

## Superconducting transition temperature of a paramagnetic material close to magnetic ordering

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**Abstract.** An explicit expression for the superconducting transition temperature  $T_c$  in a paramagnetic material is derived, when the transition occurs just before a possible magnetic ordering. As first noted by Uspenskii, such a transition may arise from electronic mechanism itself, without the necessary role played by the usual phonon-exchange mechanism. The result is discussed in terms of some recent experimental observations on the binary alloy  $Y_9Co_7$ .

**Keywords.** Superconductivity; magnetic ordering; superconducting transition temperature; binary alloy; electronic mechanism.

### 1. Introduction

In recent years, the study of the interplay between magnetism and superconductivity in the same material has been of considerable interest. In most materials, one finds that the electrons responsible for the occurrence of these two phenomena, coexisting or otherwise, are different, and the attractive phonon-exchange mechanism is still responsible for the superconductivity. We are not concerned with such a problem here. In this paper, we address ourselves to the question of calculating the superconducting transition temperature  $T_c$  for the conduction electrons in a metal, when the paramagnetic system is close to a magnetic ordering, without necessarily considering the phonon exchange mechanism. For a paramagnetic system, before the onset of magnetic ordering, the magnetic susceptibility can be considerably enhanced raising the possibility of even changing the sign of the effective Coulomb interaction between the electrons (Uspenskii 1979).

The effect of the possible onset of magnetic ordering on  $T_c$  is usually tackled (Orlando and Beasley 1981; Grest *et al* 1983; Berk and Schrieffer 1966) by calculating perturbatively the effect of spin-fluctuations on the superconducting coupling constant  $g$  arising from the usual attractive phonon-exchange mechanism in the presence of the repulsive effective Coulomb interaction. Our main concern here is not the phonon-exchange mechanism. We propose to use the general approach of the density-functional method to obtain a formal relation between  $\chi_m$  and the effective electron-electron interaction, including exchange as well as correlation effects, and explicitly find an expression for  $T_c$  as a function of average  $\langle \chi_m(\mathbf{q}) \rangle$  and other electronic parameters of the conduction electrons, close to magnetic ordering.

One indeed finds that  $T_c$  increases in the region where  $\chi_m^{-1} \rightarrow 0$ , with increasing  $\langle \chi_m(\text{peak}) \rangle$ , which is the value of  $\chi_m(\mathbf{q})$ , averaged over  $\mathbf{q}$ , just before the superconducting transition. There seems to be an interesting recent experimental observation

(Grover and Sarkissian 1983) on the binary alloy  $Y_9Co_7$ , to which our results may be applicable. The magnetic properties of this alloy arises from itinerant electrons (Huang *et al* 1983). While making a.c. magnetic susceptibility measurements on various samples of  $Y_9Co_7$  having different amounts of low level rare earth impurities, Grover and Sarkissian (1983) have observed that for each sample the susceptibility increases monotonically from the highest temperature investigated up to a lower temperature at which it starts showing a peaking behaviour. Immediately below this temperature, there is a superconducting transition with the susceptibility value going over to the Meissner value  $-1/4\pi$ . Their observations also show that a purer sample with higher susceptibility peak value has a higher superconducting  $T_c$  value. On the high temperature side, far away from the peaks,  $\chi_m$  is approximately the same for all the samples.

The mathematical formulation for calculating  $T_c$  in a paramagnetic material, close to the magnetic ordering, and a comparison of the results with the experimental observations (Grover and Sarkissian 1983) is presented in §2. We are concerned only with those experiments done at a fixed normal pressure. Limitations of our formulation of the problem and general discussion of the results are given in §3.

## 2. Superconducting transition temperature close to magnetic ordering

### 2.1 Mathematical formulation

To calculate  $T_c$ , it is usually sufficient to know the static part (Ginzburg and Kirzhnits 1972, 1982; Bhattacharyya and Jha 1978) of the electron-electron interaction in the form of separate contributions arising from the Coulomb term and other exchange-mechanisms, and their respective ranges in frequency. In the multi square-well model, the coupling parameters due to the Coulomb and other terms are obtained by averaging over the possible wavevectors  $q$  being exchanged by the conduction electrons at the Fermi surface. For obtaining a formal relation between the inverse longitudinal dielectric function  $\epsilon^{-1}(q)$  and the magnetic spin susceptibility  $\chi_m(q)$ , together with the resulting expression for the effective Coulomb interaction at  $T \ll T_F$  (the Fermi temperature), one could use, for simplicity, the zero-temperature density-functional theory of inhomogeneous electron gas. However, in this paper, we derive these expressions more rigorously by working (Mermin 1965) with the canonical free energy instead of the ground state energy  $E$ . These relations can also be generalized to take account of the periodic ionic lattice, if necessary by treating (Uspenskii 1979) functions  $\epsilon(q)$ ,  $\chi_m(q)$ , etc., to be matrices in the reciprocal lattice space instead of just numbers.

Let  $n(\mathbf{r}) = n_\uparrow(\mathbf{r}) + n_\downarrow(\mathbf{r})$  and  $\bar{n}(\mathbf{r}) = n_\uparrow(\mathbf{r}) - n_\downarrow(\mathbf{r})$  denote the electron number density and the spin density, respectively, in the electron gas under consideration. In the non-magnetic state, the free energy is minimum for  $n(\mathbf{r}) = n_0$  and  $\bar{n}(\mathbf{r}) = 0$ , in the absence of any external fields. Now, the total free energy of the system in the presence of fixed external fields coupling to these densities can be specified completely by the unique functional  $F[n(\mathbf{r}), \bar{n}(\mathbf{r})]$ , where  $n(\mathbf{r})$  and  $\bar{n}(\mathbf{r})$  are the respective density distributions produced by the fields. The exact functional form  $F[n(\mathbf{r}), \bar{n}(\mathbf{r})]$  is of course unknown for the interacting electron gas. However, for weak external fields, it can be written as a functional expansion in these densities, with the linear functional derivatives being zero for  $\delta n(\mathbf{r}) = n(\mathbf{r}) - n_0 = 0$ ,  $\bar{n}(\mathbf{r}) = 0$ . In particular, the change in free energy due to the weak fields is determined to the lowest order by the inverse longitudinal dielectric

function

$$\varepsilon^{-1}(\mathbf{q}) = 1 - V_0(\mathbf{q})\pi(\mathbf{q}), \quad V_0(\mathbf{q}) = 4\pi e^2/q^2, \quad (1)$$

$$\pi^{-1}(\mathbf{q}) = \left. \frac{\delta^2 F}{\delta n(\mathbf{q})\delta n(\mathbf{q})} \right|_0, \quad (2)$$

and the inverse magnetic spin-susceptibility

$$\chi_m^{-1}(\mathbf{q}) = \left. \frac{1}{\mu_B^2} \frac{\delta^2 F}{\delta \bar{n}(\mathbf{q})\delta \bar{n}(\mathbf{q})} \right|_0. \quad (3)$$

The total free energy  $F$  can be split in terms of the contributions arising from the noninteracting electrons, the Hartree term and the remaining exchange-correlation term, in the form

$$F[n, \bar{n}] = F_0[n, \bar{n}] + F_H[n, \bar{n}] + F_{xc}[n, \bar{n}]. \quad (4)$$

Here, for the Hartree term,

$$\left. \frac{\delta^2 F_H}{\delta n(\mathbf{q})\delta n(\mathbf{q})} \right|_0 = V_0(\mathbf{q}), \quad \left. \frac{\delta^2 F_H}{\delta \bar{n}(\mathbf{q})\delta \bar{n}(\mathbf{q})} \right|_0 = 0, \quad (5)$$

and for noninteracting electrons

$$\left. \frac{\delta^2 F_0}{\delta n(\mathbf{q})\delta n(\mathbf{q})} \right|_0 = \left. \frac{\delta^2 F_0}{\delta \bar{n}(\mathbf{q})\delta \bar{n}(\mathbf{q})} \right|_0 = \pi_0^{-1}(\mathbf{q}) \quad (6)$$

leading to the familiar expression

$$\varepsilon_f^{-1}(\mathbf{q}) = 1 - V_0(\mathbf{q})\pi_0(\mathbf{q}), \quad (7)$$

where in the limit  $q \rightarrow 0$  and  $T \rightarrow 0$  the free polarization bubble  $\pi_0 \rightarrow 3n_0/2E_F$ , the density of states of the electrons at the Fermi surface.

Equation (4) is still a formal relation since the exact  $F_{xc}[n, \bar{n}]$  is not known, even to the lowest order required here. Nevertheless, we can introduce the exchange correlation functions

$$I_e(\mathbf{q}) = \left. \frac{-\delta^2 F_{xc}}{\delta n(\mathbf{q})\delta n(\mathbf{q})} \right|_0, \quad I_m(\mathbf{q}) = \left. -\frac{\delta^2 F_{xc}}{\delta \bar{n}(\mathbf{q})\delta \bar{n}(\mathbf{q})} \right|_0, \quad (8)$$

and show that the difference

$$D_C(\mathbf{q}) = I_e(\mathbf{q}) - I_m(\mathbf{q}) = \left. -\frac{\delta^2 F_{xc}}{\delta n_{\uparrow}(\mathbf{q})\delta n_{\downarrow}(\mathbf{q})} \right|_0 = \left. -\frac{\delta^2 F_C}{\delta n_{\uparrow}(\mathbf{q})\delta n_{\downarrow}(\mathbf{q})} \right|_0, \quad (9)$$

is expected to be a positive quantity, although small compared to  $\pi_0^{-1}$ . Note that the difference  $I_e - I_m$  depends only on the correlation energy, because of its structure. The pure exchange does not contribute. Approximate calculations for the interacting electron gas confirm all these. For  $q < 2K_F$ , it is a slowly varying function of  $\mathbf{q}$ , with  $D_C(0)$  in the range of 0.05 to 0.4 times  $\pi_0^{-1}(0)$  at metallic densities (Shastry 1978; Shastry *et al* 1978) and  $T \ll T_F$ .

Equations (1)–(6) and (8)–(9), immediately lead to the result

$$\mu_B^2 \chi_m^{-1} = \pi_0^{-1} - I_m, \quad (10)$$

$$\pi^{-1} = \pi_0^{-1} + V_0 - I_e = \mu_B^2 \chi_m^{-1} + V_0 - D_C, \quad (11)$$

with the required relation

$$\varepsilon^{-1}(\mathbf{q}) = 1 - V_0(\mathbf{q})\pi(\mathbf{q}) = [\mu_B^2 \chi_m^{-1}(\mathbf{q}) - D_c(\mathbf{q})] \times [\mu_B^2 \chi_m^{-1}(\mathbf{q}) - D_c(\mathbf{q}) + V_0(\mathbf{q})]^{-1}. \quad (12)$$

The effective static Coulomb interaction, then, becomes

$$V_c(\mathbf{q}) = \varepsilon^{-1}(\mathbf{q}) V_0(\mathbf{q}) = [\mu_B^2 \chi_m^{-1}(\mathbf{q}) - D_c(\mathbf{q})] \times [\mu_B^2 \chi_m^{-1}(\mathbf{q}) - D_c(\mathbf{q}) + V_0(\mathbf{q})]^{-1} V_0(\mathbf{q}). \quad (13)$$

The above equation is the central result which shows that  $V_c$  can become negative, if  $\mu_B^2 \chi_m^{-1} < D_c$ , which is possible for a paramagnetic system close to magnetic ordering. This result has exactly the same form as first derived by Uspenskii (1979) for  $T = 0^\circ\text{K}$ . Note that the stability of the system implies  $[\partial^2 F / \partial n \partial n]_0 > 0$ , and the second bracket of (13) can be shown to be positive by using (2) and (11). At higher temperatures  $T_L$ , far away from the magnetic ordering, close to Pauli magnetic susceptibility ( $\mu_B^2 \chi_P^{-1} = \pi_0^{-1}$ ) gives  $\mu_B^2 \chi_m^{-1} > D_c$ , leading to repulsive  $V_c(\mathbf{q})$ .

Since the temperatures involved here are such that  $T \ll T_F$ , one notices that only very close to the magnetic ordering one can assume  $\chi_m^{-1}$  to be a strong function of  $T$ . Far above the magnetic ordering temperature,  $\varepsilon^{-1}$  is a very slowly varying function of  $T$ , and can be approximately taken to be  $\varepsilon^{-1}(\mathbf{q}, T_L)$ ,  $T_L$  being a high enough temperature. If we now split the temperature-dependent effective interaction  $V_c(\mathbf{q}, T)$  in to two parts

$$V_c(\mathbf{q}, T) = (\varepsilon^{-1}(\mathbf{q}, T) / \varepsilon^{-1}(\mathbf{q}, T_L)) (\varepsilon^{-1}(\mathbf{q}, T_L) V_0(\mathbf{q})) \quad (14)$$

and average over allowed  $\mathbf{q}$  values separately (as an approximation), the new Coulomb coupling constant can be written in the form

$$\mu_s(T) \simeq s(T) \mu = \left\langle \frac{\varepsilon^{-1}(\mathbf{q}, T)}{\varepsilon^{-1}(\mathbf{q}, T_L)} \right\rangle \mu(T_L), \quad (15)$$

where  $\mu = \mu(T_L)$  is the usual Coulomb coupling constant in the paramagnetic phase far above the magnetic ordering temperature. Since  $\langle \mu_B^2 \chi_m^{-1} \rangle - \langle D_c \rangle$  is expected to be small compared to  $\langle V_0 \rangle$ , the temperature-dependent multiplying factor  $s(T)$  for the Coulomb coupling constant can be approximately written as

$$s(T) \simeq \left\langle \frac{\mu_B^2 \chi_m^{-1}(\mathbf{q}, T) - D_c(\mathbf{q})}{\mu_B^2 \chi_m^{-1}(\mathbf{q}, T_L) - D_c(\mathbf{q})} \right\rangle = \frac{\langle f^{-1}(T) \rangle}{1 - d_c} - \frac{d_c}{1 - d_c}, \quad (16)$$

where

$$\langle f(T) \rangle = \langle \chi_m(T) / \chi_m(T_L) \rangle \quad (17)$$

represents the average enhancement factor for the susceptibility and

$$d_c = \left\langle \frac{D_c}{\mu_B^2 \chi_m^{-1}(T_L)} \right\rangle < 1 \quad (18)$$

is the dimensionless correlation contribution to  $s(T)$ . For a given  $\mathbf{q}$ , *eg.*,  $\mathbf{q} = 0$ , the enhancement factor  $f(T)$  can be very large ( $> 10^4$  or  $10^5$ ) just before magnetic ordering, but the average  $\langle f(T) \rangle$  is expected to be considerably smaller. Even so,  $\langle f^{-1}(T) \rangle$  can become quite small compared to  $d_c$ . Earlier estimates of  $D_c$  and  $\mu_B^2 \chi_m^{-1}(T_L)$  at metallic densities lead to  $d_c$  values in the range of  $10^{-1}$  or so.

The modification and the reduction in the effective Coulomb interaction near a

magnetic ordering is expected to reduce and modify the phonon coupling constant also from  $\lambda$  to  $\bar{\lambda}$ . However, it is not a straightforward problem to be tackled easily. In any case, in the situation in which the phonon exchange mechanism starts playing a negligible role and  $\mu_s(T) = s(T)\mu$  is negative, the usual expression for  $T_c$ , given by

$$T_c = 1.13\theta_D \exp(-1/g), \quad g = \frac{\bar{\lambda}}{1 + \bar{\lambda}} - \frac{\mu_s}{1 + \mu_s \ln(T_F/\theta_D)} \quad (19)$$

reduces to

$$T_c = 1.13T_F \exp(-1/|s(T_c)|\mu), \quad \text{for } s(T_c) < 0. \quad (20)$$

Assuming  $E_F \simeq 8 \text{ eV}$  in  $\text{Y}_9\text{Co}_7$  and taking typical value of the Coulomb coupling constant  $\mu \simeq 0.25$  at temperature  $T_L$  far above the magnetic ordering temperature, the resulting superconducting  $T_c$  is plotted against  $s$  in figure 1.

Experimentally,  $T_L$  is of course a temperature beyond which all observed susceptibility curves merge together. However, for definiteness we normalize our ratio with respect to the fixed Pauli value. We see that the experimental values (Grover and Sarkissian 1983) of  $T_c$  (see table 1) ranging from 2.90 to 2.39°K for different samples requires  $s$  to be in the band of values around 0.37. This is quite realistic. In fact,  $T_c$  for all

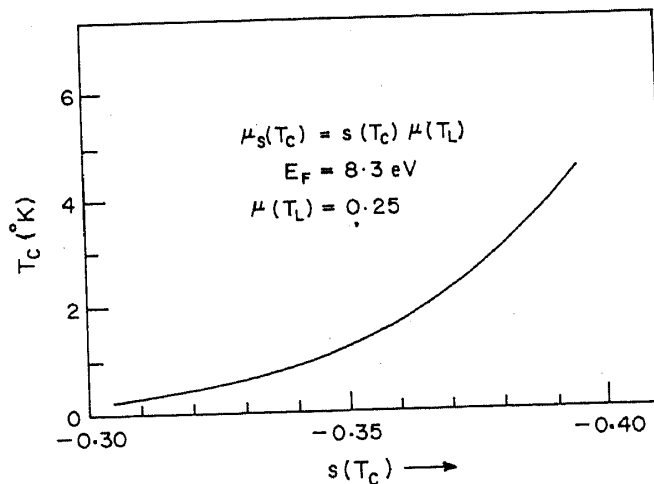


Figure 1. Theoretical plot of the superconducting transition temperature  $T_c$  as a function of the Coulomb coupling modification factor  $s(T_c)$ , close to magnetic ordering.

Table 1. Theoretical fit (equation 20) for experimental  $T_c$  in  $\text{Y}_9\text{Co}_7$ , and the resulting Coulomb coupling modification factor  $s(T_c)$ , the dimensionless correlation term  $d_c$  and the average magnetic spin susceptibility enhancement factor  $\langle f(T_c) \rangle$ .

Sample	Observed*				Theoretical			
	$T_c(^{\circ}\text{K})$	$\chi_m(\text{peak})$ $1/4\pi \text{ esu}$	$\chi_m(\text{peak})$		$s(T_c)$	$d_c$	$\langle f(T_c) \rangle$ $\times 10^2$	Relative $\langle f(T_c) \rangle$
			$\chi_p(\mathbf{q} = 0)$ $\times 10^4$	Relative peak values				
a	2.90	0.45	3.5	1	-0.378	0.276	4	1
b	2.58	0.20	1.6	0.44	-0.374		1.8	0.45
c	2.39	0.17	1.4	0.38	-0.371		1.3	0.33

\* Grover and Sarkissian (1983) and private communications.

the three experimental samples can be fitted quite well with the value of  $d_c = 0.276$ , and the average enhancement factors  $\langle f(T_c) \rangle$  as given in the table. It can be seen that the trend and relative peak values for observed susceptibility are very similar to the relative theoretical values of  $\langle f(T_c) \rangle$ . However, we must emphasize that a similar fit of data can also be obtained by including a small  $\bar{\lambda} \sim 0.1$  to  $0.2$  due to the phonon mechanism in addition to the electronic mechanism considered here.

### 3. Discussion of results and limitations

The picture of the superconducting transition which emerges from our preceding considerations is quite clear. The effective Coulomb coupling constant and the effective static Coulomb interaction in a paramagnetic system are repulsive at high temperatures, where

$$\langle \chi_m^{-1}(\mathbf{q}) \rangle > \langle D_c(\mathbf{q}) \rangle = - \langle \delta^2 F_c / \delta n_{\uparrow}(\mathbf{q}) \delta n_{\downarrow}(\mathbf{q}) \rangle > 0;$$

$F_c$  being the correlation contribution to the finite temperature free-energy of the system. As temperature is lowered towards the magnetic ordering temperature,  $\langle \chi_m^{-1}(T) \rangle$  decreases towards zero, and the effective Coulomb coupling constant  $\mu_s(T)$  as well as the effective Coulomb interaction change sign. Approaching a magnetic ordering with  $q \simeq K_F$  is obviously more favourable for superconductivity. The superconducting transition then takes place either due to this attractive interaction alone or in combination with any other mechanism which may remain attractive. The superconducting transition, of course, does not take place as soon as  $\mu_s(T)$  becomes negative, since the corresponding BCS transition temperature  $T_c$  may be too low to be equal to  $T$ . However, as  $T$  is decreased further, the negative  $\mu_s(T)$  increases in magnitude and the superconducting transition takes place when self-consistently the resulting  $T_c$  becomes exactly equal to  $T$  under consideration. On the other hand, it is possible that before this happens, the magnetic ordering in question occurs instead of the superconducting transition.

Our result discussed above does not depend upon whether it is applicable to the observations on  $\text{Y}_9\text{Co}_7$  or not, which we consider here, as a possible example. There may be a more direct explanation for those observations within the framework of the usual attractive phonon-exchange mechanism and repulsive Coulomb term. However, our result does depend on the crucial assumption that as one approaches the magnetic ordering temperature,  $\langle \chi_m^{-1}(T) \rangle$  goes to zero faster than  $\langle D_c \rangle$ . In the usual spin fluctuation theory (Orlando and Beasley 1981; Grest *et al* 1983; Berk and Schrieffer 1966) the coupling constant  $\lambda_{\text{spin}}$  is proportional to  $\ln[1 + (1 - \langle f^{-1} \rangle) / \langle f^{-1} \rangle]$ , in our notation. If we correct this for the correlation term by changing  $\langle f^{-1} \rangle$  to  $[\langle f^{-1} \rangle - d_c] / (1 - d_c)$ ,  $1 > d_c > 0$ , one indeed sees that  $\lambda_{\text{spin}}$  can change sign as  $\langle \chi_m^{-1} \rangle$  or  $\langle f^{-1} \rangle \rightarrow 0$ .

Before concluding, it may be worthwhile to point out some other important inherent assumptions and limitations in our treatment. To infer the effective interaction between two itinerant electrons relevant for superconductivity, we have used the knowledge of the static dielectric function of the medium; although the frequency range of this modified Coulomb interaction is taken to be quite large ( $|\omega| \leq \omega_F = E_F / \hbar$ ). For the over all average calculation of the effective kernel for the gap equation determining  $T_c$ , this is an acceptable approximation, (see *eg.* Ginzburg and Kirzhnits 1982) since the kernel is a smooth function of  $\omega$ . In our treatment, another very important inherent

assumption is that we have started with the dielectric function  $\epsilon$  relevant for test charges for the determination of effective interaction between two itinerant electrons of the medium. This is strictly valid only when the interaction is between two external charges, not feeling the influence of the interactions the medium itself is undergoing. Since in our case the two itinerant electrons form a part of the interacting electron-gas itself, the appropriate effective dielectric function  $\epsilon_{\text{eff}}$  is not equal to  $\epsilon$ . However, by making the assumption that the interaction is weak (Ginzburg and Kirzhnits 1982), it is possible to use the same expression for both  $\epsilon_{\text{eff}}$  as well as  $\epsilon$ , as a first approximation. Any additional effect, due to exchange, etc., may be neglected in the weak coupling theory. Note that the crucial effect leading to an attractive Coulomb interaction arises from the Coulomb correlation effects rather than the pure exchange effect. It is in fact clear from the low magnitude of the modified Coulomb coupling constants obtained from the experimental observations (Grover and Sarkissian 1983) that the weak coupling theory is quite appropriate in our case. In the case of strong coupling, higher order terms in  $V_c$  and the direct exchange interaction may play a very dominant role and it is difficult to estimate the correct effective electron-electron attraction in that case (Uspenskii 1979).

We have also ignored the important contributions of magnetic dipole-dipole interaction and thermally induced spin-flips in the temperature range of interest, while deriving an explicit relation between  $\chi_m^{-1}$  and  $\epsilon$ . However, implicitly the strong temperature dependence of  $\chi_m^{-1}$  close to magnetic ordering has to be assumed to arise from such considerations, and we must use experimental values of  $\chi_m^{-1}$ . The existence of negative values for the static dielectric function, in any case, does not contradict any general principle or the requirements of system stability (Dolgov *et al* 1981). In fact, calculations have shown that negative values of static dielectric function can occur for a rather wide class of condensed media. In that sense, the possibility of the type of superconducting transition discussed in this paper is quite plausible. However, since we have yet to calculate accurately the phonon coupling constant and its role in  $\text{Y}_9\text{Co}_7$ , from the first principles, we are still far from proving the existence of the electronic mechanism for superconductivity, of the type considered here.

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