

Technical Report

TR-2010-025

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MATHEMATICS AND COMPUTER SCIENCE

EMORY UNIVERSITY

DECAY PROPERTIES OF SPECTRAL PROJECTORS WITH APPLICATIONS TO ELECTRONIC STRUCTURE*

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Abstract. Motivated by applications in quantum chemistry and solid state physics, we apply general results from approximation theory and matrix analysis to the study of the decay properties of spectral projectors associated with large and sparse Hermitian matrices. Our theory leads to a rigorous proof of the exponential off-diagonal decay (‘nearsightedness’) for the density matrix of gapped systems at zero electronic temperature in both orthogonal and non-orthogonal representations, thus providing a firm theoretical basis for the possibility of linear scaling methods in electronic structure calculations for non-metallic systems. Our theory also allows us to treat the case of density matrices for arbitrary systems at finite electronic temperature, including metals. Other possible applications are also discussed.

Key words. matrix functions, orthogonal projectors, spectral gap, sparse matrices, electronic structure, localization, density functional theory

AMS subject classifications. Primary 65F50, 65F60, 65N22. Secondary 81Q05.

1. Introduction. Several theories for electronic structure calculations for large atomic and molecular systems require the solution of a sequence of linear eigenproblems for a one-electron Hamiltonian operator of the form $\mathcal{H} = -\frac{1}{2}\Delta + V$. Here Δ is the Laplacian in \mathbb{R}^d ($d = 1, 2, 3$) and V a suitable function (the potential). In practice, operators are discretized via Galerkin projection onto the finite-dimensional subspace spanned by a set of basis functions $\{\phi_i\}_{i=1}^n$. When linear combinations of atom-centered Slater or Gaussian-type functions (‘atomic orbitals’) are employed, the total number of basis functions is $n \approx n_b \cdot n_e$, where n_e is the number of (valence) electrons in the system and n_b is a small or moderate integer related to the number of basis functions per atom. Traditional electronic structure algorithms diagonalize the discrete Hamiltonian resulting in algorithms with $O(n_e^3)$ operation count [58, 68, 86]. In these approaches, a sequence of generalized eigenproblems of the form

$$H\Psi = S\Psi E \tag{1.1}$$

is solved, where H and S are, respectively, the discrete Hamiltonian and the overlap matrix relative to the basis set $\{\phi_i\}_{i=1}^n$. The columns of the $n \times n_e$ matrix Ψ in (1.1) are the eigenvectors corresponding to the n_e lowest generalized eigenvalues $\varepsilon_1 \leq \dots \leq \varepsilon_{n_e}$, which are represented by the $n_e \times n_e$ diagonal matrix E . The overlap matrix S is just the Gram matrix associated with the basis set: $S_{ij} = \langle \phi_j, \phi_i \rangle$ for all i, j , where $\langle \cdot, \cdot \rangle$ denotes the standard L^2 -inner product. In Dirac’s *bra-ket* notation, which is the preferred one in the physics and chemistry literature, one writes $S_{ij} = \langle \phi_i | \phi_j \rangle$. For an orthonormal basis set, $S = I_n$ (the $n \times n$ identity matrix) and the eigenvalue problem (1.1) is a standard one.

*Work supported by National Science Foundation grant DMS-0810862 and by a grant of the University Research Committee of Emory University.

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Instead of explicitly diagonalizing the discretized Hamiltonian H , one may reformulate the problem in terms of the density operator P , which is the S -orthogonal projector onto the H -invariant subspace corresponding to the occupied states, that is, the subspace spanned by the columns of the matrix Ψ in (1.1). Virtually all quantities of interest in electronic structure theory can be computed from P : for instance, in the Hartree approximation (see, e.g., [86]) the expected value for the total electronic energy of the system described by H is given by $\text{Tr}(PH)$. (Here and in the rest of the paper we ignore the spin for simplicity of notation.) For certain systems, this reformulation of the problem allows for the introduction of potentially more efficient algorithms for electronic structure including *linear scaling* methods, i.e., algorithms that asymptotically require only $O(n_e)$ arithmetic operations and storage. We emphasize that most current methodologies, including Hartree–Fock, Density Functional Theory (e.g., Kohn–Sham), and hybrid schemes (like BLYP) involve a linearization process, the so-called Self-Consistent Field (SCF) iteration, and that the density matrix P must be computed at each SCF step, typically with increasing accuracy as the outer iteration converges; see, e.g., [58, 103].

In this paper we use simple tools from linear algebra and approximation theory, together with a bit of functional analysis and elementary graph theory, to provide a rigorous basis for linear scaling electronic structure calculations for a very broad class of systems. We assume that the basis functions ϕ_i are *localized*, i.e., decay rapidly outside of a small region. For systems with sufficient separation between atoms, this ensures that the Hamiltonians are sparse, up to a small truncation tolerance. Our results are especially easy to state in the banded case,¹ but more general sparsity patterns will be taken into account as well.

We can also assume from the outset that the basis functions form an orthonormal set. If this is not the case, we perform a congruence transformation to an orthogonal basis and replace the original Hamiltonian H with $\tilde{H} = Z^T H Z$, where $S^{-1} = Z Z^T$ is either the Löwdin ($Z = S^{-1/2}$, see [65]) or the inverse Cholesky ($Z = L^{-T}$, with $S = LL^T$) factor of the overlap matrix S ; see, e.g., [17]. Here Z^T denotes the transpose of Z ; for the Löwdin factorization, Z is symmetric ($Z = Z^T$). Up to truncation, the transformed matrix \tilde{H} is still a banded (or sparse) matrix, albeit not as sparse as H . Hence, in our decay results we can replace H with \tilde{H} . This is further discussed in section 6 below. We note that the case of tight-binding Hamiltonians is covered by our theory. The same applies to ‘real space’ finite difference (or finite element) approximations [86], and possibly also to plane wave discretizations (see [86, page 34], particularly footnote 2).

For a given sparse discrete Hamiltonian H , we consider the problem of approximating the zero-temperature density matrix associated with H , that is, the spectral projector P onto the subspace spanned by the eigenvectors corresponding to the smallest n_e eigenvalues of H :

$$P = \Psi \Psi^* = \psi_1 \otimes \psi_1 + \cdots + \psi_{n_e} \otimes \psi_{n_e} \equiv |\psi_1\rangle\langle\psi_1| + \cdots + |\psi_{n_e}\rangle\langle\psi_{n_e}|,$$

where $H\psi_i = \varepsilon_i\psi_i$ for $i = 1, \dots, n_e$. Clearly, $P = P^* = P^2$. Consider now the Heaviside (step) function

$$h(x) = \begin{cases} 1 & \text{if } x < \mu \\ \frac{1}{2} & \text{if } x = \mu \\ 0 & \text{if } x > \mu \end{cases}$$

¹A square matrix $A = (A_{ij})$ is said to be m -banded if $A_{ij} = 0$ whenever $|i - j| > m$; for instance, a tridiagonal matrix is 1-banded according to this definition.

where μ , the *Fermi level* (or *chemical potential*), is such that $\varepsilon_{n_e} < \mu < \varepsilon_{n_e+1}$. If the *spectral gap* $\gamma = \varepsilon_{n_e+1} - \varepsilon_{n_e}$, also known as the HOMO-LUMO gap, is not too small, the step function h is well approximated by the Fermi–Dirac function² $f_{FD}(x) = 1/(1 + e^{\beta(x-\mu)})$ for suitable values of $\beta > 0$:

$$P = h(H) \approx f_{FD}(H) = [I_n + \exp(\beta(H - \mu I_n))]^{-1}.$$

The smaller γ , the larger β must be taken in order to have a good approximation. The parameter β can be interpreted as an (artificial) inverse temperature; the zero-temperature limit is quickly approached as $\beta \rightarrow \infty$. The advantage of the Fermi–Dirac function is that it is analytic, therefore we can replace h with f_{FD} and apply a wealth of results from approximation theory for analytic functions to it. Moreover, the Fermi–Dirac function is also important in the case of systems at a finite (positive) electronic temperature, in which case $\beta = (k_B T)^{-1}$, where k_B is Boltzmann’s constant and T the temperature. These systems are characterized by the presence of fractional occupation numbers.

We mention in passing that it is sometimes advantageous to impose the normalization condition $\text{Tr}(P) = 1$ on the density matrix; indeed, such a condition is standard and part of the definition of density matrix in the quantum mechanics literature, beginning with von Neumann [97, 99]. Of course, this simply means that we replace P with $\frac{1}{n_e}P$. With this normalization P is no longer idempotent, except when $n_e = 1$. In this paper we do not make use of such normalization.

Our main goal is to give a rigorous mathematical justification for the phenomenon of *localization*, i.e., the off-diagonal decay behavior of the entries of the density matrix, particularly in the case of systems with gaps, like insulators. This decay behavior has been long known to physicists and chemists; see, e.g., [25]. It finds a physical interpretation in Kohn’s “nearsightedness principle” [57], and is the basis for the development of linear scaling algorithms for electronic structure computations; see, e.g., [3, 4, 16, 17, 37, 38, 63, 68, 72, 73, 93, 101]. Our decay bounds are explicitly computable and can be used, at least in principle, to determine *a priori* the bandwidth or sparsity pattern of the truncation of the density matrix corresponding to a prescribed error. However, in this paper we are not concerned with specific algorithms for computing approximations to spectral projectors; we refer the reader to [86] for a recent survey of algorithms. Also, as we shall see, our decay estimates tend to be conservative and may be pessimistic in practice. Hence, we regard our results primarily as a theoretical contribution, providing a rigorous (yet elementary) mathematical justification for some important localization phenomena observed by physicists. An important aspect of our work is that our bounds are universal, in the sense that they only depend on the bandwidth (or sparsity pattern) of the discrete Hamiltonian H , on the smallest and largest eigenvalues of H , and on the gap γ (in practice, estimates of the latter quantities will suffice.) In this sense, our results are valid for a wide range of basis sets and indeed for different discretizations and representations of the Hamiltonian. Also, the significance of our results is not limited to electronic structure calculations and can be applied to the study of other problems, as discussed in section 9.

The remainder of the paper is organized as follows. A brief survey of previous, related work is given in section 2. In section 3 we formulate our basic assumptions on

²Several other analytic approximations to the step function are known, some of which are preferable to the Fermi–Dirac function from the computational point of view; see, e.g., [63]. For theoretical analysis, however, we found the Fermi–Dirac function to be the most convenient one to work with.

the matrices (Hamiltonians) considered in this paper, particularly their normalization and asymptotic behavior for increasing system size ($n_e \rightarrow \infty$). The approximation (truncation) of matrices with decay properties is discussed in section 4. A few general properties of orthogonal projectors are established in section 5, and in section 6 we discuss the transformation to an orthonormal basis set. The core of the paper is represented by section 7, where various types of decay bounds for spectral projectors are stated and proved. The case of vanishing gap is discussed in section 8. Other applications of our results and methods are mentioned in section 9. Finally, concluding remarks and some open problems are given in section 10.

2. Related work. The localization, i.e., decay properties of spectral projectors (more specifically, density matrices) associated with electronic structure computations in quantum chemistry and solid state physics have been the subject of a large number of papers. An incomplete list includes the pioneering work of Kohn [56] and des Cloizeaux [25], and the more recent contributions [2, 36, 39, 48, 52, 55, 57, 69, 76, 95, 96, 104]; see also the influential review [37]. These papers provide a variety of mathematical, numerical, and physical arguments that overall confirm the intuitive expectation that density matrices corresponding to gapped systems, like insulators, decay exponentially fast with the distance; the same holds for metallic systems at finite (positive) electronic temperature. Moreover, density matrices for metallic systems at zero temperature exhibit power-law (i.e., algebraic) decay. In these papers one can find results on the asymptotic decay behavior of the density matrix expressed in the following forms:

$$\rho(\mathbf{r}, \mathbf{r}') \sim e^{-\alpha|\mathbf{r}-\mathbf{r}'|} \quad (\alpha > 0, \text{ const.}) \quad (2.1)$$

for insulators, and

$$\rho(\mathbf{r}, \mathbf{r}') \sim \frac{\cos(|\mathbf{r}-\mathbf{r}'|)}{|\mathbf{r}-\mathbf{r}'|^{(d+1)/2}} e^{-k_B T|\mathbf{r}-\mathbf{r}'|} \quad (k_B > 0, \text{ const.}) \quad (2.2)$$

for d -dimensional ($d = 1, 2, 3$) metallic systems at electronic temperature $T > 0$. In these expressions, the density function $\rho : \Omega \times \Omega \rightarrow \mathbb{C}$ is the kernel of the density operator \mathcal{P} , regarded as a linear integral operator on $L^2(\Omega)$. Here $\Omega \subset \mathbb{R}^3$ is a reference domain, chosen sufficiently large. The vectors \mathbf{r} and \mathbf{r}' represent any two points in Ω , and $|\mathbf{r}-\mathbf{r}'|$ is their (Euclidean) distance. The above estimates are usually valid only for sufficiently large separation $|\mathbf{r}-\mathbf{r}'|$.

The density kernel can be expressed as

$$\rho(\mathbf{r}, \mathbf{r}') = \sum_{i=1}^{n_e} \psi_i(\mathbf{r})^* \psi_i(\mathbf{r}'),$$

where now ψ_i is the (normalized) eigenfunction of the Hamiltonian operator \mathcal{H} corresponding to the i th lowest eigenvalue, $i = 1, \dots, n_e$, and the asterisk denotes complex conjugation; see, e.g., [67]. Note that in the case of insulators, the density operator \mathcal{P} admits the Dunford integral representation

$$\mathcal{P} = \frac{1}{2\pi i} \int_{\Gamma} (zI - \mathcal{H})^{-1} dz, \quad (2.3)$$

where Γ is a simple closed contour in \mathbb{C} surrounding the eigenvalues of \mathcal{H} corresponding to the occupied states, with the remaining eigenvalues on the outside. For metallic

systems at finite temperature T , the density operator \mathcal{P} is defined via the Fermi–Dirac function:

$$\mathcal{P} = f_{FD}(\mathcal{H}), \quad f_{FD}(x) = \frac{1}{1 + e^{\beta(x-\mu)}}$$

(recall that $\beta = (k_B T)^{-1}$, where k_B is the Boltzmann constant, and that μ is the chemical potential). The zero-temperature limit is obtained by taking $T \rightarrow 0$ in (2.2), resulting in a power-law (algebraic) decay as the distance $|\mathbf{r} - \mathbf{r}'|$ increases.

We note, incidentally, that while the zero-temperature density operator is always an orthogonal projector for an insulator, the Fermi–Dirac operator at finite temperature is not. In the limit as $T \rightarrow 0$, we obtain a spectral projector onto the occupied subspace only if μ lies within the HOMO-LUMO gap.

In (2.1), the exponential rate of decay $\alpha > 0$ (also known as the inverse correlation length) depends on the spectral gap γ ; as γ decreases, so does α and the decay rate deteriorates. The actual functional dependence of α on the gap has been the subject of intense discussion, with some authors claiming that α is proportional to γ , and others finding it to be proportional to $\gamma^{1/2}$; see, e.g., [37, 52, 55, 69, 95, 96]. It appears that both types of behavior can occur in practice, even for the same system, depending on the direction [55]. We also note that in [55], the rate of decay in the case of an insulator is found to be of the form

$$\rho(\mathbf{r}, \mathbf{r}') \sim \frac{e^{-\alpha|\mathbf{r}-\mathbf{r}'|}}{|\mathbf{r} - \mathbf{r}'|^{d/2}},$$

where d is the dimensionality of the problem. In practice, however, the power-law factor in the denominator can be ignored, since the exponential decay dominates.

Despite this considerable body of work, the localization question for density matrices cannot be regarded as completely settled from the mathematical standpoint. In the papers mentioned above, the asymptotic estimates (2.1) and (2.2) are obtained on the basis of arguments which are either not completely rigorous (being often based on clever approximations, physical reasoning, and heuristic arguments), or that are rigorously valid only in some special cases—e.g. for a specific, albeit important, model system. We are not aware of any completely general and rigorous mathematical treatment of the decay properties in density matrices associated with general (localized) Hamiltonians. Moreover, rather than order-of-magnitude estimates, actual upper bounds would be more satisfactory.

From the mathematical standpoint, some of the most satisfactory results are perhaps those presented in [76]. In this paper, the authors use norm estimates for complex symmetric operators to obtain decay estimates for the resolvent of a broad class of Hamiltonians with spectral gap. Using the integral representation formula (2.3), these estimates yield what essentially amounts to an exponential decay result of the type (2.1) for the density kernel function $\rho(\mathbf{r}, \mathbf{r}')$. For large enough separation, these estimates lead to upper bounds. A lower bound on the decay rate α is also derived. Note, however, that the decay bounds in [76] are not valid pointwise but only for locally averaged quantities.

All the above-mentioned results concern the continuous, infinite-dimensional case. In practice, of course, calculations are performed on discrete, n -dimensional approximations H and P to the operators \mathcal{H} and \mathcal{P} . The replacement of density operators with finite density matrices can be obtained via the introduction of a system of n

basis functions $\{\phi_i\}_{i=1}^n$, leading to the density matrix $P = (P_{ij})$ with

$$P_{ij} = \langle \phi_j, \mathcal{P}\phi_i \rangle = \langle \phi_i | \mathcal{P} | \phi_j \rangle = \int_{\Omega} \int_{\Omega} \rho(\mathbf{r}, \mathbf{r}') \phi_i(\mathbf{r})^* \phi_j(\mathbf{r}') d\mathbf{r} d\mathbf{r}'. \quad (2.4)$$

As long as the basis functions are localized in space, the decay behavior of the density function $\rho(\mathbf{r}, \mathbf{r}')$ for increasing spatial separation $|\mathbf{r} - \mathbf{r}'|$ is reflected in the decay behavior of the matrix elements P_{ij} away from the main diagonal (i.e., for $|i - j|$ increasing) or, more generally, for increasing distance $d(i, j)$ in the graph associated with the discrete Hamiltonian; see section 4 for details.

In developing and analyzing $O(n_e)$ methods for electronic structure computations, it is crucial to rigorously establish decay bounds for the entries of the density matrices that take into account properties of the discrete Hamiltonians, since these are the objects one is dealing with in practice. It is, however, not easy to obtain decay estimates for finite-dimensional approximations from those at the continuous level. Moreover, estimates obtained inserting (2.1) or (2.2) into (2.4) would depend on the particular set of basis functions used in the discretization of the Hamiltonian.

In this paper we take a different approach. Instead of starting with the continuous problem and discretizing it, we establish our estimates directly for sequences of matrices of finite, but increasing order. We impose a minimal set of assumptions on our matrix sequences so as to reproduce the main features of problems encountered in actual electronic structure computations, while at the same time ensuring a high degree of generality. Since our aim is to provide a rigorous and general mathematical justification to the possibility of $O(n_e)$ methods, this approach seems to be quite natural.³

To put our work further into perspective, we quote from two prominent researchers in the field of electronic structure, one a mathematician, the other a physicist. In his excellent survey [58] Claude Le Bris, discussing the basis for linear scaling algorithms, i.e., the assumed sparsity of the density matrix, wrote (pages 402 and 404):

The latter assumption is in some sense an *a posteriori* assumption, and not easy to analyse [...] It is to be emphasized that the numerical analysis of the linear scaling methods overviewed above that would account for cut-off rules and locality assumptions, is not yet available.

It is interesting to compare these statements with two earlier ones by Stefan Goedecker. In [35] he wrote (page 261):

To obtain a linear scaling, the extended orbitals [i.e., the eigenfunctions of the one-particle Hamiltonian corresponding to occupied states] have to be replaced by the density matrix, whose physical behavior can be exploited to obtain a fast algorithm. This last point is essential. Mathematical and numerical analyses alone are not sufficient to construct a linear algorithm. They have to be combined with physical intuition.

A similar statement can be found in [37], page 1086:

³We refer the historically-minded reader to the interesting discussion given by John von Neumann in [98] on the benefits that can be expected from a study of the asymptotic properties of large matrices, in alternative to the study of the infinite-dimensional (Hilbert space) case.

Even though $O(N)$ algorithms contain many aspects of mathematics and computer science they have, nevertheless, deep roots in physics. Linear scaling is not obtainable by purely mathematical tricks, but it is based on an understanding of the concept of locality in quantum mechanics.

In the following we endeavour to provide a general treatment of the question of decay in spectral projectors that is as *a priori* as possible, in the sense that it relies on a minimal set of assumptions on the discrete Hamiltonians; furthermore, our theory is purely mathematical, and therefore completely independent of any physical interpretation. Of course, in the development of practical linear scaling algorithms a deep knowledge of the physics involved is extremely important; we think, however, that locality is as much a *mathematical phenomenon* as a physical one.

We hope that the increased level of generality attained in this paper (relative to previous treatments in the physics literature) will also help in the development of $O(n)$ methods for other types of problems where spectral projectors and related matrix functions play a central role. A few examples are discussed in section 9.

3. Normalizations and scalings. We will be dealing with sequences of matrices $\{H_n\}$ of increasing size. We assume that each matrix H_n is an Hermitian $n \times n$ matrix, where $n = n_b \cdot n_e$; here n_b is fixed, while n_e is increasing. As explained in the Introduction, the motivation for this assumption is that in most electronic structure codes, once a basis set has been selected the number n_b of basis functions per particle is fixed, and one is interested in the scaling as n_e , the number of particles, increases. Hence, the parameter that controls the system size is n_e . We also assume that the system is contained in a d -dimensional box of volume $V = L^d$ and that $L \rightarrow \infty$ as $n_e \rightarrow \infty$ in such a way that the average density n_e/L^d remains constant (*thermodynamic limit*). This is very different from the case of finite element or finite difference approximations to partial differential equations (PDEs), where the system (or domain) size is considered fixed while the number of basis functions increases or, equivalently, the mesh size h goes to zero.

Our scaling assumption has very important consequences on the structural and spectral properties of the matrix sequence $\{H_n\}$, namely:

1. The bandwidth of H_n , which reflects the *interaction range* of the discrete Hamiltonians, remains bounded as the system size increases. More generally, the entries of H_n decay away from the main diagonal at a rate independent of n_e (hence, of n). See section 4 for precise definitions and generalizations.
2. The eigenvalue spectra $\sigma(H_n)$ are also uniformly bounded as $n_e \rightarrow \infty$. In view of the previous property, this is equivalent to saying that the entries in H_n are uniformly bounded in magnitude: this is just a consequence of Geršgorin's Theorem (see, e.g., [50, page 344]).
3. For the case of Hamiltonians modeling insulators or semiconductors, the spectral (HOMO-LUMO) gap does not vanish as $n_e \rightarrow \infty$. More precisely: if $\varepsilon_i^{(n)}$ denotes the i th eigenvalue of H_n , and $\gamma_n := \varepsilon_{n_e+1}^{(n)} - \varepsilon_{n_e}^{(n)}$, then $\inf_n \gamma_n > 0$. This assumption does not hold for Hamiltonians modelling metallic systems; in this case, $\inf_n \gamma_n = 0$, i.e., the spectral gap goes to zero as $n_e \rightarrow \infty$.

It is instructive to contrast these properties with those of matrix sequences arising in finite element or finite difference approximations of PDEs, where the matrix size increases as $h \rightarrow 0$, with h a discretization parameter. Considering the case of a scalar, second-order elliptic PDE, we see that the first property only holds in the one-

dimensional case, or in higher-dimensional cases when the discretization is refined in only one dimension. (As we will see, this condition is rather restrictive and can be relaxed.) Furthermore, it is generally impossible to satisfy the second assumption and the one on the non-vanishing gap ($\inf_n \gamma_n > 0$) simultaneously. Indeed, normalizing the matrices so that their spectra remain uniformly bounded will generally cause the eigenvalues to completely fill the spectral interval as $n \rightarrow \infty$. That is, in general, given any two points inside this interval, for n large enough at least one eigenvalue of the corresponding $n \times n$ matrix falls between these two points.

Our assumptions allow us to refer to the spectral gap of the matrix sequence $\{H_n\}$ without having to specify whether we are talking about an absolute or a relative gap. As we shall see, it is convenient to assume that all the matrices in the sequence $\{H_n\}$ have spectrum contained in the interval $[-1, 1]$; therefore, the absolute gap and the relative gap of any matrix H_n are the same, up to the factor 2. It is well known, and it will become apparent in our discussion below, that the gap (more precisely, its reciprocal) is a natural measure of the conditioning of the problem of computing the spectral projector onto the occupied subspace, i.e., the subspace spanned by the eigenvectors of H_n corresponding to eigenvalues $\varepsilon_i^{(n)} < \mu$. The assumption $\inf_n \gamma_n > 0$ then simply means that the electronic structure problem is *uniformly well-conditioned*; note that this assumption is also very important for the convergence of the outer SCF iteration [58, 103]. This hypothesis is satisfied for insulators and semiconductors, but not in the case of metals.

4. Approximation of matrices by numerical truncation. Discretization of \mathcal{H} , the Hamiltonian operator, by means of basis sets consisting of linear combinations of Slater or Gaussian-type orbitals leads to matrix representations that are, strictly speaking, full. Indeed, since these basis functions are globally supported, almost all matrix elements $H_{ij} = \langle \phi_j, \mathcal{H}\phi_i \rangle \equiv \langle \phi_i | \mathcal{H} | \phi_j \rangle$ are non-zero. The same is true for the entries of the overlap matrix $S_{ij} = \langle \phi_j, \phi_i \rangle$. However, owing to the rapid decay of the basis functions outside of a localized region, and due to the local nature of the interactions encoded by the Hamiltonian operator, the entries of H decay exponentially fast with the spatial separation of the basis functions. (For the overlap matrix corresponding to Gaussian-type orbitals, the decay is actually faster than exponential.)

More formally, we say that a sequence of $n \times n$ matrices $A_n = ([A_n]_{ij})$ has the *exponential off-diagonal decay property* if there are constants $c > 0$ and $\alpha > 0$ independent of n such that

$$|[A_n]_{ij}| \leq c e^{-\alpha|i-j|}, \quad \text{for all } i, j = 1, \dots, n. \quad (4.1)$$

Corresponding to each matrix A_n we then define for a nonnegative integer m the matrix $A_n^{(m)} = ([A_n^{(m)}]_{ij})$ defined as follows:

$$[A_n^{(m)}]_{ij} = \begin{cases} [A_n]_{ij} & \text{if } |i-j| \leq m; \\ 0 & \text{otherwise.} \end{cases}$$

Clearly, each matrix $A_n^{(m)}$ is m -banded and can be thought of as an approximation, or truncation, of A_n . Note that the set of m -banded matrices forms a vector subspace $\mathcal{V}_m \subseteq \mathbb{C}^{n \times n}$ and that $A_n^{(m)}$ is just the orthogonal projection of A_n onto \mathcal{V}_m with respect to the Frobenius inner product $\langle A, B \rangle_F := \text{Tr}(B^* A)$. Hence, $A_n^{(m)}$ is the best approximation of A_n in \mathcal{V}_m with respect to the Frobenius norm.

Note that we do not require the matrices to be Hermitian or symmetric here; we only assume (for simplicity) that the same pattern of non-zero off diagonals is present on either side of the main diagonal. The following simple result from [11] provides an estimate of the rate at which the truncation error decreases as the bandwidth m of the approximation increases. In addition, it establishes n -independence of the truncation error for $n \rightarrow \infty$ for matrix sequences satisfying (4.1).

PROPOSITION 4.1. [11] *Let A be a matrix with entries A_{ij} satisfying (4.1) and let $A^{(m)}$ be the corresponding m -banded approximation. Then for any $\varepsilon > 0$ there is an \bar{m} such that $\|A - A^{(m)}\|_1 \leq \varepsilon$ for $m \geq \bar{m}$.*

The integer \bar{m} in the foregoing proposition is easily found to be given by

$$\bar{m} = \left\lceil \frac{1}{\alpha} \ln \left(\frac{c}{1 - e^{-\alpha}} \varepsilon^{-1} \right) \right\rceil.$$

Clearly, this result is of interest only for $\bar{m} < n$ (in fact, for $\bar{m} \ll n$). What is important about this simple result is that when applied to a sequence $\{A_n\} = ([A_n]_{ij})$ of $n \times n$ matrices having the off-diagonal decay property (4.1) with c and α independent of n , the bandwidth \bar{m} is itself independent of n . For convenience, we have stated Proposition 4.1 in the 1-norm; when $A = A^*$ the same conclusion holds for the 2-norm, owing to the inequality

$$\|A\|_2 \leq \sqrt{\|A\|_1 \|A\|_\infty}$$

(see [41, Corollary 2.3.2]). Moreover, a similar result also applies to other types of decay, such as algebraic (power-law) decay of the form

$$|[A_n]_{ij}| \leq \frac{c}{|i - j|^p + 1}, \quad \text{for all } i, j = 1, \dots, n$$

with c and p independent of n , as long as $p > 1$.

REMARK 4.2. *It is worth emphasizing that the above considerations do not require that the matrix entries $[A_n]_{ij}$ themselves actually decay exponentially away from the main diagonal, but only that they are bounded above in an exponentially decaying manner. In particular, the decay behavior of the matrix entries need not be monotonic.*

Although we have limited ourselves to absolute approximation errors in various norms, it is easy to accommodate relative errors by normalizing the matrices. Indeed, upon normalization all the Hamiltonians satisfy $\|H_n\|_2 = 1$; furthermore, for density matrices this property is automatically satisfied, since they are orthogonal projectors. In the next section we also consider using the Frobenius norm for projectors.

The foregoing considerations can be extended to matrices with more general decay patterns, i.e., with exponential decay away from a subset of selected positions (i, j) in the matrix; see, e.g., [11] as well as [20]. In order to formalize this notion, we first recall the notion of *geodesic distance* $d(i, j)$ in a graph [26]: it is the number of edges in the shortest path connecting two nodes i and j , possibly infinite if there is no such path. Next, given a (sparse) matrix sequence $\{A_n\}$ we associate with each matrix A_n a graph G_n with n nodes and $m = O(n)$ edges. In order to obtain meaningful results, however, we need to impose some restrictions on the types of sparsity allowed. Recall that the *degree* of node i in a graph is just the number of neighbors of i , i.e., the number of nodes at distance 1 from j . We denote by $\deg_n(i)$ the degree of node i in the graph G_n . We shall assume that the maximum degree of any node in G_n

remains bounded as $n \rightarrow \infty$; that is, there exists a positive integer D independent of n such that $\max_{1 \leq i \leq n} \deg_n(i) \leq D$ for all n . Note that when $A_n = H_n$ (discretized Hamiltonian), this property is a mathematical restatement of the physical notion of locality of interactions.

Now let us assume that we have a sequence of $n \times n$ matrices $A_n = ([A_n]_{ij})$ with associated graphs G_n and graph distances $d_n(i, j)$. We will say that A_n has the *exponential decay property relative to the graph G_n* if there are constants $c > 0$ and $\alpha > 0$ independent of n such that

$$|[A_n]_{ij}| \leq c e^{-\alpha d_n(i, j)}, \quad \text{for all } i, j = 1, \dots, n. \quad (4.2)$$

We have the following simple result.

PROPOSITION 4.3. *Let $\{A_n\}$ be a sequence of $n \times n$ matrices satisfying the exponential decay property (4.2) relative to a sequence of graphs $\{G_n\}$ having uniformly bounded maximal degree. Then, for any given $0 < \delta < c$, each A_n contains at most $O(n)$ entries greater than δ in magnitude.*

Proof. For a fixed node i , the condition $|[A_n]_{ij}| > \delta$ together with (4.2) immediately implies

$$d_n(i, j) < \frac{1}{\alpha} \ln \left(\frac{c}{\delta} \right). \quad (4.3)$$

Since c and α are independent of n , inequality (4.3) together with the assumption that the graphs G_n have bounded maximal degree implies that for any row of the matrix (indexed by i), there is at most a constant number of entries that have magnitude greater than δ . Hence, only $O(n)$ entries in A_n can satisfy $|[A_n]_{ij}| > \delta$. \square

REMARK 4.4. *Note that the hypothesis of uniformly bounded maximal degrees is certainly satisfied if the graphs G_n have uniformly bounded bandwidths (recall that the bandwidth of a graph is just the bandwidth of the corresponding adjacency matrix). This special case corresponds to the matrix sequence $\{A_n\}$ having the off-diagonal exponential decay property.*

Under the same assumptions of Proposition 4.3, we can show that it is possible to approximate each A_n to within an arbitrarily small error $\varepsilon > 0$ in norm with a sparse matrix $A_n^{(m)}$ (i.e., a matrix containing only $O(n)$ non-zero entries).

PROPOSITION 4.5. *Assume the hypotheses of Proposition 4.3 are satisfied. Define the matrix $A_n^{(m)} = ([A_n^{(m)}]_{ij})$, where*

$$[A_n^{(m)}]_{ij} = \begin{cases} [A_n]_{ij} & \text{if } d_n(i, j) \leq m; \\ 0 & \text{otherwise.} \end{cases}$$

Then for any given $\varepsilon > 0$, there exists \bar{m} independent of n such that $\|A_n - A_n^{(m)}\|_1 < \varepsilon$, for all $m \geq \bar{m}$. Moreover, if $A = A^$ then it is also $\|A_n - A_n^{(m)}\|_2 < \varepsilon$ for all $m \geq \bar{m}$. Furthermore, each $A_n^{(m)}$ contains only $O(n)$ non-zeros.*

Proof. For each n and m and for $1 \leq j \leq n$, let

$$K_n^m(j) := \{i \mid 1 \leq i \leq n \text{ and } d_n(i, j) > m\}.$$

We have

$$\|A_n - A_n^{(m)}\|_1 = \max_{1 \leq j \leq n} \sum_{i \in K_n^m(j)} |[A_n]_{ij}| \leq c \max_{1 \leq j \leq n} \sum_{i \in K_n^m(j)} e^{-\alpha d_n(i, j)}.$$

Letting $\lambda = e^{-\alpha}$, we obtain

$$\|A_n - A_n^{(m)}\|_1 \leq c \max_{1 \leq j \leq n} \sum_{i \in K_n^m(j)} \lambda^{d_n(i,j)} \leq c \sum_{k=m+1}^n \lambda^k < c \sum_{k=m+1}^{\infty} \lambda^k = c \frac{\lambda^{m+1}}{1-\lambda}.$$

Since $0 < \lambda < 1$, for any given $\varepsilon > 0$ we can always find \bar{m} such that

$$c \frac{\lambda^{m+1}}{1-\lambda} \leq \varepsilon \quad \text{for all } m \geq \bar{m}.$$

If $A_n = A_n^*$, then $\|A_n - A_n^{(m)}\|_2 \leq \|A_n - A_n^{(m)}\|_1 < \varepsilon$ for all $m \geq \bar{m}$. The last assertion follows from the bounded maximal degree assumption. \square

Hence, when forming the overlap matrices and discrete Hamiltonians, only matrix elements corresponding to ‘sufficiently nearby’ basis functions (i.e., basis functions having sufficient overlap) need to be computed, the others being negligibly small. The resulting matrices are therefore sparse, and indeed banded for 1D problems, with a number of non-zeros that grows linearly in the matrix dimension. The actual bandwidth, or sparsity pattern, will depend on the choice and numbering (ordering) of basis functions and (for the discrete Hamiltonians) on the strength of the interactions, i.e., on the form of the potential function V in the Hamiltonian operator.

It should be kept in mind that while the number of non-zeros in the Hamiltonians discretized using (say) Gaussian-type orbitals is $O(n_e)$, the actual number of non-zeros per row can be quite high, indeed much higher than when finite differences or finite elements are used to discretize the same operators. It is not unusual to have hundreds or even thousands of non-zeros per row. On the other hand, the matrices are very often not huge in size. As already mentioned, the size n of the matrix is the total number of basis functions, which is a small or moderate multiple (between 2 and 25, say) of the number n_e of electrons. For example, if $n_b \approx 10$ and $n_e \approx 2000$, the size of H will be $n \approx 20,000$ and H could easily contain several millions of non-zeros. This should be compared with ‘real space’ discretizations based on finite elements or high-order finite difference schemes [86]. The resulting Hamiltonians are usually very sparse (in the strict sense of the term), with a number of non-zero entries per row averaging a few tens at most [5]. However, these matrices are of much larger dimension than the matrices obtained using basis sets consisting of atom-centered orbitals. In this case, methodologies based on approximating the density matrix are currently not feasible, except for 1D problems. The same remark applies to discretizations based on plane waves, which tend to produce matrices of an intermediate size between those obtained using localized basis sets and those resulting from the use of real space discretizations. These matrices are actually dense and are never formed explicitly. Instead, they are only used in the form of matrix-vector products, which can be implemented efficiently by means of FFTs; see, e.g., [86].

The possibility of developing linear scaling methods for electronic structure largely depends on the localization properties of the density matrix P . It is therefore critical to understand the decay behavior of the density matrix. Since at zero temperature the density matrix is just a particular spectral projector, we consider next some general properties of such projectors.

5. Some general properties of orthogonal projectors. While our main goal in this paper is to study decay properties in orthogonal projectors associated with certain sequences of sparse matrices of increasing size, it is useful to first establish some *a priori* estimates for the entries of general projectors. Indeed, the intrinsic properties

of a projector like idempotency, positive semidefiniteness, and the relations between their trace, rank, and Frobenius norm tend to impose rather severe constraints on the magnitude of its entries, particularly for increasing dimension and rank.

We begin by observing that in an orthogonal projector P , all entries P_{ij} satisfy $|P_{ij}| \leq 1$ and since P is positive semidefinite, its largest entry is on the main diagonal. Also, the trace and rank coincide: $\text{Tr}(P) = \text{rank}(P)$. Moreover, $\|P\|_2 = 1$ and $\|P\|_F = \sqrt{\text{Tr}(P)}$.

In the context of electronic structure, we deal with a sequence of $n \times n$ orthogonal projectors $\{P_n\}$ of rank n_e , where $n = n_b \cdot n_e$ with n_e increasing and n_b fixed. Hence,

$$\text{Tr}(P_n) = \text{rank}(P_n) = n_e, \quad \text{and} \quad \|P_n\|_F = \sqrt{n_e}.$$

For convenience, we will refer to such a sequence $\{P_n\}$ as a *density matrix sequence*; the entries of P_n will be denoted by $[P_n]_{ij}$. We have the following lemma.

LEMMA 5.1. *Let $\{P_n\}$ be a density matrix sequence. Then*

$$\frac{\sum_{i \neq j} |[P_n]_{ij}|^2}{\|P_n\|_F^2} \leq 1 - \frac{1}{n_b}.$$

Proof. Just observe that $\text{Tr}(P_n) = \sum_{i=1}^n [P_n]_{ii} = n_e$ together with $|[P_n]_{ii}| \leq 1$ for all i imply that the minimum of the sum $\sum_{i=1}^n |[P_n]_{ii}|^2$ is achieved when $[P_n]_{ii} = \frac{n_e}{n} = \frac{1}{n_b}$ for all i . Hence, $\sum_{i=1}^n |[P_n]_{ii}|^2 \geq \frac{n_e}{n_b} = \frac{n_e}{n_b}$. Therefore,

$$\sum_{i \neq j} |[P_n]_{ij}|^2 = \|P_n\|_F^2 - \sum_{i=1}^n |[P_n]_{ii}|^2 \leq \left(1 - \frac{1}{n_b}\right) n_e \quad (5.1)$$

and the result follows dividing through by $\|P_n\|_F^2 = n_e$. \square

REMARK 5.2. *As we shall see in section 8, the bound (5.1) is sharp.*

THEOREM 5.3. *Let $\{P_n\}$ be a density matrix sequence. Then, for any $\varepsilon > 0$, the number of entries of P_n greater than or equal to ε in magnitude grows at most linearly with n .*

Proof. Clearly, it suffices to show that the number of off-diagonal entries $[P_n]_{ij}$ with $|[P_n]_{ij}| \geq \varepsilon$ can grow at most linearly with n . Let

$$\mathcal{I} = \{(i, j) \mid 1 \leq i, j \leq n \text{ and } i \neq j\} \quad \text{and} \quad \mathcal{I}_\varepsilon = \{(i, j) \in \mathcal{I} \mid |[P_n]_{ij}| \geq \varepsilon\}.$$

Then obviously

$$\sum_{i \neq j} |[P_n]_{ij}|^2 = \sum_{(i,j) \in \mathcal{I}_\varepsilon} |[P_n]_{ij}|^2 + \sum_{(i,j) \in \mathcal{I} \setminus \mathcal{I}_\varepsilon} |[P_n]_{ij}|^2$$

and if $|\mathcal{I}_\varepsilon| = K$, then

$$\sum_{i \neq j} |[P_n]_{ij}|^2 \geq K\varepsilon^2 \quad \Rightarrow \quad \frac{\sum_{i \neq j} |[P_n]_{ij}|^2}{\|P_n\|_F^2} \geq \frac{K\varepsilon^2}{n_e} = \frac{K\varepsilon^2 n_b}{n}.$$

Hence, by Lemma 5.1,

$$\frac{K\varepsilon^2 n_b}{n} \leq \frac{\sum_{i \neq j} |[P_n]_{ij}|^2}{\|P_n\|_F^2} \leq 1 - \frac{1}{n_b},$$

from which we obtain the bound

$$K \leq \frac{n}{\varepsilon^2 n_b} \left(1 - \frac{1}{n_b}\right) \quad (5.2)$$

which shows that the number K of entries of P_n with $|[P_n]_{ij}| \geq \varepsilon$ can grow at most as $O(n)$ for $n \rightarrow \infty$. \square

REMARK 5.4. *Due to the presence of the factor ε^2 in the denominator of the bound (5.2), for small ε the proportion of entries of P_n that are not smaller than ε can actually be quite large unless n is huge. Nevertheless, the result is interesting because it shows that in any density matrix sequence, the proportion of entries larger than a prescribed threshold must vanish as $n \rightarrow \infty$. In practice, for density matrices corresponding to sparse Hamiltonians the localization occurs already for moderate values of n .*

We already pointed out in the previous section that if the entries in a matrix sequence $\{A_n\}$ decay at least algebraically with exponent $p > 1$ away from the main diagonal, with rates independent of n , then for any prescribed $\varepsilon > 0$ it is possible to find a sequence of approximants $\{A_n^{(m)}\}$ with a fixed bandwidth m (or sparsity pattern \mathcal{S}) such that $\|A_n - A_n^{(m)}\| < \varepsilon$. This applies in particular to density matrix sequences. The next result shows that in principle, a *linear* rate of decay is enough to allow for banded (or sparse) approximation to within any prescribed *relative* error in the Frobenius norm.

THEOREM 5.5. *Let $\{P_n\}$ be a density matrix sequence and assume that there exists $c > 0$ independent of n such that $|[P_n]_{ij}| \leq c/(|i-j|+1)$ for all $i, j = 1, \dots, n$. Then, for all $\varepsilon > 0$, there exists a positive integer \bar{m} independent of n , such that*

$$\frac{\|P_n - P_n^{(m)}\|_F}{\|P_n\|_F} < \varepsilon \quad \text{for all } m \geq \bar{m},$$

where $P_n^{(m)}$ is the m -banded approximation obtained by setting to zero all the entries of P_n outside the band.

Proof. We subtract $P_n^{(m)}$ from P_n and compute $\|P_n - P_n^{(m)}\|_F^2$ by adding the squares of the non-zeros entries in the upper triangular part of $P_n - P_n^{(m)}$ diagonal by diagonal and multiplying the result by 2 (since the matrices are Hermitian). Using the decay assumption we obtain

$$\|P_n - P_n^{(m)}\|_F^2 \leq 2c^2 \sum_{k=1}^{n-m} \frac{k}{(n-k+1)^2} = 2c^2 \sum_{k=1}^{n-m} \frac{k}{[k-(n+1)]^2}.$$

To obtain an upper bound for the right-hand side, we observe that the function

$$f(x) = \frac{x}{(x-a)^2}, \quad a = n+1,$$

is strictly increasing and convex on the interval $[1, n-m+1]$. Hence, the sum can be bounded above by the integral of $f(x)$ taken over the same interval:

$$\sum_{k=1}^{n-m} \frac{k}{(n-k+1)^2} < \int_1^{n-m+1} \frac{x}{(x-a)^2} dx, \quad a = n+1.$$

Evaluating the integral and substituting $a = n + 1$ in the result we obtain

$$\|P_n - P_n^{(m)}\|_F^2 < 2c^2 \left[\ln\left(\frac{m}{n}\right) + (n+1) \left(\frac{1}{m} - \frac{1}{n}\right) \right].$$

Dividing by $\|P_n\|_F^2 = n_e$ we find

$$\frac{\|P_n - P_n^{(m)}\|_F^2}{\|P_n\|_F^2} < \frac{2c^2}{n_e} \left[\ln\left(\frac{m}{n}\right) + (n+1) \left(\frac{1}{m} - \frac{1}{n}\right) \right] < \frac{2c^2}{n_e} \frac{n+1}{m}.$$

Recalling that $n = n_b \cdot n_e$, we can rewrite the last inequality as

$$\frac{\|P_n - P_n^{(m)}\|_F^2}{\|P_n\|_F^2} < \frac{2c^2}{m} \frac{n+1}{n_e} = \frac{2c^2}{m} \left(n_b + \frac{1}{n_e} \right) \leq \frac{2c^2}{m} (n_b + 1),$$

a quantity which can be made arbitrarily small by taking m sufficiently large. \square

REMARK 5.6. *In practice, linear decay (or even algebraic decay with a small exponent $p \geq 1$) is too slow to be useful in the development of practical $O(n_e)$ algorithms. For example, from the above estimates we obtain $\bar{m} = O(\varepsilon^{-2})$ which is clearly not a very encouraging result, even allowing for the fact that the above bound may be pessimistic in general. To date, practical linear scaling algorithms have been developed for density matrix sequences exhibiting exponential off-diagonal decay only.*

In the case of exponential decay, one can prove the following result.

THEOREM 5.7. *Let $\{P_n\}$ be a density matrix sequence with $|[P_n]_{ij}| \leq c e^{-\alpha|i-j|}$, where $c > 0$ and $\alpha > 0$ are independent of n . Let $\{P_n^{(m)}\}$ be the corresponding sequence of m -banded approximations. Then there exists $k_0 > 0$ independent of n and m such that*

$$\frac{\|P_n - P_n^{(m)}\|_F^2}{\|P_n\|_F^2} \leq k_0 e^{-\alpha m}.$$

Proof. Similar to that of Theorem 5.5, except that it is now easy to evaluate the upper bound and the constants exactly. We omit the details. \square

REMARK 5.8. *It is immediate to see that the foregoing bound implies the much more favorable estimate $\bar{m} = O(\ln \varepsilon^{-1})$.*

Again, similar results holds for arbitrary sparsity patterns, replacing $|i - j|$ with the graph distance. More precisely, the following result holds.

THEOREM 5.9. *Let $\{P_n\}$ be a density matrix sequence with the exponential decay property with respect to a sequence of graphs $\{G_n\}$ having uniformly bounded maximal degree. Then, for all $\varepsilon > 0$, there exists a positive integer \bar{m} independent of n such that*

$$\frac{\|P_n - P_n^{(m)}\|_F}{\|P_n\|_F} < \varepsilon \quad \text{for all } m \geq \bar{m},$$

where $P_n^{(m)}$ is sparse, i.e., it contains only $O(n)$ non-zeros.

We conclude this section with a few remarks on the effect of approximating full, but localized matrices with sparse ones. As already mentioned, in electronic structure one is often interested in estimating the total electronic energy:

$$\langle E \rangle = \text{Tr}(PH) = \varepsilon_1 + \varepsilon_2 + \cdots + \varepsilon_{n_e},$$

where ε_i denotes the i th eigenvalue of the discrete Hamiltonian H . Now, assume that $\hat{H} \approx H$ and $\hat{P} \approx P$, and define the approximate electronic energy as $\langle \hat{E} \rangle = \text{Tr}(\hat{P}\hat{H})$. (We note in passing that in order to compute $\langle \hat{E} \rangle = \text{Tr}(\hat{P}\hat{H})$, only the entries of \hat{P} corresponding to non-zero entries in \hat{H} need to be computed.) Let $\Delta_P = \hat{P} - P$ and $\Delta_H = \hat{H} - H$. We have

$$\langle \hat{E} \rangle = \text{Tr}[(P + \Delta_P)(H + \Delta_H)] = \text{Tr}(PH) + \text{Tr}(P\Delta_H) + \text{Tr}(\Delta_HP) + \text{Tr}(\Delta_P\Delta_H).$$

Neglecting the last term, we obtain for $\delta_E = |\langle E \rangle - \langle \hat{E} \rangle|$ the bound

$$\delta_E \leq |\text{Tr}(P\Delta_H)| + |\text{Tr}(\Delta_HP)|.$$

Recalling that the Frobenius norm is the matrix norm induced by the inner product $\langle A, B \rangle = \text{Tr}(B^*A)$, using the Cauchy–Schwarz inequality and $\|P\|_F = \sqrt{n_e}$ we find

$$\delta_E \leq \sqrt{n_e} \|\Delta_H\|_F + \|\Delta_P\|_F \|H\|_F.$$

Now, since the orthogonal projector P is invariant with respect to scalings of the Hamiltonian, we can assume $\|H\|_F = 1$, so that $\delta_E \leq \sqrt{n_e} \|\Delta_H\|_F + \|\Delta_P\|_F$ holds. In practice, a bound on the *relative* error would be more meaningful. Unfortunately, it is not easy to obtain a rigorous bound in terms of the relative error in the approximate projector \hat{P} . If, however, we replace the relative error in $\langle \hat{E} \rangle$ with the normalized error obtained dividing the absolute error by the number n_e of electrons, we obtain

$$\frac{\delta_E}{n_e} \leq \frac{\|\Delta_H\|_F}{\sqrt{n_e}} + \frac{\|\Delta_P\|_F}{n_e}.$$

Using $n = n_b \cdot n_e$ and the fact that $\|A\|_F \leq \sqrt{n} \|A\|_2$ for any $n \times n$ matrix A , we finally obtain

$$\frac{\delta_E}{n_e} \leq \sqrt{n_b} \left(\|\Delta_H\|_2 + \frac{1}{\sqrt{n_e}} \|\Delta_P\|_2 \right). \quad (5.3)$$

Since n_b is constant, an interesting consequence of (5.3) is that for large system sizes (i.e., in the limit as $n_e \rightarrow \infty$), the normalized error in the total energy is essentially determined by the truncation error in the Hamiltonian H rather than by the error in the density matrix P .

On the other hand, scaling H so that $\|H\|_F = 1$ may not be advisable in practice. Indeed, since the Frobenius norm of the Hamiltonian grows unboundedly for $n_e \rightarrow \infty$, rescaling H so that $\|H\|_F = 1$ would lead to a loss of significant information when truncation is applied in the case of large systems. A more sensible scaling, which is often used in algorithms for electronic structure computations, is to divide $\|H\|$ by its largest eigenvalue in magnitude, so that $\|H\|_2 = 1$. This is consistent with the assumption, usually satisfied in practice, that the spectra of the Hamiltonians remain bounded as $n_e \rightarrow \infty$. (Note this is the same normalization used to establish the decay bounds in section 7.) With this scaling we readily obtain, to first order, the bound

$$\frac{\delta_E}{n_e} \leq \sqrt{n_b} \|\Delta_H\|_2 + n_b \|\Delta_P\|_2, \quad (5.4)$$

showing that errors in Δ_H and Δ_P enter the estimate for the normalized error in the total electronic energy with approximately the same weight, since n_b is a moderate

constant. We also note that since both error matrices Δ_H and Δ_P are symmetric, the bounds (5.3) and (5.4) remain true if the 2-norm is replaced by the 1-norm. We mention that the problem of the choice of norm in the measurement of truncation errors has been discussed in [82, 84]. These authors emphasize the use of the 2-norm, which is related to the distance between the exact and inexact (perturbed) occupied subspaces $\mathcal{X} := \text{Range}(P)$ and $\hat{\mathcal{X}} := \text{Range}(\hat{P})$ as measured by the sine of the principal angle between \mathcal{X} and $\hat{\mathcal{X}}$; see [82].

One important practical aspect, which we do not address here, is that in many quantum chemistry codes the matrices have a natural block structure (where each block corresponds, for instance, to the basis functions centered at a given atom); hence, dropping is usually applied to submatrices rather than to individual entries. Exploitation of the block structure is also desirable in order to achieve high performance in matrix-matrix products and other operations. See, e.g., [17, 18, 83].

We conclude this section with a few remarks on the infinite-dimensional case. Recall that any separable, complex Hilbert space \mathcal{H} is isometrically isomorphic to the sequence space

$$\ell^2 := \left\{ (\xi_n) \mid \xi_n \in \mathbb{C} \ \forall n \in \mathbb{N} \ \text{and} \ \sum_{n=1}^{\infty} |\xi_n|^2 < \infty \right\}.$$

Moreover, if $\{e_n\}$ is an orthonormal basis in \mathcal{H} , to any bounded linear operator \mathcal{A} on \mathcal{H} there corresponds precisely one infinite matrix $A = (A_{ij})$ acting on ℓ^2 , with $A_{ij} = \langle e_j, \mathcal{A}e_i \rangle$. Note that each column of A must be in ℓ^2 , hence the entries A_{ij} in each column of A must go to zero for $i \rightarrow \infty$. The same is true for the entries in each row (for $j \rightarrow \infty$) since $A^* = (A_{ji}^*)$, the adjoint of A , is also a (bounded) operator defined everywhere on ℓ^2 . More precisely, for any bounded linear operator $A = (A_{ij})$ on ℓ^2 the following bounds hold:

$$\sum_{j=1}^{\infty} |A_{ij}|^2 \leq \|A\|_2^2 \quad \text{for all } i \quad \text{and} \quad \sum_{i=1}^{\infty} |A_{ij}|^2 \leq \|A\|_2^2 \quad \text{for all } j, \quad (5.5)$$

since $\|A\|_2 = \|A^*\|_2$.

An orthogonal projector \mathcal{P} on \mathcal{H} is a self-adjoint ($\mathcal{P} = \mathcal{P}^*$), idempotent ($\mathcal{P} = \mathcal{P}^2$) linear operator. Such an operator is necessarily bounded, with norm $\|\mathcal{P}\| = 1$. Hence, (5.5) implies

$$\sum_{j=1}^{\infty} |P_{ij}|^2 \leq 1. \quad (5.6)$$

Denoting by $P = (P_{ij})$ the matrix representation of \mathcal{P} , the idempotency condition implies

$$P_{ij} = \sum_{k=1}^{\infty} P_{ik} P_{kj}, \quad \text{for all } i, j = 1, 2, \dots$$

In particular, for $i = j$ we get, using the hermiticity property $P_{ij} = P_{ji}^*$:

$$P_{ii} = \sum_{k=1}^{\infty} P_{ik} P_{ki} = \sum_{k=1}^{\infty} |P_{ik}|^2, \quad \text{for all } i = 1, 2, \dots \quad (5.7)$$

Now, since P is a projector its entries satisfy $|P_{ij}| \leq 1$, therefore (5.7) is a strengthening of inequality (5.6). Note in particular that the off-diagonal entries in the first row (or column) of P must satisfy

$$\sum_{j>1} |P_{1j}|^2 \leq 1 - |P_{11}|^2,$$

those in the second row (or column) must satisfy

$$\sum_{j>2} |P_{2j}|^2 \leq 1 - |P_{22}|^2 - |P_{12}|^2,$$

and in general the entries P_{ij} with $j > i$ must satisfy

$$\sum_{j>i} |P_{ij}|^2 \leq 1 - \sum_{k=1}^i |P_{ki}|^2 \quad \text{for all } i = 1, 2, \dots \quad (5.8)$$

Hence, decay in the off-diagonal entries in the i th row of P must be fast enough for the bounds (5.8) to hold. In general, however, it is not easy to quantify the asymptotic rate of decay to zero of the off-diagonal entries in an arbitrary orthogonal projector on ℓ^2 . In general, the rate of decay can be rather slow. In section 8 we will see an example of spectral projector associated with a very simple tridiagonal Hamiltonian for which the off-diagonal entries decay linearly to zero.

6. Transformation to an orthonormal basis. In this section we discuss the transformation of an Hamiltonian from a non-orthogonal to an orthogonal basis. The main point is that while this transformation results in matrices with less sparsity, the transformed matrices retain the decay properties of the original matrices, only with (possibly) different constants. What is important, from the point of view of asymptotic complexity, is that the rate of decay remains independent of system size.

We begin with a discussion of decay in the inverse of the overlap matrix. To this end, consider a sequence $\{S_n\}$ of overlap matrices of size $n = n_b \cdot n_e$, with n_b constant and n_e increasing to infinity. We make the following assumptions:

1. Each S_n is a banded symmetric positive definite (SPD) matrix with unit diagonal entries and with bandwidth uniformly bounded with respect to n ;
2. The spectral condition number (ratio of the largest to the smallest eigenvalue) of each S_n , $\kappa_2(S_n)$, is uniformly bounded with respect to n . Because of assumption 1, this is equivalent to requiring that the smallest eigenvalue of S_n remains bounded away from zero, for all n .

As always in this paper, the bandedness assumption in item 1 is not essential and can be replaced by the weaker hypothesis that each S_n is sparse and that the corresponding graphs $\{G_n\}$ have bounded maximal degree with respect to n . Actually, it would be enough to require that the sequence $\{S_n\}$ has the exponential decay property relative to a sequence of graphs $\{G_n\}$ of bounded maximal degree. In order to simplify the discussion, and also in view of the fact that overlap matrices usually exhibit exponential or even superexponential decay, we assume from the outset that each S_n has already been truncated to a sparse (or banded) matrix. Again, this is for notational convenience only, and it is straightforward to modify the following arguments to account for the more general case. On the other hand, the assumption on condition numbers in item 2 is essential and cannot be weakened. Note that since the overlap matrix represents the identity operator in a non-orthogonal basis, the

requirement that the sequence $\{S_n\}$ has asymptotically bounded condition numbers is reasonable and is usually satisfied by most basis sets.

REMARK 6.1. *We note that assumption 2 above is analogous to the condition that the sequence of Hamiltonians $\{H_n\}$ has spectral gap bounded below uniformly in n ; while this condition ensures (as we will see) the exponential decay property in the associated spectral projectors P_n , assumption 2 above insures exponential decay in the inverses (or inverse factors) of the overlap matrices. Both conditions amounts to asking that the corresponding problems be uniformly well-conditioned in n . The difference is that the decay on the spectral projectors depends on the spectral gap of the Hamiltonians and therefore on the physical properties of the system under study (e.g., insulator vs. metallic system), whereas the sparsity and spectral properties of the overlap matrices are determined exclusively by the basis set and not by the physical properties of the system [81]. Differently put: the overlap matrix, unlike the spectral projector, is not a function of the Hamiltonian.*

In the following we shall need some basic results on the decay of the inverses [23], inverse Cholesky factors [12] and inverse square roots (Löwdin factors) [9] of banded SPD matrices; see also [53].

Let A be SPD and m -banded, and let a and b denote the smallest and largest eigenvalue of A , respectively. Write κ for the spectral condition number $\kappa_2(A)$ of A (hence, $\kappa = b/a$). Define

$$q := \frac{\sqrt{\kappa} - 1}{\sqrt{\kappa} + 1} \quad \text{and} \quad \lambda := q^{1/m}.$$

Furthermore, let $K_0 := (1 + \sqrt{\kappa})^2 / (2b)$. In [23], Demko et al. obtained the following bound on the entries of A^{-1} :

$$|[A^{-1}]_{ij}| \leq K \lambda^{|i-j|}, \quad 1 \leq i, j \leq n, \quad (6.1)$$

where $K := \max\{a^{-1}, K_0\}$. Note that the bound (6.1) ‘blows up’ as $\kappa \rightarrow \infty$, as one would expect.

As shown in [12], the decay bound (6.1) and the bandedness assumption on A imply a similar decay bound on the inverse Cholesky factor $Z = R^{-1} = L^{-T}$, where $A = R^T R = LL^T$ with R upper triangular (L lower triangular). Assuming that A has been scaled so that $\max_{1 \leq i \leq n} A_{ii} = 1$ (which is automatically true if A is an overlap matrix corresponding to a set of normalized basis functions), we have

$$|Z_{ij}| \leq K_1 \lambda^{j-i}, \quad j \geq i, \quad (6.2)$$

with $K_1 = K \frac{1-\lambda^m}{1-\lambda}$; here K, λ are the same as before. We further note that while $K_1 > K$, for some classes of matrices it is possible to show that the actual magnitude of the (i, j) entry of Z (as opposed to the bound (6.2)) is actually less than the magnitude of the corresponding entry of A^{-1} . This is true, for instance, for an irreducible M -matrix; see [12].

Finally, let us consider the inverse square root, $A^{-1/2}$. In [9] the following bound is established:

$$\left| [A^{-1/2}]_{ij} \right| \leq K_2 \lambda^{|i-j|}, \quad 1 \leq i, j \leq n. \quad (6.3)$$

Here K_2 depends again on the extreme eigenvalues a and b of A , whereas $\lambda = q^{1/m}$, where q is any number satisfying the inequalities

$$\frac{\kappa - 1}{(\sqrt{\kappa} + 1)^2} < q < 1.$$

As before, the bound (6.3) blows up as $\kappa \rightarrow \infty$, as one would expect.

Introducing the positive scalar $\alpha = -\ln \lambda$, we can rewrite all these bounds in the form

$$|B_{ij}| \leq K e^{-\alpha|i-j|}, \quad 1 \leq i, j \leq n$$

for the appropriate matrix B and suitable constants K and $\alpha > 0$.

Let now $\{S_n\}$ be a sequence of $n \times n$ overlap matrices, where $n = n_b \cdot n_e$ with n_b fixed and $n_e \rightarrow \infty$. Assuming that the matrices S_n satisfy the above assumptions 1-2, then their inverses satisfy the uniform exponential decay bounds (6.1), with K and λ constant and independent of n . Hence, as discussed in section 4, for any given $\varepsilon > 0$ there exists an integer \bar{m} independent of n such that each matrix S_n in the sequence can be approximated, in norm, by an \bar{m} -banded matrix with an error less than ε . As usual, this result can be extended from the banded case to the sparse case, assuming that the corresponding graphs G_n have bounded maximal degree as $n \rightarrow \infty$. Moreover, under assumptions 1-2 above, the inverse Cholesky factors Z_n satisfy a uniform (in n) exponential decay bound of the type (6.2), and therefore uniform approximation with banded triangular matrices is possible. Again, generalization to more general sparsity patterns is possible, provided the usual assumption on the maximum degree of the corresponding graphs G_n holds. Similarly, under the same conditions we obtain a uniform rate of exponential decay for the entries of the inverse square roots $S_n^{-1/2}$, with a corresponding result on the existence of a banded (or sparse) approximation.

Let us now consider the sequence of transformed Hamiltonians, $\tilde{H}_n = Z_n^T H_n Z_n$. Here Z_n denotes either the inverse Cholesky factor or the inverse square root of the corresponding overlap matrix S_n . Assuming that the sequence $\{H_n\}$ satisfies the off-diagonal exponential decay property and that $\{S_n\}$ satisfies assumptions 1-2 above, it follows from the decay properties of the matrix sequence $\{Z_n\}$ that the sequence $\{\tilde{H}_n\}$ also enjoys off-diagonal exponential decay. This is a straightforward consequence of the following result, which is adapted from a similar one for infinite matrices due to Jaffard [53, Proposition 1].

THEOREM 6.2. *Consider two sequences $\{A_n\}$ and $\{B_n\}$ of $n \times n$ matrices (where $n \rightarrow \infty$) whose entries satisfy*

$$|[A_n]_{ij}| \leq c_1 e^{-\alpha|i-j|} \quad \text{and} \quad |[B_n]_{ij}| \leq c_2 e^{-\alpha|i-j|}, \quad 1 \leq i, j \leq n,$$

where c_1, c_2 and $\alpha > 0$ are independent of n . Then the sequence $\{C_n\}$, where $C_n = A_n B_n$, satisfies a similar bound:

$$|[C_n]_{ij}| \leq c e^{-\alpha|i-j|}, \quad 1 \leq i, j \leq n, \quad (6.4)$$

with c independent of n .

Proof. First note that the entries of each A_n clearly satisfy

$$|[A_n]_{ij}| \leq c_1 e^{-\alpha'|i-j|} \quad \text{for any} \quad \alpha' < \alpha.$$

Let $\omega = \alpha - \alpha'$. Then $\omega > 0$ and the entries $[C_n]_{ij}$ of $C_n = A_n B_n$ satisfy

$$|[C_n]_{ij}| \leq \sum_{k=1}^n |[A_n]_{ik}| |[B_n]_{kj}| \leq c_1 c_2 \left(\sum_{k=1}^n e^{-\omega|k-j|} \right) e^{-\alpha|i-j|}.$$

To complete the proof just observe that for any j ,

$$\sum_{k=1}^n e^{-\omega|k-j|} = \sum_{k=0}^{j-1} e^{-\omega k} + \sum_{k=1}^{n-j} e^{-\omega k} < \sum_{k=0}^{\infty} e^{-\omega k} + \sum_{k=1}^{\infty} e^{-\omega k} < \frac{1 + e^{-\omega}}{1 - e^{-\omega}}.$$

Since the last term is independent of n , the entries of C_n satisfy (6.4) with a constant c that is also independent of n . \square

The foregoing result can obviously be extended to the product of three (or more) matrices. Thus, the entries of the matrix sequence $\{\tilde{H}_n\}$, where $\tilde{H}_n = Z_n^T H_n Z_n$, enjoy the exponential off-diagonal decay property:

$$|[\tilde{H}_n]_{ij}| \leq c e^{-\alpha|i-j|}, \quad 1 \leq i, j \leq n,$$

for suitable constants c and $\alpha > 0$. The decay constant α is the smallest between the corresponding ones for H_n and Z_n . (In some cases, this will be the same α ; see, e.g., [2].) The constant c , on the other hand, will be larger than the constants for both H_n and Z_n .

Alternatively, one could first approximate H_n and Z_n with banded matrices \hat{H}_n and \hat{Z}_n and then define the (approximate) transformed Hamiltonian as $\tilde{H}_n := \hat{Z}_n^T \hat{H}_n \hat{Z}_n$, possibly subject to further truncation. Using the fact that both H_n and Z_n have 2-norm bounded independently of n , it is easy to show that the final approximation error can be reduced below any prescribed tolerance by reducing the error in \hat{H}_n and \hat{Z}_n . Hence, with either approach, the transformed Hamiltonians \tilde{H}_n can be approximated uniformly in n within a prescribed error by banded matrices of constant bandwidth, just like the original (“non-orthogonal”) Hamiltonians. While the bandwidth of the approximations will be larger than for the original Hamiltonians, the truncated matrices retain a good deal of sparsity and asymptotically contain $O(n)$ (hence, $O(n_e)$) non-zeros. Hence, we have a justification of the statement (see the Introduction) that in our theory we can assume from the outset that the basis set $\{\phi_i\}_{i=1}^n$ is orthonormal.

In Fig. 6.1 we show the Hamiltonian \hat{H} for a chain composed of 52 Carbon and 106 Hydrogen atoms ($C_{52}H_{106}$) discretized in an atomic orbital basis (left) and the ‘orthogonalized’ Hamiltonian $\tilde{H} = \hat{Z}^T \hat{H} \hat{Z}$ (right). This figure shows that while the transformation to orthogonal basis alters the magnitude of the entries in the Hamiltonian, the bandwidth of \tilde{H} (truncated to a tolerance of 10^{-8}) is only slightly wider than that of \hat{H} .

As usual, the bandedness assumption was made for simplicity of exposition only; similar bounds can be obtained for more general sparsity patterns, assuming the matrices H_n and S_n have the exponential decay property relative to a sequence $\{G_n\}$ of graphs having maximal degree uniformly bounded with respect to n .

It is important to emphasize that in practice, the explicit formation of \tilde{H}_n from H_n and Z_n is not needed and is never carried out. Indeed, in all algorithms for electronic structure computation the basic matrix operations are matrix-matrix and matrix-vector products, which can be performed without explicit transformation of the Hamiltonian to an orthonormal basis. On the other hand, for the study of the decay properties it is convenient to assume that all the relevant matrices are explicitly given in an orthogonal representation.

One last issue to be addressed is whether the transformation to an orthonormal basis should be effected via the inverse Cholesky factor or via the Löwdin (inverse square root) factor of the overlap matrix. Comparing the decay bounds for the two

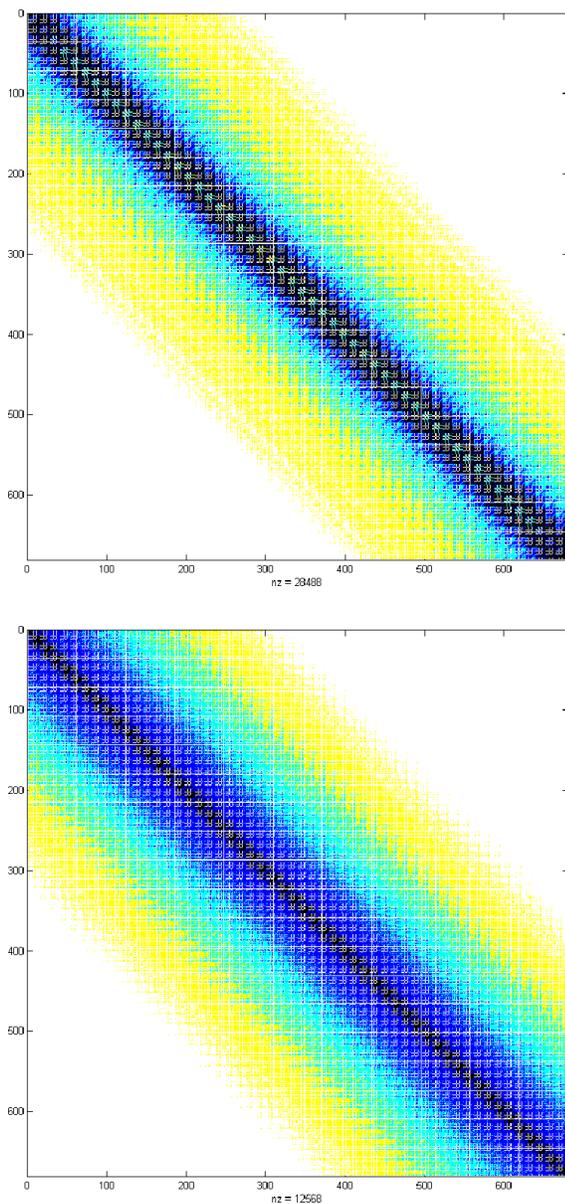


FIG. 6.1. Magnitude of the entries in the Hamiltonian for $C_{52}H_{106}$ hydrocarbon chain. Top: non-orthogonal (AO) basis. Bottom: orthogonal basis. White: $< 10^{-8}$; yellow: $10^{-8} - 10^{-6}$; green: $10^{-6} - 10^{-4}$; blue: $10^{-4} - 10^{-2}$; black: $> 10^{-2}$. Note: **nz** refers to the number of ‘black’ entries.

factors suggests that the inverse Cholesky factor should be preferred (smaller α). Also note that the inverse Cholesky factor is triangular, and its sparsity can be increased by suitable reorderings of the overlap matrix. In contrast, the Löwdin factor is a full symmetric matrix, regardless of the ordering. On the other hand, the multiplicative constant c is generally smaller for the Löwdin factor. Closer examination of a few examples suggests that in practice there is no great difference in the actual decay

behavior of these two factors. However, approximating $S_n^{-1/2}$ is generally more expensive and considerably more involved than approximating the inverse Cholesky factor. For the latter, the AINV algorithm [10] and its variants [17, 80, 102] are quite efficient and have been successfully used in various quantum chemistry codes. For other $O(n)$ algorithms for transformation to an orthonormal basis, see [54, 74, 92]. In all these algorithms, sparsity is preserved by dropping small entries in the course of the computation. Explicit decay bounds for the Z_n factors could be used, in principle, to establish *a priori* which matrix elements *not* to compute, thus reducing the amount of overhead. Notice, however, that even if asymptotically bounded, the condition numbers $\kappa_2(S_n)$ can be fairly large, leading to rather pessimistic decay estimates. This is perfectly analogous to the situation with the condition number-based error bounds for the conjugate gradient (CG) method applied to a linear system $Ax = b$; see, e.g., [41, Theorem 10.2.6]. And indeed, both the CG error bounds and the estimates (6.1) are obtained by using Chebyshev polynomial approximation to the function $f(\lambda) = \lambda^{-1}$.

7. Decay results. In this section we present and discuss some results on the decay of entries for the Fermi–Dirac function applied to Hamiltonians and for the density matrix (spectral projector corresponding to occupied states). We consider both the banded case and the case of more general sparsity patterns. The proofs, which require some basic tools from approximation theory, will be given in the next subsection.

7.1. Bounds for the Fermi–Dirac function. We begin with the following result for the banded case. As usual in this paper, in the following one should think of the positive integer n as being of the form $n = n_b \cdot n_e$ with n_b constant and $n_e \rightarrow \infty$.

THEOREM 7.1. *Let m be a fixed positive integer and consider a sequence of matrices $\{H_n\}$ such that:*

- (i) H_n is an $n \times n$ Hermitian, m -banded matrix for all n ;
- (ii) For every n , all the eigenvalues of H_n lie in the interval $[-1, 1]$.

For a given Fermi level μ and inverse temperature β , define for each n the $n \times n$ Hermitian matrix $F_n := f_{FD}(H_n) = [I + e^{\beta(H_n - \mu I)}]^{-1}$. Then there exist constants $c > 0$ and $\alpha > 0$, independent of n , such that the following decay bound holds:

$$|[F_n]_{ij}| \leq c e^{-\alpha|i-j|}, \quad i \neq j. \quad (7.1)$$

The constants c and α can be chosen as

$$c = \frac{2\chi M(\chi)}{\chi - 1}, \quad M(\chi) = \max_{z \in \mathcal{E}_\chi} |f_{FD}(z)|, \quad (7.2)$$

$$\alpha = \frac{1}{m} \ln \chi, \quad (7.3)$$

for any $1 < \chi < \bar{\chi}$, where

$$\begin{aligned} \bar{\chi} = & \frac{\sqrt{\sqrt{(\beta^2(1 - \mu^2) - \pi^2)^2 + 4\pi^2\beta^2} - \beta^2(1 - \mu^2) + \pi^2}}{\sqrt{2}\beta} + \\ & + \frac{\sqrt{\sqrt{(\beta^2(1 - \mu^2) - \pi^2)^2 + 4\pi^2\beta^2} + \beta^2(1 + \mu^2) + \pi^2}}{\sqrt{2}\beta}, \end{aligned} \quad (7.4)$$

and \mathcal{E}_χ is the unique ellipse with foci in -1 and 1 with semi-axes $\kappa_1 > 1$ and $\kappa_2 > 0$, and $\chi = \kappa_1 + \kappa_2$.

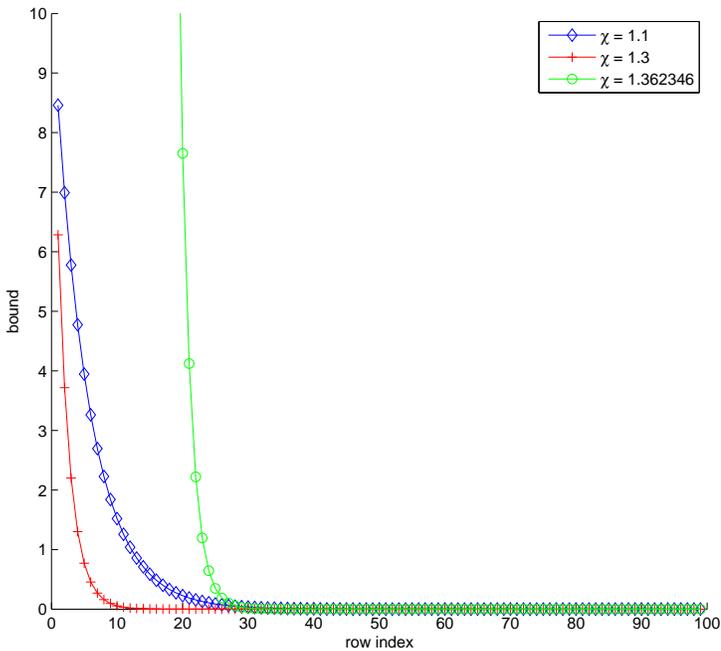


FIG. 7.1. Bounds (7.1) with $\mu = 0$ and $\beta = 10$, for three different values of χ .

REMARK 7.2. The ellipse \mathcal{E}_χ in the previous theorem is unique because the identity $\sqrt{\kappa_1^2 - \kappa_2^2} = 1$, valid for any ellipse, implies $\kappa_1 - \kappa_2 = 1/(\kappa_1 + \kappa_2)$, hence the parameter $\chi = \kappa_1 + \kappa_2$ alone completely characterizes the ellipse.

REMARK 7.3. Theorem 7.1 can be immediately generalized to the case where the spectra of the sequence $\{H_n\}$ are contained in an interval $[a, b]$, for any $a < b \in \mathbb{R}$. It suffices to shift and scale each Hamiltonian:

$$\widehat{H}_n = \frac{2}{b-a} H_n - \frac{a+b}{b-a} I,$$

so that \widehat{H}_n has spectrum in $[-1, 1]$. For the decay bounds to be independent of n , however, a and b must be independent of n .

It is important to note that there is a certain amount of arbitrariness in the choice of χ , and therefore of c and α . If one is mainly interested in a fast asymptotic decay behavior (i.e., for sufficiently large $|i - j|$), it is desirable to choose χ as large as possible. On the other hand, if χ is very close to $\bar{\chi}$ then the constant c is likely to be quite large and the bounds might be too pessimistic. Let us look at an example. Take $\mu = 0$; in this case we have

$$\bar{\chi} = \left(\pi + \sqrt{\beta^2 + \pi^2} \right) / \beta \quad \text{and} \quad M(\chi) = \left| 1 / \left(1 + e^{\beta(\zeta - \mu)} \right) \right|, \quad \text{where} \quad \zeta = i \frac{\chi^2 - 1}{2\chi}.$$

Note that, in agreement with experience, decay is faster for smaller β (i.e., higher electronic temperatures). Figures 7.1 and 7.2 show the behavior of the bound given

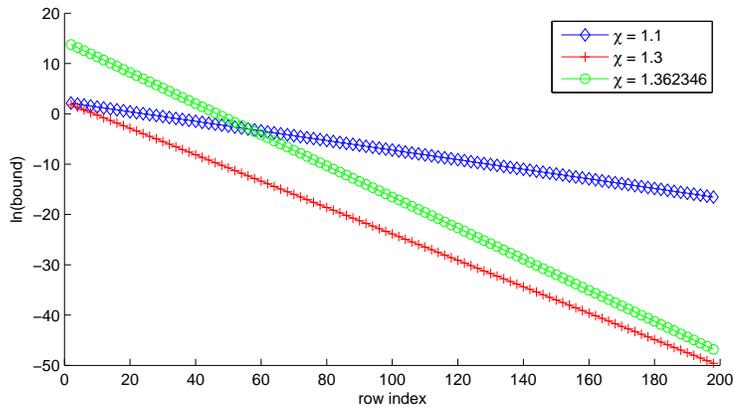


FIG. 7.2. *Logarithmic plot of the bounds (7.1) with $\mu = 0$ and $\beta = 10$.*

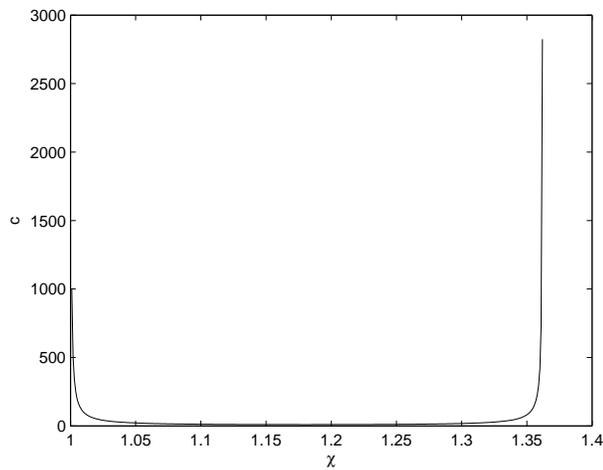


FIG. 7.3. *Plot of c as a function of χ with $\mu = 0$ and $\beta = 10$.*

by (7.1) on the first row of a 200×200 tridiagonal matrix ($m = 1$) for $\beta = 10$ and for three values of χ . It is easy to see from the plots that the asymptotic behavior of the bounds improves as χ increases; however, the bound given by $\chi = 1.362346$ is less useful than the bound given by $\chi = 1.3$. Figure 7.3 is a plot of c as a function of χ and it shows that c grows very large when χ is close to $\bar{\chi}$. This is expected, since $f_{FD}(z)$ has two poles, given by $z = \pm i\pi/\beta$ on the regularity ellipse $\mathcal{E}_{\bar{\chi}}$. It is clear from Figures 7.1 and 7.2 that $\chi = 1.3$ is the best choice among the three proposed values, if one is interested in determining a bandwidth outside of which the entries of F_n can be safely neglected. As already observed in [11, 77], improved bounds can be obtained by adaptively choosing different (typically increasing) values of χ as $|i - j|$ grows, and use as a bound the (lower) envelope of the curves plotted in Figure 7.4, which shows the behavior of the decay bounds for several values of $\chi \in (1.1, \bar{\chi})$, where $\bar{\chi} \approx 1.3623463$.

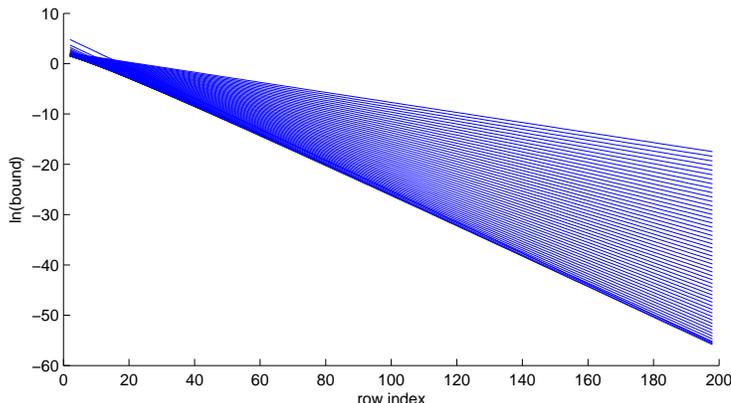


FIG. 7.4. Logarithmic plot of the bounds (7.1) with $\mu = 0$ and $\beta = 10$, for several values of χ .

The results of Theorem 7.1 can be generalized to the case of Hamiltonians with rather general sparsity patterns; see [11, 20, 77]. To this end, we make use of the notion of geodesic distance in a graph already used in section 4. The following result holds.

THEOREM 7.4. *Consider a sequence of matrices $\{H_n\}$ such that:*

- (i) *H_n is an $n \times n$ Hermitian matrix for all n ;*
- (ii) *the spectra $\sigma(H_n)$ are uniformly bounded and contained in $[-1, 1]$ for all n .*

Let $d_n(i, j)$ be the graph distance associated with H_n . Then the following decay bound holds:

$$|[F_n]_{ij}| \leq c e^{-\theta d_n(i,j)}, \quad i \neq j, \quad (7.5)$$

where $\theta = \ln(1/\chi)$ and the remaining notation and choice of constants are as in Theorem 7.1.

We remark that in order for the bound (7.5) to be meaningful from the point of view of linear scaling, we need to impose some restrictions on the asymptotic sparsity of the graph sequence $\{G_n\}$. As discussed in section 4, $O(n)$ approximations of F_n are possible if the graphs G_n have maximum degree uniformly bounded with respect to n . This guarantees that the distance $d_n(i, j)$ grows unboundedly as $|i - j|$ does, at a rate independent of n for $n \rightarrow \infty$.

7.2. Density matrix decay for systems with gap. The previous results establish exponential decay bounds for the Fermi–Dirac function of general localized Hamiltonians and thus for density matrices of arbitrary systems at positive electronic temperature. In this subsection we consider the case of gapped systems (like insulators) at zero temperature. In this case, as we know, the density matrix is the spectral projector onto the occupied subspace. As an example, we consider again the $C_{52}H_{106}$ hydrocarbon chain previously considered. The Hamiltonian for this system (in an orthogonal basis) has been shown in Fig. 6.1. The number of occupied states is 209, or half the total number of electrons in the system.⁴

Fig. 7.5 displays the zero temperature density matrix, which is seen to decay exponentially away from the main diagonal. Comparing Fig. 7.5 and Fig. 6.1, we

⁴Here spin is being taken into account, so that the density kernel is given by $\rho(\mathbf{r}, \mathbf{r}') = 2 \sum_{i=1}^{n_e/2} \psi_i^*(\mathbf{r}) \psi_i(\mathbf{r}')$; see, e.g., [67, page 10].

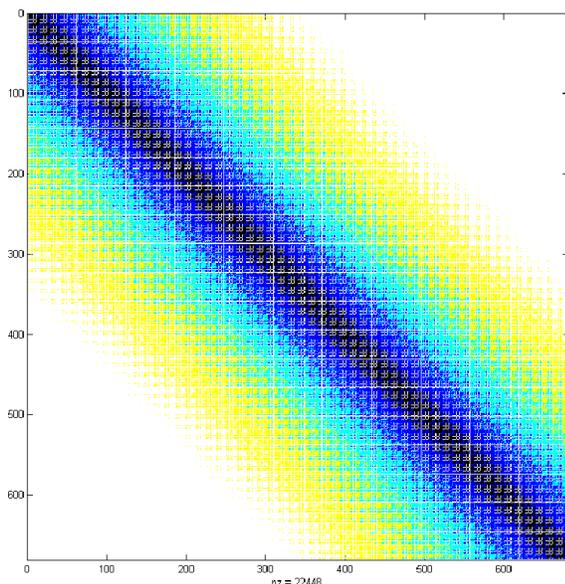
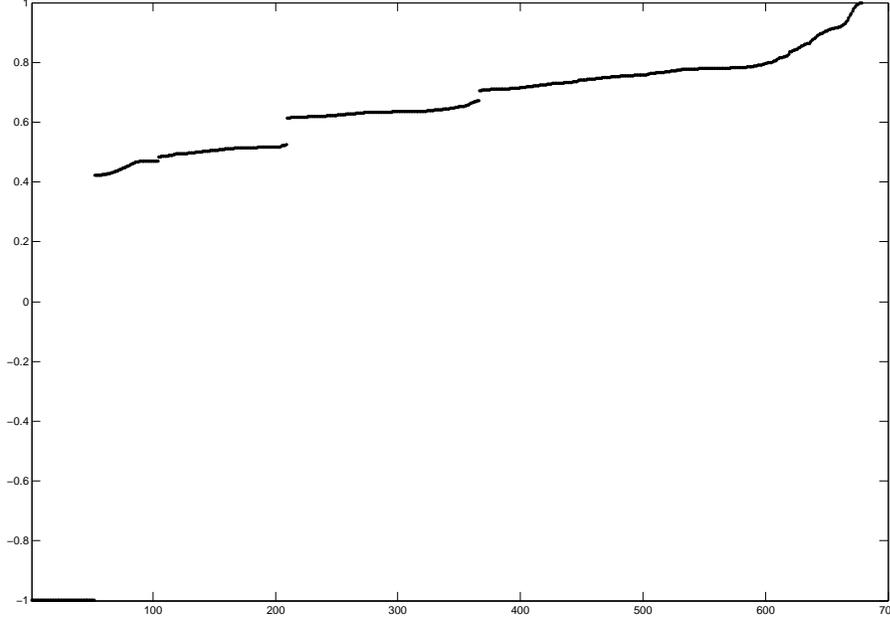


FIG. 7.5. Magnitude of the entries in the density matrix for hydrocarbon $C_{52}H_{106}$ chain, with 209 occupied states. White: $< 10^{-8}$; yellow: $10^{-8} - 10^{-6}$; green: $10^{-6} - 10^{-4}$; blue: $10^{-4} - 10^{-2}$; black: $> 10^{-2}$. Note: \mathbf{nz} refers to the number of ‘black’ entries.

can see that for a truncation level of 10^{-8} , the bandwidth of the density matrix is only slightly larger than that of the Hamiltonian. The eigenvalue spectrum of the Hamiltonian, scaled and shifted so that $\sigma(\hat{H}) \subset [-1, 1]$, is shown in Fig. 7.6. One can clearly see a large gap (≈ 1.4) between the 52 low-lying eigenvalues corresponding to the core electrons in the system, as well as the smaller HOMO-LUMO gap (≈ 0.1) separating the 209 occupied states from the virtual ones. It is worth emphasizing that the exponential decay of the density matrix is independent of the size of the system; that is, if the hydrocarbon chain was made arbitrarily long by adding C and H atoms to it, the density matrix would be of course much larger in size but the bandwidth would remain virtually unchanged for the same truncation level, due to the fact that the bandwidth and the HOMO-LUMO gap of the Hamiltonian do not appreciably change as the number of particles increases. It is precisely this independence of the rate of decay (hence, of the bandwidth) on system size that makes $O(n_e)$ approximations possible (and competitive) for large n .

Let us now see how Theorem 7.1 can be used to prove decay bounds on the entries of density matrices. Let H be the discrete Hamiltonian associated with a certain physical system and let μ be the Fermi level of interest for this system. We assume that the spectrum of H has a gap γ around μ , that is, we have $\gamma = \bar{\varepsilon} - \underline{\varepsilon} > 0$, where $\bar{\varepsilon}$ is the smallest eigenvalue of H to the right of μ and $\underline{\varepsilon}$ is the largest eigenvalue of H to the left of μ . In the particular case of the HOMO-LUMO gap, we have $\underline{\varepsilon} = \varepsilon_{n_e}$ and $\bar{\varepsilon} = \varepsilon_{n_e+1}$.

The Fermi–Dirac function can be used to approximate the Heaviside function; the larger is β , the better the approximation. More precisely, the following result is easy to prove (see [77]):

FIG. 7.6. Spectrum of the Hamiltonian for $C_{52}H_{106}$.

PROPOSITION 7.5. Let $\delta > 0$ be given. If β is such that

$$\beta \geq \left(\ln \left(\frac{1-\delta}{\delta} \right) - \ln \left(\frac{\delta}{1-\delta} \right) \right) \frac{1}{\gamma}, \quad (7.6)$$

then $1 - f_{FD}(\underline{\varepsilon}) \leq \delta$ and $f_{FD}(\bar{\varepsilon}) \leq \delta$.

In Fig. 7.7 we show Fermi–Dirac approximations to the Heaviside function (with jump at $\mu = 0$) for different values of γ between 0.1 and 1, where β has been chosen so as to reduce the error in Proposition 7.5 above the value $\delta = 10^{-6}$.

As a consequence of Theorem 7.1 and Proposition 7.5 we have:

COROLLARY 7.6. Let n_b be a fixed positive integer and $n = n_b \cdot n_e$, where the integers n_e form a monotonically increasing sequence. Let $\{H_n\}$ be a sequence of Hermitian $n \times n$ matrices with the following properties:

1. Each H_n has bandwidth m independent of n ;
2. There exist two fixed intervals $I_1 = [-1, a], I_2 = [b, 1] \subset \mathbb{R}$ with $\gamma = b - a > 0$, such that for all $n = n_b \cdot n_e$, I_1 contains the smallest n_e eigenvalues of H_n (counted with their multiplicities) and I_2 contains the remaining $n - n_e$ eigenvalues.

Let P_n denote the $n \times n$ spectral projector onto the subspace spanned by the eigenvectors associated with the n_e smallest eigenvalues of H_n , for each n . Let $\epsilon > 0$ be arbitrary. Then there exist constants $c > 0, \alpha > 0$ independent of n such that

$$|[P_n]_{ij}| \leq \min \left\{ 1, c e^{-\alpha|i-j|} \right\} + \epsilon, \quad \text{for all } i \neq j. \quad (7.7)$$

The constants c and α can be computed from (7.2) and (7.3), where χ is chosen in the interval $(1, \bar{\chi})$, with $\bar{\chi}$ given by (7.4) and β such that (7.6) holds.

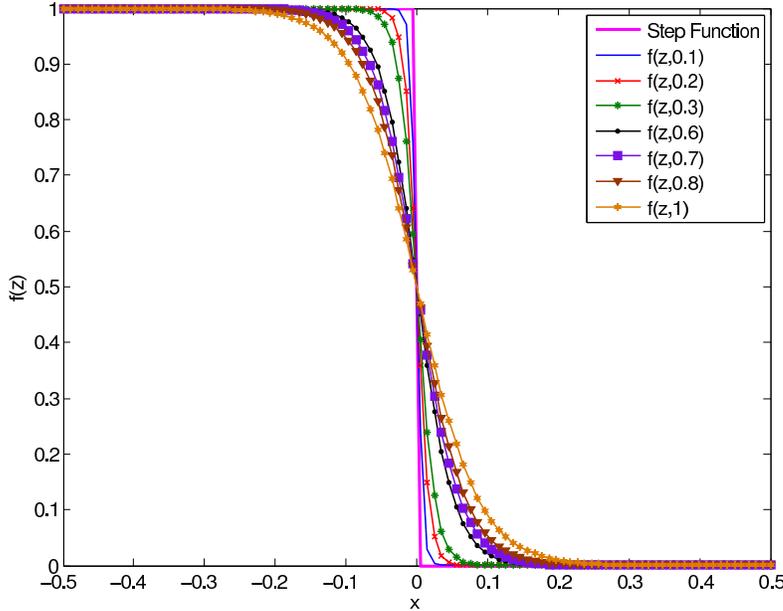


FIG. 7.7. Approximations of Heaviside function by Fermi-Dirac function ($\mu = 0$) for different values of γ and $\delta = 10^{-6}$.

Corollary 7.6 allows us to determine *a priori* a bandwidth \hat{m} independent of n outside of which the entries of P_n are smaller than a prescribed tolerance $\tau > 0$. Observe that it is not possible to incorporate ϵ in the exponential bound, but, at least in principle, one may always choose ϵ smaller than a certain threshold. For instance, one may take $\epsilon < \tau/2$ and define \hat{m} as the smallest integer value of m such that the relation $ce^{-\alpha m} \leq \tau/2$ holds.

In the case of Hamiltonians with a general sparsity pattern one may apply Theorem 7.4 to obtain a more general version of Corollary 7.6. If the fixed bandwidth hypothesis is removed, the following bound holds:

$$|[P_n]_{ij}| \leq \min \left\{ 1, ce^{-\theta d_n(i,j)} \right\} + \epsilon, \quad \text{for all } i \neq j, \quad (7.8)$$

with $\theta = \ln(1/\chi)$. Once again, for the result to be meaningful some restriction on the sparsity patterns, like the uniformly bounded maximum degree assumption already discussed, must be imposed.

7.3. Proof of decay bounds. Theorem 7.1 is a consequence of results proved in [9] (Thm. 2.2) and [77] (Thm. 2.2); its proof relies on a fundamental result in polynomial approximation theory known as Bernstein's Theorem [71]. Given a function f continuous on $[-1, 1]$ and a positive integer k , the k th best approximation error for f is the quantity

$$E_k(f) = \inf \left\{ \max_{-1 \leq x \leq 1} |f(x) - p(x)| : p \in P_k \right\},$$

where P_k is the set of all polynomials with real coefficients and degree less than or equal to k . Bernstein's theorem describes the asymptotic behavior of the best approximation error for a function f analytic on a domain containing the interval $[-1, 1]$.

Consider the family of ellipses in the complex plane with foci in -1 and 1 . As already mentioned, an ellipse in this family is completely determined by the sum $\chi > 1$ of its half-axes and will be denoted as \mathcal{E}_χ .

THEOREM 7.7 (Bernstein). *Let the function f be analytic in the interior of the ellipse \mathcal{E}_χ and continuous on \mathcal{E}_χ . Moreover, assume that $f(z)$ is real for real z . Then*

$$E_k(f) \leq \frac{2M(\chi)}{\chi^k(\chi - 1)},$$

where $M(\chi) = \max_{z \in \mathcal{E}_\chi} |f(z)|$.

Let us now consider the special case where $f(z) := f_{FD}(z) = 1/(1 + e^{\beta(z-\mu)})$ is the Fermi–Dirac function of parameters β and μ . Observe that $f_{FD}(z)$ has poles in $\mu \pm i\frac{\pi}{\beta}$, so the admissible values for χ with respect to $f_{FD}(z)$ are given by $1 < \chi < \bar{\chi}$, where the parameter $\bar{\chi}$ is such that $\mu \pm i\frac{\pi}{\beta} \in \mathcal{E}_{\bar{\chi}}$ (the *regularity ellipse* for $f = f_{FD}$). Explicit computation of $\bar{\chi}$ yields (7.4).

Now, let H_n be as in Theorem 7.1. We have

$$\|f_{FD}(H_n) - p_k(H_n)\|_2 = \max_{x \in \sigma(H_n)} |f_{FD}(x) - p_k(x)| \leq E_k(f_{FD}) \leq cq^{k+1},$$

where $c = 2\chi M(\chi)/(\chi - 1)$ and $q = 1/\chi$. The Bernstein approximation of degree k gives a bound on $|[f_{FD}(H_n)]_{ij}|$ when $[p_k(H_n)]_{ij} = 0$, that is, when $|i - j| > mk$. We may also assume $|i - j| \leq m(k + 1)$. Therefore, we have

$$|[f_{FD}(H_n)]_{ij}| \leq ce^{m(k+1)\ln(q^{1/m})} = ce^{-\alpha m(k+1)} \leq ce^{-\alpha|i-j|}.$$

As for Theorem 7.4, note that for a general sparsity pattern we have $[(H_n)^k]_{ij} = 0$, and therefore $[p_k(H_n)]_{ij} = 0$, whenever $d_n(i, j) > k$. Writing $d_n(i, j) = k + 1$ we obtain

$$|[f_{FD}(H_n)]_{ij}| \leq c(1/\chi)^{k+1} = ce^{-\theta d_n(ij)}.$$

Let us now prove Corollary 7.6. Assume that β satisfies the inequality (7.6) for given values of ϵ and γ . If we approximate the Heaviside function with step at μ by means of the Fermi–Dirac function $f_{FD}(x) = 1/(1 + e^{\beta(x-\mu)})$, the pointwise approximation error is given by $g(x) = e^{\beta(x-\mu)}/(1 + e^{\beta(x-\mu)})$ for $x < \mu$ and by $f_{FD}(x)$ for $x > \mu$. It is easily seen that $g(x)$ is a monotonically increasing function, whereas f_{FD} is monotonically decreasing. As a consequence, for each Hamiltonian H_n we have that $1 - f_{FD}(\lambda) \leq \epsilon$ for all eigenvalues $\lambda \in I_1$ and $f_{FD}(\lambda) \leq \epsilon$ for all $\lambda \in I_2$. In other words, the pointwise approximation error on the spectrum of H_n is always bounded by ϵ . Therefore, we have

$$\|[P_n - f_{FD}(H_n)]_{ij}\| \leq \|P_n - f_{FD}(H_n)\|_2 \leq \epsilon.$$

We may then conclude using Theorem 7.1:

$$|[P_n]_{ij}| \leq |[f_{FD}(H_n)]_{ij}| + \epsilon \leq ce^{-\alpha|i-j|} + \epsilon.$$

Finally, recall that in an orthogonal projector no entry can exceed unity in absolute value. With this in mind, (7.7) and (7.8) readily follow.

7.4. Additional bounds. Theorems 7.1 and 7.4 rely on Bernstein's result on best polynomial approximation. Following the same argument, one may derive decay bounds for the density matrix from any other estimate on the best polynomial approximation error for classes of functions that include the Fermi–Dirac function. For instance, consider the following result of Achieser (see [71, Thm. 78], and [1]):

THEOREM 7.8. *Let the function f be analytic in the interior of the ellipse \mathcal{E}_χ . Suppose that $|\Re(f(z))| < 1$ holds in \mathcal{E}_χ and that $f(z)$ is real for real z . Then the following bound holds:*

$$E_n(f) \leq \frac{4}{\pi} \sum_{\nu=0}^{\infty} \frac{(-1)^\nu}{(2\nu+1) \cosh((2\nu+1)(n+1) \ln \chi)}. \quad (7.9)$$

The series in (7.9) converges quite fast; therefore, it suffices to compute a few terms explicitly to obtain a good approximation of the bound. A rough estimate shows that, in order to approximate the right hand side of (7.9) within a tolerance τ , one may truncate the series after n_0 terms, where $r^{n_0} < \tau(1-r)$ and $r = \chi^{-\frac{n+1}{2}}$.

Observe that, as in Bernstein's results, there is again a degree of arbitrariness in the choice of χ . However, the admissible range for χ is smaller here because of the hypothesis $|\Re(f(z))| < 1$.

The resulting matrix decay bounds have the form

$$|[f_{FD}(H_n)]_{ij}| \leq \frac{4}{\pi} \sum_{\nu=0}^{\infty} \frac{(-1)^\nu}{(2\nu+1) \cosh((2\nu+1)(d(i,j)+1) \ln \chi)} \quad (7.10)$$

for the case of general sparsity patterns. While these bounds are less transparent than those derived from Bernstein's Theorem, they are computable. We found that the bounds (7.10) improve on (7.1) for entries close to the main diagonal, but do not seem to have a better asymptotic behavior. A possibility would be to combine the two bounds by taking the smaller between the two values.

So far we have only considered bounds based on best approximation of analytic functions defined on a single interval. In [45], Hasson has obtained an interesting result on polynomial approximation of a step function defined on the union of two symmetric intervals. Let $a, b \in \mathbb{R}$ with $0 < a < b$ and let $\text{sgn}(x)$ be the sign function defined on $[-b-a] \cup [a, b]$, i.e., $\text{sgn}(x) = -1$ on $[-b, -a]$ and $\text{sgn}(x) = 1$ on $[a, b]$. Notice that the sign function is closely related to the Heaviside function $h(x)$, since we have $h(x) = \frac{1}{2}(1 + \text{sgn}(x))$.

PROPOSITION 7.9. *There exists a positive constant K such that*

$$E_n(\text{sgn}; [-b, -a] \cup [a, b]) \leq K \frac{\left(\sqrt{\frac{b-a}{b+a}}\right)^n}{\sqrt{n}}. \quad (7.11)$$

Given a sequence of Hamiltonians $\{H_n\}$ with gapped spectra, one may choose a and b and shift H_n , if necessary, so that the spectrum of each H_n is contained in $[-b, -a] \cup [a, b]$ and the eigenvalues corresponding to occupied states belong to $[-b, -a]$. Then we obtain the following decay bound for the density matrix:

$$|[P_n]_{ij}| \leq K \frac{e^{-\xi d(i,j)}}{2\sqrt{d(i,j)}}, \quad \text{where } \xi = \frac{1}{2} \ln \frac{b+a}{b-a}. \quad (7.12)$$

Under the bounded maximal degree condition, the rate of decay is independent of n .

A few remarks on (7.12) are in order:

- Since (7.12) relies directly on a polynomial approximation of the step function, we do not need here the extra term ϵ found in (7.8).
- Unfortunately, it is not possible to assess whether (7.12) may be useful in practice without an explicit formula – or at least an estimate – for the constant K . The asymptotic decay rate, however, is faster than exponential and indeed faster than for other bounds; a comparison is shown in Fig. 7.8 (top). Notice that this logarithmic plot is only meant to show the slope of the bound (which is computed for $K = 1$).
- A disadvantage of (7.12) is the requirement that the intervals containing the spectra $\sigma(H_n)$ should be symmetric with respect to 0. Of course one may always choose a and b so that this hypothesis is satisfied, but the quality of the decay bound deteriorates if b (or $-b$) is not close to the maximum (resp., minimum) eigenvalue; see Fig. 7.8 (bottom). The blue curve shows the slope of the decay bound for $a = 0.25$ and $b = 1$, in a logarithmic scale. In green we display the behavior of the first row of the density matrix associated with a tridiagonal 100×100 matrix with spectrum in $[-1, -0.25] \cup [0.25, 1]$. The red plot refers to the first row of the density matrix associated with a matrix with spectrum in $[-0.4375, -0.25] \cup [0.25, 1]$. The first matrix is clearly better approximated by the decay bound than the second one.

As one can see from the two plots in Fig. 7.8, even for $c = K = 1$ both types of decay bounds are rather conservative, and estimating the truncation bandwidth \bar{m} needed to achieve a prescribed error from these bounds would lead to an overly large band. Hence, the bounds may not be very useful in practice. For further discussion of these issues, see section 7.8.

7.5. Further results. Let us assume again that we have a sequence $\{H_n\}$ of Hermitian $n \times n$ Hamiltonians (with $n = n_b \cdot n_e$, n_b fixed, $n_e \rightarrow \infty$) such that

- The matrices H_n are banded with uniformly bounded bandwidth, or sparse with graphs having uniformly bounded maximum degree;
- the spectra $\sigma(H_n)$ are uniformly bounded;
- the sequence $\{H_n\}$ has a “stable” spectral gap, i.e., there exist real numbers $g_1 < g_2$ such that $[g_1, g_2] \cap \sigma(H_n) = \emptyset$ for sufficiently large n .

In this subsection we let

- $\mu := (g_2 + g_1)/2$ (Fermi level),
- $\gamma := g_2 - \mu = \mu - g_1$ (absolute spectral gap).

Note that because of the uniformly bounded spectra assumption, the absolute spectral gap is within a constant of the relative gap previously defined.

Chui and Hasson study in [19] the asymptotic behavior of the error of best polynomial approximation for a sufficiently smooth function f defined on the set $I = [-b, -a] \cup [a, b]$, with $0 < a < b$. Denote as $\mathcal{C}(I)$ the space of real-valued continuous functions on I , with the uniform convergence norm. Then we have (see [19, Thm. 1] and [64]):

THEOREM 7.10. *Let $f \in \mathcal{C}(I)$ be such that $f|_{[-b, -a]}$ is the restriction of a function f_1 analytic on the left half plane $\Re(z) < 0$ and $f|_{[a, b]}$ is the restriction of a function f_2 analytic on the right half plane $\Re(z) > 0$. Then*

$$\limsup_{n \rightarrow \infty} [E_n(f, I)]^{1/n} \leq \sqrt{\frac{b-a}{b+a}},$$

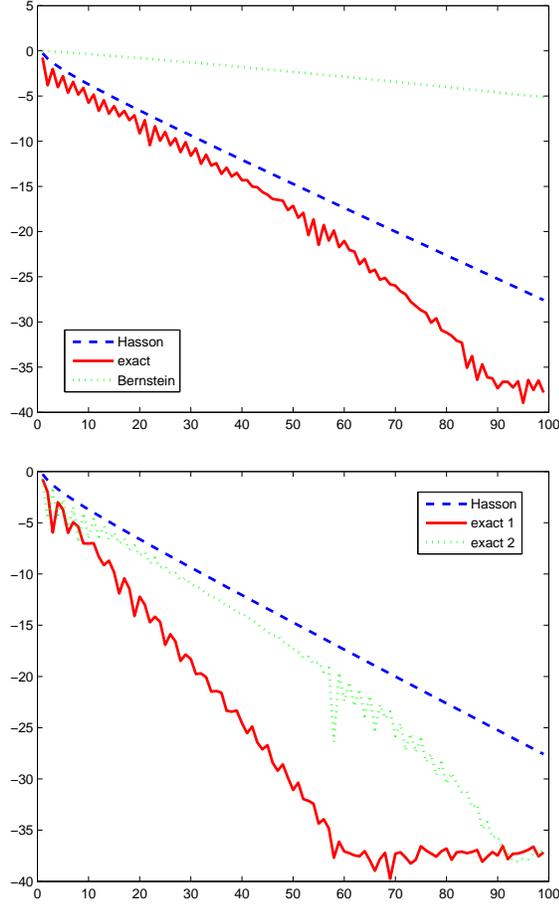


FIG. 7.8. Top: logarithmic plot of Hasson (blue) and Bernstein-type (green) decay bounds, for a 100×100 tridiagonal matrix with spectrum in $[-1, -0.25] \cup [0.25, 1]$. The first row of the “exact” density matrix is plotted in red. Bottom: logarithmic plot of Hasson decay bounds (blue) and first rows of density matrices associated with matrices with different eigenvalue distribution (red and green).

where $E_n(f, I)$ is the error of best polynomial approximation for f on I .

The authors of [19] observe that the above result cannot be obtained by extending $f(x)$ to a continuous function on $[-b, b]$ and applying known bounds for polynomial approximation over a single interval. Theorem 7.10 looks potentially useful for our purposes, except that it provides an asymptotic result, rather than an explicit bound for each value of n . Therefore, we need to reformulate the argument in [19]. To this end, we prove a variant of Bernstein’s Theorem (cf. Theorem 7.3) adapted to our goals. Instead of working on the interval $[-1, 1]$ we want to bound the approximation error on the interval $[a^2, b^2]$.

THEOREM 7.11. *Let $f \in \mathcal{C}([a^2, b^2])$ be the restriction of a function f analytic in the interior of the ellipse \mathcal{E}_{a^2, b^2} with foci in a^2, b^2 and a vertex at the origin. Then*

there exists a constant K such that

$$E_n(f, [a^2, b^2]) \leq K \left(\frac{1}{\xi} \right)^n,$$

where

$$1 < \xi < \xi_{\max} := \frac{a+b}{a-b}.$$

Proof. The proof closely parallels the argument given in [71] for the proof of Theorem 7.3. First of all, observe that the ellipse \mathcal{E}_χ in Bernstein's Theorem has foci in ± 1 and vertices in $\pm(\chi+1/\chi)/2$ and $\pm(\chi-1/\chi)/2$. The parameter χ is the sum of the lengths of the semi-axes. Similarly, the ellipse \mathcal{E}_{a^2, b^2} has foci in a^2, b^2 and vertices in $0, a^2+b^2$ and $(a^2+b^2)/2 \pm iab$. Also observe that ξ_{\max} is the sum of the lengths of the semi-axes of \mathcal{E}_{a^2, b^2} , normalized w.r.t. the semifocal length, so that it plays exactly the same role as χ for \mathcal{E}_χ . Now we look for a conformal map that sends an annulus in the complex plane to the ellipse where f is analytic. When this ellipse is \mathcal{E}_χ , a suitable map is $u = c(v) = (v+1/v)/2$, which sends the annulus $\chi^{-1} < |v| < \chi$ to \mathcal{E}_χ . When the desired ellipse has foci in a^2, b^2 , we compose $c(v)$ with the change of variable

$$x = \psi(u) = \left(u + \frac{a^2+b^2}{b^2-a^2} \right) \frac{b^2-a^2}{2},$$

thus obtaining a function that maps the annulus $\mathcal{A} = \{\xi^{-1} < |v| < \xi\}$ to an ellipse $\mathcal{E}_{a^2, b^2, \xi}$ contained in the interior of \mathcal{E}_{a^2, b^2} . Therefore we have that the function

$$f(\psi(c(v))) = f\left(\left[\frac{1}{2} \left(v + \frac{1}{v} \right) + \frac{a^2+b^2}{b^2-a^2} \right] \frac{b^2-a^2}{2} \right)$$

is analytic on \mathcal{A} and continuous on $|v| = \xi$. The proof now proceeds as in the original Bernstein Theorem. The Laurent expansion

$$f(\psi(c(v))) = \sum_{\nu=-\infty}^{+\infty} \alpha_\nu v^\nu$$

converges in \mathcal{A} with $\alpha_{-\nu} = \alpha_\nu$. Moreover, we have the bound

$$|\alpha_\nu| = \left| \frac{1}{2\pi i} \int_{|v|=\xi} \frac{f(\psi(c(v)))}{v^{\nu+1}} dv \right| \leq \frac{M(\xi)}{\xi^\nu},$$

where $M(\xi)$ is the maximum value (in modulus) taken by f on the ellipse $\mathcal{E}_{a^2, b^2, \xi}$.

Now observe that $u = c(v)$ describes the real interval $[-1, 1]$ for $|v| = 1$, so for $u \in [-1, 1]$ we have

$$f(\psi(u)) = \alpha_0 + 2 \sum_{n=1}^{\infty} \alpha_n T_n(u),$$

where $T_\nu(u)$ is the ν -th Chebyshev polynomial. Since $\psi(u)$ is a linear transformation, we have $E_n(f(z), [a^2, b^2]) = E_n(f(u), [-1, 1])$, so from the theory of Chebyshev approximation [71] we obtain

$$E_n(f, [a^2, b^2]) = E_n(f(u), [-1, 1]) \leq 2M(\xi) \sum_{\nu=n+1}^{\infty} \xi^{-\nu} = \frac{2M(\xi)}{\xi-1} \xi^{-n},$$

hence the thesis. Note that the explicit value of K is computable. \square

The following result is based on [19, Thm. 1].

THEOREM 7.12. *Let $f \in \mathcal{C}(I)$ be as in Theorem 7.10. Then there exists $C > 0$ independent of n such that*

$$E_n(f, I) \leq C\xi^{-\frac{n}{2}},$$

where ξ is as in the previous theorem.

Proof. Let P_n and Q_n be polynomials of best uniform approximation of degree n on the interval $[a^2, b^2]$ for the functions $f_2(\sqrt{x})$ and $f_2(\sqrt{x})/\sqrt{x}$, respectively. Then by Corollary 7.11 there are constants K_1 and K_2 such that

$$\max_{x \in [a^2, b^2]} |P_n(x) - f_2(\sqrt{x})| \leq K_1 \xi^{-n} \quad (7.13)$$

and

$$\max_{x \in [a^2, b^2]} |Q_n(x) - f_2(\sqrt{x})/\sqrt{x}| \leq K_2 \xi^{-n}. \quad (7.14)$$

We use the polynomials P_n and Q_n to define a third polynomial $R_{2n+1}(x) := [P_n(x^2) + xQ_n(x^2)]/2$, of degree $\leq 2n+1$, which approximates $f(x)$ on $[a, b]$ and has small norm on $[-b, -a]$. Indeed, from (7.13) and (7.14) we have:

$$\begin{aligned} \max_{x \in [a, b]} |R_{2n+1}(x) - f(x)| &\leq \frac{1}{2} \max_{x \in [a, b]} |P_n(x^2) - f(x)| + \frac{1}{2} \max_{x \in [a, b]} |xQ_n(x^2) - f(x)| \\ &\leq \frac{1}{2} K_1 \xi^{-n} + \frac{1}{2} b K_2 \xi^{-n} = \frac{K_1 + bK_2}{2} \xi^{-n} \end{aligned} \quad (7.15)$$

and

$$\max_{x \in [-b, -a]} |R_{2n+1}(x)| \leq \frac{1}{2} \max_{x \in [a, b]} |P_n(x^2) - f(x) + f(x) - xQ_n(x^2)| \quad (7.16)$$

$$\leq \frac{1}{2} \max_{x \in [a, b]} |P_n(x^2) - f(x)| + \frac{1}{2} \max_{x \in [a, b]} |xQ_n(x^2) - f(x)| \leq \frac{K_1 + bK_2}{2} \xi^{-n}. \quad (7.17)$$

Similarly, we can find another polynomial $S_{2n+1}(x)$ such that

$$\max_{x \in [-b, -a]} |S_{2n+1}(x) - f(x)| \leq \frac{K_3 + bK_4}{2} \xi^{-n} \quad (7.18)$$

and

$$\max_{x \in [a, b]} |S_{2n+1}(x)| \leq \frac{K_3 + bK_4}{2} \xi^{-n}. \quad (7.19)$$

Then, from the inequalities (7.15)-(7.19) we have

$$\begin{aligned} \max_{x \in I} |R_{2n+1}(x) + S_{2n+1}(x)| &\leq \max_{x \in [a, b]} |R_{2n+1}(x) - f(x)| + \max_{x \in [a, b]} |S_{2n+1}(x)| \\ &\quad + \max_{x \in [-b, -a]} |S_{2n+1}(x) - f(x)| + \max_{x \in [-b, -a]} |R_{2n+1}(x)| \\ &\leq (K_1 + K_3 + b(K_2 + K_4)) \xi^{-n}, \end{aligned}$$

and therefore

$$E_n(f, I) \leq \sqrt{\xi} (K_1 + K_3 + b(K_2 + K_4)) \xi^{-\frac{n}{2}}.$$

This completes the proof. \square

In order to obtain bounds on the density matrix, we apply Theorem 7.12 to the step function f defined on I as follows:

$$f(x) = \begin{cases} 1 & \text{for } -b \leq x \leq -a \\ 0 & \text{for } a \leq x \leq b, \end{cases}$$

i.e., f is the restriction of $f_1(z) \equiv 1$ on $[-b, -a]$ and the restriction of $f_2(z) \equiv 0$ on $[a, b]$. Here the polynomial approximation of $f_1(\sqrt{x})$, $f_1(\sqrt{x})/\sqrt{x}$ and $f_2(\sqrt{-x})$ is exact, so we have $K_1 = K_2 = K_3 = 0$. As for K_4 , observe that $|1/\sqrt{z}|$ achieves its maximum on the vertex of $\mathcal{E}_{a^2, b^2, \xi}$ with smallest abscissa; therefore we have

$$K_4 = \frac{2M(\xi)}{\xi - 1},$$

where

$$M(\xi) = \frac{1}{\sqrt{z_0}} \quad \text{with} \quad z_0 = \left[-\frac{1}{2} \left(\xi + \frac{1}{\xi} \right) + \frac{a^2 + b^2}{b^2 - a^2} \right] \frac{b^2 - a^2}{2}.$$

Moreover, we find $R_{2n+1}(x) \equiv 0$ and $S_{2n+1}(x) = (1 + xV_n(x^2))/2$, where $V_n(x)$ is the polynomial of best uniform approximation for $1/\sqrt{x}$ on $[a^2, b^2]$. Thus, we obtain the bound

$$E_n(f, I) \leq C \xi^{-\frac{n}{2}},$$

where C is given by

$$C = \sqrt{\xi} K_4 b.$$

Let us now apply this result to our sequence of Hamiltonians. We will assume that the matrices are shifted so that $\mu = 0$, that is, we replace each H_n by $H_n - \mu I_n$. Under this hypothesis, the natural choice for a is $a = \gamma/2$, whereas b is the smallest number such that $\sigma(H_n) \subset [-b, -a] \cup [a, b]$ for every n .

Using the same argument used in section 7.3 for the derivation of matrix decay bounds (see also [9] and [11]), we can obtain bounds on the off-diagonal entries of $f(H_n)$. If H_n is banded with bandwidth m independent of n , we have

$$|[P_n]_{ij}| = |[f(H_n)]_{ij}| \leq \sqrt{\xi} \frac{2M(\xi)}{\xi - 1} b \xi^{-\frac{|i-j|}{2m}}, \quad (7.20)$$

whereas if H_n has a more general sparsity pattern we obtain

$$|[P_n]_{ij}| = |[f(H_n)]_{ij}| \leq \sqrt{\xi} \frac{2M(\xi)}{\xi - 1} b \xi^{-\frac{n}{2} d_n(i,j)}, \quad (7.21)$$

where $d_n(i, j)$ is the distance between nodes i and j in the graph G_n associated with H_n .

Next, we compare the bounds derived in this section with those for the Fermi-Dirac approximation of the step function obtained in section 7.1, using a suitable

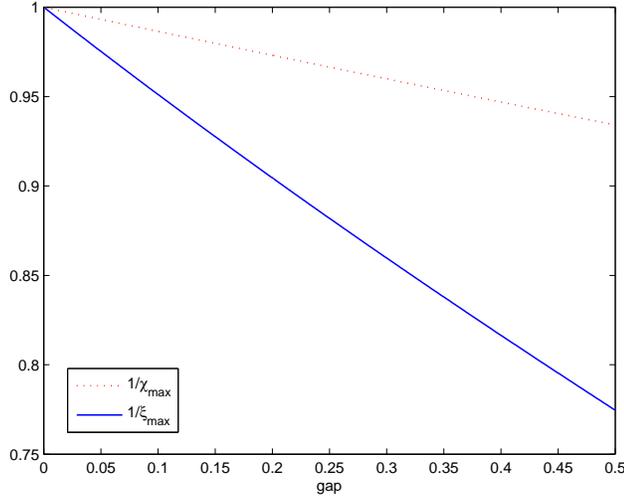


FIG. 7.9. Comparison of parameters $1/\xi_{\max}$ and $1/\chi_{\max}$ for several values of the spectral gap. Here $\epsilon = 10^{-5}$.

choice of the inverse temperature β . Recall that if \mathcal{E}_χ denotes the regularity ellipse for the Fermi–Dirac function, the earlier bounds for the banded case are:

$$|[P_n]_{ij}| \leq \frac{2M(\chi)}{\chi - 1} \left(\frac{1}{\chi} \right)^{\frac{|i-j|}{m}}. \quad (7.22)$$

For ease of computation, we assume in this section that $\mu = 0$ and that the spectrum of each matrix H_n is contained in $[-1, 1]$. As explained in section 7.1, once γ is known, we pick a tolerance ϵ and compute β so that the Fermi–Dirac function provides a uniform approximation of the step function with error $< \epsilon$ outside the gap:

$$\beta = \left(\ln \left(\frac{1-\epsilon}{\epsilon} \right) - \ln \left(\frac{\epsilon}{1-\epsilon} \right) \right) \frac{1}{\gamma}.$$

Then the maximum admissible value of χ , which ensures optimal asymptotic decay in this framework, is

$$\bar{\chi} = \left(\pi + \sqrt{\beta^2 + \pi^2} \right) / \beta.$$

Figures 7.9 and 7.10 compare the values of $1/\xi_{\max}$ and $1/\chi_{\max}$ (which characterize the behavior of the bounds (7.20) and (7.22), respectively). Note that in general we find $1/\xi_{\max} < 1/\chi_{\max}$; this means that the asymptotic decay rate is higher for the bound based on disjoint interval approximation. Moreover, the disjoint interval method directly approximates the step function and therefore does not require to choose a tolerance for “intermediate” approximation. As a result, the bounds based on disjoint interval approximation prescribe a smaller truncation bandwidth \bar{m} in the approximation to the spectral projector in order to achieve a given level of error. For instance, in the tridiagonal case ($m = 1$) we observed a factor of three reduction in \bar{m} compared to the previous bounds, independent of the size of the gap.

7.6. Dependence of the rate of decay on the spectral gap. As already mentioned in section 2, the functional dependence of the the inverse correlation length (governing the rate of decay in the density matrix) on the spectral gap has been the

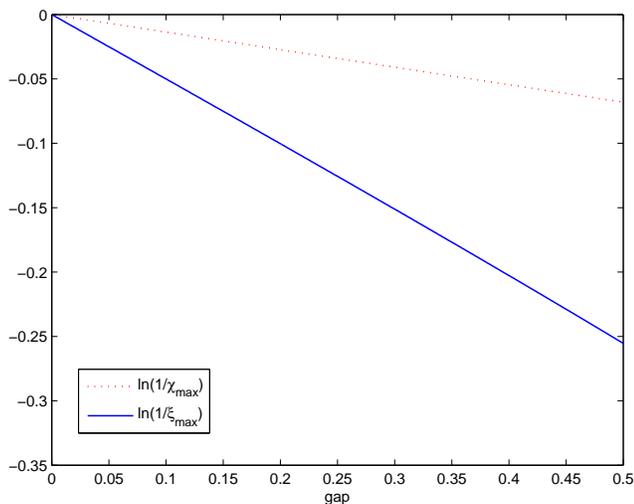


FIG. 7.10. *Logarithmic plot of parameters $1/\xi_{\max}$ and $1/\chi_{\max}$ w.r.t. several values of the spectral gap. Here $\epsilon = 10^{-5}$.*

subject of some discussion. In this section we address this problem by studying how the decay described by the bounds (7.20) and (7.21) behaves asymptotically with respect to γ or, equivalently, on a (see the notation introduced in the previous section). Let us rewrite (7.20) in the form

$$|[P_n]_{ij}| \leq C e^{-\alpha|i-j|/m},$$

where

$$\alpha = \frac{1}{2} \ln \xi = \frac{1}{2} \ln \left(\frac{a+b}{b-a} \right).$$

For a fixed m , the decay behavior is essentially described by the parameter α . Let us assume for simplicity of notation that $b = 1$, so that the spectral gap is normalized and the expression for α becomes

$$\alpha = \frac{1}{2} \ln \left(\frac{1+a}{1-a} \right).$$

The Taylor expansion of α for a small yields

$$\alpha = a + \frac{a^3}{3} + o(a^3).$$

Therefore, for small values of γ , the decay behavior is described at first order by the gap itself, rather than by a more complicated function of γ . Since we are working with upper bounds, this result is consistent with the behavior observed in the literature, where the actual decay is said to be determined sometimes by γ and sometimes by $\gamma^{1/2}$ (which would cause faster decay). Given that our theory is completely general, it is not surprising that we obtain the more conservative estimate.

7.7. Other approaches. Decay bounds on the entries of spectral projectors can also be obtained from the contour integral representation

$$P_n = \frac{1}{2\pi i} \int_{\Gamma} (zI_n - H_n)^{-1} dz, \quad (7.23)$$

where Γ is a simple closed curve (counterclockwise oriented) in \mathbb{C} surrounding a portion of the real axis containing the eigenvalues of H_n which correspond to the occupied states and only those. Componentwise, (7.23) becomes

$$[P_n]_{ij} = \frac{1}{2\pi i} \int_{\Gamma} [(zI_n - H_n)^{-1}]_{ij} dz, \quad 1 \leq i, j \leq n,$$

from which we obtain

$$|[P_n]_{ij}| \leq \frac{1}{2\pi} \int_{\Gamma} [(zI_n - H_n)^{-1}]_{ij} dz, \quad 1 \leq i, j \leq n.$$

Assume the matrices H_n are banded, with uniformly bounded spectra and bandwidths as $n \rightarrow \infty$. By [23, Prop. 2.3], there exist for all $z \in \Gamma$ constants $c(z) \geq 0$ and $0 < \lambda(z) < 1$ (independent of n) such that

$$|[(zI - A)^{-1}]_{ij}| \leq c(z)[\lambda(z)]^{|i-j|}, \quad (7.24)$$

for all $i, j = 1, \dots, n$. Moreover, c and λ depend continuously on $z \in \Gamma$. Since Γ is compact we can set

$$c = \max_{z \in \Gamma} c(z) \quad \text{and} \quad \lambda = \max_{z \in \Gamma} \lambda(z). \quad (7.25)$$

Now let us assume that the matrices H_n have spectral gaps γ_n satisfying $\inf_n \gamma_n > 0$. It is then clear that c is finite. Also, $\lambda \in (0, 1)$. Hence, we obtain the following bound:

$$|[P_n]_{ij}| \leq \left(c \cdot \frac{\ell(\Gamma)}{2\pi} \right) \lambda^{|i-j|}, \quad (7.26)$$

for all $i, j = 1, \dots, n$, where $\ell(\Gamma)$ denotes the length of Γ . Finally, letting $C = c \cdot \frac{\ell(\Gamma)}{2\pi}$ and $\alpha = -\ln \lambda$ we obtain the exponential decay bounds

$$|[P_n]_{ij}| \leq C \cdot e^{-\alpha|i-j|}, \quad 1 \leq i, j \leq n,$$

with both $C > 0$ and $\alpha > 0$ independent of n . As usual, the bounds can be easily extended to the case of general sparsity patterns. One disadvantage of this approach is that explicit evaluation of the constants C and α is rather complicated.

The integral representation (7.23) is useful not only as a theoretical tool, but also increasingly as a computational tool. Indeed, quadrature rules with suitably chosen nodes $z_1, \dots, z_k \in \Gamma$ can be used to approximate the integral in (7.23), leading to

$$P_n \approx \sum_{i=1}^k w_i (z_i I_n - H_n)^{-1} \quad (7.27)$$

for suitable quadrature weights w_1, \dots, w_k . Note that this amounts to a rational approximation of $P_n = h(H_n)$. In practice, using the trapezium rule with a small number of nodes suffices to achieve high accuracy, due to the exponential convergence of this quadrature rule for analytic functions [22]. Note that if P_n is real then it is sufficient to use just the z_i in the upper half-plane and then take the real part of the result [49, page 307]. If the spectral gap γ_n for H_n is not too small, all the resolvents $(z_i I_n - H_n)^{-1}$ decay rapidly away from the main diagonal, with exponential rate independent of n_e . Hence, $O(n_e)$ approximation is possible, at least in principle.

Rational approximations of the type (7.27) are especially useful in those situations where only selected entries of P_n are required. Then only the corresponding entries of the resolvents $(z_i I_n - H_n)^{-1}$ need to be computed. For instance, in some cases only the diagonal entries of P_n are needed [86]. In others, only entries in positions corresponding to the non-zero entries in the Hamiltonian H_n must be computed; this is the case, for instance, when computing the electronic energy $\langle E \rangle = \text{Tr}(P_n H_n)$. Computing selected entries of a resolvent is not an easy problem. However, progress has been made on this front in several recent papers; see, e.g., [59, 61, 62, 87, 94].

7.8. Practical considerations. In the preceding sections we have rigorously established exponential decay bounds for zero-temperature density matrices corresponding to finite-range Hamiltonians with non-vanishing spectral gap (‘insulators’), as well as for density matrices corresponding to arbitrary finite-range Hamiltonians at positive electronic temperatures. Our results are very general and apply to a wide variety of physical systems and discretizations. Hence, a mathematical justification of the physical phenomenon of ‘nearsightedness’ has been obtained, and the possibility of $O(n_e)$ methods firmly established.

On the other hand, the practical usefulness of these bounds is limited. While our results can be used to compute an upper bound on the bandwidth (or sparsity pattern) of the approximation which is independent of the size of the system, the bounds are generally pessimistic and tend to be overly conservative, especially for low or zero temperature cases. This is to be expected, since the bounds were obtained by estimating the degree of a polynomial approximation to the Fermi–Dirac matrix function needed to satisfy a prescribed error tolerance. These bounds tend to be rather pessimistic because they do not take into account the possibility of numerical cancellation when evaluating the matrix polynomial. For instance, the bounds must apply in the worst-case scenario where the Hamiltonian has non-negative entries and the approximating polynomial has nonnegative coefficients. Moreover, the bounds do not take into account the size of the entries in the Hamiltonian, particularly the fact that the nonzeros within the band (or sparsity pattern) are not of uniform size but may be spread out over several orders of magnitude. The situation is not unlike that of the error bounds for the convergence of the conjugate gradient method in terms of conditioning of the coefficient matrix. Hence, the theory developed in this paper is primarily a qualitative one, even if it captures many of the features of actual physical systems (like the asymptotic dependence of the decay rate on the gap size or on the electronic temperature).

A natural question is whether the bounds can be improved to the point where they can be used to obtain practical estimates of the entries in the density matrix. We think that in order to achieve this, additional assumptions on the Hamiltonians would be needed, making the theory less general. In other words, the price we pay for the generality of our theory is a lack of tightness in the bounds. Whether improved bounds would be really useful in practice is not clear, given that most linear scaling codes do not need to know *a priori* the precise rate of decay in the density matrix.⁵

Another practical issue that should be at least briefly mentioned is the fact that our bounds assume knowledge of lower and upper bounds on the spectra of the Hamiltonians H_n , as well as estimates for the size and location of the spectral gap (this is also needed in order to determine the Fermi level μ). These issues have received a

⁵We further observe that in some cases the bounds are known to be sharp; this is the case, for instance, for the decay bounds for the inverses of banded symmetric positive definite matrices, see [23].

great deal of attention in the literature, and here we limit ourselves to observe that $O(n_e)$ procedures exist to obtain sufficiently accurate estimates of these quantities; see, e.g., [37].

8. The vanishing gap case. In this section we discuss the case of a sequence $\{H_n\}$ of bounded, finite range Hamiltonians for which the spectral gap around the Fermi level μ vanishes as $n \rightarrow \infty$. Recall that this means that $\inf_n \gamma_n = 0$, where $\gamma_n := \varepsilon_{n_{e+1}}^{(n)} - \varepsilon_{n_e}^{(n)}$ is the HOMO-LUMO gap for the n -th Hamiltonian; it is assumed here that $\varepsilon_{n_e}^{(n)} < \mu < \varepsilon_{n_{e+1}}^{(n)}$ for all $n = n_b \cdot n_e$. The reciprocal γ_n^{-1} of the gap can be interpreted as the condition number of the problem, so a vanishing spectral gap means that the conditioning deteriorates as $n_e \rightarrow \infty$ and the problem becomes increasingly difficult.

As already mentioned, in the zero-temperature limit our decay bounds blow up, and therefore lose all meaning as $\gamma_n \rightarrow 0$. On the other hand, we know *a priori* that some type of decay should be present, in view of the results in section 5. A general treatment of the vanishing gap case appears to be rather difficult. The main reason is that in the limit as $\beta \rightarrow \infty$ the Fermi–Dirac approximation to the Heaviside function becomes discontinuous, and therefore we can no longer make use of tools from classical approximation theory for analytic functions. Similarly, the representation of the density matrix in terms of contour integrals (see (2.3)) breaks down in the vanishing gap limit, and so do the decay bounds based on resolvent estimates.

Rather than attacking the problem in general, in this section we give a complete analysis of what is perhaps the simplest nontrivial example of a sequence $\{H_n\}$ with vanishing gap. While this is only a special case, this example captures some of the essential features of the ‘metallic’ case, such as the rather slow off-diagonal decay of the entries of the density matrix. We hope that this analysis will shed some light on more general situations.

We begin by considering the infinite tridiagonal Toeplitz matrix

$$H = \begin{pmatrix} 0 & \frac{1}{2} & & & \\ \frac{1}{2} & 0 & \frac{1}{2} & & \\ & \ddots & \ddots & \ddots & \\ & & \frac{1}{2} & 0 & \frac{1}{2} \\ & & & \ddots & \ddots & \ddots \end{pmatrix}, \quad (8.1)$$

which defines a bounded, banded, self-adjoint operator on ℓ^2 . The graph of this matrix is just a (semi-infinite) path. The operator can be interpreted as an averaging operator, or as a centered second-difference operator with a zero Dirichlet condition at one end, shifted and scaled so as to have spectrum contained in $[-1, 1]$.

For n even ($n = 2 \cdot n_e$, with $n_e \in \mathbb{N}$) consider the n -dimensional approximation

$$H_n = \begin{pmatrix} 0 & \frac{1}{2} & & & \\ \frac{1}{2} & 0 & \frac{1}{2} & & \\ & \ddots & \ddots & \ddots & \\ & & \frac{1}{2} & 0 & \frac{1}{2} \\ & & & \frac{1}{2} & 0 \end{pmatrix}. \quad (8.2)$$

This corresponds to truncating the semi-infinite path and imposing zero Dirichlet conditions at both ends. Let now $\{e_1, e_2, \dots\}$ denote the standard basis of ℓ^2 , and let \hat{I}

denote the identity operator restricted to the subspace of ℓ^2 spanned by e_{n+1}, e_{n+2}, \dots . Letting

$$H_{(n)} := \begin{pmatrix} H_n & 0 \\ 0 & \hat{I} \end{pmatrix},$$

the sequence $\{H_{(n)}\}$ is now a sequence of bounded self-adjoint linear operators on ℓ^2 that converges strongly to H . Note that $\sigma(H_n) \subset [-1, 1]$ for all n ; also, $0 \notin \sigma(H_n)$ for all even n . It is easy to see that half of the eigenvalues of H_n lie in $[-1, 0)$ and the other half in $(0, 1]$. We set $\mu = 0$ and we label as ‘occupied’ the states corresponding to negative eigenvalues. The spectral gap of each H_n is then $\varepsilon_{n/2+1}^{(n)} - \varepsilon_{n/2}^{(n)}$.

The eigenvalues and eigenvectors of H_n are known explicitly [24, Lemma 6.1]. Indeed, the eigenvalues, in descending order, are given by $\varepsilon_k^{(n)} = \cos\left(\frac{k\pi}{n+1}\right)$ (with $1 \leq k \leq n$) and the corresponding normalized eigenvectors are given by $v_k^{(n)} = (v_k^{(n)}(j))$ with entries

$$v_k^{(n)}(j) = \sqrt{\frac{2}{n+1}} \sin\left(\frac{jk\pi}{n+1}\right), \quad 1 \leq j \leq n.$$

Note that the eigenvalues are symmetric with respect to the origin, and that the spectral gap at 0 vanishes, since $\varepsilon_{n/2+1}^{(n)} = -\varepsilon_{n/2}^{(n)} \rightarrow 0$ as $n \rightarrow \infty$. We also point to the well known fact that the eigenvectors of this operator are strongly delocalized. Nevertheless, as we will see, some localization (decay) is present in the density matrix, owing to cancellation (i.e., destructive interference).

Now let P_n be the zero-temperature density matrix associated with H_n , i.e., the spectral projector onto the subspace of \mathbb{C}^n spanned by the eigenvectors of H_n associated with the lowest n_e eigenvalues (the occupied subspace). We extend P_n to a projector acting on ℓ^2 by embedding P_n into an infinite matrix $P_{(n)}$ as follows:

$$P_{(n)} := \begin{pmatrix} P_n & 0 \\ 0 & 0 \end{pmatrix}.$$

Note that $P_{(n)}$ is just the orthogonal projector onto the subspace of ℓ^2 spanned by the eigenvectors of $H_{(n)}$ associated with eigenvalues in the interval $[-1, 0)$. Moreover, $\text{Tr}(P_{(n)}) = \text{Tr}(P_n) = \text{rank}(P_n) = \frac{n}{2} = n_e$. The limiting behavior of the sequence $\{P_{(n)}\}$ (hence, of $\{P_n\}$) is completely described by the following result.

THEOREM 8.1. *Let H , H_n , and $P_{(n)}$ be as described above. Then*

- (i) *H has purely absolutely continuous spectrum,⁶ given by the interval $[-1, 1]$. In particular, H has no eigenvalues.*
- (ii) *The union of the spectra of the n -dimensional sections H_n of H is everywhere dense in $\sigma(H) = [-1, 1]$. In other words, every point in $[-1, 1]$ is the limit of a sequence of the form $\{\varepsilon_k^{(n)}\}$ for $n \rightarrow \infty$, where $\varepsilon_k^{(n)} \in \sigma(H_n)$ and $k = k(n)$.*
- (iii) *The sequence $\{H_n\}$ has vanishing gap: $\inf_n \gamma_n = 0$.*
- (iv) *The spectral projectors $P_{(n)}$ converge strongly to $P = h(H)$ where $h(x) = \chi_{[-1, 0)}(x)$, the characteristic function of the interval $[-1, 0)$.*
- (v) *P is the orthogonal projector onto an infinite-dimensional subspace of ℓ^2 .*

⁶For the definition of absolutely continuous spectrum, see [78].

Proof. Statements (i)-(ii) are straightforward consequences of classical results on the asymptotic eigenvalue distribution of Toeplitz matrices, while (iv)-(v) follow from general results in spectral theory. Statement (iii) was already noted (the eigenvalues of H_n are explicitly known) and it also follows from (i)-(ii). More in detail, statement (i) is a special case of Rosenblum's Theorem on the spectra of infinite band Toeplitz matrices: see [79] or [13, Thm. 1.31]. For the fact that the spectrum of H coincides with the interval $[-1, 1]$ and that the finite section eigenvalues $\varepsilon_k^{(n)}$ are dense in $\sigma(H) = [-1, 1]$ (statement (ii)), see the paper by Hartman and Wintner [44] or the book by Grenander and Szegö [42, Chapter 5]. Statement (iv) can be proved as follows. For a linear operator A on ℓ^2 , write $R_\lambda(A) = (A - \lambda I)^{-1}$, with $\lambda \notin \sigma(A)$. A sequence $\{A_n\}$ of self-adjoint (Hermitian) operators is said to converge in the *strong resolvent sense* to A if $R_\lambda(A_n) \rightarrow R_\lambda(A)$ strongly for all $\lambda \in \mathbb{C}$ with $\Re(\lambda) \neq 0$, that is:

$$\lim_{n \rightarrow \infty} \|R_\lambda(A_n)x - R_\lambda(A)x\| = 0 \quad \text{for all } x \in \ell^2.$$

It is easy to check, using for instance the results in [14, Chapter 2], that the sequence $\{H_n\}$ converges in the strong resolvent sense to H . Statement (iv) (as well as (ii)) now follows from [78, Thm. VIII.24]. The fact (v) that $P = h(H)$ is an orthogonal projector onto an infinite-dimensional subspace of ℓ^2 is an immediate consequence of the spectral theorem for self-adjoint operators in Hilbert space; see, e.g., [78, Chapter VII] or [85, Chapter 12]. \square

The foregoing result implies that the Toeplitz matrix sequence $\{H_n\}$ given by (8.2) exhibits some of the key features of the discrete Hamiltonians describing conductors, in particular the vanishing gap property and the fact that the eigenvalues tend to fill the entire energy spectrum. The sequence $\{H_n\}$ can be thought of as a 1D 'toy model' that can be solved analytically to gain some insight into the decay properties of the density matrix for metallic systems. Indeed, from the knowledge of the eigenvectors of H_n we can write down the spectral projector corresponding to the lowest $n_e = n/2$ eigenvalues explicitly. Recalling that the eigenvalues $\varepsilon_k^{(n)}$ are given in descending order, it is convenient to compute P_n as the projector onto the orthogonal complement of the subspace spanned by the eigenvectors corresponding to the $n/2$ largest eigenvalues:

$$P_n = I_n - \sum_{k=1}^{n_e} v_k^{(n)} (v_k^{(n)})^T.$$

The (i, j) entry of P_n is therefore given by

$$[P_n]_{ij} = e_i^T P_n e_j = \delta_{ij} - \frac{2}{n+1} \sum_{k=1}^{n_e} \sin\left(\frac{ik\pi}{n+1}\right) \sin\left(\frac{jk\pi}{n+1}\right).$$

For $i = j$, we find

$$[P_n]_{ii} = 1 - \frac{2}{n+1} \sum_{k=1}^{n_e} \sin^2\left(\frac{ik\pi}{n+1}\right) = \frac{1}{2}, \quad \text{for all } i = 1, \dots, n \text{ and for all } n. \quad (8.3)$$

Hence, for this system the charge density P_{ii} is constant and the system essentially behaves like a non-interacting electron gas, see for example [34]. We note in passing that this example shows that the bound (5.1) is sharp, since equality is attained for this particular projector. Moreover, the trigonometric identity

$$\sin \theta \sin \phi = -\frac{1}{2} [\cos(\theta + \phi) - \cos(\theta - \phi)] \quad (8.4)$$

implies for all $i, j = 1, \dots, n$:

$$[P_n]_{ij} = \frac{1}{n+1} \sum_{k=1}^{n_e} \left[\cos\left(\frac{(i+j)k\pi}{n+1}\right) - \cos\left(\frac{(i-j)k\pi}{n+1}\right) \right]. \quad (8.5)$$

From (8.5) it immediately follows, for all i and for all n , that

$$[P_n]_{i,i+2l} = 0, \quad l = 1, 2, \dots \quad (8.6)$$

Since (8.3) and (8.6) hold for all n , they also hold in the limit as $n \rightarrow \infty$. Hence, the strong limit P of the sequence of projectors $\{P_{(n)}\}$ satisfies $P_{ii} = 1/2$ and $P_{i,j} = 0$ for all $j = i + 2l$, where $i, l = 1, 2, \dots$. To determine the remaining off-diagonal entries P_{ij} (with $j \neq i$ and $j \neq i + 2l$) we directly compute the limit of $[P_n]_{ij}$ as $n \rightarrow \infty$, as follows. Observe that using the substitution $x = k/(n+1)$ and taking the limit as $n \rightarrow \infty$ in (8.5) we obtain for all $i \geq 1$ and for all $j \neq i + 2l$ ($l = 0, 1, \dots$):

$$\begin{aligned} P_{ij} &= \int_0^{\frac{1}{2}} \cos[(i+j)\pi x] dx + \int_0^{\frac{1}{2}} \cos[(i-j)\pi x] dx \\ &= \frac{1}{\pi} \left[\frac{(-1)^{i+j}}{i+j} + \frac{(-1)^{i-j+1}}{i-j} \right]. \end{aligned} \quad (8.7)$$

It follows from (8.7) that $|P_{ij}|$ is bounded by a quantity that decays only linearly in the distance from the main diagonal. As a result, $O(n_n)$ approximation of P_n for large n involves a huge prefactor. Therefore, from this very simple example we can gain some insight into the vanishing gap case. The analytical results obtained show that the density matrix can exhibit rather slow decay, confirming the well known fact that $O(n_e)$ approximations pose a formidable challenge in the vanishing gap case.

The 2D case is easily handled as follows. We consider for simplicity the case of a square lattice consisting of n^2 points in the plane. The 2D Hamiltonian is given by

$$H_{n^2} = \frac{1}{2}(H_n \otimes I_n + I_n \otimes H_n),$$

where the scaling factor $\frac{1}{2}$ is needed so as to have $\sigma(H_{n^2}) \subset [-1, 1]$. The eigenvalues and eigenvectors of H_{n^2} can be explicitly written in terms of those of H_n ; see, e.g., [24]. Assuming again that n is even, exactly half of the n^2 eigenvalues of H_{n^2} are negative (counting multiplicities), the other half positive. As before, we are interested in finding the spectral projector associated with the eigenvectors corresponding to negative eigenvalues. Note again that the spectral gap tends to zero as $n \rightarrow \infty$. If P_{n^2} denotes the spectral projector onto the occupied states, it is not difficult to show that

$$P_{n^2} = P_n \otimes (I_n - P_n) + (I_n - P_n) \otimes P_n. \quad (8.8)$$

It follows from (8.8) that the spectral projector P_{n^2} has a natural $n \times n$ block structure, where

- Each diagonal block is equal to $\frac{1}{2}I_n$; note that this gives the correct trace, $\text{Tr}(P_{n^2}) = \frac{n^2}{2}$.
- The (k, l) off-diagonal block Π_{kl} is given by $\Pi_{kl} = [P_n]_{kl}(I_n - 2P_n)$. Hence, each off-diagonal block has a ‘striped’ structure, with the main diagonal as well as the third, fifth, etc. off-diagonal identically zero. Moreover, every block Π_{kl} with $l = k + 2m$ ($m \geq 1$) is zero.

This shows that in the 2D case, the rate of decay in the spectral projector is essentially the same as in the 1D case. The 3D case can be handled in a similar manner, leading to the same conclusion.

We can also use this example to estimate the rate of decay in the density matrix for the case of vanishing gap but positive electronic temperature $T > 0$. Recalling that the density matrix in this case is given by the Fermi–Dirac function with parameters $\beta = 1/(\kappa T)$ and $\mu = 0$, we have in the 1D case

$$P_{ij} = \frac{2}{n+1} \sum_{k=1}^n \frac{\sin\left(\frac{ik\pi}{n+1}\right) \sin\left(\frac{jk\pi}{n+1}\right)}{1 + \exp\left[\beta \cos\left(\frac{k\pi}{n+1}\right)\right]}. \quad (8.9)$$

Making use again of the trigonometric identity (8.4) and using the same substitution $x = k/(n+1)$, we can reduce the computation of the density matrix element P_{ij} for $n \rightarrow \infty$ to the evaluation of the following integral:

$$P_{ij} = \int_0^1 \frac{\cos[(i-j)\pi x] - \cos[(i+j)\pi x]}{1 + \exp(\beta \cos \pi x)} dx. \quad (8.10)$$

Unfortunately, this integral cannot be evaluated explicitly in terms of elementary functions. Note, however, that the integral

$$I_k = \int_0^1 \frac{\cos(k\pi x)}{1 + \exp(\beta \cos \pi x)} dx$$

(where k is an integer) becomes, under the change of variable $\pi x = \arccos t$,

$$I_k = \frac{1}{\pi} \int_0^1 \frac{\cos(k \arccos t)}{1 + e^{\beta t}} \frac{dt}{\sqrt{1-t^2}}.$$

Hence, up to a constant factor, I_k is just the k th coefficient in the Chebyshev expansion of the Fermi–Dirac function $1/(1+e^{\beta t})$. Since the Fermi–Dirac function is analytic on the interior of an ellipse containing the interval $[-1, 1]$ and continuous on the boundary of such an ellipse, it follows from the general theory of Chebyshev approximation that the coefficients I_k decay at least exponentially fast as $k \rightarrow \infty$; see, e.g., [71]. This in turn implies that the entries P_{ij} given by (8.10) decay at least exponentially fast away from the main diagonal, the faster the larger the temperature β^{-1} is. Hence, for this special case we have obtained in a more direct way the exponentially decaying bounds already proved in general in section 7.1. In the present case, however, for any value of β the decay rate of the entries P_{ij} given by (8.10) can be determined to arbitrary accuracy by numerically computing the Chebyshev coefficients of the Fermi–Dirac function.

9. Other applications. In this section we sketch a few possible applications of our decay results to areas other than electronic structure.

9.1. Density matrices for thermal states. In quantum statistical mechanics, the equilibrium density matrix for a system of particles subject to a heat bath at absolute temperature T is defined as

$$P = \frac{e^{-\beta H}}{Z}, \quad \text{where } Z = \text{Tr}(e^{-\beta H}). \quad (9.1)$$

As usual, $\beta = (k_B T)^{-1}$ where k_B denotes the Boltzmann constant; see [75]. The matrix P is the quantum analog of the canonical Gibbs state. The Hamiltonian H is usually assumed to have been shifted so that the smallest eigenvalue is zero [66, page 112]. Note that P as defined in (9.1) is not an orthogonal projector. It is, however, symmetric and positive semidefinite. Normalization by the *partition function* Z ensures that $\sigma(P) \subset [0, 1]$ and that $\text{Tr}(P) = 1$.

It is clear that for increasing temperature, i.e., for $T \rightarrow \infty$ (equivalently, for $\beta \rightarrow 0$) the canonical density matrix P approaches the identity matrix, normalized by the matrix size n . In particular, the off-diagonal entries tend to zero. The physical interpretation of this is that in the limit of large temperatures the system states become totally uncorrelated. For temperatures approaching the absolute zero, on the other hand, the canonical matrix P tends to the orthogonal projector associated with the zero eigenvalue (ground state). In this limit, the correlation between state i and state j is given by the (i, j) entry of the orthogonal projector onto the eigenspace corresponding to the zero eigenvalue, normalized by n .

For finite, positive values of T , the canonical density matrix P is full but decays away from the main diagonal (or, more generally, away from the sparsity pattern of H). The rate of decay depends on β : the smaller it is, the faster the decay. Application of the bounds developed in section 7 to the matrix exponential is straightforward. For instance, the bounds based on Bernstein's Theorem take the form

$$|[e^{-\beta H}]_{ij}| \leq C(\beta) e^{-\alpha d(i,j)}, \quad i \neq j, \quad (9.2)$$

where

$$C(\beta) = \frac{2\chi}{\chi - 1} e^{\beta \|H\|_2 (\kappa_1 - 1)/2} \quad \text{and} \quad \alpha = 2 \ln \chi.$$

In these expressions, $\chi > 1$ and $\kappa_1 > 1$ are the parameters associated with the Bernstein ellipse with foci in -1 and 1 and major semi-axis κ_1 , as described in section 7. Choosing χ large makes the exponential term decay $e^{-\alpha d(i,j)}$ very fast, but causes $C(\beta)$ to grow larger. Clearly, a smaller β makes the upper bound (9.2) smaller. Bounds on the entries of the canonical density matrix P can be obtained dividing through the upper bounds by Z . Techniques for estimating Z can be developed using the techniques described in [40]; see also [8].

Although the bound (9.2) is an exponentially decaying one, it can be shown that the decay in the entries of a banded or sparse matrix is actually *super*-exponential. This can be shown by expanding the exponential in a series of Chebyshev polynomials and using the fact that the coefficients in the expansion, which can be expressed in terms of Bessel functions, decay to zero super-exponentially; see [71] and also [51]. The decay bounds obtained in this way are, however, less transparent and more complicated to evaluate than (9.2).

Finally, exponential decay bounds for spectral projectors and other matrix functions can be used to give a rigorous justification for $O(n)$ algorithms recently developed for disordered systems; see, e.g., [88, 89].

9.2. Quantum information theory. A related area of research where our decay bounds for matrix functions have proven useful is the study of quantum many-body systems in information theory; see, e.g., [20, 21, 27, 90, 91]. In particular, relationships between spectral gaps and rates of decay for functions of finite range Hamiltonians have been established in [20] using the techniques introduced in [9].

Furthermore, the exponential decay of correlations and its relation to the spectral gap have also been studied in [46, 47].

As shown in [21], exponential decay bounds for matrix functions play a crucial role in establishing so-called *area laws* for the *entanglement entropy* of ground states associated with bosonic systems. These area laws essentially state that the entanglement entropy associated with a 3D bosonic lattice is proportional to the surface area, rather than to the volume, of the lattice. Intriguingly, such area laws are analogous to those governing the Beckenstein–Hawking black hole entropy. We refer the interested reader to the recent, comprehensive survey paper [27] for additional information.

9.3. Complex networks. The study of complex networks is an emerging field of science currently undergoing vigorous development. Researchers in this highly interdisciplinary field include mathematicians, computer scientists, physicists, chemists, engineers, neuroscientists, biologists, social scientists, etc. The main mathematical tools used are linear algebra and graph theory, in particular spectral graph theory. Also, statistical mechanics concepts and techniques have been found to be ideally suited to the study of large-scale networks.

In recent years, quantitative methods of network analysis have increasingly made use of matrix functions. This approach has been spearheaded in the works of Ernesto Estrada, Juan Rodríguez-Velázquez, Des Higham and Naomichi Hatano; see, e.g., [28, 29, 30, 31, 33], as well as the recent survey [32] and the references therein. Here the underlying matrices are those usually associated with graphs, such as the adjacency matrix and the graph Laplacian. Functions naturally arising in the context of network analysis include the exponential, the resolvent, and hyperbolic functions.

For example, the exponential of the adjacency matrix A associated with a simple, undirected graph $G = (V, E)$ is used to give natural definitions of important measures associated with nodes in G , such as the *subgraph centrality* associated with node i , defined as $C(i) = [e^A]_{ii}$, and the *communicability* associated with two distinct nodes i and j , defined as $C(i, j) = [e^A]_{ij}$. Other network quantities that can be expressed in terms of the entries in appropriate matrix functions of A include betweenness, returnability, vulnerability, and so forth.

Often (though by no means always), each node in a complex network is connected to only a few other nodes: that is, the degree of each node is small. Hence, the adjacency matrix is highly sparse. Our theory of decay in functions of sparse matrices can then be used to obtain bounds on quantities such as the communicability between two nodes. For example, our results imply that given a sequence of graphs G_n of increasing size n but having maximal degree uniformly bounded with respect to n , the communicability between node i and node j decays exponentially with the distance $d_n(i, j)$ of i and j in G_n , with a decay rate independent of n . The actual rate of decay can be easily estimated in terms of the maximal degree; the smaller this is, the faster the decay.

We refer the reader to [8] for recent work on bounding techniques for matrix functions arising in the analysis of complex networks.

9.4. Non-Hermitian extensions. Although the main focus of the paper has been the study of functions of sparse Hermitian matrices, many of our results can be extended, under appropriate conditions, to non-Hermitian matrices. The generalizations of our decay bounds to *normal* matrices, including for example skew-Hermitian ones, is relatively straightforward; see, e.g., the results in [11] and [77]. Further generalizations to diagonalizable matrices have been given in [11], although the bounds now contain additional terms taking into account the departure from normality. These

bounds may be difficult to use in practice, as knowledge of the eigenvectors or of the field of values of the matrix is needed. Bounds for functions of general sparse matrices can also be obtained using contour integration; see, e.g., [77] and [70]. It is quite possible that these bounds will prove useful in applications involving functions of sparse, non-normal matrices. Examples include functions of digraphs in network analysis, like returnability, or functions of the Hamiltonians occurring in the emerging field of non-Hermitian quantum mechanics; see, respectively, [30] and [6, 7].

10. Conclusions and open problems. In this paper we have developed a general theory of localization for the density matrices associated with certain sequences of banded or sparse discrete Hamiltonians of increasing size. We have obtained, under very general conditions, exponential decay bounds for the off-diagonal entries in zero-temperature density matrices for gapped systems (‘insulators’) and for density matrices associated with systems at positive electronic temperature. Our theory, while purely mathematical, recovers well-known physical phenomena such as the fact that the rate of decay is faster at higher temperatures and for larger gaps. Thus, we have provided a theoretical justification for the development of $O(n_e)$ methods for electronic structure computations. As necessary preliminaries to this theory we have also studied the approximation of rapidly decaying matrices by banded or sparse ones, the effects of transforming an Hamiltonian from a non-orthogonal to an orthogonal basis, and some general properties of orthogonal projectors.

In the case of zero-temperature and vanishing gaps (‘metallic case’), our bounds deteriorate for increasing n_e . In the limit as $n_e \rightarrow \infty$ we no longer have exponentially decaying bounds, which is entirely consistent with the physics. In this case the decay in the spectral projector is only algebraic, and we have exhibited a simple class of model Hamiltonians for which the decay in the corresponding density matrices is only linear in the distance from the main diagonal.

Because of the slow decay, the development of $O(n_e)$ methods in the metallic case at zero temperature is problematic. We refer the reader to [4, 15, 60, 100] for some attempts in this direction, but the problem remains essentially open. In the metallic case it may be preferable to keep P in the factorized form $P = XX^*$, where $X \in \mathbb{C}^{n \times n_e}$ is any matrix whose columns span the occupied subspace, and to seek a maximally localized X . Note that

$$P = XX^* = (XU)(XU)^*$$

for any unitary $n_e \times n_e$ matrix U , so the question is whether the occupied subspace admits a set of basis vectors that can be rotated so as to become as localized as possible. Another possibility is to research the use of rank-structured approximations (such as hierarchical matrix techniques [43]) to the spectral projector. Combinations of tensor product approximations and wavelets may also prove fruitful; we refer here to numerous recent works by W. Hackbusch and collaborators.

Besides the motivating application of electronic structure, our theory is applicable also to other problems where localization plays a prominent role. We hope that this paper will stimulate further research in this fascinating and important area at the crossroads of mathematics, physics, and computing.

Acknowledgements. We would like to thank David Borthwick, Matt Challacombe, Jean-Luc Fattebert and Maxim Olshanskii for useful discussions, and Jacek Jakowski for providing the data for the hydrocarbon chain.

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