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Electronic properties of CeNiX compounds

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The electronic properties of compounds of the type CeNiX with X = Al, Ga, Si and Ge, crystallizing in various crystal structures, have been studied. All compounds are found to possess the paramagnetic ground state. The intrinsic magnetic properties, which are masked by trivalent Ce impurities, were derived by combining measurements of the magnetic susceptibility and of the magnetization in fields up to 35 T. Also, the low-temperature specific heat in fields up to 5 T is presented.

Within the framework of a research program on the electronic properties of equiatomic ternary intermetallic compounds of the type CeTX with T a transition metal and X an element from the p-block in the periodic table, we have investigated CeNiX compounds with X = Al, In, Si and Ge. The purpose of the program is to clarify the influence of the constituting T and X elements on the electronic properties and the ground states of the Ce ions and to investigate the role of hybridization of the 4f states with the s, p and d states introduced by the T and X atoms.

CeTX compounds with X = Al, In, Si and Ge, were prepared by arc-melting stoichiometric amounts of the pure elements. All samples were annealed at 800 °C for one to two weeks. By X-ray diffraction, the crystal structures (ZrNiAl-structure for X = Al, In; LaPtSi-structure for X = Si; TiNiSi-structure for X = Ge) and the lattice parameters were found to agree with literature data [1–5]. By microprobe analysis, the matrix composition and the composition and concentration of impurity phases were determined. The following main impurity phases were identified: about 2% material with composition $CeNi_2Al_2$ in CeNiAl, 6% material with composition $CeNi_4In$ in CeNiIn, and a few percent of Ce_2O_3 and material with composition $Ce_3Ni_4Si_4$ in CeNiSi. In CeNiGe, no second phase was detected. In this compound, however, fluctuations of a few percent in the Ni concentration were found, which were absent in the other compounds.

Fig. 1 shows the temperature dependence of the magnetic susceptibility for all CeNiX compounds between 1.6 and 300 K. Although, at low temperatures, the temperature dependence is considerable in all cases, no indications of magnetic transitions are found. The absence of magnetic order is confirmed by the specific-heat results between 1.3 and 30 K (fig. 2). Apart from the small anomaly at about 6 K, present in all samples except CeNiAl, the curves do not show any sign of magnetic order. The anomaly observed at 6 K is likely due to small amounts (about 1%) of Ce_2O_3, that orders magnetically at 5.8 K. For all compounds the specific heat is not significantly influenced by a magnetic field of 5 T.

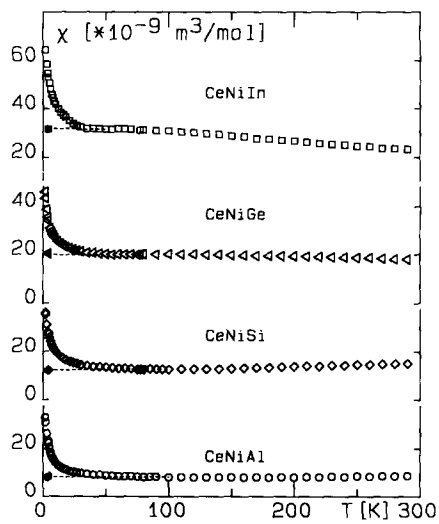

Fig. 1. As-measured temperature dependence of the magnetic susceptibility of CeNiX compounds with X = Al, In, Si and Ge. The filled symbols correspond to the intrinsic susceptibility values at 4.2 K obtained in high-field magnetization measurements (see fig. 3). The dashed lines represent the estimated temperature dependence of the intrinsic susceptibility.

From these results it can be concluded that all compounds have a paramagnetic ground state. At low temperatures, however, Ce^{3+} ions present in impurity phases like Ce_2O_3, strongly contribute to the susceptibility, thereby masking the intrinsic susceptibility of the compounds. In order to determine the intrinsic low-temperature susceptibility of the compounds, we have performed magnetization measurements in high fields, in which the Ce^{3+} magnetic moments saturate, so that the remaining high-field susceptibility can be considered to represent the intrinsic susceptibility. The re-

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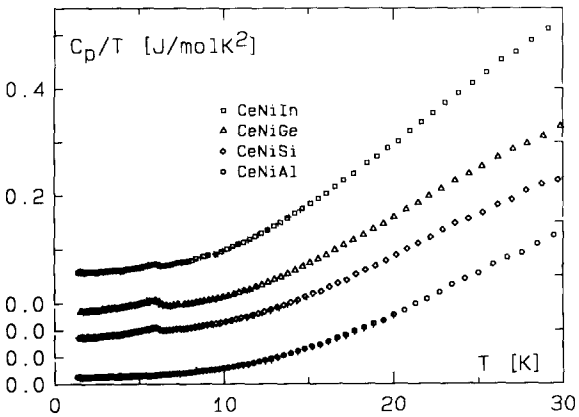

Fig. 2. Specific heat of CeNiAl, CeNiIn, CeNiSi and CeNiGe, presented as C_p/T vs. T. The +signs correspond to data taken in an applied magnetic field of 5 T.

sults of these measurements, carried out at 4.2 K in semicontinuous fields up to 35 T in the High Magnetic Field Installation at the University of Amsterdam, are presented in fig. 3. The dotted lines in fig. 3 correspond to the intrinsic susceptibilities of the compounds and are represented in fig. 1 by the filled symbols. The dotted lines in fig. 1 indicate the estimated temperature dependences of the intrinsic susceptibilities which suggest the development of (rather flat) maxima, indicative for the intermediate-valence state of Ce. The

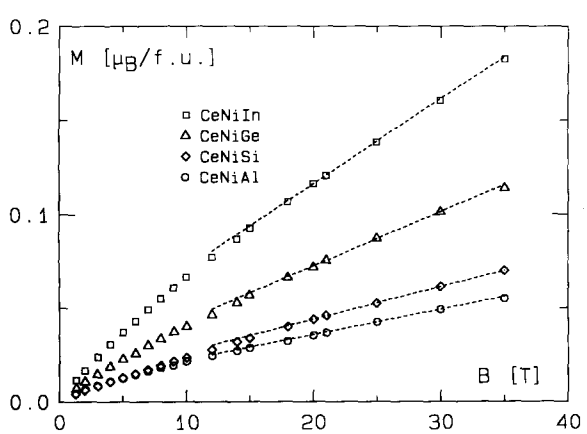

Fig. 3. Magnetization vs. applied magnetic field at 4.2 K of CeNiX compounds with X = Al, In, Si and Ge. The intrinsic susceptibility values represented by the full symbols in fig. 1, correspond to the straight lines drawn.

maxima develop at lower temperatures, the higher the value of the low-temperature susceptibility, which amounts for the compounds with X = Al, Si (see also ref. [5]), Ge and In (see also ref. [6]) to 9×10^{-9}, 12×10^{-9}, 20×10^{-9} and 31×10^{-9} m^3/mol, respectively. The coefficients γ of the electronic specific heat, giving direct evidence of the presence of 4f states at the Fermi level, show a similar dependence on X. They amount to 13, 22, 32 and 54 mJ/K^2mol for X = Al, Si, Ge and In (see also ref. [7]), respectively.

In order to obtain information on the magnetocrystalline anisotropy in these compounds, we have carried out magnetization measurements on two types of samples: (a) single-crystalline powder particles, free to rotate in the sample holder, so that the powder particles can be oriented by the applied field, and (b) powder particles fixed in a random orientation by frozen alcohol, thus simulating an ideal polycrystalline sample. For all compounds, no significant difference was observed between the results of these two types of measurements, which suggests that the anisotropy is small. However, since the magnetic moments involved are small, even in 35 T, an alternative explanation may be a poor orientation of the particles due to the weak magnetic forces.

In conclusion, the intrinsic low-temperature susceptibilities of CeNiX compounds with X = Al, In, Ge, Si, all having a paramagnetic ground state, have been derived by combining susceptibility and high-field magnetization measurements. In these compounds, a gradual development of the intermediate-valence state is observed in the sequence X = Al, Si, Ge, In.

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