THERMAL EXPANSION AND MAGNETOSTRICTION STUDIES OF A KONDO LATTICE COMPOUND: CeAgSb$_2$

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We have investigated a single crystal of CeAgSb$_2$ using low field ac-susceptibility, thermal expansion and magnetostriction measurements. The thermal expansion coefficient $\alpha$, exhibits highly anisotropic behaviour between 3K and 80K: $\alpha$ (for $\Delta L/L)$ $\perp c$ exhibits a sharp peak at $T_N$ followed by a broad maximum at 20K, while a sharp negative peak at $T_N$ followed by a minimum at 20K has been observed for ($\Delta L/L\parallel c$) the $c$ direction. The observed maximum and minimum in $\alpha(T)$ at 20K have been attributed to the crystalline field effect (CEF). The magnetostriction (MS) also exhibits anisotropic behaviour with a large MS along the $c$-axis.

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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 with the easy $c$-axis of magnetization in the magnetic ordered state (probably a complex antiferromagnetic (AFM) state with a FM component) below 10K, while in the paramagnetic state there is an easy ab-plane [1,5]. The magnetization isotherm at 2K shows that the easy magnetization direction changes from the $c$-axis to the ab-plane above 1T field with the saturated moment $\mu^{sat}=1.1\mu_B$/Ce-ion for $B\perp c$ and $0.37\mu_B$/Ce-ion for $B\parallel c$ at 5.5T field. The neutron diffraction measurements at 2K show the presence of only a single magnetic Bragg peak, (1 0 1), with moment of $0.33\mu_B$/Ce-ion [5]. The zero-field $\mu$SR study shows well-defined frequency oscillations
with an anomalously low internal field of 53mT at the muon site, which is in agreement with the extremely low frequency (0.25MG) 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 ($\chi_{ac}$), 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, $\chi'_{ac}(T)$ of the ac-susceptibility of CeAgSb$_2$ single crystal with $B_{ac}(5G) \parallel c$, exhibits a sharp peak at 9.7K which is due to the magnetic ordering of the Ce-moments (inset of Fig.1). $\chi'_{ac}(T)$ for $B_{ac} \perp c$ also exhibits a similar peak, but the peak height is only 27% of $B_{ac} || c$. 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 $\chi'_{ac}(T)$ signal of CeAgSb$_2$ with that from the ferromagnetic CePdSb [7]. Well below $T_N$ (or $T_C$) $\chi'_{ac}(T)$ of CeAgSb$_2$ is very small, while 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 ($\text{TE} = \Delta L/L$) as a function of temperature for CeAgSb$_2$ single crystal parallel to c-axis ($\text{TE} || c$) and perpendicular to c-axis ($\text{TE} \perp c$) 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 $\text{TE} \perp c$ and negative for $\text{TE} || c$, with a sudden change at $T_N$ 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 $T_N$ and 20K, positive for $\text{TE} \perp c$ and negative for $\text{TE} || c$, respectively. It should be noted that the $\alpha_M(T)$ of polycrystalline CeAgSb$_2$ also shows a broad peak at 18K, but no clear peak at $T_N$ [3]. The absence of the peak at $T_N$ in the polycrystalline sample might be due to the cancellation of positive (for $\text{TE} \perp c$) and negative (for $\text{TE} || c$) contributions observed in the single crystal. The sharp peak at $T_N$ 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 20K 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 5.1meV and 12.4meV, 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 25K 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 \( \alpha_M(T) \) in an applied magnetic field of 8T (Fig.2). The observed sharp peak at \( T_N \) in zero field was almost suppressed in 8T field for both the directions.

We estimated the value of \( dT_N/dP = -0.088 \) (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 \( dT_N/dP \) 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 \( (\Delta L/L)_c \) and \( (\Delta L/L)_{\perp c} \) with applied fields \( B||c \) and \( B\perp c \) directions. MS exhibits highly anisotropic behaviour with the largest length change for \( (\Delta L/L)_c \). Between 14K and 20K MS exhibits a quadratic behaviour, for all measured directions (Figs.3a-d), that 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.3d). At 3K MS exhibits a peak at 3.3T, 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 3K, 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 3K 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.

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Figure captions

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, $\chi_{ac}'(T)$ component of the ac-susceptibility for $B_{ac}||c$.

Fig.2 The magnetic contribution to the linear thermal expansion coefficient, $\alpha_M(T)$ of CeAgSb$_2$ single crystal in zero field and 8T field, (a) TE$\perp c$ and, (b) TE$||c$.

Fig.3 The magnetostriction isotherms at various temperatures of CeAgSb$_2$ single crystal, (a) and (b) ($\Delta L/L$)$||c$ and, (c) and (d) ($\Delta L/L$)$\perp c$, for both B $||c$ and B$\perp c$. 

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\( \chi_{ac} \) (arb. units)

\( T(K) \)

\( B_{ac} \parallel c \)

\( B_{ac} = 5G \)

\( \Delta L/L \times 10^{-4} \)

Temperatures (K)

CeAgSb\(_2\)

TE \( \perp c \)

LaAgSb\(_2\)

Polycrystal

CeAgSb\(_2\)

TE \( \parallel c \)

\( T_N \)
