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PHYSICA B

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

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Abstract

Electron spin resonance (ESR) experiments at 4.15 K of Er^{3+} in YBiPt show that Er^{3+} is in a site of cubic local symmetry, with a $\Gamma_8^{(3)}$ ground state and an overall crystal field splitting of $\sim 85(10)$ K. We inferred from the spectra the existence of lattice distortions at the rare-earth (RE) site. These results may help in understanding the heavy-fermion system YbBiPt , which has the same structure as YBiPt .

1. Introduction

The 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.

Interest in this structure [3,4] has grown since the discovery by Canfield et al. [5,6] that the compound YbBiPt is a cubic heavy-fermion and has the largest known linear specific-heat coefficient, $\gamma = 8 \text{ J/mol K}^2$. This is ten times that of typical heavy-fermions [7] and about a thousand times that of conventional metals.

Here we report on ESR experiments of Er^{3+} in YBiPt . The results give information about local symmetry and

possible distortions associated with the RE (Y and Er) site in the MgAgAs structure.

2. Experiment

Single crystals of YBiPt were grown from a Bi flux [5] and doped by including Er in the melt ($\text{Y}_{1-x}\text{Er}_x\text{BiPt}$; $x \approx 0.001$). A typical crystal size was $2 \times 2 \times 2 \text{ mm}^3$. 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 TE_{102} cavity.

3. Results and analysis

The ESR spectra of Er^{3+} in YBiPt at 4.15 K show two intense anisotropic resonances and two weak, almost isotropic ones (Fig. 1). Their increased intensities at 1.6 K indicate that all four resonances belong to the ground

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Fig. 1. ESR spectrum of $\sim 0.1\%$ Er^{3+} in YBiPt at $T = 4.2$ K for $H \parallel [001]$. Only three strongest lines are seen in this scale.

state. The resonances have a dysonian line shape [8,9], characteristic of conducting hosts. The most intense and narrow resonance has satellites due to the hyperfine lines of the $^{167}\text{Er}^{3+}$ ($I = \frac{7}{2}$) isotope. The hyperfine splitting was isotropic within the accuracy of the measurements, with a hyperfine constant $^{167}A = 75 \pm 5$ G. The poor resolution of the hyperfine lines may be due to the superposition of forbidden transitions ($\Delta m_I = \pm 1$); such transitions are allowed by nuclear quadrupole interactions [10,11], which are themselves probably caused by internal lattice distortions.

Our ESR results indicate the Er^{3+} is located at a site of cubic symmetry. The Hamiltonian describing the energy levels within a manifold of angular momentum J , in a crystal field (CF) of cubic point symmetry, with an external magnetic field, is given [12] by

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

where the first term is the Zeeman interaction and the second and third terms represent the cubic CF. 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 CF parameters, and O_n^m are the Stevens equivalent operators [12].

Following Lea et al. [13], Eq. (1) may be written

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

where

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

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

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. Solid lines are fitting of Eq. (1).

$$O_4 = O_4^0(J) + 5O_4^4(J),$$

$$O_6 = O_6^0(J) - 21O_6^4(J).$$

F_4 and F_6 are numerical factors, W is a scaling factor for the overall CF splitting (Δ).

Since the 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 CF [12]. The best fit of Eq. (2) to the experimental data (Fig. 2), using 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 $W < 0$. Hence 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.

The line widths of the transitions corresponding to the two Kramers doublets ($\frac{1}{2} \leftrightarrow -\frac{1}{2}$, $\frac{3}{2} \leftrightarrow -\frac{3}{2}$) are narrower than the line widths corresponding to the other four transitions between these two Kramers doublets ($\frac{3}{2} \leftrightarrow \frac{1}{2}$, $-\frac{1}{2} \leftrightarrow -\frac{3}{2}$ and $\frac{3}{2} \leftrightarrow -\frac{1}{2}$, $\frac{1}{2} \leftrightarrow -\frac{3}{2}$) (Fig. 3). This is expected, since the former transitions are not affected by lattice distortions. The broadening of about 25 Oe for the ($\frac{1}{2} \leftrightarrow -\frac{1}{2}$) 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 ($dH_{\text{res}}/d\theta \simeq 25$ Oe/deg, see Fig. 2). The small broadening of the $\frac{3}{2} \leftrightarrow \frac{1}{2}$, $-\frac{1}{2} \leftrightarrow -\frac{3}{2}$ transitions along the [111] direction (~ 25 Oe) is attributed to a small splitting (Fig. 2) between these two transitions, due to the admixture of excited CF levels via the applied magnetic field.

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

Assuming a splitting of ~ 25 Oe along the [111] direction, $g_J = 1.200$ and $x = 0.271$, we estimated $W = -0.17$ K. For these values, the order of the CF levels is $\Gamma_8^{(3)}$ (0 K), Γ_6 (~ 30 K), $\Gamma_8^{(2)}$ (~ 45 K), $\Gamma_8^{(1)}$ (~ 65 K), and Γ_7 (~ 85 K).

4. Conclusions

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 CF splitting of $\sim 85(10)$ K. The presence of small distortions at the cubic site can be

inferred from the broadening of the $\frac{1}{2} \leftrightarrow -\frac{1}{2}$ transition observed around 15° from the [001] direction. The unresolved hyperfine structure for the $\frac{1}{2} \leftrightarrow -\frac{1}{2}$ 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.

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