Problem 1. The material $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ has a layered crystal structure that consists of two-dimensional square lattices of CuO_2 planes (shown in Fig. 1) separated by layers of $\text{La}_{2-x}\text{Sr}_x\text{O}_2$. You may assume that La has valence 3+, Sr valence 2+, and O valence 2-; the electrons from these cations are donated uniformly to the widely separated CuO_2 layers, which thus have a two-dimensional electronic structure. Neutral atomic Cu has the configuration [Ar] $4s^23d^9$. In this compound, four of the Cu d levels are completely filled, and there is a partially filled band formed from $d_{x^2-y^2}$ orbitals. You may assume the Cu 4s levels are unoccupied, and the O 2p levels are fully occupied. Electronic dispersion perpendicular to the planes may be neglected.

The band structure in the independent particle approximation is well described by a tight-binding model incorporating a single orbital (per unit cell) of $d_{x^2-y^2}$ symmetry centered on the Cu atom, with nonzero Hamiltonian matrix elements t between nearest neighbor orbitals in the x and y directions, and matrix elements t' between second neighbors across the diagonals.

1(a) Show that in this approximation the energy dispersion of an electron is

$$E(k) = 2t[\cos(k_x a) + \cos(k_y a)] + 4t'\cos(k_x a)\cos(k_y a).$$

Solution. The band energy in the tight-binding description is given by (4.57) in the lecture notes,

$$E(k) = \epsilon_0 + t \sum_{\rho} e^{-i\mathbf{k}\cdot\boldsymbol{\rho}}.$$

In the two-dimensional CuO₂ plane, the four nearest neighbors to the origin are located at

$$\boldsymbol{\rho} \in a\{(1,0,0), (0,1,0), (-1,0,0), (0,-1,0)\},\tag{1}$$

and the four second-nearest neighbors are located at

$$\rho' \in a\{(1,1,0), (-1,1,0), (1,-1,0), (-1,-1,0)\}.$$
 (2)

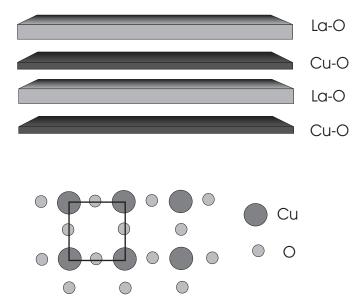


Figure 1: Schematic drawing of the layered crystal structure of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, and a drawing of the a single two-dimensional CuO_2 plane. The square is a primitive unit cell, with primitive lattice vectors a(1,0,0) and a(0,1,0).

So, assuming $\epsilon_0 = 0$, the band energy is

$$E(k) = t \sum_{\rho} e^{-i\mathbf{k}\cdot\boldsymbol{\rho}} + t' \sum_{\rho'} e^{-i\mathbf{k}\cdot\boldsymbol{\rho}'}$$

$$= t \left(e^{-iak_x} + e^{-iak_y} + e^{iak_x} + e^{iak_y} \right) + t' \left(e^{-ia(k_x + k_y)} + e^{ia(k_x - k_y)} + e^{-ia(k_x - k_y)} + e^{ia(k_x + k_y)} \right)$$

$$= t \left(e^{-iak_x} + e^{iak_x} + e^{-iak_y} + e^{iak_y} \right) + t' \left(e^{-iak_x} e^{-iak_y} + e^{iak_x} e^{-iak_y} + e^{-iak_x} e^{iak_y} + e^{iak_x} e^{iak_y} \right)$$

$$= t \left[\left(e^{-iak_x} + e^{iak_x} \right) + \left(e^{-iak_y} + e^{iak_y} \right) \right] + t' \left(e^{-iak_x} + e^{iak_x} \right) \left(e^{-iak_y} + e^{iak_y} \right)$$

$$= t \left[2\cos(k_x a) + 2\cos(k_y a) \right] + t' \left[2\cos(k_x a) \right] \left[2\cos(k_y a) \right]$$

$$= 2t \left[\cos(k_x a) + \cos(k_y a) \right] + 4t' \cos(k_x a) \cos(k_y a)$$
(3)

as we wanted to show.

1(b) What do you expect to be the signs of t and t'? Explain your reasoning.

Solution. The signs of t and t' are expected to be negative because the Coulomb potential ΔU between the two electrons at any two sites is negative [1, pp. 78–79]. We see that t depends on ΔU by Ashcroft & Mermin (10.18), and we see that t has the same sign as ΔU by (4.56) in the lecture notes:

$$t = \int d\mathbf{r} \, \psi^*(\mathbf{r} - \rho) \Delta U \psi(\mathbf{r}).$$

1(c) For the case that |t'/t| = 0, sketch the Fermi surface for Sr concentrations of x = 0, $x \approx 0.2$, and $x \approx 0.5$.

Solution. As x increases, the partially-filled band of Cu becomes more empty. When x=0, the band is mostly full. When x=0.5, it is half full. Fig. 2 (left) shows the Fermi surface in the reduced zone scheme for x=0 (blue line), $x\approx 0.2$ (gold line), and $x\approx 0.5$ (green line) when t'=0 [2, p. 231]. The figures were created in Mathematica by plotting Eq. (3) and choosing appropriate contours.

1(d) How do these contours change qualitatively if $|t'/t| \sim 0.1$? (Choose the signs of t and t' that you proposed in 1(b).)

Solution. For t and t' both negative, the contributions from t' will distort the contours by pulling their ends toward the corners of the frame. This happens because, in general, the Fermi surface is stretched toward the positions of the nearest neighbors (for t < 0) given by Eq. (1). There is a nearest neighbor at the center of each edge of the frame. When we also consider t' < 0 contributions from the next-nearest neighbors, which are located in the corners of the frame by Eq. (2), the Fermi surface is pulled toward those ions as well. The effect is not too large since |t'| is small compared to |t|. Fig. 2 (right) shows the Fermi surface in the reduced zone scheme for x = 0 (blue line), $x \approx 0.2$ (gold line), and $x \approx 0.5$ (green line) when t' = 0.1t.

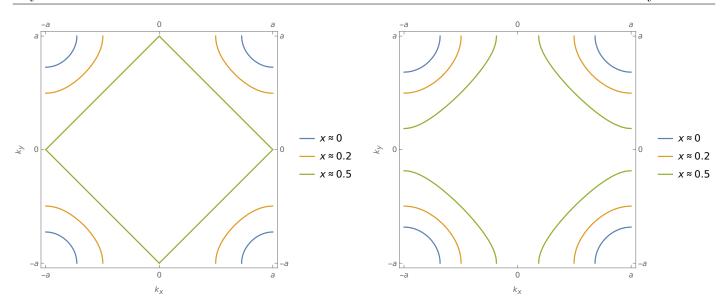


Figure 2: Fermi surfaces for t' = 0 (left) and |t'/t| = 0.1 (right) for x = 0 (blue line), $x \approx 0.2$ (gold line), and $x \approx 0.5$ (green line).



Figure 3: Density of states g(E) for the dispersion shown in Fig. 2 (right).

1(e) Assuming the dispersion of 1(d), sketch the electronic density of states in energy, paying particular attention to the behavior near the edges of the band and at a saddle point in the middle of the band.

Solution. Our sketch of g(E) is based on Fig. 4.2 in the course lecture notes, and is shown in Fig. 3. The logarithmic singularity in the middle represents the saddle point. Near the edges, the curve flattens out.

1(f) What would the independent electron model predict for the temperature dependence of the low-temperature electronic specific heat when the chemical potential is exactly at the saddle point near the middle of the band?

Solution. By (2.15) in the lecture notes, the electronic specific heat is given by

$$c_v = \int dE \, Eg(E) \frac{df(E)}{dT},$$

where f(E) is the Fermi distribution. At low temperature, the chemical potential is approximately equal to the specific heat. The saddle points are located at the positions of the nearest neighbors, given by Eq. (1). Since the lattice is periodic, we can choose $\rho = (a, 0, 0)$ without loss of generality.

1(g) La₂CuO₄ is an antiferromagnetic insulator. Suggest, and discuss, reason(s) why the ground state differs from that predicted by the band structure in the independent particle approximation. Your answer should include a qualitative explanation of both the magnetic and the insulating behavior.

Problem 2.

2(a) Write short notes on the properties of a Fermi liquid.

Solution.

- A Fermi liquid is similar to a Fermi gas, except a Fermi liquid accounts for electron-electron interactions
- The electron-electron interactions cause the electron "gas" to distort around any given electron near the Fermi surface, and the electron together with this screening by the gas is a quasiparticle: a single-particle excitation of the system of interacting electrons
- The electron carries this electron gas distortion with it as it moves, which enhances (renormalizes) the mass and magnetic moment of the quasiparticle compared to the electron
- The Fermi liquid may be thought of as a system of non-interacting quasiparticles, analogous to how the Fermi gas is a system of non-interacting electrons; in fact, there is a one-to-one correspondence between the quasiparticle states of the Fermi liquid and the fermion states in the Fermi gas
- The dispersion relation $\epsilon_{\mathbf{k}}$ of the Fermi liquid differs from that of the Fermi gas due to the effects of electron-electron interactions; this is the most significant effect of the electron-electron interactions
- Quasiparticles deacy since they are excitations, but the closer a quasiparticle is to the Fermi surface, the longer its lifetime, and the more its behavior resembles that of an actual particle
- The Fermi liquid must have a stable ground state, so it cannot exist if the electron gas system undergoes a phase transition as electron-electron interactions are adiabatically turned on
- The Fermi liquid is a good model for typical metals at low temperature, as well as more exotic systems such as liquid ³He and neutron star cores

[lecture notes, pp. 84–87] [2, p. 417] [1, p. 127–131] [3, pp.345–351]

2(b) Write short notes on effects due to the electron-phonon interaction in metals.

Solution.

- Phonons cause local distortions of the crystal lattice, which move the ions from their equilibrium positions, thereby creating an electric potential that is screened by nearby conduction electrons
- The potential scatters electrons from state \mathbf{k} to state \mathbf{k}' , which alters the density distribution of the electron gas
- The disturbance in the electron density may in turn create a new phonon or lattice distortion, and the degree of distortion is determined by the phonon susceptibility of the crystal
- The lattice distortion created by an electron density fluctuation lasts longer than the fluctuation itself, and creates more local electron density fluctuations over its lifetime
- This retarded interaction creates an effective "attraction" between conduction electrons in the metal, which can lead to superconductivity and the creation of Cooper pairs
- In addition, the interaction between phonons and electrons causes electrons to effectively carry polarized lattice distortions with them as they move, which decreases their effective velocity and increases their effective mass

[lecture notes, pp. 129–134] [2, pp. 671–672] [3, p. 512]

2(c) Write short notes on the use of density functional theory to perform practical calculations of ground state properties of solids.

Solution.

- The key idea of density functional theory is that the total ground state energy of an interacting electron system, with electron-ion interactions, can be represented as a functional of the number density of electrons, $n(\mathbf{r})$
- Finding the minimum of the functional with respect to $n(\mathbf{r})$ can tell us the number density function of the ground state, as well as its energy
- In general the analytic functional is not known, but we can make very good approximations
- The first Hohenberg-Kohn theorem states that there is a one-to-one correspondence between the groundstate density of an N-electron system and the external potential v_{ext} acting on it; this means we do not need to know the wavefunction of the ground state (which is untenable for a large system) in order to determine its properties, since we can use $n(\mathbf{r})$ instead
- The reason for this is that the ground-state wavefunction is a functional of v_{ext} , which in turn is a functional of $n(\mathbf{r})$; thus, the ground-state wavefunction is a functional of $n(\mathbf{r})$
- Likewise, the total energy, kinetic energy, and interaction energy are also functionals of $n(\mathbf{r})$ ' furthermore, they are the same functionals for any system of N electrons (that is, independent of v_{ext})
- The method of Lagrange multipliers can be used to obtain the Kohn-Sham equations (4.133),

$$\left(-\frac{\hbar^2 \nabla^2}{2m} + U_{\text{ion}}(\mathbf{r}) + U_{\text{coul}}(\mathbf{r}) + U_{\text{xc}}(\mathbf{r})\right) + \phi_i(\mathbf{r}) = \epsilon_i \phi_i(\mathbf{r})$$

which are similar in form to Schrödinger equations, and the Lagrange multipliers ϵ_i resemble one-particle energies

- The Lagrange multipliers may approximate single-particle energies when the exchange-correlation energy is small, as is the case in in many metals and semiconductors
- The exchange-correlaration potential $U_{xc}(\mathbf{r})$ may be closely approximated by that of a uniform electron gas in the local density approximation
- The Thomas-Fermi approximation is also a simple approximation of $U_{xc}(\mathbf{r})$, and so is a density functional theory

[lecture notes, pp. 59–66] [4]

2(d) Write short notes on photoemission spectroscopy.

Solution.

- Photoemission spectroscopy is used to directly measure the electron spectral function (the probability of finding an electronic state with a given energy and momentum) and the electronic density of states
- This method uses the photoelectric effect: a beam of photons is incident on the surface of a solid, which are absorbed by electrons in the crystal, exciting them and causing them to leave the crystal

- In angle-resolved photoemission spectroscopy (ARPES), both the energy and momenta of the emitted electrons are measured in a detector
- Since the incident photons are perpendicular to the surface of the crystal, the momentum of an electron parallel to the surface, p_{\parallel} , is conserved and can scanned directly by rotating the detector (assuming the crystal surface is very smooth)
- Since only p_{\parallel} (as opposed to p_{\perp}) can be probed, the cleanest measurements come from materials whose band structure has little dispersion in the direction perpendicular to the surface
- By integrating over all angles, the electronic density of states as a function of energy can be measured
- ARPES can also be used to map the Fermi surface of a crystals

[lecture notes, pp. 84, 87–89] [5] [6]

Problem 3. A long-wavelength optical phonon in an insulating ionic solid is described by the following equation of motion:

$$M\ddot{\mathbf{u}} + K\mathbf{u} = Q\mathbf{E},\tag{4}$$

where \mathbf{u} is the atomic displacement, M the mass, K a local restoring force, and \mathbf{E} is the electric field. A displacement of the ions by \mathbf{u} results in a polarization

$$\mathbf{P} = nQ\mathbf{u},\tag{5}$$

where n is the density of the ions, and Q their effective charge. We will consider only longitudinal modes, where \mathbf{D} , \mathbf{E} , \mathbf{u} are all parallel.

3(a) Neglecting any further electronic response of the solid, calculate the response of the phonon mode to a uniform external electric displacement field $\mathbf{D} = \epsilon_0 \mathbf{E} + \mathbf{P}$, oscillating with a frequency ω . Hence show that the phonon contribution to the frequency-dependent dielectric function, defined by $D = \epsilon_{\text{ion}} \epsilon_0 E$ may be written

$$\epsilon_{\rm ion}(\omega) = 1 - \frac{\Omega_P^2}{\omega^2 - \Omega_0^2},\tag{6}$$

giving formulae for Ω_P and Ω_0 in terms of the constants M, K, n, and Q.

Solution. Substituting $\mathbf{E} = (\mathbf{D} - \mathbf{P})/\epsilon_0$ and Eq. (5) into Eq. (4), we have [lecture notes, p. 75]

$$M\ddot{\mathbf{u}} + K\mathbf{u} = \frac{Q}{\epsilon_0}\mathbf{D} - \frac{nQ}{\epsilon_0}\mathbf{u} \implies \epsilon_0 M\ddot{\mathbf{u}} + (\epsilon_0 K + nQ)\mathbf{u} = Q\mathbf{D}.$$
 (7)

The susceptibility or response function $\chi(\omega)$ is given by [7, p. 185]

$$\mathbf{P} = \epsilon_0 \chi \mathbf{E}. \tag{8}$$

We recall that differentiating in the time domain is equivalent to multiplying by $i\omega$ in the frequency domain [8]:

$$\mathcal{F}_x[f^{(n)}(x)](\omega) = (i\omega)^n \mathcal{F}[f(x)](\omega).$$

Then we can easily Fourier transform both sides of Eq. (4). Let $x \equiv \epsilon_0 K + nQ$. Then

$$QD(\omega) = \epsilon_0 M(i\omega)^2 u(\omega) + xu(\omega) = \left(x - \omega^2 \epsilon_0 M\right) u(\omega) \implies u(\omega) = \frac{QD(\omega)}{x - \epsilon_0 M \omega^2}.$$

Feeding this into Eq. (5), we have

$$\mathbf{P} = \frac{nQ^2D(\omega)}{x - \epsilon_0 M\omega^2}$$

By Eq. (8) and $D = \epsilon_{\text{ion}} \epsilon_0 E$, then, the response function is

$$\chi(\omega) = \frac{nQ^2}{\epsilon_0 K + nQ - \epsilon_0 M\omega^2}.$$

The dielectric function is defined by $\epsilon \equiv 1 + \chi$ [7, p. 186]. So the phonon contribution to the dielectric function is

$$\epsilon_{\rm ion}(\omega) = 1 + \frac{nQ^2}{\epsilon_0 K + nQ - \epsilon_0 M \omega^2} = 1 - \frac{nQ^2}{\epsilon_0 M \omega^2 - (\epsilon_0 K + nQ)} = 1 - \frac{nQ^2/\epsilon_0 M}{\omega^2 - (\epsilon_0 K + nQ)/\epsilon_0 M},$$

or

$$\epsilon_{\rm ion}(\omega) = 1 - \frac{\Omega_P^2}{\omega^2 - \Omega_0^2}$$

where

$$\Omega_P = \sqrt{\frac{nQ^2}{\epsilon_0 M}},$$
 $\Omega_0 = \sqrt{\frac{\epsilon_0 K + nQ}{\epsilon_0 M}}$

as we wanted to show.

3(b) Figure 4 shows measurements of the frequency squared of an optical phonon in an insulating oxide (solid line) and measurements of the reciprocal of the static dielectric constant $1/\epsilon = 1/\epsilon_{\rm ion}(\omega = 0)$ (dotted-dashed line) in the same material. Using the model above, estimate the ion plasma frequency Ω_P and discuss whether the magnitude of your result seems appropriate for a typical ionic solid.

Solution. From Eq. (6),

$$\epsilon_{\rm ion}(\omega=0) = 1 + \frac{\Omega_P^2}{\Omega_0^2} \implies \Omega_P = \sqrt{\Omega_0^2(\epsilon_{\rm ion}(\omega=0) - 1)}.$$

Inspecting Fig. 4, it appears that $\Omega_0^2 \approx 4 \times 10^{24} \, \mathrm{s}^{-2}$ when $1/\epsilon \approx 0.002$ (or $\epsilon \approx 500$) slightly below 200 K. Plugging in these values,

$$\Omega_P \approx \sqrt{(4 \times 10^{24} \, \mathrm{s}^{-2})(500 - 1)} \approx 500 \sqrt{4 \times 10^{24} \, \mathrm{s}^{-2}} \approx 1 \times 10^{15} \, \mathrm{Hz}.$$

This is above the visible frequency range. Assuming it can be said in general that a material reflects light of all frequencies below the plasma frequency (as we discovered for the Drude model in question 5.3 of the homework), then our material would be highly reflective at optical frequencies. This is expected for a metal, but not for an insulator like the ionic solid in the problem. So the magnitude of this result does not seem quite appropriate, though it is not too far off. We would expect it to be at least one order of magnitude smaller in order not to reflect visible light.

3(c) Explain how the extrapolated vanishing of the optical phonon frequency at low temperatures, marked by the arrow in the figure, is indicative of a phase transition.

Solution. As Ω_0 decreases with temperature, the crystal lattice is changing as well, since the phonon frequencies depend on how the nearby ions are arranged. Say the lattice has a particular structure (here, cubic)

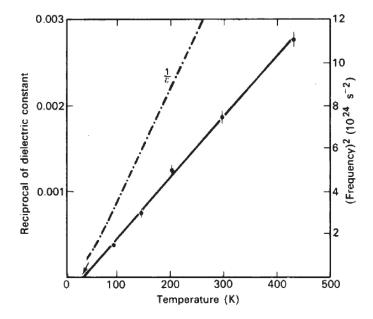


Figure 4: The solid line shows the square of the optical phonon frequency Ω_0^2 (right axis) and the dot-dashed line the inverse of the measured low frequency dielectric function $1/\epsilon_{\rm ion}(\omega=0)$ (left axis).

with a particular phonon dispersion at $T \approx 400 \,\mathrm{K}$, and a different structure (not cubic) below T_c , the temperature at which the optical phonon frequency vanishes. The low-temperature structure may have a completely different phonon dispersion than the high-temperature structure. As the temperature decreases and the crystal morphs from the high-temperature to the low-temperature structure, the occupation of the high-temperature phonon states will decrease in favor of the low-temperature phonon states. When the temperature reaches T_c and the crystal has fully transformed into the low-temperature structure, the optical phonon frequency of the high-temperature structure completely disappears. This is a structural phase transition [2, p. 467].

3(d) Construct a phenomenological Landau theory for the free energy of the solid as a power series expansion in the polarization, consistent with cubic symmetry.

Solution. Since we are constructing a Landau theory with polarization as the order parameter, it is likely our ionic solid is ferroelectric, which is consistent with the structural phase transition [2, pp. 469–470]. Since $\Omega_0 = 0$ at T_c , $\chi(\omega = 0) = \Omega_P^2/\Omega_0^2$ diverges at the critical temperature. This is characteristic of a second-order phase transition [lecture notes, p. 79].

We can write the free energy in the form

$$\mathcal{F} = \frac{a}{2}P^2 + \frac{b}{4}P^4 + \frac{c}{6}P^6 + \cdots,$$

where P, representing the polarization, can be written as a scalar instead of a three-dimensional vector since it may only point in a symmetry direction of the lattice [lecture notes, p. 103]. The coefficient $a = a_0(T - T_c)$ where $a_0 > 0$ is a constant [9]. For a second-order transition, b > 0, and since the $\mathcal{O}(P^4)$ term contributes much more than the $\mathcal{O}(P^6)$ term, so we may neglect it [2, p. 475]. Our theory is then

$$\mathcal{F} = \frac{a}{2}P^2 + \frac{b}{4}P^4 + \cdots,$$

where

$$a = a_0(T - T_c),$$
 $a_0 > 0,$ $b > 0.$

3(e) Use your model to predict how the phonon frequency Ω_0 and static dielectric constant ϵ_{ion} would vary with temperature below the critical temperature.

Solution. The equilibrium P(T) occurs at the minima of \mathcal{F} , where $d\mathcal{F}/dP = 0$ [2, p. 475]:

$$\frac{d\mathcal{F}}{dP} = aP + bP^3 = 0.$$

This implies

$$P = 0, P = \pm \sqrt{-\frac{a}{b}}.$$

Note, however, that P = 0 is a local maximum of \mathcal{F} if $T < T_c$:

$$\frac{d^2 \mathcal{F}}{dP^2}\Big|_{P=0} = \left[a + 2bP^2\right]_{P=0} = a_0(T - T_c) < 0 \text{ when } T < T_c.$$

However, the nonzero solution is imaginary for $T > T_c$. Thus the equilibrium P(T) is given by

$$P(T) = \pm \sqrt{\frac{a_0}{b}(T_c - T)} \qquad \text{when} \quad T < T_c.$$
 (9)

By including in \mathcal{F} the energy of the polarization coupled to an external electric field E, we can determine the dielectric susceptibility $\chi = dP/dE$ below the critical temperature [lecture notes, p. 103]. With the addition of the coupling term [2, p. 475]:

$$\mathcal{F} = \frac{a}{2}P^2 + \frac{b}{4}P^4 - EP.$$

Then

$$\frac{d\mathcal{F}}{dP} = aP + bP^3 - E = 0.$$

Differentiating both sides by E, we find

$$0 = a\frac{dP}{dE} + b\frac{dP^{3}}{dE} - 1 = a\frac{dP}{dE} + b\frac{dP^{3}}{dP}\frac{dP}{dE} - 1 = a\chi + 3bP^{2}\chi - 1,$$

which implies

$$\chi(T) = \frac{1}{a+3bP^2} = \frac{1}{a_0(T-T_c) + 3a_0(T_c-T)} = \frac{1}{2a_0(T_c-T)}$$
 when $T < T_c$, (10)

where we have used P(T) as defined in Eq. (9) and ignored the imaginary P(T). Although this P(T) is evaluated at E = 0, we assume the difference is negligible from P(T) evaluated at small E [lecture notes, p. 79].

We know from 3(a) that $\epsilon \equiv 1 + \chi$ and $\chi(\omega = 0) = \Omega_P^2/\Omega_0^2$. Then applying Eq. (10) tells us

$$\Omega_0(T) = \frac{\Omega_P}{\sqrt{\chi(T)}} = \Omega_P \sqrt{2a_0(T_c - T)}$$
 when $T < T_c$

and

$$\epsilon_{\text{ion}}(T) = 1 + \chi(T) = 1 + \frac{1}{2a_0(T_c - T)}$$
 when $T < T_c$.

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