Electron-correlation effects on the properties of $La_xSr_{1-x}TiO_3$

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A theoretical study of the electron-correlation effects on the properties of $La_xSr_{1-x}TiO_3$ is presented. Using a single band repulsive Hubbard model employing the Gutzwiller approximation we study the effect of electron correlations on the effective-mass, band narrowing, plasma frequency, and spin susceptibility. The doping dependence of these properties are found to be qualitatively in agreement with recent experimental studies on $La_xSr_{1-x}TiO_3$. We have also proposed a possibility for inducing superconductivity using the idea of proximity effect. The induced superconducting gap in $La_xSr_{1-x}TiO_3$ is predicted to be maximum near the half-filling of the Ti $3d$ band.

I. INTRODUCTION

Studies of electron-correlation effects in narrow-band perovskite structured materials have received renewed attention in recent years primarily due to the discovery of Cu-O based high-T_c superconductors. One of the most important manifestations of electron correlation in a narrow band is the possibility of Mott-Hubbard (MH) metal-insulator transition[1,2] when the correlation strength (U) exceeds a critical value U_c (of the order of the bandwidth). The most extensively studied Mott insulator[3] is V_2O_3, which undergoes MH metal-insulator (MI) transition when doped with Ti or Cr. Recently, a new material, $Sr_{1-x}La_xTiO_3$, has been extensively studied by Tokura *et al.*,[4] with a view of understanding the doping dependence of the electronic properties of correlated electron systems. The ground state of $LaTiO_3$ with a half-filled titanium $3d$ band is a Mott insulator with a gap of 0.1 eV.[4,5] On substitution with Sr, electrons are removed from this band and the material undergoes an insulator-metal transition. Tokura and co-workers[4,6] have done extensive investigations of the doping dependence of the properties of this compound. The end product $SrTiO_3$ is a closed-shell insulator. Thus, this system undergoes a transition from an ordinary insulator ($SrTiO_3$) to a metal with negligible correlation to a correlated metal (for electron concentrations near half-filling) and finally to a Mott-insulator at half filling. They have found that as half-filling situation is approached, spin susceptibility, effective mass, and specific heat shows a strong enhancement. It has also been found that the enhancement of spin susceptibility is not due to Stoner enhancement, but rather is due to an increase in the effective mass resulting from electron correlation effects (a Wilson ratio of ~ 2 has been deduced from experiments).

The main aim of this paper is to interpret the experimental results[4,6] on $La_xSr_{1-x}TiO_3$ on the basis of a single-band Hubbard model taking into account the localization effects of electron correlations (U). Applied to this material, the Hubbard model represents the Ti $3d$ band. Properties of this model are dependent on the band width, on-site repulsion strength and the electron concentration. It has been found that a rigid band filling occurs[4] in the above material and the metallic state has a Fermi surface consistent with Luttinger's theorem. The main purpose of this work is to understand the dependence of the properties of $La_xSr_{1-x}TiO_3$ on the filling of the Ti $3d$ band and the effect of electron correlations. The importance of electron correlations is also borne out by the temperature dependence of the resistivity, which goes as $\rho_{(T)}=\rho_0+AT^2$. Using the Gutzwiller approximation[7-9] to treat U, we calculate spin susceptibility, plasma frequency, effective mass, and the band narrowing factor and study the doping dependence of these properties of $La_xSr_{1-x}TiO_3$. It is also important to note that the removal of electrons from the d-band in $LaTiO_3$ produces a metallic state, which has a large Fermi surface,[4] not a small-hole Fermi surface corresponding to the hole doping of the insulator. So, the metallic state in this compound is in the $U<U_c$ regime of the Brinkman-Rice transition[2] (where U_c is critical correlation strength required for metal-insulator transition for the half-filled band).

The plan of the paper is as follows. In Sec. II, we introduce the Hubbard model in the Gützwiller approximation, Sec. III deals with the calculation of χ_s, m^*, band-narrowing factor, plasma frequency, etc., Sec. IV we investigate the possibility of inducing superconductivity in this material by the proximity effect and finally the conclusions are given in Sec. V.

II. ELECTRON CORRELATION EFFECTS IN THE NARROW BAND METAL

To understand the effect of repulsive on-site electron correlations in the narrow-band metal, we will employ the following Hubbard Hamiltonian,

$$H=\sum_{ij\sigma} T_{ij}C_{i\sigma}^{\dagger}C_{j\sigma}+U\sum_i n_{i\uparrow}n_{i\downarrow}\ . \tag{1}$$

We will use Gutzwiller approximation to treat the strong electron correlation effect of U. In this approximation the Hamiltonian can be written as

$$H=\sum_{ij\sigma} q_\sigma T_{ij} C^\dagger_{i\sigma} C_{j\sigma} , \tag{2}$$

where q_σ is the band width renormalization factor, which depends on U, bandwidth, and band filling and is given by,

$$q_\sigma=\frac{\{\sqrt{n_\sigma-d}\,\sqrt{1-n_\sigma-n_{-\sigma}+d}+\sqrt{(n_{-\sigma}-d)d}\,\}^2}{n_\sigma(1-n_{-\sigma})} \tag{3}$$

Here, d is the double occupancy of the sites and $n=n_\sigma+n_{-\sigma}$. The total energy is given by

$$E_g=q_\uparrow\epsilon_\uparrow+q_\downarrow\epsilon_\downarrow+Ud \tag{4}$$

and

$$\epsilon_0=\sum_{|k|<k_F}\epsilon_k<0 . \tag{5}$$

The parameter d is determined by minimizing E_g with respect to d. Since the metallic phase of $La_xSr_{1-x}TiO_3$ is a paramagnetic Fermi liquid, we have $n_\sigma=n_{-\sigma}=n/2$. Then, the condition to determine d is given,

$$\frac{U}{U_c}=\frac{1}{2n(2-n)}\left\{\sqrt{n/2-d(1-n+d)}+\sqrt{d(n/2-d)}\right\}\times\left\{\left[\frac{n/2-d}{1-n+d}\right]^{1/2}-\left[\frac{1-n+d}{n/2-d}\right]^{1/2}+\left[\frac{n/2-2d}{\sqrt{d(n/2-d)}}\right]\right\}, \tag{6}$$

where $U_c=8|\epsilon_0|$. At half filling ($n=1$), the Brinkman-Rice transition occurs at $U=U_c$. In what follows, we will use the above results obtained in the Gutzwiller approximation to calculate the doping dependence of various physical properties of $La_xSr_{1-x}TiO_3$.

III. DOPING DEPENDENCE OF THE PROPERTIES OF $La_xSr_{1-x}TiO_3$

A. Effective mass

Starting from the closed-shell insulator $SrTiO_3$, substituting Sr sites with La, electrons are introduced into the Ti $3d$ band. As the electron concentration increases in this band, correlation effects should start becoming dominant leading to the increase of m^* of the charge carriers. Using Gutzwiller approximation, the value of m^* can be calculated as a function of carrier density. The effective mass is given by

$$m^*/m=1/q , \tag{7}$$

where q is the band width reduction parameter given by Eq. (3).

In writing Eq. (7), it is assumed that the dominant contribution to the renormalization of m^*/m comes from the energy dependence of electronic self-energy. The contribution from the momentum dependence of self-energy is small in comparison and is neglected. In general,

$$\frac{m}{m^*}\approx Z\left[1+\frac{\partial\Sigma(k,\epsilon)}{\partial\epsilon_k}\right] ,$$

where Z is the Fermi surface discontinuity in the single-particle occupation number. In analogy with electron-phonon[10] and paramagnon[11] problems, we have assumed that the momentum dependence of self-energy (at $k=k_F$) is small and consequently its contribution to the electron effective mass is not significant (in comparison with the effective-mass enhancement due to the correlation effects). The assumption that the energy dependence of the electronic self-energy makes the maximum contribution to the effective mass[12] is also consistent with the value of[4] $A/\gamma^2\approx 1.0\times10^{-5}$ $\mu\Omega$ cm $(\text{mol K/mJ})^2$ (where A is the coefficient of the T^2 term in resistivity and γ is the electronic specific-heat coefficient in the metallic phase). In ordinary metals (wide-band metals), the electron-electron interaction contribution to the effective mass is small.[10] But, in the case of narrow-band metals this contribution from electron correlation becomes significant and is dependent on correlation strength and band filling. Using Eqs. (3) and (6), we have calculated m^*/m as a function of deviation from half filling and the results are given in Fig. 1. It is clear that significant enhancement of m^*/m occurs near half filing and correlations effects are not dominant for approximately $\delta>0.5$. The enhancement of m^*/m is clearly found in the experiments of Tokura and co-workers.[4]

B. Bandwidth reduction

In the Gutzwiller approximation, the effective mass diverges at $n=1$ for $U=U_c$ and the particles are local-

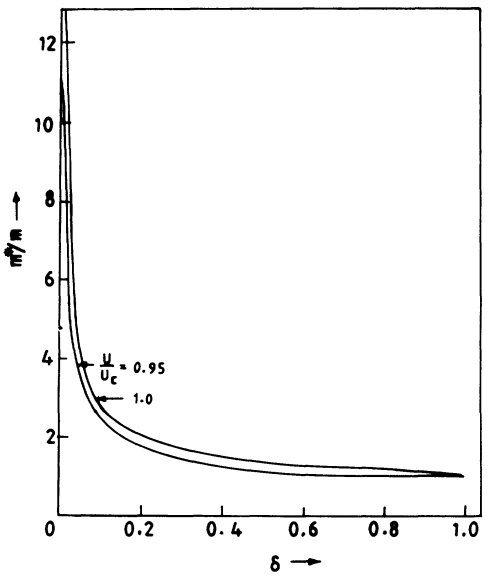

FIG. 1. Variation of effective-mass with deviation from half-filling for different values of U/U_c.

ized. The changes in the bandwidth are determined by the value of q. In Fig. 2 we have shown the bandwidth as a function of hole doping. This also means that the DOS at Fermi level is enhanced near half filling of the band leading to the enhancement of the electronic specific heat as seen in the experiments.[4]

C. Plasma frequency

The plasma frequency for a metal is given by

$$\omega_p^2 = \frac{4\pi N e^2}{\epsilon_\infty m^*} ,$$

where N is the electron density and m^* is the effective mass of the carriers. The effect of electron correlations in the narrow band is reflected in the increase of m^* near half filling of the band. In terms of the number of lattice sites per unit volume (N_s), we can write (assuming a dispersionless band of width D),

$$\omega_p^2 = \frac{4\pi N_s e^2}{\epsilon_\infty}\frac{n}{m^*} = \left[\frac{4\pi N_s e^2}{\epsilon_\infty m}\right] nq . \tag{8}$$

It should be noted that, what enters in the plasma frequency, for electrons in a narrow band with electron correlations, is the band-electron mass. The band-electron mass depends on the electron-electron interaction strength and band filling, since these parameters determine the effective band width in which the electrons are moving. The parameter which determines the band width is q, so that the plasma frequency is now determined by the correlation strength and band filling. For half filling as $U \to U_c$, the effective bandwidth becomes zero and the insulating state is obtained for $U = U_c$. The band narrowing is a consequence of electron correlations and band filling and is possible only in highly correlated narrow-band metals. The Gutzwiller approximation on which the above-mentioned arguments are based is consistent with Luttinger's theorem.

Since $m^*/m = 1/q$, where q is given by Eq. (3), in the correlated metal the plasma frequency is determined by the value of nq. For $U = U_c$, the bandwidth reduction factor goes to zero for the half-filled band and $\omega_p = 0$. To study the doping dependence of ω_p, we have calculated nq as a function of doping and the results are shown in Fig. 3. This shows that as the system is doped away from half filling, the plasma frequency first increases, goes through a maximum near $\delta = 0.3$ and decreases thereafter. This strong dependence of ω_P is consistent with experiments.[4,6] The experiments show that the value of n/m^* is anomalously suppressed as the $n = 1$ end is approached. Note that the suppression of ω_p near half filling cannot be understood by considering hole doping of the insulator ($LaTiO_3$), since close to half filling, a large Fermi surface (FS) has been found experimentally, not a small FS corresponding to hole doping of an insulator. Further, the experimentally found[4] energy dependence of effective mass can be understood from the relation between m^* and conductivity

$$\sigma(\omega) = \sigma_1(\omega) + i\sigma_2(\omega) ,$$
$$\frac{m^*}{m} = \frac{\omega_p^2}{4\pi}\frac{\sigma_2(\omega)}{\omega|\sigma(\omega)|^2} , \tag{9}$$

where ω_p^2 is given by Eq. (8).

D. Spin susceptibility

The spin susceptibility of the Hubbard model in the Gutzwiller approximation[7] is given by[9]

$$\frac{\chi_s}{\mu_B^2 N(0)} = \frac{2x^2 - x^4 - \delta^2}{x^4 - \delta^4} , \tag{10}$$

where

$$x = \sqrt{d+\delta} + \sqrt{d} . \tag{11}$$

Here $N(0)$ is the density of states (DOS) at the Fermi sur-

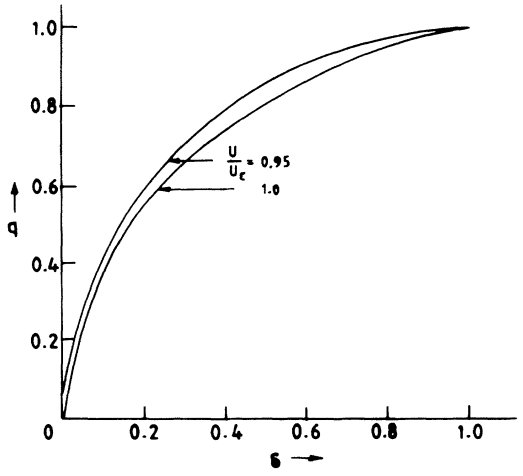

FIG. 2. Variation of bandwidth reduction factor with deviation from half filling for different values of U/U_c.

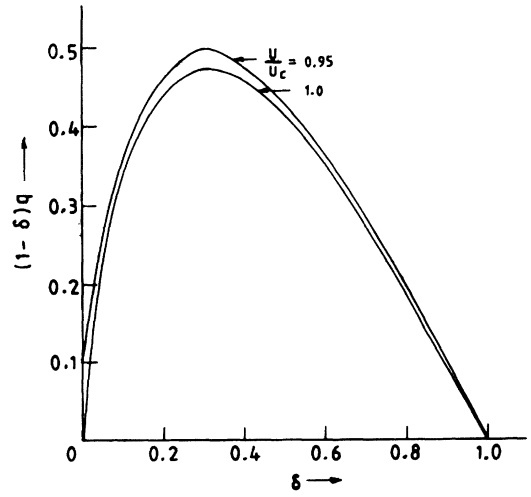

FIG. 3. Variation of the factor, $(1-\delta)q$, which determines plasma frequency with deviation from half filling for different values of U/U_c.

face for the uncorrelated system. The double-occupancy parameter (d) depends on correlation strength (U) and band filling. Using Eq. (6) we calculate d, and χ is obtained from Eq. (10), as a function of doping. The spin susceptibility shows a large enhancement as the system approaches the half filling as shown in Fig. 4. This enhancement is purely a correlation effect rather than due to Stoner enhancement factor. The Wilson ratio of 2 deduced from experiments also confirms this. These results are consistent with experimental results[4] on $La_xSr_{1-x}TiO_3$.

So far, we have discussed the properties of $La_xSr_{1-x}TiO_3$, which are strongly dependent on electron correlation effects. It is interesting to compare the correlated metallic phase in this compound (with enhanced effective mass) with heavy-fermion (HF) state. Both these systems have a T^2 term in resistivity and Wilson ratio of approximately 2. This comparison also shows that the mean free path of electrons in the titanate is not affected by electron correlations. The reason is that, in analogy with HF systems, we can write

$$\frac{m^*}{m}=\frac{\tau^*}{\tau}, \quad \frac{v_F^*}{v_F}=\frac{m}{m^*} \tag{12}$$

so that $l^*=l$. Here τ (τ^*) and v_F (v_F^*) are the relaxation time and Fermi velocity, respectively, for the uncorrelated (correlated) metal. Now, using analogy with heavy fermions[12,13] and using the fact that[4] $A/\gamma^2=1.0\times10^{-5}$ $\mu\Omega$ cm (mol K/mJ)^2, we can make a prediction: thermal conductivity in the correlated metallic phase of $La_xSr_{1-x}TiO_3$ will not show any effect due to correlation effects, since $\tau^*/m^*=\tau/m$. This experiment, then, serves as a test for the assumption that the dominant contribution to the effective mass comes from the energy dependence of electronic self-energy.

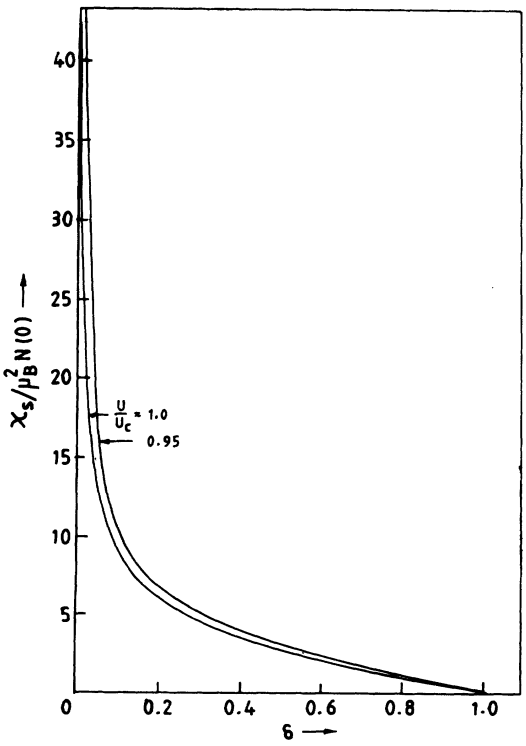

FIG. 4. Variation of spin susceptibility with deviation from half filling for different values of U/U_c.

IV. PROXIMITY-INDUCED SUPERCONDUCTIVITY IN $La_xSr_{1-x}TiO_3$

It is significant that hole doped $LaTiO_3$ is found to be not superconducting. The starting material has remarkable similarities with Cu-O based high-T_c materials. $LaTiO_3$ has cubic perovskite structure and is a Mott insulator, when well stoichiometric. The insulating state is antiferromagnetic with $T_N=120-150$ K. With slight hole doping ($\delta>0.05$), the antiferromagnetism is destroyed and the system becomes a correlated metal. The crucial difference is that the (Cu-O)-based high-T_c materials are quasi-two-dimensional with conducting Cu-O planes. The main question we are concerned with in this section is the following: is it possible to induce superconductivity in $La_xSr_{1-x}TiO_3$?

Let us consider a system consisting of a high-T_c perovskite (Cu-O)-based material with a thin film of $La_xSr_{1-x}TiO_3$ deposited on it. The proximity-induced superconducting gap in $La_xSr_{1-x}TiO_3$ is determined[14] by the condition

$$\frac{\Delta_s}{(NV)_s}=\frac{\Delta_N}{(NV)_N}, \qquad D_s\left[\frac{d\Delta}{dx}\right]_s=D_N\left[\frac{d\Delta}{dx}\right]_N. \tag{13}$$

Here $V_N(-ve)$ is the pairing field associated with the induced gap in the N layer and D is the diffusion constant. Now, everything else remaining constant, the induced superconducting gap in the N layer is controlled by the factor $(NV)_N$. It has been shown that in titanate material, there is a large enhancement of DOS at the Fermi level near the metal-insulator transition (i.e., for compositions near the half-filled $3d$ titanium band). So, the induced superconducting gap should also show a consequent enhancement due to the DOS increase in this material. So, if the $La_xSr_{1-x}TiO_3$ film has composition such that it is close to the metal-insulator transition region (i.e., in the strongly correlated metallic state), the proximity-induced gap should be strongly enhanced. Since the D_N is given by $(1/3)(m/m^*)]v_Fl_F$ [using Eq. (12)], the reduction of the bulk value of the gap Δ_s due to the interface should also be small (due to the enhanced m^*/m near half filling).

The idea of the correlation enhancement of induced superconductivity, described above, relies on the enhancement of electronic effective mass due to correlation effects. For this the system has to be close to the half filling. The experimental results of Tokura and coworkers on $La_xSr_{1-x}TiO_3$ show that there is considerable enhancement[4] of m^*/m in the correlated metallic phase ($0.5<x<0.95$). The value of m^*/m at $x=0.9$ is approximately 6, and larger values are obtained in the

range $0.9 < x < 0.95$. This material is found to be metallic for $x < 0.95$. The experimental results do not show any non-Fermi-liquid features and are consistent with what could be expected from a Fermi liquid with electron correlations in a narrow band. So, in what follows we employ the Gutzwiller approximation to model the effects of electron correlation.

The proximity-induced gap should also be strongly dependent on the band filling in the N-layer material. To understand the doping dependence of the gap, we will use a Bardeen-Cooper-Schrieffer (BCS) Hamiltonian taking into account the band-narrowing effects of the electron correlations. In the Gutzwiller approximation, this Hamiltonian can be written[15] as

$$H=\sum_{k\sigma} q\epsilon_k C^{\dagger}_{k\sigma} C_{k\sigma} + V\sum_k (\Delta_k C^{\dagger}_{k\uparrow} C^{\dagger}_{-k\downarrow} + \text{H.c.}) . \quad (14)$$

So, in the Gutzwiller approximation, the only change in the standard BCS Hamiltonian is in the electron correlation and filling dependent bandwidth of the electrons. This leads to the modification of the electronic density of states, which now is dependent on correlation strength and band filling. The standard BCS gap equations become

$$\frac{1}{|V|}=\frac{1}{2N}\sum_k \frac{1}{\sqrt{(q\varepsilon_k-\mu)^2+\Delta_0^2}} \quad (15)$$

and

$$\delta=\frac{1}{N}\sum_k \frac{q\varepsilon_k-\mu}{\sqrt{(q\varepsilon_k-\mu)^2+\Delta_0^2}} . \quad (16)$$

Here ε_k is the band structure of the uncorrelated d band. We can solve the gap equations using a constant DOS, $1/2D$, where D is half the bandwidth. The width of the band is dependent on the strength of the electron correlations and band filling. The gap equations then are

$$\frac{1}{|V|}=\frac{1}{2D}\int_{-D}^{D} \frac{d\varepsilon}{\sqrt{(\varepsilon-\mu)^2+\Delta_0^2}} \quad (17)$$

and

$$\delta=\frac{1}{2D}\int_{-D}^{D} \frac{(\varepsilon-\mu)d\varepsilon}{\sqrt{(\varepsilon-\mu)^2+\Delta_0^2}} . \quad (18)$$

Here $D=qD_0$, where D_0 is half the width of the original uncorrelated band and q is the bandwidth reduction factor given in Eq. (3). Is it reasonable to assume a constant pairing field for the induced superconductivity in the normal metal? The idea is that on any plane in the normal-metal parallel to the interface the solution of Bogoliubov-deGennes equations[14] will give a definite value of the proximity-induced superconducting gap. It is reasonable to associate a constant pairing field associated with this gap. So the gap equations with constant V refer to one of the planes parallel and close to the interface. In Fig. 5, we have shown the solutions of Eqs. (17) and (18) as a function of the deviation from half filling.

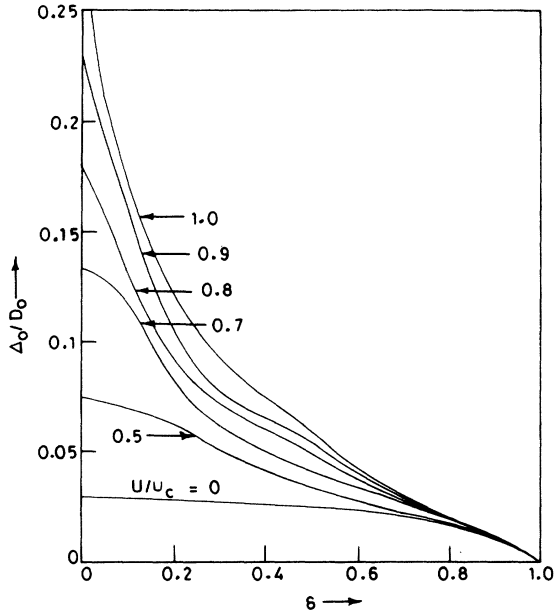

FIG. 5. Variation of induced superconducting gap with deviation from half filling for various values of U/U_c, and $|V|/D_0=0.5$.

The solutions show that if the normal-side metal is such that the d band is close to half filling, then the superconducting gap shows large enhancement (due to localization effects of electron correlations) over the gap that would be induced in an uncorrelated metal. For $U=U_c$ and $n=1$, the mean-field theory presented here is not applicable, and we expect the induced gap to go to zero due to complete localization of carriers.

The most favorable configuration for observing these effects would be a thin film of $La_xSr_{1-x}TiO_3$ deposited on a high-T_c Cu-O material. The reason is that large superconducting gaps are observed in (Cu-O)-based high-T_c materials so that the induced gap will have an observable magnitude. If deposited parallel to the Cu-O planes, the interface quality will be good, since both have perovskite structures. If observed, this N-S system would provide the first example of induced superconductivity (which is strongly doping dependent) in a correlated nonsuperconducting material.

V. CONCLUSIONS

In this paper, we have calculated effective mass, plasma frequency, spin susceptibility, and band narrowing factor for a correlated narrow-band model of $La_xSr_{1-x}TiO_3$. We have studied the doping dependence of the above properties. Near half filling of the d band in this material, we have shown the effective mass, spin susceptibility, and specific heat are strongly enhanced due to electron correlation effects. We have also studied the doping dependence of the plasma frequency. The results are qualitatively in agreement with the recent experiments. Further, we have also proposed a possibility for inducing superconductivity in this material through a proximity effect. The calculations show that the induced superconducting gap shows a large enhancement for

compositions such that the d band in $La_xSr_{1-x}TiO_3$ is near half filling.

It should also be noted that the calculated doping dependence of m^*, χ and specific heat shows a faster reduction as the system is doped away from the half-filling situation. This might arise from the deficiencies of the Gutzwiller approximation employed here to treat the electron correlation effects. One of the drawbacks of this approximation is that it leads to infinite gap for the insulator (for $U=U_c$ and $n=1$). The experimental value for insulating gap for $LaTiO_3$ is approximately 0.1 eV. A slave boson approach[16] including the fluctuations[17] about the mean-field slave-boson ground state might improve the calculations, so that a better agreement with experimental results is obtained.

Strongly correlated electron systems in narrow energy bands have a tendency to form an antiferromagnetically ordered state in the insulator. It is not clear to us how antiferromagnetic spin fluctuations very close to the metal-insulator transition would modify our results. The Gutzwiller method, as used here, is not adequate to describe these effects. $LaTiO_3$ is a Mott insulator with antiferromagnetic ordering (with a $T_N=150$ K). However, on removing electrons from the half-filled Ti $3d$ band of this material (by Sr substitution), the antiferromagnetism is destroyed and the metallic state ($x<0.95$) in $La_xSr_{1-x}TiO_3$ does not show any significant effects[11] due to spin fluctuations. It is to be noted that the energy gap in the insulator is only 0.1 eV and hole doping produces a metallic state with a large Fermi surface. Hall measurements show that the carriers in the metallic state are electrons. Furthermore, the temperature variation of resistivity in the metallic state is consistent with a Fermi-liquid picture. Also, a finite Wilson ratio (~ 2) implies that the metal-insulator transition in this material is realistically modeled as a localization-due-to-correlation transition rather than a magnetic phase transition (in which case the Wilson ratio goes to infinity). Nevertheless, detailed neutron-scattering experiments probing the spin excitations and a detailed study of the electronic properties, especially close to the metal-insulator transition, would be required to answer the effects of spin fluctuations in the metallic state close to the metal-insulator transition in this material.

As we have already mentioned, antiferromagnetic spin correlations for the strong-coupling half-filled limit of the Hubbard model does not come out in the mean-field type Gutzwiller approximation employed to study the effect of electron correlations in this paper. The development of spin correlations near the phase boundary between the metallic and insulating phases are not properly captured in this approximation. But, we have noted that the experimental results available on $La_xSr_{1-x}TiO_3$ does not show any prominent effects of the presence of spin correlations in the metallic phase. This implies that the antiferromagnetic spin correlations present in the insulator ($LaTiO_3$) is significantly rapidly suppressed when electrons are removed from the Ti $3d$ band (by Sr substitution). We believe that this is due to the small value of the insulating gap of the insulator (0.1 eV). So, for the metallic phase of this material, we can reasonably expect that retardation effects arising from such spin correlations are not significant. In this case, then, a BCS approach used in Sec. IV, in studying superconductivity, is reasonable.

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