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MAGNETIC FORM FACTOR MEASUREMENTS IN CERIUM HEXABORIDE


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Résumé. - Les résultats d'une étude par diffraction de neutrons polarisés du facteur de forme magnétique de l'ion Ce$^{3+}$ dans CeB$_6$ sont décrits. Le facteur de forme ne présente pas l'anisotropie caractéristique d'un doublet $\Gamma_7$. Le niveau fondamental de l'ion Ce$^{3+}$ dans CeB$_6$ ne peut donc pas être un doublet $\Gamma_7$ isolé ($\Delta > 20$ K).

Abstract. - The results of a polarized neutron study of the magnetic form factor of Ce$^{3+}$ ions in CeB$_6$ are described. This form factor does not show the anisotropy characteristic of a $\Gamma_7$ doublet, so the ground state of Ce$^{3+}$ ions in CeB$_6$ cannot be a well isolated $\Gamma_7$ doublet ($\Delta > 20$ K).

1. Introduction. - Cerium hexaboride, with a simple cubic structure of the CeB$_6$ type is a very interesting compound in the sense that it shows the most typical "dense Kondo behaviour" (1), (2) and very peculiar magnetic properties revealed by magnetization (3), specific heat (4), resistivity (1), (2), neutron diffraction (5), (6), (7) and NMR (8) measurements. The magnetic phase diagram of CeB$_6$ as a function of temperature and magnetic field is shown in figure 1. Three distinct phases are observed: a high temperature paramagnetic phase I, an intermediate phase II, the nature of which is not yet elucidated and a low temperature phase III in which a complex antiferromagnetic ordering has been described (5), (6), (7). In spite of the large number of experimental data obtained up to now, the magnetic behaviour of CeB$_6$ is not yet understood. In particular, the ground state of the Ce$^{3+}$ ion is not well established. Susceptibility measurements (9) have been interpreted on the basis of a $\Gamma_7$ doublet as the ground state with the $\Gamma_8$ quartet lying about 60 K above. However to fit the experimental susceptibility curve, anisotropic interactions with different amplitudes within the doublet and the quartet have been introduced in the model which allows to explain the values of $T_N$ (2.3 K) and of $\theta_p$ (-60 K) which differ by more than one order of magnitude. This model disagrees with measurement of magnetic entropy (4) and with inelastic neutron scattering experiments (10) which do not show any crystal field splitting lower than 450 K. The measurement of the magnetic form factor of the Ce$^{3+}$ ion in CeB$_6$ is then highly interesting to get information about the ground state of Ce$^{3+}$. In particular, if a well isolated $\Gamma_7$ doublet is the ground state, the magnetic form factor must shows a typical anisotropy (11).

2. Experimental. - Polarized neutron experiments have been performed at the high flux isotope reactor of the Oak-Ridge National Laboratory. The sample used is a single crystal, prepared by a floating zone method (2) using 99% enriched $^{11}$B. The crystal has the shape of a parallelepiped of $1 \times 1.8 \times 6 \text{ mm}^3$, the largest dimension being a $<110>$ direction. The measurements of the flipping ratio $I^+ / I^-$ were performed at $T = 4.2$ K with a magnetic field of 40 kOe applied along the [110] and [001] directions. An additional measurement was performed with $H = 13$ kOe applied along the [110] direction. Two neutron wave-lengths $\lambda = 1.067$ Å and $\lambda = 0.78$ Å were used allowing a collection of data up to $\sin \theta / \lambda = 0.9$ Å$^{-1}$. The data reduction has been performed using the experimental values of the polarization of the primary beam and
Temperature(K)

Figure 1 - Magnetic phase diagram of CeB6

of the efficiency of the flipping coil. Nuclear structure factors have been calculated using the Fermi lengths: $b_{Ce} = 0.476 \times 10^{-12}$ cm and $b_B = 0.61 \times 10^{-12}$ cm (12). The only position parameter involved in the calculation of the nuclear structure factors is that of the boron atoms located in the special position 6f of the Pm3m space group ($x, 1/2, 1/2$). This parameter $x = 0.200 \pm 0.003$ has been determined in a crystallographic study on a small single crystal of 0.7 mm$^3$ using a 4-circled neutron diffractometer at the CEN-Grenoble with a wave length of 0.94 Å. This study demonstrates the importance of the extinction effects, which are found to be rather large ranging up to 30% for the strongest nuclear peaks. The obtained extinction parameters have been used in the determination of the experimental form factor and the extinction corrections are appreciable for reflections with large structure factors at small values of $\sin \theta / \lambda$ and have no influence when $\sin \theta / \lambda$ is larger than 0.5 Å$^{-1}$. The experimental form factor has been compared with the calculated one corresponding to different ground state configurations. For a 4f wave function $|\Phi> = \sum \alpha_n |JM>$, the form factor has been calculated by using the tensor operator method (13)

$$\mu \tilde{f}(\tilde{H}) = \sum_{K', Q''} Y_{Q''}^{*K''}(\tilde{H}) \Sigma_{MM'} \langle \tilde{K}', Q'| \tilde{H}> \Sigma_{KK'} a_n^{*M'} \sum_{MM'} \tilde{C}_{Q''K'' \tilde{K}' \tilde{M}'}$$
The spherical harmonics $Y_{lm}^{\pm m}(\mathbf{H})$ depend on the two angles $\Theta$ and $\phi$ which characterize the orientation of the scattering vector $\mathbf{H}$, the radial integrals $\langle j^m \rangle(\mathbf{H})$ are taken from relativistic atomic calculations (14). This formula shows that it is possible to refine the coefficients of the ground state wave function which best fit the observed magnetic form factor (15).

3. Results and discussion.

Intermediate phase I I. - The experimental form factors obtained in the intermediate phase II at $T = 4.2$ K and $H = 40$ kOe applied along the [001] and [110] directions are shown in figure 2. For both magnetic field directions the form factor follows a rather continuous variation and, within the experimental accuracy, no systematic anisotropy can be detected. In particular the values of $\mu_f$ corresponding to the (h0h) and (00l) reflexions are not located on two distinct curves as expected for a projection of a $T_2$ ground state along the [110] axis. The experimental values extrapolate at $\sin \Theta / \lambda = 0$ to magnetization values of $0.43 \mu_B$ ($\mathbf{H} / [001]$) and $0.45 \mu_B$ ($\mathbf{H} / [110]$). These values are close to those obtained by magnetization measurements $\mu = 0.46 \mu_B$ and $\mu = 0.48 \mu_B$ respectively for $\mathbf{H}$ along [100] and [110] directions (3). The experimental data have been fitted by choosing several models for the ground state. The fits were performed by excluding the four reflexions at small $\sin \Theta / \lambda$ because their values are very sensitive to the extinction correction and can be affected by a contribution arising from the polarization of the conduction electrons. Whatever the starting wave function, the refinement procedure always gives a nearly pure $T_2$ state with the largest possible value of $\mu$. In figure 2 are also given the results of calculations for $\frac{1}{2}$, $\frac{3}{2}$, $\frac{5}{2}$ wave functions and for the dipole approximation ($\mu_f = Cte[j_{\frac{1}{2}} + C_{\frac{3}{2}}j_{\frac{3}{2}}]$. The $\frac{1}{2}$ state can be immediately rejected while $\frac{3}{2}$, $\frac{5}{2}$ and dipolar curves are in better agreement with the experimental form factor. However, in the range where the refinement has been performed ($0.3 < \sin \Theta / \lambda < 0.9$ Å$^{-1}$) the curve associated with the $\frac{5}{2}$ state differs slightly but significantly from the experimental one, with rather too low values at large $\sin \Theta / \lambda$ and too high values for smaller $\sin \Theta / \lambda$, while the calculation for a $\frac{3}{2}$ wave function or the dipole approximation are in correct agreement with the measured form factor. For $\sin \Theta / \lambda$ smaller than 0.3 Å$^{-1}$ the dipolar curve gives values very close to the experimental ones, especially in the case of $\mathbf{H}$ along the [110] direction and consequently the polarization of conduction electron would be very small. In contrast the $\frac{3}{2}$ wave function gives rise to smaller calculated values than the observed ones and then to a strong positive polarization of conduction electrons. Such a positive contribution has been already observed in several Ce compounds (CeAl$_2$(16)(17), CeSb (18), CeSn$_3$ (19)).

Paramagnetic phase I. - The transition between the purely paramagnetic phase I and the intermediate phase II occurs at low temperature for small values of the magnetic field (15 kOe at $T = 4.2$ K). In order to keep a low temperature value we have measured the magnetic form factor in the paramagnetic phase at $T = 4.2$ K and $H = 13$ kOe applied along the [110] direction. The induced magnetic moment has a quite small value and then the measurements are not very accurate. The experimental values are located around an average curve and no systematic anisotropy can be detected. This average curve extrapolates at $\sin \Theta / \lambda = 0$ to a value of $0.09 \mu_B$ in good agreement with magnetization measurements ($0.10 \mu_B$). Within the accuracy of the experiments, the variation of the form factor in the paramagnetic phase appears very similar to that observed in the phase II.
Figure 2 - Magnetic form factor of Ce$^{3+}$ ion in CeB$_6$. Experimental points correspond to measurements at $T = 4.2$ K and $H = 40$ kOe parallel to the [001] (a) and [110] (b) direction. The calculated values for the dipole approximation (full line), a $|1/2>$ (dash-dotted line), a $|3/2>$ and a $|5/2>$ ground state (dashed lines) are also shown.
4. Conclusion. - The main result of this study of the magnetic form factor of the Ce$^{3+}$ ions in CeB$_6$ is that the hypothesis of a well isolated $T_7$ doublet ($\Delta > 20$ K) is disproved. Unfortunately this polarized neutron study does not allow us to make any conclusion in the paramagnetic state about the nature of the ground state wave function of Ce$^{3+}$ in CeB$_6$, because of the smallness of the induced magnetic moment. The measurements in the intermediate phase II are more accurate, but one should not forget that it is actually an ordered phase as we have shown by preliminary neutron experiments. In this case the interpretation of the measurements is difficult as long as the nature of this ordering is not well established. However, the projected magnetization density corresponds to that of a nearly spherical ion. To detect a possible deviation from this isotropic shape, the accuracy of the measurements must be improved and indeed more experiments are needed.

5. References. -


