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Antiferromagnetic correlations in CeNiSn

Yumin Hou†‡, Fuming Yang‡, Duo Jin†, Yupeng Wang† and Qize Ran†
† Cryogenic Laboratory, Chinese Academy of Sciences, Beijing 100080, People's Republic of China
‡ Institute of Physics, Chinese Academy of Sciences, Beijing 100080, People's Republic of China

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Abstract. From measurements of magnetic properties of CeNiSn, it has been found that a kink appears at about 3 T in the magnetization curve at 4.5 K while there is no kink at 15 K. Moreover, magnetic field can shift the peak of magnetic susceptibility to lower temperature, which gives further evidence of the existence of antiferromagnetic correlations. This paper also discusses the role that antiferromagnetic correlations play.

1. Introduction

CeNiSn first as a semiconductor [1], and now as a semimetal [2], has attracted much attention because this compound has some properties different from other Kondo insulators. NMR experiments [3, 4] show that in CeNiSn the density of states (DOS) is of a pseudo-gap type with a V-shaped structure and a finite density of states exists at the Fermi level. The gap was also observed in tunnelling experiments [5]. Moreover, it is different from the conventional semiconductors because the band gap is formed by the highly renormalized quasiparticles near the Fermi level and thus the gap has meaning only in the low-temperature region. However, the pseudogap is not very sensitive to the presence of impurity phases [6], but it is easily suppressed by alloying [7, 8] and application of pressure [9] or magnetic field [10]. Therefore, it is interesting to investigate this compound which is near the borderlines of the magnetic–nonmagnetic (M–NM) and metal–insulator (M–I) transitions.

The mechanism of the pseudogap is not yet very clear. Some experiments [1, 11–13] suggested that the antiferromagnetic correlation and Kondo coherence developed below the coherence temperature $T_{coh} = 20$ K may be the keys to understand the formation of the pseudogap. The existence of the antiferromagnetic correlations was first suggested from the measurement of static susceptibility [1], and then it was observed in the neutron inelastic excitation spectra [14, 15]. In the spectra, the peaks at 2 and 4 meV were regarded as three-dimensional and quasi-one-dimensional dynamic antiferromagnetic correlations, respectively. In addition, the high-pressure neutron scattering and Co-substitution neutron scattering experiments showed the coexistence of the pseudogap and the peaks [12, 13].

To explain the phenomena in magnetic fields, a rigid-band model was set up in which a V-shaped quasiparticle band was split into up- and down-spin bands with a relative Zeeman shift of $\Delta(k_B) = 2gJ\mu_B H$. It gave a qualitative explanation to the experiments of magnetoresistance [16], specific heat in magnetic fields [17], magnetic field dependence of $(T_1T)^{-1}$ in NMR studies [18], and magnetic susceptibility [17] in low magnetic field. In this model, it was suggested that the decrease of χ with decreasing temperature was due to the pseudogapped DOS rather than the antiferromagnetic correlation [17]. To check whether the peak of χ–T was associated with antiferromagnetic correlation, an experiment was designed to measure magnetic susceptibility in different magnetic fields.

2. Experiments

Under the protection of flowing high-purity Ar, an ingot of CeNiSn in a 1.02:1:1 proportion of individual elements was melted three times. Then it was sealed in a quartz tube under high vacuum and annealed at 800 °C for two weeks. X-ray diffraction identifies the single phase of ε-TiNiSi structure. Magnetization and magnetic susceptibility curves were obtained by using an MPM-7 type SQUID magnetometer.

3. Results and discussion

Magnetization curves (figure 1) were measured at two temperatures, $T = 4.5$ K and $T = 15$ K, which were chosen after and before gap opening, respectively. At 4.5 K, there is a kink at about 3 T in the magnetization curve of CeNiSn, which suggests a metamagnetic transition, while the curves either side of the kink behave linearly. At 15 K, the magnetization curve is smooth. The magnetic susceptibility was measured at $H = 1, 2, 3$ and 4 T (figure 2) which are lower or higher than the magnetic field at which the kink appears. At 1 T, an obvious peak appears at 10 K in the magnetic susceptibility curve, while with increasing magnetic field the peak of χ–T shifts to lower temperature, and at 4 T the magnetic susceptibility curve is smooth and monotonic without any peak observed in the measured temperature range. This means that the peak, if it exists, may appear in the temperature range lower than 4.5 K. In addition, magnetic field has hardly any effect on the magnetic susceptibility at $T > 15$ K but a large effect at $T < 15$ K. The phenomenon that magnetic field shifts the peak of χ–T indicates that the kink of the magnetization curve at 4.5 K is associated with this peak. Moreover, no evidence of phase transition has been found in the measurement of specific heat of the same sample, which confirms that the peak of χ–T has no relation to the long-range AF correlation. This is consistent with NMR experiments [3, 4].

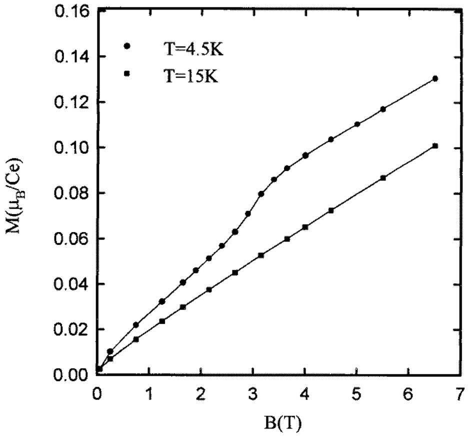

Figure 1. Magnetization against magnetic field.

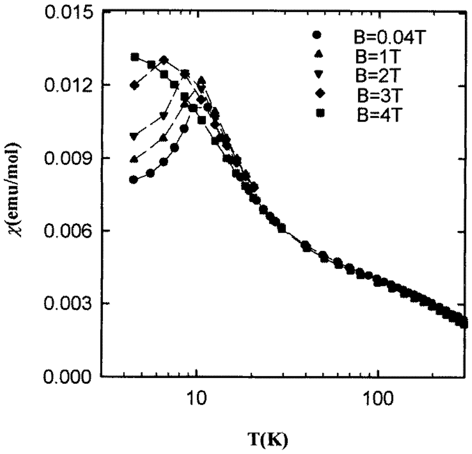

Figure 2. Magnetic susceptibility against temperature in different magnetic fields.

In the mean-field solution to the Anderson lattice, the hybridization gap is expected to close continuously with increasing magnetic field [19], so an appropriate magnetic field in which Zeeman splitting is comparable to the gap can smear the gap. In [17], the T and B dependences of C_m/T in magnetic fields were explained in terms of the Zeeman splitting of the pseudogapped DOS in the Kondo resonance band with the residual DOS at E_F. Moreover, the peak in magnetic susceptibility χ around 12 K was also imitated by using this model.

In order to explain the temperature dependence of magnetic susceptibility in different fields, here we calculated the χ–T curves in different magnetic fields by using the same model, although the model may not be practicable in magnetic fields higher than 14 T because all the parameters were obtained by the experimental results in magnetic fields less than 14 T. Neglecting the many-body effect, χ_{cal} was calculated by the following formulas [17]

$$\chi_{cal} = \frac{M}{H} = N_A \frac{g_J \mu_B |J_z|}{H} \int_{-\infty}^{\infty} \{N^-(E) - N^+(E)\} f(E,T)\,\mathrm{d}E$$

where N_A is the Avogadro number, $f(E,T)$ is the Fermi–Dirac distribution function, g_J is the Lande g-factor, J_z is the z-component of total angular momentum, μ_B is the Bohr magneton and $N^+(E)$ and $N^-(E)$ are the DOSs for the up- and down-spin bands respectively.

$$N^{\pm}(E) = \frac{A}{\pi}\frac{(D/2)}{(E - E_F \mp E_{Zeeman})^2 + (D/2)^2} \qquad \text{for } |E - E_F \mp E_{Zeeman}| \geqslant \Delta$$

$$N^{\pm}(E) = \frac{A}{\pi}\left\{\left(N_0 - \frac{(D/2)}{\Delta^2 + (D/2)^2}\right)\frac{|E - E_F \mp E_{Zeeman}| - W}{W - \Delta} + N_0\right\}$$
$$\text{for } W \leqslant |E - E_F \mp E_{Zeeman}| < \Delta$$

$$N^{\pm}(E) = \frac{A}{\pi}N_0 \qquad \text{for } |E - E_F \mp E_{Zeeman}| < W$$

where D, Δ and W are the half-widths of the Lorentzian, the V-shaped gap and the bottom in the gap respectively. N_0 is the magnitude of the residual DOS. In the calculation, the

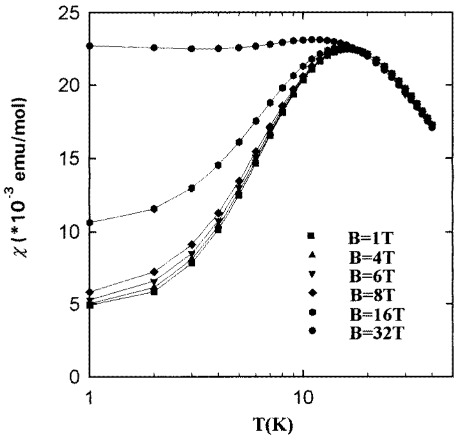

Figure 3. Calculated magnetic susceptibility against temperature in different fields.

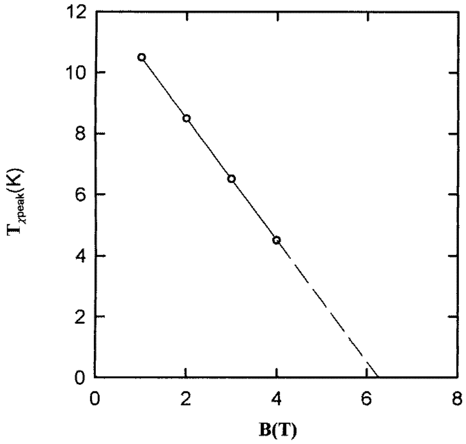

Figure 4. Peak temperature of magnetic susceptibility against magnetic field.

parameters $D = 58$ K, $\Delta = 21$ K, $W = 4.5$ K, $N_0 = 2.5 \times 10^{-3}$ K^{-1} and $g_J|J_z| = 1.1$ were adopted. The calculated results are shown in figure 3. Although the calculated χ_{cal} is field dependent, the peak of χ_{cal} is almost unshifted in different magnetic fields, i.e., the peak position of χ_{cal} is not sensitive to the magnetic fields. The contradiction between the calculated and the experimental results suggests that the peak cannot be simply explained just by the presence of the pseudogapped DOS with the effect of magnetic field attributed to the Zeeman splitting. It seems that in order to explain the experimental results the development of an antiferromagnetic correlation should be considered.

Figure 4 shows the magnetic field dependence of the $T_{\chi peak}$ at which the magnetic susceptibility peak appears. By extrapolating $T_{\chi peak}$–B to higher magnetic fields, it is found that about 6.3 T can shift the peak to 0 K although the tunnelling experiment shows that 6.3 T cannot suppress the pseudogap [20]. It is also possible that the shift of $T_{\chi peak}$ at higher magnetic fields does not behave linearly and the magnetic field corresponding to $T_{\chi peak} = 0$ K is larger than 6.3 T.

It is worth noting that the shift of the χ–T peak is consistent with the inelastic neutron scattering study of CeNiSn in high magnetic field, in which the peaks at the scattering vectors (0 1 0) and (0 0 1) shift continuously from 2 to 2.5 meV with increasing field [21]. However from the above model χ_{cal} gives a threshold value $H_T \approx 6$ T below which χ_{cal} remains unchanged, which is in agreement with the observation that there is little change of the spin excitations at 4 meV between 0 and 6 T. It seems that the pseudogap is related to 4 meV excitations while the shift of χ is related to 2 meV excitation.

The effect of short-range magnetic correlation in the non-magnetic phase of the Kondo lattice was studied in a mean-field approximation, which shows that antiferromagnetic correlations stabilize the non-magnetic phase [22]. It seems that spin excitation at 2 meV does not have a direct relation to the pseudogap, but it stabilizes the non-magnetic ground state in which the pseudogap is formed by strong hybridization of the f orbitals with a conduction band. This also suggests that the ground state is very near the 'weak' magnetic state. This is why the models without consideration of antiferromagnetic correlation also can give a reasonable explanation of some phenomena in magnetic field. However, refinement must consider magnetic correlations which are missing from the original model, especially the quasi-one-dimensional antiferromagnetic correlations at 4 meV.

4. Conclusions

To summarize, our measurements of χ–T of CeNiSn under different magnetic fields have brought new information concerning the ground state. The shift of magnetic susceptibility in magnetic field provides further evidence to confirm the existence of antiferromagnetic correlations. The result is consistent with the theoretical work.

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