

Effect of Kondo impurities on the Korringa relaxation time

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Using Nozières Fermi-liquid theory, a simple result, valid for temperatures small compared to the Kondo temperature, is obtained which describes the effect of magnetic impurities on the nuclear-spin-relaxation time T_1 of a metal in the limit of small magnetic fields. When extended to the case of finite magnetic fields, using a semiphenomenological generalization of ground-state energy results of Ishii, the theory shows quite reasonable agreement with the available experimental data. Further experiments are suggested.

Recent experiments by Edwards *et al.*¹ and Avenel *et al.*¹ show that at low temperatures T the Korringa relation $T_1 T = \text{const}$ for the nuclear-spin-relaxation time T_1 in a Pt sample containing extremely small amounts of magnetic impurities,² breaks down. Since this relation is used to determine the temperature in ultralow temperature experiments, it is of crucial practical importance to understand the mechanism causing the breakdown.

These experiments on Pt have the same gross features as those observed earlier by Gladstone³ in Cu containing Cr as an impurity. For $T \gg T_K$, $T_1 T$ is the Korringa constant for the pure host, while for $T \ll T_K$, $T_1 T = C = \text{another constant}$. The constant C is substantially smaller than the Korringa constant. The size of the deviation from the Korringa constant is a decreasing function of the applied magnetic field for field strengths such that $\mu_B H \sim K_B T_K$, where T_K is the appropriate Kondo temperature. For $\mu_B H \gg k_B T_K$ the deviation disappears. These observations strongly suggest that the formation of the Kondo state⁴ might be responsible for such a deviation. In this paper we demonstrate that the Kondo effect does account for the observed deviation and its field dependence. The theory is restricted to temperatures $T \ll T_K$.

We begin with a specification of a model which can be solved. This model assumes an impurity spin of one-half and negligible applied magnetic field. These assumptions will be removed in a semiphenomenological generalization to be presented later. The impurity concentration x is assumed small enough to permit the neglect of interactions between impurities; the effect on T_1 is, therefore, linear in x . The host-metal conduction band is assumed to contain only s electrons. The host nuclei will possess spin \bar{S} , and the coupling of \bar{S} with the electrons is treated as a perturbation. The Kondo coupling of the electronic system with the impurities is dealt with in a nonper-

turbative fashion. For small x we may restrict ourselves to the case of a single impurity located at the origin of our coordinate system.

For the problem of nuclear-spin relaxation we are interested in transitions in which the z component of the host nuclear spin $\bar{\tau}_n$ changes from M to $M \pm 1$. The couplings involving $\bar{\tau}_n$ are the host nucleus-conduction electron spin interaction and the host nucleus-impurity dipole-dipole interaction. We neglect the latter for the dilute alloys considered. Further, in the systems discussed here, the host nucleus-electron spin coupling is thought⁵ to be dominated by the contact interaction,⁶ which takes the usual form

$$H_{\text{int}} = -\frac{A}{N} [\psi_1^\dagger(\bar{\mathbf{R}}_n) \psi_1(\bar{\mathbf{R}}_n) \tau_{n-} + \text{H.c.}] \quad (1)$$

Here $\bar{\mathbf{R}}_n$ is the position of the host nucleus, τ_{n-} is the spin-lowering operator on the nucleus, and $\psi_1^\dagger(\bar{\mathbf{R}}_n)$ is a field operator creating a spin-up electron at $\bar{\mathbf{R}}_n$. A is the coupling constant and N the number of nuclei. Using Eq. (1) and standard second-order perturbation theory, the total rate for spin-flip processes becomes⁷ (for an impurity with spin $\frac{1}{2}$)

$$\frac{1}{T_1} = \frac{2A^2}{\hbar^2 N^2} \int_{-\infty}^{\infty} dt \langle S_-(\bar{\mathbf{R}}_n, t) S_+(\bar{\mathbf{R}}_n, 0) \rangle \quad (2)$$

Here

$$S_-(\bar{\mathbf{R}}_n, t) = \psi_1^\dagger(\bar{\mathbf{R}}_n, t) \psi_1(\bar{\mathbf{R}}_n, t) = S_+(\bar{\mathbf{R}}_n, t)$$

is the electron-spin-lowering operator at position $\bar{\mathbf{R}}_n$ and time t . We remark that Eq. (2) is exact to all orders in the impurity-electron coupling J if the expectation value is calculated to all orders in that coupling. However, if Eq. (2) is evaluated in the random-phase approximation (RPA) for the impurity-electron problem, $1/T_1$ becomes the sum of three terms,⁸ the standard Korringa rate in the ab-

sence of the impurity, the correction to that rate known as the GH (Giovannini and Heeger) rate, and the BGS (Benoit, de Gennes, and Silhouette) rate. For both the GH and BGS mechanisms the host nucleus and impurity are coupled via a Rudderman-Kittel-Kasuya-Yosida (RKKY) coupling. The essential point of the present calculation is to show that for temperatures $T \ll T_K$ the right-hand side of Eq. (2) can be calculated to all orders in J using known features of the Kondo ground state and the low-energy excitations above that state.

To proceed we utilize the Nozières⁹ quasiparticle picture of the low-energy excitations. In this picture the quasiparticle states are those associated with the resonant scattering of an electron from a *spin-independent* potential localized at the impurity, the impurity having had its spin screened by the binding of an opposite spin electron closely to it. It seems quite plausible that the freezing-out of the impurity spin eliminates the RKKY coupling. We assume this to be the case. Following Nozières⁹ we use *s*-wave scattering states for the quasiparticle states. These states have wave functions of the form

$$\phi_{\vec{k}}(\vec{\tau}) = e^{i\vec{k}\cdot\vec{\tau}} + \frac{e^{i\vec{k}r}}{2ikr} (e^{2i\delta_k} - 1), \quad (3)$$

where the impurity-bound electron complex is located at $\vec{\tau} = 0$. On the Fermi surface the phase shift $\delta_k = \frac{1}{2}\pi$.⁹ The field operator $\psi_{\sigma}(\vec{\tau})$ is expanded in terms of these states as

$$\psi_{\sigma}(\vec{\tau}) = \sum_{\vec{k}} \phi_{\vec{k}}(\vec{\tau}) \alpha_{\vec{k}\sigma}, \quad (4)$$

where $\alpha_{\vec{k}\sigma}$ is an operator which destroys a quasiparticle having wave vector \vec{k} and spin σ . Straightforward evaluation of the right-hand side of Eq. (2) yields

$$\frac{1}{T_1} = \frac{4\pi}{\hbar} \left(\frac{A}{N} \right)^2 \sum_{\vec{k}, \vec{k}'} |\phi_{\vec{k}}^*(\vec{R}_n) \phi_{\vec{k}'}(\vec{R}_n)|^2 \times n_k (1 - n_{k'}) \delta(\epsilon_{\vec{k}} - \epsilon_{\vec{k}'}), \quad (5)$$

where n_k is the Fermi-Dirac distribution for particles of energy $\epsilon_{\vec{k}}$. This result may be rewritten in the form

$$\frac{1}{T_1 T} = \frac{4\pi k_B}{\hbar} A^2 \rho^2(\vec{R}_n, \epsilon_F) \quad (6)$$

with a position-dependent density of states being defined by

$$\rho(\vec{R}_n, \epsilon_F) = \frac{1}{N} \sum_{\vec{k}} |\phi_{\vec{k}}(\vec{R}_n)|^2 \delta(\epsilon_{\vec{k}} - \epsilon_F). \quad (7)$$

Here ϵ_F is the Fermi energy. With the simple form of the wave function given by Eq. (3), Eq. (7)

reduces to

$$\rho(\epsilon_F, \vec{R}_n) = \rho(\epsilon_F) \left[1 + \frac{2 \cos 2k_F R_n}{(k_F R_n)^2} \right], \quad (8)$$

with

$$\rho(\epsilon_F) = \frac{1}{N} \sum_{\vec{k}} \delta(\epsilon_F - \epsilon_{\vec{k}}).$$

Since $k_F \sim 1 \text{ \AA}^{-1}$ and for the largest impurity concentration ($x = 30 \times 10^{-6}$) we use, $R_N^{-1} \sim 3 \times 10^{-2} \text{ \AA}^{-1}$ for an average nucleus. Thus even in the worst case, $(k_F R_N)^{-1} \sim 3 \times 10^{-2}$ and $\rho(\epsilon_F, R_N)$ can safely be replaced by $\rho(\epsilon_F)$. From Nozières,⁹

$$\rho(\epsilon_F) = \rho_0(\epsilon_F) [1 + (4/3\pi) \alpha (T_F/T_K) 1/N],$$

where T_F is the Fermi temperature, $\rho_0(\epsilon_F)$ is the density of states in the absence of impurities and $\alpha = 0.324$. For small impurity concentration $1/N$ is replaced by x . It should be noted that the Nozières theory and in particular, the result for the change in the density of states $\delta\rho$ assumes the presence of a single impurity. Therefore, it will be inconsistent to keep any term which is of higher than first order in $1/N$ (or x) in $\rho^2(\epsilon_F)$. No theory of the Wilson¹⁰ or Nozières⁹ type is at present capable of obtaining higher orders in x . Furthermore, though in the case of CuCr the impurity concentration used is such that $\delta\rho/\rho_0$ is of order unity and the linear approximation should begin to break down, the experimental data of Ref. 3 is indicated to be linear in x . As we demonstrate later the linear correction term does account quite well for the observed change in T_1^{-1} . In addition, Nozières⁹ theory predicts a similar change (proportional to $\delta\rho/\rho_0$) for specific heat and susceptibility and therefore the theory should also begin to break down for these quantities in the concentration range considered. However, experiments seem to indicate that in this concentration range the change in specific heat^{11,12} and susceptibility¹¹ is still linear. In the other case of interest, namely that of PtMn, again the impurity concentration is such that the linear approximation should begin to break down. However, the resistivity data¹³ indicates that we are still in the linear concentration regime. Moreover, as shown later the linear correction does explain the observed change in T_1^{-1} . Given this discussion, we keep only the first order term in x . We believe there is a need for more careful experimental studies regarding the concentration dependence of various physical quantities and thus to set precise limits for the use of present theory. Collecting our results we find

$$[T_1(x)T]^{-1} = [T_1(0)T]^{-1} \left[1 + \frac{8x}{3\pi} \alpha \frac{T_F}{T_K} \right], \quad (9)$$

where $[T_1(0)T]^{-1}$ is the standard Korringa rate for the pure metal.

Equation (9) is our central result, expected to be valid for sufficiently small x and $T \ll T_K$. For $T \geq T_K$ the quasiparticle picture breaks down, the impurity spin degrees of freedom become unlocked, and the GH and BGS mechanisms become free to operate. Of these two mechanisms the BGS is known to be the most important.¹⁴ Near but above T_K the BGS mechanism can be comparable in its effects to those of the pure Korringa mechanism.¹⁵ However, for $T \gg T_K$, the Korringa mechanism becomes dominant since the BGS rate contains an additional inverse power of $(T/T_F)^2$ compared to the Korringa rate. Given this discussion, we may replace Eq. (9) by

$$\frac{[T_1(x)T]_{T \ll T_K}}{[T_1(x)T]_{T \gg T_K}} = \left(1 + \frac{8x}{3\pi} \alpha \frac{T_F}{T_K}\right)^{-1} \quad (10)$$

From Eq. (10), it is clear that a substantial reduction in $T_1 T$ can occur as T is lowered through T_K even for very small x if T_K is small. However, the above result cannot be directly compared with the experiments on CuCr and PtMn, which are in finite magnetic field. Therefore, our next step is to generalize Eq. (10) for the case where $H \neq 0$.

At present the Kondo Hamiltonian has not been solved for finite magnetic fields, but there is an approximate ground-state calculation by Ishii,¹⁶ which gives results even for field energies $\mu_B H$ large compared to the zero-field binding energy $\tilde{E}_0 = k_B T_K$. The obtained field dependence of the binding energy \tilde{E} is given by

$$\tilde{E}^2 = \tilde{E}_0^2 + \Delta^2, \quad (11)$$

where $\Delta = \frac{1}{2} g \mu_B H$. This result does not include the effect of the magnetic field on the conduction electrons, which gives a negligible contribution ($\sim \mu_B H / E_F$).

To use the above result we note that the impurity correction in Eqs. (9) and (10) is due to a change $\delta\rho$ in the density of states which varies⁹ as $(k_B T_K)^{-1}$ and hence as \tilde{E}_0^{-1} . Nozières⁹ has calculated the impurity contribution to the susceptibility and this contribution is also proportional to $\delta\rho$. Thus if we can calculate the susceptibility in the presence of finite magnetic field, we can deduce the change in the density of states $\delta\rho(H)$ for the case of finite H . To do that we differentiate \tilde{E} twice with respect to H and find that the impurity contribution to the susceptibility, and hence the change in the density of states $\delta\rho(H)$, varies as $\tilde{E}_0 [1 + (\Delta/\tilde{E}_0)^2]^{-3/2}$. Thus, Eq. (11) leads

to

$$\frac{[T_1(x,H)T]_{T \ll T_K}}{[T_1(x)T]_{T \gg T_K}} = \left\{1 + \frac{3}{2} \frac{T_F x}{T_K} \left[1 + \left(\frac{\frac{1}{2} g \mu_B H}{k_B T_K}\right)^2\right]^{-3/2}\right\}^{-1} \quad (12)$$

Now, if we compare the impurity correction in the limit $H \rightarrow 0$ as given by Eq. (12) with that of our central result, Eq. (10), we find that they differ by a factor of $4\alpha/\pi$. Thus, generalization of Eq. (12) to the correct $H=0$ behavior involves the introduction of a single parameter α . This parameter relates the ground-state properties and the high-temperature limit ($T \gg T_K$) of the Kondo problem. In the absence of an exact theory of the ground state for $H \neq 0$, we will use the analog of the introduction of α to parameterize the field dependence of the density of states. Using Eq. (12) as a guide we write

$$\frac{[T_1(x,H)T]_{T \ll T_K}}{[T_1 T]_{T \gg T_K}} = \left\{1 + \frac{8}{3\pi} \alpha \frac{T_F x}{T_K} \times \left[1 + \left(\frac{\beta \frac{1}{2} g \mu_B H}{k_B T_K} \frac{4}{\pi}\right)^2\right]^{-3/2}\right\}^{-1} \quad (13)$$

in which β parameterizes the field dependence. Note that in Ishii's theory $\alpha = \beta = \frac{1}{4} \pi$. Since there is no reason to expect that $\alpha = \beta$ for the exact result (they describe different physical effects), we allow α to differ from β .

Now we turn to the comparison of the theory with experiment. It is always difficult to make a strict comparison⁴ of the theory of the spin- $\frac{1}{2}$ Kondo problem with experiments because in most systems the impurity spin is not one-half. One must provide a phenomenological generalization⁴ of the spin- $\frac{1}{2}$ results. We use the following prescription; insert a factor of $2S$ in front of the impurity correction in Eqs. (9), (10), and (13), as one needs $2S$ electrons instead of one to screen the impurity spin S . Replace $\frac{1}{2} g \mu_B$ in Eq. (13) by $g \mu_B S$. Also, one should allow for possibility $\alpha = \alpha(S) \neq 0.324$. Thus, Eq. (13) for spin S takes the form

$$\frac{[T_1(x,H)]_{T \ll T_K}}{[T_1(x)T]_{T \gg T_K}} = \left\{1 + \frac{8x}{3\pi} 2S \frac{\alpha T_F}{T_K} \times \left[1 + \left(\beta \frac{S g \mu_B H}{k_B T_K} \frac{4}{\pi}\right)^2\right]^{-3/2}\right\}^{-1} \quad (14)$$

First we discuss the low temperature data of Gladstone³ in CuCr. The transition in $(T_1)^{-1}$ measurements starts at about 0.05 K and extends at least up to 0.15 K. In fact, some of the experiments¹⁷ indicate that the transition continues up to about 4 K. For $T < 0.05$ K, T_1T is a constant which is less than the Korringa constant of ~ 1.28 sec K. The excess rate $\Delta(T_1T)^{-1} = (T_1T)^{-1} - (1.28 \text{ sec K})^{-1}$ for $T < 0.05$ K varies linearly with Cr concentration of up to 30×10^{-6} , which is consistent with resistivity data.¹⁸ Measurements indicate that the spin of Cr is $\frac{3}{2}$.¹⁸ The first thing to note is that the theory correctly predicts the temperature dependence, $T_1T = \text{const}$ for $T \ll T_K$. Figures 1 and 2 show the comparison of our theory, Eq. (14) with experiments for $T \ll T_K$ as regards the field dependence. We have used $T_F = 8.16 \times 10^4$ K (Ref. 19) and the measured value of $x = 30 \times 10^{-6}$.³ The best fit has been obtained by choosing $\alpha T_K^{-1} = 0.65 \text{ K}^{-1}$ and $\beta T_K^{-1} = 0.65 \text{ K}^{-1}$. Given that $T_K \sim 1$ K for CuCr, this implies that $\alpha = 0.65$, which does not appear unreasonable when compared to the value $\alpha = 0.324$ for the spin- $\frac{1}{2}$ model for $H = 0$. We see that agreement between the theory and the experiments is quite reasonable. Shown is the above-mentioned best fit along with curves illustrating the dependence on variations in the parameters α/T_K and β/T_K .

We now discuss the data of Ref. 1 on Pt, which contains, among other impurities,² Mn in a concentration of 1.4×10^{-6} and Fe in a concentration of 44×10^{-6} . There are some indications²⁰ that T_K for PtFe is ~ 0.4 K and with the above concentration Fe impurities could possibly give a sizable contribution. However, from a comparison with our interpretation

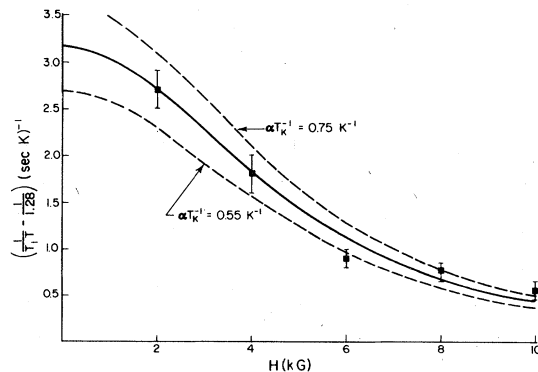


FIG. 1. Reduced Cu relaxation rate $[(T_1T)^{-1} - (T_1T)_{\text{Cu}}^{-1}]$ in CuCr, with a Cr concentration of 30×10^{-6} , as a function of external magnetic field. The low-temperature ($T < 0.05$ K) data of Gladstone are represented by solid squares. The solid line represents the best fit for which $\alpha T_K^{-1} = \beta T_K^{-1} = 0.65 \text{ K}^{-1}$. The dotted lines show the variations of the fit when one varies αT_K^{-1} .

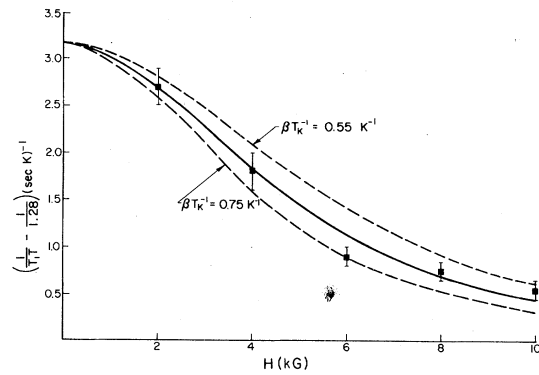


FIG. 2. Reduced Cu relaxation rate $[(T_1T)^{-1} - (T_1T)_{\text{Cu}}^{-1}]$ in CuCr, with a Cr concentration of 30×10^{-6} , as a function of external magnetic field. The low-temperature ($T < 0.05$ K) data of Gladstone are represented by solid squares. The solid line represents the best fit for which $\alpha T_K^{-1} = \beta T_K^{-1} = 0.65 \text{ K}^{-1}$. The dotted lines show the variations of the fit when one varies βT_K^{-1} .

of the CuCr data, which is better characterized, one would then expect to see a transition around $T = 0.4$ K. There is no evidence of such a transition for the Pt samples studied. Further with $T_K = 0.4$ K our theory cannot produce the field dependence of the data of Ref. 1. Given that the theory works well for the better-characterized CuCr system we conclude that it is highly probable that Fe impurities make no contribution to the excess rate. Perhaps these impurities were not uniformly distributed throughout the sample, which was not prepared with any object other than eliminating impurities.

This leaves us with Mn impurities as a possible cause for the observed deviation. The transition is centered around $T = 10$ mK, which is consistent with recent measurements¹³ of $T_K \approx 10$ mK for PtMn system. Experiment²¹ gives a Korringa constant for pure Pt of $T_1T = 0.03$ sec K for $T \geq 0.07$ K. The Pt data of Ref. 1 saturate at this value near $T = 0.02$ K, indicating that the BGS rate, due to magnetic impurities, is unimportant at and above $T = 0.02$ K for the samples studied. Again for $T \ll T_K$, T_1T settles down to a constant value, which agrees with the temperature dependence of our theory. The impurity spin is $S = 2$.¹³ To estimate T_F we remark that T_F appeared in Eq. (14) via the use of simple free-fermion relation $k_B T_F = 3N/4\rho_0$, where ρ_0/N is the density of states at ϵ_F per spin. In Pt the quantity most closely corresponding to ρ_0/N is the s -wave density of states, which has been calculated by Ketterson *et al.*²² Using their value we find $T_F = 2.12 \times 10^4$ K. Figures 3 and 4 compare the theory with the data of Ref. 1. The best fit has been obtained by choosing $\alpha T_K^{-1} = 20 \text{ K}^{-1}$ and $\beta/T_K = 40 \text{ K}^{-1}$. Given a value $T_K \approx 10$ mK, we find $\alpha = 0.2$, which seems reasonable. Note that

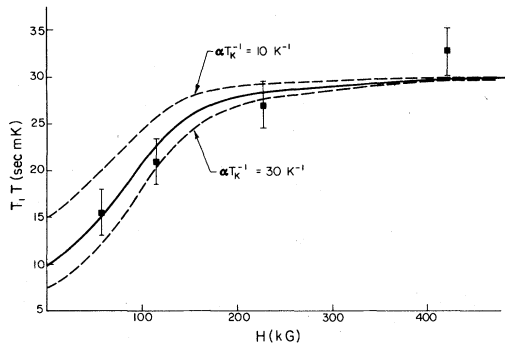


FIG. 3. T_1T of Pt as a function of external magnetic field. The low-temperature ($T \ll 10$ mK) data of Ref. 1 are represented by solid squares. The error bars represent an estimated error. The solid line gives the best theoretical fit for which $\alpha T_K^{-1} = 20$ K $^{-1}$ and $\beta T_K^{-1} = 40$ K $^{-1}$. Dotted lines show the variation about this curve when one varies αT_K^{-1} .

while $\beta = 2\alpha$ here, for the case of CuCr β was equal to α . In this case we have plotted T_1T as a function of H , rather than the excess rate, for the convenience of researchers engaged in low-temperature thermometry. Again the agreement between the theory and the experiments is quite reasonable. The high-field limit of the data of Ref. 1 does not seem to quite approach the value $T_1T = 0.03$ sec K of the pure Pt. The origin of this small discrepancy is not known.

The only other system which has been studied through its Kondo temperature (~ 29 K) is CuFe.²³ However, the clustering of Fe impurities, as indicated by magnetization measurements,²⁴ makes it a very difficult system in which to isolate and observe the one-impurity effects. We conclude that all the clear-cut available data are consistent with the predictions of our theory. However, direct experimental tests of the zero-field result, Eq. (10), which is on more sound theoretical grounds than the field-dependent result, Eq. (14) would be of great interest. This would require measuring T_1^{-1} in fields such that $\mu_B H \ll k_B T_K$. Further, more detailed experimental

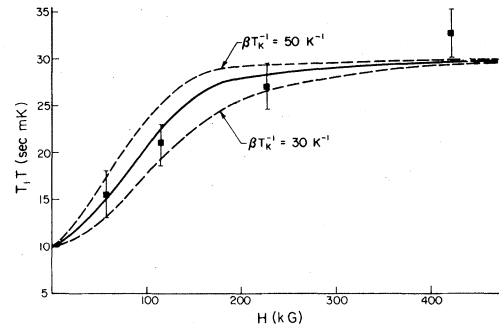


FIG. 4. T_1T of Pt as a function of external magnetic field. The low-temperature ($T \ll 10$ mK) data of Ref. 1 are represented by solid squares. The error bars represent an estimated error. The solid line gives the best theoretical fit for which $\alpha T_K^{-1} = 20$ K $^{-1}$ and $\beta T_K^{-1} = 40$ K $^{-1}$. Dotted lines show the variation about this curve when one varies βT_K^{-1} .

studies of T_1^{-1} in dilute alloys for $\mu_B H \sim k_B T_K$ could furnish valuable guides to the theorist attempting to obtain more precise descriptions of the magnetic field dependence of the nuclear-spin-relaxation rates as well as that of the ground-state energy in these systems. Lastly, we point out once again that the concentration of impurities used in both cases studied are such that our theory should begin to break down. Therefore, we strongly suggest more careful experimental studies of T_1^{-1} in these systems in order to carefully determine the concentration dependence and thus the precise limits of the present theory.

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²E. Varogaux, private communication relayed to us by D. O. Edwards. The Pt sample of both groups mentioned in Ref. 1 came from the same source and contained the same impurities and impurity concentrations; Mn(1.4 ppm), Fe(44 ppm), Ni(6.1 ppm), Co(0.15 ppm), and Cr(4.7 ppm). The Kondo temperatures of Ni, Co, and Cr in Pt are too high to be of importance here since our important result, Eq. (10), contains a factor T_K^{-1} .

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