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Application of the new LDA + GTB method for the band structure calculation of n-type cuprates ☆

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Abstract

A novel hybrid scheme is proposed and applied for band structure calculations of undoped n-type cuprate Nd_2CuO_4. The ab initio LDA calculation is used to obtain single electron and Coulomb parameters of the multiband Hubbard-type model. In strong correlation regime the electronic structure within this model is calculated by the generalized tight-binding (GTB) method, that combines the exact diagonalization of the model Hamiltonian for a small cluster with perturbation treatment of the intercluster hopping and interactions. For Nd_2CuO_4, this scheme results in charge transfer insulator with value of the gap and band dispersion in agreement to the experimental data.

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In spite of great success of the ab initio LDA calculations for metallic systems it appears to be inadequate for strongly correlated electron systems (SCES). Several approaches to include strong correlations in the LDA method are known, e.g. LDA + U and LDA-SIC. Both methods result in the correct antiferromagnetic insulator ground state for La_2CuO_4, but the origin of the insulating gap is not correct—it is formed by the local single-electron states splitted by spin or orbital polarization. The spectral weight redistribution between Hubbard subbands is very important effect in SCES. This effect is incorporated in the hybrid LDA + DMFT and LDA + + approaches. But the electron self-energy in LDA + DMFT approach is calculated by the DMFT theory in the limit of infinite dimension and is, therefore, **k**-independent [1].

A generalized tight-binding (GTB) [2] method has been proposed to study the electronic structure of SCES. The first step in this method is the exact diagonalization of the intracell part of the model Hamiltonian and construction of the Hubbard operators on the basis of the exact intracell multielectron eigenstates. Then, the model Hamiltonian is rewritten in terms of these Hubbard operators. In the obtained Hamiltonian, the intercell hoppings t are treated by the perturbation theory over $t/U \ll 1$. In the Mott–Hubbard insulators far from the Mott transition this is a good small parameter. For cuprates, instead of large $t_{pd} = 1\,\text{eV}$ the intercluster hoppings are strongly decreased and is less then 0.5 eV (see values below), while the effective U is large and given by the value of the charge-transfer gap $E_{ct} \approx 2\,\text{eV}$. The GTB calculations for undoped and underdoped cuprates [3] with fitting parameters listed in Table 1 are in good agreement to the ARPES data.

As any model approach the GTB method is not ab initio, there are many parameters. Generally, the question arises

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Table 1
Single-electron energies and hopping parameters of holes obtained by fitting [3] GTB band structure to experimental data and in the present ab initio calculations

	$\varepsilon_{d_{x^2-y^2}}$	ε_{p_x}	$\varepsilon_{d_{3z^2-r^2}}$	ε_{p_z}	t_{pd}	t_{pp}	t'_{pd}	t'_{pp}
Fitted	0	1.4	0.5	0.45	1	0.56	0	0.1
Ab initio	0	1.38	0.79	0.31	1	0.57	0.08	0.02

All values in eV, t_{pd} stands for hopping $d_{x^2-y^2} \leftrightarrow p_x, p_y$; $t_{pd}/\sqrt{3}$ for $d_{3z^2-r^2} \leftrightarrow p_{x,y}$; t'_{pd} for $d_{3z^2-r^2} \leftrightarrow p_z$, t_{pp} for $p_x \leftrightarrow p_y$; t'_{pp} for $p_{x,y} \leftrightarrow p_z$).

how unique the set of parameters is. To overcome this restriction we have proposed in this paper a novel LDA + GTB scheme that allows to calculate the GTB parameters by the ab initio LDA approach. Although LDA calculation does not give correct description of the SCES band structure, it gives ab initio parameters and reduced number of essential orbitals, that give the indication of the "minimal reliable model". Then, the effects of strong electron correlations in the framework of this model with ab initio parameters are explicitly taken into account within the GTB method and the quasiparticle band structure is derived.

LDA band calculation for n-type cuprate Nd_2CuO_4 (NCCO) with T′-structure was done within TB-LMTO-ASA [4]. It was found that the band crossing Fermi level have character of Cu-$d_{x^2-y^2}$ and O1-$p_{x,y}$ (antibonding pdσ orbital). Analysis of the LDA bands shows that the minimal reliable model for NCCO should be the 3-band p–d model [5]. To be more general and to be able to compare results with p-type cuprates further we will consider more complicated but more general multiband p–d model [6], that include Cu-$d_{3z^2-r^2}$ and O2-p_z orbitals beside Cu-$d_{x^2-y^2}$ and O1-$p_{x,y}$. To obtain hopping integrals for different sets of bands included in consideration, we apply projection procedure using Wannier functions formalism [7]. Hopping parameters decay rapidly with distance so in GTB calculation we will use only nearest Cu–O and O–O hoppings. Obtained hoppings and single-electron energies are listed in Table 1. Using these parameters it is possible to calculate parameters [8] of the effective $t-t'-t''-J$ model for n-type cuprates (all values in eV): $t = 0.410$, $t' = -0.013$, $t'' = 0.058$, $J = 0.137$. Values of the intra-atomic Hubbard repulsion $U = 10\,\text{eV}$ and Hund exchange on copper $J = 1\,\text{eV}$ were obtained in constrained LDA [9].

The GTB band structure obtained for both phenomenological and ab initio sets of parameters is almost identical (see Fig. 1): the valence band and the conductivity band divided by the insulator gap of the charge transfer origin $E_{ct} \approx 2\,\text{eV}$. The valence band have bandwidth about 6 eV and consists of a set of very narrow subbands. The dominant spectral weight in the top of the valence band stems from the oxygen p-states, while for the bottom of the empty conductivity band it is from Cu-$d_{x^2-y^2}$-states. Both methods give small, less then 0.5 eV, splitting between the

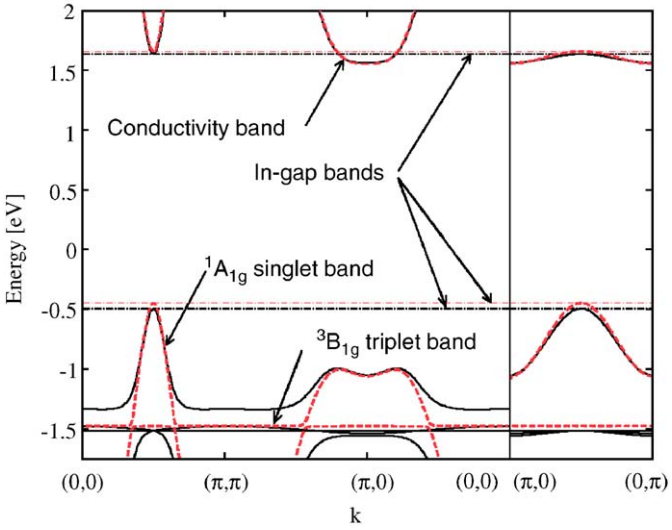

Fig. 1. The antiferromagnetic band structure of Nd_2CuO_4 obtained in the GTB method with phenomenological set of parameters (solid lines) and in the LDA + GTB method (dashed lines). Dispersionless in-gap states are shown by dot-dashed lines.

$^1A_{1g}$ Zhang–Rice-type singlet band and $^3B_{1g}$ narrow triplet band. The energy of Cu-$d_{3z^2-r^2}$ orbital plays the dominant role in this splitting in the GTB method. Both methods result in the conductivity band minima at the $(\pi, 0)$ point, consistent with experimentally observed indirect insulating gap in NCCO [10]. In the LDA + GTB method the triplet band dispersion and the singlet–triplet hybridization are much smaller mainly due to the smaller values of t'_{pp} obtained in LDA + GTB method, because it is this microscopic parameter that gives main numerical contribution to the effective hopping that determine mentioned hybridization. So, despite some minor discrepancies, both GTB method with phenomenological parameters and LDA + GTB method without free parameters gives similar band dispersion.

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References

[1] W. Metzner, D. Vollhardt, Phys. Rev. Lett. 62 (1989) 324.
[2] S. Ovchinnikov, I. Sandalov, Physica C 161 (1989) 607.
[3] V.A. Gavrichkov, et al., JETP 91 (2000) 369;
V. Gavrichkov, et al., Phys. Rev. B 64 (2001) 235124.
[4] O.K. Andersen, et al., Phys. Rev. B 34 (1986) 5253.
[5] V.J. Emery, Phys. Rev. Lett. 58 (1987) 2794;
C.M. Varma, et al., Solid State Commun. 62 (1987) 681.
[6] Yu. Gaididei, V. Loktev, Phys. Stat. Sol. B 147 (1988) 307.
[7] V.I. Anisimov, et al., Phys. Rev. B 71 (2005) 125119.
[8] M.M. Korshunov, et al., Physica C 402 (2004) 365.
[9] O. Gunnarsson, et al., Phys. Rev. B. 39 (1989) 1708.
[10] N.P. Armitage, et al., Phys. Rev. Lett. 88 (2002) 257001.