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Localized density matrix minimization and linear-scaling algorithms ☆

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ABSTRACT

We propose a convex variational approach to compute localized density matrices for both zero temperature and finite temperature cases, by adding an entry-wise ℓ_1 regularization to the free energy of the quantum system. Based on the fact that the density matrix decays exponentially away from the diagonal for insulating systems or systems at finite temperature, the proposed ℓ_1 regularized variational method provides an effective way to approximate the original quantum system. We provide theoretical analysis of the approximation behavior and also design convergence guaranteed numerical algorithms based on Bregman iteration. More importantly, the ℓ_1 regularized system naturally leads to localized density matrices with banded structure, which enables us to develop approximating algorithms to find the localized density matrices with computation cost linearly dependent on the problem size.

1. Introduction

Efficient calculation of the low-lying spectrum of operators plays a central role in many applications. In particular, in the context of electronic structure theory, given a discretized effective Hamiltonian (such as the current iterate in a self-consistent iteration), the goal is to obtain the density matrix corresponding to the number of electrons. For zero temperature, the density matrix is the projection operator onto the low-lying eigenspace; for finite temperature, the density matrix is given by the Fermi–Dirac function acting on the Hamiltonian [19,28].

In this work, we extend the variational approach for localized density matrix in our previous work [16] to finite temperature, by adding entrywise ℓ_1 penalty to the free energy of the quantum system (which, in the context of density functional theory, corresponds to the linear version of the Mermin functional [22]). We also theoretically show that the proposed localized density matrix approximates, via the Frobenius norm, the true density matrix linearly dependent on the regularization parameter $1/\eta$. In addition, convergence guaranteed numerical algorithms are also designed to solve the proposed problems based on Bregman iteration.

More importantly, this paper focuses on efficient algorithms to minimize the variational problem for localized density matrices both at zero and finite temperature. In particular, we develop linear-scaling algorithms such that the computational

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cost scales linearly with the dimension of the matrix. The key idea is to exploit the decay of the density matrix away from the diagonal. The ℓ_1 regularized localized density matrices enable us to approximate the original variational problem by restricting to banded matrices.

Linear-scaling algorithms have been an important research direction in electronic structure calculation since the 1990s. Closely related to our context is the density matrix minimization (DMM) algorithms, which is first introduced by Li, Nunes, and Vanderbilt [17] and Daw [6], and have been further developed since then, see e.g., the reviews [3,11]. These algorithms are based on the fact that for insulating system or systems at finite temperature, the density matrix decays exponentially away from the diagonal (see e.g., [1,7,15]). Thus, one may require the density matrices to satisfy prescribed sparsity structures, such as banded matrices for 1D problem. As a result, the degree of freedom and the computational cost becomes linearly scaling. Another closely related class of methods is the purification method of density matrix (see e.g., [20,21,23,27] and a review [24]). Unlike the approach of DMM and the method we take in this work, these methods are not variational.

We emphasize a crucial difference between our approach and the previous works: *Our variational problem is still convex even after truncation!* This is in stark contrast to the previous formulations where the convexity is lost by using purification [21] or other techniques to approximate the inverse of density matrix [4]. This loss of convexity often introduces local minimizers to the variational problem and also issues of convergence. We note that even when the ℓ_1 regularization is dropped from our variational problem, it is still convex and different from the standard DMM algorithms. In fact, it would be of interest to explore this convex formulation, which will be considered in our future work.

The rest of the paper is organized as follows. In the next section, we introduce the variational principles for localized density matrices for both zero and finite temperature cases, with their approximation properties. In Section 3, we introduce the Bregman iteration type algorithms to solve these minimization problems. Linear-scaling algorithms are discussed in Section 4. We validate the algorithms through numerical examples in Section 5. Some conclusive remarks are discussed in Section 6.

2. Localized density matrix minimization

In this work, we will consider a family of energy functionals with parameters β and η:

$$\mathscr{E}_{\beta,\eta} = \mathrm{tr}(HP) + \frac{1}{\beta}\,\mathrm{tr}\Big\{P\ln P + (1-P)\ln(1-P)\Big\} + \frac{1}{\eta}|||P|||_1 \tag{1}$$

where $|||\cdot|||$ denotes the entrywise ℓ_1 norm of a matrix and H and P are $n\times n$ symmetric matrices, which are respectively the (discrete) Hamiltonian and density matrix in the context of electronic structure calculation. Here, β is the inverse temperature and η is the parameter for the ℓ_1 regularization. The functional $\mathscr{E}_{\beta,\eta}$ and its terms will be further explained below.

2.1. Background of density matrix minimization

Let us recall the starting point of [16], which is also the base of the functional $E_{\beta,\eta}$, is the convex variational principle for density matrix

$$\begin{aligned}&\min_{P\in\mathbb{R}^{n\times n}} \mathscr{E}_{\infty,\infty}(P) = \min_{P\in\mathbb{R}^{n\times n}} \mathrm{tr}(HP),\\ &\text{s.t.}\quad \mathrm{tr}\,P = N,\ P = P^{\mathrm{T}},\ 0\preceq P\preceq I,\end{aligned} \tag{2}$$

where the notation $A\preceq B$ denotes that $B-A$ is a symmetric positive semi-definite matrix and N denotes the number of electrons. Note that the constraint $0\preceq P\preceq I$ is the convexification of the idempotency constraint $P=P^2$, which provides the same minimizer for non-degenerate problems (see e.g., [16, Proposition 1]). Indeed, denote $\{\lambda_i,\phi_i\}_{i=1}^n$ the eigenvalue and eigenvector pairs of H with the assumption $\lambda_N<\lambda_{N+1}$, the solution of (2) is given by

$$P_{\infty,\infty} = \sum_{i=1}^{N}\phi_i\phi_i^{\mathrm{T}}, \tag{3}$$

the projection operator on the subspace spanned by the first N eigenvectors, which is known as the density matrix in physics terms.

The variational principle (2) corresponds to the physical zero temperature case (and hence the inverse temperature $\beta=\infty$). At zero temperature, the electrons prefer to occupy lowest possible energy states with the restriction of Pauli's exclusion principle that a single state can be occupied by only one electron (we ignore spin degeneracy). Therefore, the density matrix corresponds to the projection operator to the first N eigenvectors. This is no longer the case at finite temperature. For finite temperature, the system minimizes the free energy, which is the sum of the energy and entropy contribution:

$$\begin{aligned}&\min_{P\in\mathbb{R}^{n\times n}} \mathscr{E}_{\beta,\infty}(P) = \min_{P\in\mathbb{R}^{n\times n}} \mathrm{tr}(HP) + \frac{1}{\beta}\,\mathrm{tr}\Big\{P\ln P + (1-P)\ln(1-P)\Big\}\\ &\text{s.t.}\quad \mathrm{tr}\,P = N,\ P = P^{\mathrm{T}},\ 0\preceq P\preceq I,\end{aligned} \tag{4}$$