THERMAL EXPANSION AND MAGNETOSTRICTION STUDIES OF A KONDO LATTICE COMPOUND: $CeAgSb_2$*

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(Received July 10, 2002)

We have investigated a single crystal of $CeAgSb_2$ using low field ac-susceptibility, thermal expansion and magnetostriction measurements. The thermal expansion coefficient α, exhibits highly anisotropic behaviour between 3 K and 80 K: α (for $\Delta L/L$) $\perp c$ exhibits a sharp peak at $T_{\rm N}$ followed by a broad maximum at 20 K, while a sharp negative peak at $T_{\rm N}$ followed by a minimum at 20 K has been observed for ($\Delta L/L$ $\|$) the c direction. The observed maximum and minimum in α(T) at 20 K have been attributed to the crystalline field effect (CEF). The magnetostriction (MS) also exhibits anisotropic behaviour with a large MS along the c-axis.

PACS numbers: 75.30.Mb, 65.40.De, 75.80.+q

Recent studies on $RAgSb_2$ (R=rare earth) compounds have shown that these compounds crystallize in the tetragonal $ZrCuSi_2$ type structure [1–6]. Among these compounds, $CeAgSb_2$ is the most interesting. The resistivity and thermoelectric power of $CeAgSb_2$ show a typical Kondo lattice behaviour [1]. The magnetization exhibits strongly anisotropic behaviour

* Presented at the International Conference on Strongly Correlated Electron Systems, (SCES 02), Cracow, Poland, July 10–13, 2002.

with the easy c-axis of magnetization in the magnetic ordered state (probably a complex antiferromagnetic (AFM) state with a FM component) below 10 K, while in the paramagnetic state there is an easy ab-plane [1,5]. The magnetization isotherm at 2 K shows that the easy magnetization direction changes from the c-axis to the ab-plane above 1 T field with the saturated moment $\mu^{\rm sat} = 1.1\mu_{\rm B}$/Ce-ion for $B \perp c$ and $0.37\mu_{\rm B}$/Ce-ion for $B \parallel c$ at 5.5 T field. The neutron diffraction measurements at 2 K show the presence of only a single magnetic Bragg peak, (1 0 1), with moment of $0.33\mu_{\rm B}$/Ce-ion [5]. The zero-field μSR study shows well-defined frequency oscillations with an anomalously low internal field of 53 mT at the muon site, which is in agreement with the extremely low frequency (0.25 MG) observed in a Shubnikov–de Haas study [4,6].

In the present work we have investigated a single crystal of $CeAgSb_2$ using ac-susceptibility ($\chi_{\rm ac}$), thermal expansion, and magnetostriction measurements with the aim of throwing more light on the complex electronic and magnetic ground state. The single crystal of $CeAgSb_2$ was grown in an evacuated BN-crucible at 1350°C by the Bridgemann method. The inductive component, $\chi'_{\rm ac}(T)$ of the ac-susceptibility of $CeAgSb_2$ single crystal with $B_{\rm ac}$ (5G) $\parallel c$, exhibits a sharp peak at 9.7 K which is due to the magnetic ordering of the Ce-moments (inset of Fig. 1). $\chi'_{\rm ac}(T)$ for $B_{\rm ac} \perp c$ also exhibits a similar peak, but the peak height is only 27% of $B_{\rm ac} \parallel c$. The c-axis is therefore the easy magnetization direction at low temperature, which is in agreement with the dc magnetization study [4]. It would be interesting to compare the $\chi'_{\rm ac}(T)$ signal of $CeAgSb_2$ with that from the ferromagnetic CePdSb [7]. Well below $T_{\rm N}$ (or $T_{\rm C}$) $\chi'_{\rm ac}(T)$ of $CeAgSb_2$ is very small, while

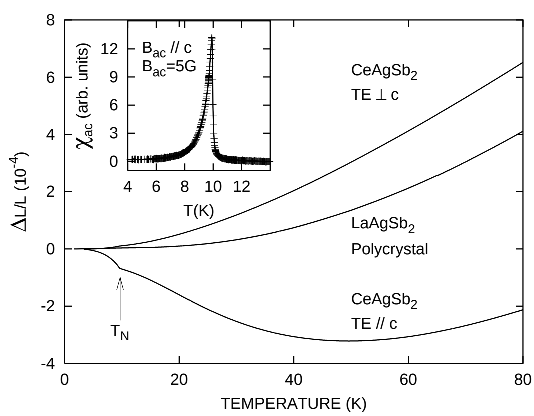

Fig. 1. Linear thermal expansion ($\Delta L/L$) *versus* temperature of $CeAgSb_2$ single crystal and $LaAgSb_2$ polycrystal. The inset shows temperature dependence of inductive, $\chi'_{\rm ac}(T)$ component of the ac-susceptibility for $B_{\rm ac} \parallel c$.

that of CePdSb retains about 92% of its peak value. This again implies that the magnetic ground state of $CeAgSb_2$ is more complicated than that of a simple AFM or FM.

Fig. 1 shows the linear thermal expansion (TE $= \Delta L/L$) as a function of temperature for $CeAgSb_2$ single crystal parallel to c-axis (TE$\,\|\, c$) and perpendicular to c-axis (TE$\perp c$) along with the isostructural nonmagnetic reference polycrystalline $LaAgSb_2$. $\Delta L/L$ of $LaAgSb_2$ exhibits a typical behaviour expected for the thermally excited phonons. On the other hand, $\Delta L/L$ of $CeAgSb_2$ shows highly anisotropic behaviour, positive for TE$\perp c$ and negative for TE$\,\|\, c$, with a sudden change at $T_{\rm N}$ in both the directions. The magnetic contribution to the thermal expansion coefficient, $\alpha_M(T)$ of $CeAgSb_2$ along both the directions was estimated by subtracting $\alpha(T)$ of $LaAgSb_2$. $\alpha_M(T)$ exhibits a sharp peak and a broad peak at $T_{\rm N}$ and 20 K, positive for TE$\perp c$ and negative for TE$\,\|\, c$, respectively. It should be noted that the $\alpha_M(T)$ of polycrystalline $CeAgSb_2$ also shows a broad peak at 18 K, but no clear peak at $T_{\rm N}$ [3]. The absence of the peak at $T_{\rm N}$ in the polycrystalline sample might be due to the cancellation of positive (for TE$\perp c$) and negative (for TE$\,\|\, c$) contributions observed in the single crystal. The sharp peak at $T_{\rm N}$ in $CeAgSb_2$ single crystal arises due to the development of anisotropic spin-spin correlations because of the magnetic ordering of Ce-moments. On the other hand the broad peak (maximum and minimum) at 20 K in both the directions has been attributed to the CEF effect on the $J = 5/2$ state of the Ce^{3+} ion. This is consistent with our recent high resolution inelastic neutron scattering measurements on $CeAgSb_2$, which show two well defined crystal field excitations, at 5.1 meV and 12.4 meV, as expected for the tetragonal point symmetry of the Ce ion [8]. It is interesting to note that the observed anisotropic behaviour of $\alpha_M(T)$ of $CeAgSb_2$ is very similar to that observed for $CeRhIn_5$ single crystal, which also has the tetragonal crystal structure [9]. The calculated $\alpha_M(T)$ for $CeRhIn_5$ on the basis of the CEF model exhibits a maximum and minimum around 25 K for [100] and [001] directions, respectively [9]. In order to investigate the effect of magnetic field on the $\alpha_M(T)$ of $CeAgSb_2$, we have measured α_M(T) in an applied magnetic field of 8 T (Fig. 2). The observed sharp peak at $T_{\rm N}$ in zero field was almost suppressed in 8 T field for both the directions.

We estimated the value of $dT_{\rm N}/dP = -0.088$ (K/kbar), using the Ehrenfest relation and the heat capacity data from Ref. [2], which is in good agreement with the experimentally measured value of -0.095 (K/kbar) on the polycrystalline $CeAgSb_2$ [3]. The negative sign of $dT_{\rm N}/dP$ indicates that $CeAgSb_2$ is on the right-hand side of the Doniach phase diagram [3].

Fig. 3 shows the magnetostriction (MS) isotherms measured at various temperatures for $(\Delta L/L) \parallel c$ and $(\Delta L/L) \perp c$ with applied fields $B \parallel c$ and $B \perp c$ directions. MS exhibits highly anisotropic behaviour with the

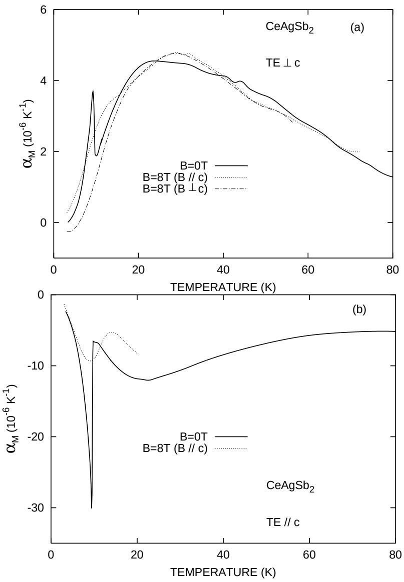

Fig. 2. The magnetic contribution to the linear thermal expansion coefficient, α_M(T) of $CeAgSb_2$ single crystal in zero field and 8T field, (a) TE $\perp c$ and, (b) TE $\parallel c$.

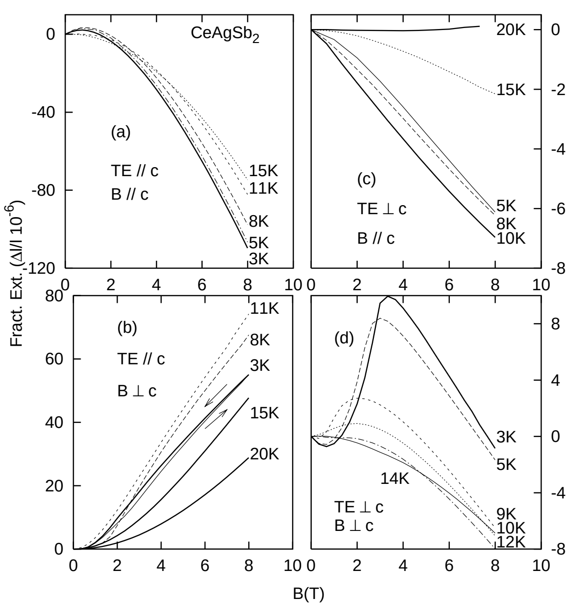

Fig. 3. The magnetostriction isotherms at various temperatures of $CeAgSb_2$ single crystal, (a) and (b) $(\Delta L/L) \parallel c$ and, (c) and (d) $(\Delta L/L) \perp c$, for both $B \parallel c$ and $B \perp c$.

largest length change for $(\Delta L/L) \parallel c$. Between 14Kand 20K MS exhibits a quadratic behaviour, for all measured directions (Figs. 3(a)–(d)), taht could be understood on the basis of the free energy of the system in an applied field. An interesting behaviour of MS is observed for $(\Delta L/L) \perp c$ and $B \perp c$ geometry at low temperatures (Fig. 3(d)). At 3 K MS exhibits a peak at 3.3 T, which is consistent with the observed peak in the magnetoresistance measurements and has been attributed to the field induced transition to the easy *ab*-plane of magnetization [4]. Furthermore, with increasing temperature from 3 K, the position of the peak moves to a lower field, which indicates that the smaller critical field for the field induce transition. A small hysteresis in MS was observed at 3 K suggesting the presence of a FM component. This result along with the absence of the domain walls contribution at low fields in MS of $CeAgSb_2$ indicates that the magnetic ground state is not a simple FM, but a complex AFM.

In conclusion, thermal expansion and magnetostriction measurements of $CeAgSb_2$ single crystal exhibit highly anisotropic behaviour. These results indicate that the anisotropic magnetic exchange and CEF-anisotropy are playing an important role.

REFERENCES

[1] M. Houshiar *et al.*, *J. Magn. Magn. Mater.* **140-144**, 1232 (1995).

[2] Y. Muro *et al.*, *J. Alloy. Compd.* **257**, 23 (1997).

[3] M.J. Thornton *et al.*, *J. Phys. Cond. Matter* **10**, 9485 (1998).

[4] K.D. Myers *et al.*, *J. Magn. Magn. Mater.* **205**, 27 (1999); K.D. Mayers *et al.*, *Phys. Rev.* **B60**, 13371 (1999).

[5] G. Andre *et al.*, *Physica* **B292**, 176 (2000).

[6] J.A. Dann *et al.*, *Physica* **B289-290**, 38 (2000).

[7] D.T. Adroja *et al.*, *Phys. Rev.* **B61**, 1232 (2000).

[8] D.T. Adroja *et al.*, unpublished.

[9] T. Takeuchi *et al.*, *J. Phys. Soc. Jpn.* **70**, 877 (2001).