Maltese Cross Anisotropy in Antiferromagnetic State of Metallic Ho$_{0.5}$Lu$_{0.5}$B$_{12}$ with Dynamic Charge Stripes

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The model fcc system Ho$_{0.5}$Lu$_{0.5}$B$_{12}$ with substitutional disorder of Ho-Lu ions, dynamic charge stripes, and a complex AF ground state has been studied by magnetoresistance at low temperatures in magnetic field up to 80 kOe. For this non-equilibrium AF metal the angular $H-\varphi-T$ magnetic phase diagram in the form of a “Maltese cross” has been constructed experimentally for the first time. It was shown that the dramatic symmetry lowering of the AF ground state in this rare earth dodecaboride should be attributed to the redistribution of charge carriers into dynamic charge stripes due to RKKY oscillations of the conduction electrons spin density. This redistribution causes extraordinary changes in the indirect exchange interaction between magnetic moments of Ho$^{3+}$ ions and results in the emergence of a number of various magnetic phases and phase transitions.

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1. Introduction

Strongly correlated electron systems (SCES) are of great interest due to their unusual properties [1]. In these compounds, charge, spin, orbital, and structural degrees of freedom are simultaneously active, leading to complex phase diagrams. Moreover, the competition of various conducting and insulating phases, such those detected in manganites, can lead to extraordinary effects that may be useful in practical applications [1, 2]. The analysis of SCES is partly hampered by their complex structure and chemical composition. Taking this into account, studies of highly symmetric compounds with a simple fcc lattice and instabilities of the electronic or magnetic subsystem are very useful. Recently it was shown that below cage-glass transition temperature ($T^* \approx 60$ K) in dodecaborides RB$_{12}$, due to the cooperative dynamic Jahn-Teller (JT) effect on cuboocahedra B$_{12}$, dynamic charge stripes appear along the principal crystallographic direction (110) [3]. Moreover, it was shown that Ho$_{0.1}$Lu$_{0.9}$B$_{12}$ are metals with AF ground state and strong concurrence of large positive and negative effects in magnetoresistance [4] similar to those detected in manganites. Therefore, in order to shed more light on the origin of both various regimes of carrier scattering and numerous magnetic phases in AF metals with dynamic charge stripes, we present here a detailed study of angular, magnetic field and temperature dependencies of magnetoresistance $\Delta \rho/\rho$.

2. Experimental details

The high quality single crystal of Ho$_{0.5}$Lu$_{0.5}$B$_{12}$ ($T_N \approx 3.4$ K) was grown by vertical crucible-free inductive floating zone melting in an inert gas atmosphere. The high quality of the crystal was controlled by X-ray diffraction, the Laue backscattering-patterns and electron microprobe analysis. Magnetoresistance was studied in a temperature range 2–5 K in magnetic fields up to 80 kOe. In experiment with sample rotation the direction of external magnetic field was varied in the range of angles $\varphi = 0-360^\circ$ (where $\varphi$ is the angle between vectors $H$ and $\mathbf{n}$, $\mathbf{n} \parallel (110)$ is the normal to the sample surface). Resistivity was measured by standard DC four-probe technique with the orientation of measuring current $\mathbf{I} \parallel [110]$.

3. Results and discussion

Figure 1 shows the temperature dependencies of magnetic susceptibility ($H = 100$ Oe), heat capacity and resistivity of Ho$_{0.5}$Lu$_{0.5}$B$_{12}$ in a wide area around the antiferromagnet (AF) — paramagnet (P) phase transition. Such behavior of magnetic susceptibility with a maximum at Neel temperature ($T_N = 3.4$ K) is typical

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for antiferromagnets. It is worth noting the sharp step-like anomaly of heat capacity and the resistivity maximum near $T_N$ with a subsequent decrease towards lower temperatures.

Figure 2 shows the magnetoresistance (MR) dependencies $\Delta \rho / \rho = (\rho(H) - \rho(0))/\rho(0)$ of Ho$_{0.5}$Lu$_{0.5}$B$_{12}$ in the temperature range 1.9–3.3 K in magnetic fields up to 80 kOe for three principal directions of the cubic structure $H \parallel [001]$, $H \parallel [110]$, and $H \parallel [111]$ (see Fig.2a, 2b and 2c, respectively). Furthermore, the amplitude of linear positive MR (pMR) in low magnetic fields (AF-state) rises with temperature decrease, reaching 20% for $H \parallel [001]$ and $T = 1.9$ K. It should be emphasized that for $H \parallel [111]$ there are two linear pMR intervals with significantly different slopes in contrast to $H \parallel [001]$ and $H \parallel [110]$ (Fig. 2a and 2b), where the slopes of MR curves are about the same.

Magnetoresistance results allow us to construct the magnetic phase diagrams of Ho$_{0.5}$Lu$_{0.5}$B$_{12}$ for magnetic fields aligned along the three principal cubic directions, namely $H \parallel [001]$, $H \parallel [110]$, and $H \parallel [111]$ (see Fig. 3). It is easy to see that the phase boundary between AF and P states is about isotropic. At the same time are the phase boundaries in the AF state very different — dependent on the crystallographic direction. In order to construct $H$–$\varphi$ phase diagram we have measured the full range angular dependencies $\rho(\varphi)$ ($\varphi = 0$–$360^\circ$) at $T = 2.1$ K for several fixed $H$ values (see Fig. 4). Below $H \sim 6$ kOe only small $\rho(\varphi)$ changes are observed. With increasing magnetic field in the range between 6 and 7 kOe a sharp resistivity minimum forms along $H \parallel [111]$. In interval 9–20 kOe we observe strong peaks with resistivity maxima for field directions $H \parallel [001]$ and $H \parallel [110]$, and an abrupt drop appears on $\rho(\varphi)$ curves between these two peaks. In higher fields (30–32 kOe) the $\rho(\varphi)$ curve changes its form, corresponding to AF-P phase transition (see Fig. 3 for $T = 2.1$ K). It is worth noting the strong anisotropy in angular dependencies even in the paramagnetic phase (Fig. 4d). These data allow us to plot a 3D view of MR in cylindrical coordinates (Fig. 5).

In Fig. 6 for visualization of phase boundaries in the $(H, \varphi)$ planes at $T = 2.1$ K, we present the projection of MR data into the (110) plane. It is easy to see that...
Fig. 3. $H$–$T$ magnetic phase diagram of Ho$_{0.5}$Lu$_{0.5}$B$_{12}$ for magnetic fields applied in three principal cubic directions. The phase boundaries are derived from magnetoresistance, heat capacity and magnetization data (see symbols in panel (a)). Roman numerals denote various phases within the antiferromagnetic state. P- paramagnetic phase.

the transverse MR of the single-domain Ho$_{0.5}$Lu$_{0.5}$B$_{12}$ crystal with $I \parallel [110]$ have in polar plots an anisotropic phase diagram in the form of a Maltese cross with lobes, which are arranged along ($H \parallel [110]$) and transverse ($H \parallel [001]$) to the dynamic charge stripes observed previously in X-ray and magnetoresistance measurements of LuB$_{12}$ [3]. The numerous magnetic phases are detected and attributed to the concurrence of dynamic charge stripes and RKKY-interaction in Ho$_{0.5}$Lu$_{0.5}$B$_{12}$.

Fig. 4. Angular dependencies of resistivity at $T = 2.1$ K and in various magnetic fields up to 80 kOe. Top axis shows the principal axes.

Fig. 5. 3D view of magnetoresistance dependencies on magnetic field at $T = 2.1$ K. The rotation axis is $I \parallel [110]$, while three principal cubic directions in the plane (110) are shown by arrows.
4. Conclusions

A model strongly correlated electron system $\text{Ho}_{0.5}\text{Lu}_{0.5}\text{B}_{12}$ with fcc lattice instability (cooperative dynamic JT effect) and nanometer size electronic phase separation (dynamic charge stripes) has been studied in detail by low temperature magnetoresistance measurements. An angular $H-\varphi-T$ antiferromagnetic phase diagram in the form of a Maltese cross has been deduced for this AF metal for the first time. It matches to three main sectors in vicinity of the principal directions (i) along $H \parallel [110]$, and (ii) $H \parallel [001]$ transverse to dynamic charge stripes, and (iii) along $H \parallel [111]$ connected with the orientation of the AF magnetic structure. We argue that the observed dramatic symmetry lowering is a consequence of strong renormalization of the indirect exchange interaction (RKKY mechanism) due to the presence of dynamic charge stripes in the matrix of AF metal.

Acknowledgments

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