Ginzburg-Landau parameters for an $ErRh_4B_4$ single crystal

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The generalized Ginzburg-Landau parameters κ_1 and κ_2 have been determined for the primitive tetragonal phase of $ErRh_4B_4$ from magnetization curves of a single-crystal sphere. Along the magnetically hard c direction, $\kappa_1 \simeq \kappa_2 = 4.5$ near T_c, but rise gently with decreasing temperature to a value of 6 as the ferromagnetic transition temperature T_f is approached. Along the magnetically easy a direction, $\kappa_1 \simeq 4.5$ at T_c but drops anomalously as T_f is approached, reflecting the tendency of the system to a type-I superconductor. Furthermore, along the c direction where the effect of magnetism is mild and the system behaves similar to an ordinary type-II superconductor, the penetration depth $\lambda(0)$ and the coherence distance $\xi(0)$ are estimated to be 830 and 180 Å, respectively.

It is well known[1] that three distinct crystalline phases exist for $ErRh_4B_4$: orthorhombic ($T_c = 4.5$ K, $T_N = 0.3$ K), body-centered tetragonal (bct) ($T_c = 7.8$ K, $T_N = 0.64$ K), and primitive tetragonal ($T_c = 8.6$ K, $T_f = 0.7$ K). Here T_c refers to the superconducting transition temperature, T_N to the Néel temperature, and T_f is the ferromagnetic transition temperature.

All three phases exhibit superconductivity; however, as the temperature is lowered, the Er magnetic moments order antiferromagnetically in the orthorhombic and bct structures without destroying superconductivity. In contrast, the primitive tetragonal structure passes through the superconducting phase to reenter a normal ferromagnetic phase below T_f. For a small temperature interval ($\Delta \simeq 0.4$ K) above T_f, there is good evidence[2,3] from neutron scattering to support the coexistence of superconductivity and long-range ferromagnetic order. Due to these intriguing properties, the primitive tetragonal phase of $ErRh_4B_4$ has attracted the most attention.

Previous work[4] has shown that in this phase the Er moments are confined to the basal plane by strong crystal-field effects. As a result, the tetragonal axis is the magnetically hard direction, whereas the two equivalent a axes in the basal plane are the easy magnetic directions along which the Er moments readily align. Consequently, the crystal exhibits great anisotropy in its response to an external magnetic field.

We have previously reported on this anisotropy in detail,[5,6] as exhibited in the magnetization curves obtained with the external field along the a and c axes. In effect, the c-axis magnetism is so mild as to reveal the bare superconducting behavior of the system, whereas the a axis response reflects the dramatic effect of strong magnetism on superconductivity. Consequently, the c-axis magnetization curves, apart from mild paramagnetism, display features similar to those of an ordinary type-II superconductor. In contrast, the magnetization curves along the highly polarizable a axis show significant departure from ordinary type-II behavior.[6]

In this paper we report on the generalized Ginzburg-Landau parameters κ_1 and κ_2, as determined from magnetization curves for a single-crystal sphere of $ErRh_4B_4$. Inasmuch as κ_1 and κ_2 reflect the magnetic response of the system, we observe the effect of anisotropy on these parameters as well.

The single crystal was grown by solidification of a nonstoichiometric Er-Rh-B melt. The ingot contained a bicrystal which was cut along its grain boundary to yield two single crystals. The larger was used for neutron diffraction experiments,[3] while part of the small crystal was used to form a sphere of diameter approximately equal to 1 mm with less than 5% variation in its radius for the work presented here. With this geometry, the internal field H_i is related to the applied field H_a by the relation

$$H_i = H_a - 4\pi n M ,$$

for all crystallographic directions. In this expression, M is the magnetization and n is the demagnetizing factor, which for a sphere equals $\frac{1}{3}$.

To obtain the magnetization curves, the specimen is placed within one of two identical opposing coils. With the magnetic field increasing at a constant rate, the net signal from the two coils is proportional to the magnetic susceptibility of the specimen. Time integration of this net signal yields the magnetization as a function of the applied field.[7]

The widely used Ginzburg-Landau parameter κ is defined[8] by the equation

$$\kappa \equiv \lambda(T)/\xi(T) , \tag{1}$$

where $\lambda(T)$ is the penetration depth and $\xi(T)$ is the Ginzburg-Landau (GL) coherence length. The value of

κ is most important, as it characterizes the magnetic response of the system and determines whether a superconductor is type I or type II. Unfortunately, direct experimental measurement of λ and ξ to determine κ is not very practical. Therefore, several closely related parameters, known as the generalized GL parameters,[8,9] are defined in terms of readily measurable quantities through the relations

$$\kappa_1=\frac{H_{c2}}{\sqrt{2}H_c}\ , \tag{2}$$

$$4\pi\left[\frac{\partial M}{\partial H}\right]_{H_{c2}}=1/[\beta(2\kappa_2^2-1)]\ , \tag{3}$$

$$\kappa_3=\frac{\sqrt{2}H_c 2\pi\lambda^2}{\phi_0}\ . \tag{4}$$

In the above equations, H is the magnetic field, M is the magnetization, H_c is the thermodynamic critical field defined through

$$\frac{H_c^2}{8\pi}=-\int_0^{H_{c2}}MdH\ ,$$

H_{c2} is the upper critical field, β is a structure constant equal to 1.16 for a triangular vortex lattice, and ϕ_0 is the flux quantum. The parameters κ_1, κ_2, and κ_3 can all be evaluated from experimental measurements and are in turn used in determining κ since they all approach κ as $T\to T_c$. A more direct way of determining κ is the widely used approximation[8]

$$H_{c2}/H_{c1}=2\kappa^2/\ln\kappa\ , \tag{5}$$

which can be obtained from the magnetization curves. Equations (2) and (3) also show that the values of κ_1 and κ_2 can be conveniently determined from magnetization curves. These relations, however, have been developed for ordinary type-II superconductors and need to be modified for use with the magnetic superconductors.

As mentioned before, the superconducting magnetization curves in the c direction, apart from a small and constant normal susceptibility, display features similar to type-II superconductors.[6] Thus, in obtaining κ_1 and κ_2 from the magnetization curves, the small but finite normal-state susceptibility must be taken into account. We follow the self-consistent formulation of κ_1 and κ_2 suggested by Matsumoto, Umezawa, and Tachiki,[10] which is valid for magnetic superconductors, independent of a specific model or the form of interaction. In analogy with the nonmagnetic case, κ_1 is defined as

$$\kappa_1\equiv B_{c2}/(\sqrt{2}H_c)\ , \tag{6}$$

where H_c is the thermodynamic critical field and

$$B_{c2}=H_{c2}+4\pi M_{c2}\ .$$

Similarly, in analogy with the nonmagnetic case, parameter κ_2 is defined through the relation

$$[4\pi(\chi_s-\chi_n)]_{H_{c2}}=[(1+4\pi\chi_n)^2]_{H_{c2}}/[\beta(2\kappa_2^2)-\beta(1+4\pi\chi_n)]\ , \tag{7}$$

where χ_s and χ_n are the superconducting and normal susceptibilities and $\beta=1.16$ for a triangular vortex lattice.

In Fig. 1, plots of κ_1 are shown as functions of temperature for the c and a directions as determined from Eq. (6). The behavior of κ_1 in the c direction is similar to ordinary type-II superconductors, that is, κ_1 increases from a value of about 4.5 near T_c by about 40% as the temperature is reduced far below T_c. In contrast, κ_1 in the a direction behaves anomalously, *decreasing* as the temperature is reduced. It is interesting to note that, in this case, the value of κ_1 extrapolates to about $1/\sqrt{2}$ near $T=1.4$ K, the temperature at which the system transforms to a type-I superconductor.

When the interaction between the local moments (Er $4f$ electrons) and the conduction electrons (Rh $4d$ electrons) is primarily electromagnetic, i.e., in the absence of the so-called s-f exchange interaction, one can estimate the value of bare κ_1 by a simple scaling,[10–12] where

$$(\kappa_1)_{\text{bare}}=\kappa_1(B_{c2}/H_{c2})^{1/2}\ .$$

When this correction is applied to the data of Fig. 1, the c-direction κ_1 is hardly affected, increasing by about 3% across the board. In the a direction, the correction raises the value of κ_1 substantially, yielding a value of $\kappa_1=4.5$ near T_c, consistent with that in the c direction, but the temperature dependence of κ_1 remains anomalous as before. This indicates that the s-f exchange interaction plays an important role in determining the magnetic behavior of the system along the a axis.

Figure 2 shows κ_2 as a function of temperature for both a and c directions obtained from the magnetization curves using Eq. (7). In the c direction, where the magnetization curves are similar to those for ordinary type-II superconductors, κ_2 shows the expected gradual in-

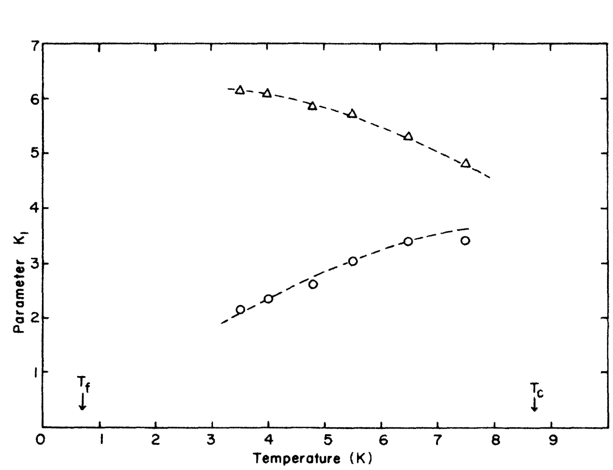

FIG. 1. Parameter κ_1 vs temperature. The upper curve shows data from the c direction; the lower curve represents the a direction data.

crease with decreasing temperature. In fact κ_2 follows the same general trend as κ_1 in the c direction.

In determining κ_2, one needs to determine the slope of the magnetization curves near H_{c2}. While this requirement presents little difficulty in the c direction, one encounters some problems in the a direction, as the magnetization curves are anomalous. In fact, as reported earlier,[6] the magnetization curves in the a direction exhibit a downward curvature between H_{c1} and H_{c2}, which becomes increasingly severe with decreasing temperature. This trend leads to the onset of a first-order phase transition at H_{c2} below 3.3 K and translates into a decreasing value of κ_2 as the temperature is lowered. However, despite the anomalous nature of the magnetization curves in the a direction, the values of κ_2, as determined from Eq. (7), are generally consistent with those for κ_1 and follow the same decreasing trend when the temperature is lowered. Indeed, κ_2 approaches a minimum value of ≈ 1.2 as the temperature nears 3.3 K. For $T < 3.3$ K, the system suffers a first-order phase transition at H_{c2} into the normal state, giving rise to an infinite slope for the magnetization curves at H_{c2}. However, due to the presence of a substantial normal-state susceptibility, Eq. (7) yields

$$\kappa_2 = [(1+4\pi X_n)/2]^{1/2}$$

and not the expected $1/\sqrt{2}$ value for $T < 3.3$ K. As the temperature is further reduced, the rise in normal-state susceptibility causes an increase in κ_2.

An estimate for the London penetration depth λ_L can be obtained by noting that the parameters κ_1, κ_2, and κ_3 all approach κ as $T \to T_c$. Hence, near T_c, Eq. (4) can be rewritten as

$$\lambda^2 = \kappa\phi_0/\sqrt{8}H_c\pi \ . \tag{8}$$

Furthermore, near T_c, the temperature dependence of the penetration depth λ is given by

$$\lambda(t) = (1/\sqrt{2})\lambda_L(1-t)^{-1/2} \ , \tag{9}$$

where, λ_L is the London penetration depth at $T=0$, and t is the reduced temperature T/T_c. When Eq. (9) is substituted in Eq. (8) and the result differentiated with respect to t, one obtains

$$\lambda_L = (\phi_0\kappa/\sqrt{2}\,|\,dH_c/dt\,|_{T_c}\pi)^{1/2} \ . \tag{10}$$

TABLE I. Values of κ as calculated from the approximate relation $(H_{c2}/H_{c1}) = (2\kappa^2/\ln\kappa)$ for several temperatures. Note that κ appears to be essentially temperature independent.

T	H_{c1}	H_{c2}	H_{c2}/H_{c1}	κ
2	440	10 000	22.7	4
3	350	9200	26.3	4.4
4	290	8000	27.6	4.6
5	250	6500	26	4.4
6	180	4800	26.6	4.4
7	125	3200	25.6	4.3
8	50	1150	23	4.0

Note that all the quantities on the right-hand side of Eq. (10) are easily obtained from the magnetization curves. Consequently, λ_L can be determined, which in turn can be used to estimate the value of the Bardeen-Cooper-Schrieffer (BCS) coherence length ξ_0 by noting that $\kappa \simeq 0.96\lambda_L/\xi_0$ in the pure limit.[8]

Bearing in mind that the resistivity ratio for our sample was only 14, one may apply the above relations with caution to the c-axis data (where the effect of local magnetism is very mild) to obtain an estimate of the superconducting parameters λ_L and ξ_0. Our data yield $\lambda_L \simeq 830$ Å and $\xi_0 \simeq 180$ Å for the c direction.

An estimate of the value of the Ginzburg-Landau coherence length at ξ can be obtained by noting that for a triangular vortex array, $B_{c2} = \phi_0/2\pi\xi^2$. For the c direction, the upper critical field H_{c2} and, hence, B_{c2} follow a nearly parabolic dependence on temperature[5] down to about 1.5 K. Extrapolation of H_{c2} to $T=0$ gives a value for $H_{c2}(0) \approx 10.3$ kG, which in turn gives $B_{c2}(0) = 10.3(1+4\pi X_n) \simeq 11$ kG. This then leads to $\xi(0) = 170$ Å, which is quite consistent with the value for $\xi_0 = 180$ Å obtained above, since, in pure superconductors, $\xi(0) = 0.74\xi_0$. Furthermore, since $\kappa = \lambda/\xi = 4.5$, assuming $\xi(0) = 170$ Å, we obtain an estimate for $\lambda(0) \simeq 765$ Å.

Finally, we may estimate the value of κ from the approximate relation in Eq. (5) using the c-direction magnetization data. Table I gives the experimental values of H_{c1} and H_{c2} as well as the value of κ for several temperatures. Clearly the average value of $\kappa \approx 4.4$ is in excellent agreement with the limiting values of κ_1 and κ_2 near T_c as obtained from Eqs. (6) and (7). Furthermore, we note that κ is temperature independent—an expected feature of type-II superconductors.

The question of whether or not the parameters κ_1 and κ_2 are of any real significance in magnetic superconductors should be examined. In their original form, these parameters were defined for ordinary type-II superconductors with no abnormal features in their magnetization curves. Broadly speaking, both parameters reflect

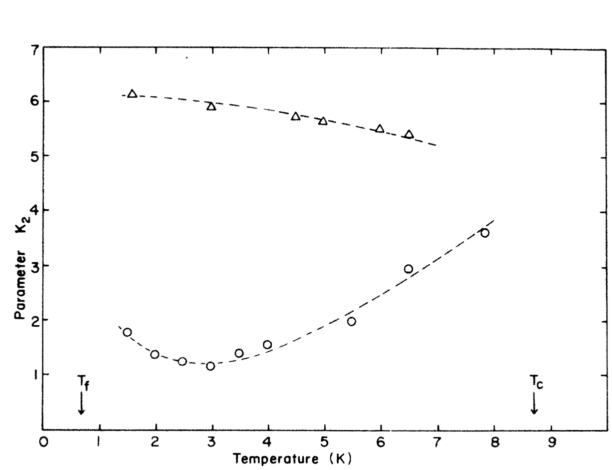

FIG. 2. Parameter κ_2 vs temperature. The upper curve shows the c direction data; the lower curve represents data from the a direction.

the ratio of H_{c2}/H_c. This is so because κ_1 is defined as $H_{c2}/\sqrt{2}H_c$, and the value of κ_2 is proportional to $[(dM/dH)_{H_{c2}})]^{-1/2}$. Note that for a given thermodynamic critical field H_c (i.e., fixed condensation energy), κ_1 increases proportionally with H_{c2}. Similarly, κ_2 becomes large when the slope $(dM/dH)|_{H_{c2}}$ decreases due to a large H_{c2}.

These considerations do not apply to materials with anomalous magnetization curves. Our data indicate that the c-direction magnetization curves are similar to the curves for ordinary type-II superconductors and, therefore, one may ascribe some relevance to the values of κ_1 and κ_2 obtained from the c-direction data. The magnetization curves for the a direction, however, are quite anomalous and cannot be used to obtain κ_1 and κ_2 reliably. Since $\kappa_1 = H_{c2}/(\sqrt{2}H_c)$ and the H_{c2}-versus-temperature curve for the a direction is severely affected by the large normal susceptibility, one can expect κ_1 to reflect this anomalous behavior as well. Furthermore, the evaluation of κ_2 for the a direction is unsatisfactory as the slope of the magnetization curves near H_{c2} is quite anomalous. Nevertheless, it is a surprising fact that if one adheres restrictly to the normal procedure for determining κ_1 and κ_2 for the a-direction data, the results are at least consistent. As Fig. 2 shows, both κ_1 and κ_2 show a *decrease* as the temperature is lowered, approaching a value close to $1/\sqrt{2}$ at about 1.4 K when the specimen changes to a type-I behavior.

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