Magnetic spin resonance in CeB_6

S. V. Demishev,[1] A. V. Semeno,[1] A. V. Bogach,[1] N. A. Samarin,[1] T. V. Ishchenko,[1] V. B. Filipov,[2] N. Yu. Shitsevalova,[2] and N. E. Sluchanko[1]

[1]*A.M. Prokhorov General Physics Institute, RAS, 38 Vavilov Str., Moscow 119991, Russia*

[2]*Institute for Problems of Materials Science, NAS, 3 Krzhizhanovsky Str., 03680 Kiev, Ukraine*

(Received 28 May 2009; revised manuscript received 5 October 2009; published 7 December 2009)

Line shape of magnetic spin resonance at 60 GHz specific to the phase II (so-called antiferroquadrupole phase) of CeB_6 was studied. The applied procedure of data analysis has allowed obtaining g factor of the oscillating magnetic moments, line width, and oscillating magnetization. It is found that the approaching to the transition temperature $T_{\text{I-II}}$ from phase II to the paramagnetic phase I results in strong broadening of the resonance (the line width increases three times in the range 1.8 K$\leq T\leq$3.8 K) whereas g factor $g=1.59$ remains temperature independent. Magnetic-resonance data suggests that the magnetization of CeB_6 in the phase II consists of several contributions, one of which is responsible for the observed magnetic resonance. This term in magnetization is missing in the paramagnetic phase and corresponds to ferromagnetically interacting localized magnetic moments. The magnitude of the oscillating part of magnetization is less than total magnetization in the range $T^*\leq T\leq T_{\text{I-II}}$ and coincides with the total magnetization for $T\leq T^*$, where $T^*\sim 2$ K. We argue that ferromagnetic correlations play a key role in the observed phenomenon in analogy with the recent experimental and theoretical results on the magnetic resonance in the dense Kondo systems. At the same time the interpretation of the magnetic-resonance data in the framework of the existing models of magnetism in CeB_6 faces substantial difficulties, which demands further development of the theory of static and dynamic magnetic properties of this heavy fermion metal.

DOI: 10.1103/PhysRevB.80.245106 PACS number(s): 76.90.+d, 75.30.Mb, 71.27.+a

I. INTRODUCTION

Up to very recent time it was believed that there are no magnetic spin resonances at low temperatures in a dense Kondo system originating from the magneto-optical response of the strongly correlated magnetic matrix.[1] As the main reasons for that the vanishing of the localized magnetic moments (LMMs) at low temperatures due to the Kondo screening effect and the broadening of the resonance by spin fluctuations were considered.[1,2] Additionally, the charge carriers in the metallic dense Kondo systems are typically heavy fermions and possible cyclotron resonance corresponds to a practically unreachable region of magnetic field. However, this understanding of the magnetic-resonance problem in this class of materials was put into question by the observation of the magnetic resonance in the dense Kondo system $YbRh_2Si_2$ below Kondo temperature $T_K=25$ K even with the use of the X-band electron paramagnetic resonance spectrometer.[2] Indeed, in this case there should be no free LMMs and expected spin-resonance line width can be estimated as $k_BT_K/\mu_B\sim 37$ T.[2]

The subsequent studies of several Yb- and Ce-based intermetallic compounds [$YbRh_2Si_2$, YbRh, $YbIr_2Si_2$, and CeRuPO (Ref. 3)] have led to a conclusion that the strong ferromagnetic correlations play a crucial role in the existence of the electron-spin resonance in any dense Kondo system.[3] This hypothesis was supported by recent theoretical works.[4,5]

However, it is worth noting that almost at the same time the low-temperature magnetic resonance was discovered in another dense Kondo system-cerium hexaboride, CeB_6,[6,7] which has Kondo temperature $T_K\sim 1$ K. In contrast to the aforementioned compounds, in majority of works CeB_6 is believed to be governed by antiferromagnetic interactions and orbital-ordering effects.[8–28] This circumstance apparently motivates the detailed study of the magnetic resonance in CeB_6 in comparison with other systems such as $YbRh_2Si_2$, YbRh, $YbIr_2Si_2$, and CeRuPO. Cerium hexaboride is known as possessing a complicated magnetic phase diagram (Fig. 1), consisting of paramagnetic (P, phase I), so-called antiferroquadrupole (AFQ, phase II), and antiferromagnetic (AF, phase III) phases.[8–14] The characteristic temperatures $T_{\text{I-II}}$ and $T_{\text{II-III}}$ for the transitions phase I$\rightarrow$phase II(P$\rightarrow$AFQ) and phase II$\rightarrow$phase III(AFQ$\rightarrow$AF) depend on the magnetic field B (Fig. 1). It is established that $T_{\text{I-II}}(B)$ increases with magnetic field whereas $T_{\text{II-III}}(B)$ decreases; for $B=0$ $T_{\text{I-II}}(0)=3.2$ K and $T_{\text{II-III}}(0)=2.3$ K.[8–14] Conventional explanation of this magnetic phase diagram is based on the models, which account interplay between spin and orbital

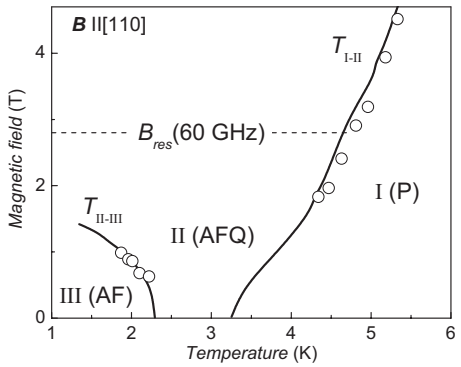

FIG. 1. Magnetic-phase diagram of CeB_6 for the [110] crystallographic direction. Solid lines are obtained from the magnetization and resistivity data (Refs. 11 and 12); points correspond to the magnetic-resonance experiments (Refs. 6 and 7). Dashed lines denote the observed resonance field in 60 GHz cavity measurements.

degrees of freedom and imply scenario with the orbital-ordering temperature T_Q higher than the spin-ordering temperature T_N.[15–28] Therefore the transition phase I $\rightarrow$phase II(P$\rightarrow$AFQ) is usually interpreted as an f-orbital-ordering phenomenon at $T_Q(B)=T_{\text{I-II}}(B)$ and no change in the spin structure at this phase boundary is expected in theory. As an experimental ground for this interpretation the observation of the magnetic field-induced antiferromagnetic vector $k=(1/2,1/2,1/2)$ corresponding to the doubling of the lattice period[8,9,28] is considered. Indeed, in absence of the noticeable atomic displacements in phase II as compared to the P phase,[8,9,27] it looks natural to explain this phenomenon in terms of orbital ordering of the f shells of the Ce^{3+} ions leading to the three-dimensional alternating structure of the electric quadrupoles of the opposite sign[8,18–23] thus motivating the application of the aforementioned term "antiferroquadrupole phase." The other transition phase II $\rightarrow$phase III(AFQ$\rightarrow$AF) is supposed to correspond to $T_N(B)=T_{\text{II-III}}(B)$ and leads to a complicated Neel-type ordering of the Ce^{3+} LMMs. It is worth noting that the exact magnetic structure in AF phase is still remains the subject of debates.[24–28] However, all modifications in the theoretical models suggested up to now for the description of the peculiar magnetic properties of this material assume that (i) magnetism of CeB_6 originate from the single source, namely, the LMMs of Ce^{3+} somehow affected by Kondo screening and (ii) the ground state of Ce^{3+} in crystal field is Γ_8 quartet rather than Γ_7 doublet as long as only this type of symmetry allows electric-quadrupolar effects.

It was shown[6,7] that the magnetic resonance in CeB_6 is a feature of the phase II (AFQ phase) for $T<T_{\text{I-II}}(B)$ and it is missing in paramagnetic phase (phase I). As long as in the conventional model for the AFQ phase spins are not frozen or form any long-range magnetic structure they are as free as in the paramagnetic phase. Therefore it is difficult to explain why possible orbital ordering induces the onset of a magnetic resonance just for $T<T_{\text{I-II}}$. Moreover, the case of CeB_6 looks even more complicated as compared to the strongly correlated systems studied in Refs. 2 and 3. The fact that temperature dependence of the integrated intensity $I(T)$ does not follow temperature dependence of the magnetization $M(T)$ (Ref. 7) has led to the conclusion that below T_Q a new (missing for $T>T_{\text{I-II}}$) contribution to magnetization develops and it is responsible for the observed magnetic oscillations.[7] Thus it is necessary either to "split into parts" LMMs on Ce^{3+} ions or assume that AFQ phase is magnetically inhomogeneous one (i.e., only some part of the sample volume contributes to magnetic resonance). The latter possibility to our best knowledge is not supported by any experimental observations including the structural analysis data.

The above consideration indicates the importance of the determination of the magnitude of the magnetic resonance in dense Kondo systems, i.e., determination of the dynamic magnetization responsible for the observed modes of magnetic oscillations. Unfortunately this problem was not considered in detail neither experimentally[2,3] nor in theoretical works[4,5] where accent was made on the magnetic-resonance line width and frequency.

The present work is aimed to check the hypothesis[7] about the presence of several magnetic contributions in the phase II of CeB_6 and quantitative characterization of the oscillating part of magnetization in this material. In this purpose, recently suggested experimental approach and data-analysis schema, which have been successfully applied to the case of magnetic resonance in EuB_6,[29] are used. When analyzing the obtained characteristics of the oscillating magnetization, we discuss applicability of various models suggested for the explanation of magnetism of CeB_6.

II. EXPERIMENTAL DETAILS AND DATA-ANALYSIS SCHEMA

A. Experimental details

The single crystals of CeB_6 have been prepared as described in.[30] The x-ray and chemical-analysis data as well as transport and magnetic measurements[31] have confirmed the high quality of the samples studied. Magnetic-resonance experiments were carried out with the help of the original backward wave-oscillators-based microwave cavity spectrometer operating in the range 40–100 GHz at temperatures 1.7–300 K for magnetic field up to 7 T. More details about spectrometer characteristics can be found elsewhere.[29,32]

At present it is established that magnetic spin resonance in phase II of CeB_6 exists in a wide frequency range $\omega/2p=44-360$ GHz.[6,7,33] For obtaining the best available signal-to-noise ratio essential for the line-shape analysis, the 60 GHz cylindrical reflecting cavity tuned to TE_{011} mode was chosen. The external magnetic field B generated by superconducting magnet was parallel to the cavity axis and was aligned along [110] crystallographic direction.

The used experimental geometry suitable for measurements of metallic samples with arbitrary magnetization is similar to that in Ref. 29. A thin copper foil with a hole located at the maximal oscillating magnetic field position is used as the endplate of the cavity. The plate of CeB_6 single crystal is attached to the foil from outside and fixed by highly conducting glue. The sample closes the hole completely and therefore only central part of it is subjected to microwave field inside the cavity. This schema allows us to exclude effects of inhomogeneity of magnetic field at the sample edges,[29] which may distort the resonance line shape. The quality factor of the described cavity was about $Q\sim10^4$.

In order to apply the procedure of the baseline subtraction and absolute calibration of cavity measurements[29] the temperature dependences of the loaded cavity factor together with the temperature and field dependences of dc resistivity have been studied on the same crystal. Details on the magnetotransport experiments are given in Ref. 31. In order to elucidate the nature of the magnetic resonance in CeB_6 the measurements of the temperature and field dependences of magnetization have been undertaken on the same crystals as investigated in the magnetic-resonance studies. These experiments have been carried out by means of the modified LDJ-1500 vibrating sample magnetometer.

B. Magnetic-resonance-data analysis schema

Let us first consider the magnetic resonance on some LMMs, which positions are fixed in space. In order to find

resonant cavity losses it is necessary to consider the relation between vectors of the oscillating magnetic induction inside the sample $\mathbf{B}_\omega$ and oscillating magnetic field outside $\mathbf{H}_\omega$.[29,34] If the field inhomogeneity effects are excluded, this relation is given by

$$\mathbf{B}_\omega = \hat{\mu}\mathbf{H}_\omega, \tag{1}$$

where microscopic magnetic-permeability tensor does not depend on spatial coordinates and possesses the following structure:

$$\hat{\mu} = \begin{vmatrix} \mu & i\mu_\alpha & 0 \\ -i\mu_\alpha & \mu & 0 \\ 0 & 0 & 1 \end{vmatrix}. \tag{2}$$

The diagonal and off-diagonal elements μ and μ_α are complex functions of frequency ω, magnetic induction B, gyromagnetic ratio γ, relaxation time, and oscillating part of the sample magnetization M_0. The functions μ and μ_α may be chosen in different model approximations, namely, Bloch-Bloembergen, Landau-Lifshitz, or Garstens symmetric forms, for which $\mu=\mu_1-i\mu_2=1+4\pi\chi_1-4\pi i\chi_2$ and $\mu_\alpha=\mu_{\alpha 1}-i\mu_{\alpha 2}=4\pi\chi_{\alpha 1}-4\pi i\chi_{\alpha 2}$ with the susceptibilities $\chi_1,\chi_{\alpha 1}(\chi_2,\chi_{\alpha 2})$ being odd (even) function of frequency ω. The analytical expressions for aforementioned model expressions are given in Ref. 29.

In the case of magnetic-resonance cavity measurements with TE_{01n} mode both $\mathbf{B}_\omega$ and $\mathbf{H}_\omega$ must be directed along cavity radius and thus not all components of these vectors suggested by Eq. (1) for infinite medium are allowed.[29] Assuming that in the considered above experimental geometry sample size (i.e., hole diameter) is small enough so that the magnetic field near the sample surface may be considered as homogeneous and using Eqs. (1) and (2), we get relation between radial complex amplitudes of the induction and field:[29]

$$B_{\omega r} = \mu_{eff} H_{\omega r}, \tag{3}$$

$$\mu_{eff} = \mu_{eff1} - i\mu_{eff2} = \mu - \mu_\alpha^2/\mu. \tag{4}$$

Simultaneously for the cavity losses caused by the metallic-sample response in a skin layer we get[29]

$$Q_{sample}^{-1} \sim (\omega\mu_R R)^{1/2} = [\omega(|\mu_{eff}| + \mu_{eff2})R]^{1/2}. \tag{5}$$

Here R stands for the dc sample resistance. Equations (3)–(5) are valid for an arbitrary sample magnetization and dynamic susceptibilities $\chi_1,\chi_{\alpha 1},\chi_2,\chi_{\alpha 2}$. In the limit $(\chi_1,\chi_{\alpha 1},\chi_2,\chi_{\alpha 2}) \ll 1$ (weakly magnetic solids with broad resonance lines) formula (4) gives $\mu_{eff} \approx \mu$ and expression for losses corresponds to one considered by Kittel.[35] It is worth noting that another widely used expression proposed by Young and Uehling $\mu_{eff}=\mu+\mu_\alpha$ (Ref. 36) is not exact in our case although it also suggests losses depending on the off-diagonal components of permeability matrix.

It follows from Eq. (5) that comparison of the cavity losses caused by the sample with the temperature and field dependences of dc resistance allow obtaining absolute calibration of cavity measurements together with the baseline subtraction as long as apart from the resonant magnetic field

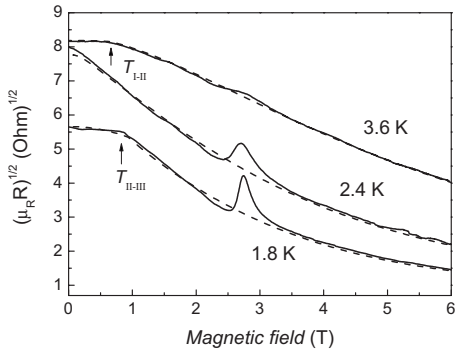

FIG. 2. Magnetic-resonance spectra in the phase II of CeB_6 (solid lines) and magnetoresistance (dashed lines). Arrows mark various transitions of the magnetic-phase diagram.

$\mu_R=1$.[29] This circumstance provides direct way for finding the proportionality coefficient between Q_{sample}^{-1} and $(\mu_R R)^{1/2}$ in Eq. (5) and finally gives μ_R as a function of magnetic field at each particular temperature (more details about the procedure of absolute calibration and justification of the suggested approach may be found in Ref. 29). When μ_R is known from experiment, it can be approximated with the help of μ_{eff} [Eqs. (4) and (5)], which in turn depends on the microscopic expressions for μ and μ_α. This algorithm of the line-shape simulation results in obtaining the spectroscopic parameters: gyromagnetic ratio γ (g factor), relaxation time (line width), and oscillating part of the sample magnetization M_0 taken at resonant field B_{res}. Of course these parameters may somehow depend on the model used[29] and therefore calculations for the different forms of permeability tensor are desirable.

Our consideration does not take into account possible effects of spin diffusion, which may be estimated with the help of Dyson theory[37] and are supposed to play essential role in magnetic resonance in metals,[1] including dense Kondo systems.[5] According to recent findings[4,5] both electron-spin resonance (ESR) on LMMs of $4f$ shell and ESR on heavy fermions may contribute to the magnetic resonance observed in Refs. 2 and 3. To shed a light on this problem, in the case of CeB_6 the μ_R experimental data should be also analyzed under assumption of dysonian line shape, which allows to estimate the relation between spin-diffusion time in the skin layer T_D and LMMs relaxation time T_2.[37] The applicability of the analysis schema suggested in Ref. 29 for LMMs without diffusion should correspond to the case of slow spatial movements $T_D \gg T_2$.

III. EXPERIMENTAL RESULTS AND DISCUSSION

A. Magnetic-resonance spectra in phase II of CeB_6

To deduce the magnetic-resonance spectra in units $(\mu_R R)^{1/2}$ the subtraction of the part of the 60 GHz cavity losses caused by the sample[29] was done in combination with both the procedure of absolute calibration (see point 2.2 and Ref. 29) and the dc magnetoresistance $R(B)$ data analysis. Examples shown in Fig. 2 demonstrate good coincidence of the results of cavity measurements outside of the resonance region (Fig. 2, solid lines) with the magnetoresistance data

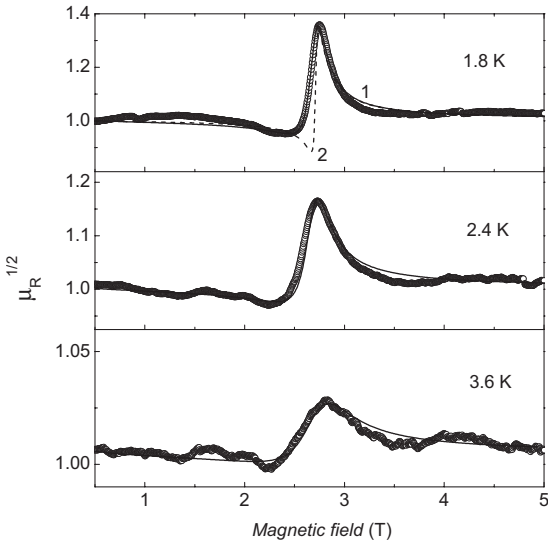

FIG. 3. Examples of the line-shape modeling for the magnetic resonance in CeB_6. Points correspond to experiment, solid line is calculated as described in section 2.2 for Garstens symmetric form of magnetic-permeability tensor. For $T=1.8$ K the fitting by Dysonian line shape with $T_D=0.1T_2$ is shown by dashed line.

(Fig. 2, dashed lines). Bends on the curves in the low-field region correspond to various phase transitions on the magnetic-phase diagram (Fig. 1) and are in good agreement with the literature data. It is visible (Fig. 2) that in accordance with[29] the $R(B)$ data may serve for experimental determination and subtraction of the background in resonant-cavity measurements as expected from Eq. (5). The obtained magnetic-resonance spectra calibrated to absolute units $\mu_R^{1/2}$ from the data of Fig. 2 are presented in Fig. 3.

It is found that resonant field $B_{res}\sim 2.8$ T is temperature independent within the experimental accuracy. The location of the B_{res} on the magnetic phase diagram is shown in Fig. 1. The increase in temperature in approaching to the II-I phase boundary $T_{\text{I-II}}(B_{res})\sim 4.7$ K induces the broadening of the magnetic resonance and damping of its magnitude (Figs. 2 and 3). We wish to point out that, in contrast to the previous findings,[38] no magnetic resonance of comparable amplitude was observed in the paramagnetic phase (phase I). As was reported previously[6,7] in this phase only the traces of the ESR were detected in the vicinity of the g factor ~ 2 which are at least an order of magnitude smaller in comparison with the magnitude of the resonance at $T=3.6$ K (Fig. 3). These negligible resonant features are difficult to distinguish from the ordinary impurity ESR and thus this effect will not be discussed in the present paper.

The application of the suggested data-analysis schema (point 2.2 and Ref. 29) allows obtaining good approximation of the observed line shapes (solid lines Fig. 3). When carrying out fitting for permeability tensor in Bloch-Bloembergen, Landau-Lifshitz, and Garstens symmetric forms[29] we find that all these models reproduce well the experimental data in the vicinity of the magnetic resonance and result in almost identical spectroscopic parameters (see below). However, the use of Garstens symmetric form gives better description of the resonance line shape in a wider range of magnetic field around the resonance (Fig. 3), which is in agreement with the previous results of modeling procedure for magnetic resonance in EuB_6.[29]

The undertaken analysis suggests that the magnetic-resonance data in the phase II of CeB_6 may be quantitatively described within the approach suggested in Ref. 29, which corresponds to negligible spin diffusion. This result is completely confirmed by application of Dyson theory to obtained experimental results. First of all, the assumption about rapid diffusion ($T_D<T_2$) does not meet our case (dashed line in Fig. 3 illustrates this statement). From fitting with the Dysonian line shape[37] we can conclude that the relation between characteristic times should satisfy condition $T_D/T_2>2$. However in this range of parameters the dependence of the Dysonian line shape on T_D/T_2 ratio is very small and only a lower limit of the ratio T_D/T_2 may be obtained.

The value of T_D may be estimated directly from the conductivity σ, effective-mass m, and carrier-concentration n data available for CeB_6.[31] Indeed, the diffusion time through skin layer δ is $T_D\sim\delta^2/D$ and diffusion coefficient may be evaluated using Einstein equation $\sigma=e^2Dg(E_F)\sim 0.16(e/h)^2Dn^{1/3}m$. This gives for the skin layer width and diffusion time at 60 GHz and $T\sim 2$ K the values $\delta\sim 1.610^{-4}$ cm and $T_D\sim 10^{-8}$ s. At the same time the line width of the resonance (Fig. 3) leads to estimate $T_2\sim 10^{-10}$ s resulting to the condition $T_D/T_2\sim 100$, which corresponds to very slow spin diffusion in the phase II of CeB_6. In other words, our analysis shows that in CeB_6 at low temperatures carriers are so heavy [$m\sim 300m_0$ (Ref. 31)] that they are "viewed" by magnetic resonance as ordinary nonmoving LMMs. Therefore (i) the suggested data-analysis schema holds even in presence of possible contribution to magnetic resonance from heavy fermions and (ii) it is not possible to distinguish *a priori* the contribution to magnetic resonance from ESR on cerium LMMs and from the ESR on heavy fermions as it was pointed out in Refs. 4 and 5. Consequently, the elucidation of the nature of the oscillating magnetization in CeB_6 appears to be very important.

B. Spectroscopic parameters

The errors of the fitting procedure applied in the vicinity of the resonance lines $\mu_R(B)$ (Fig. 3) do not exceed 3% except at the highest temperatures $T\geq 3.6$ K where the computation error is about 5%. The fitting parameters obtained assuming various model expressions for permeability tensor (Bloch-Bloembergen, Landau-Lifshitz, and Garstens symmetric forms[29]) also agree within the accuracy of 3–5 %, which is very similar to the results of calculations for the case of EuB_6. The data obtained for the g factor, line width W, and oscillating magnetization M_0 are presented in Figs. 4 and 5.

It is visible, that in the phase II of CeB_6 the g factor is temperature independent and equals to $g=1.59$ whereas line width increases when temperature tends to the phase boundary $T\rightarrow T_{\text{I-II}}(B_{res})$ (Fig. 4). The temperature independence of both g factor and B_{res} well agrees with small values of magnetization M_0 meeting the condition $\chi_1,\chi_2\ll 1$ at all temperatures. This condition gives simultaneously $\mu_{eff}\approx\mu$ in Eq.

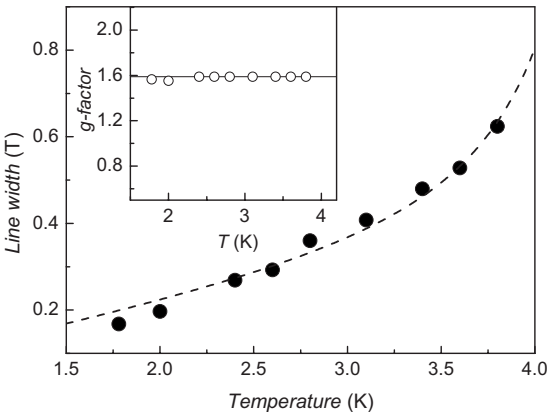

FIG. 4. Temperature dependences of the line width (main panel) and g factor (inset) for magnetic resonance in CeB_6.

(4). It is worth noting strong growth of the linewidth $W(T)$ in the phase II (three times in the range $1.8 \leq T \leq 3.8$ K).

Comparison of the oscillating magnetization $M_0(T)$ with the static magnetization taken at the resonance field $M(B_{res},T)$ clearly shows that for $T > T^* \sim 2$ K the condition $M(B_{res},T) > M_0(T)$ is valid (Fig. 5) and thus only part of the sample magnetization contributes to the observed magnetic oscillations. When temperature is lowered the $M_0(T)$ increases rapidly and for $T \leq T^*$ almost coincide with total magnetization $M_0(T) \approx M(B_{res},T)$ (Fig. 5). Therefore in the phase II of $CeB_6(T < T_{\text{I-II}})$ the sample magnetization M may be represented as a sum of the oscillating part M_0 and nonoscillating part M_1

$$M(B,T) = M_0(B,T) + M_1(B,T). \tag{6}$$

According to the results of the present study the oscillating term is missing in the paramagnetic phase (phase I, $T > T_{\text{I-II}}$), where $M(B,T) = M_1(B,T)$. We wish to emphasize that in the phase II the nonoscillating part of magnetization freezes out and does not contribute to M below T^*. The expected temperature dependence of M_1 in the range $T < T_{\text{I-II}}$ is shown in Fig. 5 by dash-dot line.

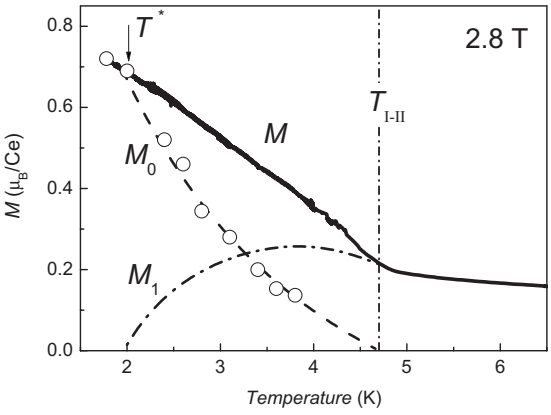

FIG. 5. Temperature dependences of the oscillating magnetization M_0 (white points) and total magnetization M (solid line) taken at the resonance field 2.8 T. The expected temperature dependence of the nonoscillating term $M_1 = M - M_0$ in the phase II is given by dash-dot line.

C. Discussion

The data presented in the previous section proves the hypothesis about complicated magnetization structure in the phase II, which was initially suggested in Ref. 7. However the observed $M_1(T)$ dependence is completely different from the "naive" linear extrapolation of $M(T)$ in the phase I to the phase II, which was postulated in our previous study.[7] Below we will consider results of the analysis of the magnetic-resonance phenomenon in CeB_6 in the frame work of various models of magnetism suggested so far for this material. In our opinion any of available approaches faces substantial difficulties, which rise important questions concerning the nature of magnetism of this strongly correlated heavy fermion metal. Indeed: (1) it is difficult to understand why the g factor is temperature independent. According to Refs. 4 and 5, the oscillating magnetic contribution for the magnetic resonance in the Kondo model is constructed of some quasiparticles which have predominantly $4f$ character,[4] i.e., the oscillating LMMs are mainly "produced" from the LMMs on Ce^{3+} ions. Thus the temperature-dependent Kondo screening of the oscillating magnetic moment $\mu^* \sim g$ should be expected, resulting in decrease in the g factor with lowering temperature. This behavior has been reported for $YbRh_2Si_2$ (Ref. 2) but does not meet the case of CeB_6 (Fig. 4). Moreover, as we will see below, even in the alternative model of magnetism of CeB_6 based on spin-polaron approach[31] the variation of screening of the Ce^{3+} LMMs with temperature in the phase II should be expected as well. (2) Why the magnetic resonance in CeB_6 is specific to the phase II and is not observed in phase I (paramagnetic phase)? Apparently this is because of $M_0(T \to T_{\text{I-II}}) \to 0$. However, according to the antiferroquadrupole model, the LMMs are free in both phases. Therefore the only reason may be found in a dramatic change in the relaxation rate at I-II phase boundary, which is supported by the observed broadening of the resonance with temperature (Fig. 2–4). However, any interactions of the LMMs in high-temperature paramagnetic phase should be weak in comparison with those ones in the low-temperature AFQ phase. For that reason the idea about drastic changes in the relaxation rate looks like *ad hoc* assumption in the antiferroquadrupole model. In our opinion this difficulty may be overcome by considering effects of ferromagnetic correlations[4,5,31] specific to the phase II. (3) In any case, the magnetism of CeB_6 in the Kondo model with antiferroquadrupole ordering is the magnetism of the LMMs on Ce^{3+} ions[18–22] although somehow screened by conduction electrons. According to the observed structure of magnetization in the phase II [Eq. (6)] the specific oscillating part of magnetization should be expected appearing for $T < T_{\text{I-II}}$. In the antiferroquadrupole model of the phase II this developing contribution is the contribution of magnetic octupole,[18–22] i.e., some symmetric combination of the orbital currents.[39,40] No reasons are known, why octupolar contribution to Ce^{3+} LMM expected in the antiferroquadrupole model should be treated separately serving as the reason for the observed

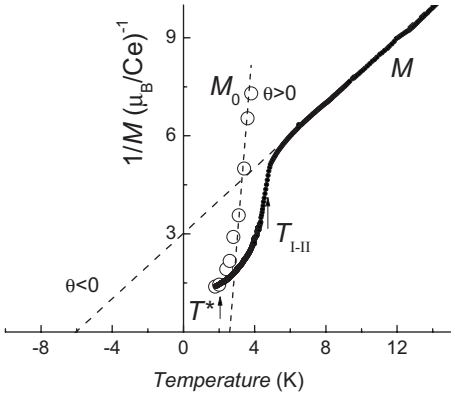

FIG. 6. Antiferromagnetic and ferromagnetic correlations in CeB_6 in the paramagnetic and antiferroquadrupole phases (see text for details.)

magnetic oscillations and there is no theory of the magnetic resonance relevant to this case. However, even within this assumption, the octupolar part is less than total magnetization at any temperature[20] and the condition $M_0 \approx M$ for $T < T^*$ (Fig. 5) makes this exotic explanation hardly possible.

Thus the antiferroquadrupole model meets serious difficulties in explanation of the character of magnetization in the phase II and the increase in $M(T)$ below $T_{\text{I-II}}$ may be interpreted as a result of ferromagnetic correlations. To illustrate this point the magnetization data in Fig. 6 are replotted in coordinates $M^{-1}=f(T)$ (Fig. 6). Although the $M(T)$ is taken at finite field, the temperature dependence of inverse magnetization in the paramagnetic phase may be reasonably fitted by Curie-Weiss law with the negative paramagnetic temperature corresponding to the antiferromagnetic exchange, which value $\Theta_{\text{AF}} \sim -6$ K agrees reasonably with the data available in literature.[31] At the same time in the phase II for the vicinity of transition temperature the M_0^{-1} follows the Curie-Weiss-type dependence with the *positive* paramagnetic temperature $\Theta_{\text{FM}} \sim 2.6$ K suggesting the importance of the ferromagnetic interaction for the oscillating part of magnetization (Fig. 6). For $T < 3$ K the saturation effects in oscillating magnetization leads to departures from Curie-Weiss behavior.

An additional argument of the domination of the ferromagnetic interactions at low temperatures may be obtained from the value of the oscillating magnetization. Indeed, knowing the g factor ($g \approx 1.59$) as well as the value of saturation magnetization ($M_{sat} \approx 0.8\mu_B$/Ce) one can estimate the expected magnetization without interaction in the magnetic field corresponding to the resonance condition ($B \sim 2.8$ T). The obtained value $M \sim 0.5\mu_B$/Ce (at $T=2$ K) is sufficiently smaller than the experimentally observed value $M_{exp} \approx 0.7\mu_B$/Ce thus suggesting ferromagnetic interactions in CeB_6 at low temperatures.

It should be mentioned that the onset of the ferromagnetic term in the phase II of CeB_6 does not contradict to the magnetization data including data on magnetic resonance obtained in the present work. However, the view on this problem in literature is contradictory. In the muon-spin-relaxation and neutron-diffraction experiments[25–27] ferromagnetic component in the phase II was not observed although ferromagnetic correlations were found in the phase III (antiferromagnetic phase). From the other hand, the analysis of transport (resistivity, magnetoresistance, Hall-effect, and thermoelectric power) and magnetic data have lead authors of Ref. 31 to a conclusion of the key role of ferromagnetic correlations in the genesis of various magnetic phases and phase transitions in CeB_6 including transition from the paramagnetic phase to phase II. The main idea of the approach suggested in Ref. 31 is to explain magnetic properties of the paramagnetic phase (phase I) of CeB_6 by magnetism of spin polarons, which are formed in the vicinity of Ce^{3+} ions. Therefore it is possible to expect that the magnetic resonance in the phase I will be damped due to strong spin fluctuations. The lowering temperature leads to increase in the spin polarization of the 5*d*-band conduction electrons resulting to the formation of the nanosize ferromagnetic domains (ferrons[41]) in the vicinity of $T_{\text{I-II}}$. Within the paramagnetic phase for $T \leq 7$ K this process is accompanied by strong enhancement of effective mass of these quasiparticles up to $m \sim 300m_0$.[31] In the range $T < T_{\text{I-II}}$ the ferrons serve as a "building material" for commensurate spin-density wave (SDW) with the double lattice period obtained by merging of the spin polarons (in analogy to the process suggested in Ref. 42 for EuB_6). Simultaneously spin density gradually shifts from the Ce^{3+} ions resulting in "release" of Ce^{3+} LLMs,[31] which may now contribute to magnetic resonance as observed experimentally. At the same time this process assumes gradual changes in effective screening of the Ce^{3+} LMMs and thus the first of the mentioned above problems persists in the considered model as well.

The increase in the phase I-phase II transition temperature $T_{\text{I-II}}(B)$ with magnetic field (Fig. 1) in the spin-polaron model[31] reflects the known effect of the SDW stabilization in magnetic field[43] rather than the orbital-ordering effects. However, at present there is no direct structural confirmation of the existence of SDW in CeB_6. In Ref. 31 the complicated spin-density distribution found in the phase II from polarized neutron diffraction by Saitoh *et al.*[24] was considered as one of the arguments favoring suggested interpretation. At the same time the result of Ref. 24 has been criticized by Givord *et al.*,[44] who claimed that all spin density is located precisely on Ce sites. Additionally the SDW formation was not observed also in the detailed study carried out in Ref. 27. From the other hand, it is worth noting that the rapid damping of the super reflex [5/2,5/2,5/2] by magnetic field, which was observed in Ref. 45, may also be a consequence of the stabilization of the SDW by magnetic field. And the last but not the least point consists in the circumstance that currently only the concept of the SDW explains the observation of the main reflex [1/2,1/2,1/2] specific to the phase II in zero magnetic field[9] as long as in the antiferroquadrupole model this reflex must disappear for $B=0$.[8]

Although suggested in Ref. 31, approach to magnetism of CeB_6 has a qualitative character and does not explain all available structural data, it is the only one at present which provides a possible explanation of the existence of several magnetic contributions in the phase II, including the oscillating component. At present it is not clear whether the considered difficulties can be resolved in somehow modified antiferroquadrupole model or it will not survive and a deeper

revision of the model of magnetism in CeB_6 will be required. When considering the possible spin-polaron version it is worth nothing that theoretically the formation of the spin polarons may be obtained for various theoretical approaches[41,42] including Kondo model[42] and thus may appear as a rather general effect. In any case the future consistent model of magnetism of CeB_6 should likely account the ferromagnetic correlations in the phase II. The onset of the ferromagnetic correlations may affect magnetic-resonance line width via spin fluctuations, which enhancement may be expected for the ferromagnetic contribution to magnetization at the phase I-phase II boundary. In the experiment this possible effect should cause rapid broadening of the magnetic resonance (Figs. 2–4).

Summarizing the results of our consideration we wish to stress that ferromagnetic correlations appear to be of crucial importance for the genesis of magnetic resonance in CeB_6 and this finding is in agreement with the modern view on magnetic resonance in the considered class of materials.[4,5]

However the existing theoretical calculations of the magnetic-resonance parameters that have been carried out for Kondo systems[4,5] possess certain difficulties with the quantitative accounting of the experimental data for CeB_6. First, in contrast to expected in theory linear-temperature dependence of the line width,[5] the observed $W(T)$ curves exhibits an essentially nonlinear form (Fig. 4). Second, the predicted renormalization of the g factor $g \sim g_0(1-m_0/m)^{-1}$ and line width $W \sim W_0 m_0/m$ (Ref. 4) should imply certain temperature dependence of these parameters as long as in CeB_6 the effective mass is a function of temperature.[31] For the g factor these corrections are less than 0.5% due to high value of $m/m_0 \sim 300$ (Ref. 31) and thus they are below the absolute accuracy of the determination of this parameter. At the same time the observed ~ 1.5 times growth of the effective mass with temperature in the range 2.9–3.8 K (Ref. 31) suggests the corresponding narrowing of the line width whereas experiment shows an increase in W by two times (Fig. 4). The aforementioned discrepancies may be merely a consequence of simplifying assumptions used in Refs. 4 and 5. However it is not possible to exclude that the explanation of all data on the magnetic resonance in strongly correlated electronic systems including those obtained in the present work will lead to certain modification in the present theory.

IV. CONCLUSIONS

In conclusion, we have shown that the magnetization of CeB_6 in the phase II consists of several contributions, one of which is responsible for the observed magnetic resonance. This term in magnetization is missing in the phase I and corresponds to ferromagnetically interacting localized magnetic moments thus confirming the idea[4,5] about the key role of ferromagnetic correlations in the genesis of magnetic resonance in the considered class of materials. The magnitude of the oscillating part of magnetization is less than total magnetization in the range $T^* < T < T_{\text{I-II}}$ and it coincides with the total magnetization for temperatures below $T^* \sim 2$ K. The approaching of the phase boundary $T_{\text{I-II}}$ results in the strong increase in the line width, probably caused by strong enhancement of spin fluctuations in vicinity of phase transition. The g factor of the oscillating LMMs equals $g=1.59$ and does not depend on temperature in the interval where magnetic resonance is observed. The examination of the obtained results within available models of magnetic properties of CeB_6 shows that any approach poses difficulties. The complicated structure of magnetization and behavior of the oscillating component in the phase II cannot be explained satisfactory in the antiferroquadrupole model. Simultaneously certain difficulties with interpretation should be marked for both classical dense Kondo system and recently suggested spin-polaron approaches. This situation demands further development of the theory of magnetism in CeB_6 aimed on the obtaining of an adequate physical picture of the magnetic resonance. As long as the technique of the magnetic resonance in CeB_6 is not appropriate to distinguish between the heavy fermions and Ce^{3+} LLMs the future theory has to address to the problem of the oscillating term in magnetization nature in this strongly correlated electron system.

ACKNOWLEDGMENTS

Authors are grateful to M. Yu. Kagan and P. I. Arseev for the helpful discussion of the spin-polaron scenario and magnetic-resonance problem in strongly correlated materials. This work was supported by the Programme of the Russian Academy of Sciences "Strongly correlated electrons" and by RFBR under Grant No. 07-02-00243.

[1] S. Barnes, Adv. Phys. **30**, 801 (1981).
[2] J. Sichelschmidt, V. A. Ivanshin, J. Ferstl, C. Geibel, and F. Steglich, Phys. Rev. Lett. **91**, 156401 (2003).
[3] C. Krellner, T. Forster, H. Jeevan, C. Geibel, and J. Sichelschmidt, Phys. Rev. Lett. **100**, 066401 (2008).
[4] E. Abrahams and P. Wolfle, Phys. Rev. B **78**, 104423 (2008).
[5] P. Schlottmann, Phys. Rev. B **79**, 045104 (2009).
[6] S. Demishev, A. Semeno, Y. Paderno, N. Shitsevalova, and N. Sluchanko, Phys. Status Solidi B **242**, R27 (2005).
[7] S. Demishev, A. Semeno, A. Bogach, Y. Paderno, N. Shitsevalova, and N. Sluchanko, J. Magn. Magn. Mater. **300**, e534 (2006).
[8] J. Effantin, J. Rossat-Mignod, P. Burlet, H. Bartholin, S. Kunii, and T. Kasuya, J. Magn. Magn. Mater. **47-48**, 145 (1985).
[9] H. Nakao, K. Magishi, Y. Wakabayashi, Y. Murakami, K. Koyama, K. Hirota, Y. Endoh, and S. Kunii, J. Phys. Soc. Jpn. **70**, 1857 (2001).
[10] K. N. Lee and B. Bell, Phys. Rev. B **6**, 1032 (1972).
[11] M. Takigawa, H. Yasuoka, T. Tanaka, and Y. Ishizava, J. Phys. Soc. Jpn. **52**, 728 (1983).
[12] M. Kawakami, S. Kunii, T. Komatsubara, and T. Kasuya, Solid State Commun. **36**, 435 (1980).

[13] D. Hall, Z. Fisk, and R. G. Goodrich, Phys. Rev. B **62**, 84 (2000).
[14] A. Takase, K. Kojima, T. Komatsubara, and T. Kasuya, Solid State Commun. **36**, 461 (1980).
[15] P. M. Levy, P. Morin, and D. Schmitt, Phys. Rev. Lett. **42**, 1417 (1979).
[16] P. Morin and D. Schmitt, Phys. Rev. B **27**, 4412 (1983).
[17] P. Morin and D. Schmitt, Phys. Rev. B **23**, 5936 (1981).
[18] O. Sakai, R. Shiina, H. Shiba, and P. Thalmeier, J. Phys. Soc. Jpn. **66**, 3005 (1997).
[19] R. Shiina, H. Shiba, and P. Thalmeier, J. Phys. Soc. Jpn. **66**, 1741 (1997).
[20] M. Sera and S. Kobayashi, J. Phys. Soc. Jpn. **68**, 1664 (1999).
[21] P. Thalmeier, R. Shiina, H. Shiba, A. Takahashi, and O. Sakai, J. Phys. Soc. Jpn. **72**, 3219 (2003).
[22] K. Hanzawa, J. Phys. Soc. Jpn. **73**, 1228 (2004).
[23] W. Sikora, F. Bialas, L. Pytlik, and J. Malinowski, Solid State Sci. **7**, 645 (2005).
[24] M. Saitoh, N. Okada, E. Nishibori, H. Takagiwa, T. Yokoo, M. Nishi, K. Kakurai, S. Kunii, M. Takata, M. Sakata, and Jun Akimitsu, J. Phys. Soc. Jpn. **71**, 2369 (2002).
[25] A. Schenck, F. N. Gygax, and S. Kunii, Phys. Rev. Lett. **89**, 037201 (2002).
[26] A. Schenck, F. N. Gygax, G. Solt, O. Zaharko, and S. Kunii, Phys. Rev. Lett. **93**, 257601 (2004).
[27] O. Zaharko, P. Fischer, A. Schenck, S. Kunii, P.-J. Brown, F. Tasset, and T. Hansen, Phys. Rev. B **68**, 214401 (2003).
[28] V. P. Plakhty, L. P. Regnault, A. V. Goltsev, S. V. Gavrilov, F. Yakhou, J. Flouquer, C. Vettier, and S. Kunii, Phys. Rev. B **71**, 100407(R) (2005).
[29] A. V. Semeno, V. V. Glushkov, A. V. Bogach, N. E. Sluchanko, A. V. Dukhnenko, V. B. Fillippov, N. Yu. Shitsevalova, and S. V. Demishev, Phys. Rev. B **79**, 014423 (2009).
[30] N. Yu. Shitsevalova and Y. Paderno, *Boron-Rich Solids*, Proceedings of tenth International Symposium on Boron, Borides, and Related Compounds (AIP, New York, 1991).
[31] N. Sluchanko, A. Bogach, V. Glushkov, S. Demishev, V. Ivanov, M. Ignatov, A. Kuznetsov, N. Samarin, A. Semeno, and N. Shitsevalova, Sov. Phys. JETP **104**, 120 (2007).
[32] S. Demishev, A. Semeno, H. Ohta, S. Okubo, I. Tarasenko, T. Ishchenko, N. Samarin, and N. Sluchanko, Phys. Solid State **49**, 1295 (2007).
[33] S. Demishev, A. Semeno, A. Bogach, N. Samarin, H. Ohta, S. Okubo, N. Shitsevalova, and N. Sluchanko, Solid State Phenom. **152-153**, 353 (2009).
[34] A. Gurevich and G. Melkov, *Magnetization Oscillations and Waves* (CRC, New York, 1996).
[35] C. Kittel, Phys. Rev. **73**, 155 (1948).
[36] J. Young and E. Uehling, Phys. Rev. **94**, 544 (1954).
[37] G. Feher and A. Kip, Phys. Rev. **98**, 337 (1955).
[38] C. Terzioglu, D. A. Browne, R. G. Goodrich, A. Hassan, and Z. Fisk, Phys. Rev. B **63**, 235110 (2001).
[39] D. Khomskii, arXiv:cond-mat./0104517 (unpublished).
[40] J. van den Brink and D. Khomskii, Phys. Rev. B **63**, 140416(R) (2001).
[41] S. L. Ogarkov, M. Y. Kagan, A. O. Sboychakov, A. L. Rakhmanov, and K. I. Kugel, Phys. Rev. B **74**, 014436 (2006).
[42] U. Yu and B. I. Min, Phys. Rev. Lett. **94**, 117202 (2005).
[43] T. Sasaki, A. G. Lebed', T. Fukase, and N. Toyota, Phys. Rev. B **54**, 12969 (1996).
[44] F. Givord, J.-X. Boucherle, P. Burlet, B. Gillon, and S. Kunii, J. Phys.: Condens. matter **15**, 3095 (2003).
[45] Y. Tanaka, K. Katsumata, S. Shinomura, and Y. Onuki, J. Phys. Soc. Jpn. **74**, 2201 (2005).