

Superconductivity and magnetism of a body-centered tetragonal ErRh_4B_4 single crystal.

II. Large anisotropy of H_{c2}

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Upper critical field H_{c2} and magnetization are measured on a single crystal of body-centered tetragonal (bct) ErRh_4B_4 which is an antiferromagnetic superconductor and behaves metamagnetically in the c plane under a magnetic field below T_N . The temperature dependences of H_{c2} along the [001] and [100] axes are roughly similar to each other in that both $H_{c2}(T)$ take maxima with decreasing temperature but the magnitude of H_{c2} along the [001] axis is quite large compared to that along the [100] axis. Furthermore, the angular dependence of H_{c2} in the c plane shows the fourfold anisotropy above T_N and the complex twofold anisotropy below T_N . The anisotropy of H_{c2} is closely related with that of the magnetization. From the analysis in which the spin polarization due to the s - f exchange interaction and the electromagnetic interaction are taken into account it is made clear that the spin polarization plays a very important role in determining H_{c2} . The s - f exchange constant and the spin-orbit scattering parameter are also determined. It is indicated that the s - f exchange interaction is ferromagnetic and the spin-orbit scattering is very strong in the bct ErRh_4B_4 crystal. Finally temperature dependence of H_{c2} in the c plane below T_N is qualitatively discussed, being connected with metamagnetism.

I. INTRODUCTION

Magnetic superconductor $\mathcal{R}\text{Rh}_4\text{B}_4$ (\mathcal{R} is a rare-earth metal) compounds have been intensively studied in order to understand the interplay between superconductivity and magnetism.¹ So far, three kinds of $\mathcal{R}\text{Rh}_4\text{B}_4$ compounds have been discovered: (i) The CeCo_4B_4 type with primitive tetragonal (pt) structure.² The pt $\mathcal{R}\text{Rh}_4\text{B}_4$ compounds are the most familiar magnetic superconductors and there are many studies on these compounds. ErRh_4B_4 with this structure is a typical ferromagnetic superconductor and shows a reentrant behavior.³ (ii) The LuRu_4B_4 type with body-centered tetragonal (bct) structure.⁴ Although the bct phase has been stabilized only in the pseudoternary $\mathcal{R}(\text{Ru}_{1-x}\text{Rh}_x)_4\text{B}_4$ system,⁴ we succeed in obtaining ErRh_4B_4 with this structure which is an antiferromagnetic superconductor.⁵ (iii) The LuRh_4B_4 type with orthorhombic structure.⁶ The superconducting transition temperatures of several compounds have been reported.⁷

It is expected that in the magnetic superconductor the magnetic anisotropy influences superconducting properties such as the upper critical field H_{c2} because exchange interaction exists between the superconducting electrons and the magnetic ions. It has been highly desirable to make a study of a single crystal of the magnetic superconductor in order to obtain precise information on the interplay between superconductivity and magnetism.

Recently, we succeeded in preparing a bct ErRh_4B_4 single crystal^{8,9} and found for the first time the anisotropy of H_{c2} among a number of antiferromagnetic superconductors. In this paper we discuss the relationship between the anisotropy of H_{c2} and magnetization. The experimental procedure is briefly described in Sec. II; the experimental results are presented in Sec. III and discussions are given in Sec. IV. A brief report of the H_{c2} anisotropy was re-

ported elsewhere.⁹ Experimental results on magnetization and a detailed analysis are described in the preceding paper¹⁰ referred to as paper I.

II. EXPERIMENTAL

H_{c2} was measured by an ac resistive method, sweeping the magnetic field at a constant temperature. H_{c2} was defined by the linearly extrapolated field of the transition curve to zero resistance. A ^3He - ^4He dilution refrigerator was used in the measurements below 1 K. The bct ErRh_4B_4 single crystal which was synthesized by induction heating is the same as that in the preceding paper I. LuRh_4B_4 , with the body-centered tetragonal structure, was also synthesized by induction heating, as a reference sample. The bct LuRh_4B_4 sample is polycrystalline and was annealed for two weeks at 1050°C, being covered with a Ta foil and sealed into a quartz tube under Ar atmosphere. This sample contains a small quantity of RhB as an impurity phase.

III. RESULTS

The residual resistivity just above T_c and the residual resistance ratio (RRR) of the bct ErRh_4B_4 sample are $\rho_0 \approx 65 \mu\Omega \text{ cm}$ and a RRR of 1.70, respectively. Those of the bct LuRh_4B_4 sample are $\rho_0 \approx 60 \mu\Omega \text{ cm}$ and a RRR of 1.65. These values are roughly common throughout the bct $\mathcal{R}\text{Rh}_4\text{B}_4$ compounds, and ρ_0 is large by a figure compared with those in the pt $\mathcal{R}\text{Rh}_4\text{B}_4$ compounds. The large residual resistivity suggests that the bct $\mathcal{R}\text{Rh}_4\text{B}_4$ compounds are dirty superconductors. The superconducting transition temperature T_c and the Néel temperature T_N of the bct ErRh_4B_4 sample are 7.80 and 0.65 K, respectively.

The magnetization process of bct ErRh_4B_4 has been

described in paper I. The results of the magnetization measurements in the superconducting paramagnetic state are summarized as follows. The [001] axis is the hard-magnetization direction. Large fourfold magnetic anisotropy exists in the c plane. The [100] axis is the easy-magnetization direction and the [110] axis is the hard-magnetization direction in the c plane. In the field range in which the sample shows the transition from the superconducting state to the normal one, the magnetization shows anisotropy due to the fourfold magnetic anisotropy. The magnetization values $M(H_{c2})$ at H_{c2} for $\mathbf{H} \parallel [100]$ and $\mathbf{H} \parallel [110]$ are nearly equal to each other in spite of the fact that H_{c2} takes different values for both directions.

Below T_N the magnetization curves for $\mathbf{H} \parallel [100]$ show two-step jumps exhibiting hysteresis for increasing and decreasing fields. Similar magnetization curves with the two-step jumps are also observed for $\mathbf{H} \parallel [010]$. On the other hand, the magnetization for $\mathbf{H} \parallel [110]$ shows a one-step jump. The magnetization process observed below T_N is understood as metamagnetism under the fourfold magnetic anisotropy.

Figure 1 shows the temperature dependence of H_{c2} in the [001], [100], and [110] directions. H_{c2} along the [001] direction which is the axis of hard magnetization is very large compared with those in the c plane. It increases monotonically down to 1.5 K, decreases a little at lower temperatures, and is almost constant below T_N . The $H_{c2}(T)$ curve in the [100] direction takes a maximum at about 4.5 K and gradually decreases at lower temperatures. The inset in Fig. 1 shows $H_{c2}(T)$ along the [100] and [110] directions around T_N in the enlarged H_{c2} scale. H_{c2} along the [100] direction takes a dip at 0.6 K, which

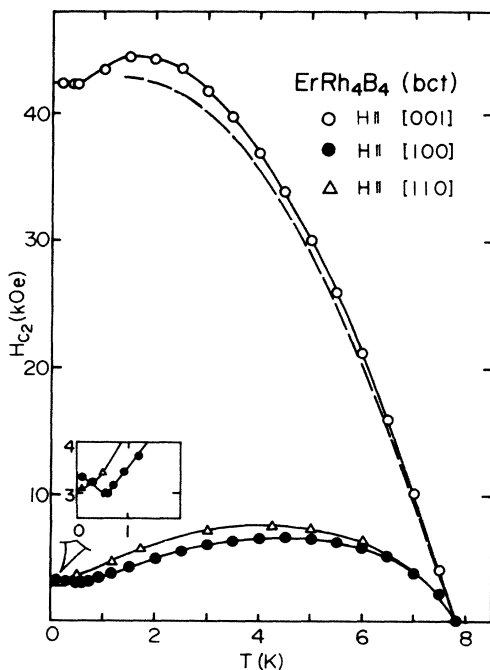


FIG. 1. Temperature dependence of H_{c2} of bct ErRh_4B_4 (a) for $\mathbf{H} \parallel [001]$, $\mathbf{H} \parallel [100]$, and $\mathbf{H} \parallel [110]$. The dashed curve in (a) shows H_{c2} after the demagnetization correction. In the inset H_{c2} in the c plane around T_N is shown in the enlarged scale.

is nearly equal to T_N . The anisotropy of H_{c2} is also observed in the c plane. H_{c2} along the [110] direction is always higher than that along the [100] direction above T_N . However, H_{c2} along both the [100] and [110] directions coincides at 0.3 K and H_{c2} along the [100] direction is higher than that along the [110] direction below 0.3 K.

The angular dependences of the upper critical field $H_{c2}(\theta)$ in the c plane at 7.0, 4.3, and 0.5 K are shown in Figs. 2(a), 2(b) and 2(c), respectively. $H_{c2}(\theta)$ at 7.0 K just

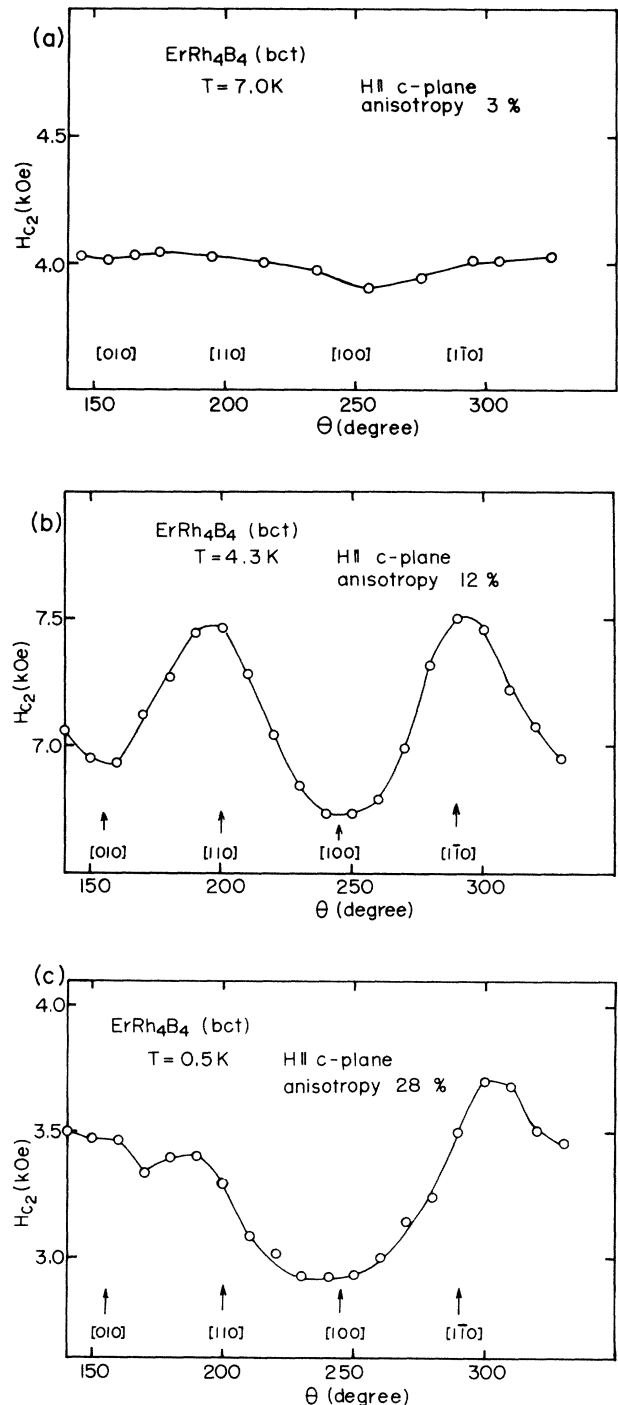


FIG. 2. Angular dependence of H_{c2} of bct ErRh_4B_4 (a) at 7.0 K just below T_c , (b) at 4.3 K, and (c) at 0.5 K, below T_N .

below T_c shows nearly no anisotropy. $H_{c2}(\theta)$ at 4.3 K almost shows the fourfold anisotropy with the magnitude of 12%, where the anisotropy is defined by

$$\frac{H_{c2_{\max}} - H_{c2_{\min}}}{H_{c2_{\min}}} \quad (1)$$

The minima of H_{c2} appear in the [100] and [010] directions which are the axes of easy magnetization, and the maxima of H_{c2} are in the [110] and $\bar{1}\bar{1}0$ directions which are the axes of hard magnetization in the c plane. It should be noted that H_{c2} values are clearly different between the crystallographically equivalent [100] and [010] directions, though both take minima. The discrepancy is beyond the limit of demagnetization correction. This is also observed in the magnetization measurements (see paper I), where the small magnetic anisotropy between the [100] and [010] axes exists in this sample. This is also apparent in the angular dependence of H_{c2} at 0.5 K just below T_N . However, Fig. 2(c) shows that $H_{c2}(\theta)$ does not show the fourfold anisotropy, but the complex twofold anisotropy. Only H_{c2} along the [100] direction takes a minimum. The magnitude of anisotropy amounts to 28%.

Figures 3(a) and 3(b) show the temperature dependence of the anisotropy of H_{c2} between principal axes in the c plane and the a plane, respectively. The definition of anisotropy is given in Figs. 3(a) and 3(b). The anisotropy of H_{c2} between the [100] and [110] directions shows significant temperature dependence. This anisotropy is very small at temperatures just below T_c , and increases gradually with decreasing temperature. After taking a maximum at 1.5 K, it becomes small again at 0.5 K, because H_{c2} along the [110] direction does not take a maximum, as shown in Fig. 2(c). The anisotropy of H_{c2} between the [100] and [001] directions has strong temperature dependence, as shown in Fig. 3(b). It is extremely large, exceeding 800% at 1.7 K.

In Fig. 4 the magnetization values at H_{c2} , $M(H_{c2})$ are plotted as a function of the temperature for $\mathbf{H}||[100]$, $\mathbf{H}||[110]$, and $\mathbf{H}||[001]$. $M(H_{c2})$ was defined by using the magnetization measurements in paper I. As mentioned before, $M(H_{c2})$ almost coincides between the [100] and [110] directions. This indicates that in the c plane superconductivity begins to be destroyed by the generation of a certain magnitude of the magnetization at every temperature. It should be noted that, exactly speaking, $M(H_{c2})$ for $\mathbf{H}||[110]$ is a little smaller than that for $\mathbf{H}||[100]$. $M(H_{c2})$ is very small for $\mathbf{H}||[001]$, whereas H_{c2} is extremely high compared with that in the c plane.

Finally, the experimental results of the bct LuRh_4B_4 sample are described. ρ_0 and the RRR are almost the same as that of the bct ErRh_4B_4 single crystal, as described before. Figure 5 shows the temperature dependence of H_{c2} . The gradient of H_{c2} is $(-dH_{c2}/dT)_{T_c} = 20.8$ kOe/K, where T_c is 8.61 K. The gradient of H_{c2} is much larger than that of LuRh_4B_4 with primitive tetragonal structure¹¹ and may come from the dirtiness of the sample as indicated by the high resistivity of this sample. The dashed-dotted curve shows H_{c2} in the dirty limit by the Werthamer, Helfand, and Hohenberg

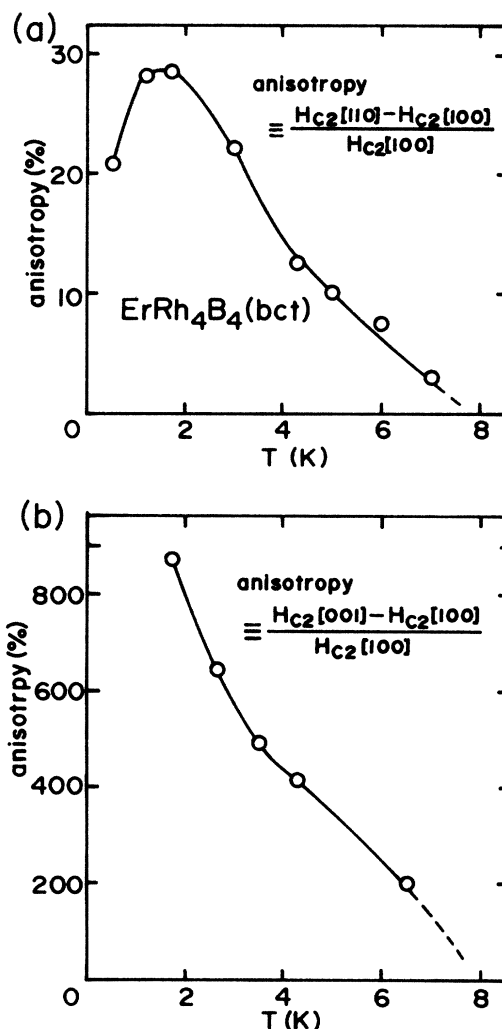


FIG. 3. Temperature dependence of the anisotropy of H_{c2} (a) in the c plane and (b) in the a plane.

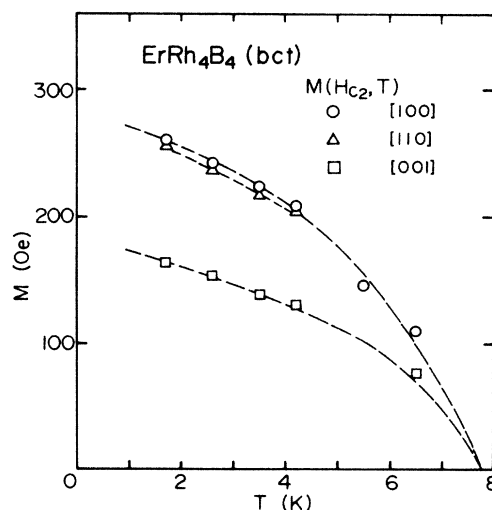


FIG. 4. Temperature dependence of the magnetization at H_{c2} for $\mathbf{H}||[100]$, $\mathbf{H}||[110]$, and $\mathbf{H}||[001]$.

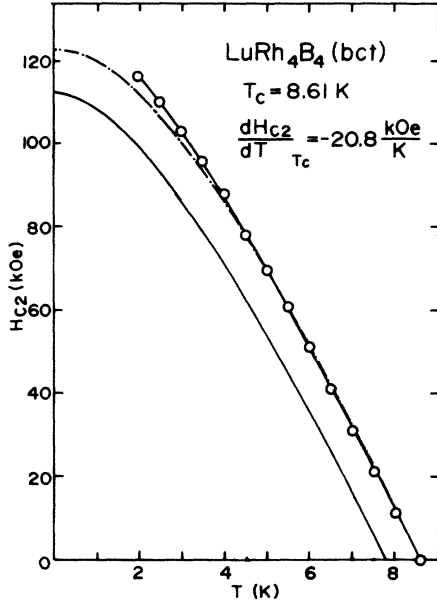


FIG. 5. Temperature dependence of H_{c2} of bct LuRh₄B₄. The dashed-dotted curve shows H_{c2} in the dirty limit by WHH theory. The solid curve shows orbital critical field H_{c2}^* of the hypothetical nonmagnetic bct ErRh₄B₄ with $T_c = 7.80$ K and $-(dH_{c2}/dT)_{T_c} = 20.8$ kOe/K.

(WHH) theory.¹² This curve corresponds to the orbital upper critical field. Though $H_{c2}(0)$, estimated from the theoretical curve, is 123 kOe, the experimental data are slightly higher than the theoretical curve.

IV. DISCUSSION

A. Anisotropy of H_{c2} of bct ErRh₄B₄ above T_N

The large anisotropy of H_{c2} between the [100] and [001] directions and the fourfold anisotropy of H_{c2} in the c plane can never be explained as long as the effect of the anisotropic magnetization is not considered, because the crystal structure of the bct ErRh₄B₄ compounds has nearly cubic symmetry as a whole.

In order to explain the large anisotropy of H_{c2} observed in bct ErRh₄B₄, we need to take account of both effects of the electromagnetic interaction¹³ and the spin polarization due to the s - f exchange interaction.^{14,15} The bct ErRh₄B₄ compounds are a dirty superconductor, as described in Sec. III. Therefore, in the analysis we use the following expression which holds for the dirty limit¹⁶:

$$H_{c2}(T) = H_{c2}^*(T) - 4\pi M(H_{c2}, T) - A[H_{c2}(T) - I'M(H_{c2}, T)]^2, \quad (2)$$

where

$$A = 0.022\alpha/\lambda_{s.o.}T_{c0} \quad (3)$$

and

$$I' = 4\pi + I, \quad I = (g_J - 1)I_0/2Ng_J\mu_B^2, \quad (4)$$

where α and $\lambda_{s.o.}$ are the Maki parameter and the spin-orbit scattering parameter, respectively, T_{c0} is the superconducting transition temperature of nonmagnetic bct ErRh₄B₄, N is the number of Er spins per unit volume, and g_J is the Landé g -factor. The term $4\pi M$ represents the effect of the electromagnetic interaction and the term IM , the effect of the spin polarization due to the s - f exchange interaction, where I is proportional to the s - f exchange constant I_0 . It should be noted that the electromagnetic interaction behaves not only as an orbital pair-breaking mechanism shown by the second term of Eq. (2) but also as the spin paramagnetic one containing the third term of Eq. (2). On the other hand, the s - f exchange interaction which is contained in the third term of Eq. (2) behaves as a spin paramagnetic pair-breaking mechanism. H_{c2}^* is the upper critical field limited by orbital effect only.

$H_{c2}(T)$ of bct LuRh₄B₄ which is shown in Fig. 5 is utilized for $H_{c2}^*(T)$ because both its resistivity and its residual resistance ratio are almost the same between the bct ErRh₄B₄ and the bct LuRh₄B₄ samples, as mentioned in Sec. III. Though H_{c2} of bct LuRh₄B₄ in the experiments is slightly higher than the orbital upper critical field H_{c2}^* by the WHH theory, the following analysis is performed on the basis of the theoretical H_{c2}^* . It is necessary to correct the discrepancy of the superconducting transition temperature between bct ErRh₄B₄ ($T_c = 7.80$ K) and bct LuRh₄B₄ ($T_{c0} = 8.61$ K). Since $(-dH_{c2}/dT)_{T_c}$ does not explicitly depend on T_c in the dirty limit according to the Bardeen-Cooper-Schrieffer-Ginzburg-Landau-Abrikosov-Gor'kov (BCS-GLAG) theory, $(-dH_{c2}/dT)_{T_c}$ is assumed to be almost constant even if T_c changes a little. Therefore, $H_{c2}^*(T)$ is the orbital upper critical field of the hypothetical nonmagnetic superconductor bct ErRh₄B₄ with $T_c = 7.80$ K and $(-dH_{c2}/dT)_{T_c} = 20.8$ kOe/K, which is shown by the solid curve in Fig. 5. Furthermore, it is also implicitly assumed that H_{c2}^* has no anisotropy because the crystal structure of the bct compounds has nearly cubic symmetry as a whole.

The experimental results of H_{c2} shown in Fig. 1 are analyzed on the basis of Eq. (2) by use of $M(H_{c2})$ shown in Fig. 4. In order to explain the anisotropy of H_{c2} observed between the [100] and [001] axes, the values of the parameters I' and A are determined under the conditions $H_{c2}^* = H_{c2}^*[100] = H_{c2}^*[001]$ at every temperature. The two solutions with positive and negative signs are obtained on I' . The mean values of the parameters obtained at several temperatures are $I' = 1112$, $A = 1.1 \times 10^{-6}$ and $I' = -140$, $A = 1.05 \times 10^{-4}$. In order to determine the solution having the physical meaning, the temperature dependence of H_{c2} along the [110] direction and the angular dependence of H_{c2} in the c plane are calculated by Eq. (2), using each set of parameters $[I', A]$. The calculated results, together with the experimental results, are shown in Figs. 6 ($H_{c2}(T)$ along the [110] direction) and 7 [$H_{c2}(\theta)$ in the c plane]. It is clear that, while I' with negative sign cannot explain the experimental results, I' with positive sign does. Because the value of I' is much larger than 4π , I' in Eq. (4) can be regarded as I . The s - f exchange constant I_0 and the parameter $\lambda_{s.o.}$ are estimated

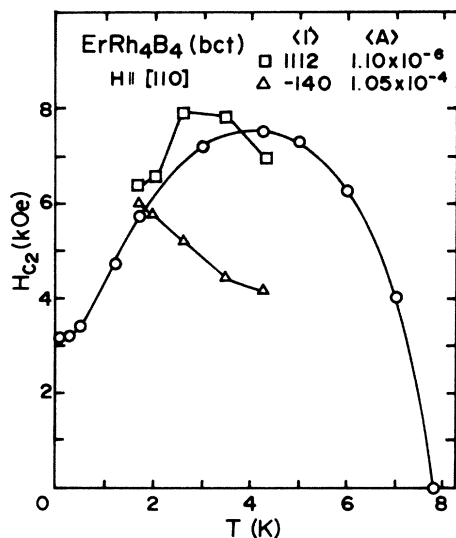


FIG. 6. Comparison of the experimental H_{c2} with the calculated one for $H||[110]$. Only the solution with a positive I' value can explain the experimental results.

to be 74 K and $\lambda_{s.o.} = 2.81$ by use of Eqs. (3) and (4). The positive sign of I' indicates that the exchange interaction between the conduction electrons and the magnetic spins is ferromagnetic. The large $\lambda_{s.o.}$ value also indicates that the spin-orbit scattering is very strong in this crystal.

Figure 8 shows the contributions of both the electromagnetic interaction and the spin polarization due to the s - f exchange interaction to H_{c2} along the [001] and the [100] axes. It is important that H_{c2}^* of hypothetical nonmagnetic bct ErRh_4B_4 (dashed curve) is greatly reduced by the effect of the spin polarization due to the s - f exchange interaction. The arrows show the contribution of the spin polarization to H_{c2} along the [001] and [100] axes. The contribution of the electromagnetic interaction represented by $4\pi M$ is much smaller compared to that of the spin polarization due to the s - f exchange interaction. It should be pointed out that the experimental value along

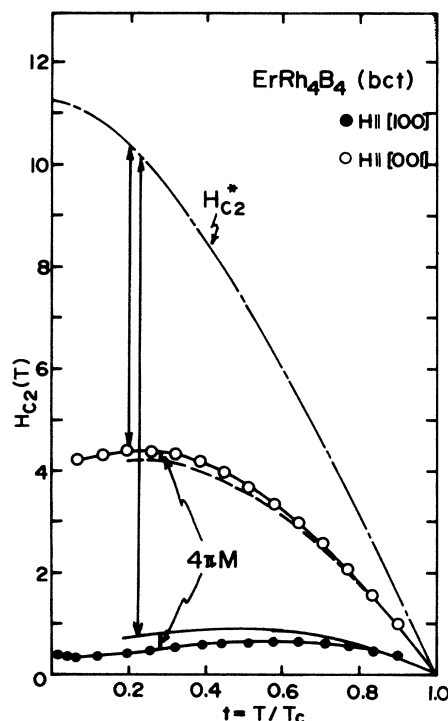


FIG. 8. Contribution of the spin polarization due to the s - f exchange interaction and the electromagnetic interaction to H_{c2} . The arrows show effects of the spin polarization. The contribution of the electromagnetic interaction is represented by $4\pi M$.

the [001] direction corresponds to $B_{c2}(T)$ because of the demagnetization and that the dashed curve represents the real $H_{c2}(T)$. It should be also noted that the experimental $H_{c2}(T)$ along the [100] direction is real $H_{c2}(T)$ and the solid curve represents $B_{c2}(T)$. It is concluded that H_{c2} along the [100] axis is almost decisively determined by the spin polarization effect and this effect also plays an important role in H_{c2} in the [001] axis. The analysis of H_{c2} is given for the temperature above T_N . However, those pair-breaking mechanisms will work for temperature below T_N .

B. Temperature dependence of H_{c2} of bct ErRh_4B_4 below T_N

As shown in Fig. 1, H_{c2} along the [100] direction increases and H_{c2} along the [110] direction decreases slightly with decreasing temperature below T_N . H_{c2} along the [110] direction becomes lower than that along the [100] direction below 0.3 K. These results are qualitatively understood as follows.

As described in the paper I, the magnetization process below T_N is understood as metamagnetism. Superconductivity coexists with the intermediate ferrimagnetic state with the magnetization of $M_0/2$ and is quenched by the occurrence of the induced ferromagnetic order for $H||[100]$. The field H_3 at which the magnetization shows the second jump becomes slightly higher with decreasing temperature (see Fig. 6 in paper I). Therefore, it is understood that H_{c2} for $H||[100]$ increases with decreasing temperature below T_N . On the other hand, the magneti-

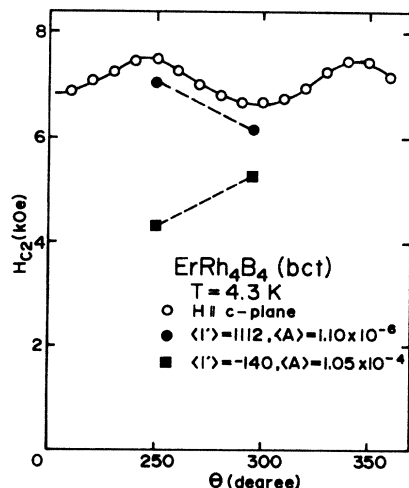


FIG. 7. Comparison of the experimental result of $H_{c2}(\theta)$ with the calculated one. From this result and Fig. 6 the solution with a negative I' value is discarded.

zation shows a one-step jump for $\mathbf{H}||[110]$ and superconductivity coexists with the canted ferromagnetic order with the magnetization of $M_0/\sqrt{2}$. The transition width at H_2 becomes narrow when the temperature decreases, and the magnetization around the transition increases abruptly at lower temperature. The magnetization after the jump at 0.1 K is larger than that at 0.5 K. Therefore, H_{c2} for $\mathbf{H}||[110]$ decreases with decreasing temperature. It is concluded that H_{c2} in the c plane is mainly determined by the effect of the magnetization through the spin polarization both above and below T_N .

H_{c2} along the $[001]$ direction decreases just above T_N and is nearly constant below T_N , as shown in Fig. 1. It seems that this behavior of H_{c2} is explained neither by the spin polarization effect due to the s - f exchange interaction nor by the electromagnetic interaction. As the magnetizations are strongly suppressed for $\mathbf{H}||[001]$, and $M(H_{c2})$ is very small compared with that in the c plane (Fig. 4), it is considered that both effects of the electromagnetic interaction and the spin polarization due to the s - f exchange interaction are small compared with the case of $\mathbf{H}||[100]$ or $\mathbf{H}||[110]$. Therefore, the other pair-breaking mechanisms such as a spin-fluctuation effect, due to the interaction between the magnetic spins, should be considered. It is not certain what kind of mechanism dominates the behavior so that H_{c2} decreases just above T_N with decreasing temperature and is nearly constant below T_N , because $M(H_{c2})$ for $\mathbf{H}||[001]$ in the temperature range around T_N has not been measured.

V. CONCLUSION

We have found the large anisotropy of H_{c2} on the bct ErRh_4B_4 single crystal, an antiferromagnetic superconductor. H_{c2} along the $[001]$ direction is extremely large compared with that in the c plane and H_{c2} in the c plane has fourfold anisotropy above T_N . In order to explain the anisotropy of H_{c2} , the electromagnetic interaction and the spin polarization due to the s - f exchange interaction are taken into account in the analysis of H_{c2} . It becomes clear that the spin polarization is very important in bct ErRh_4B_4 . The anisotropies of H_{c2} , not only between the $[001]$ axis and the $[100]$ axis but also in the c plane, are explained by this analysis. Furthermore, the s - f exchange constant I_0 and the spin-orbit scattering parameter λ_{so} are determined independently. It is confirmed that the exchange interaction between the conduction electrons and the magnetic spins is ferromagnetic and spin-orbit scattering is very strong in bct ErRh_4B_4 . Furthermore, the temperature dependence of H_{c2} in the c plane below T_N is discussed in connection with metamagnetism.

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