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Pressure effect on magnetic properties of valence fluctuating system $Ce(Ni_{1-x}Cu_x)_5$

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Magnetic susceptibility χ of the isostructural $Ce(Ni_{1-x}Cu_x)_5$ alloys ($0 \le x \le 0.9$) was studied as a function of the hydrostatic pressure up to 2 kbar at fixed temperatures 77.3 and 300 K. A pronounced pressure effect on susceptibility is found to be negative in sign and nonmonotonously dependent on the Cu content, showing a sharp maximum at $x \simeq 0.4$. The experimental results are discussed in terms of the valence instability of Ce ion in the studied alloys. For the reference $CeNi_5$ compound the main contributions to χ and their volume dependence are calculated *ab initio* within the local spin density approximation, and appeared to be in close agreement with experimental data.

PACS: 71.20.Eh Rare earth metals and alloys;
75.30.Mb Valence fluctuation, Kondo lattice, and heavy-fermion phenomena;
75.80.+q Magnetomechanical effects, magnetostriction.

Keywords: intermediate valence, magnetic susceptibility, pressure effect, electronic structure.

1. Introduction

Many of Ce intermetallics are characterized by a strong hybridization of the magnetic 4*f*-electrons with the conduction electron states resulted in delocalization of the 4*f*-level and a change of its occupancy, and hence the Ce valence. As is evident from measurements of x-ray absorption and lattice parameters [1], together with the magnetic [2,3], electric and thermoelectric properties [3], in the isostructural $Ce(Ni_{1-x}Cu_x)_5$ alloys, the Ce valence decreases consistently from Ce^{4+} to Ce^{3+} with increase of the Cu content. Accordingly, the system undergoes a series of transitions from the nonmagnetic metal with the unoccupied 4*f*-level ($x = 0$) through the intermediate valence (IV) state combined with a nonmagnetic dense Kondo state ($0.1 \le x \le 0.8$) to the magnetic 4*f*-metal ($0.9 \le x \le 1$). Thus, the reference $CeNi_5$ compound is expected to be the exchange-enhanced itinerant paramagnet [1,4,5] with the temperature dependent magnetic susceptibility exhibiting a broad maximum around 100 K, similar to those observed in YNi_5, $LaNi_5$ and $LuNi_5$ [4,6,7]. On the other side, the $CeCu_5$ compound is a Kondo lattice antiferromagnet with $T_N = 3.9$ K and $T_K = 2.2$ K [8]. The magnetic susceptibility in $CeCu_5$ obeys a Curie–Weiss law at $T \ge 50$ K with the effective magnetic moment value close to that expected for Ce^{3+} state [8–10]. Due to a direct relation between magnetic properties and the rare earth (RE) valence state, and also the strong correlation between the valence itself and RE ionic volume, the RE compounds with unstable *f* shell exhibit a large magnetovolume effect. Therefore, a study of pressure effect on magnetic properties of the systems with variable RE valence is of great interest to gain insight into a nature of the IV state.

Here we report results of our investigation of the pressure effect on the magnetic susceptibility of $Ce(Ni_{1-x}Cu_x)_5$ alloys in a wide range of Cu concentrations. The experimental results are supplemented by calculations of the magnetovolume effect value for the reference $CeNi_5$ compound, using a modified relativistic full potential approach within linearized "muffin-tin" orbital method (FP–LMTO).

2. Experimental details and results

The polycrystalline samples of $Ce(Ni_{1-x}Cu_x)_5$ alloys $(0 \le x \le 0.9)$ were prepared by arc-melting of a stoichiometric amount of initial elements in a water cooled crucible under protective argon atmosphere. The study of x-ray powder diffraction at room temperature revealed that all samples crystallize in $CaCu_5$-type hexagonal structure, and obtained data on their lattice parameters agree closely with that published in literature. Any other phases were not detected within the resolution of the x-ray technique.

The pressure effect on the magnetic susceptibility χ was measured under helium gas pressure up to 2 kbar at two fixed temperatures, 77.3 and 300 K, using a pendulum magnetometer placed into the nonmagnetic pressure cell [11]. The relative errors of our measurements, performed in the magnetic field $H = 1.7$ T, did not exceed 0.05% .

In Fig. 1 the typical pressure dependencies of the magnetic susceptibility for $Ce(Ni_{1-x}Cu_x)_5$ alloys demonstrate a magnitude of the pressure effect and its linear behavior. For each temperature the values of χ at ambient pressure and their pressure derivatives, $d\ln\chi(x,T)/dP$, are listed in Table 1. These values include corrections for a weak field dependence of χ caused by ferromagnetic impurities, which are less than 5%. The negative sign of the pressure effect is consistent with anticipation that high pressure has to increase the valence, since the Ce ion in the less magnetic higher valence state has a smaller volume.

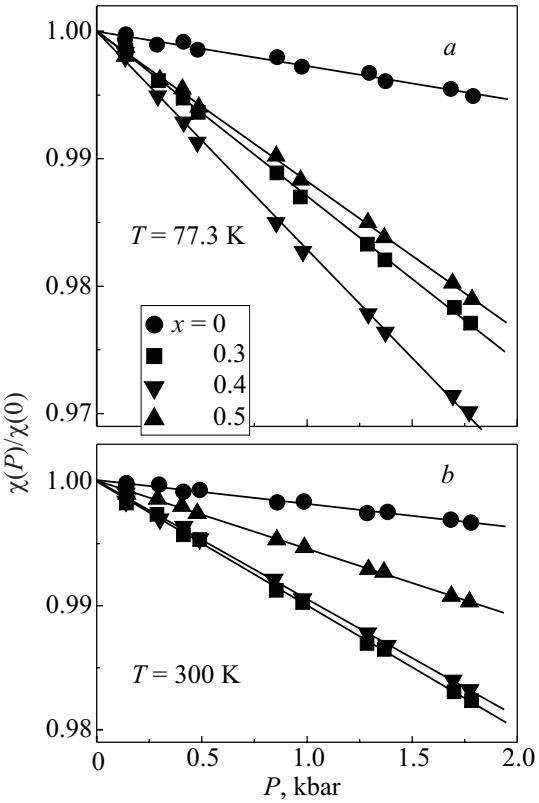

Fig. 1. Pressure dependence of the magnetic susceptibility of $Ce(Ni_{1-x}Cu_x)_5$ alloys at $T = 77.3$ K (*a*) and 300 K (*b*) normalized to its value at $P = 0$.

Table 1. The magnetic susceptibilities and their pressure derivatives for $Ce(Ni_{1-x}Cu_x)_5$ alloys at 77.3 and 300 K

x	χ, 10^{-3} emu/mole		$d\ln\chi/dP$, $Mbar^{-1}$	
	T, K		T, K	
	77.3	300	77.3	300
0.0	3.29	2.12	–2.72±0.3	–1.93±0.3
0.1	2.74	1.47	–3.41±0.4	–3.02±0.4
0.2	1.55	1.09	–4.55±0.4	–4.93±0.3
0.3	1.11	1.08	–13.0±0.5	–9.93±0.5
0.4	1.47	1.26	–17.1±1	–9.5±0.5
0.5	3.67	1.87	–11.8±0.5	–5.52±0.5
0.6	7.85	2.47	–6.63±0.5	–3.28±0.3
0.7	9.55	2.78	–3.8±0.3	–2.03±0.3
0.9	9.93	2.76	–1.42±0.2	–1.26±0.2

Of particular interest is a strong and nonmonotonous concentration dependence of the pressure effect which shows a sharp maximum in vicinity of $x \simeq 0.4$ for both temperatures, 77.3 and 300 K (Fig. 2,*a*). A comparison between the obtained experimental results and the data on concentration dependence of the lattice parameter a and the effective Ce valence ν from Ref. 1 (Fig. 2,*b*) indicates that the maximum in $d\ln\chi(x,T)/dP$ correlates with a drastic change of a (and ν) around $x \simeq 0.4$ ($\nu \simeq 3.5$).

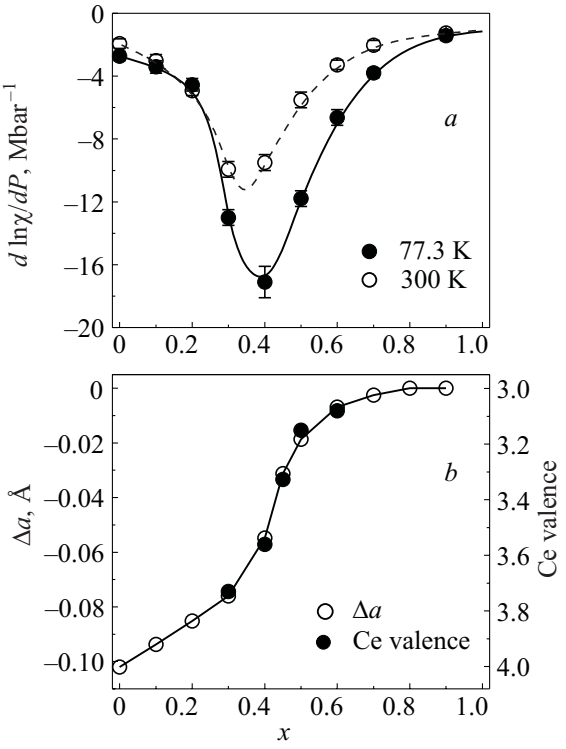

Fig. 2. Pressure derivative of the magnetic susceptibility $d\ln\chi/dP$ in $Ce(Ni_{1-x}Cu_x)_5$ alloys at 77.3 and 300 K (*a*). Deviation of the a lattice parameter, Δa, in $Ce(Ni_{1-x}Cu_x)_5$ alloys from the $a(x)$ dependence for the Ce ion assumed to be in a trivalent state (left scale) and the Ce valence deduced from x-ray absorption studies (right scale) at room temperature versus Cu content x (according to the data of Ref. 1) (*b*).

It should be noted that a similar peculiarity in $d\ln\chi/dP$ versus valence was observed for various Yb compounds at room temperature [12]. As was shown, the relative change of χ with pressure appeared to be the most pronounced also for the half-integer value of valence, $\nu \simeq 2.5$, but contrary to the Ce compounds, the pressure effect in Yb compounds has a positive sign.

3. Electronic structure and magnetic properties of $CeNi_5$

Ab initio calculations of the electronic structure were carried out for the $CeNi_5$ compound by employing a FP–LMTO method [13,14]. The exchange-correlation potential was treated in the LSDA approximation [15] of the density functional theory. In order to analyze the observed magneto-volume effect value in $CeNi_5$, the magnetic susceptibility and its volume dependence were calculated within the modified method, wherein the external magnetic field H was taken into account by means of the Zeeman operator, $H(2\hat{s}+\hat{l})$. The latter was incorporated in FP–LMTO Hamiltonian [16–18] for calculations of the field-induced spin and orbital magnetic moments. The corresponding contributions to magnetic susceptibility, χ_{spin} and χ_{orb}, were derived from these field-induced moments, which have been calculated in an external magnetic field of 10 T. This field was applied both parallel and perpendicular to the c axis, providing the components of anisotropic magnetic susceptibility, $\chi_{\|}$ and $\chi_{\perp}$, respectively.

The electronic structure calculations were performed for a number of lattice parameters a close to the experimental one. However, in doing so the c/a ratio was fixed at its experimental value 0.8226. The equilibrium lattice spacing a_{th} = 8.96 a.u. and corresponding theoretical bulk modulus B_{th} = 1.9 Mbar were determined from dependence of the total energy on the unit cell volume, $E(V)$, by using the Murnaghan equation [13]:

$$E(V) = E_{coh} + \frac{BV_0}{B'}\left(\frac{(V_0/V)^{B'-1}}{B'-1} + \frac{V}{V_0} - \frac{B'}{B'-1}\right). \quad (1)$$

By this way the bulk properties, such as the equilibrium volume V_0, the bulk modulus B, and its first pressure derivative B' are directly related to the equation (1). Here E_{coh} is the cohesive energy and it is treated as an adjustable parameter. The Murnaghan equation is based on the assumption that the pressure derivative B' of the bulk modulus B is constant. By using the evaluated from the Murnaghan equation value of $B' = 3.73$, we have estimated B_{th}^{est} = 1.45 Mbar, corresponding to the experimental a_{exp} = 9.2 a.u. [1]. This correction counterbalances the well known over-bonding tendency of the LSDA approach ([13,14]), and provides a nice agreement with the available experimental value, B_{exp} = 1.43 Mbar [19].

The strongly volume dependent spin contribution to susceptibility χ_{spin} originates predominantly from the 3*d*-states of Ni. Regarding the orbital contribution χ_{orb}, it comes mainly from conduction electrons in the atomic sphere of Ce and amounts to about 20% of total susceptibility. One would expect that the anisotropy of the susceptibility in the non-magnetic hexagonal $CeNi_5$ compound is predominantly due to the orbital Van Vleck-like contribution χ_{orb}. The calculated anisotropy is found to be small, $\Delta\chi \simeq 0.5\cdot10^{-4}$ emu/mole, in agreement with our data and that of Ref. 1.

At the theoretical lattice parameter the averaged value of the calculated susceptibility, $\overline{\chi} = (\chi_{\|} + 2\chi_{\perp})/3 = 2.9\cdot10^{-3}$ emu/mole, appeared to be very close to the experimental value of $3.0\cdot10^{-3}$ emu/mole at $T = 0$ K [2,4]. The calculated volume derivative of susceptibility, $d\ln\chi/\ln V = 4.2$, is in agreement with that resulted from our experimentally observed pressure derivative for $CeNi_5$ at T = 77.3 K, $d\ln\chi/d\ln V = 3.9\pm0.4$. Thus it is demonstrated, that LSDA provides an adequate description of the strongly exchange enhanced magnetic susceptibility of $CeNi_5$ and its pressure dependence.

4. Magnetic properties of $Ce(Ni_{1-x}Cu_x)_5$ alloys

As is demonstrated, the LSDA allows to describe the magnetic susceptibility and its volume dependence in the reference $CeNi_5$ compound. This gives grounds for future application of ab initio approaches to evaluate the itinerant background paramagnetism χ_0 in $Ce(Ni_{1-x}Cu_x)_5$ alloys, which is expected to decrease progressively with increasing of Cu content for $x \geq 0.1$, according to the experimental and calculated data on susceptibility in $Y(Ni_{1-x}Cu_x)_5$ and $La(Ni_{1-x}Cu_x)_5$ alloys [7]. At the present, however, we are unable to take into account in a rigorous way such important effects in the susceptibility of alloys as disorder, crystal electric fields, and indirect interactions of the moments. Therefore we restrict here our consideration of the experimental data in alloys within a phenomenological approach to examine effects of the localized magnetism.

4.1. Concentration dependence

Assuming that pressure effect on magnetic susceptibility arises mainly from the change of Ce valence ν, or the fractional occupation of the $4f^1$-magnetic state n_{4f} ($\nu = 4 - n_{4f}$), the pressure effect can be analyzed within a simple relation

$$\frac{d\ln\chi(T)}{dP} \approx \frac{\partial\ln\chi(T)}{\partial n_{4f}}\frac{dn_{4f}}{dP}, \quad (2)$$

in terms of the pressure dependence of n_{4f} (or ν). The most reliable results of such analysis would be expected in the Cu-rich alloys at low temperatures where the 4*f*-contribution χ_{4f} becomes dominant ($\chi \approx \chi_{4f}$).

In Fig. 3,*a* the χ versus n_{4f} dependence is shown for $Ce(Ni_{1-x}Cu_x)_5$ alloys ($0.4 \leq x \leq 0.7$) at 77.3 K, which was obtained by using the experimental $\chi(x)$ values from Table 1 and the $\nu(x)$ data in Fig. 2,*b*. A substitution of the

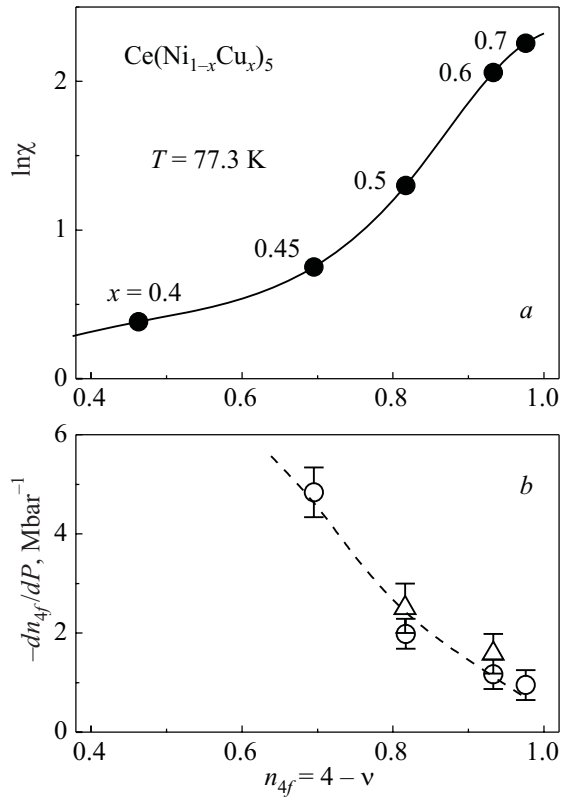

Fig. 3. Values of $\ln\chi$ at 77.3 K (*a*) and dn_{4f}/dP (*b*) plotted against n_{4f} for $Ce(Ni_{1-x}Cu_x)_5$ alloys. Symbols (○) and (Δ) denote the data obtained within Eq. (2) and Eq. (5), respectively. Points for $x = 0.45$ are interpolation of the experimental data on concentration dependence of χ and $d\ln\chi/dP$.

resulted from Fig. 3,*a* derivatives $\partial\ln\chi/\partial n_{4f}$ and experimental data on $d\ln\chi/dP$ at 77.3 K into Eq. (2) gives the value of dn_{4f}/dP which strongly depends on n_{4f} (Fig. 3,*b*). Based on the concentration behavior of valence ν and the effect of pressure in the susceptibility (Fig. 2), the maximum value of dn_{4f}/dP is expected at $n_{4f} \sim 0.5$ ($\nu \sim 3.5$) and found by extrapolation of the data in Fig. 3,*b* to be about -6.5 ± 1.5 Mbar^{-1}. The corresponding estimates of the valence change under pressure, $d\nu/dP = -dn_{4f}/dP$, are of the same order that those observed in other IV compounds, e.g., resulted from the study of the magnetovolume effect in SmB_6 (2 Mbar^{-1} [20]) and from the measurements of resonant inelastic x-ray emission in $YbAl_2$ under pressure (~ 5 Mbar^{-1} [21]).

4.2. Temperature dependence

In a simple empirical model [22] which includes interconfiguration fluctuations between f^{n+1}- and f^n-levels, the contribution of the $4f^0(J=0)$ and $4f^1(J=5/2)$ states of Ce to magnetic susceptibility is given by

$$\chi_{4f}(T) = N_A\mu^2 n_{4f}(T)/3k(T+T_f). \qquad (3)$$

Here N is the Avogadro number, μ is effective magnetic moment of the $4f^1$-state, T_f is the characteristic temperature (valence fluctuation temperature, or Kondo temperature, or heavy-fermion bandwidth). It should be noted that a quantitative analysis of the $\chi_{4f}(T)$ dependence using Eq. (3) requires the complete data on $n_{4f}(T)$ (and probably on $T_f(T)$ as well) which are actually unavailable. Furthermore, in order to separate the $\chi_{4f}(T)$ term from the experimental data on $\chi(T)$ one needs to know a background contribution χ_0, which generally can not be neglected. A simplified analysis of the experimental data can be performed assuming n_{4f}, T_f and χ_0 to be temperature independent. Then the magnetic susceptibility obeys a modified Curie–Weiss law,

$$\chi(T) = \chi_0 + \chi_{4f}(T) \equiv \chi_0 + C/(T-\Theta), \qquad (4)$$

with $C = N\mu^2 n_{4f}/3k$ and $\Theta = -T_f$. For the representative $Ce(Ni_{0.5}Cu_{0.5})_5$ alloy, the best fit of Eq. (4) to the experimental data [2] at $T \geq 50$ K (Fig. 4,*a*) is obtained with $\chi_0 = 0.6\cdot10^{-3}$ emu/mole, $C = 0.48$ K$\cdot$ emu/mole and $\Theta = -79$ K. It should be pointed out that the estimate $n_{4f} = 0.6$, resulted from C, is in a reasonable agreement with the value of 0.8 that follows from the data in Fig. 2,*b* for $x = 0.5$.

As is evident from Eq. (3), the pressure effect on the 4*f*-susceptibility is governed by changes of n_{4f} and T_f with pressure,

$$\frac{d\ln\chi_{4f}(T)}{dP} = \frac{d\ln C}{dP} - \frac{1}{(T+T_f)}\frac{dT_f}{dP} \equiv$$

$$\equiv \frac{d\ln n_{4f}}{dP} - \frac{\chi_{4f}(T)}{C}\frac{dT_f}{dP}, \qquad (5)$$

being a linear function of $(1/(T+T_f))$ or $\chi_{4f}(T)$. The data on $d\ln\chi_{4f}/dP$ for the $Ce(Ni_{0.5}Cu_{0.5})_5$ alloy were derived from the measured effect, $d\ln\chi/dP$, in the framework of Eq. (4) by using a value $d\ln\chi_0/d\ln V \sim 1$ as a rough estimate for the volume dependence of the unenhanced background susceptibility [23], which is assumed to originate from the *sp*-3*d*(5*d*) hybridized itinerant electrons. The obtained values are plotted in Fig. 4,*b* as a function of $\chi_{4f}(T)$. Its linear approximation in accordance with Eq. (5) gives

$$\frac{d\ln C}{dP} = \frac{d\ln n_{4f}}{dP} = -3.2 \pm 0.7 \text{ Mbar}^{-1},$$

$$\frac{dT_f}{dP} = 1650 \pm 250 \text{ K}\cdot\text{Mbar}^{-1}. \qquad (6)$$

The resultant value $dn_{4f}/dP = -2.5 \pm 0.5$ Mbar^{-1} is in line with the value $dn_{4f}/dP = -2.0 \pm 0.3$ Mbar^{-1} obtained above for $x = 0.5$ from analysis of the concentration dependence of the pressure effect within Eq. (2). From the

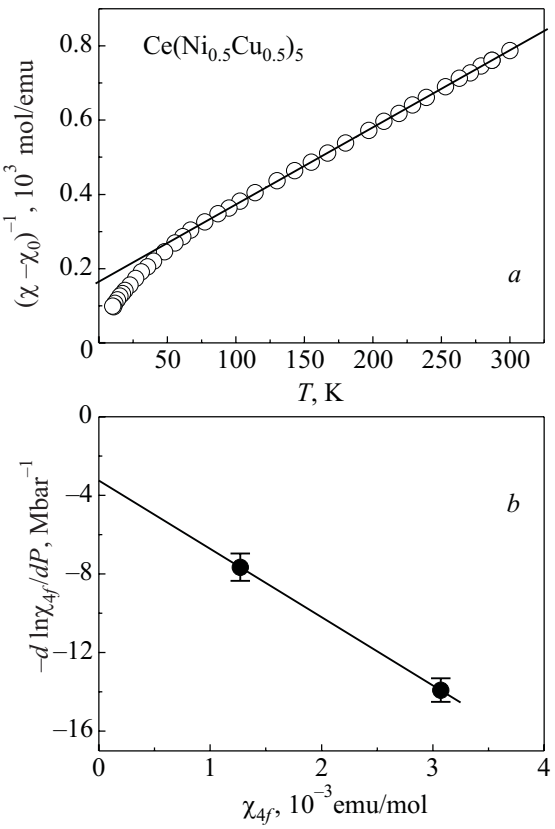

Fig. 4. Temperature dependence of the magnetic susceptibility χ (*a*) and pressure derivative $d\ln\chi_{4f}/dP$ plotted against χ_{4f} (*b*) for $Ce(Ni_{0.5}Cu_{0.5})_5$ alloy.

pressure dependence of T_f the corresponding Grüneisen parameter, Ω, is estimated to be

$$\Omega_f \equiv -\frac{d\ln T_f}{d\ln V} = B\,\frac{d\ln T_f}{dP} = 31\pm 5 \qquad (7)$$

using the experimental bulk modulus $B = 1.5$ Mbar [24].

It should be noted that the Anderson impurity model provides the Kondo temperature and its pressure derivative to be mainly described in terms of n_{4f} [25–27]:

$$T_K \propto \frac{1-n_{4f}}{n_{4f}},\quad \frac{d\ln T_K}{dP} = -\frac{1}{1-n_{4f}}\,\frac{d\ln n_{4f}}{dP}. \qquad (8)$$

Then, assuming $T_f \propto T_K$ and using in Eq. (8) the values $dn_{4f}/dP = -3.2\pm 0.7$ Mbar^{-1} and $n_{4f} = 0.8$ evaluated above for the alloy with $x = 0.5$, one obtains

$$\Omega_f \simeq \Omega_K = -\frac{d\ln T_K}{d\ln V} = 24\pm 5 \qquad (9)$$

in a reasonable agreement with the direct estimate (7).

For $Ce(Ni_{0.4}Cu_{0.6})_5$ alloy, the analogous analysis in the framework of Eq. (4) and Eq. (5) yields the following Curie–Weiss parameters: $C \simeq 0.806$ K·emu/mole, $\chi_0 \sim 0$, $T_f = -\Theta = 26$ K, and their pressure derivatives:

$$\frac{d\ln C}{dP} = \frac{d\ln n_{4f}}{dP} = -1.7\pm 0.5\ \text{Mbar}^{-1},$$

$$\frac{dT_f}{dP} = 620\pm 100\ \text{K}\cdot\text{Mbar}^{-1}.$$

The latter results in the Grüneisen parameter $\Omega_f = 35\pm 6$, assuming the bulk modulus value $B = 1.5$ Mbar, as in the $Ce(Ni_{0.5}Cu_{0.5})_5$ alloy. Within the Anderson impurity model, the similar estimate $\Omega_f \simeq \Omega_K = 36\pm 10$ follows from Eq. (8) with $n_{4f} \simeq 0.93$ derived from the data in Fig. 2,*b* for $x = 0.6$. It should be noted that the evaluated values of Ω_K are about half of that obtained for $Ce(In_{1-x}Sn_x)_3$ alloys, which exhibit similar regimes of localization of the *f*-states [28].

5. Conclusions

The pressure effect on magnetic susceptibility of $Ce(Ni_{1-x}Cu_x)_5$ alloys was studied for the first time. This effect is negative in sign, and also strongly and nonmonotonously dependent on the Cu content. For the reference $CeNi_5$ compound, the pressure effect value is successfully described within LSDA approximation by using the modified full potential relativistic FP–LMTO method. For $Ce(Ni_{1-x}Cu_x)_5$ alloys the effects of pressure and alloying on the valence state of Ce ion are the most pronounced around $x \sim 0.4$, which corresponds to the half-integer valence $\nu \sim 3.5$. In other words, the fractional occupation $n_{4f} \sim 0.5$ with the nearly degenerate f^0- and f^1-configurations of electronic states is favorable for the valence instability. It is also found that the main contributions to the pressure effect on magnetic susceptibility for the Cu-rich alloys are i) the decrease of the effective Curie constant and ii) the increase of the characteristic temperature T_f. The latter exhibits a large and positive value of the Grüneisen parameter, which can be apparently described within the Anderson impurity model. Both of these contributions have their common origin in the change of the Ce valence state caused by depopulation of the *f*-state under pressure. However, only additional experimental and theoretical studies could shed light on the relative contributions of two principle mechanisms of such depopulation, namely, the shift of the Ce 4*f*-energy level relative to the Fermi energy, or the broadening of this level.

The authors dedicate this work to the 80th birthday anniversary of V.G. Peschansky, who is one of pioneers in the field of magnetic properties studies in metallic systems.

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