Electronic and magnetic properties of heavy fermion CeCu$_4$Al

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Abstract
The studies of the magnetic susceptibility, specific heat, electrical resistivity, x-ray
photoemission spectra (XPS) and the band structure calculations for CeCu$_4$Al are reported.
CeCu$_4$Al is paramagnetic and follows the Curie–Weiss law with $\mu_{\text{eff}} = 2.53 \, \mu_{\text{B}}$/f.u. and
$\theta_{\text{p}} = -10$ K. The experimental value of μ_{eff} is close to the calculated one for a free Ce^{3+} ion
(2.54 μ_{B}), thus indicating the presence of well-localized magnetic moments carried by the
stable Ce^{3+} ions. Below the Fermi energy the total density of states contains mainly the d states
of Cu atoms, which hybridize with the Ce f electronic states. The analysis of the Ce 3d and 4d
XPS spectra indicates the localized character of the Ce f states. The determined electronic
specific heat coefficient $\gamma = 210$ mJ K^{-2} mol^{-1} confirms the heavy fermion character of the
CeCu$_4$Al compounds.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Ce intermetallics are very attractive compounds due to a variety of interesting behaviors. They are frequently characterized by an anomalous proximity of the Ce 4f level to the Fermi energy. This proximity yields a strong hybridization of the magnetic 4f electrons with the conduction electrons. The classical Kondo impurity problem and the mixed-valence behavior in Ce-, Yb-and U-based compounds laid the foundations of the extensive studies of heavy fermion physics during the previous 50 years [1–3]. Strongly correlated electron systems are still the focus of interest both in theoretical and experimental studies.

The CeCu$_4$Al compound is a derivative of the CaCu$_5$-type structure and it does not order magnetically till low temperatures. Experimental investigations have shown that CeCu$_4$Al is a heavy fermion material [4–7]. It was previously found that the estimated value of the electronic specific heat coefficient γ was equal to about 280 mJ K^{-2} mol^{-1} [5]. Below

6 K the C/T dependence showed a strong increase, which yielded a value of C/T of about 1.1 J K^{-2} mol^{-1} [5]. The measurements of γ in magnetic fields up to 12 T showed the reduction of C/T values by a factor of five at low temperatures [8].

We have previously studied the CeNi$_4$X (X = Al, Cu, Ga) compounds crystallizing in the hexagonal CaCu$_5$-type structure, space group $P6/mmm$ [9–12]. These compounds are of special interest due to the nearly filled Ni 3d band, implying a negligible contribution of Ni atoms to the resultant magnetic moment [9–11]. In the temperature dependence of electrical resistivity we have observed a shallow minimum for CeNi$_4$X below 20 K [9–12]. It has been ascribed to a Kondo-like behavior. Both the susceptibility and the x-ray photoemission (XPS) spectra show that Ce ions are in the intermediate valence state in CeNi$_4$X. In this paper we describe our studies of the structural (x-ray diffraction), magnetic (ac/dc magnetic susceptibility), thermodynamic (heat capacity), electrical transport and electronic structure (XPS, band structure calculations) properties of the CeCu$_4$Al compound.

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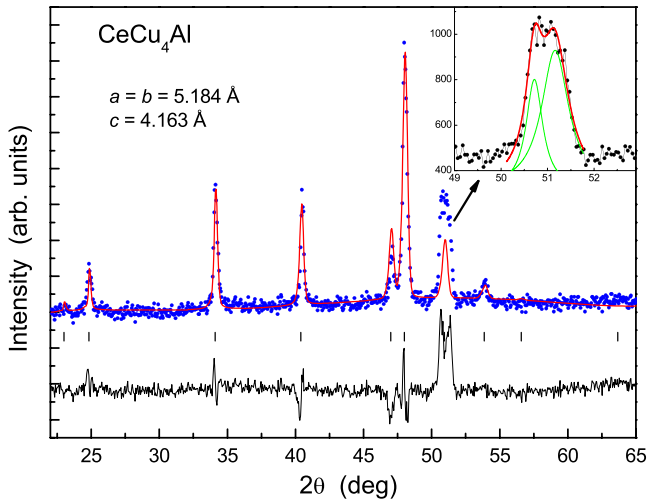
Figure 1. X-ray diffraction pattern (dots) for the $CeCu_4Al$ compound. Solid line: fitted with the program FULLPROF. Bottom solid line: difference between the experiment and the calculation. Inset: splitting for the (002) reflection.

2. Experimental details and method of calculations

The $CeCu_4Al$ compound was prepared by induction melting of stoichiometric amounts of the constituent elements in a water-cooled boat, under an argon atmosphere. The ingots were inverted and melted several times to ensure homogeneity. The crystal structure was established by a powder x-ray diffraction technique, using $Co\,K\alpha$ radiation (figure 1). The diffraction patterns have been fitted with the program FULLPROF, by the full-pattern Rietveld refinement.

The $CaCu_5$-type structure is confirmed (there is no indication of any impurity phase); however, a small distortion is probably present (see inset of figure 1) which results from the small splitting for the (002) reflection. It may be a source of the splitting in the Ce XPS 3d spectrum (see figure 6). The lattice constants are $a = 5.185$ Å and $c = 4.164$ Å.

Heat capacity measurements were carried out on the PPMS commercial device (Quantum Design) in the temperature range 0.3–300 K, by the relaxation method using the two-τ model. The magnetic susceptibility and the magnetization curves were measured on the MagLab 2000 System (Oxford Instruments Ltd).

The electrical resistivity was measured on a bar-shaped sample using a standard four-probe technique.

The x-ray photoemission spectra were obtained with an $Al\,K\alpha$ source using a PHI 5700/660 Physical Electronics Spectrometer. The energy spectra of the electrons were analyzed by a hemispherical mirror analyzer with the energy resolution of about 0.3 eV. The Fermi level $E_F = 0$ was referred to the gold 4f level binding energy at 84 eV. All emission spectra were measured immediately after breaking the sample in a vacuum of 10^{-10} Torr.

The electronic structure of $CeCu_4Al$ was calculated self-consistently by the spin-polarized tight binding linear muffin-tin orbital (TB LMTO) method in the atomic sphere approximation (ASA) with the combined correction term as

in [13]. For the exchange–correlation potential the von Barth–Hedin parameterization [14] has been employed. The Langreth–Mehl–Hu non-local correction was included [15].

In the calculations the initial atomic configurations were taken according to the periodic table of elements. We assume for Ce: $core(Xe) + 6s^25d^1\,4f^1$, for Cu: $core(Ar) + 3d^{10}4s^1$ and for Al: $core(Ne) + 3s^2p^1$. The self-consistent spin-polarized band calculations were performed for the experimental values of the lattice parameters. The primitive cell consists of six atoms, where the Ce atom occupies (1a) site, the Cu atoms are in the (2c) and (3g) positions, and the Al atom is in the (3g) position. The Brillouin zone integrations in the self-consistency loop were performed using the tetrahedron method [16] on a grid of about 1700 k-points.

The systematic errors due to the use of ASA get canceled out while determining the ground state energy differences such as the formation energy. Incorporating the so-called 'combined correction' one can partly solve the error due to the spheridization of the potential and the charge density [17, 18].

3. Results

A first step to characterize the $CeCu_4Al$ compound is to verify its magnetic properties. In figure 2(a) the temperature dependence of the inverse magnetic susceptibility $\chi^{-1}(T)$ is presented. These measurements did not reveal any magnetic transition. Above about 40 K $\chi^{-1}(T)$ follows the Curie–Weiss law with the effective magnetic moment $\mu_{eff} = 2.53\ \mu_B$ and the paramagnetic Curie temperature $\theta_p = -10$ K. The experimental value of μ_{eff} is nearly equal to the calculated one for a free Ce^{3+} ion $\mu_{eff} = g(j(j+1))^{-1/2} = 2.54\ \mu_B$, thus indicating the presence of well-localized magnetic moments carried by the stable Ce^{3+} ions. At low temperatures the $\chi^{-1}(T)$ curve deviates from the straight-line behavior, which may be partly due to the effects of the crystal electric field (CEF). However, the shape of the f band, especially its width, can also strongly influence the low temperature behavior of the magnetic susceptibility [19]. Therefore, a study of the electronic structure can be very important.

The negative paramagnetic Curie temperature is known [20] to enable a rough estimation of the Kondo temperature T_K as $T_K = |\theta_p/2| = 5$ K. In contrast, the isostructural $CeNi_4Al$ compound exhibits the Curie–Weiss behavior with a paramagnetic Curie temperature $\theta_p = -6.3$ K and the effective magnetic moment $\mu_{eff} = 0.6\ \mu_B$. This is a typical mixed-valence material [11].

The magnetic field dependences of the magnetization for $CeCu_4Al$ are presented in figure 2(b). The temperature evolution confirms the previous observations and the absence of any magnetic ordering down to 2 K.

Figure 3(a) shows the temperature dependence of the heat capacity $C(T)$ of $CeCu_4Al$ in the temperature range 0.3–300 K and in zero magnetic field. We have not observed any sign of magnetic order down to 0.3 K. In order to make comparisons with the previous results [4, 5, 8] we present in figure 3(b) the low temperature part of $C(T)/T$ as a function of T^2. An extrapolation to $T = 0$ K of the temperature range above 11 K yields a γ value of about 210 mJ K^{-2} mol^{-1}. Extrapolation
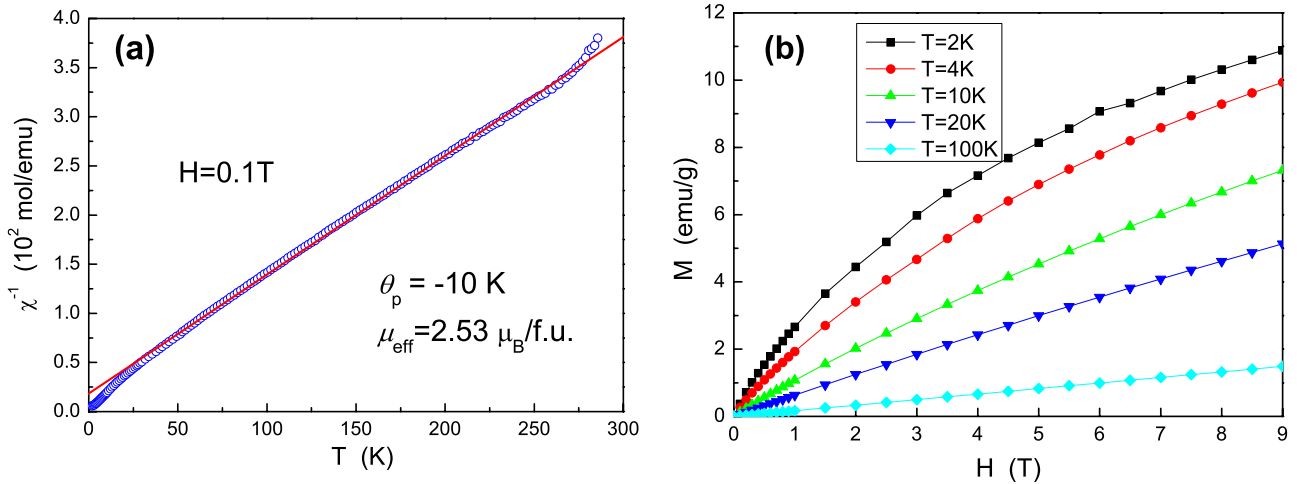
Figure 2. (a) The reciprocal magnetic susceptibility $\chi^{-1}(T)$ and (b) the magnetization $M(H)$ for the CeCu$_4$Al compound.
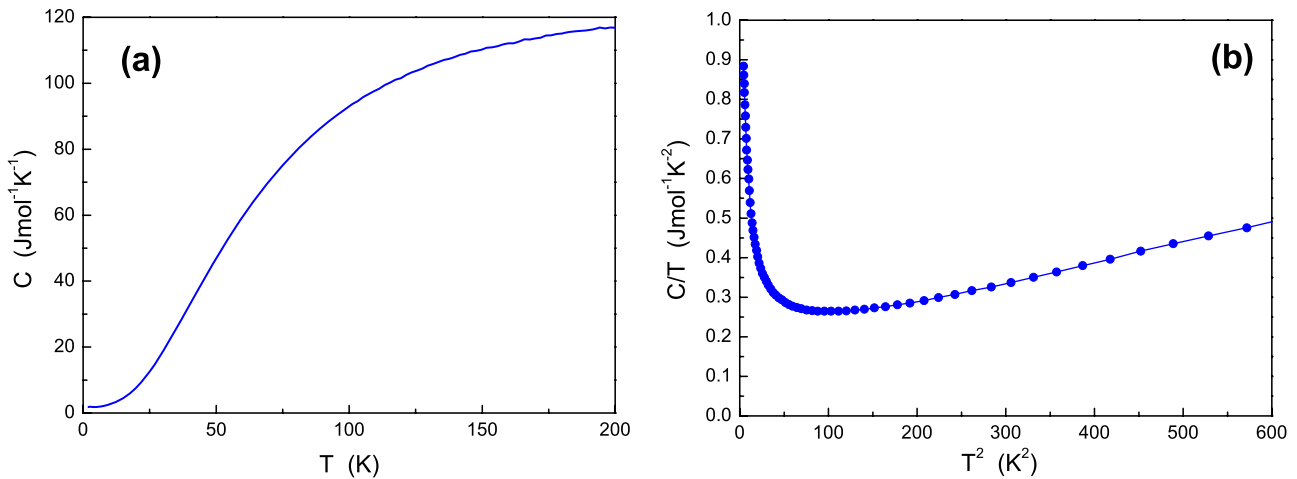
Figure 3. (a) The heat capacity $C(T)$ of the CeCu$_4$Al compound up to 200 K. (b) A low temperature part of the C/T versus T^2 dependence used to determine the γ value.

of the lowest temperature range of $C/T(T^2)$ provides a large value of 2.2 J K^{-2} mol^{-1}. In each case, these values confirm the presence of a heavy fermion state in this compound. This behavior is in qualitative agreement with [4, 5]. Our magnitudes of $C(T)$ and γ are smaller, except for the lowest temperature extrapolation. The Kondo temperature T_K derived from the standard relation $T_K = 0.68R/\gamma$ [21, 22] is equal to 2.6 K, assuming the value obtained from the lowest temperatures. For comparison CeNi$_4$Al was characterized by $\gamma = 29$ mJ K^{-2} mol^{-1} [9].

Cerium-based ternary compounds demonstrate different phenomena depending on the valence of the Ce ion. A key role is played by the hybridization between the conduction electrons and the 4f Ce electrons. A strong hybridization will decrease the localization of the Ce 4f electrons and result in a non-magnetic ground state with a mixed-valence behavior. A weak hybridization leads to localization of the 4f levels. XPS is a useful method to study these effects in rare-earth-based compounds. Figure 4 presents the XPS spectrum for CeCu$_4$Al in the whole measured range of binding energies. Only a

small contribution of oxides is present, which confirms the conclusion derived from x-ray diffraction on the good quality of the sample studied.

The valence band (figure 5) is similar to that observed for CeCu$_6$ [23]. A few humps due to the Ce 4f excitation spectrum are visible and are in agreement with those suggested in [23]. The Cu 3d band is located at about -3.5 eV. In the case of CuO, the main Cu 3d band is located at about -3.3 eV and the Zhang–Rice singlet level with the ^{1}A$_1$ symmetry is located at about -1.5 eV [24–26].

The x-ray photoemission spectrum of Ce 3d is shown in figure 6. A rather weak peak at -914 eV implies the occupation of the f level, n_f, close to 1, i.e. a localization of the f states. The most surprising observation is the splitting of the main peaks, which cannot be explained just by the contribution of the various Ce final states (f^0 and f^2) because the binding energy of these states and their intensity would be quite different. One can also exclude oxides, because their contribution has been found to be smaller than in many similar compounds we have studied previously [9–11]. This split
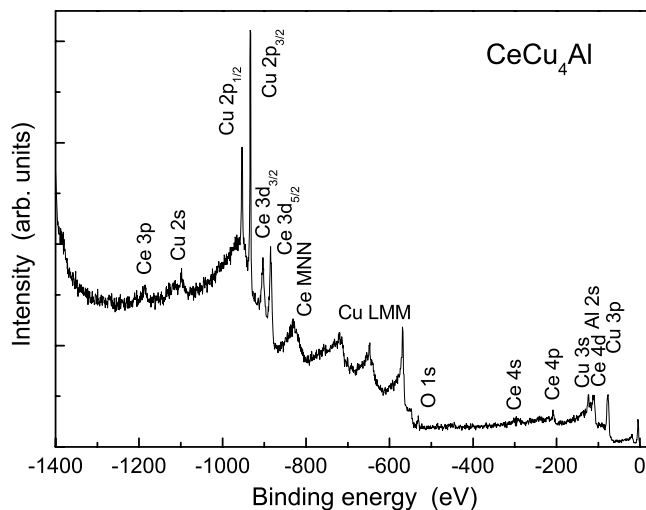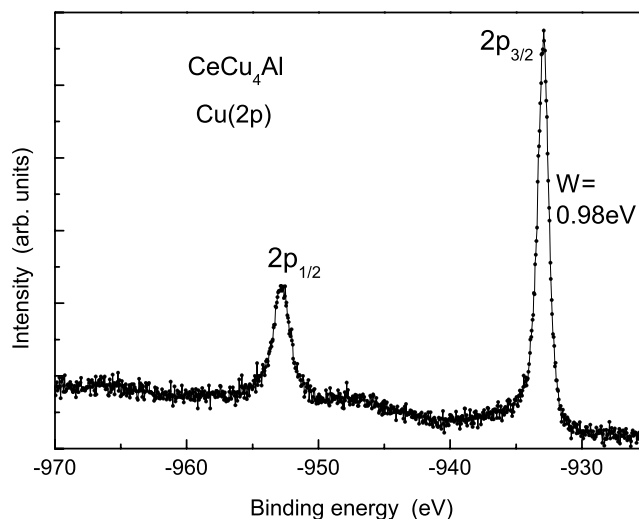
Figure 4. The XPS spectrum of CeCu₄Al in the energy range of 0–1400 eV.
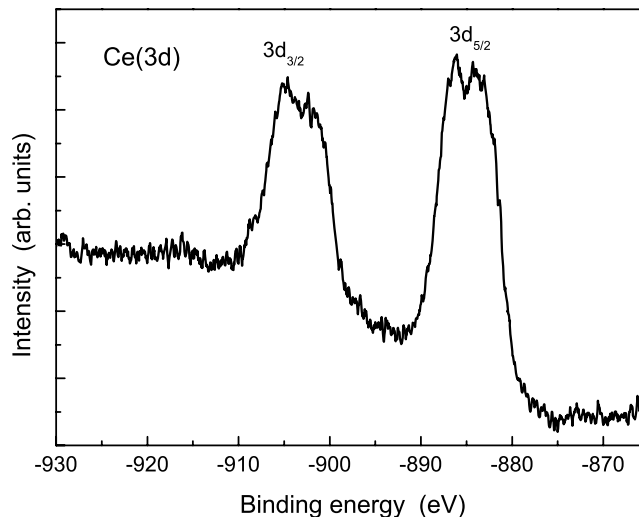
Figure 6. Experimental XPS spectrum of the Ce(3d$_{3/2,5/2}$) doublet.
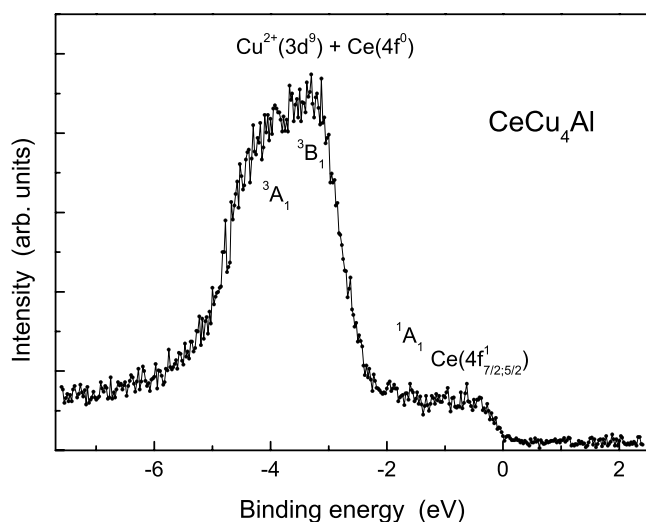
Figure 5. Valence band region of the CeCu₄Al compound measured by x-ray photoemission spectroscopy.

Figure 7. The spin–orbit split Cu(2p$_{1/2,3/2}$) doublet of the CeCu₄Al compound.

may be connected with the split displayed in figure 1, i.e. it may be due to a distortion of the crystallographic structure or a presence of crystallites with slightly different Ce locations leading to a chemical shift. The structural origin seems to be most probable; however, a final explanation remains a challenge. We do not observe a similar split in the case of any other CeNi₄X (X = Al, Cu, Ga) compounds.

The XPS spectrum of Cu(2p$_{1/2}$) and Cu(2p$_{3/2}$) is presented in figure 7. The peaks are narrow, e.g. in the case of the Cu 2p$_{3/2}$ peak the width is $W = 0.98$ eV. It was observed previously [24] that this peak for CuO and Cu₂O had a width of 3.4 eV and 1.9 eV, respectively. We obtain a binding energy of −932.95 eV, which is close to the result for CuO but the lack of peaks at about −943 eV resembles the case of Cu₂O [24]. Our observation of the sharp Cu 2p peaks may indicate that they correspond to pure copper.

The analysis of the Ce 4d spectrum (figure 8) is based on the results of Fuggle *et al* [27]. The two peaks at

−120 eV correspond exactly to the typical La spin–orbit split states, and therefore are ascribed to the 4d⁹f⁰ final states. Their appearance coincides with the presence of the peak at −914 eV for the Ce 3d spectrum (see figure 6). However, both the doublet at −120 eV and the peak at −914 eV are of small intensity. Therefore we conclude that the mixed-valence behavior is not strong in the studied CeCu₄Al compound. The 4d⁹f¹ peaks are typical of the Ce metal and the structure between −105 and −115 eV is due to the multiplet interactions [27]. Both the 3d and 4d XPS spectra suggest a localized character of the f states in this compound.

In cerium intermetallic compounds the itinerant electrons of the transition metal play a significant role in the hybridization of the Ce 4f states with the 3d bands of the transition element at the Fermi level. In our previous studies of the CeNi₄X (X = Al, Ga, Cu, Si) series we found an increase of the degree of hybridization of Ce 4f with Ni 3d bands for X = Cu compared to compounds including the non-transition
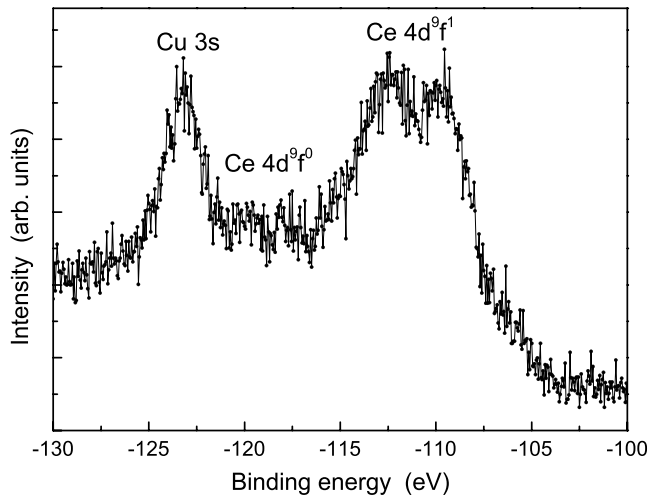
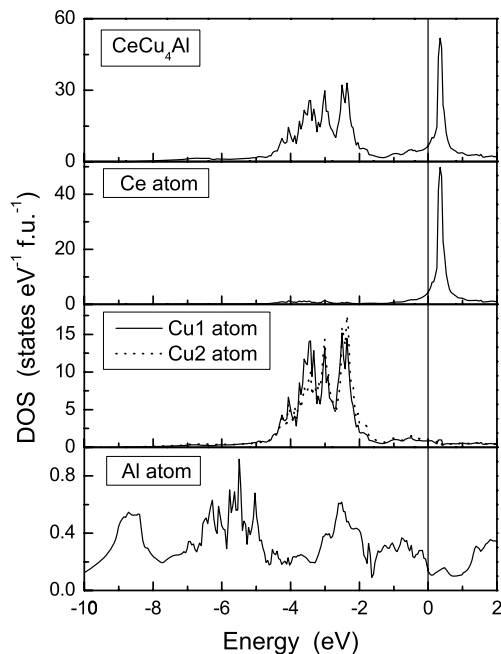**Figure 8.** The measured XPS spectrum of Ce(4d) and Cu(3s).

Figure 10. The band structure of CeCu$_4$Al along various symmetry directions.

Figure 9. The calculated densities of states for CeCu$_4$Al: (a) the total DOS; (b) the contribution of Ce atoms; (c) the contribution of Cu(2c) and Cu(3g) atoms; (d) the contribution of Al atoms.

elements (Al, Ga, Si). In CeNi$_4$Cu the presence of the filled 3d shell of copper leads to the f electrons more hybridized with the 3d electrons of Ni [28, 29]. It is, therefore, interesting to verify the predictions of the electronic structure calculations for CeCu$_4$Al. This type of calculation is not designed for heavy fermion materials but can provide useful information on the partial contributions of the elements to the electronic structure and on the hybridization effects, especially by comparison with the previously studied members of the CeNi$_4$X series.

The results of the total energy calculations have shown that the ground state is paramagnetic for this compound. The calculated total densities of states (DOS) and the contribution of the Ce, Cu and Al atoms to the total DOS for CeCu$_4$Al are presented in figures 9(a)–(d). Copper as a transition metal with
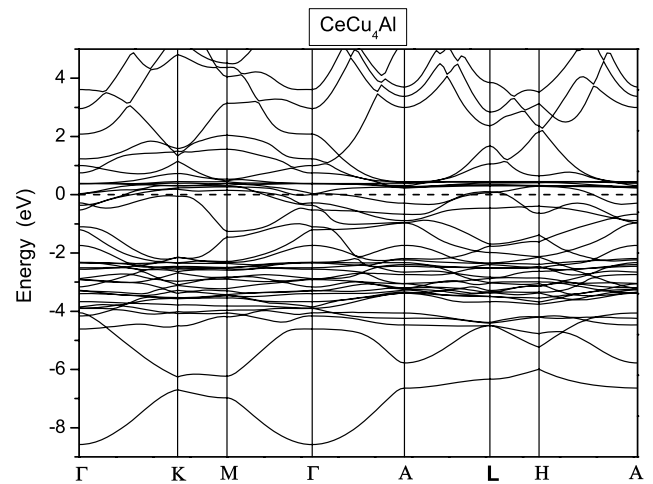
a fully occupied 3d shell influences the electronic structure of CeCu$_4$Al in a particular way. The middle part of the total DOS in the energy region from −5 to −2 eV corresponds mainly to the d states of Cu atoms in positions (2c) and (3g). However, the 4f states of Ce are close to the Fermi level. The itinerant band of copper and the 4f level of Ce move away from each other. The distance between the centers of gravity of 4f Ce and 3d of Cu in (2c) and (3g) positions is 1.72 eV and 1.58 eV, respectively. The DOS pictures can suggest a low hybridization of the 3d and 4f states. The electronic states of the Ce atom are more localized in CeCu$_4$Al than in the earlier studied CeNi$_4$Al, where the hybridization of the 3d states of Ni and 4f states of Ce was observed in the XPS experiments as well as proved by the *ab initio* studies [28]. For CeNi$_4$Al the distance between the centers of gravity of the Ce 4f and Ni 3d bands was found to be about 0.514 eV. It is clear that the filled Cu 3d states are mainly responsible for the differences between the CeCu$_4$Al and CeNi$_4$Al compounds. A dramatic difference between these compounds is also evident from the comparison of the XPS valence bands. For CeNi$_4$Al the main peaks are located between −1 and −2 eV, whereas for CeCu$_4$Al they appear between −3 and −5 eV.

A small difference in the shape of peaks of the Cu atoms observed in the region from −5 to −3.5 eV is associated with a difference in the neighborhood of the (2c) and (3g) sites. The contribution of Al atoms to the total DOS is small in the presented region of energy (from −10 to 2 eV).

The band structure of CeCu$_4$Al along various symmetry directions is shown in figure 10. The separation of the lowest band derived from the s states of Al and the bands which participate in conductivity has only a direct character. The whole separation between the low lying electronic states and the valence band is not found in the calculation. The bottom of the lowest band has a parabolic shape (below −7 eV) around the center of the Brillouin zone, i.e. in the directions Γ–M, Γ–A and Γ–K. The group of bands formed between −5 and −3.5 eV is dominated mainly by the 3d states of the Cu atoms. The Fermi level is crossed by the bands, which have mostly the Ce 4f character. The Ce f states show a flat dispersion in
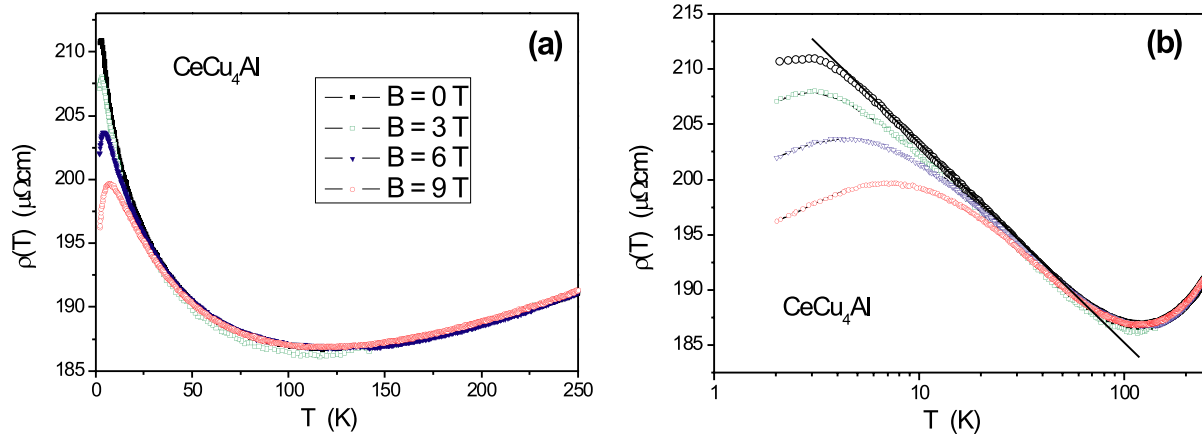
Figure 11. (a) Temperature dependence of the electrical resistivity. (b) Resistivity in the logarithmic scale.

the vicinity of the Fermi level. In the high symmetry points A, H and in the direction K–M the bands do not cross the Fermi level.

The temperature variation of the electrical resistivity of $CeCu_4Al$ as a function of the magnetic field is shown in figure 11(a). The resistivity decreases from room temperature down to about $T_{min} = 100$ K, and then increases with decreasing temperature down to 2.5 K. This behavior is attributed to the single-site Kondo scattering caused by the localized 4f electron of the Ce^{+3} ion. The resistivity begins to decrease below 2.5 K, which is better visible after application of the magnetic field, which shifts the peak position to 7.3 K for $H = 9$ T. The temperature dependence of resistivity is characteristic of the Kondo-lattice formation, in which the 4f electrons behave as heavy quasi-particles at $T < T_K$. For such materials the resistivity can be described below the minimum by the formula

$$\rho(T) = (\rho_0 + \rho_0^{\infty}) - c_K \ln T \qquad (1)$$

where the first term accounts for the scattering of the conduction electrons on the lattice defects and disordered magnetic moments and the second term represents the spin-flip scattering of the conduction electrons on the magnetic centers (Kondo effect). This dependence is better visible on the logarithmic scale in figure 11(b). Applying equation (1) to the experimental data of $CeCu_4Al$ yields a good approximation of the $\rho(T)$ curve between 5 and 60 K (note the solid line in figure 11(b)) with the fitting parameters $(\rho_0 + \rho_0^{\infty}) = 221.5\ \mu\Omega$ cm and $c_K = 7.95\ \mu\Omega$ cm. In contrast, the electrical resistivity of $CeNi_4Al$ compound was metallic. Below 16 K a shallow minimum in $\rho(T)$, typical of a Kondo impurity system was observed. The increased ρ_0 (500 $\mu\Omega$ cm) was mainly due to the random distribution of Ni(2) and Al on the (3g) site, as has been also observed for other $CeNi_4X$ (X = Cu, B) compounds [10, 11].

It should be noticed that a similar temperature dependence of the resistivity as in figure 11(a) can also be obtained from the calculations carried out by Panwar and Singh [30] based on the periodic Anderson model and by manipulating the position of the f level and the Coulomb interaction U.

We have observed that above 25 K there is no important influence of the magnetic field on $\rho(T)$. On the contrary, as displayed in figure 11, the low temperature resistivity of $CeCu_4Al$ is very sensitive to the applied magnetic field. As we already mentioned, the maximum is shifting to higher temperatures with increasing magnetic field. Besides, one can see the magnitudes decreasing with the increasing magnetic fields. It means that the magnetoresistivity is negative ($(\rho(H) - \rho(0))/\rho(0) < 0$), which is characteristic of the Kondo systems, and the value obtained in $H = 9$ T is equal to about 5% at 4 K. This value of magnetoresistivity is typical of heavy fermion systems [1].

The presented magnetic susceptibility, specific heat and resistivity measurements suggest that the Kondo temperature takes a value from the range 2.5–5 K. It allows for the estimation of the coherence temperature, which can be expected to be between 0.4 and 0.8 K based on the relation $T_{coh} = T_K/N$, where N is the degeneracy [31].

4. Conclusions

It has been observed that $CeCu_4Al$ is paramagnetic and follows the Curie–Weiss law with the effective magnetic moment $\mu_{eff} = 2.53\ \mu_B$/f.u., which corresponds well to the free Ce^{3+} ion value. The paramagnetic Curie temperature is $\theta_p = -10$ K. A typical heavy fermion behavior has been observed with a γ value of about 210 mJ K^{-2} mol^{-1} obtained from $C(T)/T$ extrapolation of the temperature range above 11 K down to $T = 0$ K. Extrapolation of the lowest temperature range yields the value of 2.2 J K^{-2} mol^{-1}. Both the 3d and 4d XPS spectra suggest a localized character of the f states in this compound. The valence state of the Ce ion is close to 3+. The electrical resistivity $\rho(T)$ of $CeCu_4Al$ shows a Kondo-like logarithmic increase up to a maximum at $T = 2.5$ K and then a steep decrease towards low temperatures. The Kondo temperature estimated from the magnetic susceptibility, specific heat and resistivity measurements is about 2.5–5 K.

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