Effect of localized spin fluctuations on superconducting properties of dilute alloys*†

Jaime Rössler and Miguel Kiwi †
Facultad de Ciencias, Universidad de Chile, Casilla 653, Santiago, Chile
(Received 24 October 1973)

The effects of localized spin fluctuations on the critical temperature T_c and the specific-heat jump $\Delta C(T_c)$ of dilute superconducting alloys is investigated, both in the weak and strong magnetic limits. In the weak magnetic limit (rapid spin fluctuations) we find that the expression of Kaiser for $T_c(n_I)$, where n_I is the impurity concentration, which was derived for resonance scattering still holds with redefined values of the pertinent parameters, while the specific-heat jump obeys a BCS law of corresponding states. In the slow-spin-fluctuation limit the result of Abrikosov and Gor'kov (AG) for $T_c(n_I)$ is recovered, with a redefined expression for the Cooper-pair lifetime, while $\Delta C(T_c)$ as function of reduced temperature is depressed below the AG value.

I. INTRODUCTION

In the years after the papers of Rivier and Zuckermann[1] and Caplin and Rizzuto[2] on localized spin fluctuations (LSF) were published, much effort has been devoted both to the theoretical[3–5] and experimental[6–10] study of their effect on the properties of dilute alloys. In this contribution we carry out a detailed analysis of how LSF affect the critical temperature T_c and specific-heat jump $\Delta C(T_c)$ of superconducting alloys; our analysis does consider a small but finite concentration of impurities $n_I \equiv N_I/N$, where N_I is the number of paramagnetic impurities and N is the total number of atoms which constitute the system.

The thermodynamic properties we calculate are important to characterize the behavior of the magnetic impurities in a particular matrix; superconductivity is a sensitive tool used to extract the pertinent information because of the very marked difference in the way a magnetic or nonmagnetic perturbation acts on a Cooper pair.

To achieve our purpose we formulate analytically the problem in Sec. II by writing the corresponding Hamiltonian and the corresponding Green's functions to extract the physical information we are looking for. In Sec. II we also solve the self-consistency equations for the propagators in the rapid-spin-fluctuation (weak magnetic) limit, up to $(\Delta/k_B T_c)^3$, where Δ is the superconducting order parameter. The thermodynamic information in this limit is obtained in Sec. III. In Sec. IV a phenomenological model due to Zuckermann[5] is generalized to obtain information on the system in the strong magnetic limit (slow spin fluctuations). Section V is devoted to compare the theoretical results thus obtained with the available experimental information. A critical analysis of the model used and the approximations invoked is given and the paper is concluded with a qualitative comparison of the results of the present LSF theory with theoretical results obtained by Müller-Hartmann and Zittartz on the basis of the Kondo effect.

II. MATHEMATICAL FORMULATION

We formulate the problem analytically writing the Hamiltonian for our system. The alloy is described by Anderson's Hamiltonian,[11] as far as its normal properties are concerned, and the BCS interaction responsible for superconductivity[12] is added to obtain a full description of the superconducting system. We thus have

$$\mathcal{H} = \mathcal{H}_A + \mathcal{H}_{BCS} , \tag{2.1}$$

where

$$\mathcal{H}_A = \sum_{\vec{k},\sigma} \epsilon_k a^\dagger_{\vec{k}\sigma} a_{\vec{k}\sigma} + \sum_j \sum_{m,\sigma} E\, c^\dagger_{jm\sigma} c_{jm\sigma} + \sum_j \sum_{\vec{k},\sigma,m} (V_{j\vec{k}m} a^\dagger_{\vec{k}\sigma} c_{jm\sigma} + \text{H.c.}) + \sum_j \sum_{m,m'} U \hat{n}_{jm\uparrow} \hat{n}_{jm'\downarrow} \tag{2.2}$$

and

$$\mathcal{H}_{BCS} = -\lambda \sum_{\vec{k},\vec{k}'} a^\dagger_{\vec{k}\uparrow} a^\dagger_{-\vec{k}\downarrow} a_{-\vec{k}'\downarrow} a_{\vec{k}'\uparrow} . \tag{2.3}$$

$a^\dagger_{\vec{k}\sigma}$ and $a_{\vec{k}\sigma}$ are the creation and destruction operators for conduction electrons of momentum $\vec{k}$, spin σ, and energy ϵ_k. $c^\dagger_{jm\sigma}$ and $c_{jm\sigma}$ are the creation and destruction operators for localized electron states at site $\vec{R}_j$, angular momentum component m, spin σ, and energy E, which, as all energies, is measured with respect to the Fermi level. The coefficient $V_{j\vec{k}m}$ is

$$V_{j\vec{k}m} = e^{-i\vec{k}\cdot\vec{R}_j} V_{\vec{k}m} , \tag{2.4}$$

where $V_{\vec{k}m}$ is the matrix element between band electrons and electrons localized on the impurity. U is the Coulomb repulsion energy between opposite-spin electrons localized around a particular atomic site and λ is the phonon-induced attraction between conduction electrons responsible for superconductivity.

We now introduce the following Matsubara-type Green's function in the Nambu formalism[13]:

$$\mathcal{G}^{\mu\nu}_{\vec{k}\vec{k}'}(t-t') = -\langle \hat{T}\{\Psi^{\mu}_{\vec{k}}(t)\Psi^{\nu\dagger}_{\vec{k}'}(t')\}\rangle \quad (2.5a)$$

and

$$\mathcal{G}^{\mu\nu}_{ij}(t-t') = -\langle \hat{T}\{\Phi^{\mu}_{i}(t)\Phi^{\nu\dagger}_{j}(t')\}\rangle \ , \quad (2.5b)$$

where $\hat{T}$ is Wick's time ordering operator, $\mu,\nu = 1$ or 2,

$$\Phi^{(1)}_j = c_{jm\uparrow}\ ,\quad \Phi^{(2)}_j = c^{\dagger}_{jm\downarrow}\ ,\quad \Psi^{(1)}_{\vec{k}} = a_{\vec{k}\uparrow}\ ,\quad \Psi^{(2)}_{\vec{k}} = a^{\dagger}_{-\vec{k}\downarrow}\ .$$

The Fourier components $\mathcal{G}^{\mu\nu}(\omega_n)$ of the propagators defined in Eq. (2.5) are given by the general expressions

$$\hat{\mathcal{G}}_{\vec{k}\vec{k}'}(\omega_n) = \hat{G}_{\vec{k}}\Big\{\delta_{\vec{k}\vec{k}'} + \sum_{j,\vec{k}'',m} e^{i(\vec{k}''-\vec{k})\cdot\vec{R}_j} \times V_{\vec{k}m}\hat{\tau}_3\hat{G}_j\hat{\tau}_3 V_{m\vec{k}''}\hat{\mathcal{G}}_{\vec{k}''\vec{k}'}\Big\} \ , \quad (2.6a)$$

$$\hat{\mathcal{G}}_{ij}(\omega_n) = \hat{G}_i\Big\{\delta_{ij} + \Big[\hat{\Sigma}_C + \sum_{l,\vec{k},m} e^{i\vec{k}\cdot(\vec{R}_i-\vec{R}_l)} \times V_{m\vec{k}}\hat{\tau}_3\hat{G}_{\vec{k}}\hat{\tau}_3 V_{\vec{k}m}\Big]\hat{\mathcal{G}}_{lj}\Big\} \ , \quad (2.6b)$$

where Σ_C is the Coulomb self-energy, $\omega_n = (2n+1)\pi k_B T$, and $\hat{\tau}_i$ are the Pauli spin matrices with $\hat{\tau}_0$ designating the unit matrix. The bare propagators $\hat{G}_{\vec{k}}(\omega_n)$ and $\hat{G}_j(\omega_n)$ are of the form

$$\hat{G}_{\vec{k}}(\omega_n) = (i\omega_n\hat{\tau}_0 - \epsilon_{\vec{k}}\hat{\tau}_3 + \Delta\hat{\tau}_1)^{-1} \ , \quad (2.7a)$$

$$\hat{G}_j(\omega_n) = (i\omega_n\hat{\tau}_0 - E\hat{\tau}_3)^{-1} \ . \quad (2.7b)$$

Here Δ is the superconducing order parameter of the BCS theory given by

$$\Delta = \lambda\sum_{\vec{k}}\langle a_{-\vec{k}\downarrow}a_{\vec{k}\uparrow}\rangle = \lambda k_B T\sum_{n,\vec{k}}\mathcal{G}^{21}_{\vec{k}}(\omega_n) \quad (2.8)$$

and λ was defined in Eq. (2.3).

Having formulated our problem analytically we now focus our interest in the solution of the self-consistent set of equations (2.6) which contain all of the thermodynamic information of our superconducting alloy. The first step in the procedure to solve the problem is to average over the spatial distribution of impurities[14]; this restores momentum conservation to the conduction-electron propagator since

$$[\hat{\mathcal{G}}_{\vec{k}\vec{k}'}(\omega_n)]_{\rm av} = \hat{\mathcal{G}}_{\vec{k}}(\omega_n)\delta_{\vec{k}\vec{k}'} \ , \quad (2.9)$$

which considerably simplifies the problem and allows us to treat the finite concentration case.

Next we consider the resonance scattering of conduction electrons by the d or f impurity, described by the matrix element $V_{\vec{k}m}$; out of all possible diagrams we only keep those like Fig. 1(a) but neglect all terms like the one of Fig. 1(b) which describe quantum interference effects between conduction-electron scattering events at different impurity sites. This limits our solution to low concentration of impurities.

Since our main concern are the LSF, it is the Coulomb scattering which we treat more carefully and with more detail. Essentially, we do perturbation theory in U since in the weakly magnetic regime it is the electron-hole multiple scattering processes which are dominant; we thus consider both the "normal" and "superconducting" electron-hole ladder diagrams shown in Fig. 2(a), but with renormalized "particle" propagator internal lines, as exemplified in Fig. 2(b). However, diagrams like those of Fig. 2(c) and 2(d) are discarded; the 2(d) diagrams have been treated by Hamman[15] and Kuroda[16] for the normal state and Kitamura[17] for the superconducting case. In the weakly magnetic limit they lead to small corrections of the results obtained from diagrams of the type of Figs. 2(a) and 2(b).

After all these considerations the set of Eqs. (2.6) reduces to

$$\hat{\mathcal{G}}_{\vec{k}}(\omega_n) = \hat{G}_{\vec{k}}\{1 + (2l+1)n_I V_{\vec{k}m}\hat{\tau}_3\hat{\mathcal{G}}_j\hat{\tau}_3 V_{m\vec{k}}\hat{\mathcal{G}}_{\vec{k}}\} \ , \quad (2.10a)$$

$$\hat{\mathcal{G}}_j(\omega_n) = \hat{G}_j\Big\{1 + \Big[\hat{\Sigma}_C + \sum_{\vec{k}} V_{m\vec{k}}\hat{\tau}_3\hat{\mathcal{G}}_{\vec{k}}\hat{\tau}_3 V_{\vec{k}m}\Big]\hat{\mathcal{G}}_j\Big\} \ , \quad (2.10b)$$

where the Coulomb self-energy matrix Σ_C is given by

$$\hat{\Sigma}_C(\omega_n) = k_B T\sum_l \hat{\mathcal{G}}_j(\omega_n - \Omega_l)\, t^{\rm LSF}(\Omega_l) \ , \quad (2.11)$$

with $\Omega_l = 2l\pi k_B T$ and

$$t^{\rm LSF}(\Omega_n) = U/[1 - U\chi_0(\Omega_n)] \ , \quad (2.12)$$

where we have used the definition

$$\chi_0(\Omega_n) \equiv k_B T\sum_l [\,\mathcal{G}^{11}(\omega_l)\mathcal{G}^{22}(\Omega_n-\omega_l) - \mathcal{G}^{12}(\omega_l)\mathcal{G}^{21}(\Omega_n-\omega_l)] \ , \quad (2.13)$$

which plays the role of a generalized susceptibility appropriate to the superconducting state. As we approach the magnetic region, $U\chi_0(0) \lesssim 1$ and the t matrix $t(\Omega_n)$ shows sharp peak for $\Omega_n \to 0$, justifying our choice of the particle-hole ladder diagrams as the dominant contribution. The electron-electron ladder leads to a density charge matrix of the form

$$[U\chi_0(\Omega)]^2/[1 + U\chi_0(\Omega)] \ ,$$

which is nonsingular at the origin, as shown by Kitamura[17] and can therefore be safely neglected for our purposes; of course we do not neglect the effect of the d or f Coulomb correlations on the superconducting state studied previously.[18]

For simplicity we specialize to the case $E = 0$, which is appropriate[5] for AlMn and AlCr. The most general forms which the Green's functions $\hat{\mathcal{G}}_{\vec{k}}(\omega_n)$ and $\hat{\mathcal{G}}_j(\omega_n)$ can take[5] are

$$\hat{\mathcal{G}}_{\vec{k}}(\omega_n) = [iz(\omega_n)\omega_n \hat{\tau}_0 + \Delta(\omega_n)\hat{\tau}_1 - \epsilon_k \hat{\tau}_3]^{-1} , \tag{2.14a}$$

$$\hat{\mathcal{G}}_j(\omega_n) = [iz_d(\omega_n)\omega_n \hat{\tau}_0 + \Delta_d(\omega_n)\hat{\tau}_1]^{-1} , \tag{2.14b}$$

which substituted into Eqs. (2.10) yield the following self-consistent set of equations:

$$z(\omega_n) = 1 + \frac{c\Gamma}{\pi N(0)} \frac{z_d(\omega_n)}{z_d^2(\omega_n)\omega_n^2 + \Delta_d^2(\omega_n)} , \tag{2.15a}$$

$$\Delta(\omega_n) = \Delta + \frac{c\Gamma}{\pi N(0)} \frac{\Delta_d(\omega_n)}{z_d^2(\omega_n)\omega_n^2 + \Delta_d^2(\omega_n)} , \tag{2.15b}$$

$$z_d(\omega_n) = 1 + \frac{\Gamma z(\omega_n)}{[z^2(\omega_n)\omega_n^2 + \Delta^2(\omega_n)]^{1/2}} + \frac{k_B T}{\omega_n} \times \sum_l \frac{z_d(\omega_{n-l})\omega_{n-l}}{z_d^2(\omega_{n-l})\omega_{n-l}^2 + \Delta_d^2(\omega_{n-l})} \frac{U}{1 - U\chi_0(\Omega_l)} , \tag{2.15c}$$

$$\Delta_d(\omega_n) = \Delta_d + \frac{\Gamma \Delta(\omega_n)}{[z^2(\omega_n)\omega_n^2 + \Delta^2(\omega_n)]^{1/2}} - k_B T \times \sum_l \frac{\Delta_d(\omega_{n-l})}{z_d^2(\omega_{n-l})\omega_{n-l}^2 + \Delta_d^2(\omega_{n-l})} \frac{U^2\chi_0(\Omega_l)}{1 - U\chi_0(\Omega_l)} . \tag{2.15d}$$

Here $c \equiv (2l+1)n_I$ is the impurity concentration times the orbital degeneracy of the $d(l=2)$ or $f(l=3)$ states, $\Gamma = \pi N(0)\langle |V_{\vec{k}m}|^2\rangle$, $N(0)$ is the conduction electron density of states per spin direction at the Fermi level, and Δ_d describes the weakening of the superconducting pairing related to Coulomb correlation effects[18] between electrons on the impurity site. In analogy to the order parameter Δ, given by Eq. (2.8), it is defined as

$$\Delta_d = -U\langle c_{j\downarrow} c_{j\uparrow}\rangle = -U k_B T \sum_n \mathcal{G}_j^{21}(\omega_n) \tag{2.16}$$

and is related to the effect of the Schrieffer-Mattis electron-electron ladder correlation on the superconducting state,[19] which as stated above we do not neglect.

The set of Eqs. (2.15) is identical to Eqs. (2.12) and (2.13) of Ref. 18 with exception of the third term of the right-hand side of Eqs. (2.15c) and (2.15d), which display explicitly the additional effects due to LSF.

Our purpose is to find an expression for the specific-heat jump associated with the superconducting phase transition. This requires a self-consistent solution of Eqs. (2.15) up to third order in Δ, which we carry out next. To start, it is easy to see that $z_d(\omega_n)$ and $\Delta_d(\omega_n)$ are, respectively, even and odd functions of Δ; we then define as $z_{dn}^{(0)}$ and $\Delta_{dn}^{(0)}$ the leading terms of $z_d(\omega_n)$ and $\Delta_d(\omega_n)$ which are of order unity and Δ, respectively, and

$$\delta z_{dn} \equiv z_d(\omega_n) - z_{dn}^{(0)} , \tag{2.17a}$$

$$\delta\Delta_{dn} \equiv \Delta_d(\omega_n) - \Delta_{dn}^{(0)} . \tag{2.17b}$$

With these definitions the set of Eqs. (2.15) expanded up to third order in Δ reads

$$z(\omega_n) = 1 + \frac{c\Gamma}{\pi N(0)} \left(\frac{1}{z_{dn}^{(0)}\omega_n^2} - \frac{\delta z_{dn}}{(z_{dn}^{(0)}\omega_n)^2} - \frac{z_{dn}^{(0)}(\Delta_{dn}^{(0)})^2}{(z_{dn}^{(0)}\omega_n)^4} \right), \tag{2.18a}$$

$$\Delta(\omega_n) = \Delta + \frac{c\Gamma}{\pi N(0)} \left(\frac{\Delta_{dn}^{(0)} + \delta\Delta_{dn}}{(z_{dn}^{(0)}\omega_n)^2} - \frac{(\Delta_{dn}^{(0)})^3 + 2z_{dn}^{(0)}\delta z_{dn}\omega_n^2\Delta_{dn}^{(0)}}{(z_{dn}^{(0)}\omega_n)^4} \right) , \tag{2.18b}$$

$$z_{dn}^{(0)} = 1 + \frac{\Gamma}{|\omega_n|} + \frac{k_B T}{\omega_n} \sum_l \frac{t_N(\Omega_l)}{z_{d,n-l}^{(0)}\omega_{n-l}} , \tag{2.18c}$$

$$\Delta_{dn}^{(0)} = \frac{\Gamma}{|\omega_n|} \left(\frac{\Delta_n^{(0)}}{z_n^{(0)}} \right) - k_B T \sum_l \frac{t_N(\Omega_l)\Delta_{d,n-l}^{(0)}}{(z_{d,n-l}^{(0)}\omega_{n-l})^2} , \tag{2.18d}$$

$$\delta z_{dn} = -\frac{\Gamma}{2|\omega_n|^3} \left(\frac{\Delta_n^{(0)}}{z_n^{(0)}} \right)^2 - \frac{k_B T}{\omega_n} \sum_l \left(\frac{(\Delta_{d,n-l}^{(0)})^2}{(z_{d,n-l}^{(0)}\omega_{n-l})^3} + \frac{\delta z_{d,n-l}}{z_{d,n-l}^{(0)}\omega_{n-l}} \right) t_N(\Omega_l) + \frac{k_B T}{\omega_n} \sum_l \frac{\delta t(\Omega_l)}{z_{d,n-l}^{(0)}\omega_{n-l}} , \tag{2.18e}$$

$$\delta\Delta_{dn} = -\frac{\Gamma}{2|\omega_n|^3} \left(\frac{\Delta_n^{(0)}}{z_n^{(0)}} \right)^3 + \frac{c_n}{|\omega_n|[\pi N(0) + c_n z_{dn}^{(0)}/\Gamma]} \left[\left(\delta\Delta_{dn} - \frac{2\delta z_{dn}\Delta_{dn}^{(0)}}{z_{dn}^{(0)}} - \frac{(\Delta_{dn}^{(0)})^3}{(z_{dn}^{(0)}\omega_n)^2} \right) + \frac{\Delta_n^{(0)}}{z_n^{(0)}} \left(\delta z_{dn} + \frac{(\Delta_{dn}^{(0)})^2}{z_{dn}^{(0)}\omega_n^2} \right) \right] + k_B T \sum_l \left[\frac{(\Delta_{d,n-l}^{(0)})^3}{(z_{d,n-l}^{(0)}\omega_{n-l})^4} - \frac{\delta\Delta_{d,n-l}}{(z_{d,n-l}^{(0)}\omega_{n-l})^2} + \frac{2\Delta_{d,n-l}^{(0)}\delta z_{d,n-l}}{(z_{d,n-l}^{(0)})^3\omega_{n-l}^2} \right] t_N(\Omega_l) - k_B T \sum_l \frac{\Delta_{d,n-l}^{(0)}\delta t(\Omega_l)}{(z_{d,n-l}^{(0)}\omega_{n-l})^2} \tag{2.18f}$$

with

$$\delta t(\Omega_l) \equiv k_B\, T\, t_N^2(\Omega_l) \sum_m \left(\frac{2(\Delta_{dm}^{(0)})^2 + 2z_{dm}^{(0)}\,\delta z_{dm}\omega_m^2}{(z_{dm}^{(0)}\,\omega_m)^3\, z_{d,l-m}^{(0)}\,\omega_{l-m}} - \frac{\Delta_{dm}^{(0)}\,\Delta_{d,l-m}^{(0)}}{(z_{d,l-m}^{(0)}\,\omega_{l-m}\, z_{dm}^{(0)}\,\omega_m)^2} \right) .$$

Above we have used $t_N(\Omega_l)$ to denote the t matrix describing the LSF in the normal state. $z_n^{(0)}$ and $\Delta_n^{(0)}$ denote the first two terms on the right-hand side of Eqs. (2.18a) and (2.18b), respectively, and $c_n = \Gamma^2 c/(z_{dn}^{(0)}\omega_n)^2$.

We notice that Eq. (2.18c) is the well-known expression for a normal state LSF alloy. In this paper we employ the linear solution to this equation due to Hargitai and Corradi[3] within the context of the self-consistent treatment of Paton and Zuckermann[4] which has the form

$$z_{dn}^{(0)} = z + \Gamma/|\omega_n| \;. \tag{2.19}$$

This solution yields good results for $\mathcal{G}_j(\omega_n)$ if $\omega_n \ll \Gamma/z$. In this approximation

$$t_N(\Omega_n) \cong \frac{\pi\Gamma^2}{\Gamma_s + |\Omega_n|} , \tag{2.20}$$

with

$$\Gamma_s \equiv \frac{\pi\Gamma^2}{U}\left(1 - \frac{U}{\pi\Gamma z}\right) . \tag{2.21}$$

$\Gamma_s(U)$ is the inverse lifetime of the LSF. (We work in units of $\hbar = 1$.) The method of Hargitai and Corradi is applicable and gives good results when $\Gamma_s \ll \Gamma/z$.

It is possible to find the solution we are seeking starting by substituting the above expressions for $z_n^{(0)}$, $\Delta_n^{(0)}$ at t_N in Eq. (2.18d); after a long calculation we obtain

$$\Delta_{dn}^{(0)} = \rho + \frac{\Gamma}{|\omega_n|}\,\xi_n , \tag{2.22}$$

where

$$\xi_n \equiv \left(\Delta + \frac{c\Gamma\rho}{\pi N(0)(\Gamma + z|\omega_n|)^2}\right) \Big/ \left(1 + \frac{c\Gamma z}{\pi N(0)(\Gamma + z|\omega_n|)^2}\right), \tag{2.23}$$

$$\rho \equiv -\Gamma^3 b_D \Delta / [(1 + cz/\pi\Gamma N(0)) \times (1 + \Gamma^2 a_\infty + c\Gamma^3 b_\infty/(\pi\Gamma N(0) + cz))] , \tag{2.24}$$

$$a_\infty \equiv \pi k_B T \sum_{n=-\infty}^{\infty} \frac{1}{(\Gamma_s + |\Omega_n|)(\Gamma + z|\omega_n|)^2} , \tag{2.25}$$

$$b_D \equiv \pi k_B T \sum_{\omega_n = -\omega_D}^{\omega_D} \frac{1}{|\omega_n|(\Gamma_s + |\Omega_n|)(\Gamma + z|\omega_n|)^2} , \tag{2.26}$$

$$b_\infty \equiv \pi k_B T \Gamma^2 \sum_{n=-\infty}^{\infty} \frac{1}{|\omega_n|(\Gamma_s + |\Omega_n|)(\Gamma + z|\omega_n|)^4} . \tag{2.27}$$

The subindices of a and b above are related to the cutoff of the summations; when electron-phonon interactions are involved we choose to cut off at the Debye frequency ω_D while Coulomb interactions are kept to arbitrary high energies. The summations above can be carried out to yield

$$a_\infty = \frac{1}{(\Gamma - z\Gamma_s)^2} \ln \frac{\Gamma}{z\Gamma_s} - \frac{1}{\Gamma(\Gamma - z\Gamma_s)} , \tag{2.28}$$

$$b_D = \frac{1}{\Gamma_s \Gamma^2}\left(\alpha_0 + x - \frac{\omega_D}{\Gamma_s} - \frac{2z\omega_D}{\Gamma}\right) , \tag{2.29}$$

$$b_\infty = \frac{1}{\Gamma_s \Gamma^2}\,(K_0 + x - \Gamma^2 a_\infty) , \tag{2.30}$$

with

$$\alpha_0 \equiv \ln\left(\frac{\omega_D}{2\pi k_B T_{c0}}\right) - \psi(\tfrac{1}{2}) , \tag{2.31}$$

$$x \equiv \ln \frac{T_{c0}}{T_c} , \tag{2.32}$$

$$K_0 \equiv \ln\left(\frac{\Gamma}{2\pi z k_B T_{c0}}\right) - \psi(\tfrac{1}{2}) - \tfrac{11}{6} , \tag{2.33}$$

where $\psi(\frac{1}{2}) = -1.96351\ldots$ is the digamma function[20] of $\frac{1}{2}$ and we have assumed $\omega_D \ll \Gamma, \Gamma_s$.

We now investigate (2.18e); noticing that $\Delta_n^{(0)}/z_n^{(0)} = \xi_n$ and invoking again the Hargitai-Corradi[3] approximation we obtain, using an iterative procedure, that to the order in Δ in which we are interested, δz_{dn} is given by

$$\delta z_{dn} \cong -(\Gamma\xi_n^2/2|\omega_n|^3). \tag{2.34}$$

Equation (2.18f) is solved in a similar fashion and yields

$$\delta\Delta_{dn} \cong \tfrac{7}{8}\zeta(3)\frac{\Gamma}{\Gamma_s}\left(\frac{\xi_0^3}{(\pi k_B T)^2}\right) \times \left[1 + \frac{c_n}{|\omega_n|(\pi N(0) + c_n z/\Gamma)}\right] \Big/ \left[1 + \Gamma^2 a_\infty + \frac{c\Gamma^2 b_\infty}{\pi N(0) + cz/\Gamma}\right] - \frac{\Gamma\,\xi_0^3}{2|\omega_n|^3} , \tag{2.35}$$

where $\zeta(s)$ is the Riemann ζ function and c_n, defined after Eqs. (2.18), has the following form in the approximation of Hargitai and Corradi:

$$c_n = \frac{\Gamma^2 c}{(z|\omega_n| + \Gamma)^2} . \tag{2.36}$$

In this way the self-consistent set of Eqs. (2.15) has been solved up to third order in Δ and we can go on to obtain the physical information in which we are interested.

III. THERMODYNAMIC PROPERTIES NEAR T_c IN THE WEAK MAGNETIC REGION

In this section we use the results obtained in Sec. II in order to obtain expressions for the transition temperature T_c of the alloy, as function of the impurity concentration n_I and of the specific-heat discontinuity associated with the normal to superconducting phase transition

$$\Delta C(T_c) = C_s(T_c) - C_N(T_c) .$$

The basic equation to achieve these purposes is Eq. (2.8), which can be cast into the form

$$\Delta = \lambda k_B T \sum_n \sum_{\vec{k}} \frac{\Delta_n}{\Delta_n^2 + \epsilon_k^2 + z_n^2 \omega_n^2} , \tag{3.1}$$

after using the explicit form of $G_{\vec{k}}^{21}(\omega_n)$ given in (2.14a). Carrying out the summation over $\vec{k}$, employing Eqs. (2.18) as well as the expressions obtained for $z_{dn}^{(0)}$, $\delta z_{dn}^{(0)}$, $\Delta_{dn}^{(0)}$, and $\delta\Delta_{dn}$ in Sec. II, the "gap equation" (3.1) can be written

$$1/\lambda = f(T) + g(T)\Delta^2 , \tag{3.2}$$

where

$$f(T) = \frac{2\pi k_B T N(0)}{\Delta} \sum_{n\geq 0} \frac{\xi_n}{\omega_n} , \tag{3.3}$$

$$g(T) = -\frac{2\pi k_B T N(0)}{\Delta^3} \sum_{n\geq 0} \left(\frac{\xi_n^3}{2\omega_n^3} - \frac{c\Gamma}{\pi N(0)\omega_n + c\Gamma/(\Gamma + z\omega_n)} \left\{ \frac{\zeta_n - \rho}{(z\omega_n + \Gamma)^4} \left[\left(\frac{\Gamma\xi_n}{|\omega_n|} + \rho \right)^2 - \frac{\Gamma\xi_n^2(z\omega_n + \Gamma)}{\omega_n^2} \right] \right. \right.$$
$$\left. \left. + \left[\frac{7}{8}\zeta(3) \frac{\Gamma\xi_0^3}{\Gamma_s(\pi k_B T)^2} \middle/ \left(1 + \Gamma^2 a_\infty + \frac{c\Gamma^2 b_\infty}{\pi N(0) + cz/\Gamma} \right) \right] \left[\left(1 + \frac{c_n}{[\pi N(0)\omega_n + c_n z\omega_n/\Gamma]} \right) \middle/ (z\omega_n + \Gamma)^2 \right] \right\} \right) . \tag{3.4}$$

Equation (3.2) has been written in a form appropriate to Landau's second-order phase transition theory[21]; therefore, we can obtain from it complete thermodynamic information about the transition under study.

A. Critical temperature

The critical temperature T_c is obtained directly from the requirement that $\Delta(T_c) = 0$, or, equivalently,

$$f(T_c) = 1/\lambda . \tag{3.5}$$

When $n_I = 0$ we have

$$f(T_{c0}) = 1/\lambda = N(0)\alpha_0 , \tag{3.6}$$

with α_0 defined by Eq. (2.31). Carrying out the summation in (3.3) and using Eqs. (3.5) and (3.6) we obtain

$$T_c = T_{c0} \exp[-An_I/(1 - Bn_I)] , \tag{3.7}$$

where

$$A \equiv \frac{(2l+1)\alpha_0 z}{\pi\Gamma N(0)} \left(1 + \frac{(K_0 + \frac{5}{6})(\Gamma/z\Gamma_s)}{1 + \Gamma^2 a_\infty} \right) , \tag{3.8}$$

$$B \equiv \frac{(2l+1)}{\pi\Gamma_s N(0)} \frac{\alpha_0 + \frac{5}{6} + \Gamma^2 a_\infty}{1 + \Gamma^2 a_\infty} . \tag{3.9}$$

Most parameters used in the above relations are given by Eqs. (2.28)–(2.33).

Equation (3.7) is a generalization of Kaiser's results[22] to include the effect of LSF and extends the work of Zuckermann[5] to finite impurity concentration; the results of Refs. 5 and 22 are correctly recovered when $z = 1$ and $\Gamma_s \sim \pi\Gamma^2/U \to \infty$ (no LSF) and when we evaluate the initial slope of $T_c(n_I)$. It is important to note that the functional form of $T_c(n_I)$ is identical for resonance scattering[22] and when LSF are included; they just imply a redefinition of the parameters A and B as suggested by Luengo *et al.*[10]

B. Specific-heat discontinuity and critical field

Relation (3.2) within the context of Landau's second-order phase transition theory[21] gives the following expression for the Helmholtz free energy F, in the region $T \lesssim T_c$:

$$F_S - F_N = \tfrac{1}{2}\Delta^4 g(T) = \tfrac{1}{2}(T_c - T)\left(\frac{\partial f}{\partial T}\right)^2 \Big/ g(T) . \tag{3.10}$$

It then follows immediately that

$$\Delta C \equiv C_S - C_N \big|_{T_c} = -T_c \left(\frac{\partial f}{\partial T}\right)^2 \Big/ g(T) \Big|_{T_c} \tag{3.11}$$

and $g(T)$ can be evaluated explicitly as

$$g(T) = -\frac{7}{8} \frac{\zeta(3)}{(\pi k_B T)^2} \left(\frac{\xi_0}{\Delta}\right)^3 \times \frac{1 + cz/\pi\Gamma N(0)}{\alpha_0 + x} f(T) . \tag{3.12a}$$

Also $f(T)$ can be obtained explicitly; it reads

$$f(T) \cong \frac{N(0)(\alpha_0 + x)}{1 + cz/\pi\Gamma N(0)} \times \frac{(\pi\Gamma N(0) + cz)(1 + \Gamma^2 a_\infty) - c\Gamma^3 a_\infty/\Gamma_s}{[\pi\Gamma N(0) + cz](1 + \Gamma^2 a_\infty) + c\Gamma(K_0 + x - \Gamma^2 a_\infty)/\Gamma_s} . \tag{3.12b}$$

From here we obtain, by direct use of Eq. (3.11) and keeping only relevant terms that

$$\Delta C = \frac{8\pi^2 k_B^2 T_c}{7\zeta(3)} \left[N(0) + cz/\pi\Gamma\right] . \tag{3.13}$$

Again we recover the functional form of ΔC valid for resonance scattering [see Eq. (4.20) of Ref. 18]; the only difference is that the impurity concentration c is multiplied by the LSF renormalization factor z. Moreover, if we define

$$N_d^{\text{eff}}(0) \equiv z/\pi\Gamma \tag{3.14}$$

as the effective density of d (or f) states at the Fermi level, then the total density of states at this energy level is

$$N_T(0) = N(0) + cN_d^{\text{eff}}(0) \tag{3.15}$$

and we see that the expression for ΔC we obtained above is the same as the one of BCS with an enhanced density of states $N_T(0)$. It follows that

$$\frac{\Delta C}{\Delta C_0} = \left(\frac{T_c}{T_{c0}}\right) \frac{N_T(0)}{N(0)} , \tag{3.16}$$

which is a law of corresponding states.

From the relation (3.10) we can also derive directly an expression for the critical magnetic field H_c which reads

$$H_c^2(T, n_I) = -8\pi(F_S - F_N)$$
$$= -4\pi(T_c - T)^2 \left[(\partial f/\partial T)^2/g(T)\right]\Big|_{T_c} \tag{3.17}$$

and therefore for $T_c - T \ll T_c$,

$$H_c^2(T, n_I) = (4\pi/T_c)(T_c - T)^2 \Delta C , \tag{3.18}$$

which provides an alternative way to proceed from the experimental point of view.

IV. STRONGLY MAGNETIC LIMIT

In this section we generalize a proposal of Zuckermann[5] for the form of the t matrix in the strong magnetic limit, in order to obtain an expression for ΔC when $U \gg \pi\Gamma$. In this limit the LSF lifetime is large compared with the Cooper pair, that is, $\Gamma_s \ll k_B T_c$, in contrast with the cases considered in Secs. II and III.

In addition to the interaction between localized and conduction electrons (described in this paper through t^{LSF}), we now also consider an exchange interaction[23] of the type $J_1 \vec{S} \cdot \vec{\sigma}$, where $\vec{S}$ describes the impurity magnetic moment, $\vec{\sigma}$ is the conduction-electron spin and J_1 is the Heisenberg exchange integral. Our treatment deviates from the usual procedure followed in this context,[24] which consists of adding to J_1 a second contribution $J_2 \cong -2|V_{\vec{k}m}|^2/|E| < 0$ obtained from the Anderson Hamiltonian through the Schrieffer-Wolff transformation[25]; we instead choose to continue describing the LSF through a t matrix in order to obtain detailed information on their effect on the superconducting properties.

Formally, we treat the term $J_1 \vec{S} \cdot \vec{\sigma}$ only to second order in J_1, as Abrikosov and Gorikov[26] (hereafter referred as AG) did; that is, we do not tackle the Kondo effect. On the other hand, we consider the effect of $J_1 \vec{S} \cdot \vec{\sigma}$ and of the LSF as contributing additively to the conduction-electron self-energy (i.e., we neglect quantum interference effects between both scattering mechanisms).

In this context Eqs. (2.15a) and (2.15b) take the form

$$z(\omega_n) = 1 + \frac{c\Gamma}{\pi N(0)} \frac{z_d(\omega_n)}{z_d^2(\omega_n)\omega_n^2 + \Delta_d^2(\omega_n)} + \frac{n_I \Gamma_1 z(\omega_n)}{[z^2(\omega_n)\omega_n^2 + \Delta^2(\omega_n)]^{1/2}} , \tag{4.1a}$$

$$\Delta(\omega_n) = \Delta + \frac{c\Gamma}{\pi N(0)} \frac{\Delta_d(\omega_n)}{z_d^2(\omega_n)\omega_n^2 + \Delta_d^2(\omega_n)} - \frac{n_I \Gamma_1 \Delta(\omega_n)}{[z^2(\omega_n)\omega_n^2 + \Delta^2(\omega_n)]^{1/2}} , \tag{4.1b}$$

where[26]

$$\Gamma_1 \equiv \tfrac{1}{4}\pi N(0) J_1^2 S(S+1) . \tag{4.2}$$

Following Zuckermann[5] we now make the basic assumption that it is sufficient to keep the dominant $\Omega_n = 0$ component of $t^{\text{LSF}}(\Omega_n)$ in the strong magnetic limit. Equations (4.1) then take the form

$$z_{dn}^{(0)} = 1 + \frac{\Gamma}{|\omega_n|} + \frac{\Xi}{z_{dn}^{(0)}\omega_n^2} , \tag{4.3a}$$

$$\Delta_{dn}^{(0)} = \frac{\Gamma}{|\omega_n|} \frac{\Delta_n^{(0)}}{z_n^{(0)}} - \frac{\Delta_{dn}^{(0)}\Xi}{(z_{dn}^{(0)}\omega_n)^2} , \tag{4.3b}$$

$$\delta z_{dn} = -\frac{\Gamma}{2|\omega_n|^3}\left(\frac{\Delta_n^{(0)}}{z_n^{(0)}}\right)^2 + \frac{\Pi}{z_{dn}^{(0)}\omega_n^2} - \frac{\Xi}{\omega_n}\left(\frac{(\Delta_{dn}^{(0)})^2}{(z_{dn}^{(0)}\omega_n)^3} + \frac{\delta z_{dn}}{(z_{dn}^{(0)})^2\omega_n}\right) , \tag{4.3c}$$

$$\delta\Delta_{dn} = -\frac{\Gamma}{2|\omega_n|^3}\left(\frac{\Delta_n^{(0)}}{z_n^{(0)}}\right)^3 + \frac{c\Gamma^2}{\pi N(0) z_n^{(0)}\omega_n}\left[\frac{\delta\Delta_{dn}}{(z_{dn}^{(0)}\omega_n)^2} - \frac{2\Delta_{dn}^{(0)}\delta z_{dn}}{(z_{dn}^{(0)})^3\omega_n^2} - \frac{(\Delta_{dn}^{(0)})^3}{(z_{dn}^{(0)}\omega_n)^4} + \frac{\Delta_n^{(0)}}{z_n^{(0)}}\left(\frac{\delta z_{dn}}{(z_{dn}^{(0)}\omega_n)^2} + \frac{z_{dn}^{(0)}(\Delta_{dn}^{(0)})^2}{(z_{dn}^{(0)}\omega_n)^4}\right)\right]$$

$$+\left(\frac{(\Delta_{dn}^{(0)})^3}{(z_{dn}^{(0)}\omega_n)^4}-\frac{\delta\Delta_{dn}}{(z_{dn}^{(0)}\omega_n)^2}+\frac{2\Delta_{dn}^{(0)}\delta z_{dn}}{(z_{dn}^{(0)})^3\omega_n^2}\right)\Xi-\frac{\Pi\Delta_{dn}^{(0)}}{(z_{dn}^{(0)}\omega_n)^2}\ , \qquad (4.3d)$$

where we have used definitions (2.17), Eqs. (2.18), and

$$k_B T\, t^{\mathrm{LSF}}(\Omega_n=0)\equiv\Xi(T)+\Pi(T)\ , \qquad (4.4)$$

with $\Xi(T)$ corresponding to the normal and $\Pi(T)$ corresponding to the superconducting elements (proportional to Δ^2) of the t^{LSF} matrix. Clearly this is a phenomenological theory, where we ignore the relation between Ξ and Π and the relation of these parameters with those of the Anderson model (U and Γ).

The set of Eqs. (4.3) has a triangular structure, where the first one can be solved algebraically and the rest by substitution of the solutions of the preceding ones.

A. Critical temperature

In order to obtain an expression for the critical temperature we have to find a solution for the self-consistent set of Eqs. (4.1) and (4.3) up to order Δ. The renormalized-random-phase approximation (RRPA) of Hamman[15] suggests that $\Xi\sim U^2\gg\Gamma$ and $\Pi\sim\Gamma\Delta^2\Xi^{1/2}/(k_BT)^2$ and thus, keeping only relevant terms, we obtain

$$z_{dn}^{(0)}=\Xi^{1/2}/|\omega_n|\ , \qquad (4.5a)$$

$$\Delta_{dn}^{(0)}=\Gamma\xi_n/2|\omega_n|\ , \qquad (4.5b)$$

$$z_n^{(0)}=1+\frac{1}{|\omega_n|}\left(\frac{c\Gamma}{\pi N(0)\Xi^{1/2}}+n_I\Gamma_1\right)\ , \qquad (4.5c)$$

$$\Delta_n^{(0)}=\Delta+\frac{\xi_n}{|\omega_n|}\left(\frac{c\Gamma^2}{2\pi N(0)\Xi}-n_I\Gamma_1\right), \qquad (4.5d)$$

where again

$$\xi_n\equiv\frac{\Delta_n^{(0)}}{z_n^{(0)}}=\frac{\Delta}{1+2\pi k_BT_c\lambda_c/|\omega_n|} \qquad (4.6)$$

and

$$\lambda_c\equiv Q/2\pi k_BT_c\ , \qquad (4.7)$$

with

$$Q\cong\tfrac{1}{2}n_I\pi N(0)J_1^2S(S+1)+\frac{c\Gamma}{\pi N(0)\Xi^{1/2}}\ . \qquad (4.8)$$

The critical temperature T_c is obtained as in Sec. III, that is, by use of (3.3) and (3.5) in combination with (4.5), which yields

$$\ln(T_{c0}/T_c)=\psi(\tfrac{1}{2}+\lambda_c)-\psi(\tfrac{1}{2})-\ln(1+Q/\omega_D)\ , \qquad (4.9)$$

where $\psi(x)$ is the digamma function.[20]

Noticing that Q/ω_D is so much smaller than unity as to make the logarithmic term above negligible and recalling the AG[26] expression

$$\ln(T_{c0}/T_c)=\psi(\tfrac{1}{2}+1/\pi k_BT_c\tau)-\psi(\tfrac{1}{2})\ , \qquad (4.10)$$

we obtain for the pair lifetime τ the relation

$$1/\tau=1/\tau_{\mathrm{AG}}+1/\tau_{\mathrm{LSF}}\ , \qquad (4.11)$$

where

$$\tau_{\mathrm{AG}}^{-1}=\tfrac{1}{4}n_I\pi N(0)J_1^2S(S+1)=\pi k_BT_c\lambda_c^{\mathrm{AG}} \qquad (4.12a)$$

and

$$\tau_{\mathrm{LSF}}^{-1}=\frac{c\Gamma}{2\pi N(0)\Xi^{1/2}}=\pi k_BT_c\lambda_c^{\mathrm{LSF}}\ . \qquad (4.12b)$$

As expected, they are the same expressions obtained by previous authors in the corresponding limits.[5,26]

The critical concentration n_{cr} is obtained by requiring $T_c=0$ in Eq. (4.9), which yields

$$\lambda_c=\frac{n_I}{n_{\mathrm{cr}}}\frac{T_{c0}}{T_c}e^{\psi(1/2)}\ . \qquad (4.13)$$

The value of n_{cr} thus obtained justifies the approximations invoked, in particular dropping the logarithmic term in Eq. (4.9).

B. Specific heat

In order to evaluate the specific-heat discontinuity we need to know δz_n and $\delta\Delta_n$ (to order Δ^2 and Δ^3, respectively); assuming $\Xi^{1/2}\gg\Gamma T_{c0}/T_c$ (strong magnetic limit) we obtain a significantly simplified version of our self-consistency equations which reads

$$\delta z_{dn}=\frac{\Pi}{2\Xi^{1/2}|\omega_n|}-\frac{\Gamma\xi_n^2}{4|\omega_n|^3}\ , \qquad (4.14a)$$

$$\delta\Delta_{dn}=-\frac{\Gamma}{4}\frac{\xi_n^3}{|\omega_n|^3}\ , \qquad (4.14b)$$

$$\delta z_n=-\frac{n_I\Gamma_1\Delta^2}{2|\omega_n|(|\omega_n|+2\pi k_BT_c\lambda_c)^2}\ , \qquad (4.14c)$$

$$\delta\Delta_n=\tfrac{1}{2}n_I\Gamma_1\Delta^3\frac{|\omega_n|+2\pi k_BT_c\lambda_c^{\mathrm{LSF}}}{(|\omega_n|+2\pi k_BT_c\lambda_c)^4}\ . \qquad (4.14d)$$

Use of (4.14) in combination with Eq. (3.2) yields

$$g(T_c)=\frac{-N(0)}{2(2\pi k_BT_c)^2}\left[\zeta(3,\tfrac{1}{2}+\lambda_c)-\lambda_c^{\mathrm{AG}}\zeta(4,\tfrac{1}{2}+\lambda_c)\right], \qquad (4.15)$$

where $\zeta(n,a)=\sum_{j=0}^{\infty}(j+a)^{-n}$ is the generalized Riemann ζ function.[20] Combination of (4.15) with Eqs.

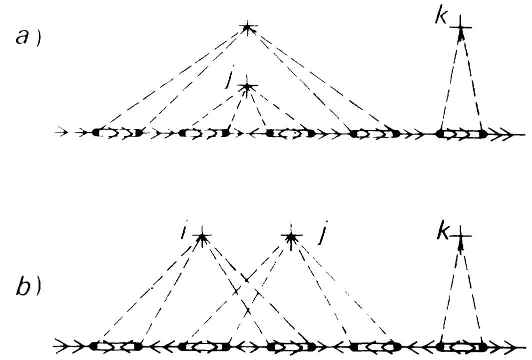

FIG. 1. Diagrams which represent the resonance scattering of conduction electrons by the d or f levels of the impurity. Here $G_{\vec{k}}^{11}$ = →→, $G_{\vec{k}}^{22}$ = ←←, $G_{\vec{k}}^{12}$ = →←, and $G_{\vec{k}}^{21}$ = ←→, while the double lines represent the analogous d- or f-electron propagators. The crosses stand for an impurity site, while the dotted lines represent the interaction $V_{\vec{k}} e^{i\vec{k}\cdot\vec{R}_j}$.

(3.3) and (3.11) yields the final expression

$$\frac{\Delta C}{\Delta C_0} = \frac{T_c}{T_{c0}} \frac{\zeta(3,\frac{1}{2})\,[1-\lambda_c\zeta(2,\frac{1}{2}+\lambda_c)]^2}{\zeta(3,\frac{1}{2}+\lambda_c) - \alpha\lambda\,\zeta(4,\frac{1}{2}+\lambda_c)}\,, \tag{4.16}$$

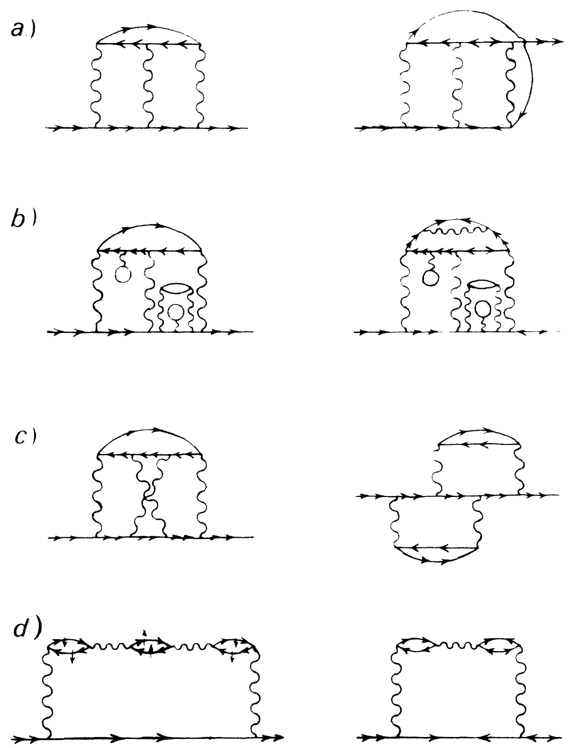

FIG. 2. Diagrams representing the Coulomb correlation of the d or f electrons. Here G_j^{11} = →→, G_j^{22} = ←←, G_j^{12} = →←, and G_j^{21} = ←→, while the wiggly lines stand for the electron-hole Coulomb interaction U.

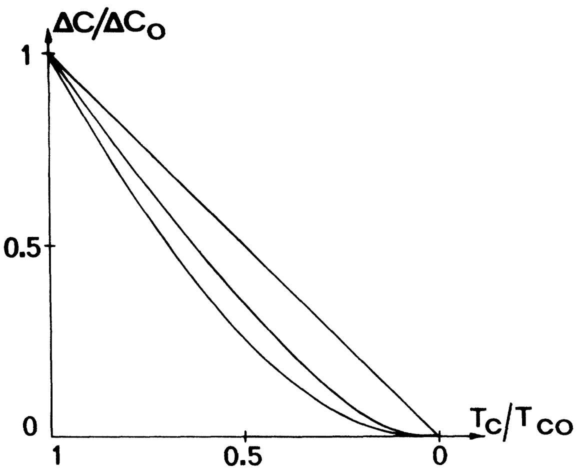

FIG. 3. Plot of the reduced specific-heat jump $\Delta C/\Delta C_0$ vs reduced critical temperature T_c/T_{c0}. The straight line represents the BCS law of corresponding states and the intermediate curve represents the relation obtained by Abrikosov and Gor'kov. The lowest curve illustrates the strong LSF case. The initial slopes are 1, 1.45, and 1.84, respectively.

with

$$\alpha \equiv \frac{\tau_{AG}^{-1}}{\tau_{AG}^{-1} + \tau_{LSF}^{-1}}\,. \tag{4.17}$$

The parameter $0 \le \alpha \le 1$ is a measure of the strength of $J_1\vec{S}\cdot\vec{\sigma}$ relative to the total scattering.

Since λ_c is a function of T_c/T_{c0} alone the graph for $\Delta C/\Delta C_0$ vs T_c/T_{c0} is universal for a fixed value of α and is illustrated in Fig. 3. The initial slope

$$\left(\frac{\partial(\Delta C/\Delta C_0)}{\partial(T_c/T_{c0})}\right)_{n_I=0}$$

varies between 1.84 ($\alpha = 0$) and 1.45 ($\alpha = 1$).

V. COMPARISON WITH EXPERIMENTS AND CONCLUSION

Here we discuss the results obtained above, mainly in Secs. III and IV, in the light of the existing experimental information. Moreover, we analyze critically the theoretical implications and limitations of this work.

There are several superconducting systems which can be classified as being in the transition region between magnetic and nonmagnetic behavior. The best documented experimentally[24] are AlMn and ThU, but AlCr also shows similar characteristics. Some of these characteristics are the following. The impurity contirubtion to the resistivity varies as

$$\Delta\rho = \rho_0[1 - (T/\theta^{LSF})^2]\,, \tag{5.1}$$

with θ^{LSF} being a temperature suggested by Caplin and Rizzuto[2] to be related to the spin-fluctuation frequency ($\tau_{\mathrm{LSF}}^{-1} \sim k_B \theta^{\mathrm{LSF}}$). These systems show a relation between T_c and n_I given by Eq. (3.7), which was derived by Kaiser[22] for resonance scattering and which with redefined parameters we have shown to be valid also for LSF. Furthermore, they obey a BCS law of corresponding states for the specific-heat jump $\Delta C/\Delta C_0$ versus reduced transition temperature T_c/T_{c0}; this is also in good agreement with our expression (3.16).

Having established that qualitatively our theory for rapid spin fluctuations (weak magnetic limit) is in good agreement with available experimental information, we now proceed to analyze quantitatively the existing data of the two best-known[24] systems: *Al*Mn and *Th*U.

For *Al*Mn the parameter $\alpha_0 = 5.9$, the pure-Al density of states $N(0) = 0.4$ states/eV atom and, according to Huber,[27] $A = 576 \pm 64$ and $B = 88 \pm 4$. Using these experimental values we find through numerical solution of the self-consistent set of Eqs. (3.8) and (3.9) the values $z\Gamma_s/\Gamma = 0.19$ and $\Gamma/z = 1.7$ eV. These values have to be considered only as a rough estimate, since they are extremely sensitive to small variations of the critical concentration which has a large experimental uncertainty. Using the relation[4]

$$\theta^{\mathrm{LSF}} = (\sqrt{3}/\pi k_B)(\Gamma/z)[1 + \tfrac{3}{2}(\Gamma/z\Gamma_s)^2]^{-1/2} , \quad (5.2)$$

we obtain $\theta^{\mathrm{LSF}} \cong 1600$ °K for *Al*Mn which is considerably larger than the experimental value[24] of 530 °K, but is significantly smaller than the value obtained from work of Maple *et al.*[28] of 3000 °K, calculated on the basis of a best fit to Eq. (3.7) without inclusion of LSF effects.

For *Th*U we know[10] that $\alpha_0 = 4.8$ $A = 630$, and $B = 130$. The pure-Th density of states is 1.84 states/eV atom, which following the same procedure outlined above for *Al*Mn yields $z\Gamma_s/\Gamma = 0.226$ and $\Gamma/z = 0.26$ eV. The result is $\theta^{\mathrm{LSF}} = 300$ °K as compared with the experimental value[28] of 100 °K.

The reason for the remaining discrepancy between rapid LSF results and experimental values has to be found in the fact that while our theory improves upon previous work, the model used still has several limitations. First, we assume that the width Γ of the localized states is much larger than their distance to the Fermi level; this seems quite reasonable for *Al*Mn, but is only a working hypothesis for *Th*U. Next, the weak magnetic limit requires that $(1 + z\Gamma_s/\Gamma)^3 \gg 1$; when this condition is not strictly satisfied, which happens to be the case of our two examples, the interaction between LSF may play a significant role; a calculation of this effect (which is quite involved) might contribute to explain the anomalously large density of *f* states at the Fermi level observed in *Th*U, which has not yet been accounted for.

As far as the strong magnetic regime is concerned the situation is not as clearcut. Vaccarone *et al.*[29] have recently suggested that superconducting magnetic alloys could be classified, on the basis of the specific-heat jump $\Delta C/\Delta C_0$ as function of T_c/T_{c0}, in three groups: (a) systems obeying a BCS law of corresponding states, (b) systems which follow the Abrikosov and Gorikov relation (see Fig. 3 for $\alpha = 1$), and (c) systems obeying a "universal" law with more depressed values of $\Delta C(T_c)$. Examples of type (a) systems were given above; type (b) systems are *La*Gd and *Th*Gd, while type (c) alloys are (*LaCe*)Al_2, *Zn*Mn, and *Zn*Cr. When comparing with theoretical results it is quite clear that the initial slope

$$\left(\frac{\partial(\Delta C/\Delta C_0)}{\partial(T_c/T_{c0})}\right)_{n_I=0}$$

of the universal curve for type (c) systems is larger than the maximum value of 1.84 allowed for by our theory and is also larger than 2.48, the maximum value permitted by the theory of Müller-Hartmann and Zittartz,[30] which is based on the Kondo effect.

On the other hand, recent work on the remarkable $(La_x Th_{1-x})$Ce system[31] shows that it goes over continously from a Kondo-like behavior for $x = 1$ to a BCS law of corresponding states for $x = 0$.

Our theory of LSF allows for either BCS-like behavior (see Fig. 3) or the region between AG ($\alpha = 1$) and the curve corresponding to $\alpha = 0$. The region between BCS and $\alpha = 1$ and below the $\alpha = 0$ curve is not excluded in principle, but the transition from one to the other requires going through the regime $\Gamma_S \sim k_B T_c$, where the approximations we have invoked breakdown completely.

It is worth comparing qualitatively our results with those of Ref. 30, in spite of the fact that they apply to different models. In the strong magnetic region ($\alpha \to 1$ or $T_K/T_{c0} \to 0$), both theories recover the AG result. In the intermediate regime ($\alpha \to 0$ or $T_K/T_{c0} \to 1$), both show maximum initial slope (1.84 and 2.48, respectively). However, in the weak magnetic region (rapid spin fluctuations or $T_K/T_{c0} \to \infty$), they differ in that we recover a BCS-like law, while Müller-Hartmann and Zittartz find an AG-like behavior. Semiquantitative analysis of the expressions for $T_c(n_I)$ in the weak and strong magnetic limits [Eqs. (3.7) and (4.9), respectively] indicate that the analogy of our results with those for the Kondo effect[32] could also apply to the dependence of the transition temperature on impurity concentration in the regime $\Gamma_s \sim k_B T_{c0}$.

In summary, we have studied the effect of LSF on superconducting properties up to third order in Δ, both in the weak and strong magnetic regimes. For rapid spin fluctuations ($\Gamma_s \gg k_B T_c$) we have found that Kaiser's formula[22] for T_c as function of n_I holds, with redefined expressions for the pertinent parameters, while a BCS law of corresponding states is obeyed by the specific-heat jump. Our expressions are in better agreement with experiment than previous work, but there still remain unexplained features, like the high density of states[10] of *Th*U. In the strong magnetic regime we have extended previous work of Zuckermann[5] which allows an alternative to the explanation based on the Kondo effect, for the depression of the specific-heat jump as function of reduced temperature.

ACKNOWLEDGMENTS

We gratefully acknowledge very informative conversations with Miguel Roth about his experimental results and thank Professor C. Caroli and Professor M. Zuckermann for helpful conversations.

*Work supported in part by the University of Chile, University of California Cooperative Program.

†This work is based on a dissertation presented by J. Rössler to the University of Chile, in partial fulfillment of the requirements for a Ph.D. degree.

‡At present John Simon Guggenheim Fellow at the University of California, La Jolla, Calif. 92037.

[1]N. Rivier and M. J. Zuckermann, Phys. Rev. Lett. 21, 904 (1968).
[2]A. D. Caplin and C. Rizzuto, Phys. Rev. Lett. 21, 746 (1968).
[3]C. Hargitai and C. Corradi, Solid State Commun. 7, 1535 (1969).
[4]B. Paton and M. J. Zuckermann, J. Phys. F 1, 125 (1971).
[5]M. J. Zuckermann, J. Phys. C 3, 2130 (1970).
[6]R. Aoki and T. Ohtsuka, J. Phys. Soc. Jap. 23, 955 (1967).
[7]R. Aoki and T. Ohtsuka, J. Phys. Soc. Jap. 26, 651 (1969).
[8]G. Boato, G. Gallinaro, and C. Rizzuto, Phys. Rev. 148, 353 (1966).
[9]G. Boato and J. Vig, Solid State Commun. 5, 649 (1967).
[10]C. A. Luengo, J. M. Cotignola, J. G. Sereni, A. R. Sweedler, M. M. Maple, and J. G. Huber, Solid State Commun. 10, 463 (1972).
[11]P. W. Anderson, Phys. Rev. 124, 41 (1961).
[12]J. Bardeen, L. Cooper, and J. R. Schrieffer, Phys. Rev. 108, 1175 (1957).
[13]J. R. Schrieffer, in *The Theory of Superconductivity* (Benjamin, New York, 1964).
[14]G. Rickayzen, *International Spring School of Physics, Bergen*, 1961 (Benjamin, New York, 1964).
[15]D. R. Hamman, Phys. Rev. 186, 549 (1969).
[16]Y. Kuroda, Prog. Theor. Phys. 43, 870 (1970).
[17]T. Kitamura, Prog. Theor. Phys. 47, No. 3 (1972).
[18]M. Kiwi and M. J. Zuckermann, Phys. Rev. 164, 548 (1967).
[19]J. R. Schrieffer and D. C. Mattis, Phys. Rev. 140, A1412 (1965).
[20]*Handbook of Mathematical Functions*, edited by M. Abramonwitz and I. Stegun (Dover, New York, 1965).
[21]L. D. Landau and E. M. Lifshitz, in *Statistical Physics* (Pergamon, London, 1958).
[22]A. B. Kaiser, J. Phys. C 3, 410 (1970).
[23]T. Kasuya, Prog. Theor. Phys. 16, 58 (1956).
[24]M. B. Maple, in *Magnetism*, edited by G. T. Rado and H. Suhl (Academic, New York, 1973), Vol. V, p. 289.
[25]J. R. Schrieffer and P. A. Wolff, Phys. Rev. 149, 491 (1966).
[26]A. A. Abrikosov and L. P. Gor'kov, Zh. Eksp. Teor. Fiz. 39, 1781 (1960) [Sov. Phys. JETP 12, 1243 (1961)]. S. Skalski, O. Betbeder-Matibet, and P. R. Weiss Phys. Rev. 136, A1500 (1964).
[27]J. G. Huber, Ph.D. thesis (University of California, 1971) (unpublished).
[28]M. B. Maple, J. C. Huber, B. R. Coles, and A. G. Lawson, J. Low Temp. Phys. 3, 137 (1970).
[29]R. Vaccarone, A. Morozzo della Rocca, A. Pilot, V. Vivaldi, and C. Rizzuto, Solid State Commun. 12, 885 (1973).
[30]E. Müller-Hartmann and J. Zittartz, Solid State Commun. 11, 401 (1972).
[31]C. A. Luengo, J. G. Huber, M. B. Maple, and M. Roth, Phys. Rev. Lett. 32, 54 (1974).
[32]E. Müller-Hartmann and J. Zittartz, Z. Phys. 234, 58 (1970).