Manifestation of dielectric correlations in the spectrum of collective excitations of a superconductor

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We consider the spectrum of collective excitations of a system with coexisting superconducting and dielectric correlations (of the charge-transfer or of the charge-density-wave type). Excitations are observed that are not connected with long-wave fluctuations of the electron density, and hence do not go over into plasma excitations when account is taken of Coulomb interaction. If the dielectric-order parameter phase is not fixed, the system undergoes low-lying collective excitations with frequency in the energy interval of the superconducting gap. We discuss the possible manifestation of this excitation in the infrared and Raman spectra of high-temperature superconductors.

1. INTRODUCTION

According to recent experiments, high-temperature superconductors have very high static dielectric constants in the long-wave limit.[1-3] This is interpreted as an indication of the proximity of these materials to the instability point with respect to a transition to a ferroelectric phase. From the microscopic standpoint an instability of this kind can be connected with electronic instability of the charge-transfer type. A convenient way of describing it in the framework of the band approach is the excitonic-dielectric model.[4] An instability of the exciton type in CuO_2 layers was considered in Refs. 5 and 6 as a cause of the radical rise of the superconducting-transition temperature T_c due to the singularities of the momentum and frequency dependences of a renormalized interelectron interactions in systems with charge transport.

In the present paper, without specifying the Cooper-pairing mechanism, we take into account, in the calculation of the spectrum of the collective modes of a superconductor, the coexistence of superconducting and dielectric (electron–hole) correlations within the framework of the model of Ref. 7. We have in mind here the following qualitative picture of the development of a superconductor in a copper-oxide system. When the electron band is half-filled, the system is unstable to transition into a dielectric antiferromagnetic state. As the system becomes alloyed with holes, the Fermi level drops into the oxygen p-band and the antiferromagnetic order is partly suppressed. This is manifested by a decrease of the antiferromagnetic splitting of the d-bands. The upper Hubbard d-band is then close to the p-band inside the Hubbard gap, an exciton-type instability becomes possible and is manifested by a redistribution of the charge either among the Cu and O sites, or among the bonds. It should be noted that $YBa_2Cu_3O_{7-\delta}$ contains at $\delta = 0$, according to band calculations,[8] almost superimposed sections of the Fermi surface, i.e., the situation is close to Fermi-surface nesting. One can therefore not exclude the possibility of formation of short-range-order regions with charge or spin density (CDW or SDW) waves. If a CDW instability develops before an exciton instability, ferroelectric fluctuations can result from the proximity of the system to the point of extrinsic ferroelectric ordering, of the type considered in Ref. 9. The main features of the onset of a superconducting state against the background of an ordering of the CDW or of the charge-transfer type are the same in the model of Ref. 7. This makes it possible to consider, within the framework of a single scheme, both copper oxide systems and systems of the $Ba_{1-x}K_xBiO_3$ type.

We describe, in the band approach, a system of correlated electrons and holes. That this approach is valid for high-temperature superconductors is convincingly confirmed by a number of experimental facts. These include first of all the observability of the Fermi surface in photoemission[10] and galvanomagnetic[11] experiments. The notion of evolution of a superconducting gap is confirmed by the appearance of corresponding singularities in photoemission spectra[12] and in tunnel spectra[13] with transition to the superconducting phase.

However, electron Raman scattering and submillimeter IR spectroscopy experiments, where the so-called combined density of states is measured, exhibit many discrepancies in the low-frequency region of the spectrum. In particular, it is impossible as a rule to observe in Raman-scattering spectra the threshold that would correspond to a nonzero minimum two-particle excitation energy.[14]

Many IR spectroscopy experiments (e.g., Refs. 15–17) have revealed spectrum singularities interpreted as superconducting gaps. Worthy of attention is Ref. 17, in which two singularities were observed in the reflection spectrum of $YBa_2Cu_3O_{6+x}$ (with superconducting compositions) at 150 and 430 cm^{-1}, corresponding to two absorption thresholds. The singularity at 430 cm^{-1} is present in spectra of samples with different oxygen contents (x) and consequently with different T_c. Moreover, a trace of it is preserved also in the normal phase. The low-frequency (150 cm^{-1}) singularities vanish on going to the normal phase, but can apparently not be reliably interpreted as a superconducting gap, owing to the weak correlation of its position with T_c for samples with $T_c \sim 90$ K. The authors of Ref. 17 indicate that the low-frequency singularity is resolved at the sensitivity limit of their apparatus. A similar singularity, however, with frequency 130 cm^{-1} ($\sim 1.9\, k_B T_c$) was observed[15] in a thin-film 1-2-3 sample, where a better signal/noise ratio than in Ref. 17 could be reached.

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It follows from the foregoing that special interest may attach to low-lying collective excitations whose characteristic frequencies land in the superconducting-gap energy interval. The question of low-lying collective modes was actively discussed earlier in connection with experiments on Raman scattering in the superconductor $2H - NSe_2$ with CDW.[18-20] The authors of these references considered Raman-active amplitudes of the CDW oscillation and of the superconducting order parameter.

We consider below the IR-active phase oscillation of the dielectric order parameter and its connection with the oscillations of the phase and amplitude differences of superconducting condensates in different bands. In the case of the single-band picture with "nesting" the latter correspond to excitations of type $s + id$ and $s + d$, respectively. In contrast to the Bogolyubov–Anderson mode,[21] whose frequency goes over to the plasma-frequency region when long-range Coulomb potential is taken into account, the collective excitations considered by us are not connected with long-wave electron-density oscillations. Such modes were phenomenologically considered in Ref. 22. Low-lying excitations were obtained in Ref. 23 microscopically in the framework of the model of a two-band superconductor without allowance for dielectric considerations. In the latter case the oscillations are not optically active.

2. HAMILTONIAN OF MODEL AND BASIC RELATIONS

The Hamiltonian of the model is given by

$$H=\sum_{i,\mathbf{p},\alpha}\varepsilon_i(\mathbf{p})a^+_{i\mathbf{p}\alpha}a_{i\mathbf{p}\alpha}+{}^1/_2\sum_{\mathbf{p},\mathbf{p}',\mathbf{q}}\lambda_{ij}a^+_{i\mathbf{p}\alpha}a^+_{j\mathbf{p}'\beta}a_{j\mathbf{p}'+\mathbf{q}\beta}a_{i\mathbf{p}-\mathbf{q}\alpha}, \tag{1}$$

where $i,j = 1,2$ are the band indices, $\varepsilon_i(p)$ is the dispersion law in band i, $a^+_{ip\alpha}$ is the creation operator for an electron with quasimomentum p and spin α in band i, while λ_{ij} are the interaction constants: λ_{11} and λ_{22} are the intraband constants that cause Cooper pairing of electrons in bands 1 and 2. We do not specify the pairing mechanism and assume λ_{ii} to be phenomenological constants that describe the Cooper attraction in the energy region $\tilde{\omega}$. The interband Coulomb interaction is simulated by the constant λ_{12}. According to Ref. 7, the interband interaction in the Cooper channel can differ from λ_{12} because it has a different renormalization when account is taken, for example, of electron–phonon interaction; we therefore assume below that it is not equal to λ_{12} and denote it by $\tilde{\lambda}_{12}$. In addition to "density–density" terms it is necessary to add to (1) terms that describe scattering processes of the type $\lambda_2 a_1^+ a_2^+ a_1 a_2$ and $\tilde{\lambda}_2 a_1^+ a_1^+ a_2 a_2$ and single-particle $g_{12}a_1^+ a_2$ transitions. Allowance for these terms leads to different effective coupling constants for different types of dielectric order parameter[24,25] (see below). In particular, the coupling constants $\lambda_{\rm Re}$ and $\lambda_{\rm Im}$ for the real and imaginary dielectric parameters are different.

The unrenormalized spectrum of the system is assumed for simplicity to be isotropic:

$$\varepsilon_{1,2}=\mu\pm\xi(\mathbf{p}),\quad \xi(\mathbf{p})=\frac{\mathbf{p}^2}{2m}-\varepsilon_0. \tag{2}$$

Here ε_0 is the Fermi energy ($\varepsilon_0 > 0$) in the case of overlapping bands 1 and 2 (the unrenormalized phase is a semimetal), or half the band gap ($\varepsilon_0 < 0$) in the semiconductor model. The parameter μ determines the position of the chemical potential, and m is the effective mass.

As shown in Refs. 4, 7, and 24, the unrenormalized phase with Hamiltonian (1) and spectrum (2) has an instability called excitonic. In the semiconductor model this instability sets in as soon as the energy of electron and hole binding into an exciton exceeds the band gap.[4] Depending on the spin structure of the electron-hole pair and the phase shift of the wave functions of the electron and hole that form the exciton, the model (1), (2) describes different types of spin, charge, or current ordering. Their classification can be found, for example, in Ref. 24 and we shall not dwell on it. We note only that if the phase shift of the wave functions of the electron and hole that form a singlet exciton is a multiple of π, and for a nonzero interband matrix element of the momentum

$$\langle 1|\frac{\nabla}{i}|2\rangle$$

ferroelectric ordering is realized in the system.[26] We consider below just this type of order. The problem of superconducting ordering in a system with exciton instability of the priming phase was solved in Ref. 7. We have indicated in the Introduction that in high-temperature superconductors with superconducting compositions only fluctuations of ferroelectric type are observed and not a true long-range order. If the reciprocal lifetimes of these fluctuations do not exceed the binding energy of the electrons in a Cooper pair, the qualitative picture of establishment of superconductivity against the background of short-range ferroelectric order will be similar to that considered in Ref. 7 in the mean-field approximation. As shown there, the phase diagram of a system with coexisting superconducting and dielectric correlations has a region in which the superconducting transition contributes to dielectric ordering. One should therefore expect an increase of the dimensions of the short-range order region and of the lifetime of the dielectric-type fluctuations, with transition of the system into a superconducting phase. This points likewise to qualitative validity of the model of Ref. 7.

The Hamiltonian of the model acquires in the mean-field approximation the form

$$H_0=\sum_{i\mathbf{p}\alpha}\varepsilon_i(\mathbf{p})a^+_{i\mathbf{p}\alpha}a_{i\mathbf{p}\alpha}+\sum_{\alpha\beta\mathbf{p}}(\Sigma_{12}{}^{\alpha\beta}a^+_{2\mathbf{p}\alpha}a_{1\mathbf{p}\beta}+\text{H.c.})$$
$$+\sum_{ij\alpha\beta\mathbf{p}}(\Delta_{ij}{}^{\alpha\beta}a^+_{i\mathbf{p}\alpha}a^+_{j-\mathbf{p}\beta}+\text{H.c.}). \tag{3}$$

The anomalous mean values describe here the dielectric ($\Sigma_{12}^{\alpha\beta}$) and superconducting intraband ($\Delta_{ii}^{\alpha\beta}$) and interband ($\Delta_{12}^{\alpha\beta}$) orderings. We shall consider hereafter anomalous mean value with a spin structure

$$\Sigma_{12}{}^{\alpha\beta}=\Sigma_{12}\delta_{\alpha\beta},\quad \Delta_{ij}{}^{\alpha\beta}=i\sigma_2{}^{\alpha\beta}\Delta_{ij}$$

(σ_2 is a Pauli y-matrix), corresponding to singlet pairing in the exciton and Cooper channels.

In the superconductivity model considered by us one can separate two independent electron-state spaces: the space of band states and the space of states inverted in time. To make the notation compact it is convenient to introduce a pseudospin representation of the creation and annihilation operators, similar to the Nambu representation and reflecting the presence of these state spaces.[19] We introduce the operator

$$A_{\mathbf{p}}^{+}=(a_{1\mathbf{p}\uparrow}^{+}, a_{2\mathbf{p}\uparrow}^{+}, a_{1-\mathbf{p}\downarrow}, a_{2-\mathbf{p}\downarrow}). \quad (4)$$

We write the Hamiltonian (3) in this representation:

$$H_0=\sum_{\mathbf{p}} A_{\mathbf{p}}^{+}\hat{H}_0 A_{\mathbf{p}}=\sum_{\mathbf{p}} A_{\mathbf{p}}^{+}\Big(\sum_{r,s=0}^{3} h_{rs}\sigma_r\otimes\sigma_s\Big)A_{\mathbf{p}}$$

$$=\sum_{\mathbf{p}} A_{\mathbf{p}}^{+}(\xi(\mathbf{p})\sigma_3\otimes\sigma_3$$

$$+\mu\sigma_3\otimes\sigma_0+\Sigma_1\sigma_3\otimes\sigma_1+i\Sigma_2\sigma_0\otimes\sigma_2-\Delta_1^s\sigma_1\otimes\sigma_0-\Delta_1^a\sigma_1\otimes\sigma_3$$
$$-i\Delta_2^s\sigma_2\otimes\sigma_0-i\Delta_2^a\sigma_2\otimes\sigma_3-\tilde{\Delta}_1\sigma_1\otimes\sigma_1-i\tilde{\Delta}_2\sigma_2\otimes\sigma_1)A_{\mathbf{p}}. \quad (5)$$

In (5) we have expanded the Hamiltonian (3), which has in the representation (4) the form of a four-row matrix $\hat{H}_0$, in terms of a complete system of matrices of form $\sigma_r\otimes\sigma_s$, where $\sigma_{1,2,3}$ are Pauli matrices, σ_0 is a unit two-row matrix. The direct product of the matrices is so defined that in the four-row matrix $\sigma_r\otimes\sigma_s$ the blocks σ_s are arranged in an order specified by the elements of the matrix σ_r, and are additionally multiplied by them, e.g.,

$$\sigma_1\otimes\sigma_3=\begin{pmatrix}0 & \sigma_3\\ \sigma_3 & 0\end{pmatrix}$$

The elements h_{rs} in (5) are expressed in terms of the parameters of the Hamiltonian (3) as follows:

$$\Sigma_{1,2}={}^1/_2(\Sigma_{12}\pm\Sigma_{21}),\quad \tilde{\Delta}_{1,2}={}^1/_4(\Delta_{12}+\Delta_{21}\pm(\Delta_{12}^{*}+\Delta_{21}^{*}))$$

$$\Delta_{1,2}^{s}={}^1/_4(\Delta_{11}+\Delta_{22}\pm(\Delta_{11}^{*}+\Delta_{22}^{*})), \quad (6)$$

$$\Delta_{1,2}^{a}={}^1/_4(\Delta_{11}-\Delta_{22}\pm(\Delta_{11}^{*}-\Delta_{22}^{*})),$$

where

$$\Sigma_{12}={}^1/_2\lambda_{12}\sum_{\mathbf{p}\alpha}\langle a_{1\mathbf{p}\alpha}^{+}a_{2\mathbf{p}\alpha}\rangle,\quad \Delta_{12}=-{}^1/_2\tilde{\lambda}_{12}\sum_{\mathbf{p}\alpha}\langle a_{1\mathbf{p}\alpha}a_{2-\mathbf{p}-\alpha}\rangle,$$

$$\Delta_{ii}=-{}^1/_2\lambda_{ii}\sum_{\mathbf{p}\alpha}\langle a_{i\mathbf{p}\alpha}a_{i-\mathbf{p}-\alpha}\rangle. \quad (7)$$

As shown in Ref. 7, one can assume without loss of generality that $\lambda_{11}=\lambda_{22}=\lambda_s$. Thus $|\Delta_{11}|=|\Delta_{22}|$ and we have two types of solution: symmetric $\Delta_{11}=\Delta_{22}\equiv\Delta$, $\Delta_{12}\equiv\tilde{\Delta}\neq 0$ and antisymmetric $\Delta_{11}=-\Delta_{22}\equiv\Delta$, $\Delta_{12}=0$. In the case of a real dielectric order parameter $\Sigma=\Sigma_1\neq 0$ the maximum superconducting-transition temperature corresponds to a symmetric solution. We have then in a wide range of parameters $\tilde{\Delta}\ll\Delta$ and we shall neglect them hereafter. The effective parameter Σ is realized if its effective coupling constant $\lambda_{\rm Re}$ turns out to be the largest.[24]

We introduce the Green's function in the representation (4);

$$\hat{G}(\mathbf{p},\mathbf{p}';\tau,\tau')=-\langle TA_{\mathbf{p}}(\tau)A_{\mathbf{p}'}^{+}(\tau')\rangle, \quad (8)$$

where $A_p(\tau)$ and $A_p^{+}(\tau)$ are operators whose components $a_{ip\alpha}^{+}$ and $a_{ip\alpha}$ are taken in the Matsubara representation.

In the mean-field approximation the system is homogeneous in space, so that

$$\hat{G}(\mathbf{p},\mathbf{p}';\tau,\tau')=\hat{G}_0(\mathbf{p},\tau-\tau')\delta_{\mathbf{pp}'}. \quad (9)$$

The mean-field order parameters (6) satisfy the self-consistency equations

$$h_{rs}={}^1/_4 g_{rs}T\sum_{i\omega}\sum_{\mathbf{p}}\mathrm{Sp}(\hat{G}_0(\mathbf{p},i\omega)\sigma_r\otimes\sigma_s), \quad (10)$$

where $i\omega=i(2n+1)\pi T$ is the fermion Matsubara frequency. The subscripts r and s run through the same values as in (5), except 30 and 33. These values of g_{rs} are expressed in terms of the interaction constants λ_s, $\tilde{\lambda}_{12}$, $\lambda_{\rm Re}$ and $\lambda_{\rm Im}$: $g_{10,13,20,23}=\lambda_s$, $g_{11,21}=\tilde{\lambda}_{12}$, $g_{31}=\lambda_{\rm Re}$, $g_{02}=\lambda_{\rm Im}$.

The elements of the matrix Green's function $\hat{G}_0(\mathbf{p},i\omega)$ were calculated in Ref. 7. Neglecting $\tilde{\Delta}$, then take for the symmetric solution the form

$$G_{11}(\mathbf{p},i\omega)=\{(i\omega+\varepsilon_2(\mathbf{p}))[(i\omega+\varepsilon_1(\mathbf{p}))(i\omega+\varepsilon_2(\mathbf{p}))-\Sigma^2]-\Delta^2(i\omega+\varepsilon_1(\mathbf{p}))\}/D,$$
$$G_{21}(\mathbf{p},i\omega)=\Sigma\{(i\omega+\varepsilon_1(\mathbf{p}))(i\omega+\varepsilon_2(\mathbf{p}))-\Sigma^2-\Delta^2\}/D,$$
$$F_{11}^{+}(\mathbf{p},i\omega)=-\Delta\{(i\omega)^2-\varepsilon_2^2(\mathbf{p})-\Delta^2-\Sigma^2\}/D, \quad (11)$$
$$F_{21}^{+}(\mathbf{p},i\omega)=-\Delta\Sigma\{\varepsilon_2(\mathbf{p})+\varepsilon_1(\mathbf{p})\}/D,$$
$$D=((i\omega)^2-\omega_+^2(\mathbf{p}))((i\omega)^2-\omega_-^2(\mathbf{p})),$$
$$\omega_\pm(\mathbf{p})=[((\xi^2(\mathbf{p})+\Sigma^2)^{1/2}+\mu)^2+\Delta^2]^{1/2},$$

where $\omega_\pm(\mathbf{p})$ is the spectrum of the elementary excitations of the superconducting phase.

The Green's functions (11) enter in $\hat{G}_0(\mathbf{p},i\omega)$ as 2×2 block components:

$$\hat{G}_0(\mathbf{p},i\omega)=\begin{pmatrix}G_{ij}(\mathbf{p},i\omega) & F_{ij}(\mathbf{p},i\omega)\\ F_{ij}(\mathbf{p},i\omega) & -G_{ji}(\mathbf{p},-i\omega)\end{pmatrix}. \quad (12)$$

The block components (12) given in (11) can be obtained in the following manner. The functions G_{22} and F_{22}^{+} are obtained from the expressions for G_{11} and F_{11}^{+}, respectively, by the interchange $\varepsilon_1\rightleftarrows\varepsilon_2$. In the case considered here, of real dielectric and superconducting order parameters (the latter can always be made real by a suitable gauge transformation) we have

$$G_{12}=G_{21},\quad F_{ij}^{+}=F_{ji},\quad F_{12}=F_{21}.$$

3. LINEAR RESPONSE TO AN EXTERNAL FIELD AND COLLECTIVE-EXCITATION SPECTRUM

To find the spectrum of the collective modes of the system we consider its linear response to an external field. As indicated in the Introduction, we are interested in low-lying modes of the collective-excitation spectrum. Therefore, before we proceed to consider the linear response, we make the following remark concerning the low-lying excitations. The principal candidates might obviously be the Goldstone modes of the system, which are the consequence of violation of the gauge invariance in the Hamiltonian (3). It is known from field theory that if the Lagrangian of a system has a continuous symmetry group G, and the vacuum state has as a result of symmetry breaking a lower symmetry with group H which is a subgroup of G, then the number of Goldstone particles is equal to the number of those group-G generators which are not generators of H (Ref. 27). The Hamiltonian (1) is invariant to phase shifts of the operators a_1 and a_2, i.e., to transformations of the $G=U(1)\times U(1)$ symmetry group having two generators. For the state described by Hamiltonian (3), one cannot indicate any subgroup H different from a unit group whose transformations leave (3) invariant. It follows therefore that the number of Goldstone modes in the discussed system is two. The foregoing pertains to a situation with a singlet dielectric parameter. In the case of a triplet parameter there is one more Goldstone mode (transverse spin wave). This case calls for separate treatment and is outside the scope of the present paper.

One of these modes corresponds to the collective Bogolyubov–Anderson mode,[21] and the second to an analogous exciton-dielectric mode.[28] In the first case, however, the long-range Coulomb potential pushes out the low-lying excitation into the plasma-frequency region, and in an excitonic dielectric fixing the phase is unavoidable when account is taken of the interaction terms corresponding to particle interband transitions,[24,25] while the second mode has a gap spectrum.[28] For the latter, however, there remains the possibility of landing in the energy interval of the superconducting gap. This possibility will be discussed below.

Let us consider the linear response of the system to an external field V in which the unrenormalized interaction vertex has in the representation (4) the form

$$\hat{\gamma}(p+k,p)=\sum_{r,s=0}^{3}\gamma_{rs}(p+k,p)\sigma_r\otimes\sigma_s, \tag{13}$$

where $p=(\rho,i\omega)$, $k=(\varkappa,i\omega)$, $i\omega=i(2n+1)\pi T$, and $i\Omega=i2n\pi T$ is the Bose frequency.

In the field V the equilibrium (mean-field) order parameters acquire induced increments which we write in the form $\hat{\eta}V$, where

$$\hat{\eta}(p+k,p)=\sum_{r,s=0}^{3}\eta_{rs}(p+k,p)\sigma_r\otimes\sigma_s. \tag{14}$$

The subscripts r and s run through the same values as in (5), except $rs=33$. The physical meaning of the quantities η_{rs} in (14) is the following. $V\eta_{33}(p+k,p)\equiv V\eta_{33}(k)$ corresponds to the scalar-potential oscillations due to the electron-density fluctuations. This potential is connected with the density by a Poisson equation, which can be treated alongside the self-consistency equations (10) if the corresponding interaction potential is taken to mean the Coulomb potential $g_{30}=4\pi e^2/\mathbf{k}^2\cdot V\eta_{31}$ and $V\eta_{02}$ correspond to the amplitude and phase oscillations of the order parameters, while $V\eta_{10}$ and $V\eta_{20}$ correspond to oscillations of the amplitude and phase of the symmetric component of the superconducting order parameter or, in other words, oscillations of the summary amplitude and phase of the intraband superconducting parameters. $V\eta_{13}$ and $V\eta_{23}$ correspond to the oscillations of the differences of the amplitudes and phases of the superconducting parameters of bands 1 and 2 or to oscillations of the antisymmetric component of the superconducting order parameter, while $V\eta_{11}$ and $V\eta_{21}$ describe oscillations of the amplitude and phase of the interband component of the superconducting order parameter.

It is appropriate to point out the similarity of the model considered by us to the model of a single-band model with inserted sections of the Fermi surface. The symmetric solution for the superconducting ordering in the model of Ref. 7 corresponds then to the s-wave of the superconducting parameter in conventional classification, and the antisymmetric corresponds to the d-wave. The antisymmetric oscillations considered by us are similar to transitions induced by an external field of the s state in the $s+d$ (amplitude oscillation) and $s+id$ (phase oscillation) states. These oscillations are produced even if the interaction potential contains only s-harmonics. This is the result of renormalization of the unrenormalized Cooper-channel constant by the dielectric ordering on the background of which the superconductivity sets in.

The total Hamiltonian of the system takes, with allowance for (13) and (14), the form

$$H=\hat{H}_0+(\hat{\eta}(p+k,\ p)+\hat{\gamma}(p+k,\ p))V(k) \tag{15}$$

To simplify the notation, we have not identified explicitly in (13)–(15) the tensor character (scalar, vector, etc.) of the external field.

The Green's function (8) satisfies the equation of motion

$$\left(\frac{\partial}{\partial\tau}-\hat{H}\right)\hat{G}(\mathbf{p},\mathbf{p}';\tau,\tau')=\delta(\tau-\tau')\delta_{\mathbf{pp}'}\sigma_0\otimes\sigma_0. \tag{16}$$

Separating in the Green's function the increment $\delta\hat{G}$ which is linear in the field and linearizing (16) we obtain after substituting $\hat{G}_0+\delta\hat{G}$ in the self-consistency equation (10) the following system of dynamic equations for $\eta_{rs}(p+k,p)$:

$$\eta_{rs}(p+k,p)=g_{rs}(\mathbf{k})T\sum_{i\omega}\sum_{\mathbf{q}}\sum_{m,n=0}^{3}\Pi_{rsmn}(q+k,q)(\eta_{mn}(q+k,q)$$
$$+\gamma_{mn}(q+k,q)), \tag{17}$$

where

$$\Pi_{rsmn}(q+k,\ q)={}^1/_4\,\mathrm{Sp}[\sigma_r\otimes\sigma_s\hat{G}_0(q+k)\sigma_m\otimes\sigma_n\hat{G}_0(q)]. \tag{18}$$

It follows from (17) that $\eta_{rs}(p+k,p)$ depends only on k. We have written in (17) $g_{rs}(k)$ in lieu of the g_{ij} employed above, so as to include together with the interaction constants the Coulomb potential produced by oscillations of the electron density. The quantity $\eta_{mn}(k)+\gamma_{mn}(q+k,q)$ is the total vertex "dressed" by the interelectron interaction. Denoting it by $\Gamma_{mn}(q+k,q)$, we obtain for it the usual random-phase-approximation equation:

$$\Gamma_{rs}(p+k,p)=\gamma_{rs}(p+k,p)+g_{rs}(k)T$$
$$\times\sum_{i\omega}\sum_{\mathbf{q}}\sum_{m,n=0}^{3}\Pi_{rsmn}(q+k,q)\Gamma_{mn}(q+k,q). \tag{19}$$

The generalized susceptibility for the action of the field V is given by

$$\chi_v(k)=T\sum_{i\omega}\sum_{\mathbf{p}}\sum_{r,s,m,n=0}^{3}\gamma_{rs}(p+k,p)\Pi_{rsmn}(p+k,p)\Gamma_{mn}(p+k,p) \tag{20}$$

from which we can calculate the contribution of the collective excitations to the system susceptibility

$$\chi_{mn}(k)=T\sum_{i\omega}\sum_{p}\sum_{rs=0}^{3}\gamma_{rs}(p+k,p)\Pi_{rsmn}(p+k,p)\eta_{mn}(k). \tag{21}$$

The system (17) contains nine unknowns. In the quasi-two-dimensional state corresponding to layered high-temperature superconductors the system (17) breaks up into two independent equations with dimensionalities 6×6 and 3×3. The first contains the dynamic equations for the amplitude oscillations of the dielectric order parameter, the scalar parameter, the scalar potentials, and the amplitude and phase oscillations of the symmetric and interband components of the superconducting parameter. We shall not solve this part of the system, since it was already investigated several times.[18-20]

The long-wave Coulomb potential impeding the onset of the Goldstone mode in an ordinary superconductor turns out not to be connected with the phase oscillations of the dielectric order parameters and with the oscillations of the phase and amplitude differences of the superconducting condensates of bands 1 and 2, since these oscillations do not cause the electron density to oscillate. The possibility of low-lying collective excitation is therefore preserved.

The investigated part of the homogeneous set of equations corresponding to (17) is

$$\begin{pmatrix} \frac{2}{\lambda_{1m}}-\Pi_{0202}(\mathbf{k},\Omega)-\Pi_{0223}(\mathbf{k},\Omega) & -\Pi_{0213}(\mathbf{k},\Omega) \\ -\Pi_{2302}(\mathbf{k},\Omega) & \frac{2}{\lambda_s}-\Pi_{2323}(\mathbf{k},\Omega) & -\Pi_{2313}(\mathbf{k},\Omega) \\ -\Pi_{1302}(\mathbf{k},\Omega) & -\Pi_{1323}(\mathbf{k},\Omega) & \frac{2}{\lambda_s}-\Pi_{1313}(\mathbf{k},\Omega)\end{pmatrix}\begin{pmatrix}\eta_{02}(\mathbf{k},\Omega)\\ \eta_{23}(\mathbf{k},\Omega)\\ \eta_{13}(\mathbf{k},\Omega)\end{pmatrix}=0. \tag{22}$$

We have introduced here the notation

$$\Pi_{rsmn}(\mathbf{k},i\Omega)=\sum_{p}\Pi_{rsmn}(p+k,p)$$

and carried out in the quantities $\Pi_{rsmn}(\varkappa,i\Omega)$ an analytic continuation from the discrete imaginary frequencies $i\Omega$ to the region of real frequencies, $i\Omega\to\Omega+i\delta$. The expressions for the quantities $\Pi_{rsmn}(\varkappa,\Omega)$ are very unwieldy. We present them taken to the limit as $k\to 0$ and for a temperature $T=0$, using the notation

$$\Pi_{rsmn}(\mathbf{k}\to 0,\Omega)=N(0)\int_{-\infty}^{\infty}d\xi P_{rsmn}(\xi,\Omega),$$

where $N(0)$ is the density of states of the unrenormalized phase on the Fermi level,

$$P_{0202}(\xi,\Omega)=\left(1+\frac{E^2-\mu^2-\Delta^2}{\omega_+\omega_-}\right)\frac{\omega_++\omega_-}{(\omega_++\omega_-)^2-\Omega^2},$$

$$P_{0223}(\xi,\Omega)=P_{2302}(\xi,\Omega)=\left(\frac{1}{\omega_+}+\frac{1}{\omega_-}\right)\frac{2\Sigma\Delta}{(\omega_++\omega_-)^2-\Omega^2},$$

$$P_{0213}(\xi,\Omega)=P_{1302}(\xi,\Omega)=\frac{\Sigma\Delta}{E}\left(\frac{1}{\omega_-}-\frac{1}{\omega_+}\right)\frac{\Omega}{(\omega_++\omega_-)^2-\Omega^2},$$

$$P_{2323}(\xi,\Omega)=\frac{2\xi^2}{E^2}\left(\frac{\omega_+}{4\omega_+^2-\Omega^2}+\frac{\omega_-}{4\omega_-^2-\Omega^2}\right)+\frac{\Sigma^2}{E^2}\left(1-\frac{E^2-\mu^2-\Delta^2}{\omega_+\omega_-}\right)\frac{\omega_++\omega_-}{(\omega_++\omega_-)^2-\Omega^2}, \tag{23}$$

$$P_{1323}(\xi,\Omega)=P_{2313}(\xi,\Omega)=\Omega\frac{\xi^2}{E^2}\left(\frac{E+\mu}{\omega_+(4\omega_+^2-\Omega^2)}-\frac{E-\mu}{\omega_-(4\omega_-^2-\Omega^2)}\right)$$

$$P_{1313}(\xi,\Omega)=\frac{2\xi^2}{E^2}\left(\frac{(E+\mu)^2}{\omega_+(4\omega_+^2-\Omega^2)}+\frac{(E-\mu)^2}{\omega_-(4\omega_-^2-\Omega^2)}\right)+\frac{\Sigma^2}{E^2}\left(1-\frac{E^2-\mu^2+\Delta^2}{\omega_+\omega_-}\right)\frac{\omega_++\omega_-}{(\omega_++\omega_-)^2-\Omega^2}.$$

Here

$$\omega_\pm=((E\pm\mu)^2+\Delta^2)^{1/2},\quad E=(\xi^2+\Sigma^2)^{1/2}.$$

Being interested in low-lying oscillations, we solve the system (22) in the frequency region $\Omega\ll 2\Delta$ in the limit as $\mathbf{k}\to 0$ and at $T=0$. Expanding in (23) up to second order in $\omega=\Omega/\Delta$ we have

$$\begin{pmatrix} 2\alpha+\frac{\Delta^2}{\mu}A-\frac{\Delta^2}{\mu^2+\Sigma^2}\omega^2 & -\frac{\Sigma\Delta}{\mu}A & -\frac{\Sigma\Delta^2}{2\mu^2}A\omega \\ -\frac{\Sigma\Delta}{\mu}A & \frac{\Sigma^2}{\mu}A-\frac{3}{2}\frac{n}{\mu}\omega^2 & \frac{\Sigma^2\Delta}{2\mu^2}A\omega \\ -\frac{\Sigma\Delta^2}{2\mu^2}A\omega & \frac{\Sigma^2\Delta}{2\mu^2}A\omega & \frac{\Sigma^2}{\mu}A-\frac{1}{36}\frac{n}{\mu}\omega^2\end{pmatrix}\times\begin{pmatrix}\eta_{02}(\Omega)\\ \eta_{23}(\Omega)\\ \eta_{13}(\Omega)\end{pmatrix}=0, \tag{24}$$

where

$$\eta_{rs}(\Omega)\equiv\eta_{rs}(\mathbf{k}\to 0,\Omega),\qquad \alpha=\frac{1}{N(0)}\left(\frac{1}{\lambda_{\rm Im}}-\frac{1}{\lambda_{\rm Re}}\right),$$

$$A=\int\limits_{\Sigma}^{\infty}\frac{dE}{(E^2-\Sigma^2)^{1/2}((E-\mu)^2+\Delta^2)^{1/2}}.$$

In the limit $\Delta\Sigma\ll 4n^2\equiv 4(\mu^2-\Sigma^2)$ we can use the asymptote of the elliptic integral A:

$$A=\frac{2}{n}\ln\frac{4n^2}{\Delta\Sigma}.$$

It follows from (24) that in the limit $\alpha=0$ the characteristic equation has a root $\omega=0$. This root corresponds to the dielectric-ordering Goldstone mode referred to at the beginning of this section. The low-lying root of the characteristic equation of system (22) is

$$\Omega_0^2=2\alpha(\mu^2+\Sigma^2). \tag{25}$$

This expression was obtained in the limit

$$\alpha\ll\frac{\Delta^2}{n^2}\ln\frac{4n^2}{\Delta\Sigma}.$$

The condition for the validity of the expansion in $\omega=\Omega/\Delta$ is

$$\alpha\frac{\mu^2}{\Delta^2}\ll 1.$$

It is more stringent. Thus, if $\alpha\ll\Delta^2/\mu^2$ the frequency of the collective mode lands in the energy interval of the superconducting gap and the mode turns out to be weakly damped.

4. MANIFESTATION OF LOW-LYING EXCITATION IN IR SPECTRA OF SUPERCONDUCTORS

Let us examine the manifestation of the obtained low-lying collective excitation in IR spectra. We describe the electromagnetic-wave vector by the vector potential $\mathbf{A}(\mathbf{k},\Omega)=\mathbf{A}\exp(i\mathbf{kr}-\Omega t)$. The vertex of the interaction of the electrons in this system with the field $\mathbf{A}(\mathbf{k},\Omega)$ can be established by replacing $\mathbf{p}$ in (5) by $\mathbf{p}-(e/c)\mathbf{A}$ and taking into account the possible interband transitions relative to the nonzero interband matrix element of the momentum

$$\mathbf{P}_{12}=\left\langle 1\left|\frac{\nabla}{i}\right|2\right\rangle=i\mathbf{P}.$$

Allowance for the term

$$\sum_{p\alpha}\mathbf{P}_{12}a^+_{1p\alpha}a_{2p\alpha}$$

in the Hamiltonian (1) adds to the spectrum of the dielectric phase (beside establishment of the phase) corrections of order $pk_F/m_0\Sigma$ (k_F is the Fermi momentum and m_0 is the free-electron mass). We assume hereafter for simplicity that this parameter is small, so that the spectrum corrections can be neglected.

The unrenormalized vertex of the interaction with the electromagnetic field is given by

$$\hat{\gamma}=-\frac{e}{m_0c}\mathbf{p}\sigma_0\otimes\sigma_3+\frac{e\mathbf{P}}{m_0c}\sigma_0\otimes\sigma_2. \tag{26}$$

It follows from (26) that the discussed low-lying mode can be excited by an electric field in proportion to P in an approximation linear in A. A similar situation was pointed out earlier in Ref. 29 as applied to an excitonic dielectric. The system (22) becomes then inhomogeneous. The column of free terms takes the form:

$$\frac{ie\mathbf{PA}(\mathbf{k},\omega)}{m_0c}(\Pi_{0202}(\mathbf{k},\Omega),\Pi_{0223}(\mathbf{k},\Omega),\Pi_{0213}(\mathbf{k},\Omega))^T,$$

where T denotes the transpose.

We determine the system susceptibility $Q_{\alpha\beta}(k,\Omega)$ in the usual manner in terms of the current $j(k,\Omega)$

$$j_\alpha(\mathbf{k},\Omega)=\sum_{\beta=x,y,z}Q_{\alpha\beta}(\mathbf{k},\Omega)A_\beta(\mathbf{k},\Omega).$$

The contribution of the obtained oscillation to the transverse part of $Q_{\alpha\beta}(k,\Omega)$ is given by

$$Q(\mathbf{k}\to 0,\Omega)=\frac{e\mathbf{P}}{4\pi m_0}\{\Pi_{0202}(\mathbf{k}\to 0,\Omega)\eta_{02}(\Omega)+\Pi_{0223}(\mathbf{k}\to 0,\Omega)\eta_{23}(\Omega)+\Pi_{0213}(\mathbf{k}\to 0,\Omega)\eta_{13}(\Omega)\}. \tag{27}$$

The contribution of the third term of (27) at low frequencies is small, since $\Pi_{0223}(k\to 0,\Omega)\sim\Omega$, as follows from (23).

Near the frequency Ω_0 [Eq. (25)], the response (27) becomes resonant

$$Q(\mathbf{k}\to 0,\Omega)=\frac{4e^2\mathbf{P}^2}{\pi m_0^2c}\frac{\mu^2+\Sigma^2}{\lambda_{\rm Im}^2N(0)}(\Omega^2-\Omega_0^2)^{-1}. \tag{28}$$

Let us discuss the response differences between the superconducting and normal phases. We disregarded in (28) the damping of the collective mode. Its damping will obviously be different in different phases, and larger in the normal than in the superconducting. This is due, first, to decay of this collective excitation into single-particle excitations with phonon emission. The probability of such a process is

$$W(\mathbf{k})=\sum_{\mathbf{p},\mathbf{q}}|M(\mathbf{p},\mathbf{k},\mathbf{q})|^2\delta(\Omega_{\mathbf{k}}+\varepsilon(\mathbf{p})-\varepsilon(\mathbf{p}+\mathbf{q})-\omega_{ph}(\mathbf{k}-\mathbf{q})), \tag{29}$$

where $M(p,k,q)$ is the matrix element of the process, $\varepsilon(p)$ is the single-particle excitation energy, and $\omega_{\rm ph}(p)$ is the phonon energy. In optical experiments $k\to 0$, so that we can put $k=0$ in (29). Such processes are energy-forbidden in the superconducting phase at $\Omega_0<2\Delta$, whereas in the nonsuperconducting phase they contribute to the damping. If the unrenormalized phase is not alloyed, such processes are energy-forbidden also in the phase with dielectric ordering. Our solution (25) turns then into the corresponding solutions of Refs. 29 and 30, dealing with collective modes of an undoped excitonic dielectric ($\mu=\Sigma$). To avoid misunderstandings, we note that doping of the unrenormalized phase is an obligatory condition for realization of superconductivity in the model of Ref. 7.

Second, in connection with the fluctuating character of the dielectric parameter Σ, the collective-mode damping is inversely proportional to the fluctuation lifetime. We have noted above that the phase diagram of the system contains a region in which superconductivity contributes to the increase of the lifetimes and sizes of the regions with short-range dielectric order. The collective-mode damping will be weakened. The obtained collective mode can therefore be resolved better in absorption spectra of the superconducting mode than in those of the normal phase.

In accordance with (28) and (25), the frequency of the response singularity due to the low-lying mode depends little on the superconducting parameters of the system, such as the gap Δ, and consequently on the temperature T_c of the superconducting transition. Within the framework of the model of Ref. 7 the connection between these parameters with Σ and μ is:

$$\Delta \approx \frac{4n^2}{\Sigma_0}\exp\left(-\frac{n}{\Sigma_0}\beta\right), \tag{30}$$

where $\beta = \ln(\Sigma_0/\Delta_0)$, while Σ_0 and Δ_0 are the dielectric and superconducting parameters in the absence of interaction between the superconducting and dielectric correlations, and $n^2 = \mu^2 - \Sigma^2$ is the free-carrier density, expressed in energy units, corresponding to the deviation of the deviation of the unrenormalized band from half-filled, with

$$\Sigma = (\Sigma_0(\Sigma_0 - 2n))^{1/2}.$$

It follows from (30) that the parameter Δ reaches as a function of n a maximum at the point $n_{\max} = 2\Sigma_0/\beta$. Near this point,

$$\Delta \approx \Delta_{max} - \frac{(n-n_{max})^2}{2\Sigma_0}. \tag{31}$$

The frequency of the collective mode (25) can be expressed in terms of Σ_0 and n:

$$\Omega_0 = 2\alpha^{1/2}(\Sigma_0 - n). \tag{32}$$

From (32) we obtain the frequency change $\delta\Omega = \Omega_0 - \Omega_{0,\max}$ when n deviates from $n_{\max}$. We shall connect this deviation next with the deviation of Δ from $\Delta_{\max}$ (31):

$$\delta\Omega_0 = \pm 2[2\alpha\Sigma_0(\Delta_{max} - \Delta)]^{1/2}. \tag{33}$$

The $\pm$ signs correspond here to the cases $n < n_{\max}$ and $n > n_{\max}$, respectively. We have assumed throughout that the dielectric-parameter phase is weakly fixed, i.e., $\alpha \ll 1$. Thus, the dependence (33) is weak in the region of small deviations of Δ from $\Delta_{\max}$, just where the expansion (31) is valid. In the weak-coupling limit we have in the weak-coupling theory[7] as well as in BCS $T_c = (\gamma/\pi)\Delta($ $\ln\gamma$ is the Euler constant), and the dependence of $\delta\Omega_0$ on $(T_{c,\max} - T_c)$ is similar to the dependence on $(\Delta_{\max} - \Delta)$ in (33).

Note that the qualitative variation of $\Omega_0 = \Omega_{0,\max} + \delta\Omega_0$ with T_c (actually with the doping) near the maximum temperature $T_{c,\max}$ agrees with that observed in experiment[17] in that there is practically no correlation between the position of the low-frequency singularity of the reflection spectrum and the temperature of superconducting $YBa_2Cu_3O_{6+x}$ specimens with $T_c \sim 90$ K. It is noted in Ref. 17 at the same time that the absorption edge of specimens with non optimal composition ($T_c < 60$ K) the absorption edge shifts towards lower frequencies when T_c is lowered.

This behavior can apparently be interpreted as follows. The singularity in (28) has the character of a steep peak in the region $\Omega_0 \ll 2\Delta$. The last condition may not be satisfied for samples with relatively low superconducting temperatures. The connection with amplitude excitation $\eta_{13}(k,\Omega)$ which lies higher than 2Δ and is therefore strongly attenuated is no longer weak in the system (22), and the low-lying mode will also attenuate. This broadens the absorption line and thus shifts the absorption edge into the low-frequency region. Note that in a number of experiments[15-17] were observed spectrum singularities whose frequency position on the T_c scale is $\omega/k_B T_c \simeq 8$. It is remarkable that a trace of these singularities is preserved also at a temperature $T \gtrsim T_c$.[16,17] In our opinion this singularity is made to contribute to the absorption by the quasiparticle generated as a result of pair breaking by ,he external field, and reflects the state-density singularity connected with the presence of dielectric correlations.[31]

5. CONCLUSION

We have considered here the spectrum of the collective excitations and their manifestations in the optical spectra of superconductors with dielectric correlations. The latter make the system close to a ferroelectric transition in a nonsuperconducting phase and is the cause of the high temperature of the superconducting transition. The proximity of the system to ferroelectricity was described by us in terms of excitonic instability of a doped semiconductor with nonzero interband momentum matrix element. The last circumstance is not decisive, although it does simplify the calculation considerably. In the case of proximity to an instability of the charge-density-wave type, which cannot be excluded from high-temperature superconductors based on $YBa_2Cu_3O_{7-\delta}$ in the region of superconductive compounds in view of the presence of almost congruent sections on the Fermi surface as $\delta \to 0$, ferroelectric fluctuations are possible as a manifestation of the proximity to the point of extrinsic ferroelectric ordering[9] and our present results remain qualitatively in force.

In contrast to Refs. 18–20, dealing with Raman-active amplitude oscillations of the superconducting and dielectric (CDW) order parameters, we consider IR-active phase oscillations. Under conditions of sufficiently strong electron–phonon interaction, the oscillations considered by us may be manifested also in Raman-scattering spectra, particularly as singularities of the form of the phonon lines (Fano antiresonance). In all likelihood, however, unequivocal separation of their contribution to Raman-scattering spectra is a very complicated task. A more reliable method of observing the considered collective excitation is IR spectroscopy.

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