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Crystalline electric field excitations in $CeAgSb_2$

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Abstract
Neutron scattering experiments have been carried out that reveal the magnetic excitation in the ferromagnet $CeAgSb_2$. Crystalline electric field excitations were observed at 5.2 and 12.5 meV in the paramagnetic state, indicative of the localized nature of the Ce 4f electron. Our analysis concluded that $|J_z = \pm\frac{1}{2}\rangle$ is the ground state, while the first and second excited levels are mainly due to $|\pm\frac{3}{2}\rangle$ and $|\pm\frac{5}{2}\rangle$, respectively. The observed ferromagnetic moment is in good agreement with the ground-state saturation moment, $g_J \mu_B J_z \sim 0.43\ \mu_B$. The spin wave dispersion is interpreted using the anisotropic Heisenberg model with ferromagnetic exchange interaction up to seventh-nearest neighbours. The spontaneous moment along the c-axis decreases with increasing field perpendicular to the c-axis, and disappears at the critical field H_c, which corresponds to the anomaly which appeared in the magnetization curve.

$CeAgSb_2$ with tetragonal crystal structure orders at $T_c \simeq 9$ K with a small net ferromagnetic component of 0.33 μ_B/Ce [1]. The unusual magnetic properties of $CeAgSb_2$ have not yet been understood. First, the magnetic susceptibility perpendicular to the c-axis is larger than that parallel to the c-axis. However, the magnetic ordered moment below T_c is parallel to the c-axis. Second, the in-plane magnetization below T_c increases linearly with magnetic field and reached about 1.2 μ_B at 3 T [2]. This induced moment is much larger than the spontaneous moment along the c-axis. The antiferromagnetic (AFM) and/or complex magnetic structure was suggested to be the origin of the unusual magnetic properties. However, no AFM peak has been reported from neutron diffraction experiments [1]. Our neutron diffraction experiments also revealed a ferromagnetic moment of 0.41 μ_B along the c-axis, but no antiferromagnetic moment. Recently, study of the thermal and magnetic properties revealed that the magnetic susceptibility could be explained by the crystalline electric field (CEF) effect [3]. It was also

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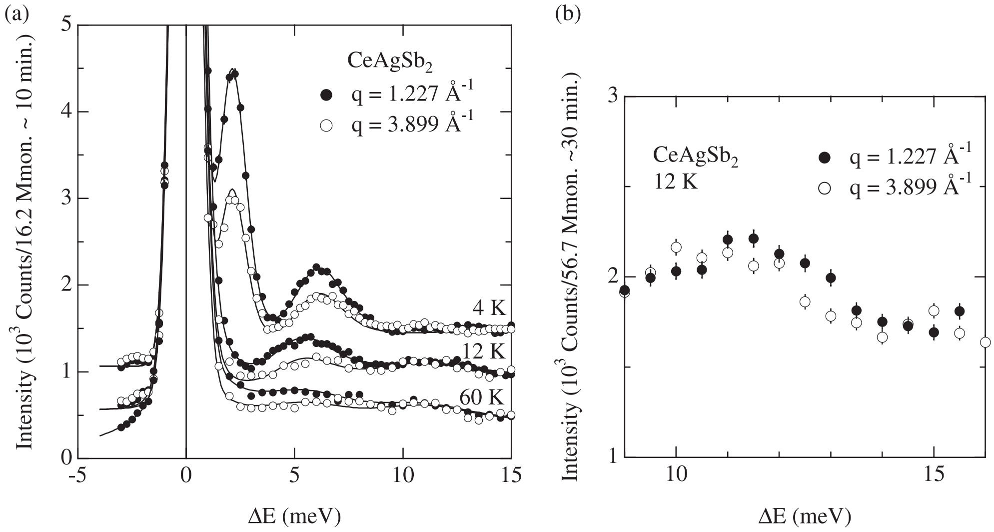

Figure 1. Inelastic neutron scattering spectra measured for a polycrystalline sample of CeAgSb$_2$. The data are shifted vertically for convenience.

revealed that the saturation moment can be understood in terms of the $|J_z = \pm\frac{1}{2}\rangle$ ground state, although why the ferromagnetic component is parallel to the c-axis remains an open question.

A polycrystalline sample was prepared by arc-melting of the constituent elements. The starting materials were 3N (99.9% pure) Ce, 4N Ag, and 5N Sb, with the ratio of 1:1:2.4, to compensate for the loss of antimony due to evaporation. The arc-melted sample was annealed in an evacuated quartz tube at 600 °C for a week. The total mass of the polycrystalline sample was about 30 g. On the other hand, single crystals were grown by the Sb-self-flux method. The starting materials were also 3N Ce, 4N Ag, and 5N Sb in this case. The typical sample dimensions were $15 \times 7 \times 2.5$ mm^3 for 0.85 g. Neutron scattering experiments were carried out at the research reactor JRR-3M in Japan Atomic Energy Research Institute, JAERI. Neutron inelastic scattering spectra were measured using TAS-1 at the final energy $E_f = 14.7$ meV and using LTAS at $E_f = 4.0$ meV. Neutron elastic scattering under magnetic fields was measured using the TAS-2 spectrometer.

Figure 1 shows the neutron inelastic scattering spectra for a polycrystalline sample of CeAgSb$_2$. At $T = 4$ K, we observed remarkable excitation peaks at the neutron energy transfers $\Delta E = 1.9$ and 5.9 meV. The q-dependence of the intensities of these peaks is more or less consistent with the square of the 4f magnetic form factor of the Ce^{3+} ion. This means that these inelastic peaks are due to magnetic excitations. On elevating the sample temperature to $T = 12$ K, just above $T_c = 9.6$ K, the excitation peak at 1.9 meV disappeared. This means that this peak is due to spin wave excitation. The 5.2 meV peak, which corresponds to the 5.9 meV excitation at 4 K, survived in the paramagnetic state, although the peak became weak and broad. At a much higher temperature, $T = 60$ K, the excitation peak spread out over a wide energy range. From the dependences on q and the temperature, we concluded that the 5.2 meV peak is due to CEF excitation. In addition to these clear excitation peaks, a very weak excitation at $\Delta E = 12.5$ meV was observed at 12 K, as shown in figure 1(b). Recently, these two CEF excitations were also observed by Adroja *et al* [4] using a HET spectrometer at ISIS, independently of our work.

This situation can be understood in terms of the $|\pm\frac{1}{2}\rangle$ ground state with the first and the second level dominated by $|\pm\frac{3}{2}\rangle$ and $|\pm\frac{5}{2}\rangle$ states, respectively. In this level scheme,

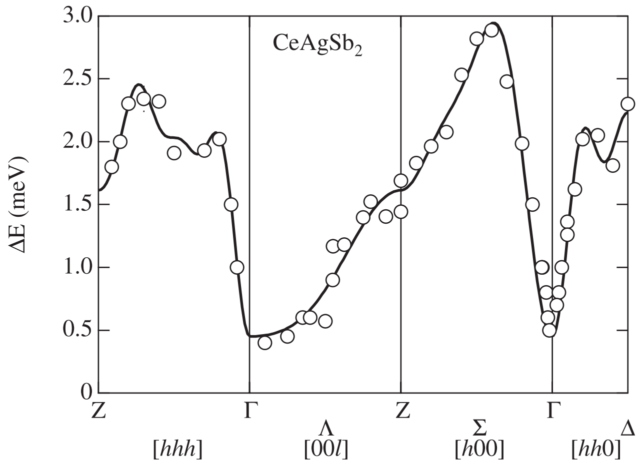

Figure 2. The dispersion relation of the spin wave excitation in $CeAgSb_2$. The solid curve shows the calculated dispersion curve.

we observe strong first and weak second excitation peaks, because $\langle \pm\frac{1}{2}|J_{x,y}|\pm\frac{3}{2}\rangle$ is nonzero and $\langle \pm\frac{1}{2}|J_{x,y,z}|\pm\frac{5}{2}\rangle = 0$. The magnetic moment of the ground state, $g_J \mu_B J_z = 0.428\ \mu_B$, is in good agreement with the observed ferromagnetic moment of 0.41 μ_B. The CEF level scheme determined by the present neutron scattering study is very similar to the result of the recent study by Takeuchi *et al* [3].

The spin wave dispersion, as shown in figure 2, has been measured with the use of a single-crystalline sample. The solid curve in figure 2 is the calculated dispersion relation within the anisotropic Heisenberg model. We regard the crystal structure as body-centred tetragonal for simplicity. The exchange interactions J_n up to ninth neighbours were taken into account in the model calculation. The J_ns obtained, up to the seventh, are ferromagnetic.

Our experimental results, (i) the absence of an AFM peak and (ii) ferromagnetic exchange interactions up to seventh-nearest neighbours, clearly indicate that $CeAgSb_2$ has a simple ferromagnetic ordering with the $|\pm 1/2\rangle$ ground state. We note that the 'unusual' properties which were interpreted as properties characteristic of antiferromagnets can be explained on the basis of a simple ferromagnet.

The open symbols in figure 3 show the in-plane magnetization measured from the neutron diffraction. A kink, indicated by arrows, appears below T_c. On the other hand, the full symbols in figure 3 show the ferromagnetic moment along the c-axis with the magnetic field parallel to [110] measured from the neutron diffraction intensity at the (110) peak position. With this configuration, the in-plane magnetization does not contribute to any magnetic scattering, because the induced in-plane magnetic moment is parallel to the scattering vector q. Therefore only the ferromagnetic component along [001] was observed. The ferromagnetic component along [001] exhibits a gradual decrease with increasing in-plane magnetic field, and disappears at the critical field H_c where the in-plane magnetic moment shows a kink behaviour. Our data indicate that H_c is the field where the ferromagnetic moment switches its direction from parallel to perpendicular relative to [001]. It is why a kink is observed in the magnetization curve for the field direction perpendicular to [001].

In conclusion, our neutron scattering study revealed simple ferromagnetic structure of $CeAgSb_2$. The CEF level scheme consists of the $|\pm\frac{1}{2}\rangle$ ground state and the first and second excited levels dominated by $|\pm\frac{3}{2}\rangle$ and $|\pm\frac{5}{2}\rangle$, respectively. We showed that the magnetic

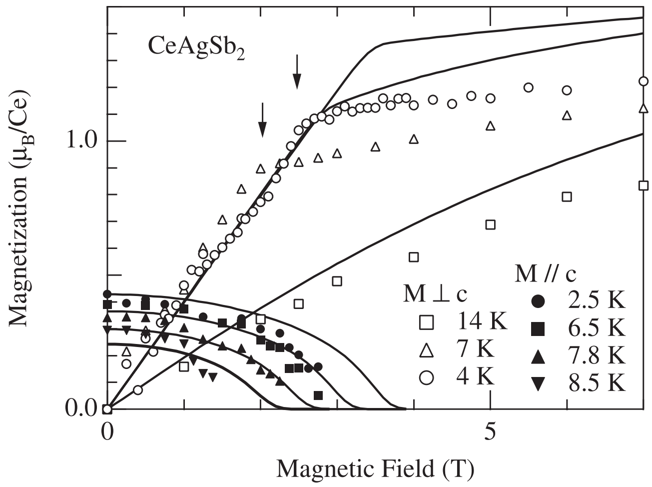

Figure 3. The in-plane magnetization ($M \perp c$) and the ferromagnetic moment ($M \parallel c$) are plotted as a function of the magnetic field perpendicular to [001].

properties of $CeAgSb_2$ can be well understood in terms of a $|\pm\frac{1}{2}\rangle$ ground state with anisotropic exchange interaction.

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