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Specific heat and de Haas–van Alphen effect in NiAs

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Abstract

The specific heat at the temperature range from 1.7 to 30 K and the dHvA effect at 0.55 K in the field up to 9.5 T have been measured for Pauli paramagnetic NiAs. The electronic specific heat coefficient is 3.0 mJ/(K^2 mol) which is in good agreement with the value estimated from the calculated density of states. It is found that the dHvA oscillation in the field along [0001] consists of four frequencies which correspond to the extremal areas in the k-space of 0.06, 0.11, 0.16 and 0.18 $Å^{-2}$.

Keywords: NiAs; Specific heat; de Haas–van Alphen effect; Fermi surface

The 3d transition metal chalcogenides and pnictides with either a NiAs-type or a pseudo-NiAs-type structure have various electric and magnetic properties. They have been intensively studied in view of the relation between the magnetism and the crystalline structure. NiAs has been considered as a prototype of the materials having the NiAs-type structure. However, it was pointed out by electron diffraction that its structure is slightly deviated from the ideal NiAs-type [1]. Its magnetism is Pauli paramagnetic; the susceptibility, χ_g, is almost independent of temperature and 0.25×10^{-6} cm^3/g [2]. The electronic band structure of NiAs was calculated theoretically [3]. However, the topology of the Fermi surfaces has never been studied experimentally. In this paper, we report the results of the dHvA effect and the specific heat measurements for NiAs.

The samples were prepared from Ni wires (99.99%) and As shots (99.9999%) by heating weighed quantities in an evacuated silica tube for 7 days at 800°C and for 2 days at 950°C. After careful gliding, they were reheated for 2 days at 950°C. It is confirmed by an X-ray powder diffraction using Cu K_α radiation that they contain only a NiAs phase. No reflection concerned with the distortion above can been observed. The lattice constants, a and c, are 3.618 and 5.033 Å, respectively. A single crystal was made from the powder sample by a Bridgman method using a silica crucible. It is confirmed by a Laue photograph that the sample is a single crystal. It is found by the temperature dependence of the electric resistivity along the c axis, ρ_c, that the ratio of $\rho_c(300\ K)/\rho_c(4.2\ K)$ is about 20. We can observe the dHvA oscillations with this single crystal.

The specific heat was measured for the 1.2455 g polycrystal from 1.7 to 30 K by an adiabatic method. The measurements of dHvA effect were carried out in the field applied along [0001] at 0.55 K by the field modulation method under the following two conditions. In the first condition, an AC modulation field, H_m, is 18 Oe at 134 Hz in the applied field from 3 to 9.5 T at the sweeping rate of 18 T/h and in the second condition $H_m = 28$ Oe at

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84 Hz in the field from 7.5 to 9.5 T at the rate of 2 T/h.

The specific heat, c, is shown in Fig. 1 in the form of c/T versus T^2. The c/T value has a linear relation to T^2 at low temperature and can be fitted using $c/T = \gamma + AT^2$ within the temperature range from 2.5 to 5.1 K. We note that the fitted region is narrower than that in the other metals. The obtained γ value, 3.0 mJ/(K^2 mol), is in good agreement with that estimated from the calculated density of states but is twice larger than that estimated from χ_g [2–4].

The dHvA oscillations were observed under both the conditions above. These oscillations were analyzed using a fast fourier transform (FFT) and a maximum entropy method (MEM) by which the frequencies can be obtained more exactly than those by the FFT analysis. It was found under the first condition that the oscillation consists of two rather broad dHvA frequencies around 300 and 800 T. These frequencies correspond to the extremal area in the k-space, $S = 0.03$ and 0.08 Å^{-2}, respectively, which are about 1% of the cross section of the Brillouin zone perpendicular to [0001]. Since the sweeping rate is too fast under this condition, it is difficult to observe the oscillation with the higher frequencies. Figs. 2 and 3 show the oscillation against H^{-1} under the second condition in the field along [0001] and the FFT and MEM spectra,

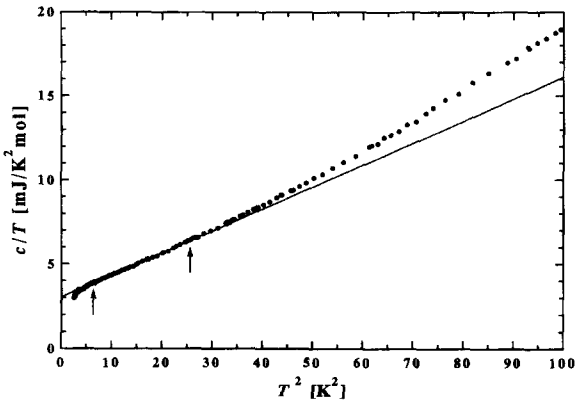

Fig. 1. Low-temperature specific heat of NiAs in a form of c/T versus T^2. The dashed line shows the least-squares fit within the range between two arrows.

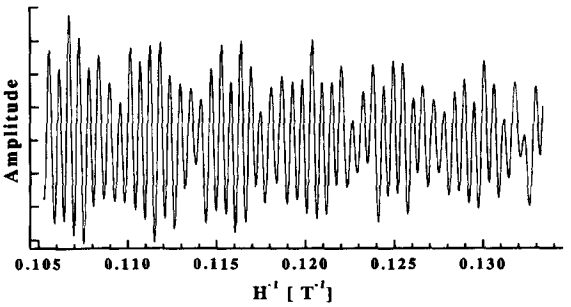

Fig. 2. dHvA oscillation versus H^{-1} in the field from 7.5 to 9.5 T at the rate of 2 T/h applied along [0001] with the modulation field 28 Oe at 84 Hz.

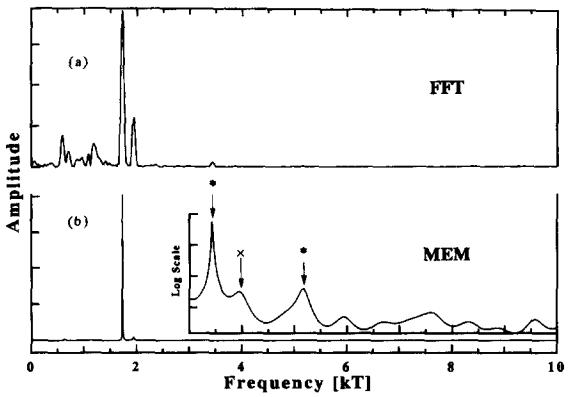

Fig. 3. FFT (a) and MEM spectra (b) of the oscillation shown in Fig. 2. The inset shows the spectrum above 3000 T in a log scale. The peaks labeled with the asterisk and the cross are the higher harmonics.

respectively. The MEM analysis reveals the four peaks of the narrow width at the frequencies of 620, 1120, 1720 and 1930 T. The peaks labeled with the asterisk in the inset of Fig. 3(b) are the second and third harmonics of the 1720 T frequency and the peak labeled with the cross the second harmonics of the 1930 T frequency. The S value of the four frequencies are estimated to be 0.06, 0.11, 0.16 and 0.18 Å^{-2}, respectively. The origin of the 1930 T frequency is most likely caused by the orbit of the hole Fermi surface lying at the center of the Brillouin zone predicted by the band calculation since this area is comparable to that estimated from the hole surface [3].

We have observed the dHvA oscillations in NiAs. The oscillation in the field along [0001] consists of four frequencies. One of them is attributed to the orbit of the hole surface at the center of the Brillouin zone and the others are not found to correspond to any orbits, though the Fermi surface was predicted by the band calculation. The γ value is in good agreement with that estimated from the calculated density of states. We have measured the oscillation in the main crystallographic planes recently and are making an analysis at present.

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