Electron spin resonance of Er^{3+} in YBiPt

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We report low-temperature electron spin resonance (ESR) measurements in YBiPt, doped with 0.1% Er substituting for Y. The results show that Er^{3+} is located at a cubic lattice site and has a $\Gamma_8^{(3)}$ as ground state and an overall crystal-field splitting of $\sim 85(20)$ K. From the ESR spectra we inferred the existence of lattice distortions at the rare-earth site. These results may help in understanding the heavy-fermion system YbBiPt, which has the same structure as YBiPt.

I. INTRODUCTION

The cubic compound YBiPt forms in the MgAgAs cubic structure (space group $F43m$) of the so-called Heusler alloys.[1] This structure contains one formula unit per primitive cell, and the primitive rhombohedral cell is generally preferred over the face-centered-cubic unit cell in describing it.[2] The atoms are located on three of the four sites along the [111] diagonal, having tetrahedral point-group symmetry; the fourth site can be thought of as an ordered vacancy.

The study of this structure[3,4] became important owing to the discovery by Canfield and co-workers[5,6] that the compound YbBiPt is a cubic heavy fermion with the same MgAgAs structure, and has the largest known linear specific-heat coefficient $\gamma = 8$ J/mol K^2, which is an order of magnitude larger than that of typical heavy fermions[7] and three orders of magnitude greater than that of conventional metals.

In this paper we report on electron spin resonance (ESR) experiments of Er^{3+} in YBiPt. The study gives valuable information about the local symmetry, crystal field, and possible distortions associated with the rare-earth (RE) (Y and Er) site in the MgAgAs structure. We hope that these results will contribute to the understanding of the electronic properties of the superheavy fermion YbBiPt.

II. EXPERIMENT

Single crystals of YBiPt were grown from a Bi flux, as reported previously.[5] Er doping was accomplished by including Er in the melt ($Y_{1-x}Er_xBiPt$, $x \approx 0.001$). A typical crystal size was $2 \times 2 \times 2$ mm^3.

The ESR experiments were done in a Varian E-line X-band spectrometer, using either a liquid helium tail Dewar or a helium gas flux system, adapted to a room temperature rectangular TE_{102} cavity.

III. RESULTS AND ANALYSIS

The ESR spectra of Er^{3+} in YBiPt at liquid helium temperature show two intense anisotropic resonances and two weak, almost isotropic ones. The temperature dependence of the resonance intensities, down to 1.6 K, indicates that all four resonances belong to the ground state. Figure 1 shows the three most intense resonances with the external magnetic field parallel to the [001] direction. The resonances have a dysonian line shape,[8,9] characteristic of conducting hosts. Figures 2, 3, and 4 show, respectively, the resonance fields, relative intensities, and linewidths for the four observed resonances, as functions of the angle of the external field in the (110) plane. The most intense and narrow resonance of the spectrum has satellites due to the hyperfine lines of the $^{167}Er^{3+}$ ($I = 7/2$) isotope. The hyperfine splitting was found to be isotropic within the accuracy of the measurements, with a hyperfine constant $^{167}\mathcal{A} = 75 \pm 5$ G. The poor resolution between the hyperfine lines may be the result of the superposition of forbidden transitions ($\Delta m_I = \pm 1$); such transitions are allowed by nuclear quadrupole interactions,[10,11] which are probably caused by internal lattice distortions.

The experimental data described above indicate that Er^{3+} is located at a lattice site of cubic symmetry. The Hamiltonian describing the energy levels within a manifold of angular momentum J, in the presence of an external mag-

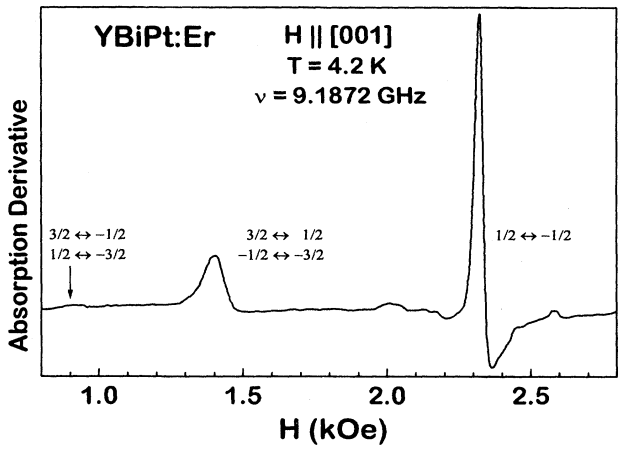

FIG. 1. ESR spectrum of ~0.1% Er^{3+} in YBiPt at $T=4.2$ K for H‖[001].

netic field and a crystal field of cubic point symmetry, is given by[12]

$$\mathcal{H}=g_J\mu_B\vec{H}\cdot\vec{J}+\beta_J A_4\langle r^4\rangle[O_4^0(\vec{J})+5O_4^4(\vec{J})]+\gamma_J A_6\langle r^6\rangle \times[O_6^0(\vec{J})-21O_6^4(\vec{J})], \quad (1)$$

where the first term is the Zeeman interaction and the second and third terms represent the cubic crystal field. β_J and γ_J are reduced matrix elements, $A_4\langle r^4\rangle$ and $A_6\langle r^6\rangle$ are the fourth- and sixth-order crystal-field parameters, and O_n^m are the Stevens equivalent operators.[12]

Following Lea, Leask, and Wolf,[13] Eq. (1) may be written

$$\mathcal{H}=g_J\mu_B\vec{H}\cdot\vec{J}+W\left[x\frac{O_4}{F_4}+(1-|x|)\frac{O_6}{F_6}\right], \quad (2)$$

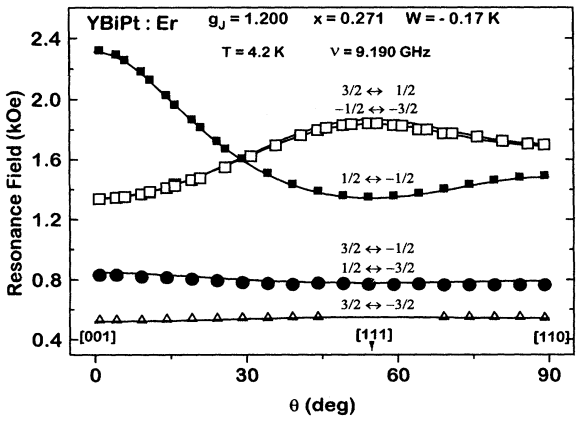

FIG. 2. Angular dependence of the resonance field for the four observed resonances of Er^{3+} in YBiPt. The external field is rotated in the (110) plane.

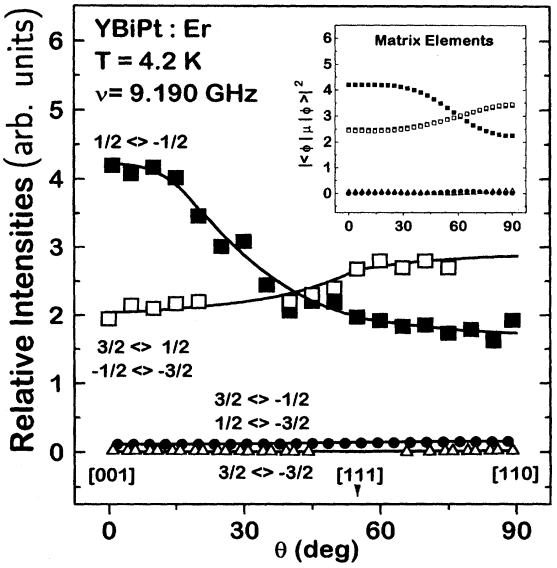

FIG. 3. Angular dependence of the relative intensities for the four observed resonances of Er^{3+} in YBiPt. The external field is rotated in the (110) plane. The solid lines are guides for the eye. The inset shows the calculated angular dependence of the magnetic dipole matrix element for the observed transitions (see text).

where

$$\beta_J A_4\langle r^4\rangle F_4=Wx,$$

$$\gamma_J A_6\langle r^6\rangle F_6=W(1-|x|),$$

$$O_4=O_4^0(\vec{J})+5O_4^4(\vec{J}),$$

$$O_6=O_6^0(\vec{J})-21O_6^4(\vec{J}),$$

F_4 and F_6 are numerical factors, W is a scaling factor for the overall crystal-field splitting (Δ), and x gives the ratio between the fourth- and sixth-order crystal-field parameters.

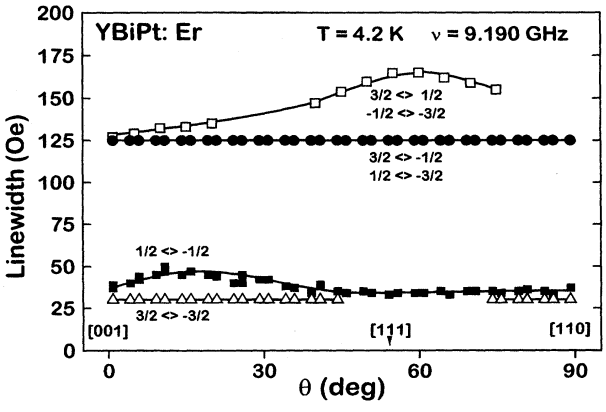

FIG. 4. Angular dependence of the linewidth for the four observed resonances of Er^{3+} in YBiPt. The external field is rotated in the (110) plane.

Since the observed resonances are anisotropic and belong to the ground state, they must come from transitions within the Zeeman levels of one of the three $\Gamma_8^{(i)}$ quartets that result from the splitting of the free Er^{3+} ion ground state ($4f^{11}$, $^4I_{15/2}$) in the cubic crystal field.[12] The best fit of Eq. (2) to the experimental data of Fig. 2, taking into consideration the exact diagonalization of the 16×16 ($J=15/2$) energy matrix when the magnetic field is rotated in a (110) plane, is obtained for a $\Gamma_8^{(3)}$ ground state with $g_J=1.200(1)$, $x=0.271(5)$, and $W<0$. Therefore the ground state of Er^{3+} in YBiPt is a $\Gamma_8^{(3)}$ quartet, and the observed resonances correspond to the transitions within this quartet.

Using the $\Gamma_8^{(3)}$ wave functions for $x=0.271$ and $g_J=1.200$, we calculated the angular dependence of the magnetic dipole matrix elements for the six possible transitions within the $\Gamma_8^{(3)}$ quartet. The inset in Fig. 3 shows the calculated matrix elements, which are in reasonable agreement with the experimental relative intensities shown in Fig. 3, both in magnitude and angular dependence.

Figure 4 shows that the linewidths of the transitions corresponding to the two Kramers doublets ($1/2\leftrightarrow-1/2$, $3/2\leftrightarrow-3/2$) are narrower than the linewidths corresponding to the other four transitions between these two Kramers doublets ($3/2\leftrightarrow1/2$, $-1/2\leftrightarrow-3/2$, and $3/2\leftrightarrow-1/2$, $1/2\leftrightarrow-3/2$). This is expected, since the former transitions are not affected by lattice distortions. The broadening of about 25 Oe for the ($1/2\leftrightarrow-1/2$) transition around 15° from the [001] direction is the result of local distortions (mosaic effects) together with the strong angular dependence of the resonance field ($dH_{res}/d\theta\simeq25$ Oe/deg; see Fig. 2). The broadening of the $3/2\leftrightarrow1/2$, $-1/2\leftrightarrow-3/2$ transitions along the [111] direction ($40\pm15Oe$) can be attributed in part to a small splitting between these two transitions, due to the admixture of excited crystal-field levels via the applied magnetic field. Attributing the minimum broadening of ~25 Oe to the splitting between these two transitions along the [111] direction, $g_J=1.200$ and $x=0.271$ (obtained above), we estimated $W=-0.17$ K. For these values, the order of the crystal field levels is $\Gamma_8^{(3)}(0K)$, $\Gamma_6(\sim30$ K), $\Gamma_8^{(2)}(\sim45$ K), $\Gamma_8^{(1)}(\sim65$ K), and $\Gamma_7(\sim85$ K). Figure 2 shows the splitting between the $3/2\leftrightarrow1/2$ and $-1/2\leftrightarrow-3/2$ transitions, calculated as discussed above. Figure 5 shows a rapid linewidth broadening above ~12 K for the transition $1/2\leftrightarrow-1/2$. This broadening—due to relaxation via thermally populated excited crystal field levels—may be the result of spin-lattice coupling[14] and/or RE-conduction-electron exchange interactions,[15] although the latter effect may not be so important in view of the small temperature dependence of the linewidth (Korringa rate) below 12 K (see Fig. 5).

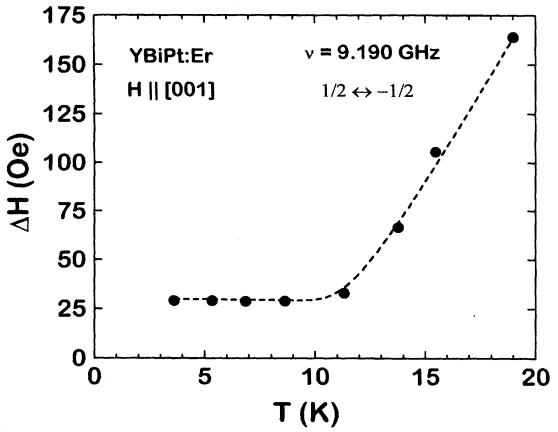

FIG. 5. Temperature dependence of the linewidth for the $1/2\leftrightarrow-1/2$ transition. The dashed line is a guide for the eye.

IV. CONCLUSIONS

In this work we have shown that the RE site in the compound YBiPt is cubic, and that for Er^{3+} the ground state is a $\Gamma_8^{(3)}$ with an overall cubic crystal field splitting of ~85(20) K. This value is comparable with recent neutron-scattering results on YbBiPt, in which an overall crystal-field splitting of ~6 meV (~70 K) is estimated.[16] The presence of small distortions at the cubic site can be inferred from the broadening of the $1/2\leftrightarrow-1/2$ transition observed around 15° from the [001] direction. The unresolved hyperfine structure for the $1/2\leftrightarrow-1/2$ transition also supports the existence of such distortions at the RE sites. The distortions may be due to the substitution of Er^{3+} for Y^{3+}, but it is unlikely, because these ions are of similar size and have the same valence. Finally, we note that in spite of the observed Er^{3+} resonance line shape being metallic (dysonian), the Er^{3+} relaxation via conduction electrons (Korringa relaxation) was found to be very small (see Fig. 5 for $T<12$ K).

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