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Novel Ce magnetism in CeDipnictide and Di-Ce pnictide structures

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Results of electrical resistance and magnetic susceptibility measurements on Ce_2Bi , Ce_2Sb , $CeScGe$, $CeScSi$ and $CeSb_2$ are presented. Ce_2Bi and Ce_2Sb have antiferromagnetic transitions at low temperatures, while $CeSb_2$, $CeScGe$ and $CeScSi$ have ferromagnetic transitions, $CeScGe$ having a $T_c = 46$ K. The data are analyzed with respect to the similarities of the two crystal structure groups that these materials fall into: $CeSb_2$ having the $LaSb_2$ structure and the other materials all having the La_2Sb structure.

Single crystals of Ce_2Sb and polycrystalline Ce_2Bi have been studied by Isobe *et al.*¹ Both compounds form in the tetragonal La_2Sb structure which has two inequivalent La sites: La1 and La2. For Ce atoms on the La1 sites, the Ce-Ce distance is substantially shorter than in metallic Ce. One of the primary conclusions drawn by Isobe *et al.* is that these two inequivalent Ce sites display significantly different magnetic behavior, with the Ce1 (La1) sites exhibiting a strongly mixed valence-type susceptibility and the Ce2 (La2) sites acting as local moments. In this paper, we examine in greater detail Ce_2Sb and Ce_2Bi , and compare results to those obtained on related compounds $CeScGe$, $CeScSi$ and $CeSb_2$. $CeScGe$ and $CeScSi$ both have the La_2Sb structure with only one of the unique Ce sites, Ce2, occupied; whereas, $CeSb_2$ has the orthorhombic $LaSb_2$ structure which is closely related to the La_2Sb structure.^{2,3} By examining these five compounds, the occupancy of the Ce sites can be varied systematically, permitting a study of occupancy on the magnetism of these systems.

Single crystals of $CeSb_2$ were grown out of Sb flux; whereas, single crystals of Ce_2Bi and Ce_2Sb were grown out of Ce flux. The $CeScGe$ and $CeScSi$ materials were arc-melted on a water-cooled hearth under a protective atmosphere of Ar. The electrical resistance was measured using a standard four-probe lock-in technique, with current flowing in the a - b plane of the crystals. Magnetic susceptibility measurements were performed with a Quantum Design superconducting quantum interference device (SQUID) magnetometer, in the case of single crystals with the field applied parallel or perpendicular to the c axis.

The La_2Sb and $LaSb_2$ structures are well known.^{2,3} Briefly, the La_2Sb structure can be thought of as consisting of two predominant features. The first feature is that the La1, 4(c) sites, form two-dimensional (2-D) sheets of La atoms. The La2 and the Sb occupy 4(e) sites which form a slab that separates the sheets of La1. This slab consists of interpenetrating La and Sb triangular prisms (see Fig. 1 of Ref. 2). In $CeScGe$ and $CeScSi$ (Ref. 4) only the La2 site is occupied by Ce. This means that there are 2-D sheets of Sc on the La1 site and Ce/Si or Ce/Ge slabs separating these sheets. The $LaSb_2$ structure is in many aspects similar to the La_2Sb structure, particularly as seen in the cases of $CeScGe$ and $CeScSi$. In the $LaSb_2$ structure there are two unique Sb sites and one unique La site; the converse of the La_2Sb structure. One of the Sb sites forms 2-D planes (sim-

ilar to the planes formed by the La1 sites in the La_2Sb structure), while the other Sb site and the La site form a slab of triangular prisms that separate the 2-D Sb planes (see Fig. 1 of Ref. 3). The details of the La/Sb slabs in the $LaSb_2$ and La_2Sb structures are somewhat different, but to first order, they are qualitatively the same.

Figure 1 shows low-temperature magnetic susceptibility and electrical resistance for Ce_2Bi . There is an antiferromagnetic phase transition at $T_N = 11$ K that manifests itself in both the resistance and susceptibility for the field applied parallel to the c axis. The large anisotropy seen in the magnetic susceptibility of Ce_2Bi is in agreement with the anisotropic susceptibility that Isobe *et al.* argue for and we have found in Ce_2Sb as well (data not shown). Figure 2 shows the low-temperature electrical resistance for $CeScSi$ and $CeScGe$. The anomalies at 26 and 46 K for $CeScSi$ and $CeScGe$, respectively, are associated with ferromagnetic phase transitions, confirmed by the presence of substantial hysteresis in magnetization versus field measurements below T_c . These are unusually high ferromagnetic transitions for intermetallic Ce compounds, and specifically make $CeScGe$ of interest on its own right. Figures 3 and 4 show the electrical resistance and anisotropic magnetic susceptibility of $CeSb_2$. The resistive anomaly at 15 K is associated with a ferromagnetic transition that results in hysteresis loops similar to those found for $CeScGe$ and $CeScSi$.

Although all compounds measured manifest virtually full Ce moments, one striking difference between the materials that have two unique Ce sites and the materials that only have one is that the Ce_2Sb and Ce_2Bi have antiferromagnetic transitions, whereas $CeScSi$, $CeScGe$ and $CeSb_2$ have ferromagnetic transitions. This supports our discussion of the La_2Sb and $LaSb_2$ structures: that for both $CeScGe/Si$ and $CeSb_2$ the environment of the Ce site is similar. By removing the Ce from the tightly packed, 2-D planes, ferromagnetism is found for both the La_2Sb and $LaSb_2$ structures. Apparently, the absence of the 2-D planes of Ce atoms at the La1 sites favors the formation of a ferromagnetic groundstate from the Ce atoms on the La2 sites. This is suggested as well from susceptibility measurements on Ce_2Bi . Although Ce_2Bi orders antiferromagnetically at 11 K, strong ferromagnetic correlations are present at low temperatures. For temperatures below approximately 100 K, the inverse susceptibility is nearly linear in temperature and extrapolates to a positive $\theta_p = 9$ K. (See

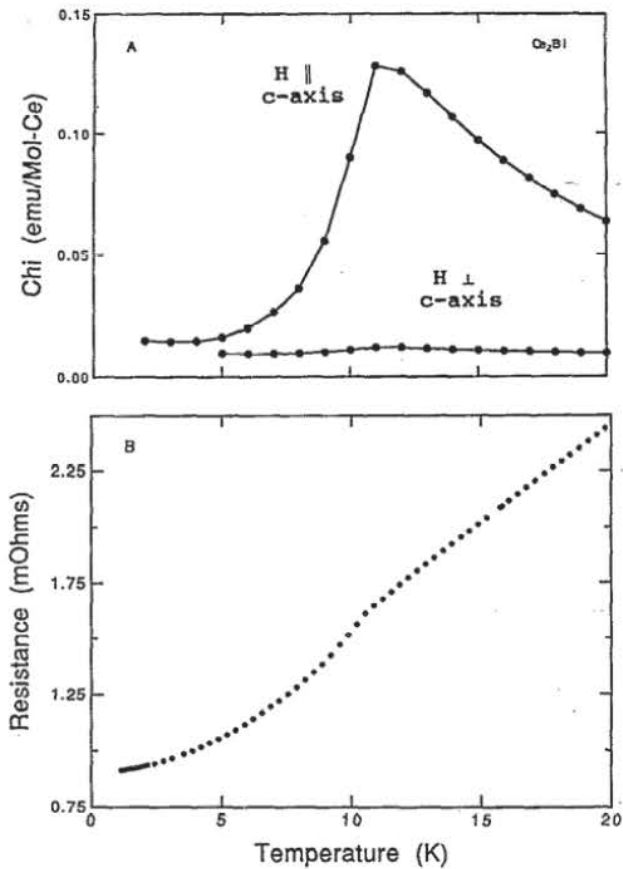


FIG. 1. Low-temperature data for Ce_2Bi . (a) Magnetic susceptibility for a field of 0.1 T applied parallel and perpendicular to c axis. (b) Electrical resistance with current running in a - b plane.

Fig. 5.) This is comparable to the $\theta_p = 26$ K found for ferromagnetic CeSb_2 . The inset of Fig. 5 clearly shows the development of ferromagnetic correlations at low temperatures. Further, we note that both CeSb_2 and Ce_2Bi have magnetic susceptibilities of comparable magnitudes at low temperature (compare Figs. 1 and 3). Thus, it seems that

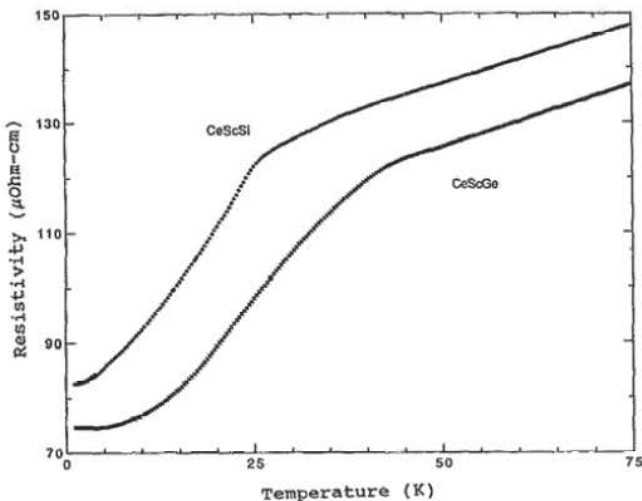


FIG. 2. Low-temperature resistivity vs temperature for CeScGe and CeScSi polycrystalline samples. Features in the resistivity mark the onset of ferromagnetic order found in magnetization experiments.

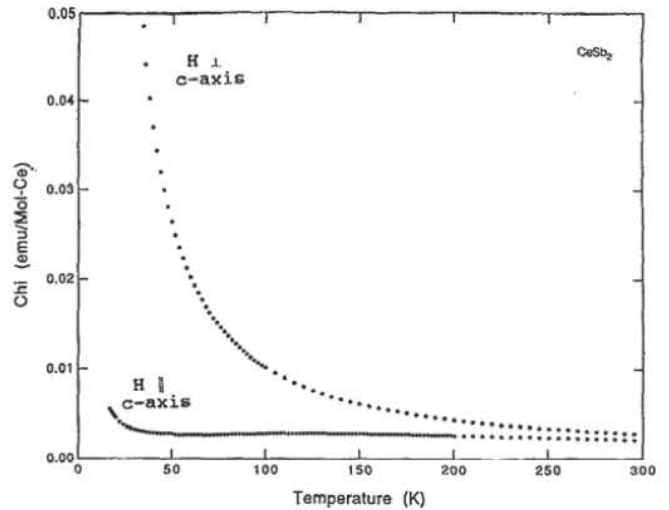


FIG. 3. Magnetic susceptibility vs temperature of CeSb_2 for a field of 0.1 T applied parallel and perpendicular to c axis.

Ce_2Bi has a strong tendency to be ferromagnetic that is inhibited by the existence of 2-D planes formed by Ce occupancy of the La1 sites.

In addition to these systematics, there are a few puzzling questions raised by the data. First, there is the question of the remarkable anisotropy seen in CeSb_2 crystals. If the field is applied perpendicular to the c axis, the system shows a clear local momentlike behavior with a positive θ_p that is consistent with a ferromagnetic transition at 15 K. On the other hand, if the field is applied parallel to the c axis, the high-temperature $\theta_p = -175$. A similarly large anisotropy has been found in Ce_5Sn_3 (Ref. 5) for which it has been argued that crystal-field effects are important. We suggest this to be the case for both CeSb_2 and Ce_2Sb . Another aspect of the magnetic anisotropies of CeSb_2 and Ce_2Bi (Figs. 1 and 4) is that the easy magnetic directions

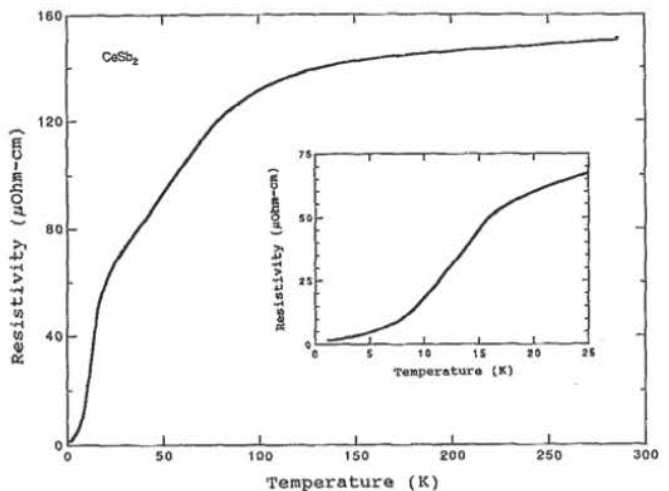


FIG. 4. Electrical resistivity vs temperature of CeSb_2 with current running in a - b plane. Inset: Low-temperature resistivity showing an anomaly in the resistance due to the onset of ferromagnetism. The nature of the slight anomaly seen at 12 K is unknown.

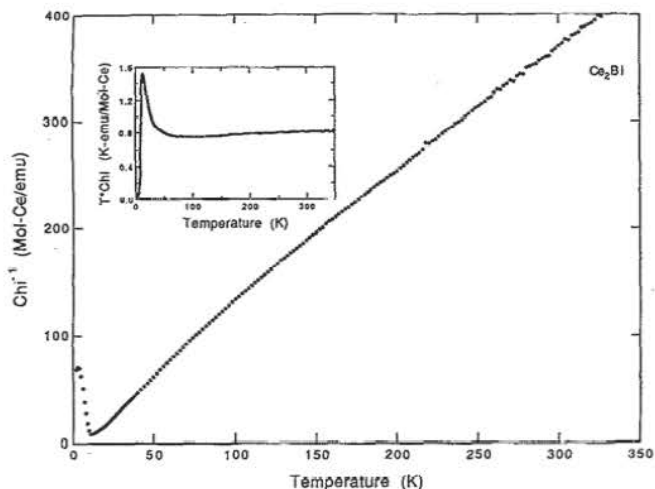


FIG. 5. Inverse magnetic susceptibility vs temperature of Ce_2Bi . Inset: Temperature times magnetic susceptibility vs temperature of Ce_2Bi . The rise in $T \cdot \chi$ below 100 K is due to the development of ferromagnetic correlations.

are orthogonal, i.e., perpendicular and parallel to the c axis for CeSb_2 and Ce_2Bi , respectively. In addition to the presence of planar Ce atoms at the La1 sites in Ce_2Bi , it is possible that this difference is a manifestation of the slightly different crystal structures of these two materials. Large magnetic anisotropies are found for both crystal

structures, implying that the La2 sites and crystal field anisotropy at these sites dominate this behavior. Finally, we note the temperature dependent resistivity of CeSb_2 (Fig. 3) which is similar to that observed in heavy-electron/Kondo-lattice compounds. In these cases, antiferromagnetic exchange is important. We suggest that this may also be the case in CeSb_2 , but that band structure and the momentum dependence of the exchange work to promote a ferromagnetic ground state.

In summary, we have studied the systematics of Ce members of the La_2Sb and LaSb_2 systems. We find that the La2 sites can be associated with a proclivity to form a ferromagnetic ground state at low temperatures and that the La1 sites, if occupied by Ce, can effectively thwart this predisposition.

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