Long-range Coulomb interactions and the onset of superconductivity in the high-T_c materials

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(Received 19 September 1988; revised manuscript received 11 August 1989)

The Ginzburg-Landau functional for a superconductor is extended to include a quantum-fluctuation term arising from imperfect screening of the long-range Coulomb interaction. At low temperatures the resulting quantum x-y model shows a second-order phase transition between a superconducting state and an insulating state as a function of the ratio of the phase stiffness to the Coulomb energy measured on the scale of the mean pair spacing. By relating the functional formulation to a BCS-type model of high-temperature superconductivity in the strongly correlated regime, we show that the phase stiffness is proportional to doping away from the $\frac{1}{2}$-full Mott insulating state. We discuss application of the model as a mechanism for the onset of superconductivity of the CuO_2-based high-T_c materials above a critical doping level. Transport and optical properties of materials with reduced transition temperature are calculated.

I. INTRODUCTION

In this paper we address the physics of the insulator-superconductor transition observed in a variety of materials of the $La_{2-x}Sr_xCuO_4$ (2:1:4) and $YBa_2Cu_3O_{7-y}$ (1:2:3) classes. The phase diagram obtained from measurements in both of these classes of superconductors shows that they are well-defined Mott insulators at a composition corresponding to a nominal one hole per copper in the CuO_2 planes ($x=0$ for 2:1:4, $y=1$ for 1:2:3) and remain insulating up to a critical hole-doping concentration of about $x=5\%$ for 2:1:4 (Ref. 1) and $y=0.5$ for 1:2:3.[2] A similar phenomenon seems to occur for the recently discovered T' class $Nd_{2-x}Ce_xCuO_4$, which may be interpreted as an electron-doped Mott insulator.[3]

Our purpose in this paper is to explore the consequences of the hypothesis that Cooper pairing between the charge carriers is the dominant energy in the problem, and furthermore that it persists even in the insulating regime below the critical doping threshold for superconductivity (which we note by δ_c).

In this limit, localization (i.e., the fact that the system develops a gap for charge fluctuations) is then a consequence of competition between the suppression of pair density fluctuations due to the long-range Coulomb repulsion on a length scale longer than an effective Cooper-pair radius, and the tendency for long-range phase coherence between pairs that we express in terms of a Ginzburg-Landau phase stiffness parameter. Our approach is based on a phenomenological description in which we assume the existence of an underlying periodicity with length scale a_0 for the idealized homogeneous system.

In Wigner's original argument for electron localization, the length scale for localization is basically the carrier spacing. This can be traced back to the fact that electron-electron repulsion is minimized with only one electron per unit cell, in contrast to charge-density-wave formation in more complex materials where Fermi-surface nesting sets the localization length scale. For our case we make the assumption that carrier repulsion on the scale of a lattice spacing has been converted to a pairing attraction by strong correlation effects, but that the Coulomb energy on the scale of the spacing between pairs remains repulsive. Thus we take a_0 to represent the mean pair spacing, hence to vary as $\delta^{-1/2}$. In practice, a nonuniform potential associated with the dopants (Sr^{2+} in the case of 2:1:4 or chain oxygens for the 1:2:3) will tend to lead to random pinning of the pair localized state, which may be sufficient to obscure the breaking of translational symmetry. Nevertheless, our basic hypothesis is that the pairing energy is stronger than the energy to localize a single carrier, so that it is energetically more favorable to localize one pair at a doping defect than to localize two individual carriers at two separate defects.

Our hypothesis is to be contrasted with alternative scenarios for the insulator-superconductor transition that have been proposed such as conventional single-particle localization,[4] the idea that magnetic-mediated pairing only switches on when Néel order disappears,[5] or the idea that some kind of exciton-mediated pairing requires a finite hole concentration to become attractive.[6] We do not attempt a microscopic justification of our approach, although we show that the underlying physics is similar to that derived on the basis of the negative-U Hubbard model by Robaszkiewicz *et al.*[7]

In our picture, the disappearance of superconductivity below the critical doping concentration, δ_c does not involve pair breaking as it would in phonon-mediated superconductors where the Bardeen-Cooper-Schrieffer

Schrieffer (BCS) coherence length $\xi_{\rm BCS}$ is in general longer than the mean free path for the carriers, but rather that it is associated with the loss of long-range superconducting phase coherence as the Coulomb energy to suppress charge fluctuations overcomes the tendency to long-range superconducting order maintained by the Ginzburg-Landau phase stiffness parameter. Thus the transition temperature for onset of superconducting phase coherence, T_c, will not, in general, coincide with the temperature below which Cooper pairing occurs, which we denote by $T_{\rm BCS}$.

One of our principal results is to show that the phase stiffness parameter is roughly proportional to doping concentration, δ, within a mean-field BCS theory for superconductivity. Consequently, the tendency to charge localization takes place progressively as δ is reduced, with corresponding reduction of the superconducting transition temperature T_c, till, for $\delta<\delta_c$ the system enters an insulating phase with an energy gap for charge excitations.

Our model is formulated in terms of a path integral with a generalized Ginzburg-Landau (GL) action in which the effect of long-range Coulomb interactions is represented by a time-dependent term. This leads to quantum fluctuations of the GL order parameter, and the insulator-superconductor transition becomes a second-order phase transition at zero temperature. Consequently, we expect to see critical fluctuation effects close to δ_c in which the fluctuations (on the superconducting side) correspond to islands of pair-localized state on a length scale $\xi_{\rm loc}$ that diverges at δ_c. The existence of this length scale may help explain the sensitivity of CuO_2-based superconductors to neutral blockers of superconducting coherence such as Zn.[8] At finite temperatures, with δ close to δ_c the carriers remain paired below the pairing temperature, $T_{\rm BCS}$, but above the insulator-superconductor phase transition temperature, T_c, so that in an intermediate temperature range, both excited pairs and ionized single carriers will be present in a kind of hot plasma.

A further consequence of the existence of this mixed state will be the appearance of an optical absorption edge below the BCS pairing gap, $2\Delta_{\rm BCS}$, corresponding to excitation of charge fluctuations in the superconducting state. Because of imperfect screening due to the pairing, the usual Josephson plasma frequency will be reduced and, in fact, go to zero at the insulator-superconductor phase boundary. This in turn leads to the optical absorption threshold going to zero as $\delta\rightarrow\delta_c$, and to disappearance of the zero-frequency London term in the optical conductivity for $\delta<\delta_c$ when the system enters the insulating state.

II. A PHENOMENOLOGICAL GINZBURG-LANDAU ACTION WITH LONG-RANGE COULOMB INTERACTIONS

The derivation of a GL action from a microscopic model for fermion pairing involves integrating out the fermion degrees of freedom for fixed values of the local order parameter variables $\psi_i(\tau)$ inserted in the microscopic theory via a Hubbard-Stratanovich transformation. This procedure has been discussed in general terms by Efetov.[9]

The resulting functional includes time-dependent (i.e., quantum) fluctuations of the order parameter, and hence describes the response of the system in terms of collective bosonic degrees of freedom. This response is characterized in terms of the q and ω dependence of the kernel for the quadratic term in ψ in the resulting functional. For disturbances on a length scale shorter than the Ginzburg-Landau zero-temperature coherence length and time scale shorter than $\hbar/2\Delta_{\rm BCS}$ ($\Delta_{\rm BCS}$ the BCS gap), one expects efficient screening due to ordinary single-particle excitations. However, at long wavelengths and low frequencies, screening will be taken over by the collective degrees of freedom and the usual single-particle screening will be less effective owing to the pairing gap in the underlying fermion system. Thus the usual Lindhard function entering into the calculation of the q- and ω-dependent dielectric function will be strongly modified in this region of q and ω. To simplify the resulting analysis, we make a basic assumption that a single effective high-frequency dielectric constant ϵ_∞ may be defined to describe screening at short length and time scales. We then start from a phenomenological Ginzburg-Landau action of the form

$$\mathcal{F}_{\rm GL}=\int_0^\beta d\tau\int d^3x\tfrac{1}{2}\left[C_{\alpha\beta}|\nabla_\alpha\psi\nabla_\beta\psi|+a|\psi|^2+b|\psi|^4+\left[\frac{\epsilon_\infty}{4e^2}\right]\left|\nabla\left[\hbar\frac{\partial\phi}{\partial\tau}\right]\right|^2\right], \tag{1}$$

where $\psi=|\psi|e^{i\phi}$ and the last term takes account of the Coulomb energy resulting from quantum fluctuations of the phase of the order parameter. In this description of the superconductor, screening effects at small q and ω are now included through the dynamics of the ψ field. As we will show in the following, this may respond either as a superconductor (with metallic screening) or as an insulator (with dielectric screening), depending on where one is in the phase diagram.

Our second basic assumption, discussed in the introduction is that there is a fundamental unit cell, length a_0 defined by the mean pair spacing, which exists in both the insulating and superconducting phases. We discuss this assumption further in relation to the negative-U Hubbard model in Sec. V.

We now put our model (1) onto a lattice of spacing a_0. Note that the Coulomb term is three-dimensional, while the Josephson coupling is of two-dimensional character (or highly anisotropic) due to weak tunneling between CuO_2 planes. The lattice version of (1) then naturally leads at $T=0$ to a Hamiltonian for the quantum GL order parameter of the form of a quantum xy model:

$$H_{\rm GL}=-J\sum_{\langle ij\rangle}\cos(\phi_i-\phi_j)+\frac{4e^2 1}{\epsilon_\infty d}\frac{1}{2}\sum_{{\rm all}\,i,j}C_{ij}^{-1}\left[i\frac{\partial}{\partial\phi_i}i\frac{\partial}{\partial\phi_j}\right], \tag{2}$$

where the inverse capacitance matrix C_{ij}^{-1} is the lattice version of $a_0/|R_{ij}|$ given by

$$C_{ij}^{-1}=\sum_q \frac{e^{i\mathbf{q}\cdot\mathbf{R}_{ij}}}{2(3-\cos q_x a_0-\cos q_y a_0-\cos q_z d_0)}, \quad (3)$$

where the sum is restricted to the first Brillouin zone and d_0 is the lattice spacing normal to the CuO_2 planes.

In (2) the amplitude fluctuations of the order parameter inherent in the original functional (1) are suppressed and all the dynamics is in the phase fluctuations. The model is thus reduced to that of a granular superconductor,[10] although here it is to be understood that the "grains" are purely intrinsic and define degrees of freedom of the postulated Cooper-pair-localized state. The appropriate Josephson coupling parameter is given by

$$J=Cd_0|\psi|^2 \quad (4)$$

where C is the Ginzburg-Landau "phase stiffness" parameter, and we have replaced the gradient by a finite difference ∇_{ij}/a_0 and introduced a volume element $a_0^2 d_0$ for the unit cell.

Coupling to an electrostatic field $\Phi(x)$ may be introduced by recognizing that

$$\rho(x)=\sum_i \delta(x-x_i)\frac{2e}{\epsilon_\infty}i\frac{\partial}{\partial\phi_i} \quad (5)$$

represents the charge fluctuation-density operator for the pair-localized state so that the coupling term becomes

$$H_{\rm int}=\int d^3x\, \Phi(x)\rho(x)\ . \quad (6)$$

By imposing Poisson's equation on the internal part, $\Phi_{\rm int}$, of the electrostatic potential

$$-\nabla^2\Phi_{\rm int}=2e\rho(x)$$

we can take care of the long-range part $C_{ij}^{-1}\approx 1/R_{ij}$ of the Coulomb interaction, and rewrite the model in terms of a separation of the total Coulomb energy into an intracell term, or short-range part and a long-range part. This leads to a quantum xy model with on-site and nearest-neighbor terms only

$$H^0=-J\sum_{\langle ij\rangle}\cos(\phi_i-\phi_j)+\frac{4e^2}{\epsilon_\infty a_0}\sum_i C_{ii}^{-1}\left[i\frac{\partial}{\partial\phi_i}\right]^2 \quad (7)$$

together with a long-range Coulomb interaction included via (6).

The physics of the quantum xy model has been explored extensively, although mostly on the context of a short-ranged (i.e., screened) inverse capacitance matrix C_{ij}^{-1} (see, however, work of Fisher[11] and Fisher and Grinstein[12]). The long-range nature of C_{ij}^{-1} will have important consequences for the predicted optical properties (see Sec. IV). We show in Sec. III, however, that it will not alter the qualitative physics of the insulator-superconductor transition at $T=0$, or of the general nature of the phase diagram.

The effect of the short-range part of the Coulomb term is to lead to quantum fluctuations in the long-range phase coherence of the superconducting order, which are sufficient to disrupt long-range order above a critical value. For the model of Eq. (7), the superconductor-insulator instability occurs at the mean-field value J_c defined by[17]

$$r=\frac{4e^2}{\epsilon_\infty a_0}\left[\frac{4e^2}{J_c\epsilon_\infty a_0}-2\right]=0 \quad (8)$$

given in terms of the diagonal part C_{ii}^{-1} of the Coulomb interaction. Above the critical value $(4e^2/\epsilon_\infty a_0 J_c)=2$ of the ratio of the Coulomb energy to Josephson coupling, the long-range phase order parameter $\langle \exp(i\phi)\rangle$ goes to zero and the weight of the London superfluid term in the electromagnetic response $\sigma(\omega)\propto\delta(\omega)$ also goes to zero, so that the response becomes that of an insulator. (As already mentioned, we assume that the pair-localized state is pinned so that collective charge-density-wave conduction of Fröhlich type is suppressed.)

For $J>J_c$, the system remains a superconductor where, for J close to J_c, the weight of the δ function in the electromagnetic response is reduced and goes to zero at $J=J_c$. In this regime the system behaves as a metal and screens out an applied static electric field on the length scale of the lattice spacing a_0. At finite frequencies a longitudinal electric field can in principle penetrate the superconductor in the limit of zero interplane tunneling. (See the discussion in Sec. IV.)

In order to relate this instability to an underlying microscopic model for the CuO_2-based superconductors, we use the mean-field approach based on a superexchange type of electron-electron interaction. Strong on-site Coulomb correlations are taken care of via a slave-boson representation treated in a mean-field limit. The phase-stiffness parameter may then be computed from an expansion of the appropriate Cooper-pair correlation functions in powers of q. For a superconductor at $T=0$ this involves calculating the effect of a distortion $\delta\psi(x)$ from the ground-state broken symmetry order parameter ψ_0. This has been conveniently summarized by Kleinert.[13] A detailed calculation is discussed in Appendix A. To get a feeling for the doping dependence of the phase stiffness at this stage, we consider the diagonal contribution[13]

$$J\approx\lim_{q\to0}\frac{\partial}{\partial q^2}L_{11}(q)\ , \quad (9)$$

where

$$L_{11}=\sum_k \frac{E_kE_{k+q}+\varepsilon_k\varepsilon_{k+q}}{E_kE_{k+q}(E_k+E_{k+q})} \quad (10)$$

and E_k are the quasiparticle energies in the BCS theory.

We first consider the simplest mean-field approximation of Basaran *et al.*[14] and Ruckenstein *et al.*[15] In this approximation the E_k have the form

$$E_k=[\varepsilon_k^2+(I\tau_k\Delta)^2]^{1/2}\ , \quad (11)$$

where $\varepsilon_k=(\langle b^2\rangle\varepsilon_k^0-\mu)$, $\langle b^2\rangle$ is the slave-boson mean-field expectation value, $\langle b^2\rangle=\delta$ (hole concentration),

$$\tau(k)=2[\cos(k_xa)+\cos(k_ya)]\ ,$$

Δ is the BCS gap parameter, and I is the pairing interaction. For small δ, this form of the strongly correlated electron model has the property that the mean-field gap,

Δ_0 is independent of δ. This is because at very small carrier concentration, the kinetic energy term in the BCS equation $\to 0$ and all the pairing comes from the potential term. It then turns out that the phase-stiffness response (10) becomes singular in the $\delta \to 0$ limit for this model. This may readily be seen from (11) since $E_k = -I\Delta|\tau_k|$ for occupied states in this limit. Then

$$L_{11}|_{\delta=0} \approx \frac{1}{I\Delta} \sum_k \frac{1}{|\tau_k| + |\tau_{k+q}|} \approx \frac{1}{I\Delta} \ln \left[\frac{1}{qa_0} \right] . \quad (12)$$

In order to achieve a physically sensible result for small δ, it is necessary to take account of the fact that the strongly correlated Hubbard model has long-range antiferromagnetic order at low doping, at least in a mean-field limit.[16] In this case the kinetic energy part of E_k is split by a k-independent antiferromagnetic gap (see Appendix A). This removes the divergence in Eq. (12). Using

$$\left. \frac{\partial}{\partial q^2} E_{k+q} \right|_{q=0} = \frac{2[\varepsilon_k \langle b^2 \rangle 2t + \tau_k (I\Delta)^2]}{E_k} \left. \frac{\partial \tau_{k+q}}{\partial q^2} \right|_{q=0} \quad (13)$$

and the fact that $\Delta \propto \sqrt{\delta}$ for small δ in the presence of antiferromagnetic order, we see that the phase-stiffness parameter J [Eq. (9)] is proportional to δ for small δ. An equivalent way of saying this is to remark that the Ginzburg-Landau coherence length in BCS theory varies as $\xi_{\rm GL} \approx v_F / \Delta_0$ with Δ_0 the BCS gap.[13] In this case this becomes modified by replacing Δ_0 with the magnetic gap and adding an additional contribution from the k dependence of the gap $\Delta(k) \equiv \Delta\tau_k$ in Eq. (13). Since both v_F and Δ vary as $\sqrt{\delta}$, this leads to $\xi_{\rm GL}^2 \propto \delta$.

As a function of hole concentration the stability criterion (8) then shows that the system will remain localized at small doping up to a critical value δ_c depending on the strength of the effective high-frequency dielectric constant ϵ_∞. To make contact with experiment we fit ϵ_∞ by assuming that the experimental critical doping ($\approx 5\%$) is given from (8). $J(\delta)$ is calculated numerically using the extended mean-field theory of Inui *et al.*[16] At finite temperature, we use the relation $J \propto |\psi|^2$, where $|\psi|$ is the Ginzburg-Landau order parameter and assume a linear relation $|\psi|^2 \simeq (1 - T/T_{\rm BCS})$ to derive

$$J_c(T) = J_0(\delta)(1 - T/T_{\rm BCS}) = \frac{e^2}{\epsilon_\infty 2a_0} , \quad (14)$$

which defines the insulator-superconductor phase boundary $T_c(\delta)$. The results are shown in Fig. 1. As δ increases, the relation between J_0 and δ rapidly becomes nonlinear and $T_c(\delta)$ approaches $T_{\rm BCS}$ for $\delta \simeq 0.15$.

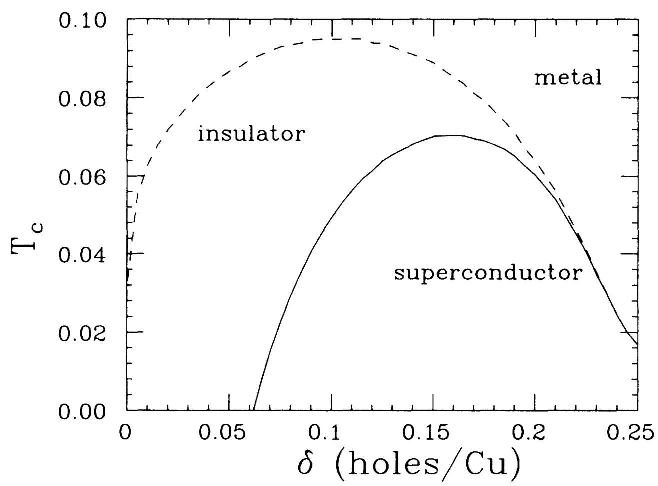

FIG. 1. Phase diagram [from Eq. (12)].

III. EFFECTIVE-FIELD THEORY IN THE REGION OF THE INSULATOR-SUPERCONDUCTOR TRANSITION

In this section we discuss the effects of the long-range part of the Coulomb interaction on the insulator-superconductor phase transition already formulated in terms of the nearest-neighbor quantum x-y model. To do this we make a further coarse graining following earlier work[17] by introducing a coarse grained complex order parameter $\chi(r,\tau)$ that measures a local average of $\langle \exp(i\phi) \rangle$. We then perform an expansion about a mean-field saddle point based on the dominant, diagonal short-ranged part of the Coulomb interaction, C_{ii}^{-1}. In the presence of an electrostatic potential Φ the resulting free-energy functional may be written in gauge-invariant form as

$$\mathcal{F} = \int_0^\beta d\tau \int d^3x \left[r|\chi|^2 + u|\chi|^4 + C|\nabla_\perp \chi|^2 + d \left| \left[\partial_\tau - \frac{2e}{\epsilon_\infty} \Phi \right] \chi \right|^2 \right] , \quad (15)$$

where $r = \varepsilon_C(\varepsilon_C / ZJ - 2)$, $C = (\varepsilon_C a_0^2 / 3ZJ)$, $u = (ZJ)^2 / 6\varepsilon_C$, $d = 2/\varepsilon_C$, and Z is the number of nearest neighbors, and $\varepsilon_C = 4e^2/\epsilon_\infty a_0$. By including the internal electrostatic potential in Φ,

$$\Phi(x,\tau) = \Phi_{\rm int} + \Phi_{\rm ext} , \quad (16)$$

where

$$-\nabla^2 \Phi_{\rm int} = 4\pi\rho_{\rm int} , \quad (17)$$

and

$$\rho_{\rm int}(x,\tau) = \left[2e \left[\chi^* \frac{\partial \chi}{\partial \tau} - \frac{\partial \chi^*}{\partial \tau} \chi \right] + 2e\Phi\chi^*\chi \right] \quad (18)$$

is the appropriate gauge-invariant charge (fourth component of the current operator), we have included the effects of long-range Coulomb interactions as already discussed.

The effects of these long-range interactions on the physical properties in the localized regime, $J < J_c$, where J_c is a renormalized critical phase stiffness parameter, may be understood by calculating the bare dielectric polarizability response function in the Gaussian approximation:

$$R^0_{\alpha\beta}(x,\tau)=\langle T\{\nabla^{-1}\rho_{\rm int}(x,\tau)\}_\alpha\{\nabla^{-1}\rho_{\rm int}(0,0)\}_\beta\}\rangle \ , \quad (19)$$

where α and β are cartesian suffixes. For $r>0$, the Gaussian boson modes are massive with energy

$$\Omega_k=\left[\frac{Ck^2+r}{d}\right]^{1/2} , \quad (20)$$

and the resulting response function may be written in terms of the boson propagator

$$g^0(x,\tau)=\langle T\{\chi(x,\tau),\chi^*(0,0)\}\rangle_0 \quad (21)$$

from which $g^0(k,\varepsilon_n)=1/(\varepsilon_n^2+\Omega_k^2)$, where ε_n is a Matsubara frequency $\varepsilon_n=2\pi nT$.

In terms of g, the response function is given by

$$R^0(q,\omega_n)=\frac{1}{q^2}\sum_{\varepsilon_n,k} g^0(k+q,\omega_n+\varepsilon_n)g^0(k,\varepsilon_n) \times(\varepsilon_n+\omega_n)\omega_n \ . \quad (22)$$

At $T=0$, the appropriate analytic continuation gives for the retarded propagator

$$R^0(q,\omega)=\frac{1}{q^2}\sum_k \frac{\omega(\Omega_k-\Omega_{k+q})}{4\Omega_k\Omega_{k+q}[\omega^2-(\Omega_k+\Omega_{k+q}+i\epsilon)^2]} \ . \quad (23)$$

As $q\to 0$, the numerator may be expanded in q to give

$$R^0(0,\omega)=\frac{c}{d}\sum_k \frac{-\omega}{4\Omega_k^2[\omega^2-(2\Omega_k+i\epsilon)^2]} \ . \quad (24)$$

For $\Omega_0=\sqrt{r/d}>0$, the dielectric response therefore has a gap of $2\Omega_0$, corresponding to excitation of a pair of Bose fluctuations, so that the system is an insulator. The effect of the long-range part of the Coulomb interaction may now be understood qualitatively by thinking of the system as a medium of polarizable oscillators. Neglecting local-field effects, the long-wave dielectric constant may then be written

$$\epsilon(0,\omega)=1-4\pi\left[\frac{4e^2}{\epsilon_\infty}\right]R^0(0,\omega) \ ,$$

which, in the limit of large r, we can expand in Ck^2/r to give

$$\epsilon(0,\omega)\simeq 1+\frac{4\pi n^0}{m}(4e^2/\epsilon_\infty)\frac{1}{(2\Omega_0^2)-\omega^2} \ . \quad (25)$$

where n_0 is the density of oscillators, $n_0=(1/V)\times\sum_{k<k_{\rm max}}1$, where V is the volume. As $J\to J_c$, $r\to 0$ and the dielectric response will diverge. In terms of diagrams, the preceding approximation is equivalent to an RPA summation of polarization bubbles for the dielectric response function

$$\epsilon^{-1}(q,\omega)=1+\frac{4\pi(4e^2/\epsilon_\infty)R^0(q,\omega)}{1-4\pi(4e^2/\epsilon_\infty)R^0(q,\omega)} \ . \quad (26)$$

Calculation of leading (one-loop) corrections to the boson mass term will now involve polarization diagrams in addition to the short-range part coming from the $u\chi^4$ term (Fig. 2).

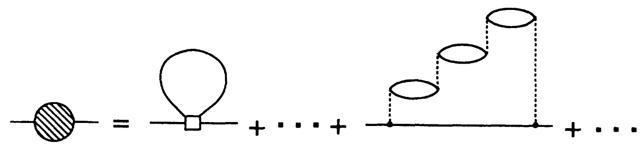

FIG. 2. Leading contributions to boson self-energy.

Thus we do not expect a basic change in the physics as a result of the long-range part of the interaction, although the scaling character of critical fluctuations close to J_c may well alter as in classical critical systems with long-range interactions.

VI. DIELECTRIC RESPONSE AND OPTICAL ABSORPTION IN THE SUPERCONDUCTING STATE

For $J>J_c$, on the other hand, $r<0$, and the system develops long-range superconducting phase order with mean-field order parameter

$$\chi_0=\sqrt{|r|/2u} \ . \quad (27)$$

In order to calculate the electromagnetic response of the system it is now important to distinguish between longitudinal and transverse excitations. It then turns out that the model field theory introduced in (7) is not adequate to deal with the long-range Coulomb effects in the superconducting phase. To understand this, it is necessary to go back to the quantum x-y model of Eq. (2). Let us consider the system far into the superconducting regime. Then the long-range phase coherence is well established and one can expand the cosine to give a harmonic "phason" model

$$H_{\rm phason}=-\sum_{{\rm all}\ ij} C_{ij}^{-1}\frac{4e^2}{a_0\epsilon_\infty}\frac{\partial}{\partial\phi_i}\frac{\partial}{\partial\phi_j}+\frac{J}{2}\sum_{\langle ij\rangle}(\phi_i-\phi_j)^2 \ . \quad (28)$$

It is important to remember that the first term is three-dimensional, while the second is essentially two-dimensional.

Expanding in phason modes

$$\phi_q=\frac{1}{\sqrt{N}}\sum_q e^{i\mathbf{q}\cdot\mathbf{R}_i}\phi_i, \quad \pi_q=\frac{i}{\sqrt{N}}\sum_q e^{-i\mathbf{q}\cdot\mathbf{R}_i}\frac{\partial}{\partial\phi_i} \quad (29)$$

we see that the long-range form of C_{ij}^{-1} leads to a set of phason modes where

$$\phi_q=\sqrt{(1/2\alpha_q)}(b_q+b_{-q}) \quad (30)$$

with $\alpha_q^2=(J\epsilon_\infty a_0/4\pi e^2)q_\perp^2 q^2$ and

$$H_{\rm phason}=\sum_q \Omega_J(q)(b_q^\dagger b_q+\tfrac{1}{2}) \ , \quad (31)$$

where

$$\Omega_J(q)=\left[\frac{4\pi e^2}{a_0\epsilon_\infty}J\frac{q_\perp^2}{q_\perp^2+(q_z d_0/a_0)^2}\right]^{1/2} \quad (32)$$

are a set of Josephson plasma frequencies appropriate to layered materials (i.e., the long-wavelength regime for the plasmon modes derived by Fetter[18]).

These modes couple [via $\Phi_{\rm ext}$ in (6)] to the longitudinal part of the electromagnetic field. For $q_z=0$ the Goldstone modes are pushed up to finite frequency as pointed out many years ago by Anderson.[19] However, the plasma branch for finite q_z has $\Omega_J \propto q_\perp$ in the limit of zero interplane tunneling, so it stretches all the way down to zero frequency. For real layered superconductors, however, there would be a threshold plasma frequency

$$\Omega_J|_{\rm min}=(\xi^z_{\rm GL}/\xi^\perp_{\rm GL})^2\Omega_J(q=0)$$

that may be considerably reduced from the bulk value.

To deal with the effects of Coulomb-induced fluctuations on the superconducting phase order, which one may think of as spontaneous creation and annihilation of vortex loops in 2+1 dimensions, the effective-field theory (7) needs to be extended in the superconducting phase. Once a long-range superfluid order parameter has been established (i.e., for $J>J_c$), the effects of the long-range part of the Coulomb interaction must be included by an additional time-dependent term of the form postulated in Eq. (1) that acts on the phase variable of the coarse grained order parameter $\chi(r,\tau)$.

To do this, we work to quadratic order in fluctuations about the mean-field minimum phase coherent superfluid order parameter χ_0:

$$\chi(\mathbf{r},\tau)=\chi_0 e^{i\phi(\mathbf{r},\tau)}[1+\eta(\mathbf{r},\tau)] , \tag{33}$$

where ϕ and η are purely real fields.

A reasonable form for an effective-field theory in the superconducting phase may then be written as

$$\mathcal{F}=\int_0^\beta d\tau \int d^3x \left[2|r|\chi_0^2\eta^2+C\left|\left[\nabla_\perp-\frac{e}{c}\mathbf{A}\right]\chi\right|^2+d\chi_0^2\left|\frac{\partial\eta}{\partial\tau}\right|^2+\frac{\epsilon_\infty}{4e^2}\left[\left|\nabla_\perp\hbar\frac{\partial\phi}{\partial\tau}\right|^2+\left|\frac{d_0}{a_0}\frac{\partial}{\partial z}\hbar\frac{\partial\phi}{\partial\tau}\right|^2\right]\right] , \tag{34}$$

where we have introduced an external electromagnetic field $\mathbf{A}$ in transverse gauge $\nabla\cdot\mathbf{A}=0$.

In effect, the model of (34) supposes that the time dependence of the amplitude part of the order parameter χ is given in terms of the short-range part of the Coulomb interaction, while the time dependence of the phase of the order parameter is dominated by the long-range part. We give a further discussion of this postulated form, based on perturbation theory, in Appendix B.

Given this model form, it is straightforward to calculate the optical response function for the system in the superfluid state. Writing the conductivity in terms of a diamagnetic and paramagnetic part [cf. Schrieffer (Ref. 20, Chap. 8)] we have

$$K_{ij}(q,t)=\mathcal{R}_{ij}(q,t)+\left[C\chi_0^2\frac{4\pi e^2}{c^2}\right]\delta(t)\delta_{ij} , \tag{35}$$

where

$$\mathcal{R}_{ij}(q,t)=-i\Theta(t)\langle[j_i(q,t),j_j(q,0)]\rangle \tag{36}$$

and

$$\mathbf{j}(q,t)=\int d^3r e^{i\mathbf{q}\cdot\mathbf{r}}(\chi^*\nabla\perp\chi-\chi\nabla\perp\chi^*) . \tag{37}$$

In (35), the London term, proportional to the superfluid density, goes to zero at the superconductor-insulator phase transition. Substituting (33), we see that the phase-only modes couple purely longitudinally to the electromagnetic field, hence do not contribute to the optical absorption. On the other hand, the mixed term

$$\mathbf{j}^T(x)=2i\chi_0\eta(x)\nabla_\perp\phi(x) \tag{38}$$

will couple transversely producing an absorption band formed from a phason together with an amplitude fluctuation (see Fig. 3)

$$\mathcal{R}^T_{ij}(q,t)=\left[1-\frac{q_iq_j}{q^2}\right]\sum_k g_k^\phi(t)g_{k+q}^A(t) , \tag{39}$$

where

$$g_k^\phi(t)=\frac{k_\perp^2}{(\epsilon_\infty/4e^2)(k_\perp^2+k_z^2)}\frac{\exp(i\Omega_k^Jt)}{2\Omega_k^J} , \tag{40}$$

$$g_k^A(t)=\frac{\exp(i\Omega_k^At)}{2\Omega_A d} , \tag{41}$$

and

$$\Omega_k^J=[C\chi_0^2(4\pi e^2/\epsilon_\infty a_0)k_\perp^2/(k_\perp^2+(k_zd_0/a_0)^2]^{1/2} ,$$

$$\Omega_k^A=[(2|r|+Ck^2)/d]^{1/2} .$$

The optical conductivity is then given as

$$\sigma(\omega)={\rm Im}K(q=0,\omega)$$
$$=C\chi_0^2\frac{4\pi e^2}{c^2}\delta(\omega)+\sigma^P(\omega) , \tag{42}$$

where

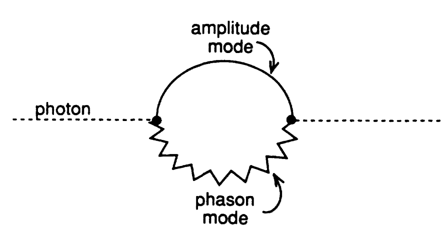

FIG. 3. Contribution to optical absorption in the superconductor.

$$\sigma^P(\omega)=\mathrm{Im}R(q=0,\omega)=\sum_{k_\perp,k_z}\frac{1}{4Cd}\frac{\Omega_k^J}{\Omega_k^A}\delta(\omega-\Omega_k^J-\Omega_k^A)\ . \tag{43}$$

Thus we do expect to see optical absorption below the BCS gap frequency, where the threshold frequency

$$\omega_0=\Omega^J|_{\min}+\Omega_0^A=\sqrt{2|r|/d} \tag{44}$$

will go to zero as $|r|\to 0$ and the amplitude of the superconducting order parameter, $\chi_0\to 0$. So in the region just above the critical doping concentration, where T_c is reduced below T_{BCS}, we expect to see a precursor absorption with threshold frequency scaling with the thermodynamic transition temperature, T_c. Expanding J about δ_c [from Eq. (12)] we see that the threshold absorption frequency then varies as

$$\omega_0 \propto T_c^{1/2}\ . \tag{45}$$

A numerical evaluation of the optical absorption (41) is plotted in Fig. 4 for a selection of values of $\delta>\delta_c$. The frequency scale is set by the basic unit $4e^2/\epsilon_\infty a_0$ of the phenomenological theory, which, however, bares no simple relationship to the BCS energy gap of an underlying microscopic model. The curves are computed using an arbitrary choice of model parameters and are given for illustration purposes only.

V. RELATIONSHIP TO THE NEGATIVE-U HUBBARD MODEL

The basic thesis of this paper is the idea that the competition between short-range effective attraction between fermions and longer-range repulsion can lead to a transition between a superconducting state and a pair-localized state that breaks translational symmetry. Although we have derived the phenomenological phase-stiffness parameter from a mean-field treatment of the strongly correlated Hubbard model, we have not attempted to justify the localization hypothesis starting from a microscopic model. One can, however, make an analogy with the negative-U Hubbard model by considering an idealized system in which a set of strontium acceptors (for the case of 2:1:4 compounds) form a regular superlattice with period $a_0/\sqrt{2}$. We may then suppose that the added holes form a set of impurity bands with Wannier orbitals built out of the hole states of the original, underlying Hubbard model. If we now restrict ourselves only to the lowest superlattice band (i.e., average over all details on a length scale $\ll a_0/\sqrt{2}$), the resulting model Hamiltonian may be written as a negative-U model with general form

$$H_{\text{superlattice}}=-t_{\text{eff}}\sum_{\langle ij\rangle,\sigma}c_{i\sigma}^\dagger c_{j\sigma}-U_{\text{eff}}\sum n_{i\uparrow}n_{\downarrow}+W_{\text{eff}}\sum_{\langle ij\rangle,\sigma,\sigma'}n_{i\sigma}n_{j\sigma'}\ , \tag{46}$$

where we neglect the Coulomb repulsion beyond the first neighbor cell and U_{eff} is an appropriate average over the superexchange coupling of the original, underlying Hubbard model. Since one additional hole per Sr is added, the superlattice band is $\frac{1}{2}$ full.

Clearly this approximation would not represent the kinetic energy for an uncorrelated system since the true band width for the holes is much wider than the superlattice band width. However, it does simulate the correlated band in the mean-field slave-boson model band, since there the band width $\simeq\delta(8t)$, and $\delta=(a/a_0)^2$ where a is the lattice spacing of the original Hubbard model. Hence (46) may not be such a bad description for the system close to $\frac{1}{2}$ filling of the underlying model.

We may now use the results of studies of the negative-U Hubbard model to discuss properties of (46). Following Robaszkiewicz *et al*;[7] we can map the charge degrees of freedom of (46) onto spin degrees of freedom of a transformed system in which down-spin particles are replaced by down-spin holes. In the strong-coupling limit $t_{\text{eff}}/U_{\text{eff}}\ll 1$, this becomes an anisotropic Heisenberg model:

$$H_{\text{eff}}=-J_{\text{eff}}\sum_{\langle ij\rangle}(S_i^xS_j^x+S_i^yS_j^y)+W_{\text{eff}}\sum_{\langle ij\rangle}S_i^zS_j^z\ . \tag{47}$$

At $\frac{1}{2}$ filling the external magnetic field (equivalent to a chemical potential in the original representation) is zero.

For $J_{\text{eff}}>W_{\text{eff}}$ the lowest state has $\langle S^x\rangle\neq 0$ and $\langle S^z\rangle=0$, equivalent to a superconducting state (SS). For $J_{\text{eff}}<W_{\text{eff}}$ the ground state is Néel-like with sublattice $\langle S^z\rangle\neq 0$ corresponding to a charge-density wave, or charge-ordered state (CO) with two lattice sites per unit cell.

A corresponding between the quantum Ginzburg-Landau model of Sec. II and the preceding model may be made by writing

$$e^{i\varphi_i}\leftrightarrow S_{i1}^+ + S_{i2}^+\ ,\qquad i\frac{\partial}{\partial\varphi_i}\leftrightarrow S_{i1}^z+S_{i2}^z\ , \tag{48}$$

where 1 and 2 refer to the two sites in the extended unit

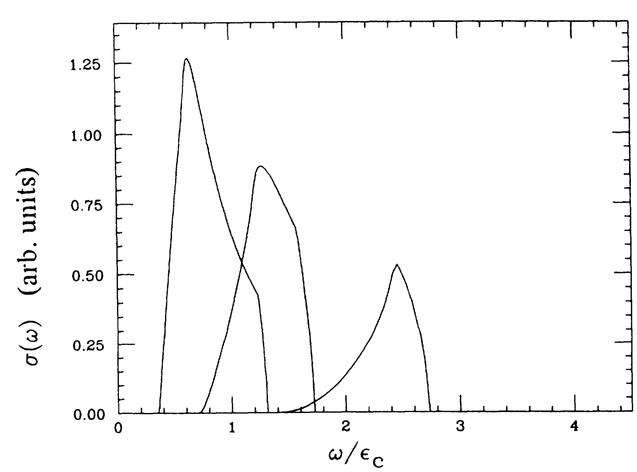

FIG. 4. Diagram representing contribution to intragap infrared absorption at three different values of the hole concentration, δ. Frequency is in units of $\varepsilon_C=4e^2/\epsilon_\infty a_0$.

cell, since the two sides of (48) have the same commutation relations.

In mean-field theory for the spin model, the transition between the SS and CO states is first order as a function of J/W. This is because of the constraint $(S_i^x)^2+(S_i^y)^2+(S_i^z)^2=\frac{3}{4}$. The analogous transition for the quantum rotor model is second order in mean field since this constraint does not apply in the latter model. For the anisotropic Heisenberg model it seems likely that the inclusion of zero-point fluctuations would also drive the transition second order.

While the preceding analogy helps in visualizing the physics of the insulator-superconductor transition, it should not be taken too seriously as it neglects the coupling to the degrees of freedom of the original Hubbard model for the CuO_2 planes from which the doping dependence of J (i.e., the phase stiffness) was derived in Sec. II.

VI. DISCUSSION OF PROPERTIES OF YBa_2CuO_{3-y}: PREFORMED PAIRS ABOVE T_c IN OXYGEN DEFICIENT SAMPLES

In this section we compare the qualitative physics expected from our model with available experimental data. There are many experimental indications that fully oxygenated Y-Ba-Cu-O has a BCS-like phase transition at $T_c \simeq 90$ K, albeit with $2\Delta/kT_c \simeq 6-8$.[21] However, oxygen deficient samples appear to show a number of anomalies. In particular the Pauli susceptibility appears to decrease with decreasing temperature below $T \simeq 200$ K (Johnston[22]) as does the Y Knight shift (Alloul *et al.*[23]).

Optical data of Thomas *et al.*[24] for Y-Ba-Cu-O appears to show a gap-like feature in the optical conductivity at $\omega \simeq 54$ meV whose energy is rather independent of oxygen concentration, but give indications of further absorption at lower frequencies, which is much more sensitive to oxygen deficiency.

Although we cannot claim to give a quantitative interpretation of the data, the above behaviors are qualitatively consistent with our phase diagram of Fig. 1 if it is assumed that the fully oxygenated sample lies close to the top of the T_c curve, i.e., where the charge fluctuation effects are small. On removal of oxygen, δ decreases and the system becomes an insulator: we then assume δ_c corresponds to $O_{6.5}$. For intermediate doping our model suggests $T_c < T_{BCS}$ and the appearance of optical absorption below the BCS gap. The onset of BCS pairing above T_c may be indicated by the susceptibility and Knight-shift data. It is also consistent with data on the nuclear relaxation rate, $1/T_1$, below $\simeq 100$ K comparing a $T_c = 90$ K sample and a $T_c = 60$ K sample,[25] which shows evidence of Cu-singlet formation well above T_c for the oxygen deficient sample.

Finally, as mentioned in the introduction, the sensitivity of both the 1:2:3 and 2:1:4 compounds to Zn doping may be interpreted in terms of closeness of the system to a second-order phase transition.

A quantitative inconsistency with our basic pairing hypothesis appears to be a lack of sharp transition associated with pair localization in the vicinity of the pairing temperature, T_{BCS} as seen in the lack of an abrupt change with temperature in the susceptibility or Knight-shift data. However, it seems possible that the randomness of distribution of the dopants (Sr^{2+} for 2:1:4 or oxygen vacancies for 1:2:3) may have the effect of smearing out this transition.

Equally puzzling at first sight is the absence of any upturn in dc resistivity above T_c on lowering the temperature for samples with $\delta > \delta_c$. However, the observed behavior may be understood qualitatively by noting that charge-density fluctuations, represented by time dependence of the GL order parameter in Eq. (13), will carry current. At temperatures $T > T_c(\delta)$ one therefore has a hot plasma of pseudo relativistic positive and negatively charged Bose particles with energy $\Omega_k = (k^2+r)^{1/2}$ and "mass" $\sqrt{r}$ which tends to zero as $T \to T_c$. Simultaneously, there will be thermally activated single fermions due to breakup of Cooper pairs. Evaluation of the mean boson density

$$n = \langle n_{\text{bosons}} \rangle = \sum_k \frac{1}{e^{\beta\Omega_k}-1} \tag{49}$$

for $r \propto (T-T_c)$ leads to a parameter-dependent function which increases roughly linearly with T provided the cutoff in the k sum is not too large, i.e., $Ck_{\max}^2 \approx (\varepsilon_C/2ZJ) = O(1)$.

The scattering cross section for charged bosons in the resulting classical plasma will be given in terms of the Debye screening length

$$a_D = (4\pi n e^2/\epsilon_\infty k_B T)^{-1/2}$$

which is roughly temperature independent for $n \simeq T$. The scattering rate may then be estimated in Born approximation via the scattering rate (in the center-of-mass frame) for two bosons:

$$\frac{1}{\tau_k} = 2\pi \sum_{k'} \left[\frac{4\pi e^2}{(k-k')^2 + a_D^{-2}} \right]^2 \delta(\Omega_k - \Omega_{k'}) , \tag{50}$$

leading to

$$\frac{1}{\tau} \approx \left[\frac{T}{n} \right]^2 \sum_k n_k \Omega_k . \tag{51}$$

Since $\langle \Omega_k \rangle \simeq T$, then provided one is in a parameter regime where $n \approx T$, the resistivity varies approximately as

$$\rho \approx \frac{\overline{m}}{ne^2\tau} \propto T , \tag{52}$$

where $\overline{m}$ represents an average over the effective mass, $\overline{m}^{-1} = \langle d^2\Omega_k/dk^2 \rangle$. Since our phenomenological Ginzburg-Landau approach is only valid close to δ_c, the above result is justifiable only for temperatures well below T_{BCS}.

For $\delta < \delta_c$, r remains finite at all T, and n drops to zero exponentially at low temperatures leading to a rapid increase in resistivity (see Fig. 5). Thus the resistance minimum observed for samples with low doping[26] may be interpreted in terms of freezing out of Cooper-pair fluctuations which lead to a finite conductivity at higher temperatures.

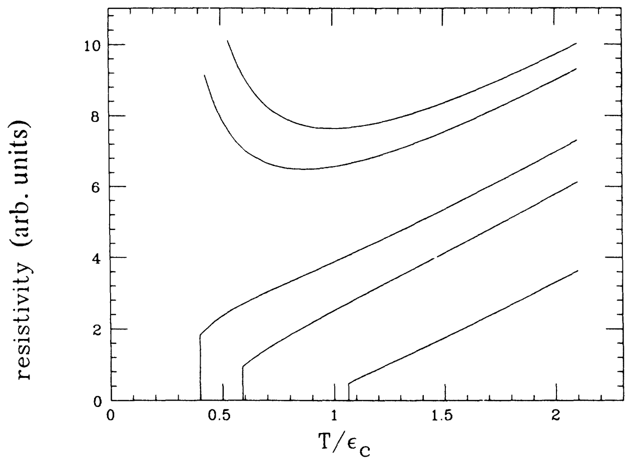

FIG. 5. Resistivity vs temperature for a variety of values of δ from Eqs. (49) and (50). [Upper curves: $(\delta/\delta_c-1)=-0.01,-0.001$; lower curves: 0.3, 0.5, 1.0]. Temperature is in units of ε_C.

VII. DISCUSSION

Our approach to the physics of the insulator-superconductor transition in the CuO_2-based materials is based on the hypothesis that pairing occurs even in the insulating state. Our main evidence to support his hypothesis is the observation that conventional lowering of T_c due to unbinding of Cooper pairs (e.g., due to magnetic impurities) does not seem to occur in these materials since this would be expected to lead to a normal-metal–superconductor transition as opposed to the insulator-superconductor transition universally observed in both the p-type and the n-type superconductors.

Our assumption of Cooper pairing in the insulating state implies that conduction at finite temperatures close to the critical concentration, δ_c, must take place by preformed pairs, i.e., by carriers of charge $2e$ rather than e. We have argued that this is not in qualitative conflict with currently available transport or optical data. Probably the best experimental test of this picture will be a more precise study of the intragap infrared absorption in materials close to δ_c such as oxygen-depleted Y-Ba-Cu-O.

On the other hand, our phenomenological approach has not taken into account the effects of randomness of dopants such as Sr^{2+} or oxygen vacancies, which will certainly modify the quantitative details of our conclusions if included in a more comprehensive theoretical treatment.[27]

ACKNOWLEDGMENTS

We thank J. R. Schrieffer, A. I. Larkin, and C. Varma for helpful criticism. We are grateful to the National Science Foundation (NSF) and the U. S. Office of Naval Research (ONR) for support.

APPENDIX A: DOPING DEPENDENCE OF THE PHASE-STIFFNESS PARAMETER

In this appendix we derive the phase-stiffness parameter for the Ginzburg-Landau model of Eq. (1) using the extended mean-field theory for a Hubbard superconductor of Inui *et al.*[16] In this treatment, both BCS ordering and Néel ordering are allowed to occur simultaneously. For the real system, one expects magnetic frustration effects to remove the long-range order.[28] Nevertheless, short-range magnetic order is likely to persist so that the mean-field theory may be expected to give qualitatively reasonable answers for a local parameter like the phase stiffness.

The mean-field Hamiltonian of Inui *et al.*[16] may be written as

$$H_{\text{eff}}=\frac{1}{2N}\sum_{k,q}\psi^\dagger_{kq}M_{k,q}\psi_{kq} \tag{A1}$$

with $\psi_{kq}=(c_{k+\frac{q}{2}\uparrow},\, c_{k-Q+\frac{q}{2}\uparrow},\, c^\dagger_{-k+\frac{q}{2}\downarrow},\, c^\dagger_{-k+Q+\frac{q}{2}\downarrow})$ and

$\mathbf{Q}=(\pi/a_0)(\hat{\mathbf{x}}+\hat{\mathbf{y}})$ is the antiferromagnetic nesting vector.

The matrix $m_{k,q}$ is given by

$$M_{k,q}=\begin{bmatrix} e^+_{k+q/2} & s_{k+q/2} & p_{kq} & 0 \\ s_{k+q/2} & e^-_{k+q/2} & 0 & -p_{kq} \\ p^*_{kq} & 0 & -e^+_{k-q/2} & s_{k-q/2} \\ 0 & -p^*_{kq} & s_{k-q/2} & -e^-_{k-q/2} \end{bmatrix}, \tag{A2}$$

where

$$e^\pm_k=(t^2/U')\{\pm[\delta(U'/t)+(6\delta+2)r]g_1(k) -[\tfrac{1}{2}\delta g_2(k)+4]\bar{\rho}\}\ ,$$

$$p_{kq}=\sum_s p'_{kqs}\Delta_{qs} =\sum_s (t^2/U')[\delta g_5(k,q,s)+2g_4(k,s)]\Delta_{qs}\ ,$$

and

$$s_k=-(t^2/U')[\delta g_2(k)+8]m_z\ .$$

Here

$$g_1(k)=2(\cos k_x a+\cos k_y a)\ ,$$

$$g_2(k)=g_1^2(k)-4\ ,$$

$$g_3(k,l)=g_1(k)g_1(l)-g_1(k-l)\ ,$$

$$g_4(k,l)=g_1(k+l)+g_1(k-l)\ ,$$

$$g_5(k,l,s)=g_3(k+l/2,l/2-s)+g_3(k+l/2,l/2+s) +g_3(-k+l/2,l/2-s) +g_3(-k+l/2,l/2-s)\ ,$$

and $r=\langle c^\dagger_{i\uparrow}c_{i\uparrow}\rangle=\langle c^\dagger_{i\downarrow}c_{i\downarrow}\rangle$, $\bar{\rho}=\langle n_{i\uparrow}+n_{i\downarrow}\rangle$, $m_i=e^{i\mathbf{Q}\cdot\mathbf{R}_i}\langle n_{i\uparrow}-n_{i\downarrow}\rangle/2$,

$$\Delta_{qs}=\frac{1}{N}\sum\nolimits_{x,x'}\exp[-i(x+x')q/2-i(x-x')s]\Delta_{x,x'}\ ,$$

and $\Delta_{x,x'}=\langle c_{x\uparrow}c_{x'\downarrow}\rangle$.

We now follow Kleinert[13] to calculate the response of the ground state to a change of order parameter relative to the broken symmetry direction:

$$\Delta(x,x')=\Delta_0(x-x')+\Delta'(x,x') . \quad \text{(A3)}$$

Then we need the propagators of the generalized mean-field Hamiltonian (A1). To obtain these we perform a rotation on (A1) in order to diagonalize with respect to the antiferromagnetic coupling.

Defining

$$\mathcal{B}_{kq}=\begin{bmatrix} c_{k+q/2\uparrow}\cos\Theta_k+c_{k-Q+q/2\uparrow}\sin\Theta_k \\ c^{\dagger}_{k+q/2\downarrow}\cos\Theta_k-c^{\dagger}_{-k+Q+q/2\downarrow}\sin\Theta_k \\ -c_{k+q/2\uparrow}\sin\Theta_k+c_{k-Q+q/2\uparrow}\cos\Theta_k \\ c^{\dagger}_{-k+q/2\downarrow}\sin\Theta_k-c^{\dagger}_{-k+Q+q/2\downarrow}\cos\Theta_k \end{bmatrix}$$

this leads to

$$H_{\text{eff}}=\frac{1}{2N}\sum_{kq}\mathcal{B}^{\dagger}_{kq}\tilde{M}_{kq}\mathcal{B}_{kq} , \quad \text{(A4)}$$

with

$$\tilde{M}_{qk}=\begin{bmatrix} \bar{e}^{+}_{k+q/2} & p_{kq} & 0 & 0 \\ p^{*}_{kq} & -\bar{e}^{+}_{k-q/2} & 0 & 0 \\ 0 & 0 & \bar{e}^{-}_{k+q/2} & -p_{kq} \\ 0 & 0 & -p^{*}_{kq} & -\bar{e}^{-}_{k-q/2} \end{bmatrix} \quad \text{(A5)}$$

where

$$\bar{e}^{\pm}_{k}=(e^{+}_{k}/2)(1\pm\cos 2\Theta_k)+(e^{-}_{k}/2)(1\mp\cos 2\Theta_k)\pm s_k\sin 2\Theta_k$$

and

$$\sin 2\Theta_k=2s_k/[(e^{+}_{k}-e^{-}_{k})^2+4s^2_k]^{1/2} .$$

The Hamiltonian is now expressed in terms of a pair of 2×2 matrices involving BCS coupling within upper and lower antiferromagnetic bands, respectively. We now follow Kleinert to compute the dependence of the action on Δ' to quadratic order in terms of Green's functions given by the inverse of $\tilde{M}_{k,0}$. The relevant diagrams are given in Fig. 6. We have evaluated expressions $L_{\alpha\beta}$ corresponding to L_{11} in Eq. (10) numerically. The results were fitted to a quadratic form for small q in order to extract $\lim_{q\to 0}(\partial/\partial q^2)L_{\alpha\beta}$. On diagonalizing the resulting quadratic in Δ', we found the Ginzburg-Landau parameter C of Eq. (1) to vary linearly with δ for small δ, but to become strongly nonlinear for δ in the region of 15%.

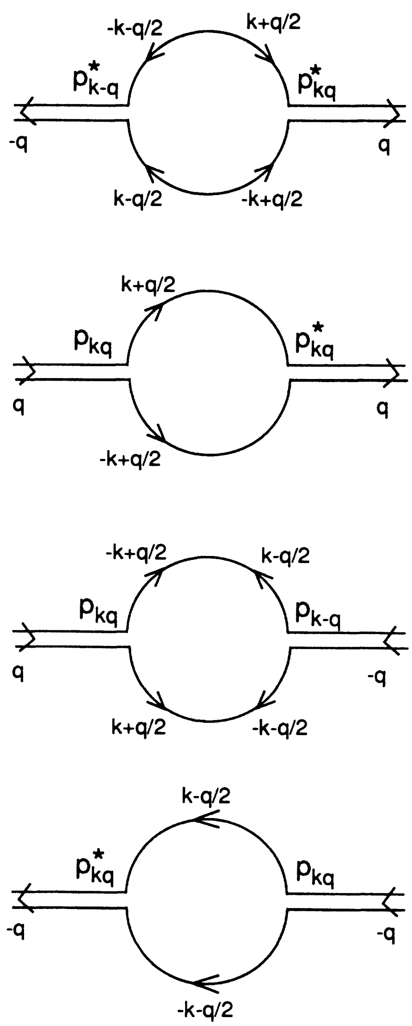

FIG. 6. Contributions to the response functions $L_{\alpha\beta}$.

APPENDIX B: OPTICAL ABSORPTION IN THE QUANTUM *XY* MODEL

In this Appendix we give some justification for the phenomenological model of Sec. IV by considering the effect of coupling to a transverse electromagnetic field on the basic quantum xy model of Eq. (2). The gauge-invariant coupling to an external vector potential may be put in the Josephson coupling term by replacing

$$-J\sum_{\langle ij\rangle}\cos(\phi_i-\phi_j)\to -J\sum_{\langle ij\rangle}\cos\left[\phi_i-\phi_j-\int_i^j \mathbf{A}\cdot d\mathbf{l}\right] . \quad \text{(B1)}$$

Then the corresponding current operator connecting sites i and j is

$$j_{ij}=Ja_0\sin(\phi_i-\phi_j) . \quad \text{(B2)}$$

In the phason limit of Eq. (26), this may be expanded in powers of $\nabla\phi$ to give

$$\mathbf{j}(r)=J[a_0\nabla\phi-\tfrac{1}{6}(a_0\nabla\phi)^3+\cdots] . \quad \text{(B3)}$$

On using this operator in the paramagnetic response function (35), the $\nabla\phi$ term contributes only to the longitudinal response and does not lead to optical absorption. So the leading contribution to the optical absorption is a three-phason term.

$$R^{(3)}_{\alpha\beta}=-i\Theta(t)\langle\{\tfrac{1}{6}[a_0\nabla_\alpha\phi(r,t)]^3,\tfrac{1}{6}[a_0\nabla_\beta\phi(0,0)]^3\}\rangle . \quad \text{(B4)}$$

Comparing this to the result derived from our phenomenological model (38), we see that the role of an amplitude fluctuation is played by a phason-pair propagator

$$\Pi_{\alpha\beta}=i\langle T\{[a_0\nabla_\alpha\phi(r,t)]^2,[a_0\nabla_\beta\phi(0,0)^2]\}\rangle \; . \qquad \text{(B5)}$$

Using the phason limit (28), this may be evaluated as

$$\Pi_{\alpha\beta}(\mathbf{k},t)=\delta_{\alpha\beta}\sum_{k_1} g^{\phi}_{k+k_1}(t)g^{\phi}_{k_1}(t) \; , \qquad \text{(B6)}$$

where the phason propagator is given by

$$\begin{aligned} g^{\phi}_k(t) &= \frac{i}{3}\langle T[\nabla_\perp\phi(r,t)\cdot\nabla_\perp\phi(0,0)]\rangle \\ &= k_\perp^2\left[\frac{4e^2/\epsilon_\infty a_0}{J}\frac{1}{k_\perp^2 k^2}\right]^{1/2}\exp(-i\Omega^{\phi}_k|t|) \\ &= \frac{\Omega^J_k}{J}\exp(-i\Omega^J_k|t|) \end{aligned} \qquad \text{(B7)}$$

and the plasmon mode frequency Ω^J_k is given by

$$\Omega^J_k=\Omega^J_0\left[\frac{k_\perp^2}{k_\perp^2+(d_0/a_0)^2k_z^2}\right]^{1/2} \qquad \text{(B8)}$$

with $\Omega^J_0=\sqrt{J}\,4e^2/\epsilon_\infty a_0$.

Comparing $\Pi(k,\omega)$ with $g^A(k,\omega)$ given in Eq. (40), we see that the amplitude spectral density with a δ response at mode frequency Ω^A_k is now broadened out into a continuum stretching down to zero frequency in the limit that interplane Josephson tunneling is zero. This means there is no finite threshold for optical absorption. However, on inspecting the spectral density of $\Pi(k,\omega)$ with positive frequency part given by

$$\text{Im}\Pi(k,\omega)=\sum_{k_1}\frac{\Omega^J_{k+k_1}\Omega^J_{k_1}}{J^2}\delta(\omega-\Omega^J_{k_1}-\Omega^J_{k+k_1}) \; , \qquad \text{(B9)}$$

we find that at frequencies $(\omega/\Omega^J_0)\ll 1$,

$$\text{Im}\Pi\propto\left[\frac{\omega}{\Omega^J_0}\right]^3 \qquad \text{(B10)}$$

so that the bulk of the spectral weight occurs in the vicinity of $2\Omega^J_0$. Hence in the phason limit, we can think of amplitude fluctuations of the effective order parameter $\langle\exp(i\phi)\rangle$ as occurring at twice the phason frequency. Then the main optical absorption will occur at an approximate threshold frequency $2\Omega^J_0$, even though there will be a weak tail stretching down to zero frequency. Including higher-order terms in the phason expansion of Eq. (2) will eventually renormalize the phason frequency to zero, leading to the loss of long-range phase coherence, so that the amplitude mode energy will go to zero and the threshold for strong intragap optical absorption will go to zero at the critical doping concentration δ_c.

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