Magnetic anisotropy in the AFM and SDW phases of CeB$_6$

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Abstract. The angular dependences of magnetization and Hall resistance have been investigated both in paramagnetic and magnetically ordered phases by the technique of sample rotation in magnetic field up to 6T on the high-quality single crystals of CeB$_6$. It has been shown that, as CeB$_6$ undergoes the transition from the antiferromagnetic modulated (AFM) to the so-called antiferroquadrupolar phase, the easy-magnetization axis in the [110] plane changes from <100> to <110>. The behavior of the magnetization anisotropic component differs radically in these two magnetically ordered phases. The analysis provides evidence in favor of a spin density wave (SDW) state formation in cerium hexaboride in the temperature range $T_N \approx 2.3K < T < T_Q \approx 3.3K$. Moreover, the SDW antinodes’ formation was established to be the main factor of the Hall effect renormalization and anisotropy in both AFM and SDW magnetic phases of CeB$_6$. As a result, the complex magnetic phase diagram of CeB$_6$ may be interpreted as caused by a competition between itinerant and 4f-antiferromagnetism in this strongly correlated electron system.

1. Introduction.

Since the 1960s the cerium hexaboride has been attracting significant interest, because it was considered to be an archetypal example of the dense Kondo system with about equal concentration of conduction electrons and the localized magnetic moments (LMM) of Ce$^{3+}$ ions ($n_e \approx n_{4f}$) [1]. Additionally it was believed for a long time that along with the Kondo compensation mechanism at $T_Q \approx 3.3K$ an unusual antiferroquadrupole (AFQ) orbital order appears in CeB$_6$ (see H-T phase diagram in Fig.1). As the temperature decreases the AFQ ordered state changes to the antiferromagnetic modulated (AFM) structure at $T_N \approx 2.3K$ [1-2] (phase III in Fig.1). However recently it was shown certainly [3-4] that a considerable (~30%) spin density is located outside the Ce sites both in the immediate vicinity of the B octahedra and inside the B$_6$ clusters. The authors [3-4] found that these magnetic polarization regions, which do not directly correspond to the $^2F_{5/2}$ state of the Ce$^{3+}$ ions, hold in the paramagnetic (P) phase at $T_C < T < 10K$ (I in Fig.1). The polarization of these nanosize domains in the P phase of CeB$_6$ appears to be antiparallel to the LMM of Ce$^{3+}$ ions, in contrast to their co-directional arrangement in the magnetically ordered AFQ(II) state [3-4]. The next step to the explanation of the unusual magnetism in CeB$_6$ was done in [5] where a considerable magnetic polarization of 5d-states in the conduction band was found at $T \leq 7K$. Then, a strong arguments in favor
of itinerant 5d-component spin density wave (SDW) magnetism in the range between $T_N \approx 2.3K$ and $T_Q \approx 3.3K$ were presented in [6] and based on the results of the comprehensive study of charge transport and magnetic properties of CeB$_6$.

2. Experimental results and discussion.

To shed more light on the nature of unusual low temperature magnetism of CeB$_6$, it is important to investigate the magnetic and transport anisotropy in this compound with strong electron correlations. Indeed, according to the predictions of the authors [7-8] the modulation amplitude of the magnetic structure should be expected to increase linearly with the external magnetic field in the case of the SDW magnetic ordering mechanism. As a result it seems important to carry out the precision measurements of the angular dependences within the wide range of magnetic field at temperatures corresponding both to paramagnetic I (P) and magnetically ordered phases III (AFM) and II(AFQ) (see Fig.1a) for high-quality single crystalline samples. Since the anomalous Hall effect is also observed in the preliminary measurements [6], useful additional information may be acquired from the measurements of the Hall resistance angular dependences in I-III phases of cerium hexaboride.

![Figure 1. (a) Magnetic phase diagram of CeB$_6$ [6], I(P) –paramagnetic, II(AFQ) and III(AFM)-magnetically ordered phases (see text). (b) Magnetic field dependences of anisotropic component of magnetization $\Delta M$ at helium temperatures and peak-to-peak Hall resistance amplitude $\Delta \rho_H$ in CeB$_6$.](image)

In this study we have performed precision measurements of the angular dependences of the magnetization and the Hall resistance in the magnetic field up to 70 kOe at liquid helium temperatures 1.8÷4.2K. The magnetization $M(\phi, H_0, T_0)$ was measured on the vibrating sample and SQUID magnetometers. The Hall effect was measured by the sample rotation technique with a stepwise fixation of the sample position in magnetic field [6].

The magnetization anisotropy of CeB$_6$ was studied at temperatures 4.2K (phase I), 3K (phase II) and 1.85K (phase III) (see the dashed lines in Fig.1a) in the range of magnetic fields $H<60$ kOe where the points $H_N$ and $H_Q$ of III-II and I-II phase transitions are included. For example, Fig.2a shows the family of non-monotonous angular dependences of the CeB$_6$ magnetization obtained at $T_0=1.85K<T_N$ for the magnetic fields in the interval 3.5÷50 kOe. For comparison, the angular independent curve $M(\phi, H_0=9.4$ kOe, $T_0=4.2K$) corresponding to paramagnetic phase of cerium hexaboride is also presented on Fig.2a. It should be mentioned here that the <001> direction was chosen as a rotation axis of the sample in the magnetization experiments. As a result, we analyzed the magnetic anisotropy related to variation of the external magnetic field $H$ between directions $H||<100>$ (0° in Fig.2) and $H||<110>$ (45° in Fig.2).

From the data of Fig.2a the dramatic transformation of the character of magnetic anisotropy can be evidently deduced. Indeed, on the III-II phase interface ($H_\gamma \approx 10$ kOe, Fig.1) the shape of the $M(\phi,H_0,T_0)$ angular dependences of CeB$_6$ changes from the combination of smooth maxima in <100> and sharp drops along <110> directions (Fig.2a, curve $H_\gamma=5$ kOe) to the sinusoidal dependence...
ΔM~sinφ, which is shifted in phase by 180° with respect to the maximums of the low field curves. Moreover, approaching the III-II phase transition the amplitude of the anisotropic component ΔM(H) decreases drastically in phase III and then, about linear increase ΔM(H)~H is observed above the H_N (Fig.1b). Similarly, the anisotropic component ΔM(H) in phase II is also a linear function of the

Figure 3. (a) Angular dependences of the Hall resistance ρ_H(φ, H, T=4.15K) of CeB_6. The dashed and dash-dotted lines correspond to the main and even harmonics contribution to the Hall signal (see text). (b) Magnetic field dependences of the Hall coefficient R_H(H) and even harmonics amplitude at helium temperature. H_Q corresponds to the I-II phase transition (see also Fig. 1a).
The most unusual behavior was observed in the present study in the measurements of the Hall resistance angular dependences in phases III and II (Fig.2b). Indeed, the harmonic dependence $\rho_H \sim \cos \phi$ is typical for compounds with metallic conduction and it results from variation of the normal component of $H$ vector when the sample rotates in magnetic field (see inset on Fig.3a). Instead of that, very large amplitude Hall resistance was detected in phase III (curves $H_0=5$ kOe and 10 kOe in Fig.2b) that demonstrates a complicated meander-type angular dependence with switching between different Hall resistance levels at the angles around $H//<110>$ orientation. Additionally considerable contribution of the Hall signal from the even harmonics of the form $\rho_H(\phi) \sim \rho_{H1} \cos \phi + \rho_{H2} \cos(2n\phi - \phi_0)$, with $n=1, 2...$ appears on the Hall resistance curves in the magnetic field $H>H_N$ (Fig.2b) and $H>H_Q$ (Fig.3a). We note also the similarity of the anisotropic magnetization $\Delta M(\phi)$ (Fig.2a) and Hall resistance (Fig.2b) curves in the phase II (see, for example $H_0=30$ kOe in Fig.2). Moreover, the $\Delta M(H)$ decrease in phase III is accompanied with very strong depression (~40 times) of the Hall resistance amplitude and it is followed by its elevation with magnetic field in the range $H>25$ kOe in phase II (see Fig.1b, $T_0=1.85$ and 1.95K). Fig.3b shows an example of the analysis of the harmonic’s amplitudes at helium temperature. We note from the results of Fig.3b (i) very strong magnetic field dependence of the Hall coefficient both through the I-II phase transition and in II phase and (ii) an appearance of the even harmonics $\rho_{H2}$ and $\rho_{H4}$ and their enhancement in the range $H>2H_Q$.

It should be mentioned that the appearance of even harmonics in the Hall signal was earlier observed in the Ce-based strongly correlated electron systems CeAl$_2$, CeCu$_6$-Au$_x$ [9-10] and it was attributed to the formation of spin polarization in nanosize regions of the 5d-states in these compounds. In the case of CeB$_6$ in phase II the antiferromagnetic state is induced by external magnetic field and increases with increase in $H$, thus, the mentioned spin-polarized nanodomains should probably be attributed to the formation of the spin density wave antinodes and SDW enhancement in magnetic field [7-8]. As a result, the magnetic anisotropy and charge transport in CeB$_6$ at liquid helium temperatures exhibits the singularities associated with the magnetic structure of the SDW state. We emphasize that the revealed sharp variation of the magnetic and charge transport anisotropy as well as the proposed scenario of phase transition in SDW (II) phase agree with the data of ESR-spectra studies [11] and allow us to interpret successfully the NMR, $\mu$SR and magnetic neutron scattering data [2-5] for CeB$_6$.

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