A model of the T-dependent pseudogap and its competition with superconductivity in copper oxides.

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Results for pseudogaps are obtained from a band model, where the stability of the gap depends on the amplitudes of vibrational displacements, or magnetic moments, and their coupling to electrons. A one-particle gap is favored by normal thermal excitations of phonons or spin waves. Another gap can be generated by spontaneous waves at lower temperature, if the electronic energy gain overcomes the elastic/magnetic energy needed for increased amplitudes of the oscillations. This state is characterized by charge or spin density waves. The pseudogap has many features in common with the superconducting gap, and the model lends support to the interpretation that the pseudogap is a precursor of, and competes with, superconducting pairing.

PACS numbers: 74.20.-z,74.20.Fg,74.20.Pq

The understanding of high-$T_C$ cuprates has increased a lot since the discovery of these systems more than 20 years ago [1], although a convincing mechanism behind the high superconducting $T_C$ is not found. A pseudogap with transition temperature $T^* > T_C$ at low hole doping seems to be characteristic for all cuprates [2–4]. Ab-initio band calculations for long supercells of copper oxides containing phonon distortions or spin waves show a partial gap (pseudogap) in the density-of-state (DOS) [5, 6]. The energy position of this gap depends on the length of the supercell, i.e. on the wave length of the periodic potential perturbation induced by the phonon/spin wave. Hence, if the one-particle gaps appear at the Fermi energy, $E_F$, then there must be a correlation between doping (determines $E_F$) and wave length. The appearance of the gap can be understood from the nearly-free-electron model (NFE), which has also been used to interpolate ab-initio results [7]. The pseudogap is an important part of cuprate physics, but the question if it helps or competes with superconductivity is not settled. On the other hand, it can be argued that artificial periodic waves such as given by periodic doping or oxygen ordering will lead to higher $T_C$, if the waves are tuned correctly [8, 9].

Presented here is a many-particle extension of the NFE band model, which can be used for the simulation of the T-dependence of the pseudogap. The model is based on one-dimensional potential perturbations from phonons or anti-ferromagnetic (AFM) spin waves. The criterion for having gaps at $E_F$ is made from the estimated total energy (kinetic electronic energy and energy of phonon/spin fluctuations) for an interacting electron-phonon (electron-spin) system, with many-body coupling parameters $\lambda$ or $\lambda_{sf}$ for phonons or spinfluctuations, respectively. A continuous metallic band can be gapped because of a periodic potential perturbation, as in the appearance of a gap for semiconductors [10]. The potential perturbation is such that two regions of k-space, at $\vec{k}$ and $-\vec{k}$, are affected equally. Only waves which modify electronic states around the Fermi surface (FS) are of interest, since changes of the bands far from the Fermi energy ($E_F$) make no change in the total kinetic energy.

This puts a constraint on the $q$-vectors of the perturbations. The results of the model imply similar mechanisms for the pseudogap and the superconducting gap in high-$T_C$ cuprates.

Phonons and spin fluctuations are normally excited thermally following the Bose-Einstein occupation, $g(T, \omega)$, of the phonon- or spin wave density-of-states (DOS), $F(\omega)$ or $F_m(\omega)$, respectively. The averaged atomic displacement amplitude for phonons, $u$, can be calculated as function of $T$ [10, 11]. Approximate results make $u^2_T \to 3k_B^2D/2K$ at low $T$ ("zero-point motion", ZPM) and $u^2_T \to 3k_B^2T/K$ at high $T$ ("thermal excitations"), where $\omega_D$ is a weighted average of $F(\omega)$. The force constant, $K = M_A^2$, where $M_A$ is a atomic mass, can be calculated as $K = d^2E/du^2$ ($E$ is the total energy). The corresponding relations for averaged fluctuation amplitudes of the magnetic moments, $m$, are the same, but without the polarization factor 3 and with the replacement of $K$ with $K_m = d^2E/dm^2$ [12]. The force constants $K$ and $K_m$ do not change with the oscillation amplitudes for harmonic oscillations. The time scales of phonons and electrons are sufficiently different for adiabatic relaxation of electrons and band gaps. The energy of an atomic oscillation, $U$, has an elastic contribution because of $u$, and a kinetic contribution because of the velocity, $v$. The time dependence of the sum, $2U(t) = Ku^2\cos^2(\omega t) + M_Au^2\sin^2(\omega t)$, is a constant in the harmonic approximation. $U(0) = \frac{1}{2}Ku^2$, where $u$ refers to the maximal atomic displacement, permits us to calculate $U$ from static conditions, and to identify $Ku^2$ as an ingredient of the standard expression for $\lambda$. Acoustic phonons are more efficient for having a clear gap, while optical phonons sometimes have $u=0$ everywhere and a smeared time average of the band gap.

Thermally activated phonons and spin waves create some disorder and will change the electronic states and the DOS, $N(E)$ [13]. Individual phonon- or spin-waves which cause a gap close to $E_F$ in the normal state are particularly interesting for high-$T_C$ copper oxides [3]. The gap is largest in the CuO bond directions, while the FS remains sharp in the diagonal direction, to form an FS-
Band calculations show that atomic distortions of phonons create a periodic potential along a chain of atoms, which can be modeled by

\[ V(x) = V_0 e^{-ix \cdot q} \]  \hspace{1cm} (1)

for phonon propagation along \( x \) with wave vector \( q \). The maximum amplitude, \( V_0 \), can be obtained from ab-initio band results as half of the band gap, or as the maximum difference of the potential shift within the unit cell. A spin wave makes an analogous perturbation within the spin polarized part of the potential, where the differences of opposite spins differ by a phase factor of \( \pi \). The result is an AFM spin configuration with wave length given by \( 2\pi/q \). Phonons and spin waves can be considered separately, but an important spin-phonon coupling (SPC) in the cuprates leads to unusual properties \([6, 14, 15]\). The model for finding the maximal temperature for \( T^* \) is obtained from eq. \( 4 \) but with the

The electron-phonon coupling \( \lambda \) is active for energies \( \pm \hbar \omega \) around \( E_F \), where it can be written \( N M^2/K \).  \hspace{1cm} (10)

The matrix element \( M \) for energies inside the interval \( \pm \hbar \omega \) can be evaluated as \( \langle \Psi^* (E_F, r) dV(r)/du \Psi (E_F, r) \rangle \), which is the first order change in energy caused by the perturbation \( dV(r) \) for \( du \rightarrow 0 \). For a finite value of \( u \) the change in energy will be finite and equal to the gap \( V_q \), since \( V_q/u \) is constant for harmonic vibrations. Thus, instead of calculating \( M \) as a matrix element it is convenient to take the value directly from the band gap, and \( M \) can be written \( V_q/u \) for energies close to \( E_F \), which makes \( \lambda = NV_q^2/Ku^2 \).

Totally there is a gain in energy if \( |U| \leq |E| \). The system will spontaneously increase \( u \) of the vibrations (from the normal value given by thermal excitations) in such a case. Other effects such as electron-electron correlation and potential terms add to the energy costs and can prevent a gap in many systems. With the DOS, \( U \) and \( E \) per unit cell the condition \( |U| = \langle E \rangle \) is written

\[ 1/2 Ku^2 = \int_{-\hbar \omega}^{0} \epsilon (N(\epsilon) - \tilde{N}(\epsilon)) d\epsilon \]  \hspace{1cm} (4)

for \( T = 0 \), where \( \tilde{N}(\epsilon) \) is the DOS with the gap and \( N(\epsilon) \), the DOS of the normal state, is assumed constant within \( \hbar \omega \) around \( E_F \). The integration is to \( \hbar \omega \) since \( \lambda \) is zero for energies larger than \( \pm \hbar \omega \).

With a substitution \( \epsilon^2 = \epsilon^2 + V_q^2 \) we obtain \( \tilde{N} = N|\epsilon|/\sqrt{\epsilon^2 - V_q^2} \) and,

\[ 1/2 Ku^2 = \int_{-\hbar \omega}^{0} N \epsilon d\epsilon - \int_{-\hbar \omega}^{-V_q} N \epsilon^2/\sqrt{\epsilon^2 - V_q^2} d\epsilon \]  \hspace{1cm} (5)

The result is

\[ Ku^2 = NV_q^2 \ln (2\hbar \omega/V_q) \]  \hspace{1cm} (6)

and

\[ V_q = 2\hbar \omega e^{-1/\lambda} \]  \hspace{1cm} (7)

since \( NV_q^2/Ku^2 \) turns out to be equal to \( \lambda \). The coupling determines the gap through constant ratios of \( V_q/u \). This result is similar to the BCS equation for the superconducting gap \([16, 17]\). The present state is different from a normal gapped state obtained via standard band calculations. It only appears because of electronic interaction near \( E_F \), and it disappears for small \( \lambda \).

Eqn. \( 5 \) has no solutions for \( \hbar \omega < V_q \), so the concurrent state with phonon softening, where all electronic energy goes into a renormalization of the phonon with \( K, \omega \rightarrow 0 \), is not described. Neither using \( W \) instead of \( \hbar \omega \) in eqn \( 5 \) gives a proper description of a static case, because states far below \( E_F \) do not have the correct wavelength and cannot contribute to \( V_q \).

The model for finding the maximal temperature for having a gap (\( T^* \)) is obtained from eq. \( 4 \) but with the
This is solved numerically through integrations of $(\int_{-\hbar\omega}^{\hbar\omega} e|e|/\sqrt{(e^2 - V_{q}^2)}| fde)/V_{q}^2$ for $V_q \rightarrow 0$. The ' in the second integral means that the energies where $|e| < V_q$ are excluded.

The results, shown in Fig. 1, has the same form as the solution for superconductivity:

$$k_B T^* = 1.13\hbar \omega e^{-1/\lambda}$$

For example, it can be verified from this formula and Fig.11 that a $\lambda$ of 0.5 makes $k_B T^* \approx 15$ meV when $\hbar \omega$ is 100 meV.

A non-constant DOS, with $N(\epsilon)$ from eqn. 11 inserted in eqn. 8 and $\hbar \omega$ being a large fraction $W$, tends to decrease $T^*$.

For AFM spin waves there is a cost in magnetic energy, which in the harmonic approximation can be written $U_m = \frac{1}{2}K_m m^2$. The change in potential on some site, $V_m$, is positive for one spin and negative for the other spin direction. This defines a $\lambda_{sf} = N1/2Mk/m^2$ as a coupling constant for spin-fluctuations. The rest of the equations are applicable with $\lambda_{sf}$ replacing $\lambda$ and with $\hbar \omega_{sf}$ being the energy of the spin wave.

Typical atomic displacements and magnetic moments from phonons and spin waves in the normal state can be determined from the effective force constants $K$ and $K_m$ and the gap values, $u = \sqrt{(N/K\lambda)V_q}$ and $m = \sqrt{(N/K_m\lambda_{sf})V_q}$, respectively.

The charge/spin wave state has low resistivity if the gap is complete over the FS, i.e. $N(E_F) = 0$. An electric field $E_x$, applied during a time $\tau$, accelerates free electrons until their velocity $\dot{x}$ becomes $eE_x/\tau m$, where $e$ and $m$ are the electron charge and mass, respectively. The velocity $v_x(k)$ changes from $\hbar k/m$ to $\hbar k/m+\dot{x}$ ($k$ is the momentum along $\vec{x}$), and the free electron band from $\hbar^2k^2/2m$ to $\hbar^2k^2/2m+\dot{x}$, as shown in fig. 2.

For a metal, with $E_F$ indicated by the horizontal line in fig. 2 occupied states near $k = +1$ $(kF)$ are pushed upward, while the states near $k = -1$ $(+kF)$ decrease their energy. The net velocity $\int v_x(k)dk \approx 2eF\dot{x}$ and the conductivity is approximately $e^2\tau Nv_F^2/m$, as from the Boltzmann equation. But the total kinetic energy $E_t = \int N(\epsilon)\epsilon d\epsilon$ is enhanced by the field, so the net current will decay through resistive dissipation when the field is switched off. A band insulator, with a gap at the zone boundary, will also have a higher $E_t$ from an applied field, and by switching off the field, $E_t$ decays back to its low value for the ground state.

However, if the free electron band without field (thin line in fig. 2) is shifted horizontally by the electric field to a new position (bold line), then $E_t$ will remain at the minimum, no decay is possible, so the state with the net current can persist. It is equivalent to moving the origin of $k$ to $-2m/\hbar \dot{x}$. The electron occupation of the gapped band will not be asymmetric as the bold broken line in fig. 2 but will be symmetric relative to the new origin as for the unshifted band. This can be achieved through phonons with slightly different momentum so that phonons and electrons have a common drift. (Two phonons with momentum $\pm P$ have energy $\hbar \Omega_P = \sqrt{K/M_A}\sin(Pa)$ and velocity $\pm a\sqrt{K/M_a}\cos(Pa)$, where $a$ is the real space periodicity. Together they have energy $2h\Omega_P$ and zero velocity. If the phonons change to $P + P$ and $-P + P$ (and $p << P$), there will be a net velocity, $V_p \approx 2a\sqrt{K/M_A}\sin(Pa)/pa = \dot{x}$. The phonon energy, $2h\Omega_P\cos(pa)$, will not increase, since $\cos(pa) < 1$.)

A common drift of electrons and phonons implies different lattice conduction along opposite directions relative to an imposed current, which also will be different for a state with no current. Effects on lattice and spin-wave dependent properties should be observable although very small.

From the selective $q$-dependence it can be shown that a weak magnetic field will destroy the pseudogap. As mentioned, one $\vec{q}$ generates the gap on the paramagnetic FS at $E_F$. A magnetic field, $H$, will split the FS into two, one for each spin ("up" or "down"), and $K_{up,down} = E_F \pm \mu_B H$. Two independent phonons are required for having the gaps optimally on two FS. One has a potential perturbation at $e^{-i(\vec{q}+\delta)\vec{x}}$, and the other at $e^{-i(\vec{q}+\delta)\vec{x}}$, where $\delta$ is determined by the band dispersion and $\mu_B H$. The sum of these two potentials is $2\cos(\delta \cdot \vec{x})e^{-i\vec{q}\vec{x}}$, and therefore, even if there is a modulation given by the cosine function,
the effective $\vec{q}$ remains the same and cannot fit optimal values for two FS. The resulting gaps do not appear at $E_F$ on the two spin-split bands, which will reduce the gain in energy. The energy difference, $D(H, T)$, between the kinetic energy for the gapped DOS with and without field, which is calculated as

$$D(H, T) = \int_{-\hbar\omega'}^{\hbar\omega'} e\tilde{N}(e)(f(e+H, T)+f(e-H, T)-2f(e, T))de$$

(10)

increases quadratically with the field amplitude $H < V_q$ for low and high $T$ ($\approx V_q$). This is because the thermal occupation can be made more efficiently if $E_F$ is closer to the DOS peak (on $N$) above the gap for "majority" and closer to the DOS peak below the gap in the "minority" states, than if $E_F$ is in the middle of the gap.

In conclusion, three mechanisms promote gaps at $E_F$. First, thermal normal state excitations of phonons/spin waves generate potential perturbations and weak band gaps near $E_F$. The total energy for the perturbed state is not lower than for the static state, but phonons can be softened. Second, systems with large $\lambda$ can spontaneously generate phonons/spin waves because of a gain in total energy, as has been described above for NFE bands. The third mechanism is through superconducting pairing. The equations for $T^*$ and $T_C$ are similar. Also conductivity and the effect of a magnetic field behave in similar ways. Calculations show increased $\lambda$ for low doping, which would agree with the evolution of $T^*$. But the superconducting $T_C$, which is maximal at a doping of 0.12-0.15 holes/Cu, does not follow the doping dependence of $\lambda$. The shape of the FS and differences in the $k, k'$-dependence of $\lambda$ (nesting etc.) are probably important for which state will win. In 3-D it is hard to imagine that a multitude of density waves can coexist to make a full gap everywhere on the FS sphere. For 2-D cuprate bands it is simpler since $\lambda$ is only large for k-points near X and Y, where a pseudogap can appear. These results support the picture that the pseudogap competes with superconductivity, and that it seems to depend on preformed pairs between $k$ and $-k$ states.

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