Anomalous spin and lattice coupling in La_2CuO_4 calculated by first-principles quantum Monte Carlo

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Introduction

After ~25 years since their discovery, the high temperature (~120 K) of the superconducting transition in copper oxide materials is unexplained.

A major confounding property of these materials is that their behavior is very complicated near superconductivity, exhibiting structure, magnetic, and charge reordering.

Accurate computer simulations could help untangle the different effects in the copper oxides. However, the materials are strongly correlated, making it challenging to find good approximate numerical solutions.

We use Monte Carlo methods to approximately solve the Schroedinger equation for the copper oxides in order to assess the quality of this approximation and to help disentangle the magnetic and structural degrees of freedom. We find excellent accuracy and help explain some confusing experiments on these materials.

Computational Method

Input

$$\hat{H} = -\frac{1}{2}\sum_i \nabla_i^2 + \sum_{i<j}\frac{1}{r_{ij}} - \sum_{i,\alpha} V_\alpha(r_{i\alpha}) + \sum_{\alpha<\beta}\frac{Z_\alpha Z_\beta}{r_{\alpha\beta}}$$

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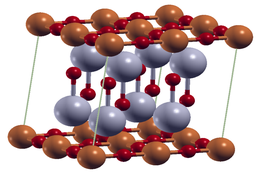
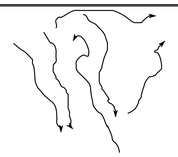


Variational wave function

$$\Psi(r_1, r_2, r_3, \ldots, r_N) = \mathrm{Det}[\phi_i^\uparrow(r_j^\uparrow)]\mathrm{Det}[\phi_i^\downarrow(r_j^\downarrow)]\exp\left[\sum_{i,j,\alpha} u(r_{i\alpha}, r_{j\alpha}, r_{ij})\right]$$

Stochastic projection (FN-DMC)

$$\lim_{\tau\to\infty} \exp[-\tau\hat{H}]\Psi(r_1, r_2, r_3, \ldots, r_N)$$



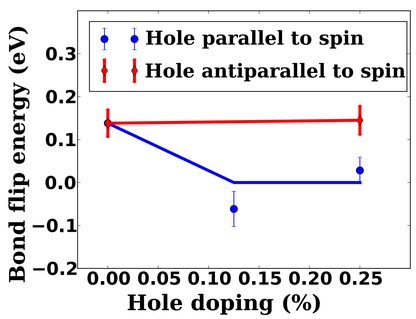
Details:
First principles Hamiltonian with all atoms
Ground state calculated by fixed node diffusion quantum Monte Carlo

Slater-Jastrow trial wave function with hybrid PBE0 orbitals
Correlation consistent pseudopotentials[1]

Packages:
CRYSTAL (DFT) (www.crystal.unito.it)[3]
QWalk (QMC) (www.qwalk.org)[2]

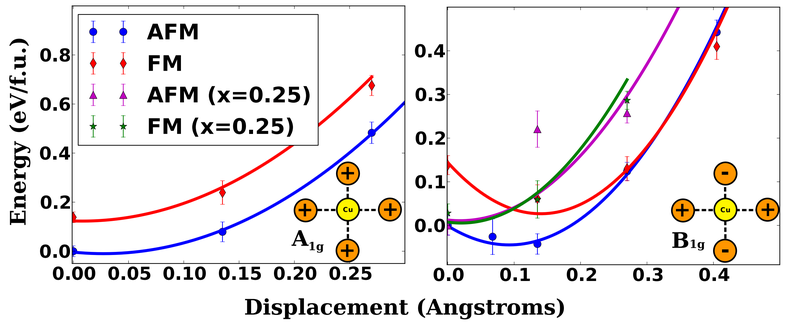
Benchmarking

Quantity	FN-DMC	Experiment
J (eV)	0.14(3)	0.12
Magnetic moment of Cu (Bohr)	0.6	0.6
Quasiparticle gap (eV)	2.0(3)	2.2
B_{1g} phonon frequency (meV)	36(2)	40-42
A_{1g} phonon frequency (meV)	46(2)	>42



The material parameters are within statistical errors of the experiment. We also obtain a reduction of magnetic excitation energy when the material is doped, indicating our computer solution is a good representation of the realistic material

Spin-lattice coupling



Oxygen buckling modes of vibration studied. The A1g mode does not couple with spin, but the B1g mode couples strongly.

The spin-structure coupling is removed when the material is doped.

Is there a connection between the the spin-lattice coupling and the reduction of magnetic excitation energy?

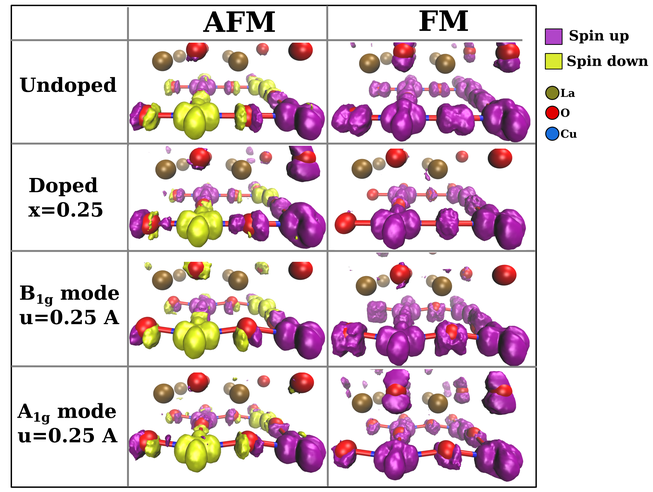
Connection between spin-lattice coupling and doping dependence of magnetic order

At right is an isosurface of the electronic spin density in the spin aligned and spin anti-aligned states.

The spillover from the copper atoms onto the oxygen atoms is a major difference between the different simulations. This is a way of visualizing the so-called superexchange interaction that causes magnetic order.

The spillover can be modified either by doping the material, which removes an electron on the oxygen atoms, or by moving the oxygen atoms in the correct way.

The B_{1g} mode couples with the spin because the oxygen interacts with the overhead lanthanum atoms two at a time, contrary to the A_{1g} which has four oxygens approaching the lanthanum.



Summary

First application of quantum Monte Carlo techniques to superconducting copper oxides.

High accuracy attained.

New physical understanding of the relationship between charge, spin, and lattice degrees of freedom.

The authors gratefully acknowledge funding from DOE FG02-12ER46875

References
[1] Burkatzki et al. J. Chem. Phys. **126** 234105, Lee et al. PRB **62** 13347
[2] Wagner et al. J. Comp. Phys **228** 3390
[3] Dovesi et al. Zeit. Kristallographie **220** 571