Hopping and Correlation Effects in Atomic Clusters and Networks

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ABSTRACT

Exact solution for hopping and correlation effects in atomic clusters and mesoscopic/nanoscopic networks is outlined. The program translates the Hamiltonian operator of the cluster written in terms of second-quantized creation and annihilation operators, to sparse matrix of (extremely) large dimension and solves the latter with the help of new compiler termed ABC ("Advanced Basic-C" compiler/convertor/programmer). The ABC creates a stand-alone executable or, if proved necessary, source C-code received from the original program written in a simplified Quick Basic dialect. ABC employes mathematical functions including the complex variables, arbitrary precision floating-point numbers, special functions, standard mathematical routines (mulidimensional integrals, eigenvalues of Hermitian marices, in particular a new algorithm for sparce Hermitian matrices, etc.) and is appropriate to practically all software/hardware environments (Windows, OS/2, Linux and UNIX machines).

Keywords: Atomic cluster, mesoscopic system, persistent current, superconducting network, sparse matrix.

1 FORMULATION OF THE MODEL

The understanding of electron transport and binding energy in strongly correlated electronic systems (high-temperature superconductors; molecular conductors, e.g. carbon nanotubes and fullerenes; mesoscopic structures; biological systems and soft matter; quantum computers) is one of demanding tasks in modern condensed matter physics and microelectronics. The present paper aims at the goal of exact solution of electron transport and correlation in atomic clusters and networks the examples of which are presented in Fig.1.

The simplest Hamiltonian of cluster has form [1]

$$H = -t \sum_{<i,j>\sigma} a_{i\sigma}^+ a_{j\sigma} e^{i\alpha_{ij}} + U \sum_{i=1}^{N} n_{i\uparrow} n_{i\downarrow} + \sum_{i\sigma} V_i n_{i\sigma}$$

$$+ \sum_{<i,j>\sigma} a_{i\sigma}^+ a_{j\sigma} \{ V n_{i\bar\sigma} n_{j\bar\sigma} + W(n_{i\bar\sigma} + n_{j\bar\sigma}) \} e^{i\alpha_{ij}}$$

$$+ \sum_k \Omega_k b_k^+ b_k + V_{ph} \sum_{<i,j>\sigma} \sum_k a_{i\sigma}^+ a_{j\sigma} (b_k^+ + b_k) \quad (1)$$

where $a_{i\sigma}^+ (a_{i\sigma})$ is fermionic second-quantized operator creating (annihilating) electron at atomic site i with spin projection $\sigma = \pm 1/2 = \uparrow\downarrow$, $n_{i\sigma} = a_{i\sigma}^+ a_{i\sigma}$ is the site occupation operator, and b_k the bosonic operator of the deformation field mediating the electron-electron attraction. $e^{\alpha_{ij}}$ is the Peierls substitution phase factor taking into consideration the effect of external magnetic field,

$$\alpha_{ij} = 2\pi \frac{\Phi}{N_s \Phi_0} (j - i) \quad (2)$$

where N_s is the number of sites in z-projected cluster with magnetic field in z direction producing a magnewtic flux Φ. V_i is the (random) potential at site i, and U, V, W and V_{ph} are the coupling constants:

U: Hubbard potential;

V, W: occupation-dependent hopping potentials;

V_{ph}:   electron-phonon coupling strength.

Operators a_m are presented as matrices

$$a_m = (v \otimes)^{m-1} a (\otimes u)^{N-m}, \quad m = 1...N \quad (3)$$

where a, u, v are 2×2 matrices

$$a = \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix}, u = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}, v = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \quad (4)$$

and $\otimes$ is the symbol of Kronecker matrix product. In particular, for $N = 3$ we receive matrices

$$a_1 = \begin{pmatrix} 0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix}, \quad (5)$$

$$a_2 = \begin{pmatrix} 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & -1 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix}, \quad (6)$$

Figure 1: (a)Cubic cluster centered with a vibrating two-level oscillator; (b)Icosahedral cluster; (c)Network of octahedral clusters.

$$a_3 = \begin{pmatrix} 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & -1 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix} \qquad (7)$$

satisfying the commutation relations

$$a_i^+ a_j + a_j a_i^+ = \delta_{ij}. \qquad (8)$$

The bosonic sector of operators continues the line in Eq.(3) to left by replacing in it v to u, if we choose an approximation for the vibrational modes as the two-level vibrators.

The full Hilbert space of the Hamiltonian (1) without the bosonic operators has dimension 2^{2N} where N is the number os sites. The dimension may be reduced by using symmetries including the one related to conservation of spin-up and spin-down particle numbers, as well as geometrical symmetries of the cluster but not in the case of nonzero flux which is important in phase-sensisitive phenomena like quantum computation, superconducting weak links (Josephson effect and Andreev reflection), as well as Aharonov-Bohm effect and persistent currents in mesoscopic loops. In case when $\Phi \neq 0$, the only symmetry allowing for the reduction of the matrix dimension is the spin-up/spin-down particle number conservation

$$N_\sigma = 2 \sum_i \sigma a_{i\sigma}^+ a_{i\sigma}. \qquad (9)$$

Using Eqs.1,5, matrix operator H is block-diagonalized to partial matrices $H_{s_1 s_2}$ of smaller dimension (see Table 1) which are solved with the help of ABC.

2 THE "ABC" COMPILER

We focus on the numeric algorithm for coupled fermi- and fermi-bose systems allowing easy calculation of eigenvalues and eigenvectors of extremely large (of dimension up to 1000000, when executed on a standard Pentium PC) sparse complex Hermitian matrices. The program was devised with a newly developed Advanced Basic/C Compiler/Convertor/Programmer ("ABC") which produces C-codes as well as executables fit for various hardware/software environments (Windows, Linux and UNIX machines). The ABC C-code is translated from the QuickBasic dialect source code extended for easy use of mathematical routines such as complex numbers, arbitrary precision arithmetics, multidimensional integration, eigenvalue problem for sparse and conventional complex Hermitian matrices, etc.

ABC assumes a mathematical subspace of Basic dialect as it was specified in the Microsoft QuickBasic. By using the QuickBasic compiler as an **editor**, we have an additional advantage of testing the initial program code for possible errors by trying to execute (but not actually executing) the program thus eleminating most of (possible) syntax errors. The ABC code accepts complex numbers, special functions, arbitrary precision floating-point variables and a number of standard (and sometimes new) mathematical algorithms written in compliance with the (pseudo)QuickBasic dialect, so that the error checking is also applicable to these QuickBasic extensions within the QuickBasic rules. As an example, below is a **full** program in ABC

$DIM\ a, b, c, x, y\ AS\ DOUBLE:\ a = 0.111: b = 0.222$

$c = integ(x, 0, 1, y, 1 - x, 1 + x, SIN(pi * a * x * y + b)^2)$

PRINT a; b; c

for calculating an integral

$$c = \int_0^1 dx \int_{1-x}^{1+x} dy \sin^2(\pi a x y + b). \qquad (10)$$

In case when program execution is assumed on a machine different from the one of the ABC (e.g., faster, allowing larger RAM), the C-code appropriate to that

Table 1: Maximal reduced dimensions and other parameters for various clusters. N_s - number of sites, N_e - maximal number of electrons on cluster, D_H - dimension of the Hilbert space of cluster's Hamiltonian matrix, D_R - maximal dimension of the reduced matrix $H_{s_1 s_2}$.

Cluster type	N_s	N_e	D_H	D_R
Tetrahedron	4	8	256	36
Octahedron	6	12	4096	400
Cube	8	16	65536	4900
Icosahedron	12	24	16777216	853776
Ring	8	16	65536	4900
Ring	10	20	1048576	63504
Prism	2×6	24	16777216	853776
Prism	3×3	18	262144	15876

machine is generated. The codes thus produced are generally equal, or faster, than the conventional C-codes on same machine. Unlike similar programs for mathematical calculations (Maple or Matlab), ABC doesn't support any sophisticated graphics and, generally speaking, is **not** an advanced interactive routine. Also, dynamic strings are limited to the scope necessary for easy communication with the compiler (command-line data input and output, helps, etc.). The goal is rather in easy programming for nonprofessionals (physicists, mathematicians), on a professional level.

3   PHYSICAL IMPLEMENTATION

An example of numeric solution, Fig.2, represents the mesoscopic parity effect [2], i.e., number-parity sensitive dependence of the energy of cluster (mesoscopic superconductivity [3]), and the energy versus magnetic flux threading the cluster dependence (representing the persistent-current [4]) and supercurrent effects. The program allows calculation of the energy and other relevant physical characteristics of cluster with the single algorithm in which the cluster type (cubic, orthohedral, etc.) as well as the coupling strengthes are specified as parameters. In previous works, cubic cluster [5] and the cluster 4×4 [6] have been examined within the Hubbard model at $\Phi = 0$ for restricted value of electron filling.

3.1   The Hubbard Model

The Hubbard model (Hamiltonian (1) with $U > 0$ and $V = W = V_{ph} = 0$) was suggested for explanation of high-temparature superconductivity in ceramic metals ($La_{2-x}Sr_xCuO_4$, $YBa_2Cu_3O_{7-x}$). Some authors claim that superconductivity may exist in crystal without the electron-phonon interaction and with the repulsive interaction between opposite-spin electrons at sites. The problem was analized, in particular, within the Quantum Monte-Carlo computational method [7] near the

half filling (corresponding to the number of electrons nearly equal to the number of sites) without the conclusive results.

In small specimens, the question arizes whether superconductive pairing can survive in case when the energy level spacing approaches, or becomes larger than the superconducting energy gap [8]. It was suggested [2] that lowering of system energy at even number of electrons compared to the odd number, the so called **parity gap**

$$\Delta_p = E_{2n+1} - \frac{1}{2}(E_{2n} + E_{2n+2}), \qquad (11)$$

may serve for discrimination between the superconductive and nonsuperconductive behavior. Our calculation showed that the parity gap doesn't appear in case of positive Hubbard U but the negative-U Hubbard Hamiltonian is indeed superconductive. We present, as an example, the energy versus the number of particle dependence for cubic cluster $E(N)$ (Fig.2,left panel) which clearly shows the existence of the parity gap.

3.2   Occupation-dependent Hopping

Electon transport in oxides is determined by a peculiarity specific to atoms in the lowest part of the periodic table (H, O, B and, possibly, C). Specifically, in case of oxygen, delocalization of electron from the oxygen site (localization of hole at the site) results in significant increase of positive charge near the atom and therefore in shrinking of the electronic cloud near the atom thus reducing the transfer integral between the oxygens (or between the oxygen and the near metallic atom) sites. This will cause significant change in the transfer integral between the sites resulting in strong interatomic interaction (which is neither attractive nor repulsive but nevertheless results in electron pairing). The Hamiltonian responsible for this interaction is displayed as a second line in Eq.(1) and consists of the multiplicative (V)
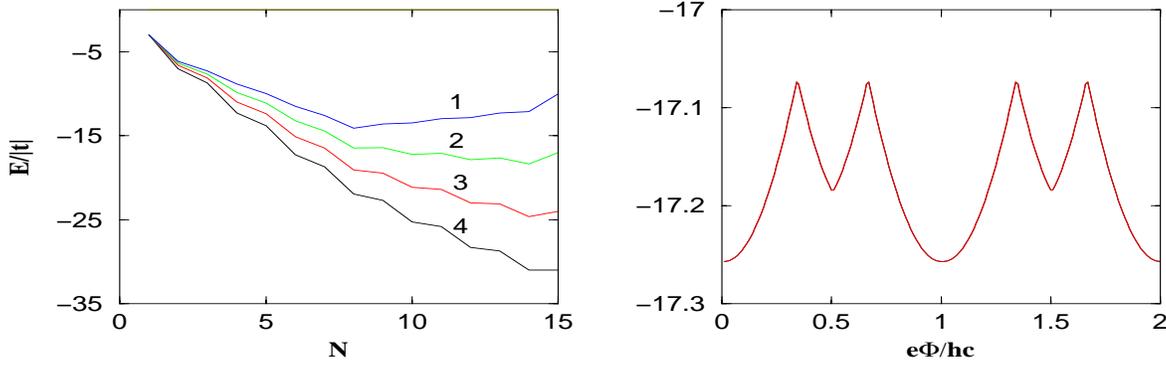
Figure 2: (a)Energy versus number of particles in negative-U cubic cluster. 1 - $U/|t| = -1$, 2 - $U/|t| = -2$, 3 - $U/|t| = -3$, 4 - $U/|t| = -4$; (b)Energy versus magnetic flux threading cubic cluster. hc/e-periodicity represents the persistent current effect, the $hc/2e$-periodicity is accounting for the pairing (superconductive) correlation.

and additive (W) occupation-dependent hopping amplitudes. Depending on the values of V and W, energy versus particle number dependence shows dips with a nonzero parity gap. This may serve as a possible mechanism of high-temperature superconductivity in oxide metals [1], [9], [10].

3.3 Persistent Current and Flux Quantization

Magnetic flux dependence of cluster energy produces a current

$$J = -\partial E/\partial \Phi. \tag{12}$$

Such currents, termed persistent currents, exist even in the noninteracting Fermi gas [4] and have periodicity in magnetic flux equal to the flux quantum $\Phi_0 = hc/e = 4.14 \cdot 10^{-7} G \cdot cm^2$. Superconducting cluster (the one with the negative value of U or the nonzero value of V, W, in certain domain of the ratio V/t, W/t), develops the $E(\Phi)$ dependence with twice shorter periodicity than in noninteracting Fermi gas [1], as it evidenced in Fig.2,b (right panel). Similar dependences have been calculated earlier for mesoscopic rings [11], [12].

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