Interplay between spin-glass and non-Fermi-liquid behavior in $Y_{1-x}U_xPd_3$

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We report the results of a study of the ac and dc magnetic susceptibilities of $Y_{1-x}U_xPd_3$. The magnetic properties of the system gradually evolve from the spin-glass (SG) behavior observed for $x=0.4$ to the non-Fermi-liquid (NFL) ground state that develops at $x=0.2$. For $x=0.4$, the SG properties are remarkably similar to those found in canonical spin glasses. We have been able to analyze our data in terms of critical slowing down when T approaches the critical temperature associated with a spin-glass freezing phase transition. The values obtained for the dynamic and static critical exponents [$z\nu=10\pm1$, $\beta=0.9(1)$, $\gamma=1.8(2)$] support the existence of a true spin-glass phase transition in $Y_{0.6}U_{0.4}Pd_3$. On the other hand, the evolution of magnetic properties with a decreasing U concentration suggests an interplay between the SG and NFL ground states. The ground state that finally appears for $x=0.2$ displays a temperature dependence of its low-field ac susceptibility consistent with the prediction of the Griffith's phase model of NFL behavior. The role played by compositional disorder in the development of the NFL state is discussed.

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I. INTRODUCTION

The pseudobinary compound $Y_{1-x}U_xPd_3$ displays a rich variety of phenomena, including Fermi-level tuning,[1] a spin-glass state,[2] and the thoroughly studied non-Fermi-liquid (NFL) behavior[2,3] that develops at $x=0.2$. Several models have been proposed to explain the unconventional thermal, electrical, and magnetic properties of the NFL state detected for $x=0.2$. We will mention first the multichannel Kondo model,[4] used by Seaman *et al.*,[2] which explains their experimental results in terms of a two-channel quadrupolar Kondo effect. Almost coetaneous with the former, the quantum critical point model (QCP) model proposes a quantum phase transition that takes place at $T=0$ as the source of the NFL behavior.[3] Other authors have presented alternative explanations, including the Kondo disorder model[5] and the proposal of a Griffith's phase that develops at low temperatures.[6] Such models emphasize the role of structural and compositional disorder that is almost unavoidable in the U- and Ce-based intermetallics and alloys. Nevertheless, all the reported NFL systems are close to points of the phase diagram where the critical temperature of a magnetic instability tends to zero, i.e., to quantum critical points. Thus, the relationship between the actual character of this magnetic phase and the onset of NFL behavior has been discussed in several papers.[7,8] Moreover, the spin-glass state observed in some NFL systems has itself been the subject of attention.[9,10] Wu *et al.* analyzed their μ SR data in the theoretical framework of the mean-field Sherrington-Kirkpatrick model, concluding that above $x=0.3$, the spin-glass behavior can be understood in terms of induced moments.[11] In this work we focus on the case of $Y_{1-x}U_xPd_3$ in the concentration range between $x=0.2$ and 0.45. We present a detailed study of the dc and ac magnetic susceptibility, with the aim of ascertaining the existence of a possible spin-glass phase transition, following the spirit of mean-field models for spin-glass systems. The relevance of our results with respect to the actual mechanism that leads to the development of the NFL ground state at $x\approx0.2$ is discussed.

II. EXPERIMENTAL

Polycrystalline samples of $Y_{1-x}U_xPd_3$ ($x=0.2$, 0.3, and 0.4) were prepared by arc-melting high-purity materials in an argon atmosphere. The ingots were melted and flipped over several times to increase homogeneity, and subsequently annealed at 900 °C for 1 week. Powder x-ray-diffraction patterns did not show evidence of secondary phases. The sample composition, determined using x-ray energy-dispersive spectroscopy (XEDS), was found to be very close to the nominal value, consistent with the negligible losses produced during the melting procedure. dc magnetization and ac susceptibility measurements were performed for different dc fields and frequencies in the temperature range between 2 and 25 K, using Quantum Design SQUID magnetometers (dc measurements), a Quantum Design MPMS system (ac measurements), and a MagLabExa from Oxford Instruments (ac and dc measurements).

III. RESULTS

In Fig. 1 we present the results of selected dc magnetic susceptibility curves ($\chi=M/H$) measured for $Y_{0.6}U_{0.4}Pd_3$ and $Y_{0.7}U_{0.3}Pd_3$ in the zero-field-cooled (ZFC) and field-cooled (FC) regimes. They show the features typically related to a spin-glass-like state developing below a freezing temperature T_f. From the results shown in Fig. 1, which show strong irreversibilities between the ZFC and FC curves for both materials, T_f is easily determined considering $T_f\cong T_{irr}$ (low field); T_f values of 9.6 and 4.6 K are obtained

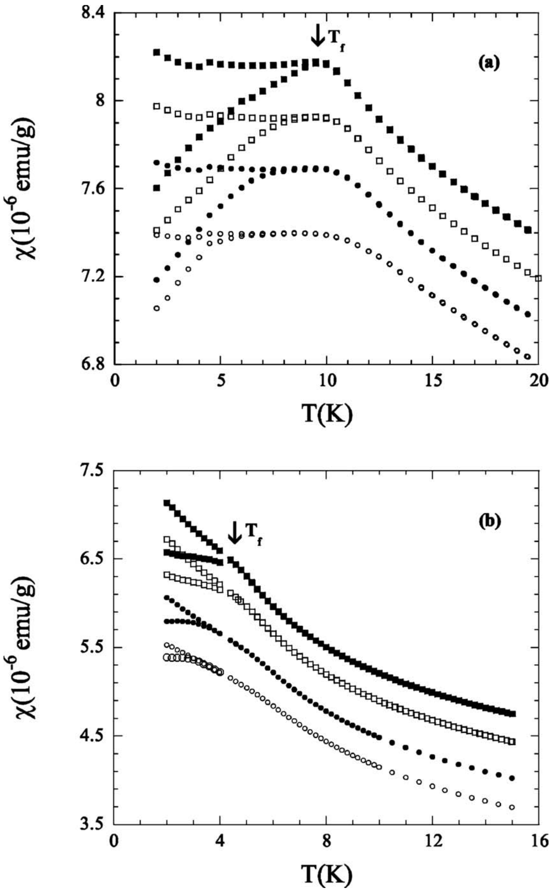

FIG. 1. (a) dc magnetic susceptibility ($\chi = M/H$) vs T of $Y_{0.6}U_{0.4}Pd_3$ shown for selected values of the applied field: 1 kOe (top); 5, 10, and 30 kOe (bottom). The 5, 10, and 30 kOe curves have been shifted down by amounts of 0.2×10^{-6}, 0.4×10^{-6}, and 0.6×10^{-6} emu/gram. (b) dc magnetic susceptibility ($\chi = M/H$) vs T of $Y_{0.7}U_{0.3}Pd_3$ for selected values of the applied field: 0.1 kOe (top), 1, 5, and 10 kOe (bottom). The 1, 5, and 10 kOe curves have been shifted down by amounts of 0.3×10^{-6}, 0.6×10^{-6}, and 0.9×10^{-6} emu/gram. For both samples, we show data measured in the ZFC and FC regimes. The arrows mark T_f=9.5 K ($Y_{0.6}U_{0.4}Pd_3$) and T_f=4.6 K ($Y_{0.7}U_{0.3}Pd_3$).

for x=0.4 and 0.3. In the case of $Y_{0.6}U_{0.4}Pd_3$, in Fig. 1(a) we display part of our previous results in this material (see Ref. 10). As we pointed out in that work, the static magnetic properties of $Y_{0.6}U_{0.4}Pd_3$ are similar to those of canonical spin glasses, although the spin-glass state seems to be remarkably robust against the action of magnetic field,[10] a peculiarity that has been reported for other U-based spin-glass systems.[12] As the uranium concentration approaches the value x=0.2, the freezing temperature tends to zero, whereas the peak displayed by $\chi(T)$ and the irreversibility between the ZFC and FC branches are progressively broadened and reduced, as shown in Fig. 1(b) for $Y_{0.7}U_{0.3}Pd_3$. All the spin-glass-like features practically disappear for uranium concentrations close to 0.2 (see Ref. 9), and a non-Fermi-liquid ground state develops instead.[2]

In Figs. 2 and 3 we present the results of our ac susceptibility measurements. Figure 2(a) displays the frequency-dependent maximum of the real (in-phase) component χ' for

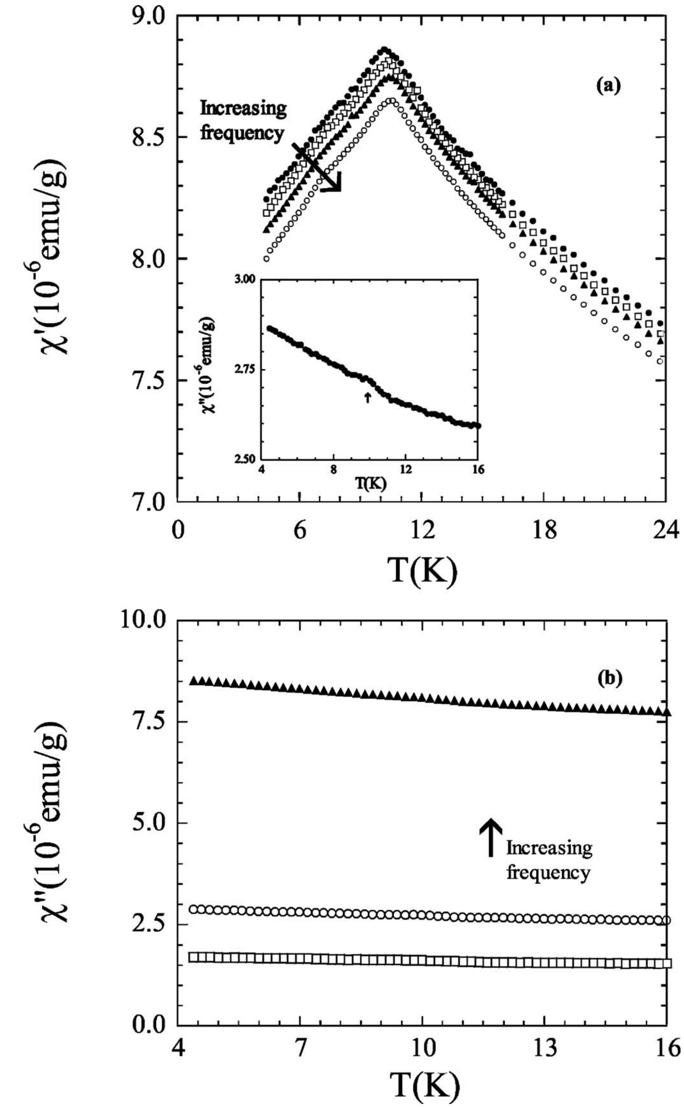

FIG. 2. (a) Real component (χ') of the ac susceptibility of $Y_{0.6}U_{0.4}Pd_3$ for frequencies of 66, 111, 666, and 1000 Hz. Inset: imaginary component (χ''), measured for 111 Hz. (b) χ'' measurements in a broader temperature range and for frequencies of 66, 111, and 333 Hz. All the experiments shown were performed with an applied ac field of 10 Oe.

$Y_{0.6}U_{0.4}Pd_3$, which is the other typical signature of spin-glass behavior. On the other hand, the imaginary (out-of-phase) component χ'' [Fig. 2(b)] shows a weak anomaly at T_f, clearly visible only for low frequencies [see the inset of Fig. 2(a)]. This last feature is superimposed on a frequency-dependent contribution that is also present for x=0.3 and 0.2 (see Figs. 3 and 4). For $Y_{0.7}U_{0.3}Pd_3$, there is a shoulder in χ' instead of a peak, as was found in the dc measurements, although in this case a clear jump in the imaginary component χ'' is observed for certain frequencies. Once again, this jump is superimposed on a frequency-dependent contribution (see the inset of Fig. 3).

Finally, in Fig. 4 we present results of ac susceptibility measurements for $Y_{0.8}U_{0.2}Pd_3$, which do not exhibit the signatures of spin-glass behavior or magnetic order in the temperature range of our measurements. As in the two other

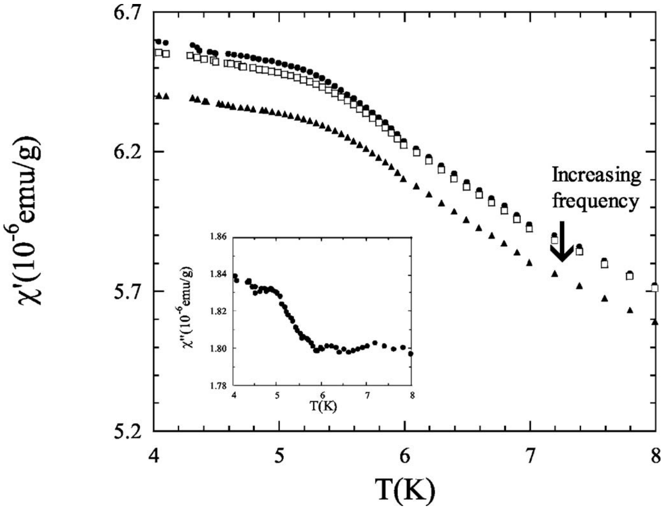

FIG. 3. Real component (χ') of the ac susceptibility of $Y_{0.7}U_{0.3}Pd_3$ measured between 4 and 8 K in a 1 Oe ac applied field for frequencies of 100, 300, and 1000 Hz. The inset shows χ'' for a frequency of 100 Hz.

compositions, a dramatic frequency dependence of the dissipative component χ'' is observed (see the inset of Fig. 4). Actually, in the range of frequencies studied in this work, χ'' is directly proportional to the frequency. We have found other examples of a similar behavior in the heavy fermion antiferromagnetic systems $CeNi_2Sn_2$ (Ref. 13) and $U_{1-x}Y_xRu_2Si_2$ (Ref. 14). As we will show below, it is due to a contribution to the susceptibility arising from currents induced in the sample by the alternating magnetic field.

IV. DISCUSSION

In view of the results outlined above, the static and dynamic magnetic properties can be interpreted in terms of an

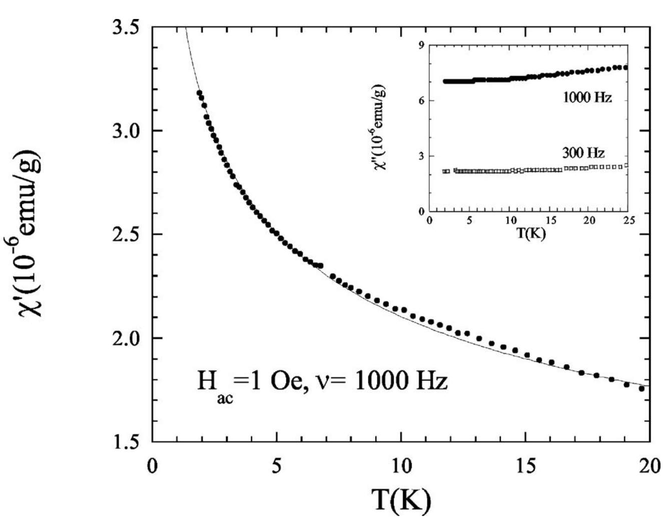

FIG. 4. Real component (χ') of the ac susceptibility of $Y_{0.8}U_{0.2}Pd_3$ measured between 2 and 25 K in a 1 Oe ac applied field for a frequency of 1000 Hz; the solid line is a fit of the data to the expression $\chi'(T)=\chi_0 T^{-1+\lambda}$. The inset shows χ'' for frequencies of 300 and 1000 Hz.

interplay between SGL and NFL behavior. Focusing first on the $Y_{0.6}U_{0.4}Pd_3$ sample, its static magnetic properties are very similar to those of canonical spin-glass behavior. In a previous paper,[10] we reported the observation of a strong increase of the nonlinear susceptibility coefficients (b_3 and b_5), which show a peak right at T_f. The observation of a divergence of these coefficients when the temperature approaches T_f is usually interpreted as a proof of the existence of a true thermodynamic phase transition.[15] On the other hand, the ac magnetic susceptibility displays a particular frequency dependence that is only partially related to the spin-glass behavior. As it was already pointed out above, the frequency dependence of the maximum in χ' is as expected for a spin glass. The frequency shift is usually quantified as $p=\Delta T_f/T_f\Delta(\log\omega)$, the relative variation of the freezing temperature per frequency decade. In our case, it takes a value of about 0.02, which is much smaller than the values typically found for superparamagnetic systems, and comparable to those usually observed in real spin-glass systems.[15] However, we have already pointed out that the imaginary component χ'' only displays a modest feature at T_f, superimposed on a frequency- and temperature-dependent background. The magnitude of the background contribution to χ'' shows a linear dependence on frequency. This is just the predicted behavior of χ'' in terms of the classical theory of the propagation of electromagnetic waves in conducting media.[14,16] Following Očko *et al.*,[14] if the measurements were performed in the low shielding limit, as is the case here, the dissipative component of the ac susceptibility should exhibit a contribution coming from the conduction electrons that is proportional to (ν/ρ), where ρ is the electrical resistivity of the sample

$$\chi'' \approx k\left(\frac{\nu}{\rho}\right). \quad (1)$$

The same theory predicts also a frequency-dependent *negative* contribution to the real component, given by

$$\chi' \approx -\frac{4}{3}k^2\left(\frac{\nu}{\rho}\right)^2. \quad (2)$$

The coefficient k is proportional to the cross section of the sample. Our present measurements were performed on the same samples ($\sqcup 3\times 3\times 8\ \mathrm{mm}^3$) previously used for dc magnetization and thermal expansion measurements.[10,17] This explains also the anomalous frequency dependence of χ' that is observed even above T_f (see Fig. 2). To extract the genuine spin-glass contribution to χ'', we calculate the conduction electron contribution using Eq. (1) and the results of our electrical resistivity measurements performed on the same sample [Fig. 5(b)]. The coefficient k can be calculated from the data at 25 K. This means that we assume that there is no spin-glass contribution to χ'' at that temperature, which is a reasonable hypothesis. Using data corresponding to measurements taken with frequencies of 66, 111, and 333 Hz, we obtain k values of 3.37×10^{-12}, 3.39×10^{-12}, and 3.38×10^{-12} emu Ω cm/gHz. In Fig. 5(a) we display our experimental and calculated values of χ'' vs T for 111 Hz. Well above the freezing temperature, the agreement is perfect,

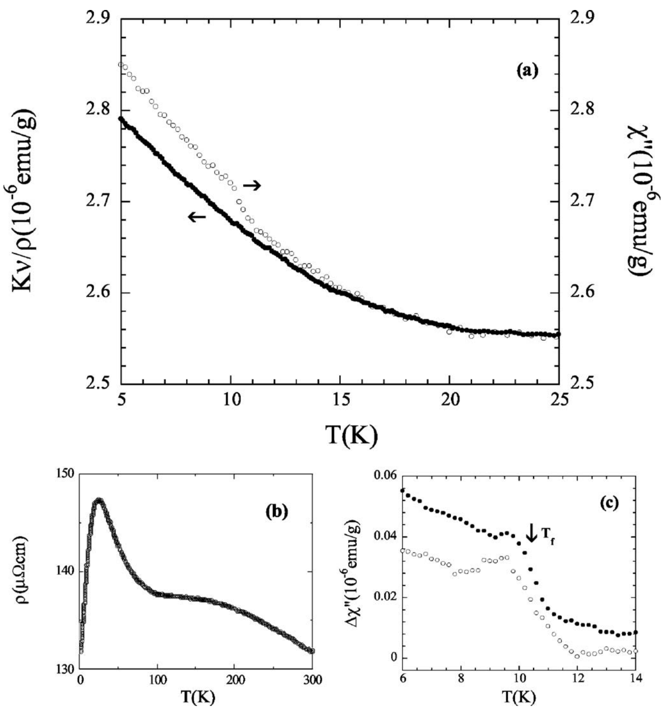

FIG. 5. (a) Calculated conduction electron contribution $(k\nu/\rho)$ and experimental χ'' results for the $Y_{0.6}U_{0.4}Pd_3$ sample measured with 111 Hz (see text). (b) Electrical resistivity measured on the same $Y_{0.6}U_{0.4}Pd_3$ sample. (c) Spin-glass contribution $\Delta\chi''=\chi''-k\nu/\rho$ extracted from the experimental data for 66 (open circles) and 111 Hz (closed circles).

while the curves separate from each other below $T\sqcup 15$ K. Subtracting the calculated χ'' from the experimental data, we obtain the spin-glass contribution to χ'', shown in Fig. 5(c), for the cases of $\nu=66$ and 111 Hz. Notice the similar behavior observed for both frequencies. As expected for spin-glass systems, a step feature appears below T_f; the maximum slope of $\Delta\chi''(T)$ is assumed to occur at $T_f(\omega)$. In our case, $T_f\sim 10.4$ K for both frequencies. The jump in $\Delta\chi''(T)$ signals the onset of dissipative phenomena associated with the magnetic freezing transition. We note that in the case of $Y_{0.7}U_{0.3}Pd_3$, the experiments were performed on a smaller sample, and thus the influence of the frequency-dependent contribution was less significant, although the data were somewhat noisier.

As the results of static and dynamic susceptibility measurements in $Y_{0.6}U_{0.4}Pd_3$ are so similar to those observed in canonical spin glasses, we have attempted an analysis of the results of both sets of measurements in terms of the Sherrington-Kirkpatrick (SK) model for spin-glass behavior.[18] This is a mean-field theory, which assumes that a second-order phase transition takes place at a critical temperature T_c close to the maximum of the FC and ZFC magnetization curves and the low-frequency limit of T_f obtained from ac susceptibility measurements. A true phase transition at T_c should reveal itself in the form of characteristic divergences of different quantities, with critical exponents having values that are predicted theoretically.[19]

In principle, it should be possible to obtain the spin-glass order parameter $q(T)$ and the static critical exponent β from the susceptibility, since in the framework of the SK model,[18]

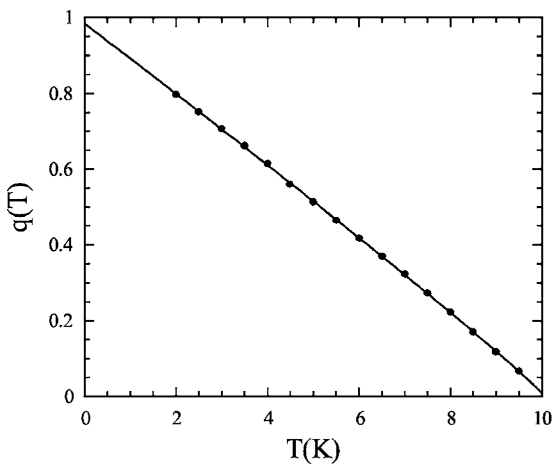

FIG. 6. Spin-glass order parameter $q(T)$ computed using Eq. (1) and the FC data measured for $Y_{0.6}U_{0.4}Pd_3$ with an applied magnetic field $H=1000$ Oe.

$$\chi(T) = C(T)[1-q(T)]\{T-\theta(T)[1-q(T)]\}^{-1}. \qquad (3)$$

Below T_c, the order parameter q should follow a power law dependence $q(T)\propto(T_c-T)^{\beta}$. The value predicted by the SK theory for the critical exponent β is 1.

On the other hand, the frequency dependence of the maxima observed in the ac susceptibility can be used to obtain the dynamic coefficients *via* the critical slowing down (CSD) law for the relaxation time

$$\tau=\frac{1}{\nu_{\max}}=\tau_0\left(\frac{T_{\max}}{T_c}-1\right)^{-z\nu}. \qquad (4)$$

For conventional three-dimensional (3D) spin glasses, $z\nu$ values around 10 are usually obtained from fits to the CSD law.

In Fig. 6 we present the spin-glass order parameter $q(T)$ computed using Eq. (1) and the FC data for $H=1000$ Oe. The parameters $C(T_c)$ and $\theta(T_c)$ were determined from a fit of the data above T_c to the expression $\chi(T)=\chi_0+C/T-\theta$. Finally, a fit of the computed $q(T)$ data to $q(T)\propto(T_c-T)^{\beta}$ gives $T_c=10$ K and $\beta=0.94$. In Fig. 7 we display a fit of $\nu(T_{\max})$ in terms of a CSD law, from which we deduce $z\nu=10\pm1$ and $T_c=9.6\pm0.1$ K.

In order to justify these results, obtained from the SK model [Eq. (3)], and using extrapolations that could bias the results towards mean-field theory, we have used the data reported in Ref. 10 to perform a supplementary static scaling analysis of the nonlinear susceptibility,[20] defined as

$$\chi_{dc}(T)=\chi_0(T)+\chi_{nl}(T)=\chi_0(T)-b_3\chi_0^3H^2+b_5\chi_0^5H^2+\cdots . \qquad (5)$$

If the nonlinear susceptibility shows the divergent behavior expected for a spin-glass transition, critical exponents γ and β can be obtained by means of the universal scaling equation[15,20,21]

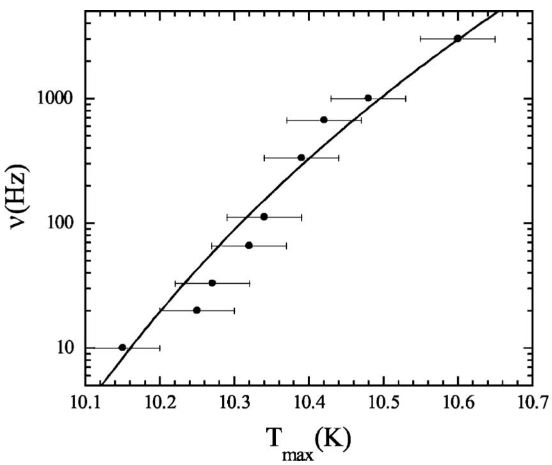

FIG. 7. Fit of ν (T_{max}) in terms of the CSD law, yielding $z\nu=10\pm1$ and $T_c=9.6\pm0.1$ K.

$$\chi_{nl} = t^{\beta}\Phi\left(\frac{H^2}{t^{\beta+\gamma}}\right), \tag{6}$$

where $t=(T-T_c)/T_c$ is the reduced temperature. In Fig. 8 the universal curve obtained from our experimental results (Ref. 10) is displayed as a log-log plot. Due to rounding effects close to T_c, and the scatter of data far away from T_c, only data in the temperature range $1.1T_c<T<1.9T_c$ and fields above 5 kOe were used for this plot. A reasonable overlap was obtained with $T_c=9.8(2)$, $\beta=0.9(1)$, and $\gamma=1.8(2)$, to be compared with $\beta=0.94$ obtained in the previous analysis. These values of the static coefficients are in good agreement with the experimental results typical of canonical *metallic* spin glasses, such as CuMn and AgMn ($\beta=1$, $\gamma=2.2$) or PdMn ($\beta=0.9$, $\gamma=2$) (Ref. 22). Moreover, the curve approaches the asymptotic slope $\beta/\gamma+\beta$ predicted from Eq. (6), clearly supporting the existence of a true spin-glass phase transition in $Y_{0.6}U_{0.4}Pd_3$.

A similar complete analysis of the dc and ac magnetic susceptibility could not be performed for $Y_{0.7}U_{0.3}Pd_3$, due to the smearing of the spin-glass features observed in that sample. Nevertheless, a simple inspection of the dc magnetization reveals that some of the typical spin-glass features have disappeared. There is no plateau in the FC magnetization below the freezing temperature, and the peak at T_f is barely observed. The irreversibility between the FC and FC branches is also much less marked. Even the frequency dependence of the freezing temperature is difficult to quantify from the ac susceptibility measurements, as can be readily observed in the figures. We have estimated a value of $p\leqslant0.037$, considering $T_f=5.2$ K, and $\Delta T_f\leqslant0.2$ K per decade in frequency. In any case, we extracted the nonlinear contribution to the dc susceptibility following the method previously used by us for $Y_{0.6}U_{0.4}Pd_3$ (see Ref. 10). The results are displayed in Fig. 9. Only an inflection in the b_3 coefficient is observed at 5.3 K, consistent with the results of the ac and dc susceptibility measurements. However, it appears as if the nonlinear susceptibility comprises two contributions, one of them the spin-glass term responsible for the change of slope at T_f, and the other a background contribution that decreases monotonically with temperature. The dc and ac *linear* susceptibilities [Figs. 1(b) and 3] can be interpreted in a similar way.

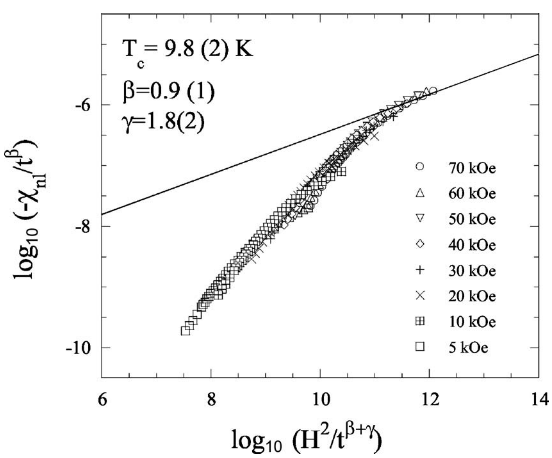

FIG. 8. Nonlinear susceptibility χ_{nl} for $Y_{0.6}U_{0.4}Pd_3$ obtained from data reported in Ref. 10 and analyzed according to Eq. (6). The solid line represents the asymptotic slope $\beta/\gamma+\beta$ in the limit $t\rightarrow0$.

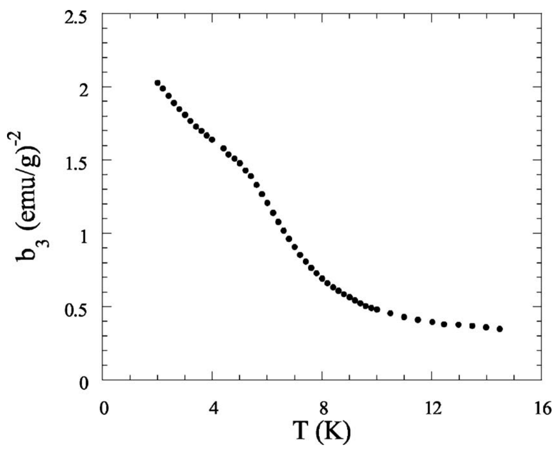

FIG. 9. Nonlinear coefficient b_3 vs T of $Y_{0.7}U_{0.3}Pd_3$ computed from the FC magnetization data shown in Fig. 1(b).

Thus, it seems that, at least well above $x=0.2$, the magnetic properties of the $Y_{1-x}U_xPd_3$ system can be described in terms of a conventional spin-glass state, with distinct evidence for the possibility of a true thermodynamic phase transition. As we approach $x=0.2$, additional contributions to the magnetic properties enhance their role at the expense of the spin-glass features, which finally disappear for $x=0.2$. It is well known that in the $Y_{1-x}U_xPd_3$ system the compositional disorder due to local fluctuations in U concentration increases as the x approaches $x=0.2$ from above.[23] This compositional disorder is a logical candidate for the destabilizing factor of the robust spin-glass phase transition detected at $x=0.4$ and the source of the NFL behavior.[24] What we could be observing in our system is the formation, with decreasing U concentration, of a magnetically inhomogeneous state in which there is interplay between a spin-glass phase and a nonmagnetic phase. This latter phase reveals itself through the additional contributions detected in the magnetic properties. We recall the background contribution to the nonlinear susceptibility found for $x=0.3$. All NFL systems, including $Y_{0.8}U_{0.2}Pd_3$, certainly show a nonlinear contribution to the susceptibility, whose origin has been the subject of some attention.[25,26] The Griffith's phase model makes a precise prediction about the temperature dependence of this nonlinear contribution.[6] If our interpretation is correct, the background contribution found for $x=0.3$ would be no more than a precursor of the NFL properties fully developed for $x=0.2$.

It is tempting to relate the NFL properties observed in this system to the progressive disorder-induced weakening of the spin-glass transition observed for $x=0.4$, giving rise to a Griffith's phase as x approaches the quantum critical point at 0.2, as has already been suggested by different authors.[6,27] We point out that the results of our ac susceptibility measurements in $Y_{0.8}U_{0.2}Pd_3$ can be analyzed in terms of the Griffith's phase model, which predicts[6]

$$C/T \propto \chi(T) = \chi_0 T^{-1+\lambda}. \tag{7}$$

The solid line in Fig. 4 is a fit of our ac susceptibility results to that equation, from which we obtain $\lambda=0.77$, in comparison with $\lambda=0.75$ and 0.7 reported by De Andrade *et al.* from specific heat and dc magnetic susceptibility measurements.[27] Notice that Eq. (7) was originally derivated for the case of a Griffith's phase developing in a *quantum spin glass*,[28] what we believe is quite appropriate for the case of the $Y_{1-x}U_xPd_3$ system.

To illustrate this point further, we show in Fig. 10 the results of our own dc susceptibility measurements performed on the same $x=0.2$ sample, with applied fields of 1 and 10 kOe. Within our experimental temperature range, both curves are fully reversible, and can be fitted to $\chi(T)=\chi_0 T^{-1+\lambda}$, with slightly different values of the λ coefficient ($\lambda=0.79$ for 1 kOe and $\lambda=0.80$ for 10 kOe). The 10 kOe curve reveals a nonlinear contribution to the susceptibility. Following the same method used by Vollmer *et al.* in UCu_4Pd (Ref. 26), we have computed this contribution as $\chi_{nl}=(M/H)_{1\ \mathrm{kOe}}-(M/H)_{10\ \mathrm{kOe}}$. The results are shown in Fig. 10, and attached to them a fit to the expression $\chi_{nl}=\chi_{nl_0}+\chi_1 T^{-2.21}$. The Griffith's phase model predicts $\chi_{nl}\propto T^{-3+\lambda}$, from which we conclude that the temperature dependence of the nonlinear susceptibility of $Y_{0.8}U_{0.2}Pd_3$ can also be reasonably explained in terms of this model.

V. CONCLUSIONS

In summary, we have performed an experimental study of the static and magnetic properties of the $Y_{1-x}U_xPd_3$ system between $x=0.2$ and 0.4. The evolution of the magnetic properties points to an interplay between a genuine spin-glass state (at $x=0.4$) and the NFL behavior (at $x=0.2$) induced by disorder. Between them, at $x=0.3$, the magnetic properties show the coexistence of contributions from both states. The contribution that we relate to the emerging NFL behavior grows at the expense of that associated with the SG state. Finally, the SG state disappears at $x=0.2$. Our low field ac susceptibility measurements, dc linear and nonlinear susceptibility for $Y_{0.8}U_{0.2}Pd_3$, can be adequately interpreted in terms of a disorder-induced Griffith's phase, which develops close to the spin-glass quantum critical point at $x=0.2$. Further experimental work is underway to monitor with more detail the evolution of the low-temperature magnetic properties, especially the nonlinear magnetic susceptibility, in the region around this last U concentration.

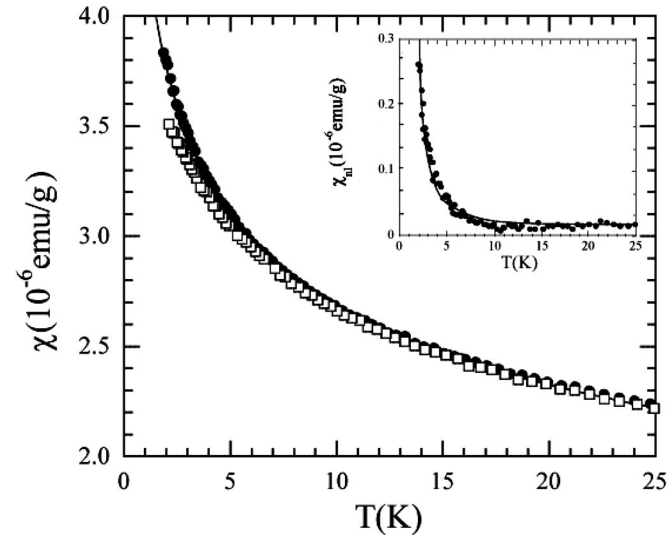

FIG. 10. dc magnetic susceptibility ($\chi=M/H$) vs T of $Y_{0.8}U_{0.2}Pd_3$ measured in applied fields 1 kOe (open squares) and 10 kOe (closed circles). The solid line is a fit of the 1 kOe data to a law $\chi(T)=\chi_0 T^{-1+\lambda}$ ($\lambda=0.79$). The inset shows $\chi_{nl}=(M/H)_{1\ \mathrm{kOe}}-(M/H)_{10\ \mathrm{kOe}}$ and a fit to $\chi_{nl}=\chi_{nl_0}+\chi_1 T^{-2.21}$.

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