Suppression of superconductivity of $Dy_{0.6}Y_{0.4}Rh_{3.85}Ru_{0.15}B_4$ in inclined magnetic fields

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(Submitted December 19, 2014)
Fiz. Nizk. Temp. **41**, 350–354 (April 2015)

The electrical resistance of polycrystalline $Dy_{0.6}Y_{0.4}Rh_{3.85}Ru_{0.15}B_4$ is measured for the first time near the superconducting transition temperature ($T_{c1/2} \approx 6.66\,K$) in various inclined magnetic fields ($\varphi = 0$, 45, and 90°) and plots are made of $R(T)$ and the upper critical field in dimensionless units $h^*(t)$. These dependences exhibit strong anisotropy with respect to the transport current through a sample when the orientation of the magnetic field is changed from $\varphi = 0$ to 90°. This appears to be related to texturing of the polycrystals. An analysis of the $R(T)$ and $h^*(t)$ curves shows that the magnetic field suppresses the superconducting state more strongly at $\varphi = 45°$ than at the other angles of inclination. The $h^*(t)$ dependences are analyzed in terms of the Werthamer-Helfand-Hohenberg theory and the Maki parameter α and the spin-orbital scattering parameter λ_{SO} are estimated. It is shown that for $\varphi = 0$ and 90°, $\alpha = 0$ and $\lambda_{SO} = 0$ and only the orbital contribution is observed, while for $\varphi = 45°$, $\alpha = 4.2$ and $\lambda_{SO} = 0$ and there is a substantial increase in the relative contribution of spin paramagnetic effects. It is suggested that the character of the $R(T)$ and $h^*(t)$ curves in inclined magnetic fields is influenced by the effect on the superconducting state of the internal magnetism of the dysprosium atoms, which depends on the inclination of the field.
[http://dx.doi.org/10.1063/1.4918570]

Recent studies of the rare-earth borides of rhodium in which nonmagnetic yttrium is partially replaced by magnetic dysprosium have shown that superconductivity and uncompensated internal magnetism can coexist in these materials.[1,2] Distinctive features have shown up in the behavior of some physical parameters of these compounds that are not typical of for systems with traditional superconductivity. These include a paramagnetic Meissner effect,[3,4] and an increase in the second critical field H_{c2} in $Dy_{0.8}Y_{0.2}Rh_4B_4$[2,5] and in the superconducting gaps Δ in $Dy_{0.8}Y_{0.2}Rh_4B_4$[2,5] and $Dy_{0.6}Y_{0.4}Rh_{3.85}Ru_{0.15}B_4$[6] with rising temperature (in a constant magnetic field) and magnetic field (at constant temperature). Since there are no published data on H_{c2}(T) for $Dy_{0.6}Y_{0.4}Rh_{3.85}Ru_{0.15}B_4$ we have decided to make a detailed study of the temperature dependences of the electrical resistivity of this rare-earth boride in different magnetic fields near the superconducting transition and to determine the temperature dependence of the upper critical field.

The $Dy_{0.6}Y_{0.4}Rh_{3.85}Ru_{0.15}B_4$ samples were prepared by argon-ion melting of the initial constituents followed by annealing for several days to produce a single-phase crystalline sample with an $LuRu_4B_4$ structure (spatial group *I*4/*mmm*) as determined by x-ray phase and x-ray structural analysis. Partial substitution of Rh by Ru made it possible to synthesize samples with a specified type of crystalline structure at normal pressure, which is not possible without the substitution.

Resistive and magnetoresistive measurements were made using a standard four-probe circuit on a Quantum Design PPMS-9 automatic system. A constant magnetic field was created by a superconducting solenoid. The ac electrical resistance was measured ($I = 8$ mA, $f = 97$ Hz) for different orientations of the magnetic field. The holder in the cryostat was equipped with a system for automatically rotating the backing with the sample around a horizontal axis using a high-resolution stepping motor. The angle φ is the angle between the directions of the current and the external magnetic field. For $\varphi = 0°$, the measurement current in the sample was directed parallel to the magnetic field ($I||H$), and an angle $\varphi = 90°$ corresponds to having the measurement current in the sample perpendicular to the magnetic field. For orientations $\varphi = 0°$ and $\varphi = 45°$, measurements were made for magnetic fields of 0–9 kOe, and for orientations $\varphi = 90°$, for fields in the range 0–36 kOe.

$H_{c2}(T)$ was determined from plots of the resistive transitions obtained in different constant magnetic fields as the temperature was varied. $H_{c2}(T)$ was taken to be the value of H for which $R = 0.5R_N$ (R_N is the normal resistance of the sample at a temperature above the superconducting transition temperature). Since the resistance curves $R(T)$ for the transitions in the neighborhood of $R \sim 0.5R_N$ are quite steep, there is no reason to expect significant errors in the determination of $H_{c2}(T)$ owing to broadening of the resistive transitions caused by the motion of free flux vortices or the inhomogeneity of the samples.

Figure 1 shows the temperature dependences of the electrical resistance of one of the test samples of $Dy_{0.6}Y_{0.4}Rh_{3.85}Ru_{0.15}B_4$ in different magnetic fields for three orientations of the field relative to the sample ($\varphi = 0°$ (Fig. 1(a)), $\varphi = 45°$ (Fig. 1(b)), $\varphi = 90°$ (Fig. 1(c))). For all field orientations, in fields below 8 kOe the $R(T)$ curves have a shape typical of the transition into the superconducting state, which shows up as a sharp drop in the electrical resistance below some temperature (T_c) with the resistance going to zero at lower temperatures. With increasing magnetic field the $R(T)$ curves shift toward lower temperatures and become broader, which is typical of superconductors of the second kind (the appearance of resistance associated with the penetration of vortices). Here the highest values of T_c for equal magnetic fields are observed for $\varphi = 90°$ and the lowest, for $\varphi = 45°$.

The unusual behavior of the ohmic losses for $\varphi = 45°$ in fields above 8 kOe is noteworthy. Instead of a sharp drop in the electrical resistance, as happens for the other orientations, at temperatures below T_c here the electrical resistance falls only to a finite value $R_{\min} \approx 0.4R_N$ in the $R(T)$ curve and the resistance starts to rise rapidly when the temperature is reduced further. With increasing field the minimum shifts toward lower temperatures and its depth is greatly reduced ($R_{\min} \approx 0.9R_N$). It follows from this that below the superconducting transition temperature the $R(T)$ curves depend more strongly on the magnetic field for $\varphi = 45°$ (Fig. 1(b)) than for $\varphi = 0$ and 90°.

This strong anisotropy of the $R(T)$ curves in a magnetic field for certain orientations of the sample may indicate the presence of texturing (predominant orientation of individual crystallites) in the polycrystalline sample. If this is actually so, then the presence of a minimum in $R(T)$ for $\varphi = 45°$ below T_c can be explained by the fact that, for this orientation of the field, a magnetically ordered state that coexists with superconductivity changes in the magnetic field in a way such that the uncompensated total magnetic moment increases. The latter also leads to strong suppression of the superconducting state even in fields on the order of 8 kOe. A minimum in $R(T)$ in a magnetic field has been observed previously in other magnetic superconductors, such as $NdRh_4B_4$[7] and $Dy_{1.2}Mo_6S_8$,[8] and has been attributed to field induced magnetic ordering of the Nd or Dy ions for $T_M < T_c$, which suppressed or entirely destroyed the superconducting state. Our case differs in that the superconductivity and the magnetic ordering, which shows up for $T_M > T_c$, coexist below T_c and the minimum is a consequence of a change in the already existing magnetic structure under the influence of a magnetic field with a certain orientation. We are unaware of any other reports of similar observations.

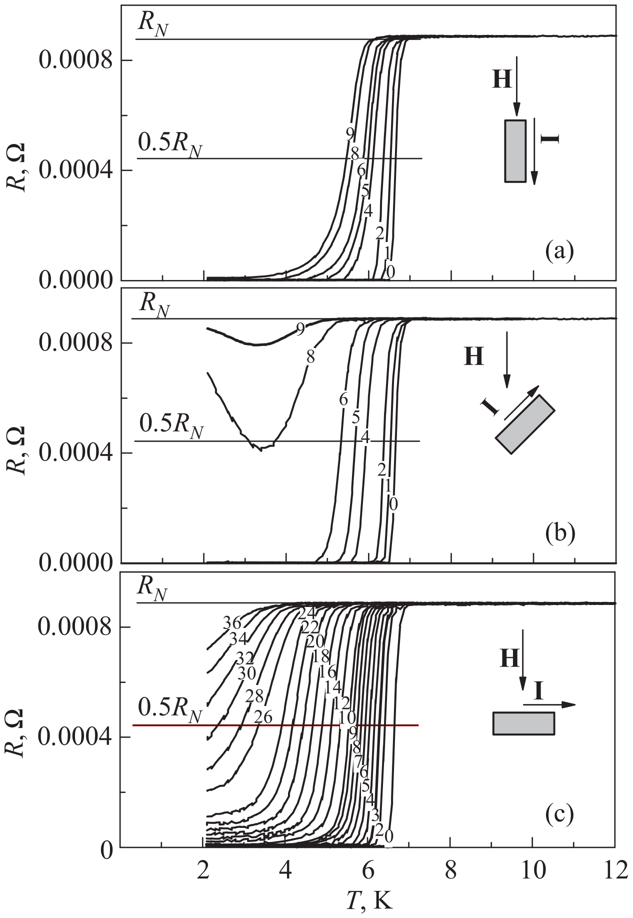

FIG. 1. Electrical resistance as a function of temperature for three orientations of the magnetic field relative to the sample ($\varphi = 0°$, $I||H$ (a); $\varphi = 45°$ (b); $\varphi = 90°$, $I_{\|}H$ (c)) in magnetic fields of 0–9 kOe for $\varphi = 0°$ (a) and 45° (b), and 0–36 kOe for $\varphi = 90°$ (c). The numbers on the curves indicate the magnetic fields in kOe for which the $R(T)$ curves were obtained.

We have used the data of Fig. 1 to find experimental values of the upper critical fields and to plot their temperature dependence ($h^*(t)$, where $t = T/T_c$) in dimensionless units for different orientations of the magnetic field relative to the test sample (see Fig. 2). h^* is determined from the experimental data in the following way:[9]

$$h^* = -\frac{H_{c2}}{(dH_{c2}/dt)|_{t=1}}. \quad (1)$$

A representation of the temperature dependence of the upper critical field in dimensionless units is needed in order to analyze our results in terms of the Werthamer-Helfand-Hohenberg

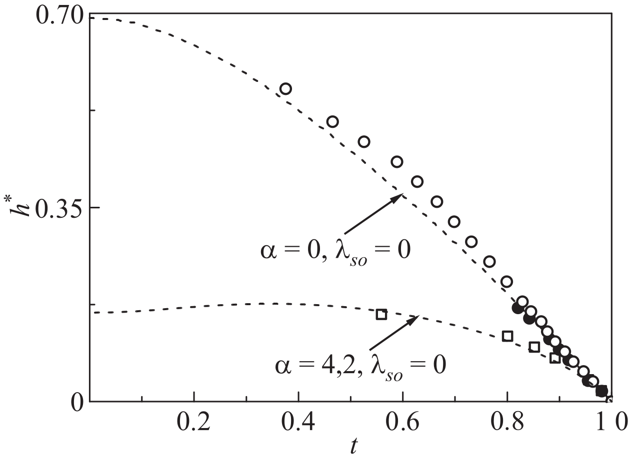

FIG. 2. The dimensionless upper critical magnetic field h^* as a function of the dimensionless temperature $t = T/T_c$ for $\varphi = 0°$ (●), 45° (○), and 90° (□). The dashed curves were calculated using the WHH theory.

(WHH) theory, which includes orbital, paramagnetic, and spin-orbital mechanisms for the suppression of superconductivity in simple superconductors.[9]

In Fig. 2 the symbols ●, ○, and □ show the rescaled experimental $h^*(t)$ curves for $\varphi = 0$, 45, and 90°, respectively. The dashed curves are plots of $h^*(t)$ calculated using the WHH theory:

$$\ln\frac{1}{t} = \left(\frac{1}{2} + \frac{i\lambda_{so}}{4\gamma}\right)\psi\left(\frac{1}{2} + \frac{\bar{h} + \frac{1}{2}\lambda_{so} + i\gamma}{2t}\right) + \left(\frac{1}{2} - \frac{i\lambda_{so}}{4\gamma}\right)\psi\left(\frac{1}{2} + \frac{\bar{h} + \frac{1}{2}\lambda_{so} - i\gamma}{2t}\right) - \psi\left(\frac{1}{2}\right), \quad (2)$$

where ψ is the digamma function

$$\gamma \equiv \left[\left((\alpha\bar{h})^2 - \left(\frac{1}{2}\lambda_{so}\right)^2\right)\right]^{\frac{1}{2}}, \quad (3)$$

and

$$h^* \equiv (\pi^2/4)\bar{h}. \quad (4)$$

The following parameters were used in the calculations: α, the Maki parameter which describes the relative contribution of the paramagnetic and orbital effects in the absence of spin-orbital scattering, and λ_{SO}, the spin-orbital scattering parameter. The best fits were obtained for $\varphi = 0°$ and $\varphi = 90°$ with $\alpha = 0$ and $\lambda_{SO} = 0$ and for $\varphi = 45°$ with $\alpha = 4.2$ and $\lambda_{SO} = 0$. These values of the fit parameters indicate that, according to the WHH theory, only the orbital contribution exists for $\varphi = 0$ and 90° ($\alpha = 0$). For $\varphi = 45°$ and $\alpha = 4.2$, there is a substantial increase in the relative contribution of spin paramagnetic effects, apparently associated with changes in the magnetic subsystem of dysprosium. In both cases, $\lambda_{SO} = 0$, which indicates an insignificant spin-orbital contribution. It can be seen in Fig. 2 that the experimental $h^*(t)$ curve lies somewhat above the theoretical curve. This behavior of $h^*(t)$ can be explained by an anisotropy (which can lead to an increase of 20%–30% in the critical field)[10] and by the presence of strong coupling in the superconducting condensate (H_c2 can increase by a factor of 1.3).[11] The possibility of strong coupling follows from a study of the superconducting gap,[6] which showed that the ratio $2\Delta/k_BT_c$ in this material can be as high as 4 or more, which exceeds the value of 3.52 for traditional superconductors with weak coupling.

To summarize, in this paper we have made the first detailed measurements of the electrical resistance of $Dy_{0.6}Y_{0.4}Rh_{3.85}Ru_{0.15}B_4$ near the superconducting transition temperature ($T_{c1/2} \approx 6.6$ K) in various inclined magnetic fields ($\varphi = 0$, 45, and 90°). Temperature dependences $h^*(t)$ of the upper critical magnetic fields in dimensionless units have been obtained for inclinations $\varphi = 0$, 45, and 90° of the magnetic field relative to the sample. An anisotropy has been observed in the $R(T)$ and $h^*(t)$ curves for different orientations of the sample in a magnetic field. It may be indicative of texturing in the polycrystalline sample. An analysis of the $R(T)$ and $h^*(t)$ curves shows that the magnetic field suppresses the superconducting state most strongly at $\varphi = 45°$. It has been found that, within the range of temperatures studied here, the upper critical field increases monotonically as the temperature is lowered. The behavior of $h^*(t)$ has been analyzed in terms of the WHH theory. For this analysis, the Maki parameter α, which corresponds to the relative contributions of spin paramagnetic and orbital effects, and the spin-orbital scattering parameter λ_{SO} were determined. It was found that for $\varphi = 0$ and 90°, $\alpha = 0$ and $\lambda_{SO} = 0$ (i.e., only an orbital contribution exists), while for $\varphi = 45°$, $\alpha = 4.2$ and $\lambda_{SO} = 0$ (which indicates a substantial increase in the relative contribution of spin paramagnetic effects). It is suggested that these features of the superconducting parameters may be related to an increase in the internal total magnetic moment of the dysprosium magnetic subsystem under the influence of a magnetic field in certain directions. The latter leads to considerable suppression of the superconducting state.

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Translated by D. H. McNeill