DEFINING METALLICITY AND MOTT LOCALIZATION IN CORRELATED NANOSCOPIC SYSTEMS*

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Exact ground state properties are presented by combining the exact diagonalization in the Fock space (including the *long-range* Coulomb interaction) with an *ab initio* optimization of the single-particle (Wannier) functions. The quasiparticle mass is almost divergent at the localization threshold, where the particle distribution of the Fermi-Dirac type gets smeared out. The analysis is performed using $1s$-like Gaussian-type orbitals.

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1. Introduction

In spite of the development of the physics of one dimensional systems such as nanotubes, quantum rings and wires, and organic metals, the understanding of these *correlated fermionic systems* is still lacking. This is because in their description the role of the *long-range* Coulomb interaction is crucial, as the charge screening becomes less effective. The exact solutions of the parametrized models with inclusion of intersite interactions [1] prove the existence of the metal–insulator transition for the half-filled-band case, in contradistinction to the corresponding Hubbard-model solution [2], for which the system is insulating even for an arbitrarily small Coulomb repulsion. A separate question concerns the appearance of the Tomonaga–Luttinger liquid behavior [3] in the metallic state, for which some evidence has been gathered [4]. In brief, the delocalization of the states in $d = 1$ provides a crucial case for the analysis of the localization as a quantum phase transition in a rigorous manner. Here we address the question of the metallicity appearance in a correlated nanoscopic atomic chain or ring.

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2. Model Hamiltonian and the optimized ground state energy

We focus here on the so-called *Extended Hubbard Model* with Hamiltonian of the form

$$H = \epsilon_a^{\rm eff} \sum_i n_i + t \sum_{i\sigma} \left(a_{i\sigma}^\dagger a_{i+1} + \text{h.c.} \right) + U \sum_i n_{i\uparrow} n_{i\downarrow} + \frac{1}{2}{\sum_{ij}}' K_{ij} \delta n_i \delta n_j, \quad (1)$$

where $\delta n_i \equiv n_i - 1$, $\epsilon_a^{\rm eff} = \epsilon_a + N^{-1} \sum_{i<j} (2/R_{ij} + K_{ij})$ (Ry) is the effective atomic level defined including the mean-field part of the Coulomb interaction, t is the nearest-neighbor hopping integral, and U, K_{ij} are the intra- and inter-site Coulomb repulsion amplitudes, respectively.

The Hamiltonian (1) is diagonalized in the Fock space with the help of Lanczos technique. As the microscopic parameters $\epsilon_a^{\rm eff}$, t, U, and K_{ij} are calculated numerically in the Gaussian STO-3G basis, the inverse orbital size α of the $1s$-like state is subsequently optimized to obtain the ground state energy E_G. This procedure follows the idea of recently developed method combining first- and second-quantization schemes [5, 6]. We have already shown [6] that such a combined exact diagonalization — *ab initio* study of the one dimensional system leads to the precise values of the localization threshold, the electron-lattice couplings, and the dimerization magnitude. However, the convergence of the results obtained with the Slater-type orbitals is not sufficient to perform the finite-size scaling with the lattice size $N \to \infty$. This is because, when calculating the microscopic parameters in the single-particle (Wannier) basis, one ignores the three- and the four-site interaction terms, that represents an uncontrolled approximation.

In contrast, for the Gaussian-type orbitals, we can treat the three- and four-site terms exactly. Their effects on the convergence of the results for the ground-state energy E_G and the optimal inverse orbital size $\alpha_{\rm min}$ are shown in Fig. 1 for $N = 6 \div 10$ atoms. These results were used to extrapolate the value of the variational parameter $\alpha_{\rm min}$ to larger N to speed up the computations. Fig. 1 illustrates also the Hubbard localization criterion. Namely, for the interatomic distance $a \approx 3a_0$ (a_0 is the Bohr radius) the energy of the metallic state crosses over to that representing the Mott insulating state. The critical value of a is very close to obtained for the $1s$ Slater-type orbitals [6].

3. The quantum critical behavior

The the electron momentum distribution $n_{k\sigma} \equiv \langle a_{k\sigma}^\dagger a_{k\sigma} \rangle$ is shown in Fig. 2 for $N = 6 \div 14$ atoms. The continuous lines represent the formula

$$n_{k\sigma} = \frac{1}{2} + \text{sgn}(k - k_F)[\alpha |k - k_F|^2 + \beta |k - k_F| - \gamma], \quad (2)$$

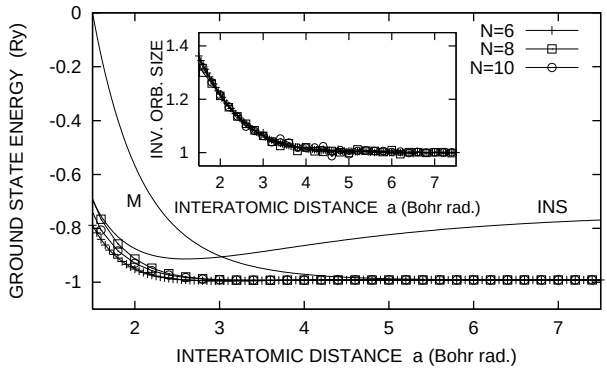

Fig. 1. The ground state energy per atom for the linear chain of $N = 6 \div 10$ with periodic boundary conditions. The Gaussian-type orbitals (STO-3G basis) have been used. The energies of the *purely* metallic (M) and insulating (INS) states are shown for comparison [7]. The *inset* provides the optimal inverse orbital size $\alpha_{\min}$.

with fitted parameters α, β, and γ. We obtain the critical distance in the range $a_{\rm crit} = 3.56 \div 4.18\, a_0$ (*cf.* Table I), at which the interpolated Fermi-ridge discontinuity $\Delta n_F = 2\,\gamma$ disappears, signaling the electron localization. No Luttinger-liquid effects were observed in the system with one-electron per atom. A clear metallic behavior is evidenced by the presence of the Fermi ridge, since the energy-level spread produces remarkably smaller variations of $n_{k\sigma}$ than $\Delta n_{\rm F}$ for $a < a_{\rm crit}$.

TABLE I

The fitted parameters of the critical exponent formula for the quasiparticle mass $m^\star/m_{\rm B} \equiv (\Delta n_{\rm F})^{-1} = A|a - a_{\rm crit}|^{-\gamma}$.

N	A	$a_{\rm crit}$	γ	N	A	$a_{\rm crit}$	γ
8	3.5 ± 2.6	4.18(1)	1.39(14)	12	7.0 ± 1.1	4.030(5)	1.29(10)
10	6.5 ± 2.3	3.900(3)	1.32(7)	14	7.4 ± 0.4	3.56(2)	1.58(7)

With the help of the interpolation formula (2) we can determine the quasiparticle mass enhancement $m^\star/m_{\rm B}$ by equating it with $(\Delta n_{\rm F})^{-1}$. The resulting parameters of fitted critical formula $m^\star/m_{\rm B} = A|a - a_{\rm crit}|^{-\gamma}$ are gathered in Table I. The values of critical exponent are in agreement with those obtained for Slater-type orbitals [6]. It would be very important to test experimentally this result, as it represents the critical behavior of the Fermi discontinuity $\Delta n_{\rm F} \sim |a - a_{\rm crit}|^{\gamma}$ at the localization border reached from the metallic side.

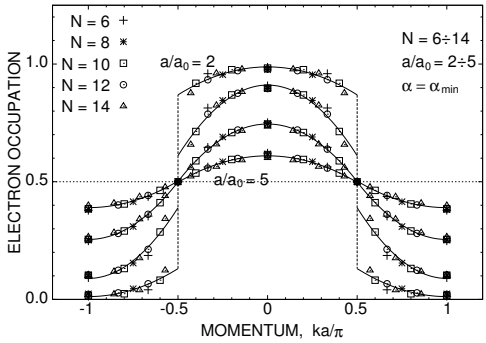

Fig. 2. Momentum distribution $n_{k\sigma}$ for electrons on a linear ring of $N = 6 \div 14$ atoms; the interatomic distance a is specified in units of Bohr radius a_0. The continuous line represents the parabolic interpolation (2).

In summary, we have determined the microscopic criterion for the *cross-over* transition from the nanoscopic metal to the localized spin system in one dimension. The new method of optimizing the single-particle wave functions in the correlated state proves thus executable in an exact treatment of nanoscopic systems for both Slater- and Gaussian-type basis sets. A quantum critical behavior at the metal–insulator boundary is suggested.

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