Muon spin relaxation in a random ferromagnet: *Pd*Mn

S. A. Dodds and G. A. Gist
Department of Physics, Rice University, Houston, Texas 77251

D. E. MacLaughlin
Physics Department, University of California, Riverside, California 92521

R. H. Heffner, M. Leon, and M. E. Schillaci
Los Alamos National Laboratory, Los Alamos, New Mexico 87545

G. J. Nieuwenhuys and J. A. Mydosh
Kamerlingh Onnes Laboratory, University of Leiden, 2300-RA Leiden, The Netherlands
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Transverse and longitudinal muon spin-relaxation experiments have been carried out in the ferromagnetic and paramagnetic states of a Pd—2.0 at.% Mn alloy to determine effects of spin-configuration disorder on the distribution of local (dipolar) fields at positive muon (μ^+) sites, and on dynamic-relaxation processes. The giant-moment polarization cloud appears to make no contribution to the local-field distribution in the paramagnetic state ($T > T_c = 5.8$ K). The Walstedt-Walker formula for the μ^+ linewidth is in good agreement with the data. In the ferromagnetic phase the distribution width is less than predicted by a mean-field model of a uniform spin system. This "shortfall" is accounted for qualitatively by the Sherrington-Kirkpatrick (replica-symmetric) theory for disordered ferromagnets. Spin-lattice relaxation rates in zero applied field give clear evidence for critical slowing down of Mn spin fluctuations over a wide range of temperatures. Muon-relaxation rates well below T_c are consistent with a two-magnon (Raman) scattering mechanism.

I. INTRODUCTION

Dilute alloys of iron-group transition metals in palladium have been of considerable interest in recent years,[1] due to the remarkable effect of exchange coupling between conduction electrons on the effective exchange interactions between localized impurity moments. In *Pd*Mn, especially, the competing effect of long-range ferromagnetic and near-neighbor antiferromagnetic interactions leads to a complicated magnetic phase diagram.[2] Prior work has disclosed a consistent picture of this alloy system, which can be summarized as follows.

(i) Mn in Pd possesses a "giant moment" of $\sim 7.8\mu_B$, described by a spin $S = \frac{5}{2}$ and effective g factor $g_{\rm eff} \approx 2.7$. This giant moment consists of a localized Mn impurity moment together with an induced polarization cloud of host Pd conduction electrons.

(ii) At low Mn concentrations ($c \lesssim 3$ at.%) the Mn moments order ferromagnetically, due to a long-range indirect exchange interaction.

(iii) At higher concentrations the average Mn separation decreases, so that direct Mn-Mn interactions become important. For $c \gtrsim 5$ at.% Mn, the ordering appears to be random, short ranged, and in some respects similar to that in canonical spin-glasses.[3] This suggests that direct Mn-Mn interactions are antiferromagnetic in nature and compete with the ferromagnetic long-range coupling. A complicated "mixed" region, with no well-defined ordering temperature, is found for Mn concentrations between 3 and 5 at.%.[2]

This paper reports a study of positive muon (μ^+) spin relaxation in a ferromagnetic *Pd*Mn alloy, using the muon spin-rotation (μSR) technique.[4–6] (A μSR study of giant-moment *Pd*Fe alloys in a transverse applied field has been reported by Nagamine *et al.*[7]) The chief goal of the present work is to obtain information on the effect of spatial inhomogeneity on magnetic ordering in *Pd*Mn. Comparison between μSR data, magnetization measurements, and mean-field theories of uniform[8] and disordered[9] ferromagnets can be used to infer microscopic features of impurity-spin configurations. In particular, the observed "shortfall" of the saturation magnetization in ferromagnetic *Pd*Mn alloys[10] is mirrored in the μSR transverse relaxation rate $\lambda_\perp$. This suggests that disorder in the ferromagnetic spin configuration is indeed microscopic in scale.

The article is organized as follows. Section II describes details of the sample characterization and data analysis. The results of measurements of inhomogeneous quasistatic μ^+ local-field distributions are discussed in Sec. III, and Sec. IV treats μ^+ spin-lattice relaxation in zero and longitudinal applied fields. Conclusions are given in Sec. V. It is noted that the μSR behavior of *Pd*Mn is very different from that of the canonical spin-glasses[11,12] *Ag*Mn, *Cu*Mn, and *Au*Fe, and it is argued that this difference is due to the nonzero (ferromagnetic) mean of the exchange interaction.

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II. EXPERIMENT

Muon spin-rotation and relaxation experiments were carried out on a *Pd*Mn alloy of Mn concentration $c=2.0$ at. % at the stopped muon channel of the Clinton P. Anderson Meson Physics Facility (LAMPF), Los Alamos National Laboratory. The sample was in the form of a polycrystalline disk 3 mm thick and 30 mm in diameter, which had been prepared from ultrapure starting materials by arc melting. After rolling and spark cutting to the desired shape the sample was heat treated at 800 °C for 1 h and returned to room temperature within 15 min. The nominal composition was verified by chemical analysis to be 2.06 at. % Mn. A ferromagnetic Curie temperature $T_c=5.8$ K was obtained from ac susceptibility measurements as well as the μSR data (see Sec. IV). Experiments were carried out in zero field, or with an external magnetic field applied in either the transverse or longitudinal directions relative to the initial μ^+ polarization direction. Measurements[13] of μ^+ diffusion in Pd lightly doped with Gd showed a negligible μ^+ diffusion rate below ~ 100 K. Therefore, the behavior of μ^+ local fields is not complicated by muon motion over most of the temperature region of interest.

The μ^+ transverse relaxation rate $\lambda_\perp$ was measured at fields of 0.2, 1, and 5 kOe over the temperature range 3–300 K. Time-differential histograms were analyzed by least-squares fitting to standard expressions for differential count rates.[4–6] The transverse depolarization function used in the fitting procedure was usually the exponential $G_\perp(t)=\exp(-\lambda_\perp t)$; fits to Gaussian or "root exponential" ($\exp[-(\lambda_\perp t)^{1/2}]$) functional forms yielded appreciably larger values of the statistic χ^2. No frequency shift was found to within the accuracy ($\sim 0.1\%$) of the measurements.

Muon longitudinal depolarization functions $G_{||}(t)$ were obtained from time-differential measurements in zero and nonzero longitudinal fields. Above the Curie temperature, $G_{||}(t)$ was exponential to within the accuracy of the data.

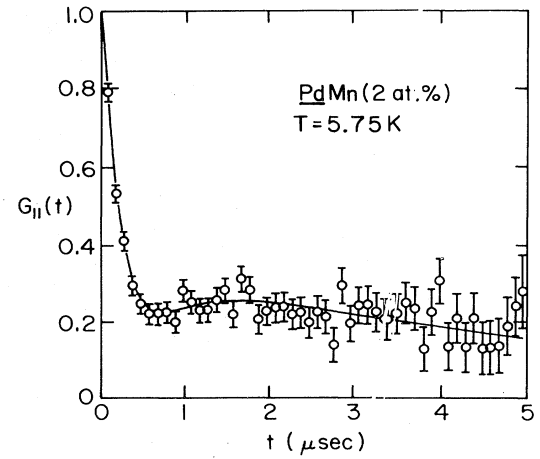

FIG. 1. Representative zero-field relaxation function $G_{||}(t)$ in Pd–2.0 at. % Mn below the Curie temperature ($T_c=5.8$ K). The curve is a fit of Eq. (1) to the data at $T=5.75$ K.

Well below T_c, the zero-field data followed the "spin-glass" form[12]

$$G_{||}(t)=\tfrac{2}{3}(1-at)\exp(-at)+\tfrac{1}{3}\exp(-\lambda_{||}t)\ , \qquad a\gg\lambda_{||}\ . \tag{1}$$

A representative fit of Eq. (1) to the data is shown in Fig. 1. The parameter a, which is the width (in frequency units) of the quasistatic local-field distribution in zero field,[12] and the spin-lattice relaxation rate $\lambda_{||}$ were both temperature dependent. In nonzero fields (up to 5 kOe), spin-lattice relaxation rates were obtained from exponential fits to the portion of the data fulfilling the condition $t\gg 1/a$, since $G_{||}(t)$ for shorter times is not a simple analytic function.

It was determined that longitudinal relaxation rates were significantly lower than transverse rates over nearly the entire range of temperature and field. Only dynamic (spin-lattice) relaxation processes affect the longitudinal μ^+ polarization. The observation that longitudinal relaxation is considerably weaker than transverse relaxation implies, therefore, that the latter is only weakly affected by dynamic processes. Thus the transverse relaxation rate reflects the width of the distribution of quasistatic[14] internal fields.

III. LOCAL-FIELD DISTRIBUTIONS AT μ^+ SITES

Walstedt and Walker[15] (WW) have treated the problem of local-field distributions at spin-probe sites in dilute magnetic alloys. The probe spins interact with impurity moments via two mechanisms: (i) dipolar fields and (ii) indirect [Ruderman-Kittel-Kasuya-Yosida (RKKY)] coupling mediated by conduction electrons. The former dominates in μSR, since the μ^+ hyperfine contact interaction is relatively weak in metals.[6] WW considered two limits for the fluctuation rate of the impurity spin, according to whether transitions between impurity-spin Zeeman levels are slow or fast compared to the probe-spin linewidth induced by the impurities. We call these the "quasistatic" and "rapid" limits, respectively. In both cases the r^{-3} spatial dependence of the coupling leads to a Lorentzian line shape in transverse field (i.e., an exponential polarization decay), with a width $\lambda_\perp$ given by

$$\frac{\lambda_\perp}{\lambda_{\perp 0}}=\begin{cases}\langle\,|S_z|\,\rangle_{\rm th} & \text{(quasistatic limit)}\\ \langle S_z\rangle_{\rm th} & \text{(rapid limit),}\end{cases} \tag{2a}$$

where

$$\lambda_{\perp 0}=5.065\rho c\gamma_\mu\, g\mu_B \tag{2b}$$

in the case of dipolar coupling. Here ρ is the impurity-site number density, c is the impurity concentration, γ_μ is the μ^+ gyromagnetic ratio ($\gamma_\mu/2\pi=13.554$ MHz/kOe), S_z is the impurity-spin component in the direction of the field, and $\langle\ \rangle_{\rm th}$ indicates a thermal average.

The above results are valid for equivalent impurity spins, i.e., a set of spins which all have the same values of $\langle S_z\rangle_{\rm th}$ and $\langle\,|S_z|\,\rangle_{\rm th}$. If, on the other hand, the impurities possess randomly distributed values of $\langle S_z\rangle_{\rm th}$

or $\langle\,|S_z|\,\rangle_{\rm th}$, the appropriate generalization of Eq. (2) is[15]

$$\frac{\lambda_\perp}{\lambda_{\perp 0}}=\begin{cases}\langle\langle\,|S_z|\,\rangle_{\rm th}\rangle_{\rm dis} & \text{(quasistatic)}\\ \langle\,|\langle S_z\rangle_{\rm th}|\,\rangle_{\rm dis} & \text{(rapid)},\end{cases} \tag{3}$$

where $\langle\ \rangle_{\rm dis}$ indicates an average over the distribution of thermal average values. The absolute values in Eqs. (2) and (3) follow from the invariance of the dipolar field distribution under inversion of the impurity-spin system. Note that the average over random impurity spatial configurations has been carried out in deriving the Lorentzian shape of the probe-spin field distribution and in calculating its width. The averages over disorder in Eq. (3) are of a different kind, namely over the disorder induced by random exchange fields, etc., in the impurity-spin system. Note also that $\lambda_\perp$ is proportional to the magnetization (itself proportional to $\langle\langle S_z\rangle_{\rm th}\rangle_{\rm dis}$) for equivalent spins in the rapid limit, but not necessarily in general.

The above results hold strictly only in the dilute limit $c\to 0$. We have carried out numerical calculations of the dipolar-field distribution at the μ^+ site in the fcc Pd lattice at finite Mn concentrations, following WW, to determine the approximate range of validity of the dilute-limit WW line shape and width. The calculations were carried out for the equivalent-spin case. Both octahedral and tetrahedral μ^+ stopping sites were studied, since the actual site is unknown. For $c=2$ at.% "satellite" lines appear in the μSR spectrum, as expected,[15] due to specific Mn near-neighbor configurations. The satellite frequencies depend strongly on crystal orientation with respect to the field. Consequently, the satellites average to a broad background in a polycrystalline sample. The width of the remaining central line is within 10% of that given by Eq. (2). We shall therefore use the dilute-limit results in the following analysis.

In deriving Eq. (2), WW considered only the z component of the impurity moments. Fiory has pointed out[16] that in a frozen-spin system with dipolar coupling to the muon, transverse components of the impurity moments will contribute to the distribution of z components of the μ^+ local fields. This turns out not to affect the Lorentzian shape of the local-field distribution, but the width $\lambda_\perp$ is changed. In the case of randomly oriented impurity spins, $\lambda_\perp$ is reduced by $\sim 10\%$ with respect to Eq. (2). Fiory treated the distribution in zero field explicitly, but his result is not changed by the application of a transverse field as long as the impurity-spin system does not precess in the field. Precession of the coupled-spin system would occur if interactions between spins were completely isotropic, because then the exchange energy would be invariant under rotation of the spin system; however, anisotropy effects are likely to prevent such precession, at least in modest fields. We therefore use Eq. (2) to describe our data in the frozen state.

The magnetization data of Star *et al.*[10] showed that at a Mn concentration of 2.45 at.% the giant moment is described by an enhanced g factor: $g_{\rm eff}=2.65$. The increase in moment above the bare Mn value ($g=2$) is due to an induced conduction-electron polarization cloud in the Pd host, which has a spatial extent of ~ 5 Å.[17] For a Mn concentration of 2 at.% the average impurity spacing is 4.8 Å (Ref. 18) so that to a first approximation the polarization cloud can be taken to be uniform throughout the system. Such a uniform magnetization density does not contribute to the spread in μ^+ local fields, and thus we expect that the observed values of $\lambda_\perp$ will not reflect the giant moment. We therefore use $g=2$ instead of $g_{\rm eff}=2.65$ in Eq. (2b).

A. Paramagnetic state

At high temperatures ($T\gg T_c$) the rapid-fluctuation limit should apply. Here the magnetization is found to obey a Curie-Weiss law,[10] with a paramagnetic Curie-Weiss temperature $\Theta_p=7.1$ K. If the exchange interactions do not introduce frozen-in disorder in the paramagnetic state, so that the μ^+ linewidth tracks the magnetization [Eq. (2a)], we can use

$$\langle S_z\rangle_{\rm th}=M_{\rm mol}/Ng_{\rm eff}\mu_B\ , \tag{4}$$

where $M_{\rm mol}$ is the molar magnetization and N is Avogadro's number, to obtain

$$(\lambda_\perp)_{\rm para}=5.79\frac{H}{T-\Theta_p}\ , \tag{5}$$

with H in kOe and $\lambda_\perp$ in $\mu{\rm s}^{-1}$. Equation (5) is plotted together with the experimental results in Fig. 2.

The excellent fit shows that the μ^+ linewidth does indeed track the bulk magnetization down to $\sim 1.5T_c$. This behavior is to be contrasted with μSR measurements[19] in spin-glass *Ag*Mn, where the observed linewidth

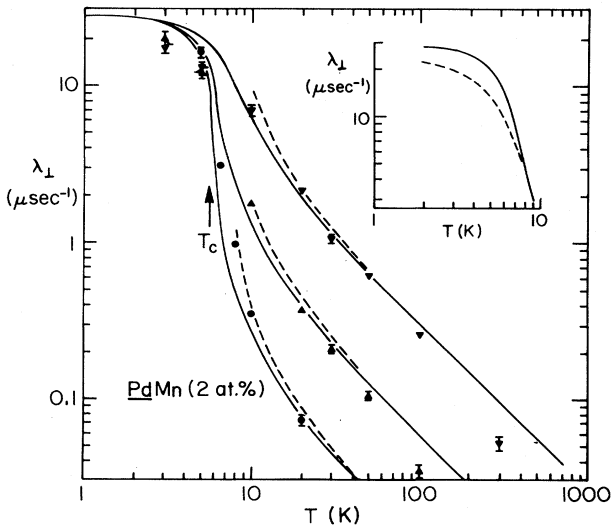

FIG. 2. Temperature and field dependence of the transverse μ^+ relaxation rate $\lambda_\perp(H,T)$ in Pd—2.0 at% Mn. Circles indicate $H=200$ Oe. Upright triangles show $H=1$ kOe. Inverted triangles indicate $H=5$ kOe. The dashed curves are obtained from susceptibility data. The solid curves are obtained from the uniform mean-field model. The inset shows the temperature dependence of $\lambda_\perp$ in the SK model [Eq. (11)] with $H=5$ kOe. Dashed curve represents $J_0=5.8$ K, $J_0/J=1.1$. Solid curve represents $J_0=5.8$ K, $J=0$.

increases considerably more rapidly than predicted from the calculated magnetization as the glass temperature T_g is approached from above. It should be stressed that inhomogeneity in the local field at μ^+ sites is due to a combination of (i) random placement of impurity spins around the various muon sites in the dilute alloy, and (ii) inhomogeneity in the values of $\langle S_z \rangle_{\rm th}$ and $\langle\,|S_z|\,\rangle_{\rm th}$. The good agreement between μ^+ linewidth and measured magnetization in *Pd*Mn suggests that (ii), if present above T_c, is not evident in the μSR data.

B. Ferromagnetic state

If the impurity-spin system is saturated, then $|\langle S_z \rangle_{\rm th}| = \langle\,|S_z|\,\rangle_{\rm th} = S$. The corresponding μ^+ linewidth is easily calculated from Eq. (2):

$$(\lambda_\perp)_{\rm sat} = 27.9\ , \tag{6}$$

in units of μs^{-1}. As can be seen from Fig. 2, the measured $\lambda_\perp$ at $T = 3$ K $= 0.52 T_c$ is only $\frac{2}{3}$ of this value. Because of the inadequacies of existing models, we can only discuss this "shortfall" in a general way. It is similar to the reduced spontaneous magnetization observed below T_c and attributed[10] to the formation of local antiferromagnetic pairs. The μSR data do not determine the nature of the spin-system disorder directly, but the onset of disorder is expected on general grounds to affect the μ^+ linewidth as discussed below. Before introducing two possible models for such an effect, we discuss the question of which fluctuation-rate limit of Eq. (3), quasistatic or rapid, is applicable below T_c.

The question of the time scale of impurity fluctuations is also of considerable importance in the canonical spin-glasses,[11,12] for which a wide range of fluctuation rates has been inferred from both μSR (Ref. 20) and neutron spin-echo[21] experiments. In *Ag*Mn the width of the static field distribution in zero field is noticeably temperature dependent below T_c, which suggests that spin disorder is not frozen in quasistatically[11] (except, perhaps, at very low temperatures). In Fig. 3 we plot the width $a(T)$ of the μ^+ static local-field distribution in *Pd*Mn for zero applied field, obtained from fits of the data to Eq. (1). As in the case of transverse field, the temperature dependence of $a(T)$ suggests that rapid fluctuations average out the frozen-in disorder as T_c is approached from below. Indeed, for $0.5 < T/T_c < 0.9$, $a(T)$ in zero field (Fig. 3) and $\lambda_\perp(T)$ in transverse field (Fig. 2) are nearly the same. Henceforth, we shall assume that fluctuations remain rapid in ferromagnetic Pd—2.0 at.% Mn, at least above $\sim T_c/2$.

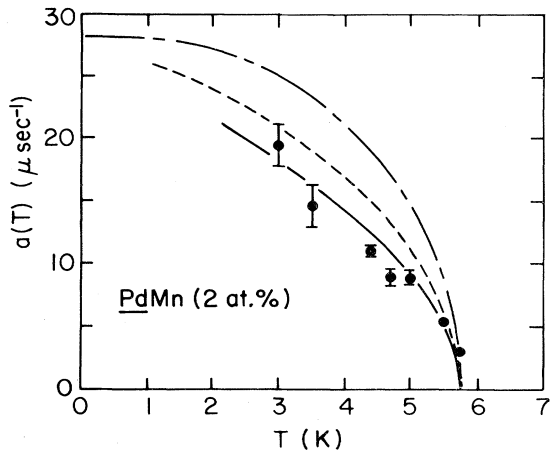

FIG. 3. Temperature dependence of the zero-field μ^+ quasistatic field distribution width $a(T)$ (in frequency units) in Pd—2.0 at.% Mn. Solid curve is the SK model [Eq. (11)] with $J_0 = 5.8$ K, $J_0/J = 1.1$. Dashed curve is the SK model, $J_0 = 5.8$ K, $J_0/J = 1.5$. Dashed-dotted curve is the homogeneous MF model, $S = \frac{5}{2}$.

To proceed further, we require a model for the behavior of the spin thermal averages for all temperatures above and below T_c. The mean-field (MF) model[8] yields the simplest calculation of $\langle S_z \rangle_{\rm th}$, but it must be used only as a qualitative guide to understanding the data. Star *et al.* have pointed out shortcomings of the MF model in accounting for more than the trend of the magnetization data in ferromagnetic *Pd*Mn.[10] The MF model for equivalent spins (which we call the homogeneous MF model) gives the familiar result

$$\langle S_z \rangle_{\rm th} = S B_S[g_{\rm eff}\mu_B S(H + \gamma\sigma_z)/k_B T]\ , \tag{7}$$

where B_S is the Brillouin function for spin S, γ is the molecular-field constant, and $\sigma_z = N g_{\rm eff}\mu_B \langle S_z \rangle_{\rm th}$ is the total (impurity plus host polarization) magnetization. The homogeneous MF result for $\lambda_\perp$, obtained from Eqs. (2) and (7), has been plotted in Fig. 2 along with the data. The value of γ has been chosen to reproduce the experimental Curie-Weiss temperature $T_c = 5.8$ K, rather than the paramagnetic state Curie-Weiss temperature $\Theta_p = 7.1$ K, so that there is some difference between the MF and high-temperature magnetization results [Eqs. (7) and (5), respectively]. Qualitative agreement with the homogeneous MF curves is seen, but they tend to exceed the data at low temperatures. We therefore consider a model which allows inequivalent Mn spins.

In spite of a great deal of theoretical work, the only tractable model which explicitly accounts for the disorder in a random magnet remains the infinite-range mean-field Ising model of Sherrington and Kirkpatrick (SK).[9] To the shortcomings of the homogeneous MF model are now added the questions of applicability of an Ising model to a vector-spin (Heisenberg) system[22] and the validity of assuming an infinite range for exchange interactions. In addition, the so-called "replica trick" required for a unique order parameter[23] is known to fail below the de Almeida—Thouless curve[24] in the H-T plane. Nevertheless, the model treats the effect of disorder on thermodynamic quantities, and will be considered as a guide to understanding the observed μSR behavior.

In the SK model, the impurity spins ($S_i = \pm 1$) are coupled by random interactions, which are independently distributed according to a Gaussian probability density. SK show that the replica-symmetric thermodynamics of the model is obtained from the solution of the coupled equations

$$m=(2\pi)^{-1/2}\int dz\exp(-z^2/2)\tanh\Xi \tag{8a}$$

and

$$q=(2\pi)^{-1/2}\int dz\exp(-z^2/2)\tanh^2\Xi\ , \tag{8b}$$

where

$$m=\langle\langle S_i\rangle_{\rm th}\rangle_{\rm dis},\quad q=\langle\langle S_i\rangle_{\rm th}^2\rangle_{\rm dis}\ , \tag{9a}$$

$$\Xi=[J_0m+J(q)^{1/2}z+Sg_{\rm eff}\mu_BH]/k_BT\ , \tag{9b}$$

and J_0/N and J^2/N are the mean and variance, respectively, of the Gaussian exchange-interaction distribution. Once m and q are known, the quantity

$$r=\langle\,|\,\langle S_i\rangle_{\rm th}\,|\,\rangle_{\rm dis}$$
$$=(2\pi)^{-1/2}\int dz\exp(-z^2/2)\,|\tanh\Xi| \tag{10}$$

may be calculated. Then, from Eq. (3), the μ^+ field distribution is proportional to r in the fast-fluctuation limit.

SK found that for $J_0<J$, a spin-glass phase with no spontaneous magnetization is formed for $T<T_g=J/k_B$. For $J_0>J$ a ferromagnetic state occurs below $T_c=J_0/k_B$ with, however, a reduced spontaneous magnetization due to the competing antiferromagnetic interactions. It is this latter region which we consider as a model for ferromagnetic *Pd*Mn. We find that for $J_0/J\gtrsim 2.5$ the model reproduces a homogeneous (Ising) MF solution, so that the effect of disorder in the ferromagnetic phase is confined to the range $1<J_0/J<2.5$. It seems reasonable to apply this model to *Pd*Mn, since the effect of increasing Mn concentration is known to be an increase of the tendency towards spin-glass behavior.[2] Indeed, the *Pd*Mn system undergoes a broad transition from a ferromagnet to a spin glass with increasing Mn concentration above about 3 at.%. We therefore take $J_0/k_B=5.8$ K, and attempt to find a value of J such that the trends of both the μSR linewidth and the spontaneous magnetization[10] are reproduced. Only values of J very close to J_0 fulfill both criteria.

The temperature dependence of the SK prediction for $a(T)$ is given in Fig. 3 for several values of J_0/J. These curves are obtained by finding solutions of Eqs. (8) for m and q, calculating r from Eq. (10), and taking

$$a(T)=(\lambda_\perp)_{\rm sat}r\ , \tag{11}$$

where $(\lambda_\perp)_{\rm sat}$ is given by Eq. (6).

It can be seen that, for the lower values of J_0/J, the SK model accounts qualitatively for the data in that the predicted $a(T)$ rises rather slowly towards its zero-temperature value compared to the homogeneous MF magnetization. In the inset of Fig. 2, we plot $\lambda_\perp$ calculated from the SK and MF models for an applied field of 5 kOe. The SK model is also seen to reproduce the observed shortfall in $\lambda_\perp$ relative to the mean-field value. Finally, one can compare the SK-model spontaneous magnetization with the data[10] from a Pd—2.5-at.% Mn specimen. This is done in Fig. 4, where it can be seen that rough agreement is found for the smaller values of J_0/J. It should be noted that the downturn in the SK magnetization at low temperatures is due to the nearby ferromagnetic—spin-glass transition found in the replica symmetric solution. Evidence for such a transition has been found in both the *Pd*Mn and *Pd*FeMn alloy systems.[25]

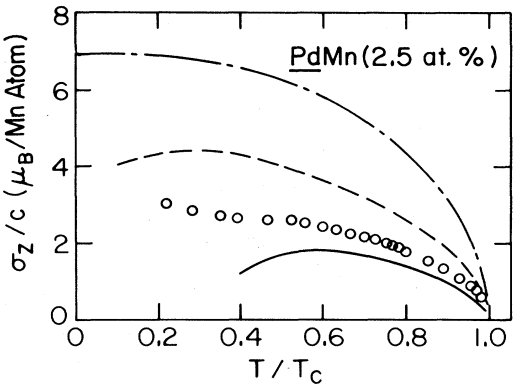

FIG. 4. Temperature dependence of the normalized spontaneous magnetization $\sigma_z(T)/c$ in Pd—2.5 at.% Mn. Data from Ref. 10. Solid curve is the SK model (see text), $J_0=5.8$ K, $J_0/J=1.1$. Dashed curve is the SK model, $J_0=5.8$ K, $J_0/J=1.35$. Dashed-dotted curve is the homogeneous MF model, $S=\frac{5}{2}$.

IV. SPIN-LATTICE RELAXATION

Figure 5 gives the temperature dependence of $\lambda_{||}(T)$, obtained as discussed in Sec. II, in zero field and 5 kOe. We interpret the sharp peak in $\lambda_{||}$ as being due to critical slowing down of Mn spin fluctuations in the neighborhood of T_c.[26] Indeed, the data enable us to identify the Curie temperature within ± 0.1 K (Sec. II). In addition, application of a relatively weak field ($g_{\rm eff}\mu_BH\ll k_BT$) sharply reduces the μ^+ relaxation rate, presumably by suppressing the critical fluctuations.

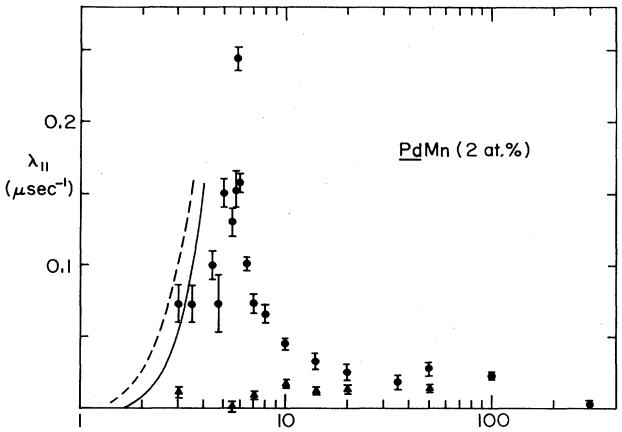

FIG. 5. Temperature dependence of the μ^+ spin-lattice relaxation rate in Pd—2.0 at.% Mn at zero field (circles) and 5 kOe (triangles). The curves are predictions of the two-magnon (Raman) scattering mechanism, Eq. (13) (see text). Solid curve is for the anisotropy energy $\hbar\omega_A=0.3k_BT_c$. Dashed curve is for $\hbar\omega_A=0.2k_BT_c$.

These results are in dramatic contrast with spin-lattice relaxation measurements[11] in spin-glass *Ag*Mn, where $\lambda_{||}(T)$ exhibited a very large value ($\sim 2\ \mu s^{-1}$) at T_g and remained $\geq 0.1\ \mu s^{-1}$ in fields up to 5 kOe. This difference further emphasizes the difference between local dynamics in the two systems, which in other respects (impurity concentration, ordering temperature) are quite comparable. Clearly, a powerful relaxation mechanism is at work in *Ag*Mn which is not present in *Pd*Mn.

The wide temperature range over which dynamical critical effects appear to be observed is in contrast with the relatively narrow range reported[27] for uniform ferromagnets. Our temperature resolution and control during these measurements were not sufficient to enter the critical region very near T_c, so we do not attempt to obtain dynamical critical exponents from the data shown in Fig. 5. It is clear, however, that fluctuations play an important role in impurity-spin dynamics relatively far from T_c. Verbeek *et al.*[28] have also concluded, on the basis of elastic neutron scattering studies, that the critical region of ferromagnetic *Pd*Mn alloys is anomalously wide. They have proposed an explanation for this observation, in which the formation of ferromagnetic clusters above T_c and the persistence of finite clusters (and "tag ends" connected to an infinite cluster) below T_c serve to broaden the temperature range of critical behavior. It would be of considerable interest to obtain from such a model both the distribution of quasistatic local fields and the fluctuation spectrum, for comparison with the present μSR results.

The width of the critical region cannot be explained with a long-range MF model of Ising critical dynamics[29] as in the case of a uniform ferromagnet, and it seems unlikely that disorder would decrease the effect of fluctuations. We conclude that the spin-lattice relaxation data do not support the use of a homogeneous MF model to describe ferromagnetic *Pd*Mn. This comes as no surprise, given the magnetization and static μSR linewidth results, but it further underlines the qualitative nature of the discussion of Sec. III above.

At low temperatures, the dominant relaxation mechanism in ferromagnets is often Raman scattering of magnons by probe spins.[30] Turov and Petrov[31] have described a continuum theory of this mechanism in the case where the magnons couple to the probe spins via the dipolar field. Their results are for the case where the magnon frequency $\omega(k)$ is given by

$$\omega(k)=\omega_A+Dk^2\ , \tag{12}$$

where $\hbar\omega_A$ is the anisotropy energy and D is the spin-wave stiffness constant. Then the probe-spin relaxation rate $\lambda_{||}$ is given by

$$\lambda_{||}=\frac{2\gamma_\mu^2\gamma_e^2k_B^2T^2}{15\pi D^3}\ln\left[\frac{k_BT}{\hbar\omega_A}\right]\ , \tag{13}$$

where γ_e is the electron gyromagnetic ratio. The magnetization data[10] yield a value for D of $\approx(0.7k_BT_c\ \text{Å}^2)/\hbar$. Because the value of $\hbar\omega_A$ is unknown, we plot in Fig. 5 curves from Eq. (13) for two values of $\hbar\omega_A/k_BT_c$. Although the data do not really extend to sufficiently low temperatures to justify the spin-wave approximation, it can be seen that the trend is consistent with the curve for $\hbar\omega_A\approx 0.3k_BT_c$. It is clear, at any rate, that Raman magnon scattering can explain the order of magnitude of the observed relaxation. This is again in contrast with the situation in *Ag*Mn, where the observed relaxation rates at $\sim T_g/2$ are almost 2 orders of magnitude more rapid than estimates obtained from simulated low-temperature magnon spectra.[32] In ferromagnetic *Pd*Mn, there appears to be no additional relaxation due to the random nature of the spin system, whereas some such mechanism must clearly be sought in the case of spin-glass *Ag*Mn.

V. CONCLUSIONS

μ^+ relaxation rates in the random ferromagnet Pd—2.0 at.% Mn have been measured for transverse, zero, and longitudinal applied fields. In the paramagnetic state the transverse relaxation is determined by the bare impurity moment; the giant-moment polarization cloud seems to make no observable contribution. Here, the WW linewidth formula for dilute impurities and dipolar coupling to the μ^+ moment is in good agreement with the transverse relaxation rate. In the ferromagnetic phase, the relaxation rates are less than those predicted by a mean-field model assuming a uniform spin system. Our data can be accounted for qualitatively by the SK theory for disordered ferromagnets.

Spin-lattice relaxation rates from zero-field μSR give clear evidence for critical slowing down of Mn spin fluctuations near T_c. Although this slowing down occurs over a wide temperature range, a well-defined value of T_c can be determined, which is in excellent agreement with macroscopic measurements. The relaxation mechanism in *Pd*Mn is much less effective than that in spin-glass *Ag*Mn.

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