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## ESR in a Heavy-Fermion Alloy (UBe<sub>13</sub>) Doped with Local Moments

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We have determined the ESR properties of UBe<sub>13</sub> doped with dilute local moments of Er, Dy, or Gd over the temperature region where there is a large variation in the enhanced specific heat. We find that neither the enhancement, the temperature variation, nor any other anomalous behavior appears to be mirrored in the ESR data. We suggest that this unexpected result must be incorporated into current models of heavy-fermion systems.

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At cryogenic temperatures the heavy-fermion systems are characterized by an anomalously large specific heat which is interpreted as the consequence of a greatly enhanced and temperature-dependent density of states,  $\rho_c$ .<sup>1</sup> We have sought further insight into the nature of this enhancement by a study of the temperature dependence of electron-spin-resonance (ESR) measurements of UBe<sub>13</sub> doped with Dy, Er, or Gd.

The Dy and Er moments are certainly unbottlenecked,<sup>2</sup> and were the local-moment-conduction-electron system characteristic of a simple metal host (such as Ag),<sup>3</sup> the ESR line would have a  $g$  shift proportional to  $J\chi_p$ , and the spin-relaxation rate would be given by the usual Korringa relation<sup>4</sup>  $1/\gamma T_1 = \pi/h(\rho J)^2 kT$ . Here,  $\chi_p$  is the electronic susceptibility,  $J$  is the exchange constant,<sup>5</sup> and  $\rho$  is the density of states at the Fermi level in the host metal. In a heavy-fermion system where there may be narrow bands of width comparable to  $kT$ , and other significant differences from a simple metal host, we would expect that the Korringa relation might no longer be appropriate, but that some aspect of the enhanced  $\rho_c$  would be manifested in the temperature dependence of the ESR properties. Measurements of the temperature dependence of the nuclear  $T_1$  in UBe<sub>13</sub> have recently been reported by MacLaughlin *et al.*<sup>6</sup> They find that there is a dramatic enhancement of the Be relaxation rate ( $\cong 10^4$  compared to pure Be), with clear deviations from a Korringa relation over the temperature range between 1 and 10 K. Since both the rare-earth local-moment and the Be-nucleus interactions with the conduction electrons are described by the same form of contact interaction, one might expect to find similarly an anomalous temperature dependence in the observed ESR. From the experimental results to be described, we have concluded that whatever the

mechanism is that results in the appreciable temperature dependence of  $\rho_c$  (as deduced from the specific heat), it does not appear to be manifested in the spin-relaxation rate of the substituted local moments. We believe that this observation provides an important additional criterion for clarification of possible models for the heavy-fermion systems, as is discussed in the following paper.<sup>7</sup>

In Fig. 1 we present both the  $g$  value and the spin-relaxation rate,  $1/\gamma T_2$ ,<sup>8</sup> as a function of temperature for powdered UBe<sub>13</sub> doped with  $\sim 1\%$  of Er. The spectrometer frequency was 9.3 GHz.<sup>9</sup> As can be seen, the relaxation rate is well represented by the usual form,  $1/\gamma T_2 = A + BT$ , where  $A$  and  $B$  have the values listed in Table I. We have observed completely consistent data for a 1%-Er single-crystal sample (then powdered), and also a polycrystalline sample with an Er concentration of  $\approx 1500$  ppm.<sup>10</sup> We also find a similar value for the thermal broadening,  $B$ , at 35 GHz. The temperature dependence of the relaxation rate and of the  $g$  value for samples doped with Dy or Gd are similar to that shown in Fig. 1 and these data are also presented in Table I.

The dotted line in Fig. 1(b) represents the results of normalizing the spin-relaxation rate at  $T = 7$  K, and taking  $1/\gamma T_2 \propto \rho_c^2 T$ , with  $\rho_c$  having the temperature dependence above and below 7 K as determined from the experimental data for the specific heat.<sup>1</sup> For the reasons mentioned earlier, we do not propose that this procedure is the correct physical interpretation of any model; we simply present it to illustrate just how dramatically the temperature dependence implied by the enhanced-specific-heat data fails to show up in the measured spin-relaxation rate of the local moment.

It is useful to compare the values of the thermal-broadening coefficient for Er, Dy, and Gd in UBe<sub>13</sub>

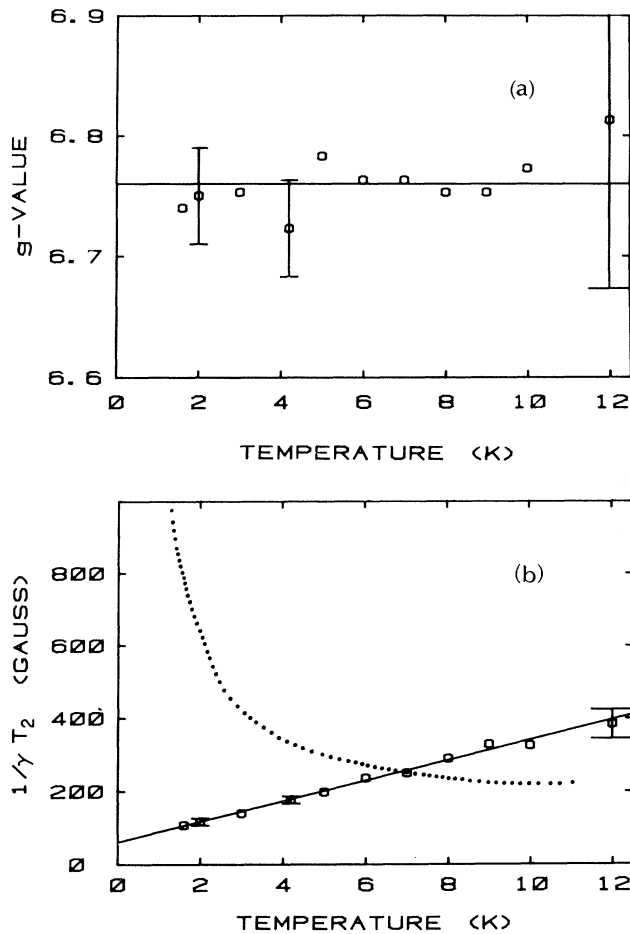


FIG. 1. (a) The  $g$  value and (b) the relaxation rate,  $1/\gamma T_2$ , for a  $\text{UBe}_{13}$ -1%-Er powdered sample as a function of temperature. The mean  $g$  value and the slope and intercept of the straight line fitted to the relaxation-rate data are presented in Table I. As discussed in the text, the dotted line in (b) represents the temperature dependence that one would expect if the relaxation rate was normalized to the data at 7 K and then presumed to obey a Korringa relation, i.e.,  $\propto \rho^2 T$ , with the density of states  $\rho$  taken to have the temperature dependence inferred from the enhanced electronic specific-heat data of Ref. 1.

with those for other  $M\text{Be}_{13}$  intermetallic compounds ( $M = \text{La}, \text{Lu}, \text{etc.}$ ). We present such data, as well as those for noble-metal hosts, in Table II. We suggest that the comparisons may be summarized as follows: (i) The thermal broadening for Gd appears to be relatively constant for all the  $M\text{Be}_{13}$  compounds, including  $\text{UBe}_{13}$ . (ii) The thermal broadening for both Er and Dy in  $\text{UBe}_{13}$  is larger than that for the other compounds by a factor of at most 3 to 5. (iii) The thermal broadening in most cases is not much larger than in noble-metal hosts, and the variation of the relaxation between noble-metal hosts (i.e., Er in Ag and Au) is also comparable to the variation among the  $M\text{Be}_{13}$  compounds.

Similarly, we note that the  $g$  value as presented in Fig. 1(a) is independent of temperature within the experimental error. Furthermore, were there a large enhanced Knight shift due to the relevant conduction-electron susceptibility being comparable to that measured for the bulk  $\text{UBe}_{13}$ , we would have expected a significant shift from the  $\Gamma_7$ -doublet group-theoretic value of  $g = 6.8$ ,<sup>19</sup> or for Dy from 7.55. In all cases the  $g$  values that we observe and their corresponding small  $g$  shifts are comparable to those for Er and Dy in simple metals and in other  $M\text{Be}_{13}$  compounds as shown in Table II. In summary, there is no anomalous behavior seen in the ESR  $g$  values of the local-moment-doped  $\text{UBe}_{13}$  samples.

Although it is true that the thermal broadening for Dy and Er in  $\text{UBe}_{13}$  is larger than in the other  $M\text{Be}_{13}$  intermetallics, the factor of only 3–5 is within the variations that happen to occur within other local-moment systems. In any event, it should be contrasted with the factor of  $\sim 10^5$  that would be expected (compared to Er in Ag) if there was an enhancement of the effective density of states relevant to spin relaxation comparable to that implied by the specific-heat data at 1.5 K. We must emphasize that even if one expected a greatly reduced effective density of states at the Er site, for example, due to hybridization between the conduction and the  $4f$  electrons,<sup>20,21</sup> one might still expect the temperature dependence to be mirrored in

TABLE I. Values of the gyromagnetic  $g$  factor, the residual ESR linewidth  $A$ , and the linear-thermal-broadening coefficient,  $B$ , for  $\text{UBe}_{13}$  doped with Er, Dy, and Gd.

Impurity	Concentration (ppm)	$g$ value	$A$ (G)	$B$ (G/K)	Frequency (GHz)
Er	1500	$6.77 \pm 0.04$	$5 \pm 2$	$25 \pm 2$	9.3
	1500	$6.68 \pm 0.03$	$35 \pm 3$	$30 \pm 1$	35
	10 000	$6.76 \pm 0.05$	$60 \pm 10$	$28 \pm 3$	9.3
Dy	2500	$7.63 \pm 0.1$	$35 \pm 15$	$56 \pm 5$	9.3
Gd	1000	$2.08 \pm 0.01$	$60 \pm 3$	$21 \pm 2$	9.3

TABLE II. Values of the gyromagnetic  $g$  factor, and ESR-linewidth linear-thermal-broadening coefficient,  $B$ , for different metallic and intermetallic hosts doped with Er, Dy, and Gd.

Host: Moment	$g$ value	$B$ (G/K)	Concentration (ppm)	Reference
Ag:Dy	$7.66 \pm 0.03$	$18.5 \pm 2$	26	3
Ag:Er	$6.83 \pm 0.02$	$7.5 \pm 1$	50	11
Ag:Gd	$2.09 \pm 0.01$	$25 \pm 5$	17	2
Au:Dy	. . .	$2.5 \pm 10$	500	12
Au:Er	6.80	$2.5 \pm 0.3$	100	13
Au:Gd	2.041	9.8	100	14
LaBe <sub>13</sub> :Dy	$7.42 \pm 0.10$	$16 \pm 4$	1000	15
LaBe <sub>13</sub> :Er	$6.74 \pm 0.05$	$6.5 \pm 1$	500	15
LaBe <sub>13</sub> :Gd	$2.040 \pm 0.005$	$18 \pm 5$	1000	15
ThBe <sub>13</sub> :Dy	$7.46 \pm 0.10$	$17 \pm 6$	1000	15
ThBe <sub>13</sub> :Er	$6.76 \pm 0.05$	$5 \pm 1$	1000	15
ThBe <sub>13</sub> :Er	$7.10 \pm 0.10$	$4 \pm 1$	10 000	a
ThBe <sub>13</sub> :Gd	$2.032 \pm 0.005$	$14 \pm 2$	1000	15
LuBe <sub>13</sub> :Er	$6.74 \pm 0.05$	$7 \pm 1.5$	1000	15
LuBe <sub>13</sub> :Gd	$2.003 \pm 0.005$	$16 \pm 3$	1000	15
CeBe <sub>13</sub> :Er	$6.70 \pm 0.05$	$10 \pm 1$	2000	16
CeBe <sub>13</sub> :Gd	$2.05 \pm 0.05$	15	. . .	17
UBe <sub>13</sub> :Dy	$7.63 \pm 0.1$	$51 \pm 5$	2500	a
UBe <sub>13</sub> :Er	$6.76 \pm 0.05$	$28 \pm 3$	10 000	a
UBe <sub>13</sub> :Gd	$2.08 \pm 0.01$	$21 \pm 2$	1000	a
UBe <sub>13</sub> :Gd	$2.08 \pm 0.01$	. . .	2000	18

<sup>a</sup>This work; nominal concentration.

the linewidth data. As can be seen in Fig. 1(b), the experimental data show a negligible variation from a linear behavior. There are systems such as CePd<sub>3</sub>,<sup>20</sup> substitutional alloy variants,<sup>22</sup> and other intermediate-valence compounds<sup>16</sup> where the observed low-temperature thermal broadening of the ESR linewidth is greatly reduced compared to that expected from a comparison of their specific heat to that of a normal metal. However, in all cases known to us, when one analyzes the effects of the temperature variation of the specific heat on the observed linewidth in a manner similar to that which we used to obtain the dotted line in Fig. 1(b), one finds that the result deviates from linear behavior very little; the deviations are comparable to the measurement uncertainties. Thus, such data are not an effective test as to whether the observed ESR linewidths are directly related to the electronic specific heat. In contrast, the difference between the dotted and solid lines in Fig. 1(b) is readily discernible, thereby clearly demonstrating that the temperature dependence of the ESR linewidth is not simply related to that of the electronic specific heat.

The preceding observations of the observed linearity of the linewidth with temperature, the absence of any significant enhancement to the thermal broadening,

and the lack of any anomaly in the  $g$  value all combine to suggest that the local moments substituting at the U sites simply do not appear to be significantly coupled to the "heavy-fermion system" either directly or indirectly. This would appear to be contrary to the implications of the Be NMR data of MacLaughlin *et al.*,<sup>6</sup> where they observe both a greatly enhanced nuclear relaxation rate and a decided deviation from a simple linear behavior over the range of temperature covered by our data. A possible explanation compatible with both sets of data is that the actual Korringa relaxation rate in UBe<sub>13</sub> is extremely small, and that the observed  $T_1$  is dominated by another process, such as dipolar fluctuations of the U moments.<sup>23</sup>

A most succinct way of stating the nature of our data is to note that if one did not know that UBe<sub>13</sub> was a heavy-fermion system, one would have no significant indication that in fact it was, simply by knowledge of the ESR data presented. Alternatively, had the fully enhanced  $\rho_c$  or any appreciable fraction of it been appropriate to the relaxation process, we should not have been able to even see the local-moment ESR because the line would have broadened beyond observation.<sup>24</sup> We suggest that these observations and the full consequences of our data must be taken into account to test

any models being proposed to explain the electronic nature of the heavy-fermion systems such as  $UBe_{13}$ .

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<sup>1</sup>H. Ott, H. Rudigier, Z. Fisk, and J. Smith, in *Moment Formation in Solids*, edited by W. J. Buyers (Plenum, New York, 1984); G. R. Stewart, Z. Fisk, J. L. Smith, H. R. Ott, and F. M. Mueller, in *Proceedings of the Seventeenth International Conference on Low Temperature Physics*, edited by U. Eckern, A. Schmid, W. Weber, and H. Wühl (North-Holland, Amsterdam, 1984), p. 321.

<sup>2</sup>The observed  $g$  values for the Er and Dy resonances are so far from  $g = 2$  that the local-moment resonance is "unbottlenecked" for all conditions. For the Gd resonance occurring near  $g = 2$  we cannot be sure, although we expect a large spin-relaxation rate to the lattice. The question should be tested; see D. R. Fredkin and S. Schultz, *Phys. Rev. Lett.* **35**, 682 (1975), and S. Oseroff, S. Schultz, B. L. Gehman, and C. Rettori, *Phys. Rev. Lett.* **35**, 679 (1975).

<sup>3</sup>S. Oseroff, S. Schultz, M. Passeggi, and D. Wohlleben, *Phys. Rev. B* **15**, 1283 (1977).

<sup>4</sup>S. E. Barnes, *Adv. Phys.* **30**, 6801 (1981).

<sup>5</sup>The appropriate  $J$  for the  $g$  shift and linewidth may be different; see D. Davidov, K. Maki, R. Orbach, C. Rettori, and E. P. Chock, *Solid State Commun.* **12**, 621 (1973), and Ref. 3.

<sup>6</sup>D. E. MacLaughlin, Cheng Tien, W. G. Clark, M. D. Lan, Z. Fisk, J. L. Smith, and H. R. Ott, *Phys. Rev. Lett.* **53**, 1833 (1984).

<sup>7</sup>C. M. Varma, following Letter [*Phys. Rev. Lett.* **55**, 2723 (1985)], and *J. Appl. Phys.* **57**, 3064 (1985).

<sup>8</sup>For a metal system we take  $T_2 = T_1$ , and we determine  $1/\gamma T_2$  from the usual line-shape analysis assuming that the observed signal is a phase-shifted Lorentzian.

<sup>9</sup>The ESR data were taken on laboratory-built superheterodyne spectrometers using standard reflection techniques.

<sup>10</sup>Samples were made by means of same procedures discussed by H. R. Ott, H. Rudigier, Z. Fisk, and J. L. Smith, *Phys. Rev. Lett.* **50**, 1595 (1983).

<sup>11</sup>S. Schultz, D. R. Fredkin, B. L. Gehman, and M. R. Shanabarger, *Phys. Rev. Lett.* **31**, 1297 (1973).

<sup>12</sup>D. Davidov, R. Orbach, C. Rettori, L. J. Tao, and E. P. Chock, *Phys. Rev. Lett.* **28**, 490 (1972).

<sup>13</sup>D. Davidov, C. Rettori, A. Dixon, K. Baberschke, E. P. Chock, and R. Orbach, *Phys. Rev. B* **8**, 3563 (1973).

<sup>14</sup>K. Baberschke and Y. v. Spalden, *Phys. Rev. B* **19**, 5933 (1979).

<sup>15</sup>J. Bloch, D. Davidov, D. Dokter, I. Felner, D. Shaltiel, *J. Phys. F* **8**, 1805 (1978).

<sup>16</sup>F. Hoekstra, D. Davidov, G. Nieuwenhuys, and I. Felner, *Phys. Lett.* **88A**, 420 (1982).

<sup>17</sup>G. Heinrich and A. Meyer, *Solid State Commun.* **26**, 1 (1977).

<sup>18</sup>J. Bloch, D. Davidov, I. Felner, and D. Shaltiel, *J. Phys. F* **6**, 1979 (1976).

<sup>19</sup>We identify the Er resonance as that of a  $\Gamma_7$  doublet since the  $g$  value is so close to the value generally found for Er in metals. However, we should expect some deviation from the group-theoretic value of  $g = 6.8$  since that is for a simple cubic symmetry. One typically finds shifts comparable to those displayed in Table II.

<sup>20</sup>T. Gambke, B. Elschner, and L. L. Hirst, *Phys. Rev. Lett.* **40**, 1290 (1978).

<sup>21</sup>N. F. Mott, *Philos. Mag.* **30**, 403 (1978).

<sup>22</sup>H. Schaeffer and B. Elschner, *Z. Phys. B* **53**, 109 (1983).

<sup>23</sup>W. G. Clark, private communication.

<sup>24</sup>Our initial experiments were in polycrystalline samples and we were concerned that perhaps we were simply seeing the Er resonance from atoms located at grain boundaries or other anomalous locations, i.e., those that were not substitutional for the U atoms. To check this important point we deliberately used a single-crystal sample, which gave the same results before and after powdering. Further evidence is as follows. The only phases formed by U and either Gd, Er, or Dy with Be, are the isostructural  $UBe_{13}$  and, say,  $GdBe_{13}$  which have nearly identical lattice parameters. Lattice-parameter measurements on a number of rare-earth (and Th) impurities in  $UBe_{13}$  reveal, for all cases investigated, a Vegard-law behavior. [For the x-ray lattice parameter, see J. L. Smith, Z. Fisk, J. O. Willis, A. L. Giorgi, R. B. Roof, H. L. Ott, H. Rudigier, and E. Felder, in *Proceedings of the International Conference on Materials and Mechanisms of Superconductivity*, Ames, Iowa, 1985, *Physica (Amsterdam) B* (to be published).] In addition, arc-melted samples of  $U_{1-x}Gd_xBe_{13}$  show a smooth and continuous decrease of  $T_c$  with  $x$ . Metallography and microprobe analysis of  $U_{1-x}Th_xBe_{13}$  alloys find no second phases in the  $x$  range investigated; Th, in most cases, acts similarly to the rare earths. We conclude that it is highly unlikely that our samples have two-phase regions for the small values of  $x$  involved in our investigations.