Letter

Theory of superconductivity due to Ngai's mechanism in lightly doped $SrTiO_3$

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We develop a theory of superconducting pairing in low-density strontium titanate due to the quadratic coupling of electron density to soft transverse optical phonons first proposed by Ngai [Phys. Rev. Lett. **32**, 215 (1974)]. This leads to a static attractive potential between electrons with decay length $l_{\rm eff}$ that scales inversely with soft optical gap ω_T. For low electron densities $n \leqslant 10^{18}$ cm^{-3}, attraction between electrons is static and local in space; thus the transition temperature T_c was found using known results for low-density electron gas. The $T_c(n)$ dependence for low doping was calculated and found to be in agreement with experimental data. In addition, we show that suppression of T_c by hydrostatic pressure and strong increase in T_c due to isotope substitution ^{16}O$\rightarrow^{18}$O observed experimentally also can be explained within our theory.

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I. INTRODUCTION

Strontium titanate (STO) is a wide-gap band insulator known for more than half a century for its properties related to its proximity to a ferroelectric transition; see Refs. [1–3] for recent reviews. It can be transformed into a dilute metal by tiny doping. The key feature of this metal which makes it very different from the majority of metals and doped semiconductors is that Coulomb interaction is nearly absent due to extremely high low-temperature static dielectric permeability $\epsilon_0 \approx 20\,000$; see Ref. [4]. This results in a huge value of the effective Bohr radius, $a_B \approx 600$ nm, compared with normal values of $\sim$0.1 nm for usual metals. Therefore Coulomb interaction, which is the major interaction in metals normally, appears to be nearly irrelevant for a very dilute STO-based conductor. Surprisingly enough, lightly doped STO becomes a superconductor in a wide range of conduction electron densities, 10^{17} cm$^{-3} \leqslant n \leqslant 10^{20}$ cm^{-3}.

Although the first report of superconductivity in STO is dated as early as 1964 (see Ref. [5]), the mechanism of electron pairing is still under active debate (examples can be found in Refs. [1–3]). The key feature of superconductivity in STO is due to its very low Fermi energy E_F, which is much less than Debye energy $\hbar\omega_D \approx 100$ meV. As a result, the classical theory of superconductivity based on Migdal-Eliashberg equations [6] could not be applied here. An attempt to circumvent this problem was made in Ref. [7], where pairing due to exchange by very soft plasmons was proposed. However, the results obtained for T_c were found to be strongly off the data, if the measured value of dielectric constant $\epsilon_0 \approx 20\,000$ is used. The proximity of STO to the ferroelectric critical point is surely the crucial feature of this material. Edge *et al.* [8] pointed out the relevance of this criticality to superconductivity; specifically, they predicted a strong increase in T_c upon isotope substitution ^{16}O$\rightarrow^{18}$O, since the latter is known to produce ferroelectricity when about $\frac{1}{3}$ of O atoms are replaced by heavy oxygen [9]. Such an effect was indeed observed soon [10]: 35% substitution ^{16}O$\rightarrow^{18}$O increases T_c by a factor of $\sim$1.5 and also increases the upper critical field nearly twice. An important step forward was made in Ref. [11], which gave support to an early idea [12] about the relevance of coupling between electron density and two soft transverse optical (TO) phonons, the existence of which was known since the publication of Ref. [13]. Namely, Ref. [11] provides arguments based on analysis of optical absorption spectra in favor of a large magnitude of this electron coupling to two TO phonons, of the form $\psi^\dagger\psi\mathbf{u}^2$, where ψ is the electron annihilation operator and $\mathbf{u}$ is the TO phonon displacement amplitude. The very low gap known for these phonons at low temperatures, $\hbar\omega_T \sim 1.5$ meV, is directly related to the large ϵ_0 value, $\omega_T \propto 1/\sqrt{\epsilon_0}$.

In this Research Letter we employ the idea first proposed by Ngai [12] and recently developed in Ref. [11]. We use quadratic coupling between electron density and TO phonons as phenomenological input for our theory. We concentrate on the lowest-density limit $n < n_{c1} \approx 1.5 \times 10^{18}$ cm^{-3}, where a single-band Fermi liquid in realized [14], and demonstrate that electron-electron interaction mediated via two TO phonons leads to a consistent description of superconducting $T_c(n)$ evolution with n (Ref. [14]) and of its giant isotope effect [10]. Pairing by two-phonon exchange differs a lot from single-phonon exchange but leads to a simple picture: Electrons attract each other via a static potential which decays with distance as $-V(r) \propto r^{-3}e^{-2r/l_0}$, where $l_0 = s/\omega_T \approx 3.3$ nm is the characteristic length related to the soft polarization TO mode; for the velocity of the TO mode we use $s = 7.5 \times 10^5$ cm/s (see Ref. [15]). At the low electron densities that we consider, $k_F l_0/2 < 0.6$ and electron-electron (e-e) scattering can be considered short range. Frequency dispersion of e-e scattering occurs then at the energy scale $\epsilon \geqslant \omega_T$, and it is relatively weak in the low-density region, since $E_F(n)$ does not exceed ω_T considerably.

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Superconductivity in a bulk Fermi gas with local attraction was studied theoretically by Gor'kov and Melik-Barkhudarov [16]; see also a more recent paper [17]. They found an expression for T_c that is similar to the one known for the usual BCS theory, with the major exception that Debye energy is replaced by Fermi energy in the prefactor, $T_c \approx 0.27 E_F e^{-1/\lambda_0}$. Dimensionless coupling constant $\lambda_0 = \nu_0 V_0$, where V_0 is the renormalized electron-electron scattering potential in the $l = 0$ scattering state (s wave) and ν_0 is the density of states at the Fermi level per single spin projection. We demonstrate that attractive short-range potential does indeed appear due to two-TO-phonon exchange and calculate T_c as a function of electron density n.

The rest of this Research Letter is composed as follows: In Sec. II we derive an effective attraction potential between electrons, and Sec. III is devoted to the calculation of superconducting transition temperature T_c as a function of the conduction electron density. Section IV contains analysis of the T_c suppression due to hydrostatic pressure, while Sec. V contains a theory of the anomalous isotopic effect in STO. Finally, Sec. VI contains our conclusions.

II. ELECTRON-ELECTRON INTERACTION MEDIATED BY A PAIR OF TO PHONONS

We start with the action for a coupled electron-phonon system close to the ferroelectric transition:

$$\begin{aligned} S &= S_e^{(0)} + S_{ph}^{(0)} + S_{\rm int}, \\ S_{ph}^{(0)} &= \frac{\rho_m}{2}\int d^3x dt \left[\dot{u}_\alpha^2 - s^2(\nabla_\beta u_\alpha)^2 - \omega_T^2 u_\alpha^2\right], \\ S_{\rm int} &= -g\rho_m \int d^3x dt (\bar{\psi}\psi) u_\alpha^2, \end{aligned} \tag{1}$$

and $S_e^{(0)}$ is just the action of free-electron gas with effective mass $m_e = 1.8 m_0$, according to the data from Ref. [18] for low electron densities in STO, m_0 being the free-electron mass. Here, u_α is the displacement coordinate for a TO soft optical phonon, $\psi(x)$ is the electron field operator, and $\rho_m = 5.11$ g/cm^3 is the mass density of STO. The action $S_{ph}^{(0)}$ describes long-wavelength TO phonons with momenta $q \ll K_{\rm BZ}$, where $K_{\rm BZ}$ is the boundary of the Brillouin zone (BZ). Whenever a high-q cutoff is needed in the further calculations, we introduce it by using the simplest TO phonon lattice spectrum of the form appropriate for a cubic lattice, with the BZ including $p_{x,y,z} \in (-\frac{\pi}{a}, \frac{\pi}{a})$:

$$\omega^2(p) = \omega_T^2 + \frac{4s^2}{a^2}\left(\sin^2\frac{p_x a}{2} + \sin^2\frac{p_y a}{2} + \sin^2\frac{p_z a}{2}\right), \tag{2}$$

where $a \approx 0.4$ nm is the lattice spacing. The coupling constant g in Eq. (1) has natural dimension length^3/time^2; we represent it in the form

$$g = \lambda a^3 \omega_L^2, \tag{3}$$

where $\hbar\omega_L = 0.1$ eV is the largest longitudinal optical gap of STO and $\lambda \sim 1$ is the dimensionless coupling constant of the problem. This is the only fitting parameter in our model, so the exact way of making it dimensionless is unimportant.

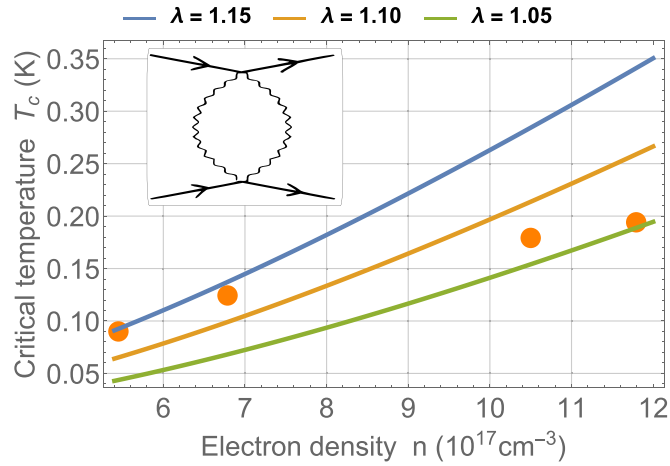

FIG. 1. Critical temperature as a function of conduction electron density for several values of electron-phonon interaction constant λ. Orange points represent experimental data from Ref. [14]. Inset: diagram corresponding to the static interacting potential between electrons. Thick (thin wavy) lines denote electron (phonon) Green's functions.

The static interacting potential between two electrons can be obtained from the action (1) by integrating out the phonons; the corresponding diagram is shown in Fig. 1.

Its analytical expression in Fourier space is

$$\begin{aligned} V(\omega, q) = &-4\hbar g^2 \rho_m^2 \int \frac{d\epsilon}{2\pi} \\ &\times \int_{\rm BZ} \frac{d^3p}{(2\pi)^3} D_0(\epsilon, \mathbf{p}+\mathbf{q}) D_0(\omega-\epsilon, \mathbf{p}), \end{aligned} \tag{4}$$

where the momentum integration goes over the simple cubic Brillouin zone and the phonon Green's function $D_0(\epsilon, \mathbf{p})$ is given, in the imaginary-time formalism, by

$$D_0(\epsilon, \mathbf{p}) = \frac{1}{\rho_m}[\epsilon^2 + \omega^2(p)]^{-1}. \tag{5}$$

The minus sign in Eq. (4) appears since we deal here with the second order of expansion over virtual phonons. Factor 4 in Eq. (4) comes about due to two variants of pairing in the average $\langle u^2(0)u^2(x)\rangle$, and two independent polarizations of TO phonons.

Integration over $d\epsilon$ is trivial; using Eqs. (2)–(4), we come to

$$V(0,0) = -\hbar\lambda^2 \frac{\left(a^3\omega_L^2\right)^2}{s^3} \int_{\tilde{\rm BZ}} \frac{d^3\tilde{p}}{(2\pi)^3} \frac{1}{[\kappa^2 + a^2\omega^2(\tilde{p})/s^2]^{3/2}}, \tag{6}$$

where $\tilde{\mathbf{p}} = a\mathbf{p}$ is dimensionless momentum and $\kappa = a\omega_T/s = a/l_0$. The dimensionless integral in Eq. (6) is equal to $J/2\pi^2$, where J is logarithmically large due to the smallness of $\kappa \approx 0.12$. We compute J numerically and find

$$V(0,0) = -\hbar\lambda^2 \frac{\left(a^3\omega_L^2\right)^2}{2\pi^2 s^3} \ln\frac{\eta}{\kappa} \equiv -W \ln\frac{\eta}{\kappa}, \tag{7}$$

where $\eta \approx 5.76$. To estimate T_c below, we will need to know $V(0,q)$ more accurately, up to the term $\sim q^2$. We find this additional term by expansion over q in the integral (4). The

resulting integral converges fast at large p, so no lattice cutoff is needed. Finally, we get

$$-V(0,q) = W\left[\ln\frac{\eta}{\kappa} - \frac{(ql_0)^2}{12}\right]. \quad (8)$$

Note that dependence on q is relatively weak, as well as dependence of $V(\epsilon, q)$ on ϵ (to be discussed below); the reason is that the major (logarithmic) contribution to the integral in Eq. (4) comes from TO phonons with momenta p in a broad range $k_F \leqslant p \leqslant \pi/a$. For the same reason, the effects of renormalization of the phonon spectrum due to interaction with electrons are weak at low concentrations $n_e \leqslant 1.5 \times 10^{18}$ cm^{-3}; we will discuss these effects later on. For completeness, we provide the e-e potential in coordinate space at $r \geqslant a$ (K_1 is the Macdonald function):

$$-V(r) = \frac{W}{2\pi l_0 r^2} K_1\left(\frac{2r}{l_0}\right). \quad (9)$$

At large distances, $V(r)$ decays exponentially with length $l_0/2$, while at shorter lengths $a < r < l_0/2$ this potential behaves as $V(r) \approx -W/4\pi r^3$.

III. SUPERCONDUCTING TRANSITION TEMPERATURE

Attractive e-e interaction defined by Eqs. (8) and (9) decays exponentially at $r > l_0/2$, so it can be considered as nearly local in the range of electron densities

$n < n_{c1} = 1.5 \times 10^{18}$ cm^{-3}. Then we can employ the theory [16] for superconductivity in a Fermi gas with local instantaneous attraction. The result [16] for T_c is

$$T_c = \zeta\, E_F\, \exp\left(-\frac{1}{\nu_0 V_0}\right), \quad \zeta = \frac{e^C}{\pi}\left(\frac{2}{e}\right)^{7/3} \approx 0.27, \quad (10)$$

where $\nu_0 = m_e k_F/2\pi^2\hbar^2$ is the density of states (DOS) per one spin projection and V_0 is the $l = 0$ harmonics of the pairing potential (8) evaluated at the Fermi surface (FS). Assuming the FS to be spherical (which is a good approximation for STO at low densities), we find (here, θ is the azimuth angle at the FS, so $q = 2k_F \sin\frac{\theta}{2}$)

$$V_0 = \frac{1}{2}\int_0^\pi |V(0,q)| \sin\theta d\theta = W\left(\ln\frac{\eta}{\kappa} - \frac{1}{6}k_F^2 l_0^2\right). \quad (11)$$

The plot of $T_c(n)$ dependence which follows from Eqs. (10) and (11), together with the definition of W in Eq. (7), is shown in Fig. 1 together with the data from Ref. [14]. The choice of electron-phonon coupling constant $\lambda = 1.1$ provides the best correspondence with the data. Some discrepancy is still present, and it will be discussed below. In a recent paper [19] the same type of electron coupling to TO phonons was employed to study theoretically high-temperature transport properties of lightly doped STO; comparison of this theory's predictions with the data [20] provides the value of the coupling constant $\lambda \approx 0.9$.

IV. SUPPRESSION OF T_c BY HYDROSTATIC PRESSURE

The relevance of STO proximity to the ferroelectric critical point to the origin of superconductivity was discussed by Enderlein *et al.* [21]. In particular, they present data on the effect of hydrostatic pressure on T_c and on dielectric constant ϵ (see Fig. 2 in Ref. [21]) which demonstrate that decrease in ϵ leads to a sharp fall in T_c. According to the standard Lyddane-Sachs-Teller relation, $\epsilon(P)/\epsilon = [\omega_T/\omega_T(P)]^2$, where argument P identifies pressure-modified values. Using the data from Fig. 2 b of Ref. [21], we calculated $T_c(P)$ for two values of electron density, $n_e = 5.4 \times 10^{17}$ cm^{-3} and $n_e = 1.2 \times 10^{18}$ cm^{-3}. The result is presented in Fig. 2. Accurate comparison of our prediction for T_c suppression with the corresponding data given by Enderlein *et al.* [21] is not possible since they studied the sample with much higher electron density $n = 3.4 \times 10^{19}$ cm^{-3}, but the overall trend is similar. Our theory predicts a bit smaller suppression effect—a factor of 2.5 between $P = 0$ and $P = 4$ kbar—while the experiment in Ref. [21] provides a suppression factor close to 4, at an electron density which is 30 times higher. One cannot exclude that hydrostatic pressure may decrease a little the coupling constant λ, which would lead to additional suppression of T_c, not accounted for in our results in Fig. 2.

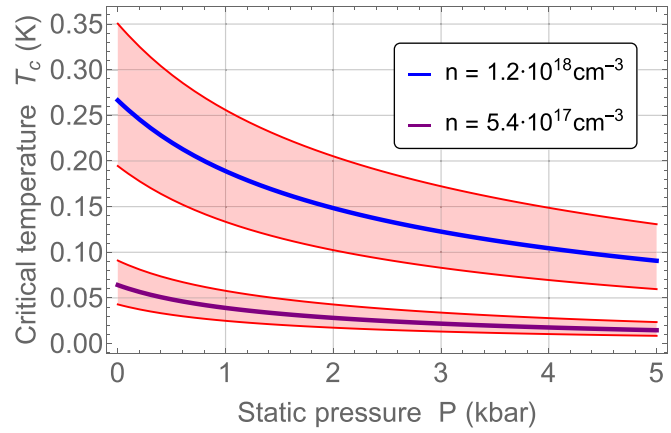

FIG. 2. Plot of critical-temperature–pressure dependence for the two values of electron density indicated in the plot, obtained with $\lambda = 1.1$ and $\omega_T(P)$ dependence extracted from the data of Ref. [21]. Shaded areas around each line represent variation in λ from 1.05 (bottom red line) to 1.15 (top red line).

V. ISOTOPIC ENHANCEMENT OF T_c

In classical weak-coupling superconductors with a phonon mechanism of e-e attraction, isotopic substitution of some part of the atoms by their heavier isotopes leads usually to weak suppression of T_c with an increase in the typical atom mass M. The reason is that $T_c = 1.13\omega_D e^{-1/\lambda_{\rm eff}}$ is proportional to the Debye frequency $\omega_D \propto 1/\sqrt{M}$, while the effective coupling constant $\lambda_{\rm eff}$ is independent of ω_D; the latter statement follows from Eliashberg theory [6]. Experimental data presented in Ref. [10] demonstrate a sharp departure from the usual behavior: Substitution of 35% of oxygen atoms ^{16}O by their heavy isotope ^{18}O resulted in a factor of $\approx$1.5 rise in T_c for the whole range of studied electron densities, 4×10^{18} cm^{-3} $< n < 4 \times 10^{20}$ cm^{-3}. Another set of data demonstrating the same effect (for higher electron densities) can be found in Ref. [22].

To explain this giant positive isotope effect, we note that under such an isotope substitution, insulating STO becomes

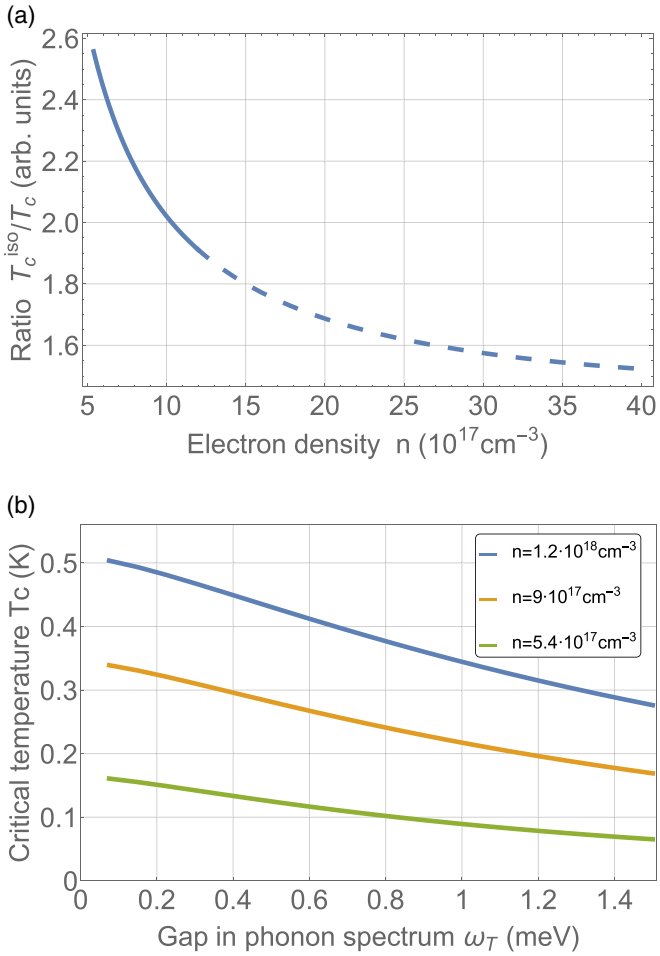

FIG. 3. (a) Plot showing the dependence of the ratio between critical temperature for isotope-substituted STO and that for native STO on electron density with $\lambda = 1.1$; the right part of the line is dashed since at these higher densities our present theory is expected to be valid qualitatively. (b) Critical temperature as a function of the TO phonon gap ω_T for different values of the electron density.

ferroelectric [9]. This means that in the isotope-modified material the TO gap $\omega_T \to 0$. To take this into account within our theory, we need just to calculate the amplitude V_0 at $\omega_T = 0$. Now the logarithmic integral in Eq. (6) diverges at the lower limit, while for $V(q)$ we find, instead of Eq. (8), the following result:

$$\tilde{V}(q) = -W \ln \frac{\eta e}{qa}. \tag{12}$$

Calculating the $l = 0$ scattering amplitude as in Eq. (11), we find

$$\tilde{V}_0 = -W\left(\ln \frac{\eta e^{3/2}}{2k_F a}\right). \tag{13}$$

We calculated, using Eqs. (13) and (11), the ratio of transition temperatures for isotope-substituted and native STO for the range of electron densities n; the results are shown in Fig. 3(a).

The data in Ref. [10] provide an increase in T_c by a factor of 1.5 at higher concentrations, starting from $n = 4 \times 10^{18}$ cm^{-3}, which is in agreement with our results in Fig. 3(a). For a weaker isotopic substitution, ω_T can be only partially suppressed, so the enhancement of T_c will be smaller. We calculated the transition temperature $\tilde{T}_c$ as a function of the partially suppressed TO gap ω_T. This was done by means of numerical integration starting from the $\omega = 0$ version of Eq. (4); the result is presented in Fig. 3(b) and can serve as a prediction for future experiments with isotope-substituted STO.

VI. CONCLUSIONS

We developed a theory able to predict the superconducting transition temperature in lightly doped STO as a function of conduction electron density $n \leqslant n_{c1} = 1.5 \times 10^{18}$cm^{-3}; the theory has a single fitting parameter λ which determines the electron coupling to a pair of TO phonons. Comparison with the data for $T_c(n)$ from Ref. [14] selects the optimal value of this parameter $\lambda = 1.1$, which is close to the value 0.9 found by a totally different method in Ref. [19]. The small discrepancy between these numbers may result from (i) inaccuracy of the method used in Ref. [19] (we estimate it as 5–10%) and (ii) possible modification of the coupling strength λ at high temperatures where nonharmonicity of the TO mode is substantial. We note that the estimate for the effective strength of e-e attraction $\lambda_{2ph} \approx 0.28$ found in Ref. [11] for much higher values of n_e does not include the large logarithmic factor we discovered; see Eq. (7). It is due to integration over a large phase volume of two virtual TO phonons.

Our theory provides good qualitative agreement with experiments on $T_c(n)$ dependence and on T_c effects due to isotopic substitution [10,22] and hydrostatic pressure [21]. We emphasize that strong suppression of Coulomb repulsion by large ϵ is crucial for the local static attraction to prevail, since the usual dynamic mechanism of Coulomb suppression (due to the Tolmachev-McMillan logarithm) is not at work due to low Fermi energy.

Lightly doped STO is not the only low-density material where coupling due to near-critical ferroelectric modes may be instrumental for the electron pairing and superconductivity. In particular, superconductivity in monolayer WTe_2 was observed in Ref. [23] in a very dilute regime with electron-electron distance of about 5 nm; moreover, bilayer WTe_2 is known [24] to be ferroelectric, which may indicate proximity to ferroelectricity in the WTe_2 monolayer. It would be very interesting to search for the presence of a soft ferroelectric mode in a monolayer of WTe_2; its possible observation would pave a possible way to a theory of superconductivity in this interesting material.

The limitation of low densities used in our theory was chosen, in the first place, in order to concentrate on the simplest situation of a single band filled by conduction electrons; at higher n, the second band starts to be filled [14], and more involved calculations are necessary. There are a few effects which we have neglected so far because they are present, in principle, in the single-band problem as well. The first of these is the renormalization of the effective phonon gap ω_T due to the presence of the coupling to electrons. Apparently, it is given by replacement $\omega_T^2 \to \omega_T^2 + 2gn$, which could lead to a considerable effect even in our range of n. However, comparison with the data [25] for $n > 10^{19}$ cm^{-3} shows a much smaller increase in the gap, compatible with $\Delta\omega_T^2 \approx 0.3gn$,

which does not lead to any noticeable effect at $n \leqslant n_{c1}$. The difference between the data from Ref. [25] and naive expectations is probably due to the fact that TO phonons interact both with conduction electrons and with ion defects (O deficiency or Nb substitution), and these defects partially suppress the increase in the gap caused by electrons.

The second relevant effect is due to the frequency dependence of the effective e-e interaction, Eq. (4). Indeed, the static approximation is definitely fine when $E_F < \hbar\omega_T$. In fact, E_F starts to exceed $\hbar\omega_T$ already at $n > 7 \times 10^{17}$ cm^{-3}. However, analysis of Eq. (4) shows that $V(\epsilon, 0) - V(0, 0) \approx W \frac{\epsilon^2}{3\omega_T^2}$ while $-V(0, 0) = W \ln \frac{\eta}{\kappa} \approx 3.8W$. Thus we expect that retardation effects are relatively minor up to n_{c1}, while at higher n the theory [16] should be augmented to take them into account; the same is needed for the accurate analysis of T_c enhancement due to isotope substitution leading to ω_T suppression.

Both effects mentioned in the previous paragraphs can lead to the dome-shaped $T_c(n)$ dependence with decrease in transition temperature at higher electron concentrations. At the low densities we consider here, the modifications due to these effects are expected to be minor; nevertheless, they might be responsible for the slight deviations between our theory and experiment seen in Fig. 1.

The absence of Coulomb interaction in doped STO makes it a rare representative of a superconductor where the universal effect of T_c suppression by disorder [26,27] is not operating. Moreover, it may demonstrate the opposite effect of T_c enhancement by strong disorder, predicted earlier in bulk [28] and two-dimensional [29] materials with suppressed Coulomb interaction. It might be possible to reach the necessary range of strong disorder, $k_F l \sim 1$, by heavy-dose electron irradiation of a STO crystal, along the lines of Ref. [30]. However, such irradiation may lead to an increase in the gap ω_T and thus a decrease in effective attraction, so the ω_T dependence on irradiation should be controlled.

Note added. Recently, we became aware that a similar approach to superconductivity in doped $SrTiO_3$ was developed in Ref. [31].

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