

Ground state of giant moments associated with Fe impurities in Pd

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(Received 2 April 1997)

We identify the isolated *giant moments* associated with Fe impurities in a Pd matrix as a spin-fluctuation system with a spin-fluctuation temperature of ≈ 250 mK. This conclusion is achieved from the temperature dependence of the resistivity of 2.5 and 16 ppm Fe containing samples, extracted from the real part of the ac electrical impedance measured at $0.1 \text{ mK} \leq T \leq 600 \text{ mK}$, $0.1 \mu\text{T} \leq B < 10 \text{ mT}$, and $f = 175 \text{ Hz}$. The effect of giant moments, which interact with others, on the resistivity is also discussed. The imaginary part of the ac electrical impedance, which is proportional to the ac susceptibility, reveals spin-glass freezing at around 1 mK, two orders of magnitude smaller than the spin-fluctuation temperature. This spin-glass freezing is confirmed by the characteristic slow relaxation of dc magnetization below the spin-glass transition temperature. [S0163-1829(97)09634-3]

I. INTRODUCTION

The formation of local magnetic moments of *3d* impurities, especially Fe atoms, in nonmagnetic host metals has been attracting a lot of attention and is still a very active field¹ because of its practical as well as scientific importance. Even if an impurity in a host metal forms a local magnetic moment, it sometimes loses its moment at low temperatures below a certain characteristic temperature.² In the case of certain impurity-host combinations, this characteristic temperature is estimated to be above the melting temperature of the host metal: The impurity in the host is always nonmagnetic. Most of so-called *magnetic* impurities are expected to be *nonmagnetic* at 0 K. In this work, we investigate Fe impurities in palladium in the low-temperature limit to seek its ground state.

Palladium is an exchange enhanced metal and close to the ferromagnetic transition, which means that magnetic properties of Pd are very sensitive to *3d* impurities.^{3,4} Fe impurities polarize the surrounding Pd matrix: The polarization is ferromagnetic and decays exponentially at short distance, while it is oscillatory at long distance.⁵ The strength and the range of the ferromagnetic polarization near Fe impurities were studied from the Fe concentration dependence of the ferromagnetic transition temperature of samples with more than 800 ppm Fe.^{6,7} The range r_{FI} of the ferromagnetic interaction is weakly (logarithmically) temperature dependent and is about 5 nm at 100 mK.⁶⁻⁸ The complex of Fe impurity plus ferromagnetic polarization cloud is recognized as a *giant magnetic moment* $\mu_{\text{gm}} \sim 10\mu_B$.^{9,10} The oscillatory interaction at long distance was confirmed by NMR (Ref. 11) and muon spin rotation experiments.¹² This oscillatory interaction with the anisotropic dipole interaction lead to a spin-glass transition¹³ at millikelvin temperatures for samples with less than 200 ppm Fe impurities.^{10,14,15} The spin-glass freezing temperature T_f is reported as $T_f/x = 0.19 \text{ mK/ppm}$,¹⁰ where x is the Fe concentration in at. ppm unit. We discuss here the ground state of isolated giant moments from the temperature dependence of the resistivity between 0.1 and 600 mK at $0.1 \mu\text{T} \leq B < 10 \text{ mT}$. In addition we investigate the properties of interacting giant moments.

It turns out to be very difficult to achieve isolated *giant moments* associated with Fe impurities in a Pd matrix because of the interactions among them. Even our very dilute samples are not free from interactions as we will discuss later. When Fe impurities are distributed randomly in a Pd matrix, the number-density probability $P(r)$ that the nearest Fe impurity can be found at distance r is

$$P(r) = n_0 4\pi r^2 \exp\left(-n_0 \frac{4\pi}{3} r^3\right), \quad (1.1)$$

where n_0 is the Fe impurity number density of PdFe.⁸ This calculation is a continuous medium approximation which can be justified for samples with small Fe concentrations. A similar discussion was made by Jaccarino and Walker.¹⁶ The number density of isolated giant moments n_1 is estimated from the number density of Fe impurities that are separated from all other Fe impurities with a characteristic distance $r_c \geq r_{\text{FI}}$. We introduce the new length scale r_c , which will be justified in Sec. IV. We expect that r_c is only weakly temperature dependent since r_{FI} is logarithmically temperature dependent. Therefore, we assume that r_c is constant in the temperature range from 0.1 to 600 mK of our measurements. n_1 is calculated from

$$n_1 = n_0 \int_{r_c}^{\infty} P(r) dr = n_0 \exp\left(-n_0 \frac{4\pi}{3} r_c^3\right). \quad (1.2)$$

This function is plotted for different r_c values in Fig. 1. For comparison, n_0 is also shown. The largest n_1 's are obtained for rather dilute samples with $x \sim 5$ to 30 ppm. To minimize the contamination by interacting giant moments, one has to practically investigate samples with as small as possible impurity concentrations, and thus minute effects of *giant moments*.

We have measured the ac electrical impedance Z of very dilute samples with a SQUID as a preamplifier. The resistivity ρ can be obtained from the real part $\text{Re}(Z)$ of the impedance, while the changes in the imaginary part $\text{Im}(Z)$ are proportional to the ac susceptibility changes: This is a great benefit for our investigations since we discuss the resistivity

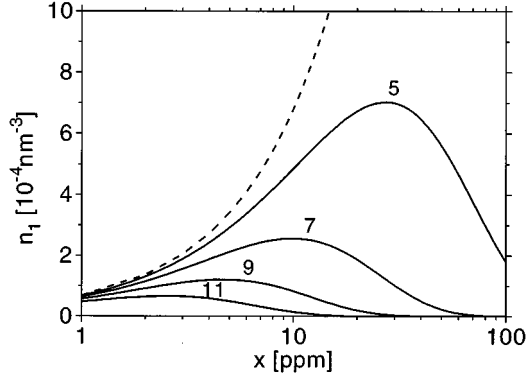


FIG. 1. Number density n_1 of isolated giant moments as a function of Fe concentration x . It is calculated from Eq. (1.2) with $r_c = 5, 7, 9$, and 11 nm. The dashed line is the total Fe number density, n_0 .

changes caused by magnetic impurities. From the temperature dependence of the resistivity, we conclude that the isolated giant moments are a spin-fluctuation system with a spin-fluctuation temperature $T_{sf} \approx 250$ mK. The ac susceptibility shows the spin-glass freezing in the samples at T_f which is two orders of magnitude smaller than T_{sf} . We confirm this spin-glass freezing from the characteristic slow dc magnetization relaxation below T_f in addition to the behavior of the ac susceptibility.

II. EXPERIMENT

A. Sample

Our two samples were prepared from commercially available *pure* Pd (Ref. 17) with Fe concentrations of 2.5 and 16 ppm. Impurities are analyzed by two complimentary methods: mass spectroscopic¹⁸ and susceptibility¹⁰ analysis at $T \geq 1.6$ K. The susceptibility analysis counts the total amount of magnetic impurities although their species cannot be known, while the mass spectroscopy can identify the species although the absolute value might not be very reliable because of the small amount of impurities. The spectroscopic analysis identifies Fe impurities as the only significant magnetic impurity, while the susceptibility determined their amounts. The results of the susceptibility analysis for our samples are reported by Herrmannsdörfer *et al.*¹⁰ A small error in the Fe concentration determination does not alter our discussions.

The samples for the ac electrical impedance measurements are thin foils of typical size of $20 \times 2 \times 0.1$ mm³, which were prepared from a wire (2.5 ppm) or a rod (16 ppm), without any further heat treatments. The surface of the samples was chemically etched to remove contaminations. The residual resistivity ratio (RRR) is 80 for both samples. The skin depth is larger than the thickness of the samples at all temperatures of our measurements. The sample for the dc magnetization measurements is a wire of 1 mm diameter as received.

B. Setup I: Impedance measurement

For the electrical impedance measurements, the experimental setup is the same as the one which we have described

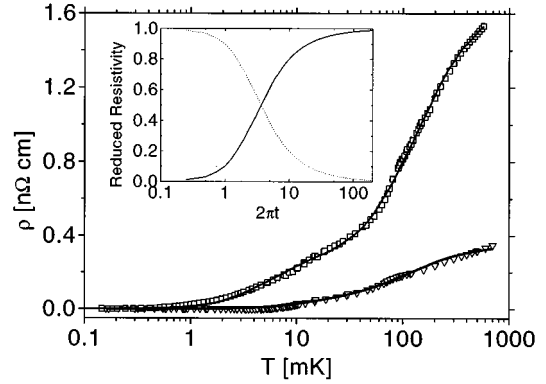


FIG. 2. Temperature dependence of the resistivity of the 2.5 (\square) and 16 ppm (∇) samples at zero field. $f = 175$ Hz. The solid line is $\rho(T) = c_1 \rho_{sf}(T/T_{sf1}) + c_2 \rho_{sf}(T/T_{sf2})$ with $T_{sf1} = 250(260)$ mK, $T_{sf2} = 15(30)$ mK, $c_1 = 1390(290)$ p Ω cm, and $c_2 = 330(85)$ p Ω cm for the 2.5 (16) ppm sample. The inset shows the normalized temperature dependence of the resistivity of *Coles* (solid line) and *Kondo* (dotted line) alloys as a function of reduced temperature $t = T/T_{sf}$.

in our previous publication⁸ except for a small difference in the measurement frequency (175 Hz instead of 159 Hz in our previous work) and larger excitations because of larger RRR (80 instead of 15). We note here that the so-called “zero” field is of the order of $0.1 \mu\text{T}$ with which $\mu_{gm}B \ll k_B T$ can be satisfied even at 0.1 mK.

C. Setup II: Magnetic measurement

For the magnetic measurements, one end of the 1 mm diameter wire with 2.5 ppm Fe impurities is squeezed to a Cu sample holder with a Cu screw. The direction of the magnetic field is along the wire and thus we can ignore the diamagnetic effect. An astatic superconducting pickup coil is wound on a Teflon tube into which the other end of the sample is inserted till the middle of the pickup coil. The measurements are performed with a SQUID. The samples are surrounded by a series of magnetic shields and a static magnetic field coil as in the case of the impedance measurements to obtain a low magnetic field.

D. Cryogenics

We employ a nuclear demagnetization cryostat¹⁹ for both the impedance and dc magnetization measurements.

III. RESULTS

A. Resistivity

The resistivity data at zero field for both samples are shown in Fig. 2. Note that the resistivity value extrapolated to zero temperature is referred to as zero in this work. The resistivity is corrected by subtracting the Pd matrix contribution which is $30T^2$ (p Ω cm/ T^2) as taken from the literature.^{20,21} This correction is negligible below 1 K. The temperature dependences of the resistivity of the two samples are similar, but the sample with *less* Fe impurities shows a *larger* temperature dependence than that with *more* Fe impurities.

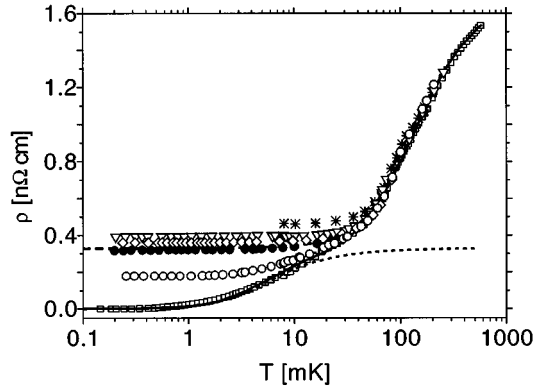


FIG. 3. Temperature dependence of the resistivity of the 2.5 ppm sample at zero (\square), 0.05 (\circ), 0.25 (\bullet), 0.5 (\diamond), 0.75 (∇), and 6.6 ($*$) mT. The solid line is $\rho(T) = c_1 \rho_{sf}(T/T_{sf1}) + c_2 \rho_{sf}(T/T_{sf2})$ with $T_{sf1} = 250$ mK, $T_{sf2} = 15$ mK, $c_1 = 1390$ p Ω cm, and $c_2 = 330$ p Ω cm. The dashed and dotted lines are $\rho(T) = c_1 \rho_{sf}(T/T_{sf1}) + c_2$ and $\rho(T) = c_2 \rho_{sf}(T/T_{sf2})$, respectively.

The effect of a magnetic field on the resistivity is investigated for the 2.5 ppm sample; see Fig. 3. In this work, the field is applied at high temperatures and then the sample is cooled down if it is not mentioned specially. When increasing the magnetic fields, the resistivity deviates at low temperatures from that at zero field. The resistivity is constant at low temperatures. The resistivity at all fields, however, merge to that at the zero field above 50 mK. The field dependences of the resistivity at different temperatures are shown in Fig. 4. We observe the largest field dependence at 0.18 mK, while the resistivities at $T \geq 56$ mK are constant below 2 mT.

B. ac susceptibility

The ac susceptibility of the samples can be obtained from the imaginary part of the ac electrical impedance.⁸ The ac susceptibility at zero field shows a maximum at $T_{\text{peak}} = 0.85(3.5)$ mK for the 2.5 (16) ppm Fe sample as shown in Fig. 5. This maximum is identified as the signature of the spin-glass transition.^{10,14,15} The solid lines in Fig. 5 are

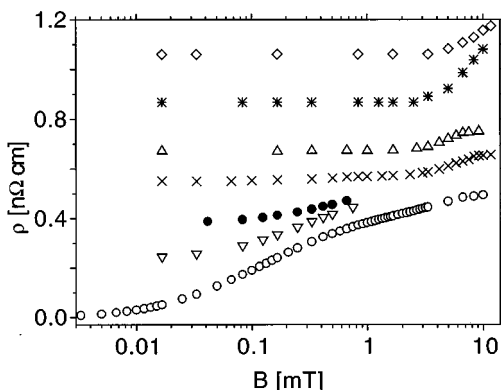


FIG. 4. Field dependence of the resistivity of the 2.5 ppm sample at 0.18 (\circ), 12 (∇), 30 (\bullet), 56 (\times), 76 (\triangle), 110 ($*$), and 170 (\diamond) mK and $f = 175$ Hz.

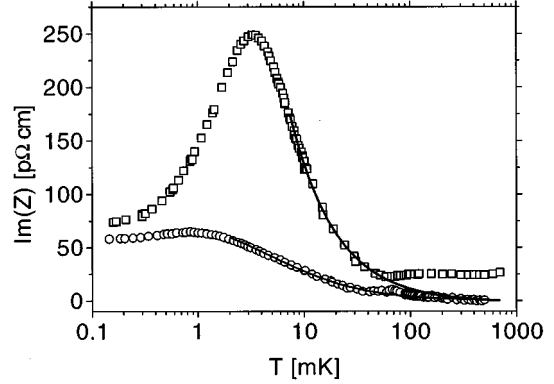


FIG. 5. Temperature dependence of the imaginary part $\text{Im}(Z)$ of the impedance of the 2.5 (\circ) and 16 (\square) ppm samples at zero field and $f = 175$ Hz, which is proportional to the ac susceptibility change. The solid line is a fit to the data of the 2.5 (16) ppm sample with $\text{Im}(Z) = A + C/(T - \Theta)$, where $\Theta = -5$ (-0.5) mK for the 2.5 (16) ppm sample. The data are vertically shifted so that $A = 0$.

fits to the data with a Curie-Weiss law, $\text{Im}(Z) = A + C/(T - \Theta)$ between 10 and 100 mK. We shift the y axis so that $A = 0$ in Fig. 5.

A magnetic field shifts T_{peak} towards higher temperatures, as shown in Fig. 6. Similar behavior was observed before for PdFe samples.^{8,10}

C. dc magnetization

It is often reported that a maximum in the temperature dependence of the ac susceptibility is not sufficient evidence for a spin glass freezing.²² Therefore, we checked the time response after switching on a field of 20 μ T following the zero field cooling. The results are shown in Fig. 7. Because of the time constant of the persistent switch for the magnet, the response during the first 100 s cannot be measured. Afterwards the magnetization changes slowly, which is typical for a spin glass.

IV. DISCUSSION

We discuss our observations in terms of spin fluctuations of magnetic moments in Coles alloys,²³ in this case fluctua-

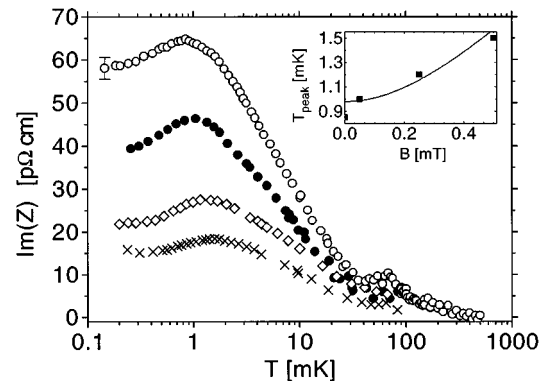


FIG. 6. Temperature dependence of the imaginary part of the impedance of the 2.5 ppm sample at zero field (\circ), 0.05 (\bullet), 0.25 (\diamond), and 0.5 (\times) mT and $f = 175$ Hz. The inset shows the field dependence of T_{peak} ; see text for more details.

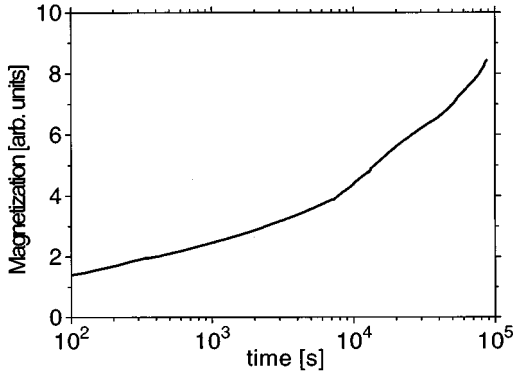


FIG. 7. Relaxation of dc magnetization after switching on a magnetic field of $20 \mu\text{T}$ at $75 \mu\text{K}$ following zero field cooling.

tions of giant moments. A Coles alloy is an alloy that exhibits a characteristic logarithmic increase of resistivity with increasing temperature, which is opposite to a Kondo alloy.^{2,23} The normalized resistivity $\rho_{\text{sf}}(t)$ of Coles alloy as a function of reduced temperature $t = T/T_{\text{sf}}$ can be found in the literature;^{2,23,24} it is reproduced in the inset of Fig. 2. Both the behavior of Coles and Kondo alloys can be explained by the spin fluctuation model²³ and then T_{sf} is equivalent to the Kondo temperature T_K .^{2,4}

In our previous publication,⁸ we have already discussed the spin fluctuation effect of Fe impurities in a Pd matrix. We realized this effect from the observation that the resistivity decreased when the temperature was lowered from 1.6 to 0.5 K. We obtained a spin fluctuation temperature much higher than those reported before by Mydosh⁴ and Riegel *et al.*²⁵ This discrepancy was mysterious for us at that time. Since we had a possible complication by clustering of giant moments (which was discussed, too) in our previous measurements, our discussion was not very convincing. Therefore, one of the motivations of the present work is to check if our previous discussion of spin fluctuation effects is correct or not, with less complication by clustering (in other words, by investigating samples with far less Fe impurities).

We also discuss the difference between the characteristic times in the resistivity and ac susceptibility measurements. This difference leads to two characteristic length scales r_c and r_{F1} which were introduced before.

A. Resistivity

The temperature dependence of the resistivity shown in Fig. 2 cannot be described with $\rho_{\text{sf}}(T/T_{\text{sf}})$ of Coles alloys alone. This is also obvious from the fact that the more concentrated sample shows a smaller temperature dependence. As we discussed in Sec. I, interactions among giant moments may not be avoidable even in our very dilute samples. In this case, the temperature dependence of the resistivity may include the contribution from isolated as well as interacting giant moments. The resistivity change per magnetic impurity due to spin fluctuations can be larger at smaller impurity concentration, as it was found for magnetic moments in noble metal hosts, $Au\text{Fe}$,²⁶ and transition metal hosts, $Rh\text{Fe}$,²⁷ $Pt\text{Mn}$,²⁸ because of their interactions at larger concentrations. Since the spin fluctuation temperature of interacting spins decreases when it interacts with more other

moments,²⁹ we only consider two kinds of giant moments: isolated ones and those which interact with only one other moment. Giant moments which interact with more moments have very small spin fluctuation temperatures and thus will give only a constant offset in our data. According to this hypothesis, the data are analyzed with

$$\rho(T) = c_1 \rho_{\text{sf}}(T/T_{\text{sf1}}) + c_2 \rho_{\text{sf}}(T/T_{\text{sf2}}). \quad (4.1)$$

c_1 (c_2) is a scaling factor for isolated giant moments (giant moments interacting with only one other), while T_{sf1} (T_{sf2}) is the spin fluctuation temperature for isolated (interacting) giant moments. Equation (4.1) reproduces the temperature dependences very well as shown by the solid lines in Fig. 2. We obtain $T_{\text{sf1}} = (250 \pm 30)$ mK, $T_{\text{sf2}} = (15 \pm 10)$ mK, $c_1 = (1390 \pm 150)$ p Ωcm , and $c_2 = (330 \pm 100)$ p Ωcm for the 2.5 ppm sample. The result $T_{\text{sf1}} \gg T_{\text{sf2}}$ is consistent with our hypothesis. In the case of the 16 ppm sample, $T_{\text{sf1}} = (260 \pm 30)$ mK, $T_{\text{sf2}} = (30 \pm 10)$ mK, $c_1 = (290 \pm 50)$ p Ωcm , and $c_2 = (85 \pm 5)$ p Ωcm . The agreement of T_{sf1} and T_{sf2} for the two samples supports our analysis. Spin fluctuation temperatures T_{sf} 's were found to be about 100 mK for $Pd\text{Co}$ ^{30,31} and 400 mK for $Pt\text{Fe}$.^{32,33} Since in both cases the hosts are also exchange enhanced, and the moments caused by the magnetic impurities, $10\mu_B$ for Co in Pd³⁴ and $8\mu_B$ for Fe in Pt,¹⁰ are giant moments as well, $T_{\text{sf}} \approx 250$ mK for $Pd\text{Fe}$ is quite reasonable.

The field dependences of the resistivity at different temperatures shown in Fig. 4 are consistent with our hypothesis. At $T = 170, 110, 76,$ and 56 mK which are all higher than T_{sf2} , the resistivity is constant up to about 2 mT. This result can be understood as follows.² The spin fluctuation effects of interacting *giant moments* are already suppressed by the high temperatures ($T > 2T_{\text{sf2}}$) and thus the magnetic field has no effect on the resistivity. On the other hand, the effects of the magnetic fields on the spin fluctuations of isolated giant moments are still small since the magnetic field is not large enough. We estimate the characteristic magnetic field B_{sf} which corresponds to T_{sf} from $\mu_{\text{gm}} B_{\text{sf}} \sim k_B T_{\text{sf}}$:³⁵ $B_{\text{sf}} \sim 30$ (3) mT for isolated (interacting) giant moments. These values are in good agreement with the observed threshold of the magnetic field ~ 1 mT. At $T = 30, 12,$ and 0.18 mK ($\leq T_{\text{sf2}}$), the magnetic field influences the resistivity even below 1 mT. The effect is larger at lower temperatures as expected from $T_{\text{sf2}} \approx 20$ mK. The different effects of a magnetic field on the isolated and interacting giant moments can be more clearly seen in the temperature dependence of the resistivity at 0.5 mT ($\leq B_{\text{sf2}}$). The field of 0.5 mT quenches the spin fluctuation effects of interacting giant moments, while it has almost no effect on isolated ones, as demonstrated in Fig. 3. The temperature dependence of the resistivity at 0.5 mT is well described by the effect from isolated giant moments [$c_1 \rho_{\text{sf}}(T/T_{\text{sf1}})$] plus that from the quenched interacting giant moments (c_2). This agreement supports our hypothesis.

According to theoretical prediction,³⁵ a magnetic field influences the ground state of a Kondo system in the similar way as a thermal fluctuation does. This implies that the free energy F which describes the impurity state has a universal character:

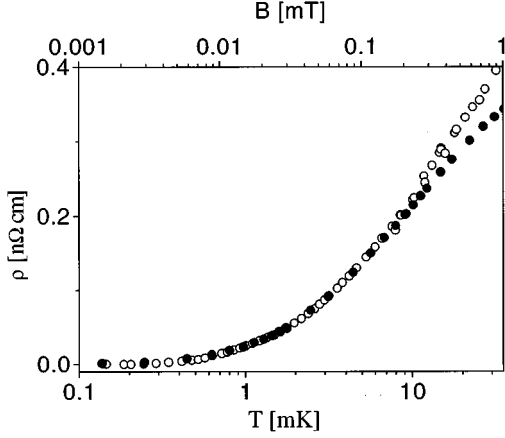


FIG. 8. Comparison between the temperature dependence of the resistivity (○) at zero field ($\ll B_{sf2}$) and its field dependence (●) at 0.18 mK ($\ll T_{sf2}$).

$$\lim_{T, B \ll D} F(T, B; D, J) = Tf(T/T_0, H/T), \quad (4.2)$$

where D , J , T_0 , and f are a momentum cutoff of excitations, a coupling constant between conduction electrons and impurities, a characteristic temperature which is a function of D and J , and a universal function, respectively. Although this relation is for Kondo systems,³⁵ we believe that it is also applicable for spin fluctuation systems.²³ We compare the temperature dependence of the resistivity at zero field ($\ll B_{sf}$) and its field dependence at 0.18 mK ($\ll T_{sf2}$) as shown in Fig. 8. We only compare the low temperature and low field data in order to satisfy $T, B \ll D$. It seems that the theoretical prediction is correct. We find

$$\rho(T, B) = \rho(\sqrt{(B/B_{sf})^{1.7} + (T/T_{sf})^2}). \quad (4.3)$$

This small difference between the magnetic and thermal effects may not be very surprising since there is a subtle difference between them. The magnetic field excites from the ground state only excitations which have maximum spin for a given energy. The temperature, on the other hand, excites all of them.³⁵

Within the framework of the n_1 calculation given in Sec. I, we evaluate n_2 , the number density of Fe impurities which are interacting with only one other Fe impurity. We first note that Eq. (1.2) can be reformulated to $n_1 = n_0 \exp(-V_1 n_0)$ by noticing that $4\pi r_c^3/3$ is a volume V_1 where a second giant moment is excluded. Then, n_2 can be written,

$$n_2 = n_0 \int_0^{r_c} n_0 \exp(-V(r) n_0) 4\pi r^2 dr, \quad (4.4)$$

where $V(r) = 2 \int_{-r}^{r/2} \pi(\sqrt{r_c^2 - t^2})^2 dt$ is a volume from which a third giant moment is excluded when the second giant moment is at the distance r from the first one. We note that Eq. (4.4) is well approximated with $n_2 = n_0(V_1 n_0) \exp(-V_2 n_0)$, where $V_2 \approx 1.5V_1$ can be considered as the averaged volume that a third *giant moment* is excluded from the interacting giant moments.³⁶ n_1 and n_2 are plotted as a function of the Fe concentration with $r_c = 9.6$ nm in Fig. 9.

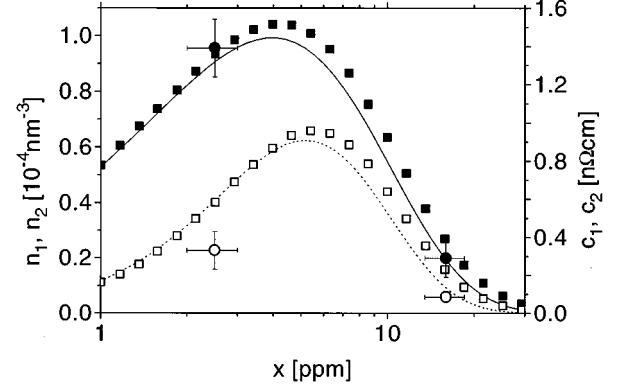


FIG. 9. Number density n_1 of isolated *giant moments* (solid line) and that n_2 of interacting-with-only-one-other (dotted line) for $r_c = 9.6$ nm as a function of concentration x calculated with Eqs. (1.2) and (4.4). The results of Monte Carlo calculation are shown with ■ (□) for n_1 (n_2). c_1 's (●) and c_2 's (○) obtained in Fig. 2 are also shown with the right side axis.

We also performed Monte Carlo simulations with $r_c = 9.6$ nm to obtain n_1 and n_2 as shown in Fig. 9. For these calculations, Fe impurities are randomly distributed in the fcc Pd lattice. The criterion for isolated and interacting-with-one-other giant moments are the same as in the previous paragraph. We obtained essentially identical results as in the previous paragraph for the simpler approach.

We expect that $c_i \propto n_i$, where i is 1 or 2. Therefore, the obtained c_1 and c_2 from the temperature dependence of the resistivity can be compared with n_1 and n_2 , respectively. Such a comparison is performed in Fig. 9. As one can see, the observed concentration x dependence of c_1 and c_2 for the two samples can well be reproduced when $r_c = 9.6$ nm is assumed.

B. ac susceptibility

It is surprising at first that we can observe a spin-glass freezing at $T \ll T_{sf}$. T_f 's usually cover the temperature regime at and above T_{sf} or T_K .⁴ However, we argue like in the case of canonical spin glasses,² that there is a competition between the interimpurity interactions and the spin fluctuations. This competition enables the magnetic moments, at least partly, to freeze into a spin-glass at $T \ll T_{sf}$. This can be understood from Fig. 1. Some giant moments are interacting in resistivity and those are magnetic even at $T \sim T_f$ which is not very far from T_{sf2} (above the curve corresponding to $r_c \approx 10$ nm in Fig. 1). We also point out the difference between the interactions in the canonical spin glasses and in *PdFe*. In the case of canonical spin glasses, the most important interaction is a RKKY interaction which decays approximately as r^{-3} with distance, while in the case of *PdFe* the central ferromagnetic part of the interaction decays exponentially. This implies that the range of the ferromagnetic interaction depends only logarithmically (*weakly*) on temperature in the case of *PdFe*, while it depends more strongly on temperature in the case of the canonical spin glasses. We point out that the ratio T_f/T_{sf} is not relevant quantity, but the ratio of the interaction ranges at T_f and T_{sf} is relevant.

In our previous publication,⁸ we have discussed the spin-cluster glass freezing for an ~ 150 ppm Fe sample and pro-

posed a method to estimate the size of the cluster and the internal field B_i . The temperature T_{peak} , at which the ac susceptibility shows a maximum, can be described with

$$T_{\text{peak}} = 4.9N_c \sqrt{B^2 + B_i^2} \text{ (mK/mT)}, \quad (4.5)$$

where an infinitely large spin number and $\mu_{\text{gm}} = 14\mu_B$ [instead of $10\mu_B$ (Ref. 8)] are assumed.¹⁰ B_i represents an internal field and N_c is the size of a cluster. We apply this equation for the 2.5 ppm sample.³⁷ This function with $N_c = 0.5$ and $B_i = 0.4$ mT is plotted as the solid line in the inset of Fig. 6. The very large B_i (compared to $B_i \approx 1$ mT for the 150 ppm sample⁸) indicates a strong anisotropic energy, presumably from the Pd matrix. This strong anisotropic energy may explain that T_f/x for the 2.5 ppm sample is larger than that of the 16 ppm sample. See also the discussions by Herrmannsdörfer *et al.*¹⁰ Although $N_c = 0.5$ seems to be far from 1, we believe that the T_{peak} data are consistent with no clustering for the 2.5 ppm sample in ac susceptibility measurements.

C. Characteristic times

Our model calculation might be criticized to be too crude, since it is based on an all-or-nothing approximation where giant moments are divided into two groups: those isolated, and the others interacting. We, however, argue as follows. The strength of the interactions among spins and the thermal fluctuations must be compared. As long as the interaction is stronger than the thermal fluctuation, one must take the spins as interacting regardless of their strength. This implies that there is a relatively clear cutoff which divides isolated and interacting giant moments.

In order to understand both the resistivity and ac susceptibility behavior, we have to introduce two length scales, r_c and r_{FI} . We justify these two length scales from the difference between the characteristic times of the resistivity and ac

susceptibility. The characteristic time τ_r of the resistivity is a mean free path divided by the Fermi velocity of conduction electrons (of the order of 10^{-14} s),² while the characteristic time τ_m of the ac susceptibility measurements is the inverse of the measurement frequency (of the order of 10^{-2} s in our measurements). The longer the waiting time, the more probable a system is disturbed by phonons. It implies that the stronger an interaction between two impurities is, the longer the correlation time can be expected. Therefore, a stronger condition for giant moments to be correlated in the ac susceptibility is necessary than that in the resistivity measurements: $r_{\text{FI}} < r_c$.

V. SUMMARY

The isolated giant moments associated with Fe impurities in Pd matrix are identified as a *spin fluctuation* system with a spin fluctuation temperature $T_{\text{sf}} = 250 \pm 30$ mK: The ground state of Fe impurity at 0 K is nonmagnetic. We also observe a spin-glass freezing at a temperature which is two orders of magnitude lower than the spin-fluctuation temperature.

We introduce two characteristic length scales in order to understand the resistivity and ac susceptibility behavior. These two different characteristic length scales are based on the very different characteristic times for those measurements: $\sim 10^{-14}$ s for the resistivity and $\sim 10^{-2}$ s for the ac susceptibility.

ACKNOWLEDGMENTS

We wish to thank T. Herrmannsdörfer, S. Rehmann, and H. Uniewski for contributions to the experiments. This work was supported by the Deutsche Forschungsgemeinschaft through Graduiertenkolleg Po 88/13 and Sonderforschungsbereich 279, through the HCM-LSF-Programme of the European Union.

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