### APPENDIX

We will give the long-wavelength limit of  $\Phi_{1234}^{(i)}$  for both the Dyson-Maleev Hamiltonian and the Holstein-Primakoff Hamiltonian arising from Eq. (2). We denote the coefficients for the Holstein-Primakoff Hamiltonian by  $\tilde{\Phi}_{1234}^{(i)}$ . It is readily seen that the Holstein-Primakoff Hamiltonian  $\tilde{\mathcal{R}}$  is related to the Dyson-Maleev Hamiltonian by

Hence,

 $\tilde{\Phi}_{1234}^{(9)} = \tilde{\Phi}_{1234}^{(1)} = \frac{1}{2} (\Phi_{1234}^{(1)} + \Phi_{3412}^{(1)}),$ (A2a)

 $\tilde{\mathcal{K}} = \frac{1}{2}(\mathcal{K} + \mathcal{K}^{\dagger}).$ 

$$\begin{split} \tilde{\Phi}_{3412}{}^{(3)} &= \tilde{\Phi}_{1234}{}^{(2)} = \tilde{\Phi}_{2143}{}^{(6)} = \tilde{\Phi}_{3421}{}^{(5)} \\ &= \frac{1}{2} \left( \Phi_{1234}{}^{(2)} + \Phi_{3412}{}^{(3)} \right), \quad (A2b) \end{split}$$

$$\tilde{\Phi}_{1234}^{(4)} = \frac{1}{2} \left( \Phi_{1234}^{(4)} + \Phi_{3412}^{(4)} \right), \qquad (A2c)$$

 $\tilde{\Phi}_{1234}^{(7)} = \tilde{\Phi}_{1234}^{(8)} = \frac{1}{2} (\Phi_{1234}^{(7)} + \Phi_{3412}^{(8)}).$ (A2d)

#### PHYSICAL REVIEW

Thus, it is only necessary to give expressions for the  $\Phi_{1234}$  (*i*). For no anisotropy these coefficients are proportional to two powers of momenta, whereas for  $D \neq 0$  they approach a constant value. We will evaluate only these two terms in each case. We use

$$x_i = (1 - \epsilon_i)^{1/2} (1 + \epsilon_i)^{-1/2} = 1 - \epsilon_i + \frac{1}{2} \epsilon_i^2 + \cdots, \quad (A3a)$$

$$\gamma_i = 1 - \frac{1}{8}k_i^2, \tag{A3b}$$

whence

(A1)

$$\Phi_{1234}^{(1)} \sim 4q + \left(\frac{1}{2}k_3 \cdot k_4 - 2\epsilon_3 \epsilon_4\right), \qquad (A4a)$$

$$\Phi_{1234}{}^{(2)} \sim -4q - \left(\frac{1}{2}k_3 \cdot k_4 - 2\epsilon_3 \epsilon_4\right), \qquad (A4b)$$

$$\Phi_{1234}{}^{(3)} \sim -4q - \left(\frac{1}{2}k_3 \cdot k_4 + 2\epsilon_3 \epsilon_4\right), \qquad (A4c)$$

$$\Phi_{1234}{}^{(4)} \sim 4q + \left(\frac{1}{2}k_3 \cdot k_4 + 2\epsilon_3 \epsilon_4\right), \qquad (A4d)$$

$$\Phi_{1234}^{(7)} \sim 4q + \left(\frac{1}{2}k_3 \cdot k_4 - 2\epsilon_3\epsilon_4\right). \tag{A4e}$$

These formulas differ from those of Ref. 2 by the terms in  $\epsilon_3 \epsilon_4$ .

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# Spin Fluctuations Associated with the Formation of Localized **Magnetic Moments in Superconductors**

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A new theory of superconductors containing transition-metal impurities is presented, explaining the observed anomalous magnetic behavior of the impurities with quenched or nearly quenched magnetic moment resulting from the localized spin fluctuations associated with the formation of localized magnetic moments. From this theory, the superconducting transition temperatures of AlMn, AlCr, and VFe are calculated. The puzzling "slowing down" of the decrease in T<sub>c</sub> observed for higher concentrations of transitionmetal impurities is shown to arise partly from the temperature dependence of the electron scattering by the localized spin fluctuations and also partly from exchange and Coulomb coupling among the transition-metal impurities, which damp the formation of the localized magnetic moments and the spin fluctuations.

## I. INTRODUCTION

 $\mathbf{E}^{\mathrm{LECTRON}}$  scattering by localized spins strongly weakens superconductivity,<sup>1,2</sup> in sharp contrast to the small mean-free-path effects of nonmagnetic impurities.3 Consequently, superconductors should reflect sensitively the quenching of localized magnetic moments.<sup>4</sup>

Grant No. AFOŠR-611-67. <sup>1</sup> R. A. Hein, R. L. Falge, Jr., B. T. Matthias, and E. Corenzwit, Phys. Rev. Letters **2**, 500 (1969); J. E. Crow and R. D. Parks, Phys. Letters **21**, 378 (1966); A. A. Abrikosov and L. P. Gor'kov, Zh. Eksperim. i Teor. Fiz. **39**, 1781 (1960) [English transl.: Soviet Phys.—JETP **12**, 1243 (1961)]. <sup>2</sup> G. Boato, G. Gallinaro, and C. Rizzuto, Phys. Rev. **148**, 353 (1966); J. Muller, Helv. Phys. Acta **32**, 141 (1959); E. W. Collings and F. T. Hedgcock, Phys. Rev. **126**, 1654 (1962); F. T. Hedgcock, W. B. Muir, and E. E. Wallingsford, Can. J. Phys. **38**, 376 (1960). (1960). <sup>3</sup> D. Markowitz and L. P. Kadanoff, Phys. Rev. 131, 563 (1963).

<sup>4</sup> P. W. Anderson, Phys. Rev. 124, 41 (1961); P. A. Wolff, P. W. Anderson, A. M. Clogston, B. T. Matthias, M. Peter, and H. J. Williams, J. Appl. Phys. 33, 1173 (1962).

The observed superconducting transition temperatures  $T_c$  of Al, Zn, In, Sn, and V containing transition-metal impurities like Cr, Mn, Fe and Ni, for example, confirm this expectation.<sup>2</sup> However, the measurements of  $T_c$ revealed an anomalous magnetic behavior of the transition-metal impurities with quenched or nearly quenched localized magnetic moment. The unexpectedly large suppression of  $T_{e}$  in such alloys is demonstrated in Fig. 1. This is very puzzling, since magnetic measurements and the absence of a resistivity minimum in AlMn, AlCr, and VFe, for example, have shown that localized magnetic moments are absent or at least are very faint in these alloys.<sup>2</sup> Furthermore, the very puzzling and interesting observation is made that  $T_c$  decreases less rapidly for larger transition-metal impurity concentrations c. It is the purpose of this paper to explain this puzzling behavior of  $T_c$  resulting from localized spin excitations associated with the formation of localized magnetic moments.

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The anomalous magnetic behavior of the transitionmetal impurities with quenched or nearly quenched magnetic moment suggests strongly that the transition of the impurity from the nonmagnetic state to the magnetic state gives rise to localized spin fluctuations. These localized spin fluctuations arising from multiple electron-hole scattering at the impurities<sup>5</sup> and accompanying the formation of localized magnetic moments enhance the electron scattering by the transition-metal impurities. Assuming a static coupling between the electrons and the transition-metal impurities, for example, one neglects the very important electron spinflip scattering due to the localized spin fluctuations generated by the nonmagnetic transition-metal impurity near its paramagnetic state. The localized spin fluctuations give rise to a dynamic coupling between the electrons and transition-metal impurities that is somewhat similar to the interaction between electrons and spin fluctuations in nearly ferromagnetic transition metals.<sup>6</sup> Hence, the previous theoretical treatments of superconductors with transition-metal impurities that assume a static electron-impurity coupling<sup>7</sup> need to be generalized by taking into account the spin fluctuations associated with the formation of a localized magnetic moment. Note that the presence of localized spin fluctuations in metals containing transition-metal impurities with quenched or nearly quenched magnetic moments is strongly suggested by the electrical resistivity observed for Al-Mn<sup>8</sup>, which seems difficult to explain by a high Kondo temperature.<sup>8,9</sup>

One expects that with increasing concentration of magnetic impurities the increasing interatomic exhange and Coulomb coupling among the magnetic states of different impurities tend to suppress the spin fluctuations and the formation of the localized magnetic moments.<sup>10</sup> Consequently, the decrease of the superconducting transition temperature  $T_{c}$  is expected to "slow down" for larger concentrations of transition-metal impurities having quenched or nearly quenched magnetic moments.

#### **II. THEORY**

For a quantitative discussion of the effects on superconductivity of the localized spin excitations accompanying the formation of localized magnetic moments, the system of conduction electrons and electrons in the unfilled shells of the transition-metal impurities is described by the Hamiltonian

$$H = H_s + H_d + H_{ex} + H_{mix}.$$
 (1)

Here,  $H_S$  is the BCS Hamiltonian for the conduction electrons,  $H_{ex}$  describes the exchange interaction between the conduction electrons and the electrons in the unfilled shells of the transition-metal impurities, and  $H_{\rm mix}$  describes the electronic transitions between the conduction-electron states and the localized electronic states of the transition-metal impurities. Note that  $H_{\rm mix}$ , which describes the hybridization of the conduction-electron states and the localized electronic states of the transition-metal impurities, causes the quenching of the localized magnetic moments.<sup>4</sup> The electrons in the unfilled shells of the transition-metal impurities are described by

$$H_{d} \equiv \sum_{i,m,\sigma} E_{im\sigma} n_{im\sigma} + \sum_{im,jm'} U_{im,jm'} n_{im\sigma} n_{jm'-\sigma} + \sum_{im,jm'} J_{im,jm'} \boldsymbol{\sigma}_{im'} \cdot \boldsymbol{\sigma}_{jm'}. \quad (2)$$

Here,  $U_{im,jm'}$  and  $J_{im,jm'}$  denote the direct Coulomb and exchange integrals between two localized orbitals specified by *im* and jm'.<sup>10</sup>  $n_{im\sigma}$  and  $E_{im\sigma}$  are the occupation number operator and kinetic energy, respectively, for the state characterized by imo. The spin operator  $\sigma_{im}$ is defined as  $\sigma_{im} = \sum_{ss'} \alpha_{ims}^{\dagger} \sigma_{ss'} \alpha_{ims'}$ , where the components of the spin vector  $\sigma$  are the Pauli-spin matrices.  $\alpha_{ims}^{\dagger} = N^{-1/2} \sum_{q} e^{iq,r_i} a_{qms}^{\dagger}$  is a Wannier creation operator for the state ims. The Wannier annihilation operator  $\alpha_{ims}$  is similarly defined.  $J_{im,im'}$  gives Hund's-rule intra-atomic exchange interaction.<sup>10</sup> The interimpurity exchange coupling  $J_{im,jm'}$  can be split into  $J_{im,jm'}$  $=J_{im,jm'}^{RKY}+J_{im,jm'}^{dir.ex}$ , where

$$U_{im,jm'}^{\rm RKY} \equiv \frac{9\pi}{2} Z^2 \frac{J^2}{E_F} \frac{x \cos x - \sin x}{x^4},$$

 $x \equiv 2p_F r_{ij}$ , describes the Rudermann-Kittel-Yosida indirect-exchange coupling,<sup>11</sup> and where

$$J_{im,jm'}^{\mathrm{dir.ex.}} \sim \sum_{r_{ij} \to \infty} \left( e^{-\mathrm{const} \times r_{ij}} / r_{ij} \right)$$

gives the direct-exchange coupling<sup>11</sup> between the localized states *im* and *jm'*. Here  $r_{ij}$  denotes the distance between the impurities i and j,  $p_F$  is the Fermi wave number and  $E_F$  the Fermi energy, Z is the number of conduction electrons per atom, and J is the average exchange integral for the exchange interaction between the conduction electrons and the electrons in the unfilled shells of the transition-metal impurities. Considering now dilute alloys with randomly distributed transitionmetal impurities, and then applying the extended Abrikosov-Gor'kov theory<sup>1</sup> for dilute superconducting alloys with localized spin excitations,<sup>12</sup> one obtains for

<sup>&</sup>lt;sup>5</sup> H. Suhl, Phys. Rev. Letters 19, 442 (1967).

<sup>&</sup>lt;sup>6</sup> N. F. Berk and J. R. Schrieffer, Phys. Rev. Letters 17, 433 (1966).

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the superconducting transition temperature  $T_c$  the approximate expression<sup>1,6,12</sup>

$$\ln(T_c/T_{c0}) + \psi[\frac{1}{2} + (2\pi T_c \tau)^{-1}] - \psi(\frac{1}{2}) = 0, \quad (3)$$

where  $T_{c0}$  is the superconducting transition temperature in the absence of transition-metal impurities,  $\psi$  is the digamma function, and  $\tau^{-1} = \text{Im}\Sigma(p_F,\omega)$  is the conduction-electron relaxation time resulting from their interaction with the localized spin fluctuations associated with the transition-metal impurities.  $\Sigma(p_F,\omega)$  is the selfenergy of an electron with Fermi wave vector  $\mathbf{p}_F$  and energy  $\omega$  due to its interaction with the localized spin fluctuations, and it is obtained from<sup>6,12</sup>

$$\Sigma(p_F,\omega_n) = -cT \sum_{n'=-\infty}^{\infty} \int \frac{d^3p'}{(2\pi)^3} |J_{\text{eff}}(\mathbf{p}-\mathbf{p}')|^2 \\ \times G(p',\omega_{n'})E(\mathbf{p}'-\mathbf{p},\,\omega_{n'}-\omega_n)\,, \quad (4) \\ \omega_n = (2n+1)\pi T\,,$$

by performing the analytical continuation  $i\omega_n \rightarrow \omega$ . In Eq. (4) the electronic Green's function is denoted by G, E is the Green's function for the spin excitation, and  $J_{\text{eff}} = J + J'$  denotes the effective potential describing the coupling between the conduction electrons and the localized spin fluctuations.

$$J' \sim \left\langle -V_{kim} V_{kim}^* \frac{U_{im,im'}}{|E_{im}|(E_{im}+U_{im,im'})} \right\rangle$$

which is averaged with respect to i, m, m', results from

$$H_{\rm mix} \equiv \sum_{kim\sigma} \left( V_{kim} a_{k\sigma}^{\dagger} a_{im\sigma} + \text{H.c.} \right)$$

when put approximately into exchange form via a canonical transformation.<sup>13</sup> Now using the spectral representation for the Green's functions G and E, rewriting the sum over n' as a contour integral, performing the analytical continuation  $i\omega_n \rightarrow \omega$ , and writing

$$\frac{d^3 p'}{(2\pi)^3} \simeq \frac{N(0)}{2p_F^2} dq \ q dE_{p'}, \quad q \equiv |\mathbf{p} - \mathbf{p}'|$$

one obtains

$$\Sigma(p_{F},\omega) = \frac{3}{2} \frac{cN(0)}{4p_{F}^{2}} \int_{-\infty}^{\infty} \frac{dz}{2\pi} \int_{-\infty}^{\infty} dz'$$

$$\times \frac{\coth[(z'-\omega)/2T] - \tanh(z'/2T)}{z'-z-\omega-i\delta} \operatorname{Im} \frac{u(z')}{[1-u^{2}(z')]^{1/2}}$$

$$\times \int_{0}^{2p_{F}} dq \ q |J_{\text{eff}}(q)|^{2}B(q,z), \quad (5)$$

where N(0) is the conduction-electron normal-state

<sup>13</sup> J. R. Schrieffer, J. Appl. Phys. 38, 1143 (1967).

density of states at the Fermi surface, B(q,z) is the spectral function for the spin excitations, and  $u(\omega) \equiv \tilde{\omega}/\tilde{\Delta}$ . The quantities  $\tilde{\omega}$  and  $\tilde{\Delta}$  are the electronic energy and superconducting order parameter renormalized as a result of the coupling between electrons and spin excitations.<sup>1,12</sup> Note that the factor  $\frac{3}{2}$  in Eq. (5) results from taking into account the longitudinal and transverse spin excitations. Using that now for  $T \rightarrow T_c u \rightarrow \infty$ and  $\operatorname{Im}\{u(z)/[-u^2(z)]^{1/2}\} \to 1$ , and writing B(q,z)=2 $\text{Im}\chi(q,z)$ , where  $\chi(q,z)$  is the Fourier transform of the susceptibility, one finds from Eq. (5) for  $\tau^{-1}$  the approximate expression<sup>12</sup>

$$\tau^{-1} = \frac{3\pi c N(0)}{4p_F^2} \int_0^{2p_F} dq \; q \left| J_{\text{eff}}(q) \right|^2 \\ \times \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \operatorname{Im} \chi(q,\omega) \left( \operatorname{coth} \frac{\omega}{2T} - \operatorname{tanh} \frac{\omega}{2T} \right). \tag{6}$$

Note, that for  $T \rightarrow 0$  the main contribution to the integral in Eq. (6) results for  $\omega \ll T$ . Using the randomphase approximation, one finds for  $\chi(q,\omega)$  the approximate expression<sup>10,12</sup>

$$\chi(q,\omega) = \chi_0(q,\omega) [1 - (U + J_{\text{ex}})\chi_0(q,\omega)]^{-1}, \qquad (7)$$

where  $\chi_0(q,\omega)$  denotes the Pauli susceptibility of the magnetic impurity states. The average effective intraatomic Coulomb interaction between the electrons and holes in the unfilled shells of the transition-metal impurities is given by U.  $J_{ex}$  describes the average exchange interaction among different impurities. It is obvious from Eq. (7) that for  $J_{ex}0$ , as is expected,<sup>10,11</sup> the interatomic exchange coupling tends to reduce the spin fluctuations and hence  $\tau^{-1}$ . Also, it is clear from Eq. (6) that the electron scattering is enhanced due to the spin fluctuations. Assuming orbital degeneracy, one finds approximately within the Hartree-Fock approximation for  $\text{Im}\chi(q,\omega)$  the expression

$$\operatorname{Im} \chi(q,\omega) \cong \operatorname{Im} \chi_0(q,\omega) \{ [1 - (U + J_{ex})\chi_0(q,0)]^2 + (U + J_{ex})^2 [\operatorname{Im} \chi_0(q,\omega)]^2 \}^{-1}, \quad (8)$$
with
$$\chi_0(q,0) \approx N_d(0) \approx \sum (1/\pi\Delta). \quad (9)$$

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Here  $N_d($ face contributed by the unfilled impurity shells. m sums over all magnetic states with average width  $\Delta$ . One finds in the case of d states that  $\sum_{m} = 5$ . Performing an ensemble average over all possible impurity positions yields, for very dilute alloys for which the directexchange coupling between different transition-metal impurities can be neglected, for  $J_{ex}$  the expression

$$J_{\text{ex}} \simeq c \frac{9\pi}{2} Z^2 \frac{J_{\text{eff}}^2}{E_F} \sum_{i \neq i_0}^{\infty} \frac{x \cos x - \sin x}{x^4} \approx -cN(0) J_{\text{eff}}^2,$$
$$x \equiv 2p_F r_{ii_0},$$

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where *i* sums over all lattice sites and *c* denotes the concentration of transition-metal impurities. Note that Eq. (7) is expected to hold if  $(U+J_{ex})N_d(0)$  is not close to 1, e.g., for the case of fast spin fluctuations. However, for the case  $(U+J_{ex})N_d(0) \sim 1$ ,  $\chi(q,\omega)$  or  $\chi_0(q,\omega)$  need to be determined self-consistently<sup>5,14</sup> by taking into account the effect of the spin fluctuations on  $\chi$ , in order to obtain in accordance with experiment a smoothly varying susceptibility near the formation of the localized magnetic moment. Close to the formation of a localized magnetic moment one obtains approximately<sup>5</sup> for the static susceptibility the expression

where

$$\tau_{1s} \equiv \pi N_d(0) [1 - (U + J_{ex}) N_d(0)]^{-1}$$

 $\chi \simeq \operatorname{const} \times (4k_BT + \pi \tau_{1s}^{-1})^{-1},$ 

gives the average lifetime of the localized spin excitations.

Since a self-consistent determination of X, using renormalized Green's functions for the particles in the localized magnetic states, is very difficult,<sup>5</sup> it is of interest to point out that the effect of the spin fluctuations associated with the formation of the localized magnetic moments on the electrical resistivity is given by<sup>8</sup>

$$\rho(T) = \rho(0) \left[ 1 - \frac{1}{3} \pi^2 (\frac{1}{5} \pi)^2 (k_B T \chi)^2 + \cdots \right] \sim (1/\tau_{\rm tr}). \quad (10)$$

Here, the transport collision time  $\tau_{tr}$  is obtained from Eq. (4) by replacing the factor q in the integrand by  $q^3/2p_F^2$ . Similarly, the coefficient  $\gamma$  of the electronic specific-heat contribution, which is linear in temperature, is in the presence of spin fluctuations given by<sup>6</sup>

$$\gamma = \gamma_0 \bigg[ 1 + N(0) V_{\rm ph} + \frac{3cN(0)}{4p_F^2} \int_0^{2p_F} dq \; q \, |J_{eff}(q)|^2 \\ \times \frac{N_d(0)}{1 - (U + J_{\rm ex})N_d(0)} \bigg], \quad (11)$$

where  $\gamma_0$  is the value of  $\gamma$  in the absence of spin fluctuations and phonon coupling and  $N(0)V_{\rm ph}$  results from the electron-phonon interaction. Consequently, one may determine  $\tau$  from  $\gamma$ ,  $\chi(0,0)$ , or  $\rho$ .

It is clear that the localized spin excitations cease if their lifetime becomes larger than the lifetime of an electron in the unfilled shells of the impurities that is of the order of  $\Delta^{-1}$ . Thus, spin-orbit scattering, for example, can significantly damp the localized spin excitations. Also, the Hund's-rule intra-atomic exchange coupling strongly affects the localized spin excitations. It would be interesting to verify experimentally the importance of these effects by using systematically different transition-metal impurities.

Now using Eq. (3), one can calculate the superconducting transition temperature  $T_c$  of various dilute alloys containing transition-metal impurities. Note



FIG. 1. Initial slope of the superconductiong transition temperature as a function of concentration for various transitionmetal impurities.

that for the very dilute alloys the interatomic exchange coupling is essentially given by the indirect interaction mediated by the conduction electrons. The coupling between the impurities is assumed to be antiferromagnetic.  $\tau$  is determined by fitting experimental results for small c. Note that one finds for  $T \rightarrow 0$  from Eq. (6)



FIG. 2. Suppression of the superconducting transition temperature  $T_e$  for VFe, AlCr, and AlMn arising from fast localized spin fluctuations associated with the formation of localized magnetic moments.  $T_e$  is determined by Eq. (3).  $\tau c T_e^2$  is determined by fitting experimental results for small transition-metal impurity concentrations c. The dashed curves result for  $J_{ex}=0$ , assuming a temperature-independent  $\tau$ .

<sup>&</sup>lt;sup>14</sup> M. J. Levine, T. V. Ramakrishnan, and R. A. Weiner, Phys. Rev. Letters **20**, 1370 (1968).

TABLE I. Numerical results for  $N(0)J_{eff}$  and  $N(0)V_{spin} \equiv [3c\tau_{1s}/2\pi N(0)][N(0)J_{eff}]^2$  calculated by determining  $\tau_{1s}$  (column 5) from the susceptibility  $\chi^8$  and then  $N(0)J_{eff}$  from  $T_e$ . Since  $\chi$  is not very accurately known,  $N(0)J_{eff}$  is also calculated by using the values given in column 8 for  $\tau_{1s}$ . The results indicate that  $N(0)J_{eff}$  depends sensitively on  $\tau_{1s}$ . N(0) is approximately given by 0.2 (eV)<sup>-1</sup> for AlMn and AlCr, and by 1.3 (eV)<sup>-1</sup> for VFe. It is  $\gamma' \equiv \gamma_0 (1+N(0)V_{ph})$  equal to  $1.35[mJ/mole(^{\circ}K)^2]$  for AlMn and AlCr and equal to 9.90  $\times [mJ/mole(^{\circ}K)^2]$  for VFe.

Alloy	c = 0.05  at. % $(\tau c T_c^2)^{-1} (^{\circ}\text{K})^{-1}$	c = 0.1  at. % $(\tau c T_c^2)^{-1} (^{\circ}\text{K})^{-1}$	$c=0.2 \text{ at.}\% (\tau c T_{c}^{2})^{-1} (^{\circ}\text{K})^{-1}$	$\begin{array}{l}(\mathrm{From}\;\chi)\\\tau_{\mathrm{ls}}^{-1}(\mathrm{eV})\end{array}$	(From $\chi$ and $T_c$ ) $N(0)J_{eff}$	c = 0.1  at. % $N(0) V_{spin}$	${ au_{1s}}^{-1}({ m eV})$	(From $\tau_{\rm ls}$ and $T_c$ ) $N(0)J_{\rm eff}$	$c = 0.1 \text{ at.} \% N(0) V_{\text{spin}}$
<i>Al</i> Mn	1.75 · 10 <sup>3</sup>	1.84 · 103	1.86 · 103	0.09	10	0.3	0.18	5	0.07
<i>Al</i> Cr	2.09 · 10 <sup>3</sup>	2.11 · 10 <sup>3</sup>	2.15 · 10 <sup>3</sup>	0.15	8	0.2	0.30	4	0.05
VFe	No experimental results			0.65	1.7	0.0003	1.3	0.8	6.10-5

for  $\tau$  the approximate expressions

$$\tau^{-1} \simeq (\pi^3/4Z) c [N(0)J_{\text{eff}}]^2 \chi^2(0,0) N_d^{-1}(0) T^2, \quad (12)$$
$$U N_d(0) < 1$$

and

$$\tau^{-1} \underline{\sim}_{2}^{3} c \pi N(0) J_{\text{eff}}^{2} \chi(0,0) [UN_{d}(0)]^{-1} T, \qquad (13)$$
$$UN_{d}(0) \approx 1.$$

Furthermore, in the limit of  $c \to 0$ , one obtains from Eq. (3)  $T_{c0} - T_c \simeq \pi/4\tau$ , and for  $T_c \ll T_{c0}$  one obtains

$$T_{c^{2}} \simeq (6/\pi^{2}\tau^{2}) \ln(\pi T_{c0}\tau/3.56).$$

(See Fig. 2.)

### III. DISCUSSION

According to Eqs. (12) and (13), the electron scattering by the localized spin fluctuations decreased with decreasing temperature. This explains the anomalously slow decrease of  $T_c$  observed at higher impurity concentrations. As shown in Table I, determining  $\tau$  by fitting  $T_c$  at small concentrations yields indeed  $c\tau T_c^2$ = const, as predicted by Eq. (12) for small  $J_{\text{ex}}$ . Also, it follows from Eq. (12) that knowing  $\chi$ , one can determine from  $T_c$  the quantity  $N(0)J_{\text{eff}}$  and then calculate the enhancement of the specific-heat contribution  $\gamma T$  arising from the localized spin fluctuations. Note that  $N(0)V_{\rm ph}$  can be calculated from  $T_{c0}$ . The results obtained are given in Table I.

Since the temperature dependence of  $\tau^{-1}$  reflects sensitively the presence of localized spin fluctuations, it would be very desirable to measure carefully the superconducting transition temperature over a wider concentration range of transition-metal impurities, where  $T_c$  changes nonlinearly with c. Note that for AlFe, ZnNi, and InMn, for example,<sup>2</sup> one observes also that the decrease of  $T_c$  slows down for higher concentrations of transition-metal impurities in a similar fashion, as for AlMn.

Unfortunately, so far no direct determination of  $N(0)J_{\rm eff}^2$  is possible. However, from comparison with previous analyses, we expect for U the assumed values and for  $N(0)J_{eff}$  values of the order of those given in column 9 of Table I. Unfortunately, no sufficiently accurate experimental results are available for  $N(0)V_{\text{spin}}$ . Obviously, a very accurate experimental determination of the enhancement of the specific-heat coefficient  $\gamma$  arising from spin fluctuations would be very useful, since according to Eqs. (10) and (11) it permits one to calculate  $N(0)J_{eff^2}$  by using the experimental results from the electrical resistivity. Note that for AlMn and AlCr the values obtained<sup>8</sup> for  $\tau_{es}^{-1}$  from the electrical resistivity are 0.08 and 0.2 eV, respectively. For VFe no sufficiently accurate experimental resistivity results are available.