

Pressure-dependent studies of CeSbNi_x ($0 \leq x \leq 0.35$)

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We studied the effect of Ni incorporation on the magnetic properties of CeSbNi_x ($0 \leq x \leq 0.35$) using resistivity, susceptibility, and magnetization measurements under high pressure and magnetic field. With increasing pressure antiferromagnetic transition temperature (T_N) of CeSb increases continuously leading to a high pressure phase. Similarly, T_N of CeSbNi_x increases with pressure for $x \leq 0.15$ while increasing pressure weakens a Cr-like anomaly found in the resistivity of $\text{CeSbNi}_{0.08}$. Interestingly, resistivity of $\text{CeSbNi}_{0.08}$ under higher pressure becomes very similar to that of CeSb at ambient pressure. Magnetic hysteresis, metamagnetic transition, and saturation behavior, which are absent in the ambient-pressure magnetization of $\text{CeSbNi}_{0.05}$ and $\text{CeSbNi}_{0.08}$, begin to appear with pressure too. On the other hand, pressure effects in both resistivity and magnetization of $\text{CeSbNi}_{0.15}$ and $\text{CeSbNi}_{0.35}$ are different from those of $\text{CeSbNi}_{0.05}$ and $\text{CeSbNi}_{0.08}$. We discuss these findings with regard to possible Ni-induced changes in the nature of the magnetic interactions of CeSb.

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I. INTRODUCTION

Cerium mononictides CeX ($X = \text{P, As, Sb, and Bi}$) having a very low carrier concentration show various interesting magnetic properties.¹⁻³ Although crystal structure of CeSb is cubic and isotropic with a modest Néel temperature of 16 K, its magnetic properties are very anisotropic in the ordered phase with an easy axis along [001] direction.^{4,5} Estimated anisotropy field is substantially larger than 100 T. Moreover, in spite of the relatively simple NaCl-type structure the magnetic phase diagram of CeSb is very complicated: altogether no fewer than fifteen different magnetic phases are found in the (H, T) phase diagram.^{6,7} For example, at 16 K there is a first-order transition to an antiferromagnetic (AFP) phase with the periodicity of three lattice spacings along the c axis: which is then followed by successive five new AFP transitions with slightly different periodicities. Below 8 K is another AF-IA state found, and it remains unchanged to the lowest measured temperature. With increasing magnetic field, CeSb moves from these AFP phases to new ferro-para phases while the AF-IA state changes to an antiferro-ferro phase before entering into a ferromagnetic state above 4 T. Interestingly enough, most these phases are also found in the (P, T) phase diagram; of which origin will be discussed shortly. It is noticeable too that a critical end point exists at 2.5 kbar: at which the first-order AFP transition meets another transition of second-order type, and for higher pressure a new transition of AF-I type is found. At 10 kbar there is a horizontal phase boundary between the AFP and AF phases. These complex magnetic phases found in CeSb have been qualitatively explained by the so-called anisotropic p - f mixing model.⁸ Apart from the p - f mixing effect, two other effects are also known to be equally important in determining the magnetic states of CeSb. One is a crystal field splitting

energy Δ_{CEF} between the Γ_7 ground state and the Γ_8 excited state of Ce^{3+} ion, and the other is the hole concentration of Sb 5 p band. While magnetic field acts on the crystal field levels and reduces Δ_{CEF} , pressure increases the p - f mixing strength. Therefore, despite the different mechanisms of magnetic field and pressure, their overall effects are more or less the same.

Recently Ni atoms were successfully incorporated into the (1/4, 1/4, 1/4) interstitial site of CeSb, while keeping the cubic structure intact up to the solubility limit of $x = 0.4$ in the formula of CeSbNi_x .⁹⁻¹¹ Both the absence of strong magnetic anisotropy and an increased crystal-field splitting as seen in inelastic neutron scattering experiments for $\text{CeSbNi}_{0.15}$ have been attributed to the collapse of p - f mixing due to Ni incorporation.^{9,10} Recent transport property measurements¹¹ of CeSbNi_x pointed out too that the decrease of Sb 5 p hole concentration due to Ni incorporation might as well be important to understanding the observed magnetic properties of CeSbNi_x . Furthermore, we found a sharp increase just below T_N in the resistivity for $x \geq 0.08$ that arises from the formation of a superzone gap as the type-I antiferromagnetic state is stabilized at low temperatures. It is also interesting to note that the unit cell volume increases with increasing Ni concentration, thus giving rise to a negative chemical pressure effect.

To our best knowledge, it is not quite clear at the moment which one of the two effects of Ni incorporation: one is the negative chemical pressure effect and another the decrease of the Sb 5 p hole concentration, is more important in determining the magnetic ground state of CeSbNi_x . At this particular point, our pressure-dependent studies of CeSbNi_x is useful, and thus help us understand how the magnetic properties of CeSbNi_x evolve with x and, more importantly, how Ni incor-

poration affects the p - f mixing in CeSb. In this study, we investigated the effects of both Ni incorporation and pressure on the magnetic properties of CeSbNi $_x$ ($x=0, 0.05, 0.08, 0.15, \text{ and } 0.35$).

II. EXPERIMENTAL DETAILS

Polycrystalline samples of CeSbNi $_x$ ($x=0, 0.02, 0.05, 0.08, 0.15, \text{ and } 0.35$) were prepared by arc melting stoichiometric amounts of the constituent elements on a water-cooled copper hearth under a high-purity argon atmosphere. The composition and crystal structure of the samples were determined by electron probe microanalysis and x-ray powder diffraction. Cubic lattice parameter (a) increases almost linearly at a rate $da/dx=0.085$ Å with x . Details of the sample preparation and characterization were described in Ref. 9.

Resistivity measurements were made on bar-shaped samples using a standard four-probe dc method at the Korea Basic Science Institute. We used a Be-Cu pressure cell, which is capable of producing pressures up to 9 kbar, with distilled petroleum as a pressure transmitting medium. Absolute pressure values were calibrated against the pressure-dependent T_c of Pb.¹² All our measurements were made in the temperature range of 2 to 300 K while warming.

We also measured magnetic susceptibility and magnetization measurements under pressures up to 12 kbar and magnetic field up to 12 T at the Institute of Solid State Physics, University of Tokyo. We used a pressure cell made of a high-purity TiCu alloy with fluorinert as a liquid pressure medium. Susceptibility measurements were made in a magnetic field of 5 kOe after zero-field cooling in the temperature range of 2 to 60 K. We also measured magnetization measurements at 4 K up to 12 T after zero-field cooling.

III. RESULTS

A. Effects of Ni incorporation at ambient pressure

In order to help one better understand our results of pressure-dependent measurements, we summarize below the effects of Ni incorporation observed at ambient pressure. Temperature dependence of the susceptibility and resistivity of CeSbNi $_x$ shows a nonlinear variation of T_N with varying x : T_N decreases rapidly with x reaching a minimum at 8.5 K for $x=0.08$ and then increases slowly up to $x=0.4$ (see Fig. 1). This x dependence of T_N is in good agreement with previous studies.^{9,11} At the same time increasing x weakens metamagnetic behavior and smears out a hysteresis found in the magnetization data of CeSb. Furthermore no magnetic anisotropy was observed in the magnetization of CeSbNi $_{0.15}$.

What is also noticeable is that $\rho(T)$ begins to increase rapidly below T_N for $x \geq 0.08$ (see Fig. 1). This $\rho(T)$ anomaly is suppressed upon applying magnetic field larger than a value at which a metamagnetic transition is observed. The rapid rise in $\rho(T)$ coincides with a strong increase in $|R_H(T)|$ obtained from Hall coefficient measurements.¹¹ These observations can be ascribed to the formation of AF superzone gap as magnetic structure of CeSbNi $_x$ changes to an AF-I type for $x \geq 0.08$; elastic neutron scattering studies found that CeSbNi $_{0.15}$ has a simple type-I AF structure.¹⁰

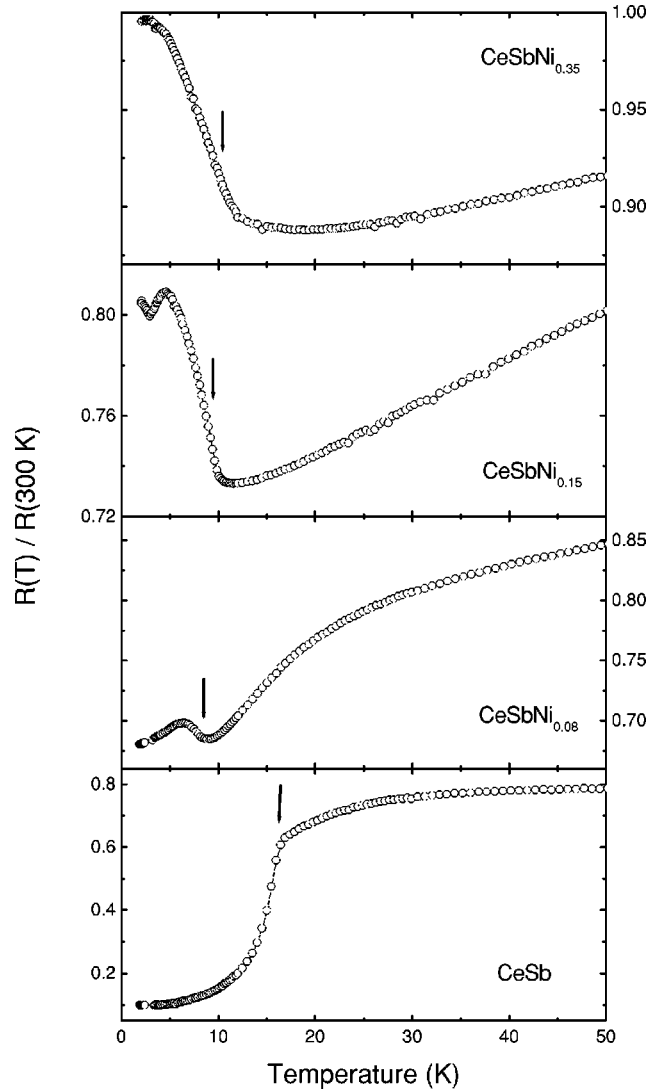


FIG. 1. Resistivity data taken at ambient pressure for several Ni compositions. T_N are marked by arrows in the figure.

B. Pressure-dependent resistivity

Figures 2 and 3 show temperature- and pressure-dependent resistivity of CeSbNi $_x$ ($x=0, 0.08, 0.15, \text{ and } 0.35$) up to 9 kbar. We normalized the resistivity with respect to the room-temperature resistivity values at ambient pressure for the sake of presentation. Arrows indicate antiferromagnetic transition temperatures T_{N^*} of AFP type and T_N of AF-I type as determined from pressure-dependent susceptibility measurements (see Fig. 4). For CeSb does a sudden drop in the resistivity coincide with a sharp peak at T_{N^*} in the ambient-pressure susceptibility. With increasing pressure, T_{N^*} increases continuously and, at the same time, a strong resistivity peak develops in good agreement with published results.¹³ At pressure higher than 2.5 kbar, an AF-I high pressure phase appears in CeSb as observed in high-pressure neutron scattering experiments.¹⁴ Transition temperature of this AF-I phase was marked by T_N in Fig. 2.

$\rho(T)$ of CeSbNi $_{0.08}$ at ambient pressure shows a minimum around $T_N=8.5$ K followed by a Cr-like anomaly at lower temperature; which was previously attributed to formation of

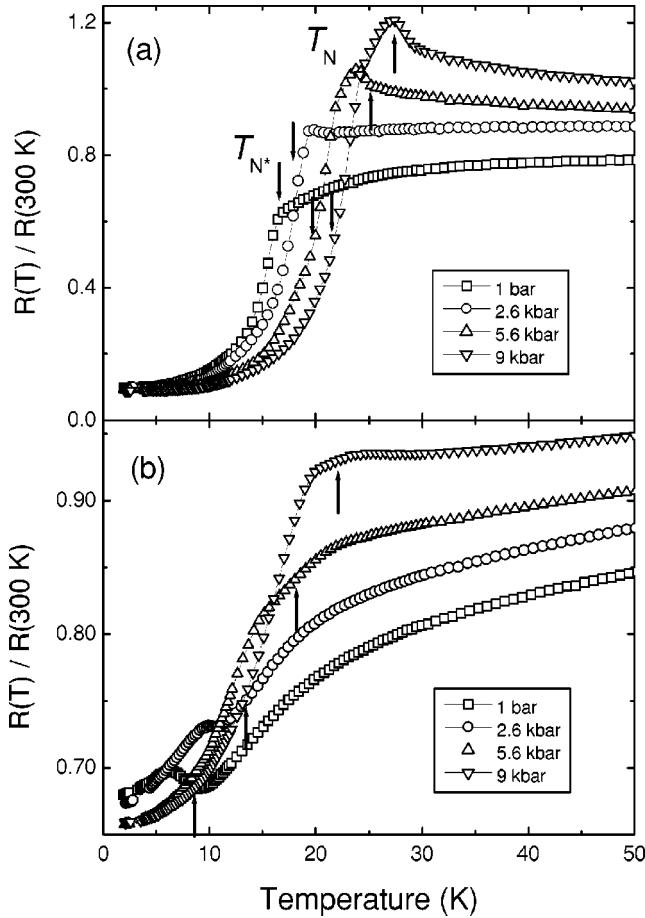


FIG. 2. Temperature dependence of resistivities for CeSb (a) and $\text{CeSbNi}_{0.08}$ (b) at different pressures. Arrows indicate T_N of the AF-I phase and T_{N^*} of the AFP phase as obtained from susceptibility data (see the text).

an antiferromagnetic superzone gap.^{9,11} With increasing pressure, T_N increases quite rapidly at a rate of approximately 1.3 K/kbar. At the same time, the Cr-like anomaly gets subdued and becomes absent altogether above 5.6 kbar. Previous studies on CeSbNi_x showed that the low temperature magnetic state of $\text{CeSbNi}_{0.08}$ is an AF-I phase¹¹ while that of CeSb is an AF-IA phase.⁶ That the Cr-like anomaly is weakened by pressure implies that the AF-I phase of $\text{CeSbNi}_{0.08}$ becomes unstable against pressure and a new magnetic state is formed at high pressure. Interestingly, $\rho(T)$ of $\text{CeSbNi}_{0.08}$ measured under 9 kbar appears very similar to the ambient-pressure $\rho(T)$ of CeSb. These results suggest that $\text{CeSbNi}_{0.08}$ under pressure should have very similar magnetic properties to that of CeSb. Therefore, we propose that pressure strengthens the p - f mixing interaction in $\text{CeSbNi}_{0.08}$ that is weakened by Ni incorporation. Similar behavior was also observed in our magnetization measurements under pressure, which will be discussed later.

$\rho(T)$ of $\text{CeSbNi}_{0.15}$ also exhibits a minimum around T_N and increases continuously at lower temperatures. We note that a small dip at 3 K in the ambient-pressure $\rho(T)$ disappears with pressure. With increasing pressure, the Cr-like anomaly in the $\rho(T)$ of $\text{CeSbNi}_{0.15}$ hardly changes unlike

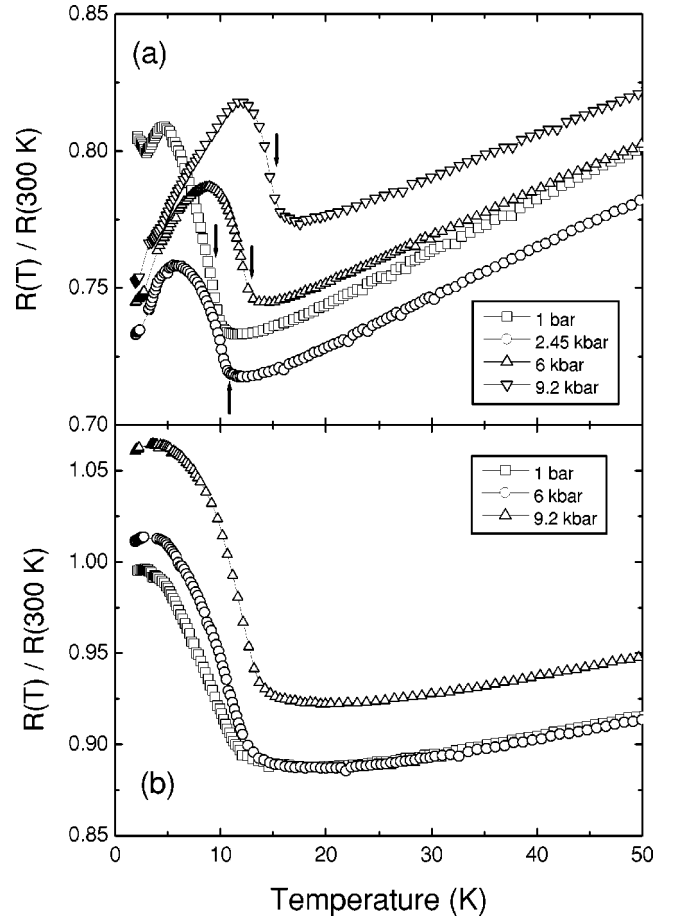


FIG. 3. Temperature dependence of resistivities for $\text{CeSbNi}_{0.15}$ (a) and $\text{CeSbNi}_{0.35}$ (b) at different pressures. Arrows indicate T_N as obtained from susceptibility data. Resistivity of $\text{CeSbNi}_{0.35}$ measured at 2.45 kbar is almost the same as the ambient-pressure data.

that of $\text{CeSbNi}_{0.08}$ while T_N increases continuously. On the other hand, T_N of $\text{CeSbNi}_{0.35}$ is found at 10.5 K at ambient pressure, and with increasing pressure T_N increases only slightly. Surprisingly enough, this pressure dependence of T_N of $\text{CeSbNi}_{0.35}$ is exactly opposite to what we would expect from the x dependence of both T_N and the lattice parameter: T_N has a minimum at around $x=0.08$. Here we should point out that the pressure dependence of T_N found in both $\text{CeSbNi}_{0.15}$ and $\text{CeSbNi}_{0.35}$ cannot be explained at all by negative chemical pressure effects. Therefore, our results suggest that there should be a considerable change in the nature of magnetic properties between below and above $x=0.15$.

C. Pressure-dependent susceptibility

Figure 4 shows the magnetic susceptibility of CeSb, $\text{CeSbNi}_{0.08}$, and $\text{CeSbNi}_{0.15}$ measured with 5 kOe. A sharp peak at 16.5 K in the ambient-pressure susceptibility of CeSb corresponds to a first-order transition to an antiferroparamagnetic (AFP) phase. This transition temperature denoted by T_{N^*} in Fig. 5 increases almost linearly with pressure, reaching 23.5 K at 12 kbar. It is noticeable that the sharp peak in the ambient-pressure data becomes all of sudden much less

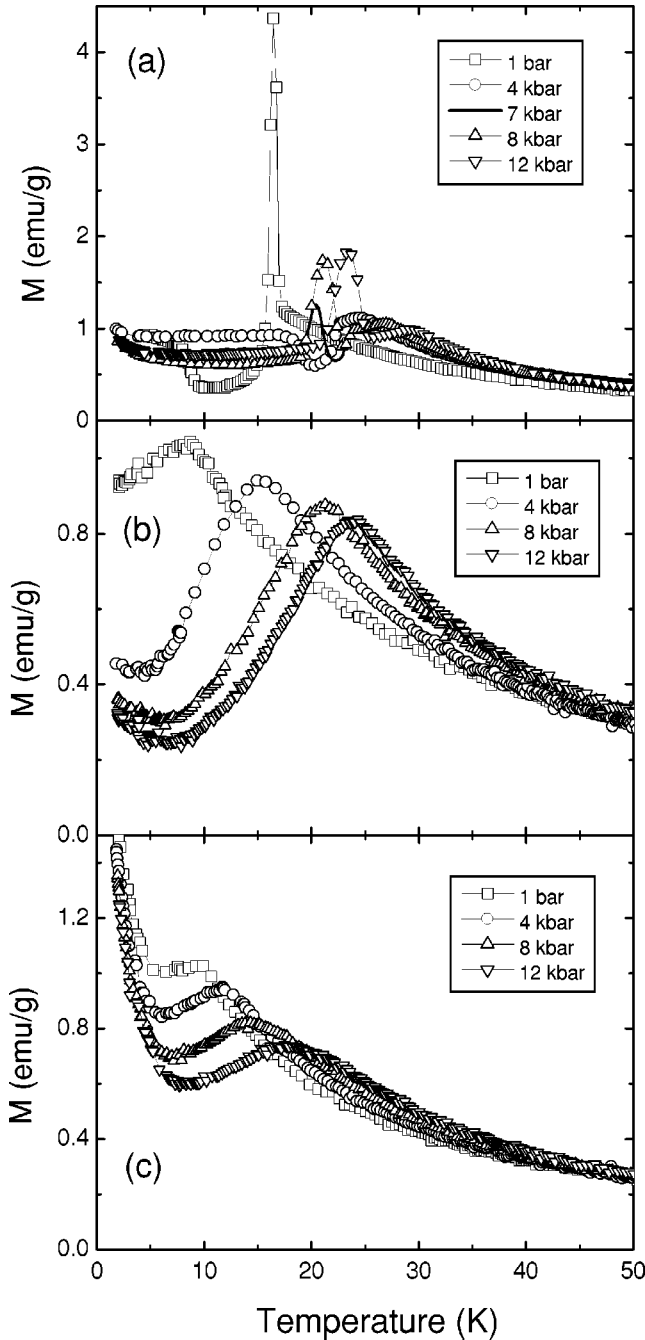


FIG. 4. Pressure dependence of magnetic susceptibility for CeSb (a), CeSbNi_{0.08} (b), and CeSbNi_{0.15} (c) measured with 5 kOe after zero-field cooling.

pronounced in data taken at 4 kbar before gaining strength at higher pressure; which is due to the fact that there exists a critical end point at 2.5 kbar. For pressures above 4 kbar, a new broad feature is observed at higher temperature than T_{N^*} . This broad feature is identified as a second-order transition to the high pressure AF-I phase, and this second-order transition temperature (T_N) increases continuously with pressure. T_N in Figs. 1 and 4 indicates this second-order transition. Pressure dependence of the magnetic transition temperatures of CeSb obtained from the susceptibility mea-

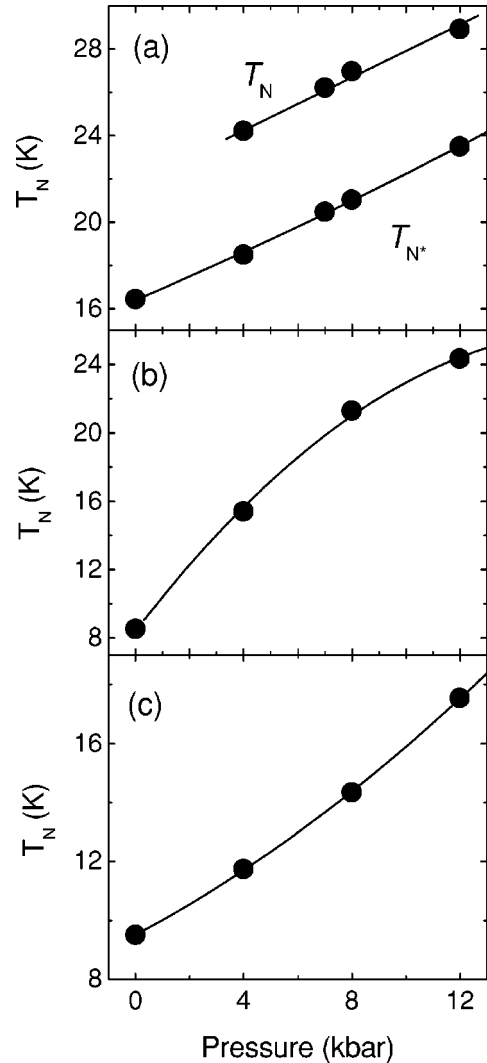


FIG. 5. Pressure dependence of Néel temperature T_N and T_{N^*} for CeSb (a), T_N for CeSbNi_{0.08} (b), and CeSbNi_{0.15} (c) as determined from susceptibility data. Solid line is guide to the eye.

surement are consistent with the (P, T) phase diagram from earlier neutron scattering experiments.¹⁴

Susceptibility of CeSbNi_{0.08} displays a peak at T_N and Curie-Weiss behavior at higher temperatures. The Néel temperature increases considerably with pressure: for CeSbNi_{0.08} $T_N=8.5$ K obtained at ambient pressure increases to 24.3 K at 12 kbar. We also note that the peak in the susceptibility of CeSbNi_{0.08} becomes sharpened with pressure, but even at 12 kbar it is not quite as sharp as the AFP peak observed in the ambient-pressure susceptibility of CeSb [see Fig. 4(b)]. Susceptibility of CeSbNi_{0.15} shows similar behavior too: T_N increases from 9.5 K at ambient pressure to 17.5 K at 12 kbar. A steep rise seen at lower temperature in the susceptibility of CeSbNi_{0.15} is likely to be due to a small amount of nonmagnetic impurities present in the sample. Pressure dependence of T_N is shown in Fig. 5 for all the three samples.

D. Pressure-dependent magnetization

Magnetization of single crystal CeSb exhibits three step-like metamagnetic transitions with a large hysteresis as it

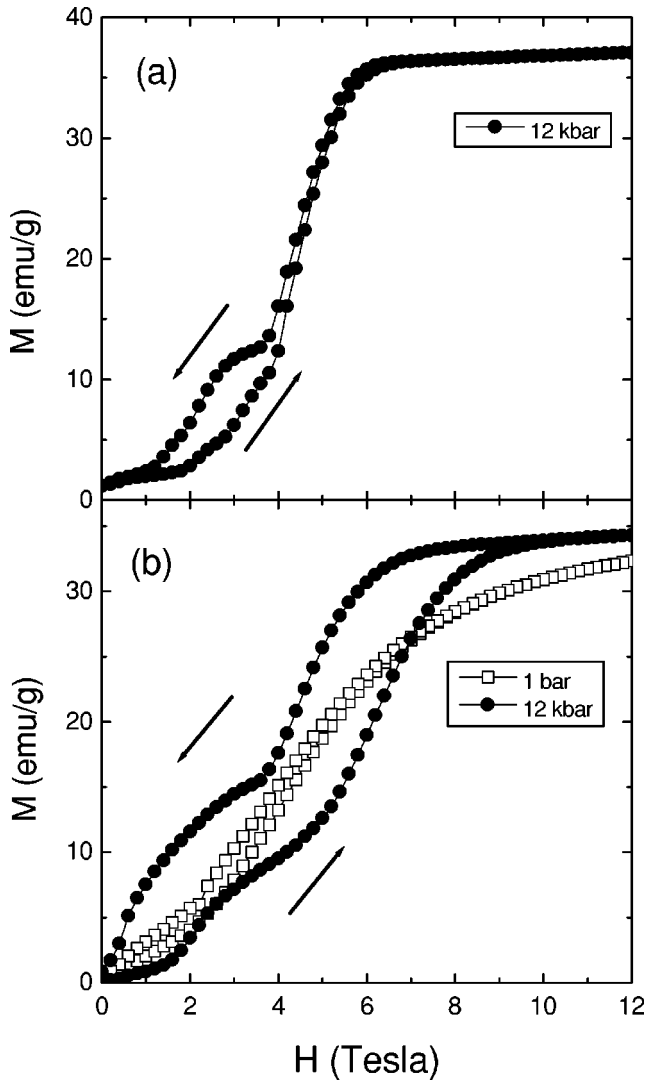


FIG. 6. High field magnetization for CeSb (a) and CeSbNi_{0.05} (b), measured at 4 K after zero-field cooling.

crosses field-induced phase boundaries.¹ For comparison, magnetization of our polycrystalline CeSb sample taken at 12 kbar shows three weak metamagnetic transitions at 2, 3, and 4 T reflecting polycrystalline nature of our sample [see Fig. 6(a)]. This observed hysteresis at high pressure demonstrates that even at 12 kbar the metamagnetic transition still remains to be of first-order nature. The magnetization saturates above 6.5 T as it enters into a high-field ferromagnetic state.

Figure 6(b) shows the high-field magnetization of CeSbNi_{0.05} up to 12 T at both ambient pressure and 12 kbar. The magnetization of CeSbNi_{0.05} taken at ambient pressure shows only one metamagnetic transition at 2.2 T with a relatively smaller hysteresis. Moreover the magnetization does not saturate even at 12 T. However the hysteresis becomes larger at 12 kbar and two metamagnetic transitions are clearly observed at 1.6 and 5.2 T with increasing field. Interestingly enough, saturation behavior now appears in the magnetization above 10 T, which means that the high-field magnetic phase of CeSbNi_{0.05} at 12 kbar is a ferromagnetic

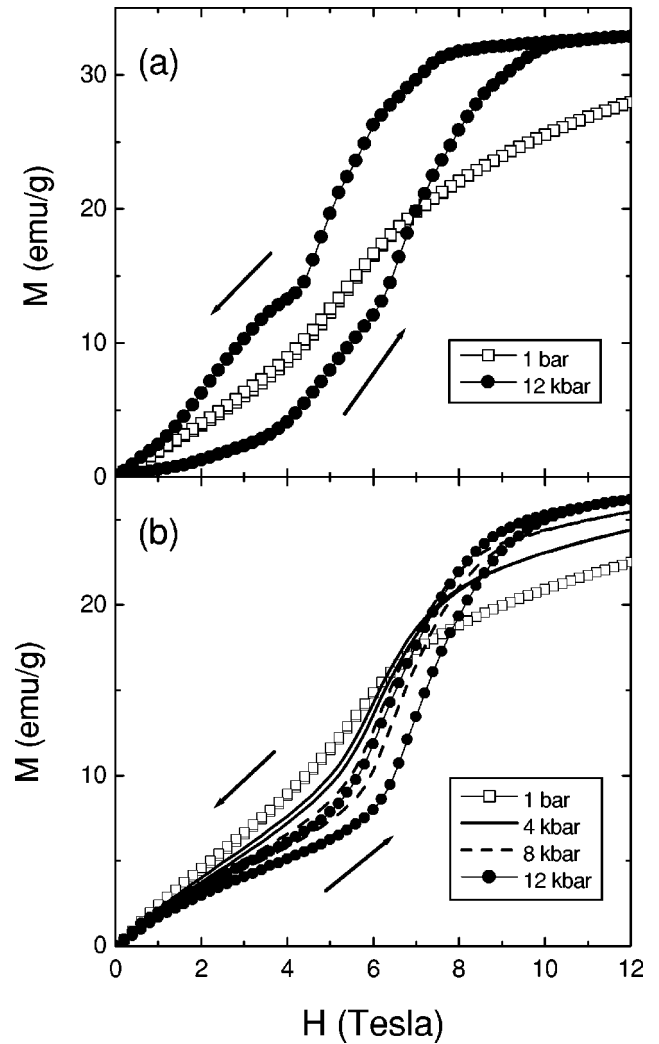


FIG. 7. High field magnetization for CeSbNi_{0.08} (a) and CeSbNi_{0.15} (b), measured at 4 K after zero-field cooling.

state like CeSb above 6 T. On the other hand, magnetization of CeSbNi_{0.08} at ambient pressure exhibits much smaller hysteresis and only a weak metamagnetic transition around 4.5 T without saturation behavior up to 12 T [see Fig. 7(a)]. At 12 kbar, however there also appear a larger magnetic hysteresis and two metamagnetic transitions at 3.9 and 6.2 T with a clear sign of saturation behavior above 10 T. This implies that a ferromagnetic state is realized above 10 T and 12 kbar for CeSbNi_{0.08} too. All these results indicate that although at ambient pressure the magnetic properties of CeSbNi_{0.05} and CeSbNi_{0.08} are different from that of the parent compound CeSb, upon applying external pressure the magnetic properties of CeSbNi_{0.05} and CeSbNi_{0.08} become similar to that of CeSb. These findings are also consistent with the results of pressure-dependent resistivity measurements.

Figure 7(b) shows the pressure-dependent magnetization of CeSbNi_{0.15} from ambient pressure to 12 kbar. The ambient-pressure magnetization shows a small magnetic hysteresis and a very weak metamagnetic transition. However, the hysteresis is made larger and the metamagnetic transition becomes clearer with increasing pressure. The metamagnetic

transition field also increases with pressure reaching 6.2 T at 12 kbar. Even at 12 kbar, however, only one metamagnetic transition is observed and the magnetization still continues to increase without a clear sign of saturation behavior. These observations confirm our conclusion drawn from the resistivity measurements that there should be a significant change in the nature of the magnetic properties of CeSbNi_x between below and above $x=0.15$.

IV. DISCUSSION

The complex and strongly anisotropic magnetic properties of CeSb were explained by the nonlinear mixing interaction between Sb 5*p* valence holes and Γ_8 state of Ce 4*f*.⁸ This so-called *p-f* mixing effect gets stronger with pressure as shown in the pressure dependence of our resistivity and magnetization results. On the other hand, strong suppression of T_N with Ni incorporation implies that thereby the *p-f* mixing gets considerably weakened; of which main effect is obviously the volume expansion, i.e., the negative chemical pressure effects. However, it is not clear yet which one of the negative chemical pressure effect and the decrease of Sb 5*p* hole concentration due to Ni incorporation is relatively more important in determining the magnetic ground state of CeSbNi_x . In order to answer this question, we have analyzed our results quantitatively so as to separate pure pressure effects from nonpressure related ones in our results. In order to do this, we used bulk modulus value of 715 kbar for CeSb (Ref. 15) and estimated that about $\Delta V/V \cong -0.14\%$ of fractional volume contraction can be induced by 1 kbar of pressure. Using the observed *x* dependence of the lattice constant ($da/dx = 0.085 \text{ \AA}$), we reckon that a fractional volume expansion for $x=0.1$ is about $\Delta V/V \cong 0.4\%$. Therefore, purely from the point of view of volume changes pressure of 4 kbar is expected to induce a bigger volume contraction than is possible with Ni incorporation for $x=0.1$.

As we commented on $\rho(T)$ of $\text{CeSbNi}_{0.08}$, the Cr-like anomaly disappears with pressure and $\rho(T)$ at high pressure looks similar to the resistivity of CeSb taken at ambient pressure. We interpret this pressure dependence that the magnetic properties of $\text{CeSbNi}_{0.08}$ under pressure become similar to that of CeSb. This conclusion is also consistent with our high-field magnetization under pressure. From these results, we can therefore assume that the weakened *p-f* mixing due to Ni incorporation is at least partly recovered by external pressure. If we consider the lattice constant changes either due to pressure or Ni incorporation alone, however, $\text{CeSbNi}_{0.08}$ under pressure of 12 kbar is expected to be already at a similar state of CeSb at 9.7 kbar. Nevertheless, there is a clear difference between the high-pressure phase of $\text{CeSbNi}_{0.08}$ and the ambient-pressure phase of CeSb. For example, we cannot observe a sharp peak in the susceptibility of $\text{CeSbNi}_{0.08}$ unlike CeSb (see Fig. 4). This then implies that apart from the negative chemical pressure effect there ought to be other effects of Ni incorporation on the magnetic properties. One obvious, and natural candidate for such effects is a change in the Sb 5*p* hole concentration that is reduced

with increasing Ni amount as shown in the Hall coefficient measurements;¹¹ which subsequently weakens the *p-f* mixing.

For $\text{CeSbNi}_{0.15}$, however, the resistivity and magnetization show qualitatively different behavior under pressure compared to $\text{CeSbNi}_{0.08}$. First, the continuous increase of T_N with pressure is not anticipated from the observed *x* dependence of T_N (Refs. 9,11) (see Fig. 5). Second, although pressure of 9.2 kbar is larger than the calculated value of 4.3 kbar; which is estimated to fully compensate for the volume expansion due to Ni incorporation of $x=0.15$, the resistivity anomaly at low temperature is not affected as much as one would expect. Finally, the high-field magnetization of $\text{CeSbNi}_{0.15}$ under pressure shows quite different behavior from those of $x \leq 0.08$ as described previously: neither clear sign of saturation behavior nor hysteresis is found. These observations lead us to conclude that the magnetic state of CeSbNi_x for $x \geq 0.15$ has different nature compared to that of CeSbNi_x for $x \leq 0.08$. Here, one might have to invoke that the hole concentration of Sb 5*p* band decreases with Ni incorporation; which according to our results seems to be more important than the negative chemical pressure effects. This decrease in the hole concentration together with the increase of the crystal field splitting energy (Δ_{CEF}) as observed in inelastic neutron scattering experiments,¹⁰ we believe, tip the total energy of CeSbNi_x in favor of the much simpler AF-I phase.

Therefore, our studies reinforce a long-held view that the complex magnetic states of CeSb arise through a delicate balance among the three factors; the strength of the *p-f* mixing, the hole concentration of Sb 5*p* bands, and the crystal field splitting. Furthermore, our results together with previous works⁹⁻¹¹ on CeSbNi_x open up a new possibility that CeSbNi_x systems can be used as a test ground for any realistic theories of the magnetic phases of CeSb, including the anisotropic *p-f* mixing model.

V. SUMMARY

We measured pressure-dependent resistivity, susceptibility, and magnetization of CeSbNi_x system for $0 \leq x \leq 0.35$ to investigate effects of Ni incorporation on the magnetic properties of CeSbNi_x . For $\text{CeSbNi}_{0.08}$, a resistivity anomaly was suppressed with pressure and the temperature dependence of the resistivity became very similar to that of CeSb while $\rho(T)$ of CeSbNi_x for $x \geq 0.15$ does not show such behavior. Metamagnetic transitions, magnetic hysteresis, and saturation behavior become clearly observable with pressure in the magnetization of CeSbNi_x for $x \leq 0.08$ while such features are not found for $\text{CeSbNi}_{0.15}$ either at ambient pressure or high pressure up to 12 kbar. From these results, we conclude that there is clearly different pressure dependence of the magnetic transitions between below and above $x=0.15$. Moreover, we find that apart from the volume expansion due to Ni incorporation, the concomitant decrease in the number of Sb 5*p* hole is also necessary for us to understand both the *x* and pressure dependence of our data.

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