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MAGNETIC BEHAVIOUR IN $CeNiGa_2$ AND Ce_2Ni_2Ga*

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The bulk properties of $CeNiGa_2$ and Ce_2Ni_2Ga were studied by means of magnetic susceptibility, magnetization and electrical resistivity measurements. It was found that $CeNiGa_2$ is an antiferromagnetically ordered dense Kondo system whereas Ce_2Ni_2Ga exhibits a fluctuating valence behaviour.

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Though the cerium ternary phases $CeNiGa_2$ and Ce_2Ni_2Ga are known since many years [1, 2], their magnetic behaviour has been studied in the past only down to 78 K [1, 3]. This paper describes for the first time the low-temperature magnetic properties of both materials.

Polycrystalline samples of $LaNiGa_2$, $CeNiGa_2$ and Ce_2Ni_2Ga were prepared by arc-melting the constituent elements in an argon atmosphere and subsequent annealing in vacuum at 650°C for two weeks. The ingots were examined by X-ray powder diffraction and found to be single phases. Magnetic measurements were carried out in the temperature interval 1.7–300 K (4.2–800 K for Ce_2Ni_2Ga) and magnetic fields up to 50 kOe using a Quantum Design MPMS-5 SQUID magnetometer. The electrical resistivity was measured in the range 4.5–300 K by conventional dc four-probe technique.

In Fig. 1 the temperature variation of the magnetic susceptibility of $LaNiGa_2$ and $CeNiGa_2$ is shown. As the La-based compound is a Pauli paramagnet, the magnetism in $CeNiGa_2$ is apparently due to the cerium atoms only. Above 100 K the susceptibility of this gallide can be well described by the Curie–Weiss law with the effective magnetic moment of $2.55\mu_B$ and the paramagnetic Curie temperature of 44 K. The pronounced deviation of $\chi(T)$ from this law, observed at lower temperatures, probably originates from crystal field interactions. The experimental value

*Dedicated to Professor Wojciech Suski on the occasion of his 60th birthday.

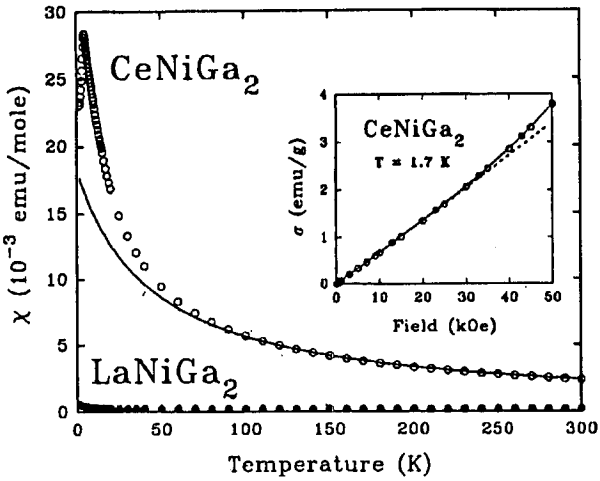

Fig. 1. Temperature dependence of the molar magnetic susceptibility for $LaNiGa_2$ and $CeNiGa_2$. The solid line is a fit of $\chi(T)$ of $CeNiGa_2$ to the Curie–Weiss law. The inset shows the field variation of the magnetization for $CeNiGa_2$, measured at 1.7 K with increasing (filled symbols) and decreasing (open symbols) magnetic field. The dashed line marks a straight-line behaviour of $\sigma(T)$ below about 30 kOe.

of $\mu_{\rm eff}$ is almost equal to that predicted by the Hund rules for a $4f^1$ configuration which indicates the presence in $CeNiGa_2$ of stable Ce^{3+} ions. The negative value of θ_p suggests the occurrence of some antiferromagnetic exchange interactions between the cerium atoms but its rather high magnitude may also be due to the Kondo-like effects.

Both a sharp maximum in $\chi(T)$ and a metamagnetic-like anomaly in $\sigma(H)$ (see the inset to Fig. 1) indicate that $CeNiGa_2$ orders antiferromagnetically at 4.4 K. In $H = 50$ kOe the cerium magnetic moment at 1.7 K reaches only $0.23\mu_{\rm B}$. This strong reduction in the moment may be a combined effect of crystal field and Kondo-like screening interactions.

The temperature variations of the electrical resistivity of $LaNiGa_2$ and $CeNiGa_2$ are presented in Fig. 2. Whereas the former compound behaves as a metallic conductor in the whole temperature range studied, the Ce-based gallide exhibits some characteristic anomalies in its $\rho(T)$. In order to estimate the magnetic scattering resistivity for this compound it was assumed that the phonon contribution to its total measured resistivity is similar to that contribution for $LaNiGa_2$, i.e. $\rho(CeNiGa_2) = \rho_0 + \rho_m + [\rho(LaNiGa_2) - \rho_0(LaNiGa_2)]$. The so-derived temperature variation of $\rho_0 + \rho_m$ is shown in the inset to Fig. 2. It appears that for $T > 90$ K the magnetic resistivity of $CeNiGa_2$ changes with temperature in a Kondo-like manner with the Kondo coefficient $c_{\rm K} = -22.8$ $\mu\Omega$ cm. The pronounced maximum in $\rho_{\rm m}(T)$, centered at 18 K, probably arises due to an interplay of Kondo effect and crystal field interactions, while a kink at 4.4 K clearly manifests the antiferromagnetic phase transition.

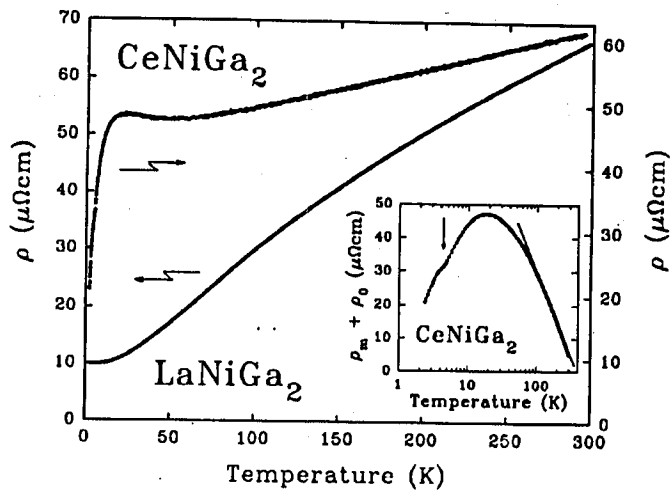

Fig. 2. Temperature variation of the electrical resistivity for $LaNiGa_2$ and $CeNiGa_2$. The inset presents in a semilogarithmic scale the temperature dependent magnetic contribution to the total electrical resistivity of $CeNiGa_2$ (enlarged by the residual resistivity). The solid line is a fit of the magnetic resistivity to the Kondo formula. The arrow marks the Néel temperature.

In Fig. 3 the temperature dependence of the inverse magnetic susceptibility of Ce_2Ni_2Ga is shown. In agreement with previous results [3] the susceptibility of this gallide is small and only weakly temperature dependent. Around $T_{\min} = 200$ K the $\chi^{-1}(T)$ curve goes through a broad minimum, characteristic of intermediate valence (IV) systems [4]. A small tail in $\chi^{-1}(T)$, observed below 50 K, presumably results from the presence in the sample of some stable Ce^{3+} ions. To account for this impurity contribution the procedure proposed in Ref. [5] has been applied. As a result of this analysis, the impurity concentration of about 9.7×10^{-4} Ce^{3+} ions/mole, the intrinsic susceptibility $\chi(0) = 1.44\times10^{-3}$ emu/mole Ce-atom and the corrected $\chi^{-1}(T)$ variation were derived.

As seen from Fig. 3, below 100 K the corrected susceptibility of Ce_2Ni_2Ga varies with temperature in a manner predicted by Béal-Monod and Lawrence within the paramagnon model of IV materials [5]

$$\chi(T) = \chi(0)\left[1 + \left(\frac{T}{T_{\rm sf}}\right)^2\right],$$

where the spin fluctuation temperature $T_{\rm sf} = 270$ K was found from the relation $T_{\rm sf} = C/2\chi(0)$ ($C = 0.807$ emu/(mole K) is the Curie constant of free Ce^{3+} ions). It is worth to note that the above value of $T_{\rm sf}$ is quite consistent with the estimate [4] $T_{\rm sf} = \frac{3}{2}T_{\min} = 300$ K.

Another approach describing the behaviour of IV systems is the interconfiguration fluctuation (ICF) model developed by Sales and Wohlleben [6]. According to this theory the magnetic susceptibility of a cerium compound with the $4f^0$

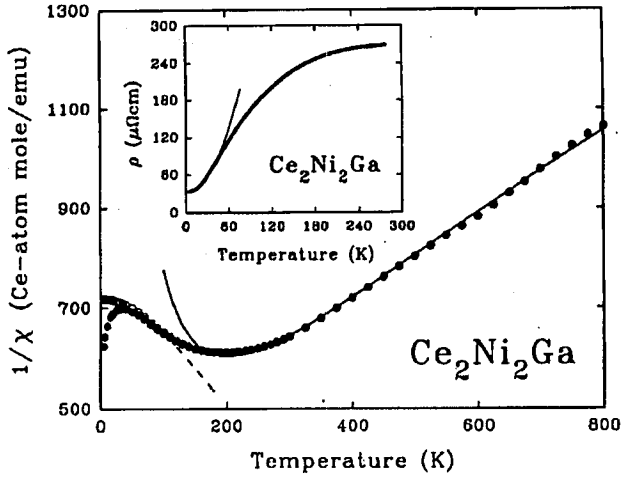

Fig. 3. Temperature dependence of the inverse molar magnetic susceptibility for Ce_2Ni_2Ga. The filled and open symbols represent the measured and corrected data (see text), respectively. The dashed and solid lines are fits of $\chi^{-1}(T)$ according to the paramagnon and ICF models, respectively. The inset shows the temperature variation of the electrical resistivity for Ce_2Ni_2Ga. The solid line marks a T^2-variation of the resistivity below 50 K.

ground state may be expressed as

$$\chi(T) = \frac{0.806\,[1-\vartheta(T)]}{T+T_{sf}^{*}} + \chi_0,$$

where the mean occupation of the ground state (the effective valence) $\vartheta(T)$ is given by

$$\vartheta(T) = \frac{1}{1+6\exp\left[-E_{ex}/\left(T+T_{sf}^{*}\right)\right]},$$

χ_0 stands for the sum of temperature independent conduction-electron paramagnetic, core-electron diamagnetic and Van Vleck contributions, while E_{ex} is the energy difference between the $4f^1$ and $4f^0$ states. From Fig. 3 it is clear that in the case of Ce_2Ni_2Ga the ICF model works well for $T > 150$ K. The fitting parameters are as follows: $E_{ex} = 509$ K, $T_{sf}^{*} = 66$ K and $\chi_0 = 2.3 \times 10^{-4}$ emu/mole Ce-atom, and the effective cerium valence is found to increase gradually from 3.07 at 800 K to 3.19 at 150 K. It is worthwhile mentioning at this place that a similar change in the cerium valence has recently been determined for Ce_2Ni_2Ga by means of L_{III}-absorption edge measurements [7].

Further evidence for a fluctuating valence behaviour in Ce_2Ni_2Ga comes from the temperature variation of its electrical resistivity, displayed in the inset to Fig. 3. The overall shape of this $\rho(T)$ is reminiscent of that observed for many IV systems [8]. In particular, below 50 K the resistivity may be well described by the function $\rho(T) = \rho_0 + AT^2$ (with $\rho_0 = 32.6$ $\mu\Omega$ cm and $A = 0.029$ $\mu\Omega$ cm/K^2) which is characteristic of a Fermi liquid [4].

In summary, $CeNiGa_2$ and Ce_2Ni_2Ga have been found to exhibit quite different magnetic properties. Whereas the first compound shows features of a well localised magnetism due to the presence of stable Ce^{3+} ions, the latter one has a nonmagnetic ground state with the noninteger cerium valence. The magnetic behaviour in both ternaries is presumably mainly governed by the hybridization of the Ce $4f$-states with the $3d$-states of neighbouring Ni-atoms and these interactions are apparently much stronger in Ce_2Ni_2Ga than in $CeNiGa_2$. It seems that the observed features may be due to a different number of Ce-atom nearest neighbours which is six and four nickel atoms for the former and the latter compound, respectively.

Acknowledgments

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