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Letter

Electronic specific heat of Mn_2Sb and $Mn_{1.9}Cr_{0.1}Sb$

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Abstract

In order to investigate the difference between the electronic structures of the ferrimagnetic and antiferromagnetic states in $Mn_{2-x}Cr_xSb$ compounds the specific heat has been measured for Mn_2Sb and $Mn_{1.9}Cr_{0.1}Sb$ which have ferrimagnetic (FR) and antiferromagnetic (AF) structures respectively at low temperatures. This revealed a great difference between the electronic specific heat coefficients γ for these two compounds: $\gamma_{FR} = 71.4$ mJ mol^{-1} K^{-2} and $\gamma_{AF} = 20.9$ mJ mol^{-1} K^{-2}. This difference allows us to suggest that the AF–FR magnetic transition in $Mn_{2-x}Cr_xSb$ is caused by the peculiarities of electronic structure and the band magnetism model is necessary to explain the magnetic properties of these compounds.

The Mn_2Sb-based compounds with manganese atoms substituted by a number of elements (chromium, copper, zinc, cobalt and vanadium) and antimony atoms substituted by arsenic and germanium reveal magnetic phase transitions of first order from the antiferromagnetic (AF) to the ferrimagnetic (FR) state as the temperature increases [1–3]. In the region of this transition the hysteresis phenomenon, great changes in the lattice parameters and electrical resistivity were observed [4, 5].

The well-known Kittel exchange inversion model was applied to explain the nature of phase transitions in Mn_2Sb-based compounds [6]. In this theory the exchange interaction depending on the interatomic distance changes its sign when the lattice parameter reaches the critical value. However, some experimental results obtained for the Mn_2Sb-based compounds cannot be explained in terms of this model, *e.g.* the dependence of the critical temperature of the phase transition not only on the variations in lattice parameters but

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also on the types of substitutional atom or the large change in the electrical resistivity (about 100 $\mu\Omega$ cm) at the AF–FR transition [5].

Similar effects were observed in FeRh-based compounds [7, 8]. The earlier interpretation of the first-order phase transitions in FeRh-based compounds was also based on Kittel's model, but the studies of the concentration dependence of the critical transition temperatures for the different substitutional elements [8, 9], the measurements of the electronic part of the specific heat [10], the measurements of electrical properties [9] and the calculations of the band structure [11] allow us to conclude that their magnetic properties can be described in terms of the band model.

In this paper the temperature dependences of the specific heat C_p for Mn_2Sb and $Mn_{1.9}Cr_{0.1}Sb$ at low temperatures are presented.

Mn_2Sb is a ferrimagnet and its Curie temperature is 550 K. The magnetic moments of manganese atoms situated in different crystallographic positions of the tetragonal lattice have different values and antiparallel orientations [12]. In $Mn_{1.9}Cr_{0.1}Sb$ an AF arrangement is observed at low temperatures and, when the temperature increases, a magnetic phase transition to the FR state takes place [3].

Mn_2Sb and $Mn_{1.9}Cr_{0.1}Sb$ alloys were melted in a helium atmosphere in an induction furnace. Doubly distilled manganese and antimony of 99.999% purity were used. The alloys were annealed at 750 °C for 1 week. X-ray diffraction of the powders was carried out using Cr Kα radiation, and a main Mn_2Sb phase with a small amount of MnSb phase was observed.

The temperature dependence of magnetization for $Mn_{1.9}Cr_{0.1}Sb$ allows us to establish the temperature of the AF–FR magnetic transition as equal to 250 K. This value differs from that given in ref. 5, probably as a result of the high sensitivity of the transition temperature to the method of synthesis and to the small variations in chromium content. The measurements of the specific heat at low temperatures were carried out by an adiabatic method.

Good linear dependence of C_p/T on T^2 was observed for both compounds (Fig. 1). This allows us to obtain the value of the electronic specific heat coefficient γ. For the FR compound Mn_2Sb, γ_{FR} is equal to 71.4 mJ mol^{-1} K^{-2} and for the AF compound $Mn_{1.9}Cr_{0.1}Sb$, γ_{AF} is equal to 20.9 mJ mol^{-1} K^{-2}. The large difference between γ_{FR} and γ_{AF} demonstrates the essential difference between the density of electronic states in the AF and FR structures and also shows that the Fermi level lies in the region of large variations in $N(E)$.

Neutron diffraction data show that the period of the AF structure along the tetragonal axis for $Mn_{1.9}Cr_{0.1}Sb$ is equal to a double period of the FR structure for Mn_2Sb along the same axis [3]. Therefore the transition from the AF to the FR state should be accompanied by the disappearance of superzones [13] and energy gaps on superzone boundaries. Thus the presence of these gaps can lead to a large difference between the electrical resistivities of the AF and the FR states.

The band structure of the Mn_2Sb-based compounds has not yet been estimated. However, the difference obtained between γ_{FR} and γ_{AF} allows us

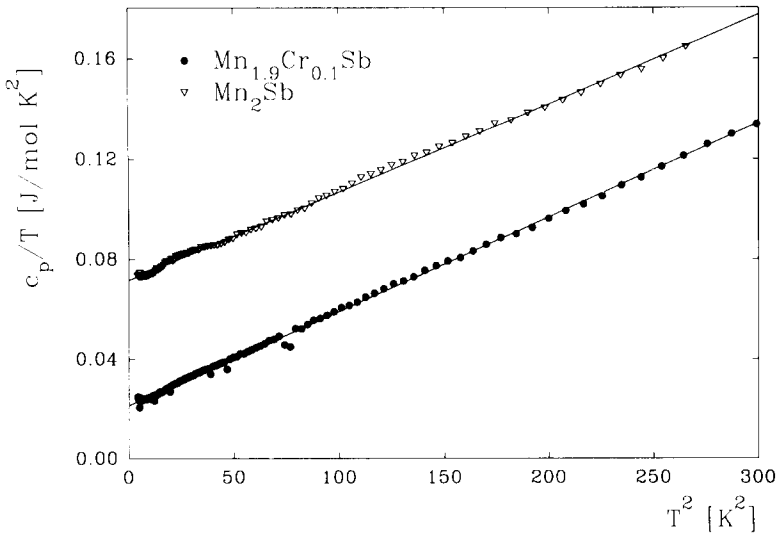

Fig. 1. C_p/T *vs.* T^2 for Mn_2Sb and $Mn_{1.9}Cr_{0.1}Sb$.

to suggest that, similarly to FeRh, the nature of the AF–FR transitions in $Mn_{2-x}A_xSb_{1-y}B_y$ is defined by the peculiarities of the electronic structure and that the description of their magnetic properties requires the use of the band model.

The possibility of the AF–ferromagnetic transition under the influence of an applied magnetic field in the band model was suggested by Wohlfarth [14]. According to ref. 14, such a transition can take place under conditions determined by the character of the $N(E)$ curve near E_F, namely when dN/dE is large and/or when $N(E)$ has a large negative curvature. The first condition, as follows from the present work, is probably correct in $Mn_{2-x}A_xSb_{1-y}B_y$.

It would undoubtedly be interesting to make measurements of the electronic specific heat coefficient γ in the AF and FR states of the same sample in an external magnetic field and without an external magnetic field. Such experiments will be carried out in the near future.

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