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Spin waves and large electron-phonon coupling near the metal-insulator transition in hole-doped high-T_c oxides

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The effects of spin and lattice modulations on the band structure of high-T_c oxides are described, with $HgBa_2CuO_4$ as an example. A simple model for band gaps induced by potential perturbations with long-range Fourier components is applied to the barrel band. The opening of gaps is confirmed in self-consistent band calculations for modulations in elongated supercells, where spin waves and phonon coupling compete for equal q vectors. It is argued that the wavelengths of spin-wave modulations, and of the phonon modes with large coupling, depend on the doping. This mechanism supports the idea that pseudogaps are caused by stripelike spin modulations in underdoped systems, while superconductivity is attributed to enhanced electron-phonon coupling for long-wavelength phonons.

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The mechanism behind high-T_c superconductivity in copper oxides is still unknown. From the vast amount of experimental information that has been collected from high-T_c oxides, one can recall some facts that seem to distinguish these materials from ordinary low-T_c superconductors.[1–3] Phase sensitive tests of the pairing indicate *d*-wave symmetry of the superconducting gap.[4] This is also implied from photoemission and tunneling measurements where the gap is highly anisotropic over the Fermi surface (FS).[5,6] Such measurements detect pseudogaps, which extend well above T_c in underdoped systems. They appear to have the same symmetry as the superconducting gap, making the FS visible only along the diagonal. The isotope effect, which is small and variable, exists also for the pseudogap.[7] Spin fluctuations follow the same type of antiferromagnetic (AFM) orientation of ordered Cu moments that can be seen in undoped, insulating copper oxides. Separation of metallic and insulating regions form static stripes for some compositions,[8] but they might be dynamic in other cases. Band theory based on the density functional approximation fails to describe the undoped, insulating cuprates, while the bands and FS of metallic, superconducting cuprates agree well with experimental data. The electron-phonon coupling (λ) calculated from band theory[9,10] is quite large in view of the low density of states (DOS) in these materials, but still not high enough to explain the high T_c's.

Here we perform self-consistent electronic structure calculations based on the local (spin) density approximation (LDA),[11] for long-wavelength excitations of either spin fluctuations or phonons, by use of large supercells. As will be shown, spin waves and phonons seem to compete for the same q vectors, and the perturbations will be large at the FS, leading to a tendency for gap formation in both cases. We focus on $HgBa_2CuO_4$ (HBCO), since it is relatively simple with one CuO-plane and only one cylindrical FS, a so-called ''barrel.'' Such a piece of FS is common to all high-T_c oxides and probably involved in the mechanism for superconductivity. The lattice constant a is 7.32 a.u. The bands are calculated using the linear muffin-tin orbital method, where five empty spheres are introduced in the most open parts of the structure. The structure, details of the method, and band results for the basic unit cell, are described earlier.[12,10] Only Cu, and to some extent the planar O, have a significant DOS at E_F. The band structure of the normal cell is shown in Fig. 1 for k points in the $k_z=0$ plane. The z projection of the FS for the undoped case is shown in Fig. 2. The FS reaches the zone boundary at X for a doping of about 0.25 hole per formula unit (f.u.). The cell of the AFM insulating case has two Cu in the xy plane with opposite magnetic moments. This cell of 2 f.u. has 26 sites (16 atoms and 10 empty spheres). The AFM state can be reached by introducing large correlation, increased exchange or by application of external field. Here we include a staggered field (positive and negative with 30 mRy amplitudes) on the two Cu in the AFM cell. The undoped material is insulating with this field, with a gap of 10 mRy and local moments of about $0.5\mu_B$. The critical field for having a gap is about 20 mRy.

The 26-site cells are put together in the [1,1,0] direction, to form larger cells in order to study q waves modulated along the diagonal (with respect to the Cu-O bonding). Four supercells containing 104, 130, 156, and 208 sites are con-

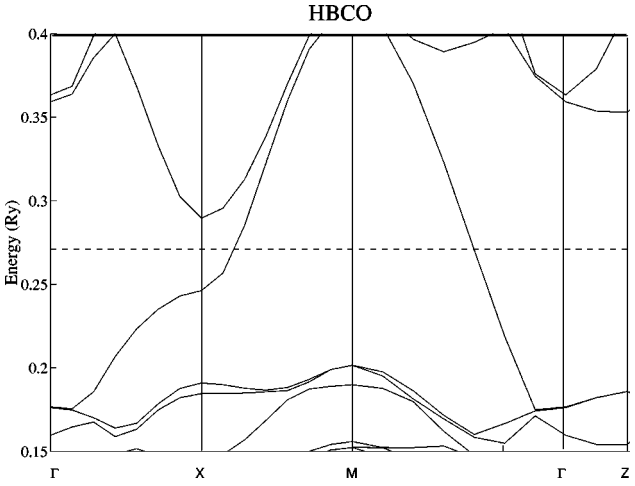

FIG. 1. Band structure of $HgBa_2CuO_4$ along symmetry directions in the $k_z=0$ plane of the tetragonal BZ. The Fermi energy is indicated by the broken line.

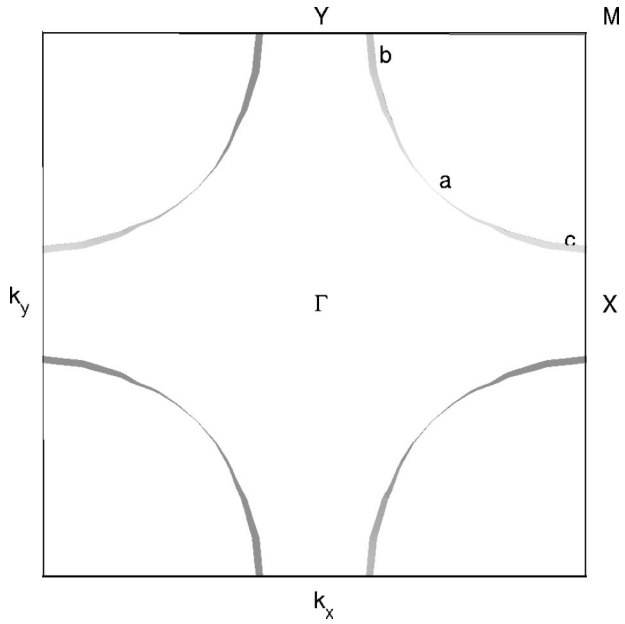

FIG. 2. The *z*-projected FS of (undoped) $HgBa_2CuO_4$ at a band filling of 63 electrons per f.u.. The M-centered barrel will swell for increasing hole doping. At 62.75 electrons per f.u the band will reach the X point to form a van Hove singularity.

sidered, corresponding to a maximal wavelength of $8\sqrt{2}a$. AFM modulations along [1,0,0] require a two-layer wide unit cell (along y). A cell of 208 sites will describe spin modulation with wavelength $8a$ along x. Phonon modulations (without spin) use a one-layer wide cell, so that a 104-site cell describes a modulation vector $[8a,0,0]$. In each case we calculate the bands in 24 k points in the respective irreducible Brillouin zones (BZ's). The DOS is determined by tetrahedron k-point integration, where it is assumed that the energies vary linearly within each tetrahedron.

The z dispersion of the barrel band is very small, as seen from the widths of the FS in Fig. 2, but the dispersion is large in the xy plane, as along the diagonal Γ-M (cf. Fig. 1). A modulation of the potential $V_q \sin(qr)$, expressed by the Fourier coefficient V_q, where q is along [1,1,0], will open a gap for a free-electron-like band. From the well-known solution for bands near the zone boundary in semiconductors,[13]

$$E(k)=\frac{1}{2}(\epsilon_k+\epsilon_{k-q})\pm\frac{1}{2}\sqrt{(\epsilon_k-\epsilon_{k-q})^2+4V_q^2}, \qquad (1)$$

which at the zone boundary $k=\frac{1}{2}q$, becomes $E(q)=\epsilon_{(1/2)q}\pm V_q$. Thus, there are two states with a gap, $2V_q$, in between, at the new BZ limit at $\frac{1}{2}q$. The square of the wave function belonging to the state below the gap has the largest amplitude where the potential is most attractive. The corresponding function for the state above the gap has its phase shifted, so that the large amplitude coincides with the repulsive potential.

The AFM state in the undoped case is, apart from exchange energy, stabilized by the opening of the gap at E_F. Hole doping made in a rigid-band manner on the same DOS leads to a metal, without the stabilizing effect of the gap. As will be seen, modulations of the AFM moments will open gaps at different doping levels, with possible energy gains. An example of a modulation is when the Cu sites projected on the [1,1,0] direction have moments: zero, up, down, up, zero, down, up, down. Such a ''stripe'' has $\pm V_q$ components in the spin potentials, with a wavelength covering totally 8 Cu layers. If E_F is far below (or far above) the gap, both states will be occupied (or unoccupied) with no gain in total energy. It is only if E_F is near the gap that there will be a possibility to gain total energy. The occupied state reduces its kinetic energy, while the rise in energy of the upper, empty state does not contribute to the total energy. The process is self-supporting via the exchange potential, since an increased spin density in regions where the potential is spin splitted will increase the potential splitting further. Nesting, which becomes more evident at higher doping, helps to generalize the process over a large portion of the FS, but Eq. (1) is valid locally. Thus, the argument is that the CuO-plane systems can chose the wavelength of the modulation according to its doping to make an optimal gain in total energy. This mechanism always opens the gap (or pseudogap) close to E_F, leading to a correlation between the wavelength and doping. These arguments can be used as long as there are no other bands near E_F, which may involve an increase of the total energy when a modulation is imposed.

Similar arguments can be used to discuss the coupling between phonon modes and the electronic states near E_F. For example, a phonon-induced modulation of the potential on Cu sites will also have its Fourier component V_q. (Since the local Cu DOS is the highest near E_F, its largest effects will be from potential changes on Cu sites.) The (non-spin-polarized) band will open a gap as for each of the spin bands in the AFM case, if the gap is close to E_F. As before, one particular wavelength will be preferred, depending on the doping. It is worth noting that for a given doping, the same q vector is involved for both spin waves and phonons. This mechanism will be dynamic for phonons, unless the gain in energy will make the phonon completely soft to make a lattice instability. Spin waves could be pinned to impurities and therefore be static, but coupling is likely between spin waves and phonons of equal q vectors.

Next, we consider the band results. The basic results from the simple, one-dimensional model are confirmed by the band calculations, and the gap can be formed over the whole FS for varying doping. AFM modulations are induced by sinuslike modulations of staggered fields (on Cu sites, throughout the iterations) with the same maximal amplitude as for the undoped case. The undoped system contain 63 electrons per f.u.. However, the gaps (or pseudogaps) found in the four cases with elongated cells are always found at one state (one electron of each spin) below this electron filling. This means at band fillings of 502, 628, 754, and 1006 electrons, for the four supercells of increasing lengths. The doping is adjusted to coincide with these fillings, by use of the virtual crystal approximation. The DOS near E_F with and without AFM modulation, are shown in Fig. 3 for the 104-site cell. The gap is small, like a pseudogap, and it coincides with E_F when the doping is 0.25 holes per f.u., corresponding to the particular q. Modulations in the longer cells give

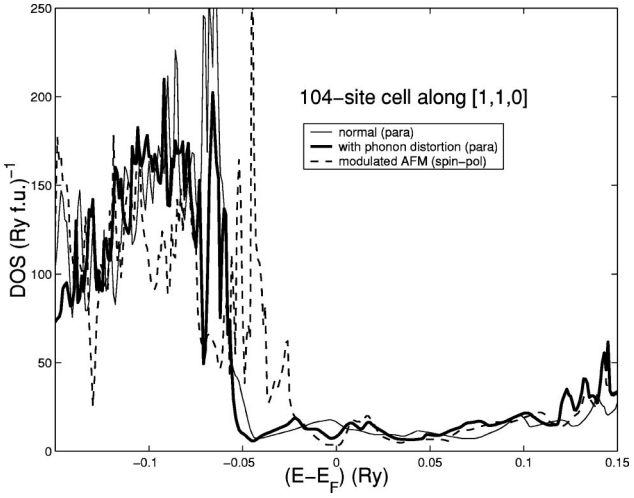

FIG. 3. Paramagnetic DOS for a 104-site unit cell of HBCO (doped with 0.25 holes per f.u.) along [1,1,0] (thin line), paramagnetic DOS when the structure contains a sinus modulation of Ba_z positions (heavy line), and the DOS for a sinus-modulated AFM state along the cell (broken line). The latter is obtained via applied fields (maximum amplitude 30 mRy) on the Cu sites. A higher field will open the gap completely, but also move the Cu-*d* band edge closer to E_F.

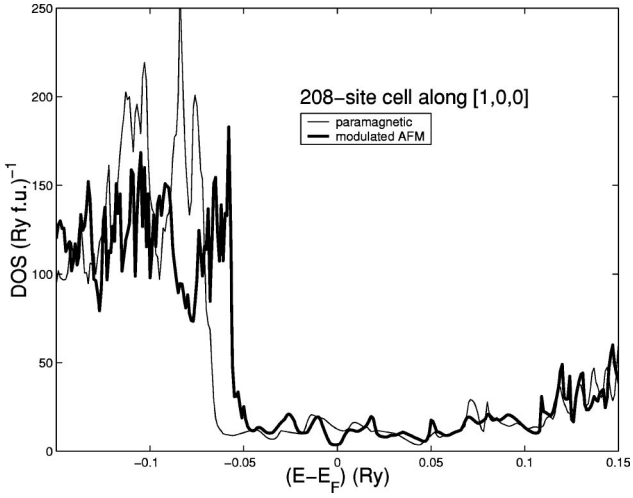

FIG. 4. Paramagnetic (thin line) and AFM (heavy line) DOS for a 208-site unit cell of HBCO (doped with 0.125 holes per f.u.) along [1,0,0].

similar small gaps, or pseudogaps, but at positions corresponding to decreased hole doping, at 0.2, 0.1667, and 0.125 holes per f.u., for the cells with 130, 156, and 208 sites, respectively. The opening of the pseudogap starts near the point ''a'' in Fig. 2. Stronger fields form a complete gap extending over the whole FS. Thus, in agreement with the simple model, the band results show that the barrel band can be cut to form gaps at different positions along Γ-M, depending on the Fourier component of the potential perturbation.

AFM modulations in the *x* direction are made for the 208-site cell, where the pseudogap is found for a doping of 0.125 hole per f.u., as for the longest cell along the diagonal direction (see Fig. 4). By folding the *k* points in the BZ back to the original BZ, it is seen that the partial opening of a gap is largest near point ''b'' in Fig. 2. This is consistent with the observations made by photoemission, where the FS seems to ''disappear'' except near the diagonal.[5] (It is assumed that stripes are running both along *x* and *y*,[1] making their effect superimposed at ''b'' and ''c.'') Also the observation of static stripes along [1,0,0] in one high-T_c material[8] suggests that [1,0,0]-modulations are more probable than diagonal ones. This can be understood from the band results by the larger *z* dispersion near points ''b'' and ''c'' on the FS, making the DOS higher on this part of the FS. A band opening on this part will lead to a larger gain in total energy than if the gap opens near point ''a.'' As the orientation in real space can be associated with the different FS points, it is therefore preferable for the system to choose the [1,0,0] orientation leading to the largest gain in energy.

Other theories based on strong on-site correlation have been shown to lead to stripe formation and spin-charge separation.[1] Here, in a band description, the spin, charge, and local DOS information show that the charges on Cu of different polarization are very similar. The DOS at E_F is largest on the nonpolarized Cu sites, but still sizable on the polarized Cu sites, at least for one of the spins. Thus, conduction is not only restricted to the zero-moment part of such stripes. Further, the connection between the doping and the wavelength implies longer wavelengths for low doping. It is probable that long-wavelength modulations are more stable than short ones. This is because at least two Cu sites have no moment in each modulation, i.e., no exchange energy can be gained from a polarization of these two sites. The fraction of zero moment Cu sites is larger for short-wavelength modulations, making them less stable. This cannot be verified by *ab initio* total energy calculations, but for the short modulation of only six Cu layers along [1,0,0] it is more difficult to open a gap even for large field amplitudes, and there is a topologic change of the FS at the X point when the doping approaches 0.25. Therefore, a possibility is that pseudogaps, caused by spin waves, cannot exist for too large doping. This is consistent with observations of pseudogaps only in underdoped high-T_c oxides. A critical concentration of about 0.19 has been suggested.[3] This is to be compared with the values 0.125 for the shortest modulation along [1,0,0] and 0.25 along [1,1,0] to produce a ''good'' pseudogap in the calculations.

The non-spin-polarized band calculations for phonon modes are *ab initio* within LDA, for the imposed lattice distortions. Coupling between spin waves and phonons is plausible, but we do not study such cases here. Different lattice modulations along [1,0,0] and [1,1,0] are considered in the elongated supercells, like the change of the distance between Cu and apical O, or between Ba and the CuO plane. ''Breathing'' modes are modeled by moving the four planar O sites alternatively towards or away from the central Cu. The modulations are chosen to be sinuslike. Distortion amplitudes of the order $0.01a$ are expected at low temperature for typical force constants and zero-point motions,[10] but in order to enhance the effects to be seen in the DOS, we also consider amplitudes of $0.03a$.

The DOS for the 104-site cell is shown in Fig. 3. By comparing the backfolded k points with the points in the normal BZ in Fig. 2, one can verify that the effects due to modulations along the diagonal or along the CuO bonding, indeed are concentrated to the expected parts of the FS. Moreover, if the sinuslike modulation is replaced by the absolute value of sinus, one finds about one order of magnitude smaller energy shifts near E_F. Results for the other cells are similar, so that the distorted structures show some different DOS compared to the undistorted cases, but no real gaps are developed. However, strong changes of the energies mean that λ is large for these modes. Full calculations of λ will not be presented here, but a few observations can be made. As was argued above, the wavelengths are determined via doping so that only energies close to E_F are changing. Calculations of electron-phonon coupling made for the undoped compositions[9,10] will miss this effect, since the sensitivity to V_q of a certain wavelength appears below the position of E_F for the undoped system. The largest change in energy from the model above, is equal to V_q, i.e., equal to the maximal amplitude of a sinusoidal potential perturbation. Two examples of O and Ba distortions with V_q from the potential in the band calculation, give λ in the range 1–2. This is from an approximate expression[10] for the long-range coupling, $\lambda = NV_q^2/\Sigma K(\Delta R)^2$, where N is the DOS at E_F per cell. K is a force constant,[10] ΔR the displacement of each site, so that K times the sum over all ΔR^2 gives the total elastic energy of the deformation.

In conclusion, the simple model of gap formation, supported by band results for $HgBa_2CuO_4$, suggests a connection between spin waves, stripes, pseudogaps, and strong electron-phonon coupling, because of a sensitivity to perturbations V_q. The amplitudes of V_q due to phonons of realistic deformation amplitudes at low T, are smaller than in the case of AFM modulation, and fully developed gaps are normally not visible in the DOS. But a particular V_q induced by phonons favors band shifts at some sections of the FS and a large λ. Any perturbation, phonons, spin, or valence fluctuations leading to a similar V_q component will show similar effects on the electronic states. This scenario explains a number of features of the high-T_c oxides mentioned above. Energy arguments suggest that the spin waves are less stable for increasing doping. If this picture is true, and spin fluctuations are harmful to superconductivity, one can expect that pressure should suppress the spin fluctuations more at low doping and increase T_c, while the effect of pressure should be smaller in overdoped samples. This is partly seen experimentally although the results are sensitive to pressure induced oxygen ordering at high temperature.[14] It can be speculated that large electron-phonon coupling coexisting with spin stripes open a possibility for superconducting fluctuations between regions of spin modulations, and that this will contribute to the pseudogap as well. Competition between different mechanisms can make interpretation of experiments difficult.

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