Spontaneous superconductivity and optical properties of high-T_c cuprates

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(Received 15 June 2006; published 31 August 2006)

We argue that the high temperature superconductivity in cuprate compounds may be supported by interaction between copper-oxygen layers mediated by *in-plane* plasmons. The strength of the interaction is determined by the *c*-axis geometry and by the *ab*-plane optical properties. Without making reference to any particular in-plane mechanism of superconductivity, we show that the interlayer interaction favors spontaneous appearance of the superconductivity in the layers. At a qualitative level the model describes correctly the dependence of the transition temperature on the interlayer distance, and on the number of adjacent layers in multilayered homologous compounds. Moreover, the model has a potential to explain (i) a mismatch between the optimal doping levels for critical temperature and superconducting density and (ii) a universal scaling relation between the dc conductivity, the superfluid density, and the superconducting transition temperature.

The layered structure of the cuprates is a well established fact[1] related to a strong anisotropy in their electronic and optical properties.[2] The common feature of all cuprates—famous for their ability to exhibit superconductivity at high transition temperatures—is the presence of conducting CuO_2 layers separated by the so called charge reservoirs. These reservoirs are nearly insulating even in superconducting phase.[2]

One of the most fascinating features of high temperature superconductivity is a strong dependence of basic superconducting properties (including the most important quantity, the transition temperature T_c) on the *c*-axis structure of cuprates. In particular, there is a systematic dependence of the critical temperature on the number n of the closely packed CuO_2 layers per a structural *c*-axis unit. In typical homologous series of superconducting cuprates the separation between the n multilayers lies in the range $d \sim 6-15$ Å which is large compared to the spacing $c_{int} \approx 3.5$ Å separating individual layers inside the multilayer. Since c_{int} is numerically close to the in-plane Cu-O bond length a(Cu-O) ≈ 3.8 Å, it seems reasonable to treat the multilayer as a single thick layer.

Reflectance data indicate that the dielectric functions of the cuprates may qualitatively be described by the so-called two-fluid model[2,3]

$$\varepsilon(\omega) = \varepsilon_\infty - \frac{\omega_{ps}^2}{\omega(\omega + i0^+)} - \frac{\omega_{pn}^2}{\omega[\omega + i\gamma_{pn}(\omega)]}, \quad (1)$$

which is a generalization of a simplest metallic dielectric function $\varepsilon(\omega) = 1 - \omega_p^2/[\omega(\omega + i\gamma_p)]$. The carriers are divided into normal and superfluid components which have different impacts on optical and conducting properties of the cuprates. In Eq. (1) ω_{ps} and ω_{pn} are the plasma frequencies of the superconducting and normal components, respectively, and ε_∞ is the high-frequency limit of ε. The relaxation processes of the normal-state electrons are described by (in general, frequency-dependent) scattering rate $\gamma_{pn}(\omega)$. In ordinary metals the relaxation rate is small compared to the plasma frequency (i.e., $\gamma_p^{Al}/\omega_p^{Al} \approx 5 \times 10^{-3}$ for aluminium). Contrary to the metals, *both* the finite conductivity and the relaxation processes are essential for optical properties of the cuprates[2] because in a typical cuprate $\gamma_{pn}(\omega_{pn})/\omega_{pn} \sim 1$.

Both in-plane and out-plane reflectance data show a sharp drop at the frequencies higher than the plasma edge [4] $\omega_{ps}/\sqrt{\varepsilon_\infty}$, which determines a boundary of a transparency window. Despite existence of other descriptions of the optical conductivities[2,5] we take Eq. (1) as our starting formula for the sake of concreteness.

The form of the dielectric function (1) suggests the presence of plasmon-mediated phenomena at the energy scales governed by the characteristic plasma frequencies. These phenomena are usually studied with respect to the *c*-axis conductivity ("transverse plasmon").[2,6] The importance of the *ab* plasmons for a proper description of the superconducting state in layered materials such as high-T_c cuprates was clearly stressed in Ref. 7. In our complimentary study we show that despite the *ab*-plane plasmon is heavily damped[6] it induces the spontaneous appearance of the superconductivity in the layers. Philosophically, our approach resembles mechanisms based on the interlayer Josephson tunneling[8] and interplane Coulomb interaction,[9] as well as other approaches[10] including phenomenological models of the Ginzburg-Landau type.[11]

The free energy per one d period per unit layer area S in the absence of external fields is given by a sum of the contributions from the normal (F_n) and the superconducting (F_s) states of the layer, and the plasmon-mediated interaction between the multilayers (F_{pl}):

$$F = F_n(\omega_{pn}, \gamma_{pn}) + F_s(\omega_{ps}) + F_{pl}(\omega_{ps}, \omega_{pn}, \gamma_{pn}). \quad (2)$$

In each term we explicitly indicate the leading-order dependence on the optical parameters ω_{ps}, ω_{pn}, and γ_{pn}. Long-range modulations of the *c*-axis structure are neglected. We imply that the effect of the intralayer media is solely insulating[2] thus neglecting a small finite out-plane conductivity in the normal state.

The free energy of the normal state F_n in Eq. (2) should depend on the optical parameters ω_{pn} and γ_{pn} related to a specific (in fact, model dependent) behavior of electrons in individual CuO_2 layers. Since we would like to keep our approach as general as possible we exclude F_n from our analysis concentrating on the difference in the free energies of the normal and superconducting states

$$\delta F = F_s(\omega_{ps}) + F_{pl}(\omega_{ps}, \omega_{pn}, \gamma_{pn}) - F_{pl}(0, \omega_{pn}, \gamma_{pn}). \quad (3)$$

The free energy density of the superconducting state in Eq. (2) is written in the Ginzburg-Landau (GL) form[12]

$$F_s = \frac{1}{S} \int_{V_l} \left[\frac{\hbar^2}{4m^*} \left| \left(\vec{\nabla} - \frac{2ie}{\hbar c} \vec{A} \right) \psi \right|^2 + \mathcal{A}|\psi|^2 + \frac{\mathcal{B}}{2}|\psi|^4 \right] dV, \quad (4)$$

where the integration is going over the volume V_l of the superconducting layer, $2m^*$ is the effective mass of the superconducting carrier, and $\mathcal{A}$, $\mathcal{B}$ are the GL phenomenological parameters describing the behavior of the order parameter ψ which is related to the density of the condensed electrons $n_s = |\psi|^2$. The GL approach has known limitations, while being usually correct near the point of the superconducting transition. Universality arguments suggest that the GL parameters must depend on the intrinsic layer properties while being generally less dependent on the intralayer structure.

Under assumption of a spatial homogeneity of the order parameter ψ and negligence of fluctuations of the electromagnetic field $\vec{A}$, the supercurrent in Eq. (4) vanishes and we arrive to the simple expression

$$F_s(\omega_{ps}) = w_n \left(\mathcal{A} \eta \omega_{ps}^2 + \frac{\mathcal{B} \eta^2}{2} \omega_{ps}^4 \right), \quad \eta = \frac{m^*}{16\pi^3 e^2}, \quad (5)$$

where w_n is the multilayer width. We used the relations

$$|\psi|^2 \equiv n_s = \frac{m^* c^2}{4\pi e^2 \lambda_L^2} \equiv \eta \omega_{ps}^2, \quad \lambda_L \omega_{ps} = 2\pi c, \quad (6)$$

where λ_L is the London penetration depth.

Naively, if the layers were structureless very thick solid plates made of alike atoms interacting with the van der Waals potential $U(r) = -\kappa r^{-6}$, then the interaction energy of the layers would be described by the well-known Hamaker form[12] $U_{pl}(d) = -H/(12\pi d^2)$, where $H = \kappa \pi^2 \rho^2$ is the Hamaker constant and ρ is the number density of atoms in the planes.

None of the above assumptions is satisfied by the cuprate layers because of significance of retardation, relaxation, dielectric absorbtion and geometrical suppression effects. These effects are known [12] to diminish the interaction which still follows the Hamaker law $U_{pl} \propto d^{-2}$. Up to an inessential numerical coefficient

$$U_{pl} = -\hbar \Omega \frac{G(w_n/d)}{16\pi^2 \epsilon_{int} d^2}, \quad \Omega = \int_0^\infty \left[\frac{\varepsilon(i\xi) - 1}{\varepsilon(i\xi) + 1} \right]^2 d\xi, \quad (7)$$

where ϵ_{int} is the intralayer dielectric function. The geometrical factor G takes into account the "multilayer-insulator" periodic structure[24]

$$G(r) = \frac{1}{(1+r)^2} \left[\psi^{(1)}\left(\frac{1}{1+r} \right) + \psi^{(1)}\left(\frac{1+2r}{1+r} \right) - \frac{\pi^2}{3} \right], \quad (8)$$

where $\psi^{(1)}$ is the first derivative of the digamma function. The geometric factor G (shown in Fig. 1 by the solid line) is a monotonically increasing function of $r \equiv w_n/d$.

The interaction energy (7) is of an electromagnetic origin. In an idealized limit of perfectly conducting layers the interaction energy may be imagined as the Casimir energy[13] of the electromagnetic field stored between the layers.[12] In the
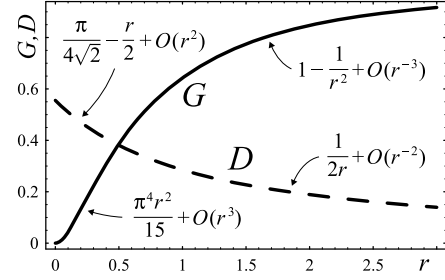

FIG. 1. The suppression factors geometrical G, Eq. (8), and dissipative D, Eq. (10), and their asymptotics at $r \to 0, \infty$.

case of real materials we follow Ref. 14 and interpret Eq. (7) as the interaction energy between the layers caused by the inlayer plasmons. The plasmons give a dominant contribution to the interlayer energy at short interlayer separations[14] $\omega_{pn} d \ll 1$. This condition is satisfied by typical cuprates (e.g., $\omega_{pn} d \lesssim 10^{-3}$ for the La$_{2-x}$Sr$_x$CuO$_4$ compound discussed below). The characteristic frequency[12] Ω in Eq. (7) gives account of the absorbtion spectra of the layers which, in turn, characterize the strength of the interaction between the ab plasmons.

The frequency Ω can be expressed via the dielectric function $\varepsilon(\omega)$ evaluated at the imaginary axis $\omega = i\xi$. The dispersion relation[12] expresses $\varepsilon(i\xi)$ via the conductivity Re $\sigma = \omega$ Im $\varepsilon/(4\pi)$ at the real axis:

$$\varepsilon(i\xi) = 1 + \frac{2}{\pi} \int_0^\infty \frac{\omega \operatorname{Im} \varepsilon(\omega)}{\omega^2 + \xi^2} d\omega \equiv 1 + 8 \int_0^\infty \frac{\operatorname{Re} \sigma(\omega)}{\omega^2 + \xi^2} d\omega. \quad (9)$$

Thus, the interlayer interaction (7) is fixed by the dissipative part of the in-plane conductivity Re σ, the interlayer dielectric parameter ε_{int}, and the c-axis geometry.

Despite the scattering rate γ_{pn} in cuprates is a complicated frequency-dependent function[2] one can approximately evaluate the order of the characteristic frequency Ω in the normal state ($\omega_{ps} = 0$) assuming that γ_{pn} is ω independent: $\Omega = \omega_{pn} D(\gamma_{pn}/\omega_{pn})$. The dissipative suppression factor [derived from Eqs. (1), (7), (9)],

$$D(r) = \frac{r\sqrt{r^2 - 2} - 4 \operatorname{arcsinh}\sqrt{(r/\sqrt{2} - 1)/2}}{2(r^2 - 2)^{3/2}}, \quad (10)$$

is a monotonically decreasing function of $r \equiv \gamma_{pn}/\omega_{pn}$. We plot $D(r)$ in Fig. 1 by the dashed line.

To estimate the energy scales related to the plasmon-mediated interactions, we consider La$_{2-x}$Sr$_x$CuO$_4$ (La214) compound. In La214 the CuO$_2$ layers are perfectly flat and are separated by two LaO layers at the distance $d = c_0/2 \approx 6.6$ Å. The basic cell has the tetragonal structure $a_0 \times b_0 \times c_0$ with the base parameters $a_0 \approx b_0 \approx 3.8$ Å. The condensation energy of the optimally doped ($x = 0.16, T_c = 38$ K) La214 compound is known to be $\mathcal{E}_{cond}^{(Cu)} \approx 13$ μeV per one atom of copper.[15] The normal state of the slightly underdoped La214 is characterized[16] by the plasma frequency $\omega_{pn} \sim 6000$ cm^{-1} while the typical scattering rate is of the order $\gamma_{pn} \sim 2000$ cm^{-1} for frequencies higher than the ab-plane "pseudogap" $\omega_{ab} \approx 700$ cm^{-1}.[17,18]Fixing the scatter-

ing rate to be constant and taking into account the dissipative suppression factor $D(1/3) \approx 0.4$, we get $\Omega_{La214} \approx 2500$ cm^{-1}. The characteristic frequency is of the order of a typical superconducting gap $\Delta \sim 10$–50 meV in the cuprates,[25] $\hbar\Omega_{La214} \sim 50$ meV.

In order to avoid suspicious fine-tuning of parameters in Eq. (7) we roughly set $\varepsilon_{int} \sim 1$, $c_0 \sim d$ [then $a_0 \sim d$ and the geometrical suppression is $G(1) \approx 0.6$]. Then the plasmon-mediated interaction energy per copper atom is

$$U_{pl}^{(Cu)}(La214) = -\frac{G\,\hbar\,\Omega_{La214}}{16\pi^2\epsilon_{int}}\left(\frac{a_0}{d}\right)^2 \approx -200 \ \mu eV. \quad (11)$$

This value is by an order of magnitude higher than the condensation energy $U_{pl}^{(Cu)} \sim 10\mathcal{E}_{cond}^{(Cu)}$. In other words, the condensation energy may well be explained by a 10% deviation in the interplane interaction, which in turn, should be related to a change of similar magnitude in the optical parameters of a cuprate as it cools down from critical to lower temperatures. In fact, the optical characteristics of cuprate compounds vary essentially in this range[2] exhibiting, e.g., a sharp drop of the scattering rate γ_{ps} and dominance of the superconducting component ω_{ps} at $T < T_c$. This argument stresses importance of the *ab*-plasmon mediated interlayer interactions. Below we ignore all interlayer interactions except for Eq. (7).

The crucial property of the interlayer interaction term U_{pl} is that it favors appearance of the scatterless superconducting component with $\omega_{ps} \neq 0$. To illustrate this property we expand the characteristic frequency Ω, Eq. (7), at $T = T_c$ in powers of $\omega_{ps}/\omega_{pn} \ll 1$,

$$\Omega(\omega_{ps}) = \omega_{pn}\left\{u_0 + u_2\left(\frac{\omega_{ps}}{\omega_{pn}}\right)^2 + O\left[\left(\frac{\omega_{ps}}{\omega_{pn}}\right)^4\right]\right\}. \quad (12)$$

The dimensionless coefficients u_m are certain functionals of the scattering rate $\bar{\gamma}_{pn}(y) = \gamma_{pn}(y \cdot \omega_{pn})/\omega_{pn}$, e.g.,

$$u_2[\bar{\gamma}_{pn}] = \int_0^\infty \frac{4[y+\bar{\gamma}_{pn}(y)]^2 dy}{\{2y[y+\bar{\gamma}_{pn}(y)]+1\}^3}. \quad (13)$$

As one can see from Eq. (13) the second coefficient of the expansion (12) is *always* positive, $u_2 > 0$, regardless of particularities of the scattering rate $\gamma_{pn}(\omega)$. The behavior of Ω at $\gamma_{pn} = const$ is illustrated in Fig. 2.

Since the characteristic frequency Ω enters Eq. (7) with the minus sign, the interaction energy (7) may provoke a tachyonic instability against emergence of the superconducting condensate (6), $|\psi|^2 \propto \omega_{ps}^2$. In other words, the interlayer interaction (7) supports the appearance of a superconducting ($\psi \neq 0$) state provided the layers are intrinsically able—via *any* microscopic mechanism—to generate this superconducting state.

At high temperatures the energy density associated with the superconducting condensate (4), (5) is higher compared to the gain in free energy (3) which would be achieved by the plasmon interaction (7). This makes the superconductivity energetically unfavorable. As the temperature decreases the coefficient $\mathcal{A}(T)$ in the GL free energy (5) gets gradually smaller and at the certain temperature the quadratic ω_{ps}^2 term of the GL free energy (5) cancels the same term in the
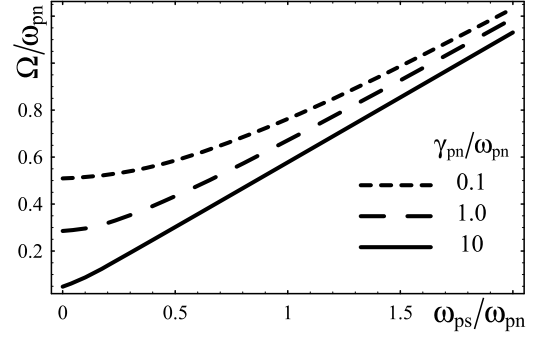

FIG. 2. The characteristic frequency (7) Ω vs the superconducting frequency ω_{ps} for various scattering rates γ_{pn}.

plasmon-mediated interaction (7), (12). This cancelation marks the critical temperature T_c. At lower temperatures, $T < T_c$, the overall coefficient in front of the quadratic term turns negative and the system becomes unstable against spontaneous development of the superconductivity $\omega_{ps} \sim |\psi| \neq 0$.

The relation between the critical temperature (T_c), optical (γ_{pn}, ω_{pn}) and geometrical (w_n, d) parameters of the cuprates can be derived from Eqs. (3), (7), (12):

$$\mathcal{A}(T_c)\frac{\omega_{pn}m^*\varepsilon_{int}}{\pi\hbar\,e^2 u_2(\gamma_{pn}/\omega_{pn})} = \frac{1}{w_n d^2}G(w_n/d), \quad (14)$$

where the left hand side (LHS) contains optical and microscopic parameters at $T = T_c$ while the right hand side (RHS) is of the purely geometrical origin. Below we list a few universal features of the cuprate superconductors which are described by Eq. (14).

Transition temperature T_c vs n. The GL coefficient $\mathcal{A}(T)$ is a monotonically increasing function of temperature. Thus, the higher (lower) value of the RHS in Eq. (14), the higher (lower) value of T_c is.[26] The RHS of Eq. (14) is a linearly rising function of w_n at $w_n \ll d$. At $w_n = 0.4448\,d$ the RHS has a maximum and then it decreases as $1/w_n$ at $w_n \gg d$ [we used Eq. (8) as well as the asymptotics of G given in Fig. 1]. In n-layered cuprates the width of the multilayer is a monotonically rising function of n, which can approximately be estimated as $w_n = (n-1)(c_{int} + \delta_g) + \delta_c$, where δ_g is the geometrical width of a single layer, ranging from $\delta_g(La-214) = 0$ Å and $\delta_g(Bi-2212) = 0.013$ Å to $\delta_g(YBCO) = 0.274$ Å,[19] $c_{int} \approx 3.5$ Å is the interlayer spacing inside the multilayer, and δ_c is a "coherence width" of external layers which should be of the order of the c-axis coherence length ξ_c (a few Å). For typical crystallographic parameters the RHS of Eq. (14), and, consequently, the transition temperature T_c, are peaked around $n_{max} = 3$. This behavior is in fact a universal feature of the homologous series.[1]

Transition temperature T_c vs d. The RHS of Eq. (14) is a monotonically decreasing function of the separation d between the multilayers provided the other geometrical parameters of the c-axis structure are fixed. Thus, the larger d the lower temperature must be. This is another universal behavior observed in the cuprates.[20]

Transition temperature T_c vs x. One may expect that the highest T_c is achieved at the doping x at which the density n_s

of the superconducting carriers is highest. However, this expectation is not confirmed experimentally:[21] the optimal doping for the transition temperature is noticeably lower compared to the one for the carriers (i.e., in La-214, Y-123, Bi-2212 cuprates one has $x_{\mathrm{opt}}^{T_c} \approx 0.16 < x_{\mathrm{opt}}^{n_c} \approx 0.19$). The plasmon-mediated interaction may explain this behavior. If the RHS of Eq. (14) were independent of x then the maximum temperature would be achieved at a certain value of $T = T_c(x_{\mathrm{opt}}^{n_s})$ corresponding to the highest carrier density. However, the interlayer distance d increases with the doping x,[22] lowering the plasmon interaction energy [proportional to the RHS of Eq. (14)]. Thus, the equality (14) is achieved at a lower value of the GL parameter, $\mathcal{A}[T_c(x_{\mathrm{opt}}^{n_s})] < \mathcal{A}[T_c(x_{\mathrm{opt}}^{T_c})]$, implying $T_c(x_{\mathrm{opt}}^{n_s}) < T_c(x_{\mathrm{opt}}^{T_c})$.

Scaling between T_c, ω_{ps}, and dc conductivity. At sufficiently low temperatures the normal component is almost invisible in the dielectric function.[2,17] In this case $\Omega = \pi \omega_{ps}/(4\sqrt{2})$ [we used Eq. (10) as well as the $\gamma_{pn} \to 0$ asymptotic, Fig. 1], and Eq. (7) becomes linear in ω_{ps}:

$$U_{pl}(\omega_{ps}, T=0) = -\frac{G(w_n/d)}{64\sqrt{2}\pi\epsilon_{\mathrm{int}}d^2}\hbar\,\omega_{ps}(0). \qquad (15)$$

Neglecting the quartic term in Eq. (5) we get the superconducting frequency at $T=0$ as a minimum of Eq. (3):

$$\omega_{ps}(0)\mathcal{A}(0) = \frac{\pi\mathcal{A}(T_c)}{8\sqrt{2}}u_2^{-1}\left(\frac{\gamma_{pn}(T_c,\omega)}{\omega_{pn}(T_c)}\right)\omega_{pn}(T_c), \qquad (16)$$

where we used Eq. (14) and disregarded the variation of the crystallographic parameters in the range of temperatures between T_c and $T=0$. The relation (16) links the ratio of the GL layer's parameter $\mathcal{A}$ at $T=T_c$ and $T=0$ with both superconducting and normal optical properties of the *ab* planes. Note that (i) the LHS (RHS) of Eq. (16) depends solely on $T=0$ ($T=T_c$) quantities; (ii) the relation (16) is universal: it is does not depend on the *c*-axis structure and should hold for all cuprate materials with the same in-plane parameters.

Since the relation (16) is dependent on $\omega_{pn}(T_c,\omega)$ further analytical calculations are difficult. We notice, however, that the integral (13) is saturated at low frequencies relevant to the normal-state dc conductivity:

$$\sigma_{dc} \equiv \lim_{\omega \to 0} \mathrm{Re}\,\sigma(\omega) = \frac{\omega_{pn}^2(T_c)}{4\pi}\lim_{\omega \to 0}\gamma_{pn}^{-1}(T_c,\omega). \qquad (17)$$

Therefore we take γ_{pn} to be equal to its low-frequency extrapolation, and work in the "dirty" limit $\gamma_{pn} \gg \omega_{pn}$, arriving to $u_2 = \gamma_{pn}/\omega_{pn}$. The requirement of the dirty limit is rather mild since even at $\gamma_{pn} = \omega_{pn}$ the above relation holds within 10%. Next, we take the standard GL-like prescription $\mathcal{A}(T) = \alpha(T+\widetilde{T}_c)$, with $0 < \widetilde{T}_c \ll T_c$, where α and $\widetilde{T}_c$ are the GL parameters describing the free energy associated with the condensation. We consider the most energetically unfavorable case: the positive $\widetilde{T}_c$ indicates that the layers alone are not able to support the superconductivity. Curiously, if the intrinsic layer properties are related, $\widetilde{T}_c = \beta\omega_{ps}(0)$ with $\beta \approx 10$ K cm, then from Eqs. (16) and (17) we get the scaling relation $\omega_{ps}^2(0) \approx 120\sigma_{dc}T_c$ observed experimentally.[23]

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[24] For simplicity we treat all layers equally while in general (i.e., for $n \geq 2$ Hg-based series) this is not the case.

[25] We ignore finite temperature corrections to the interlayer interaction (7) because $T_c \ll \hbar\Omega_{\mathrm{La214}}/k_B \approx 600$ K.

[26] We assume that the normal-state parameters are weakly dependent on temperature around T_c.