Anomalies in the isotope effect in high-T_c superconductors: evidence of a plasmon mechanism for superconductivity

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Previous research has revealed that the exponent of the oxygen isotope shift decreases with increasing superconducting transition temperature T_c in the layered cuprate metal oxides. It is shown in the present paper that this decrease can be explained by a plasmon mechanism for superconductivity. This mechanism involves a Cooper pairing of light current carriers in a wide 2D band. The pairing results from the exchange of virtual quanta of low-frequency collective excitations of the charge density of heavy carriers in a narrow 2D band. These virtual quanta are acoustic plasmons, which hybridize with oxygen vibrational modes.

1. The high-T_c superconductors exhibit an anomalously weak isotope effect.[1,2] Various types of cuprate metal oxides also display a tendency for the exponent of the isotope shift in terms of oxygen to decrease with increasing T_c (Refs. 2 and 3). These pieces of evidence suggest a nonphonon mechanism for high-T_c superconductivity,

since the conventional (low-temperature) superconductors usually[1)] exhibit the opposite tendency: a decrease in the isotope-shift exponent with decreasing T_c (Ref. 3).

Frank *et al.*[7] recently measured the isotope effect resulting from the substitution of ^{18}O for ^{16}O in the compound $(Pr_xy_{1-x})Ba_2Cu_3O_{7-\delta}$. They varied the concentration of the dopant Pr, which suppresses the superconductivity (i.e., which lowers T_c), by reducing the density of holes (because the valence of Pr is higher than that of Y). Frank *et al.*[7] observed that the exponent (α_0) of the oxygen isotope shift drops sharply with decreasing x and with increasing T_c (it drops from $\alpha_0 \approx 0.5$ at $x = 0.5$ and $T_c \approx 30$ K to $\alpha_0 \approx 0.02$ at $x = 0$ and $T_c \approx 90$ K).

In the present letter we show that a sharp drop of this sort in the isotope-shift exponent with increasing T_c is characteristic of a "plasmon" superconductivity mechanism[8] in a layered metal in which a narrow 2D band near the Fermi level is partially filled by "heavy" (h) charge carriers. A necessary condition here is that the Cooper pairing of the degenerate "light" (l) current carriers in a broad 2D band result from an exchange of virtual acoustic plasmons. According to Refs. 8 and 9, such plasmons easily hybridize with optical phonons (in particular, with dipole-active vibrations of oxygen ions in CuO_2 layers).

2. According to Ref. 10, when h carriers are nearly localized at lattice sites in a narrow 2D band,[2)] with a one-particle spectrum $E_h(k_xk_y) = W_h/4(\cos k_xa + \cos k_ya)$, the spectrum of acoustic plasmons lies above the region of a strong quantum Landau damping involving h carriers. That assertion holds in the strong-coupling approximation (W_h is the width of the narrow band, and a is the lattice constant in the plane of the layers) and under the condition $W_h < e^2/a\epsilon_\infty$, where ϵ_∞ is the rf dielectric constant of the crystal. Under these conditions, the spectrum of acoustic plasmons hybridizes with optical phonons (oxygen vibrational modes) throughout the Brillouin zone.[8]

We assume that there is an electron–plasmon coupling with acoustic plasmons, and we assume that there is an electron–phonon coupling with only one optical branch: that corresponding to vibrations of O^{2-} oxygen ions in the cuprate (CuO_2) layers. The effective retarded interaction between the degenerate l carriers can be written in this case as[8,10]

$$\tilde{V}_{ll}(\vec{q},\omega) = D_{pl}(\vec{q},\omega) + \tilde{V}_C(\vec{q}). \tag{1}$$

Here $D_{pl}(\vec{q},\omega) = V_C(\vec{q})\cdot[\tilde{\epsilon}^{-1}(\vec{q},\omega) - \epsilon^{-1}(\vec{q},\omega_{\max})]$ is the plasmon Green's function, $V_C(\vec{q})$ and $V_C(\vec{q}) \equiv V_C(\vec{q})/\epsilon(\vec{q},\omega_{\max})$ are matrix elements of the seed and screened Coulomb repulsion, and $\tilde{\epsilon}(\vec{q},\omega)$ is a generalized dielectric constant of a two-band metal. This dielectric constant is given by

$$\tilde{\epsilon}(\vec{q},\omega) = \epsilon_i(\vec{q},\omega) - V_C(\vec{q})[\Pi_l(\vec{q},\omega) + \Pi_h(\vec{q},\omega)] - \frac{\omega_{LO}^2(\vec{q}) - \omega_{TO}^2(\vec{q})}{\omega^2 - \omega_{TO}^2(\vec{q})}, \tag{2}$$

where ϵ_i is the part of the dielectric constant which stems from the polarization of the ion lattice and interband transitions; Π_l and Π_h are the polarization operators of the l and h carriers; ω_{LO} and ω_{TO} are the frequencies of longitudinal and transverse optical phonons; and $\omega_{\max}$ is the limiting or cutoff energy of the electron–plasmon coupling. This energy is equal in order of magnitude to the Fermi energy of the l carriers, E_{Fl}.

At energies $W_h \sin(q_\| a/2) < |\omega| < \Omega_\pm(\vec{q})$, where $\Omega_\pm$ are the frequencies of hybrid phonon–plasma vibrations,[8,9] there is an interelectron attraction $\mathrm{Re}\,\tilde{V}_{ll}(\vec{q},\omega) \equiv V_C(\vec{q})\mathrm{Re}\,\tilde{\epsilon}^{-1}(\vec{q},\omega) < 0$. At momentum-transfer values $q_\| \approx 2k_{Fl}$ (k_{Fl} is the Fermi momentum of the l carriers)—these are the values which dominate the Cooper pairing of quasiparticles in a quasi-2D metal with a cylindrical Fermi surface—we have $\mathrm{Re}\,\tilde{\epsilon}(\vec{q},\omega) = 0$, and $\mathrm{Im}\,\tilde{\epsilon}^{-1}(\vec{q},\omega)$ goes through a maximum (under the condition $\omega_{LO}^2 \gg \omega_{TO}^2$) at the frequency

$$\tilde{\Omega}_+ \equiv \Omega_+(2k_{Fl}) \approx \left[\frac{\Omega_h^2 k_{Fl} d + \omega_{LO}^2}{1+\alpha_\infty}\right]^{1/2} \tag{3}$$

Here Ω_h is the plasma frequency of the h carriers in the narrow 2D band, d is the distance between CuO_2 layers (or between stacks of CuO_2 layers), $\alpha_\infty = e^2/\epsilon_\infty v_{Fl}$ is a dimensionless density parameter, and $v_{Fl} = k_{Fl}/m_l^*$ is the Fermi velocity of the l carriers, with an effetive mass m_l^*.

Using the Kramers–Kronig relation for $\tilde{\epsilon}^{-1}(\vec{q},\omega)$, we can show that the dimensionless electron–plasmon interaction constant is given by

$$\lambda_{pl} = -\frac{2}{\pi}\nu_l \int_0^\infty \frac{d\omega}{\omega} < V_C(\vec{q})\mathrm{Im}\tilde{\epsilon}^{-1}(\vec{q},\omega) >, \tag{4}$$

where $\nu_l = m_l^*/2\pi$ is the density of states in the wide 2D band, and $\langle ... \rangle$ means an average over the Fermi surface. In crystals with strong ionic binding, in which the static dielectric constant of the crystal satisfies $\epsilon_0 \gg \epsilon_\infty$, the electron–plasmon coupling constant is the same, within small terms on the order of $\epsilon_\infty/\epsilon_0$ and $\tilde{\Omega}_+/E_{Fl}$, as the constant of the Coulomb repulsion at high energies ($\omega \sim \omega_{\max} \sim E_{Fl}$). In this case the static screening by l carriers is suppressed by dynamic retardation effects:

$$\mu_C = \nu_l < V_C(\vec{q})\mathrm{Re}\tilde{\epsilon}^{-1}(\vec{q},\omega_{max}) > \approx \alpha_\infty/2. \tag{5}$$

In the cuprate metal oxides of the $Bi_2Sr_2Ca_{n-1}Cu_nO_x$ and $Tl_mBa_2Ca_{n-1}Cu_nO_x$ types ($m = 1,2$), with stacks of n ($n = 1$–5) CuO_2 layers in a unit cell of the crystal, the constants λ_{pl} and μ_C are multiplied by a structure factor $\beta(n)$. This can be seen easily by working from the equations for the normal and anomalous eigenenergy parts of the l carriers. If the tunneling of electrons between layers is slight, this structure factor is essentially equal to the number of layers, n (in other words, the contributions of the individual CuO_2 layers are nearly additive). In this case the polarization operators for the l and h carriers, which contain products of two one-particle Green's functions, are multiplied by a structure factor $\tilde{\beta}(n) \approx n^2$. The Coulomb matrix element at $q_\| \approx 2k_{Fl}$ is

$$V_C(q_\|, n) \approx \frac{2\pi e^2}{q_\|} c(n); \quad c(n) = d + (n-1)d_0, \tag{6}$$

if $2k_{Fl}d \gg 1$ and $2k_{Fl}d_0 \gg 1$, where d_0 is the distance between CuO_2 layers in the stacks, d is the distance between stacks, and $c(n)$ is the lattice constant along the z axis.

3. To calculate T_c, we work from a refined version of the McMillan formula[13] which was derived in the intermediate-coupling approximation in Ref. 14. For a spectrum with a single vibrational mode, of frequency $\tilde{\Omega}_+$, and under the condition $\lambda_{pl} \approx \mu_C$, this formula takes the simple form

$$T_c \approx \tilde{\Omega}_+ \exp\{-1/\Lambda\}, \tag{7}$$

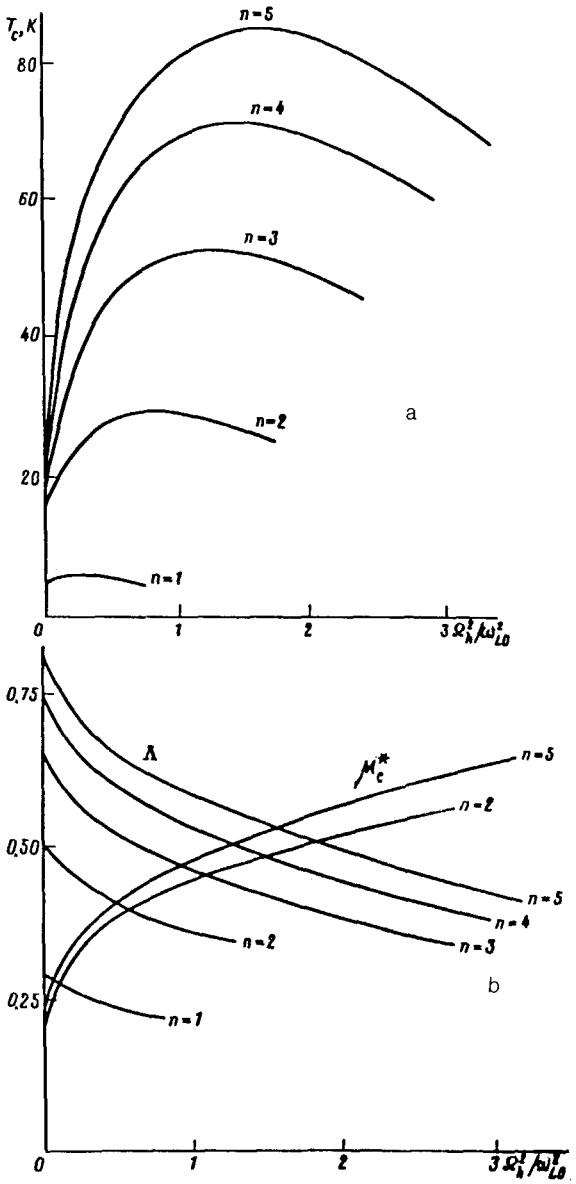

FIG. 1.

where

$$\Lambda = \frac{\mu_C - \mu_C^*(1+\mu_C \ln 2)}{1+\mu_C \ln 2}; \quad \mu_C^* = \frac{\mu_C}{1+\mu_C \ln(E_{Fl}/\tilde{\Omega}_+)}. \tag{8}$$

Figure 1, a and b, shows $T_c\Lambda$ and μ_C^* versus the ratio Ω_h^2/ω_{LO}^2, according to expressions (7) and (8) for $E_{Fl} \approx 0.25$ eV, $\omega_{LO} \approx 320$ K, $k_{Fl} \approx 4\times 10^7$ cm^{-1}, $d \approx 10$ Å, $m_l^* \approx 2m_0$ (m_0 is the mass of a free electron), and $\epsilon_\infty \approx 4$, so we have $\alpha_\infty \approx 2$ and $\mu_C \approx 1$. We see that the theoretical behavior of T_c as a function of Ω_h^2, i.e., as a function of the density of the h carriers, N_h, is in qualitative agreement with the experimental behavior of T_c as a function of the dopant concentration or the oxygen

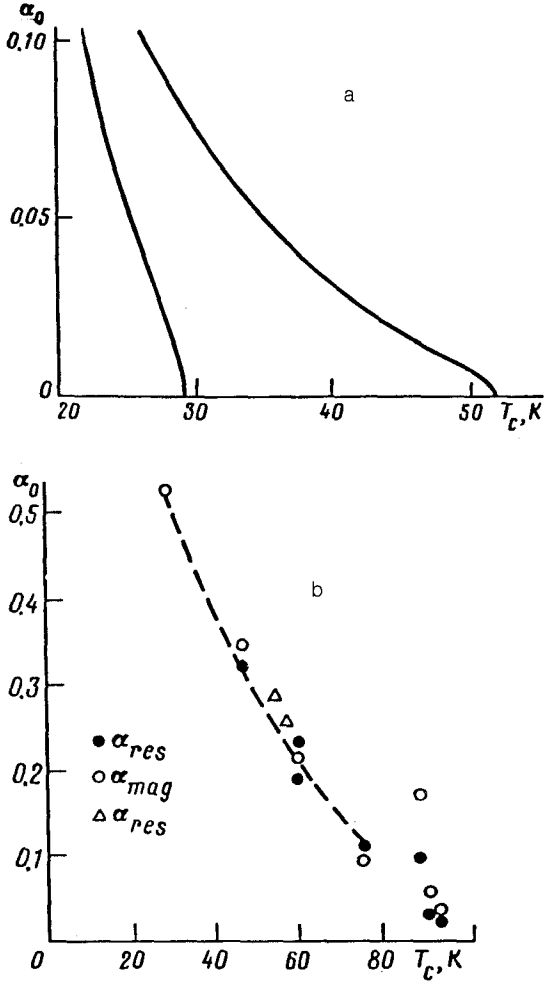

FIG. 2.

concentration (i.e., as a function of the number of holes per layer in the unit cell[15,16]). The reason is that the Fermi level is "pinned" ($E_{Fl} \approx$const) as the narrow band is being filled, and an increase in the number of holes corresponds to an increase in the density of h carriers, while the density of l carriers remains nearly constant ($k_{Fl} \approx$const, $\alpha_\infty \approx$const). Even for $T_c^{\max} \approx 85$ K (with $n = 5$), the condition for the applicability of the intermediate-coupling approximation, $\Lambda(T_c/\tilde{\Omega}_+)^2 \ll 1$, holds.

Working from (7) and (8), and using (3), we find the following expression for the exponent of the oxygen isotope shift (for $n \geqslant 1$):

$$\alpha_0 \equiv \frac{1}{2}\frac{\partial \ln T_c}{\partial \ln \omega_{LO}} \approx \frac{\omega_{LO}^2}{2(\omega_{LO}^2 + n^2\Omega_h^2 k_{Fl} d)}[1 - (\mu_C^*/\Lambda)^2]. \tag{9}$$

We have replaced the constant μ_C by $n\mu_C$ everywhere. It follows from (9) that the isotope effect is weakened in this case because the Coulomb repulsion is far stronger than in conventional superconductors ($\mu_C^* \approx 0.25$–0.65 in contrast with $\mu_C^* \approx 0.11$; Ref. 13) and also because LO phonons (oxygen vibrational modes) hybridize with acoustic plasmons. Figure 2a shows the theoretical behavior of α_0 as a function of T_c for the ascending branch[3)] of the plot of T_c versus Ω_h^2, up to $T_c^{\max}$, for various values of n. These results were found from Eqs. (7)–(9) for the same parameter values as in Fig. 1a. Figure 2b shows the experimental behavior[4)] of α_0 versus T_c according to Ref. 7, for the variable-composition compound $(Pr_x Y_{1-x})Ba_2Cu_3O_{7-\delta}$, with x from 0.5 to 0. We see a complete qualitative agreement between the theoretical and experimental curves.

In summary, the clear agreement of the theory with experimental data on the behavior of T_c as a function of N_h and n (Fig. 1) and on the behavior of α_0 as a function of T_c (Fig. 2) is evidence that a plasmon superconductivity mechanism[8] may play an important role in high-T_c superconductivity.

[1)]Exceptional cases are the hydrides PdH(D) and PtH(D), which have a negative isotope effect with $T_c \approx 10$ K (Ref. 4), and $La_{1-x}Sr_xCuO_4$, in which the exponent of the oxygen isotope shift drops sharply at the point of the maximum, $T_c \approx 40$ K (the drop is from $\alpha_0 \approx 0.6$ to $\alpha_0 \approx 0.1$).[5] This drop apparently results from a pronounced anharmonicity of the phonons in the vicinity of a lattice instability.[6]

[2)]The notion of a narrow band with a high density of states has found support in the "pinning" of the Fermi level which was observed in Refs. 11 and 12 at various doping levels of cuprate metal oxides.

[3)]For the descending branch of the plot of T_c versus Ω_h^2, the isotope effect is negative ($\alpha_0 < 0$) in this model. When the electron–phonon coupling with other oxygen modes is taken into account, the isotope-shift exponent becomes positive, but it remains anomalously small.

[4)]A similar result was found for $YBa_{2-x}La_xCu_3O_x$ in Ref. 17.

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