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Anisotropy of electric resistance and upper critical field in magnetic superconductor $Dy_{0.6}Y_{0.4}Rh_{3.85}Ru_{0.15}B_4$

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ABSTRACT

We have measured temperature dependencies of the electric resistance R and upper critical magnetic field H_{c2} of a magnetic superconductor $Dy_{0.6}Y_{0.4}Rh_{3.85}Ru_{0.15}B_4$. The measurements were made for different angles φ of the magnetic field inclination to the direction of measuring current and revealed strong anisotropy of the behavior of $R(T)$ and the values of $H_{c2}(T)$. By using the Werthamer–Gelfand–Hohenberg theory, we determined the Maki parameter α and the parameter of the spin-orbital interaction. For $\varphi = 0^\circ$ and 90° both parameters are close to zero, thus the magnitude of $H_{c2}(0) \approx 38$ kOe is basically limited by the orbital effect. At $\varphi = 45^\circ$, a large value of $\alpha = 4.2$ indicates dominating role of the spin-paramagnetic effect in the suppression of $H_{c2}(0)$ down to 8.8 kOe. We suggest that such behavior of $R(T)$ and $H_{c2}(T)$ is caused by internal magnetism of the Dy atoms which may strongly depend on the magnetic field orientation.

1. Introduction

Ternary compounds whose structures include a regular sublattice of magnetic moments are attractive objects for studying the influence of magnetism on superconductivity. Among these materials, the most famous are $PbMo_6S_8$-type “Chevrel phases” and ternary rare-earth rhodium borides [1]. The physical properties of quadruple compounds $Dy_{1-x}Y_xRh_4B_4$ having a body-centered tetragonal $LuRu_4B_4$-type crystal structure deserve special attention due to a great number of interesting features of these materials. For instance, it was found [2,3] that the magnetic ordering of Dy atoms may occur at the temperature T_M higher than the superconducting transition temperature T_c and thus may coexist with superconductivity down to very low temperatures. It was established in [3] that $Dy_{1-x}Y_xRh_4B_4$ belongs to materials with intrinsic ferrimagnetism, and the transition temperature T_M strongly depends on the concentration of non-magnetic yttrium: it falls with increasing Y concentration from 37 K in $DyRh_4B_4$ down to 7 K in $Dy_{0.2}Y_{0.8}Rh_4B_4$. On the contrary, T_c increases with the Y concentration from 4.7 K for $DyRh_4B_4$ to 10.5 K in YRh_4B_4 [2]. Measurements of the specific heat of $Dy_{0.8}Y_{0.2}Rh_4B_4$ ($T_M = 30.5$ K, $T_c = 5.9$ K) indicate emergence of another magnetic phase transition below 2.7 K [3]. Recently, anomalies of some physical quantities, unusual for systems with conventional superconductivity, were discovered in $Dy_{1-x}Y_xRh_4B_4$: the paramagnetic Meissner effect [4,5] and non-monotonic temperature dependencies of the upper critical magnetic field H_{c2} and the superconducting gap [3,6,7].

Another specific feature of this class of magnetic superconductors is the change of the type of magnetic interactions in the Dy subsystem under partial replacement of rhodium by ruthenium. As shown in [8], antiferromagnetic ordering in $Dy(Rh_{1-y}Ru_y)_4B_4$ holds for $y < 0.5$ and changes to a ferromagnetic one for $y > 0.5$. The superconducting transition temperature also varies with the Ru concentration [8].

Thus, the study of physical properties of the borides family $Dy_{1-x}Y_x(Rh, Ru)_4B_4$ with various content of dysprosium (responsible for the magnetic interactions) and of ruthenium and rhodium (responsible for both the magnetic interactions and superconductivity) is of great interest to explore the coexistence of superconductivity and magnetism and the possibility of appearance of unconventional superconductivity. For this purpose, we studied in this paper the behavior of the electric resistance in the vicinity of

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the superconducting transition and the upper critical field in the compound $Dy_{0.6}Y_{0.4}Rh_{3.85}Ru_{0.15}B_4$ at different orientations of external magnetic field with respect to the direction of measuring electric current. Preliminary results were briefly reported in [9].

2. Results and discussion

The $Dy_{0.6}Y_{0.4}Rh_{3.85}Ru_{0.15}B_4$ compound has been prepared by the argon arc melting of initial components, followed by annealing within a few days. The resulting single-phase polycrystalline ingot had a $LuRu_4B_4$-type crystal structure (space group I4/mmm) testified by the X-ray phase and structural analyses. At this concentration of ruthenium, it is possible to synthesize samples with such structure at the normal pressure, in contrast to the quadruple compounds $Dy_{1-x}Y_xRh_4B_4$, which gain the required structure only during the synthesis under a pressure of 8 GPa. The samples were cut from the ingot in the form of parallelepipeds whose lengths were about 6 mm and the cross-sectional area was 1 × 1 mm^2. The measurements of the electrical resistance $R(T)$ were performed on a Quantum Design PPMS-9 automatic system using a standard four-probe circuit with an alternating current ($I = 8$ mA, $f = 97$ Hz) in the temperature range 2 – 12 K and magnetic fields up to 36 kOe produced by a superconducting solenoid. The sample holder was equipped with a system for automatic rotation of the substrate with the sample by a stepping motor of high resolution which allowed the measurement of $R(T)$ for different angles φ of inclination of the external magnetic field H to the direction of the current. The superconducting transition temperature measured in the middle of the resistive transition in zero magnetic field was 6.66 K.

Fig. 1 presents the temperature dependencies of the sample resistance in different magnetic fields of three orientations: $\varphi = 0°$, 45° and 90° (panels (a), (b) and (c), respectively). For the angles $\varphi = 0°$ and 45°, the experiments were made in magnetic fields $H = 0 - 9$ kOe and for $\varphi = 90°$ – at $H = 0 - 36$ kOe. The shape of $R(T)$ in the range of fields 0 – 6 kOe is typical for the superconducting transition: a sharp fall of the resistance below a certain temperature followed by its disappearance at lower temperatures. Another type of the behavior of $R(T)$ was observed at $\varphi = 45°$ in the fields larger than 6 kOe. In this case, the resistance decreases only down to a certain finite value $R_{min} \approx 0.4R_N$ (R_N is the sample resistance of in the normal state), and then, as the temperature is lowered further, $R(T)$ rapidly increases. With further increase of the field, the observed minimum of $R(T)$ shifts to lower temperatures and reduces in its depth up to $R_{min} \approx 0.9R_N$. Thus it can be argued that the destruction of superconductivity at the magnetic field orientation $\varphi = 45°$ (Fig. 1b) begins in the fields much smaller than at the orientations $\varphi = 0°$ and 90°.

Such strong anisotropy of $R(T)$ is apparently due to the presence of the texture (preferred orientation of individual crystallites) in a polycrystalline sample and the coexistence of the magnetic and superconducting orderings. Along this line of reasonings, the minimum in $R(T)$ for $\varphi = 45°$ can be attributed to stronger (compared to other orientations) enhancement of an uncompensated magnetic moment with increasing magnetic field. This excess magnetism leads to significant suppression of the superconducting state in the fields > 8 kOe at $\varphi = 45°$, while for other orientations, superconductivity holds up to several tens of kOe.

We note that a minimum of $R(T)$ in magnetic fields has been earlier observed in other magnetic superconductors such as $NdRh_4B_4$ [10] and $Dy_{1.2}Mo_6S_8$ [11]. It has been attributed to the induction of the magnetic ordering of Nd and Dy ions by an external magnetic field at the transition temperature $T_M < T_c$, which leads to destruction of the superconducting state (reentrant superconductivity). However, in our case, the superconductivity and the magnetic order, which emerges at $T_M > T_c$, coexist below T_c, and

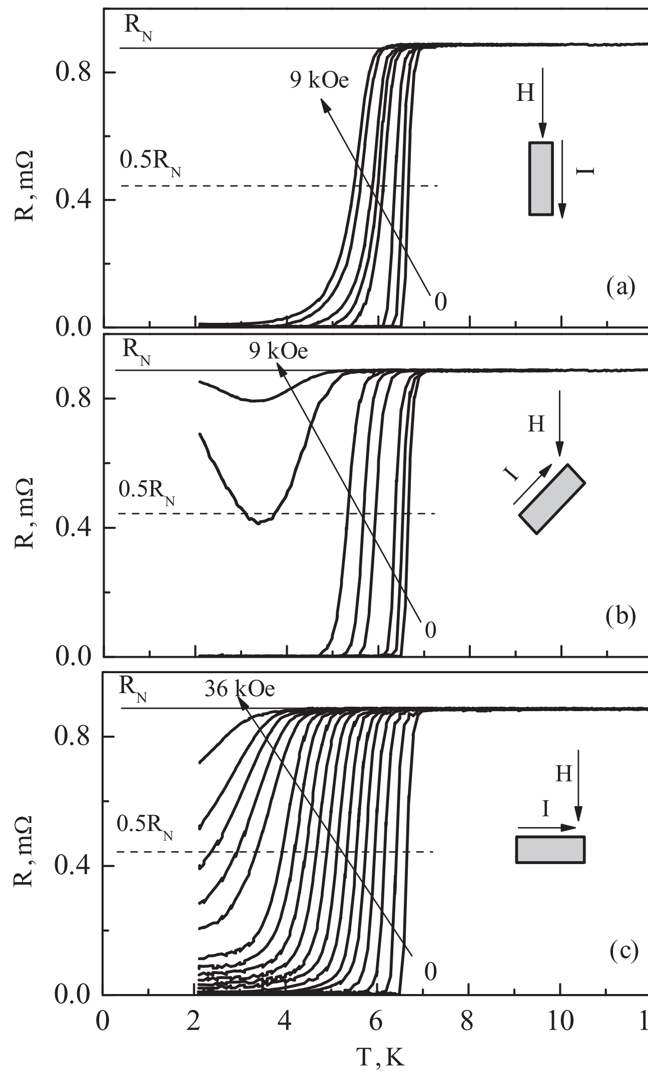

Fig. 1. Temperature dependencies of the resistance for three orientations of the magnetic field relative to the longitudinal sample axis: $\varphi = 0°$, $H \| I$ (a); $\varphi = 45°$ (b); $\varphi = 90°$, $H \perp I$ (c) in magnetic fields 0, 1, 2, 4, 5, 6, 8, 9 kOe for $\varphi = 0°, 45°$ and 0 – 36 kOe through 2 kOe for $\varphi = 90°$.

the minimum in $R(T)$ can be explained as the result of changes in the existing magnetic structure caused by the magnetic field of specific orientation.

Using the data of Fig. 1 and accepting for $H_{c2}(T)$ the values of the external magnetic field and the temperature, at which $R(H, T) = 0.5R_N$, we plotted the experimental temperature dependencies of the upper critical field depicted by circles and squares in Fig. 2. In contrast to the previous Andreev spectroscopy data [3,6,7], we did not found any non-monotony in the behavior of $H_{c2}(T)$ that possibly reflects certain ambiguity in the interpretation of the results of the point-contact measurements in nonhomogeneous samples; a certain role in this difference may be also played by the admixture of ruthenium in our samples. Dashed curves in Fig. 1 show the dependencies $H_{c2}(T)$ calculated from the equation of the Werthamer–Gelfand–Hohenberg (WHH) theory [12]:

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

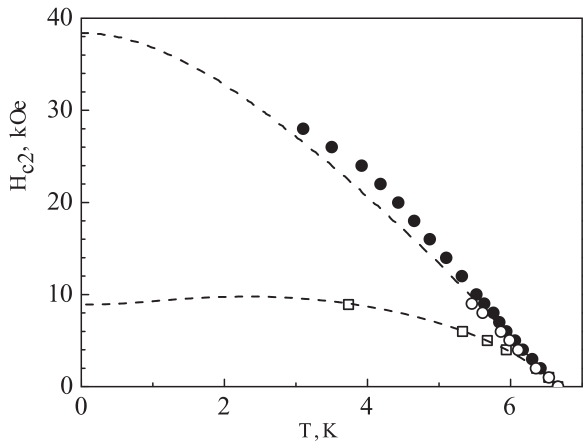

Fig. 2. Temperature dependencies of the upper critical magnetic field H_{c2} for $\varphi = 0^\circ$ (•), 45° (□) and 90° (○). Dashed lines show the results of the WHH theory with fitting parameters of the spin-paramagnetic and spin-orbital interaction.

where $\psi(x)$ is the digamma function, $\gamma = \sqrt{(\alpha\bar{h})^2 - (\lambda_{so}/2)^2}$, and

$$t = \frac{T}{T_c}, \qquad \bar{h} = -\frac{4}{\pi^2}\frac{H_{c2}}{(dH_{c2}/dt)_{t=1}} \tag{2}$$

are the reduced temperature and critical magnetic field, respectively. In our calculations, we use the fitting values of the Maki parameter α which describes relative contribution of the spin-paramagnetic effect and the parameter λ_{so} of the spin-orbit scattering. The best fit for $\varphi = 0^\circ$ and 90° gives $\alpha = \lambda_{so} = 0$, i.e., only the orbital effect is responsible for the suppression of superconductivity, whereas at $\varphi = 45^\circ$ we obtain a rather large value of $\alpha = 4.2$ ($\lambda_{so} = 0$) which indicates essential contribution of the spin-paramagnetic effect.

Fig. 2 shows that for $\varphi = 0^\circ$, the experimental values of $H_{c2}(T)$ at the temperatures below $0.8T_c$ slightly exceed the maximum possible calculated ones. This could be explained either by the effect of anisotropy (may lead to increase in H_{c2} by 20 – 30% [13]) or by the presence of strong coupling in the superconducting condensate (can enhance H_{c2} by 1.3 times [14]), which are beyond the frameworks of the WHH theory. The possibility of the strong coupling in this material follows from the results of the Andreev spectroscopy of the superconducting gap Δ [7], according to which the ratio $2\Delta/kT_c$ can reach 4 or even higher values, larger than the value 3.52 for conventional superconductors with the weak coupling.

The orbital critical field at $T = 0$ can be calculated by using the formula of the WHH theory for the dirty limit [12],

$$H_{c2}^{orb}(0) = -0.69T_c\,(dH_{c2}/dT)_{T=T_c}, \tag{3}$$

while the upper critical field can be estimated as [15]

$$H_{c2}(0) = \frac{H_{c2}^{orb}(0)}{\sqrt{1+\alpha^2}}. \tag{4}$$

As is obvious from Fig. 2, initial slopes of $H_{c2}(T)$ near T_c are approximately equal for all orientations of the magnetic field. According to (3), this results in a universal (angle-independent) value of $H_{c2}^{orb}(0) \approx 38$ kOe. Since at $\varphi = 0^\circ$ and 90° the fitting value of the Maki parameter is close to zero, we conclude from (4) that the orbital field at these orientations fully determines the magnitude $H_{c2}(0) = H_{c2}^{orb}(0)$ of the upper critical field at zero temperature. The estimate of a small paramagnetic contribution can be obtained from the relation $\mu_B H_{c2}^{p}(0) = 1.84kT_c$ for the Chandrasekhar-Clogston limit [16,17] (μ_B is the Bohr's magneton) which gives the critical paramagnetic field $H_{c2}^{p}(0) = 122.5$ kOe.

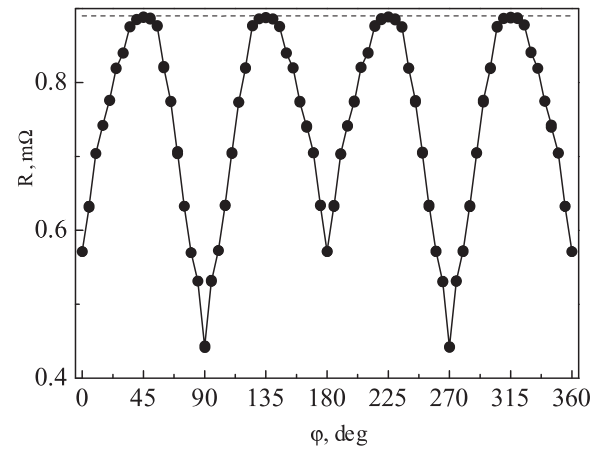

Fig. 3. Angle dependencies of the electric resistance within the range $\varphi = 0^\circ - 360^\circ$ at the temperature 5.75 K corresponding to the middle of the superconducting transition in the field of 8 kOe. Dashed line depicts the angle-independent resistance at $T = 9$ K, $H = 8$ kOe (normal state).

Then, using the equation [15]

$$\alpha = \sqrt{2}\frac{H_{c2}^{orb}(0)}{H_{c2}^{p}(0)} \tag{5}$$

we found a rather small value $\alpha \approx 0.4$ for the Maki parameter which explains unobservability of the spin effects at the experimental dependencies $H_{c2}(T)$ at $\varphi = 0^\circ$ and 90°.

As noted above, the large value of the Maki parameter $\alpha = 4.2$ at $\varphi = 45^\circ$ implies that the spin-paramagnetic effect plays the main role in the suppression of superconductivity at this orientation of the magnetic field. In this case, equation (4) gives $H_{c2}(0) \approx 8.8$ kOe, i.e., by 4.3 times smaller than its value for the orientations $\varphi = 0^\circ$ and 90°. Correspondingly, the magnitude of the critical paramagnetic field $H_{c2}^{p}(0) = 12.8$ kOe found from (5) appears to be much smaller than at $\varphi = 0^\circ$ and 90°. These results give additional arguments in the benefit of our assumption about rearrangement of the magnetic structure and formation of an excess magnetic moment induced by the external magnetic field. This considerably enhances the effective magnetic field acting on the electron spins which causes destruction of the Cooper pairs.

In order to obtain additional information about the effect of the magnetic field inclination on the suppression of superconductivity, we measured the angle dependencies of the sample resistance within the range $\varphi = 0^\circ - 360^\circ$ (see Fig. 3) at the temperature 5.75 K which corresponds to the middle of the superconducting transition at $\varphi = 0$ in the field of 8 kOe. Fig. 3 shows that with the increase in the angle, the resistance first grows to a maximum value R_N at $\varphi = 45^\circ$, then begins to drop with a minimum at $\varphi = 90^\circ$. All highs in $R(\varphi)$ repeat themselves through 90°, and lows – through 180° (the magnitude of minimum ohmic losses for $\varphi = 0^\circ$ and 180° is smaller than that at $\varphi = 90^\circ$ and 270°). Thus, the superconductivity is most strongly suppressed in the magnetic fields directed at the angles 45° plus multiple of 90° relative to the longitudinal sample axis. The fields inclined at the angles by multiple of 90° have the weakest impact on the superconducting state. The dashed line in Fig. 3 indicates the experimental data obtained at $T = 9$ K in the field of 8 kOe and demonstrates independence of the sample resistance of the field direction in the normal state.

3. Summary

We have measured the resistance R and the upper critical magnetic field H_{c2} of the magnetic superconductor $Dy_{0.6}Y_{0.4}Rh_{3.85}Ru_{0.15}B_4$ at different angles φ of inclination of the magnetic field relative to the longitudinal sample axis. The temperature dependencies $R(T)$ and $H_{c2}(T)$ are strongly anisotropic in the superconducting state, whereas the rotation of the magnetic field in the normal state has no effect on its resistive properties. Suppression of superconductivity is most pronounced at $\varphi = 45^\circ$ plus multiple of 90° ($H_{c2}(0) \approx 8.8$ kOe), while at the angles $\varphi = 0^\circ$ and 90° the effect of the magnetic field on the superconducting state is weakest, and the calculated magnitude of $H_{c2}(0)$ reaches 38 kOe. A minimum in $R(T)$ in large enough fields was observed at $\varphi = 45^\circ$ which resembles reentrance effects in some magnetic superconductors near the point of transition to the magnetically ordered state. However, in our case, this minimum most likely occurs due to restructuring of already existed magnetic ordering.

Analysis of the behavior of $H_{c2}(T)$ within the framework of the WHH theory shows that for $\varphi = 0^\circ$ and 90° the Maki parameter α and the parameter λ_{so} of the spin-orbit scattering are close to zero, i.e., only the orbital effect is responsible for the suppression of superconductivity. This is confirmed by the estimate of α obtained from the calculated paramagnetic limit. On the contrary, at $\varphi = 45^\circ$, the Maki parameter was found to be large ($\alpha = 4.2$, $\lambda_{so} = 0$) which manifests the dominating role of the spin-paramagnetic depairing mechanism. We suggest that the above mentioned features of the behavior of superconducting parameters may be associated with the growth of a spontaneous magnetic moment of the dysprosium subsystem induced by an external magnetic field of specified orientation. At the same time, one cannot exclude the existence of an unconventional pairing mechanism, such as a triplet pairing, in this material.

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