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MAGNETIC FIELD DEPENDENCE OF THE SUSCEPTIBILITY AND SPECIFIC HEAT IN A \((\text{La}_{0.92}\text{Gd}_{0.08})\text{Al}_2\) SPIN GLASS

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Résumé.- La chaleur spécifique magnétique linéaire, \(C_m = \gamma T\), de \((\text{La}_{0.92}\text{Gd}_{0.08})\text{Al}_2\) est réduite par un champ magnétique \(H\). Pour \(H \gtrsim 1\) tesla, \(\gamma(H)\) est expliqué à l'aide du modèle RKKY. La forte diminution de \(\gamma\) observée dans des champs faibles (\(H \lesssim 0.1\) tesla) est comparée à la forte réduction de la susceptibility et attribuée au blocage de nuages superparamagnétiques.

Abstract.—The linear magnetic specific heat, \(C_m = \gamma T\), of \((\text{La}_{0.92}\text{Gd}_{0.08})\text{Al}_2\) is reduced in a magnetic field \(H\). For \(H \gtrsim 1\) tesla, \(\gamma(H)\) is explained within the RKKY model. The steep decrease of \(\gamma\) already in small fields (\(H \lesssim 0.1\) tesla) is compared to the strong depression of the susceptibility and interpreted as being due to the blocking of superparamagnetic clouds.

The predominant features of spin glasses, the "cusp" in the low field susceptibility \(\chi\) and onset of remanence below a temperature \(T_f\) show up in magnetic measurements. A phenomenological description \(1/1\) invokes the existence of superparamagnetic clouds which are blocked below \(T_f\): the blocking of a few clouds with large moments can account for the magnetic effects. Calorimetric investigations have not revealed any 'incident' in the temperature variation of the zero field magnetic specific heat \(C_m\). They show a rather large contribution already well above \(T_f\) as well as a characteristic law \(C_m = \gamma T\) near \(T_f\) and down to lowest temperatures. This suggests that only a few degrees of freedom can be connected with the blocking mechanism, the majority being related to the internal formation of clouds.

The (\(\text{La}_{1-c}\text{Gd}_c\))\text{Al}_2 system has been shown to be a spin glass through previous investigations of \(\chi\), \(1/2\) and \(C_m\) \(3/3\). Here we present a comparative study for a \(c = 8\%\) single crystal in fields up to 5 tesla with particular emphasis on the low field region.

The main results on \(\chi\) are reported in figure 1: (i) the susceptibility maximum \(\chi_{\text{max}}\) is decreased by 85\% in a field of 0.1 tesla and (ii) \(T_f\) is displaced from 2.3 K to 1 K. In the model of superparamagnetic clouds \(1/1\), we interpret this as the clouds being progressively locked in as \(H\) is increased, most of the clouds are blocked in a field of 0.1 tesla, i.e. their relaxation times become larger than the measuring time of \(\chi\) (\(\sim 10^{-1}\) s).

\[\begin{align*}
\text{Fig. 1 : Susceptibility maximum } \chi_{\text{max}} \text{ and temperature } T_f \text{ of } \chi_{\text{max}} \text{ vs. } H. \text{ Lines are intended as visual guide.}
\end{align*}\]

The magnetic specific heat \(C_m\) per impurity spin, obtained by subtraction of the matrix contribution, varies linearly with \(T_f\), as found previously for this system \(3/3\). This holds from the lowest accessible temperature 0.4 K up to \(\sim 4\) K. The two important results we wish to report are: (i) in the presence of a magnetic field, the linear \(C_m\) dependence on \(T\) is not altered up to \(H = 5\) T (ii) \(\gamma\) decreases with \(H\), hence all data can be described by the equation

\[\begin{align*}
\chi \text{ work performed within the research program of Sonderforschungsbereich 125 Aachen-Jülich-Köln}
\end{align*}\]
\[ C_\gamma (H, T) = y(H) T^2 \] where \( a = 1.00 \pm 0.05 \) as determined from log-log plots. The experimental \( y(H) \) values divided by \( k_B \) are shown in figure 2. In a field of 5 tesla, \( y \) is reduced by 50%. However, a strong initial decrease in as low as 0.1 tesla affects approximately 10% of \( y \).

We can compare the experimental \( y(H) \) curve with the coefficient \( y_{\text{RKKY}} \) of \( C_\gamma \) at low temperatures which was calculated from the contribution of small clusters to the free energy in the scaling limit of the dilute RKKY model /4/: 

\[ \gamma_{\text{RKKY}} (H) = \frac{k_B^2}{6 c W_1} \Re \left( \left( 2 S + 1 + i \frac{g_H \mu_B^H}{\pi c W_1} \right) - \psi \left( 1 + i \frac{g_H \mu_B^H}{\pi c W_1} \right) \right) \]  

(1)

Here \( \psi \) is the digamma function, \( S \) the spin value and \( W_1 = \frac{4}{3} V_0 / v \) an energy proportional to the RKKY amplitude \( V_0 \) divided by the atomic volume \( v \). We first note that from eq. (1) the area under the \( y(H) \) curve should depend only on \( S \) and \( g \): 

\[ \int_0^\infty \gamma_{\text{RKKY}} (H) dH = \frac{\pi^2 k_B^2 S}{6 g_H \mu_B^H} = 2.46 \frac{k_B^S}{g} \text{ tesla} K^{-1} \] 

The area under the measured \( y(H) / k_B \) curve, including a long tail as suggested by the upward curvature of the last points, cf. figure 2, is estimated roughly as 2.4 tesla \( K^{-1} \) against 4.3 obtained from eq. (2) with \( S = 7/2 \) and \( g = 2 \). The order of magnitude agreement supports that the magnetic specific heat is indeed dominated by the freezing of degrees of freedom within small RKKY clusters.

We can now try a fit to our data simply by scaling \( \gamma_{\text{RKKY}} (H) \), eq. (1), with a reduction factor 0.55 to account for the numerical discrepancy in the areas. The solid curve in figure 2 represents \( \gamma_{\text{RKKY}} (H) = 0.55 y_{\text{RKKY}} (H) \) for \( W_1 / k_B = 13.4 \). For \( H \geq 0.5 \) tesla, \( \gamma_{\text{RKKY}} (H) \) describes the experimental points quite well. At high fields, i.e. \( g_H \mu_B^H \gamma (2 S + 1) c W_1 \), \( \gamma_{\text{RKKY}} (H) \) shows a long tail with a \( H^{-2} \) decay. This tail is due to the Lorentzian wings in the RKKY energy spectrum. The quantitative disagreement in the areas as well as the values for \( W_1 \) determined from different experiments (\( V_0 \) as obtained by high field magnetization /2/ yields \( W_1 / k_B = 3.5 \) K) is probably due to three factors: (i) the Gd concentration is rather high (8%) (ii) for the calculation of \( V_0 \) from \( M \) only the second virial approximation was used /5/ (iii) high energy cut-offs are present in the spectrum /6/. We estimate \( H_{1a} \approx 0.5 \) tesla and \( H_{1c} \approx 12 \) tesla for the antiferromagnetic and ferromagnetic cut-offs, respectively. The asymmetry is caused by a ferromagnetic first and second neighbor RKKY interaction in this system /2/. Qualitatively, the presence of cut-offs leads to a rapid decay of \( y(H) \) in the range of corresponding external fields.

At low fields, \( H \ll 0.5 \) tesla, \( y_{\text{RKK}} \) varies smoothly \( \sim H^2 \) and cannot account for the strong initial variation of the experimental \( y(H) \) curve. One can empirically describe the "excess" above the RKKY "background" by \( \Delta y(H) / k_B = \gamma (H) - \gamma_{\text{RKKY}} (H) / k_B = 0.033 K^{-1} (t - (H/1 \text{ tesla})^3) \) for \( H \lesssim 1 \) tesla (see dotted line in figure 2). This anomalous field dependence with a half width of 0.1 tesla suggests that \( \Delta y(H) \) is related to the same irreversible properties of spin glasses which also govern the \( \chi(H) \) behavior discussed above.

One then might think that \( \Delta y(0) \) is due to those degrees of freedom associated with the turning of superparamagnetic clouds between two nearly degenerate blocking positions. The observed linear \( \Delta y T \) term would imply a constant density \( n(2) \) of their energy splittings. On the other hand, the strong reduction of \( \Delta y(H) \) even for small fields \( (g_H \mu_B^H \ll k_B T) \) could be due to a strong increase of the relaxation times in the presence of \( H \). Although the argument concerning the origin of \( \Delta y(H) \) is somewhat speculative it yields a size of clouds which is in qualitative agreement with that determined from...
the remanent magnetization of spin glasses /1/. Multiplying the density of these "superparamagnetic excitations", \( n(E) = \frac{3\gamma(0)}{s^2 k_B^2} \), by the half width of 0.1 tesla, we estimate a corresponding fraction of \( \sim 10^{-3} \) degrees of freedom of the total magnetic entropy. Since each Gd spin has 8 degrees of freedom this corresponds to \( \sim 10^2 \) spins in a superparamagnetic cloud.

In conclusion, our estimate of the small fraction of degrees of freedom associated with the blocking of superparamagnetic clouds is based on the observation of two distinct characteristic field scales in the coefficient \( \gamma(H) \). This estimate should be valid quite independently of the detailed physical picture for the blocking.

References

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