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Kadowaki–Woods plot of exchange-enhanced Pauli paramagnetic Laves phase quasi-binary compounds $Lu(Co_{1-x}M_x)_2$

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Abstract

The electrical resistivity and low-temperature specific heat of exchange-enhanced Pauli paramagnetic Laves phase quasi-binary compounds $Lu(Co_{1-x}M_x)_2$ (M = Al, Ga and Si) have been investigated in order to discuss the spin fluctuation characteristics and the Kadowaki–Woods plot. In these three kinds of quasi-binary system, both the electrical resistivity coefficient A and the specific heat coefficient γ_{tot} at low temperatures decrease at first and then significantly increase with increasing x. The concentration dependence of magnetic susceptibility at 0 K, $\chi(0)$, is also similar to that of A and γ_{tot}. The Wilson ratio becomes very large because of the enhancement of magnetic susceptibility. The Kadowaki–Woods plot is fitted to the same universal line of heavy-fermion compounds, A15-type Nb_3Sn and V_3Si compounds because of significant spin fluctuations in the present quasi-binary compound systems.

1. Introduction

Spin fluctuations influence various magnetic and electrical properties of exchange-enhanced Pauli paramagnetic Laves phase compounds such as $ScCo_2$, YCo_2 and $LuCo_2$ [1, 2]. Metamagnetic transition behaviours of Laves phase quasi-binary compounds such as $Y(Co_{1-x}Al_x)_2$ [3], $Lu(Co_{1-x}Al_x)_2$ [4, 5], $Lu(Co_{1-x}Ga_x)_2$ [6, 7], $Lu(Co_{1-x}Sn_x)_2$ [8, 9] and $Lu(Co_{1-x}Si_x)_2$ [9, 10] have been investigated extensively. The relation between the critical field of the itinerant-electron metamagnetic transition and the magnetic susceptibility maximum has been investigated systematically [3, 11]. Furthermore, the magnetovolume effects have been discussed within the framework of spin fluctuations [11–15].

The significant temperature dependence of electrical resistivity ρ of the exchange-enhanced Pauli paramagnetic Laves phase compounds has been inferred to be caused by spin

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fluctuations, because spin fluctuations contribute to the transport phenomena. That is, spin fluctuations scatter conduction electrons through the exchange interactions. Low-temperature resistivity is dominated by the quadratic increase due to the spin fluctuation scatterings.

The low-temperature specific heat is also affected by spin fluctuations, accompanied by the enhancement of the specific heat coefficient γ_{tot}. Kadowaki and Woods [16] pointed out universality between the coefficient A of the quadratic increase in low-temperature resistivity ρ and the low-temperature coefficient γ_{tot} for heavy-fermion compounds. Later on, Miyake *et al* [17] discussed the relation between the resistivity ρ and the effective mass for heavy-fermion compounds, A15-type Nb_3Sn and V_3Si compounds and ordinary transition metals, and reported that the Kadowaki–Woods plot of the former two systems is located on the same straight line. They also pointed out that the sharp peak structure in the density of states near the Fermi level in A15-type compounds is due to many-body dynamical effects as in heavy-fermion compounds. Wada *et al* [18] explained the Kadowaki–Woods plot for $Hf_{1-x}Ta_xFe_2$ within the framework of significant spin fluctuations. Gratz *et al* [2] applied the Kadowaki–Woods plot to exchange-enhanced Pauli paramagnetic Laves phase YCo_2, $LuCo_2$, $ScCo_2$ and demonstrated that the data of these compounds fitted to the same universal line of heavy-fermion compounds. More recently, a full-dress discussion of the Kadowaki–Woods plot for heavy-fermion systems has been carried out on the basis of the spin fluctuation theory [19].

Laves phase quasi-binary compounds $Lu(Co_{1-x}Al_x)_2$, $Lu(Co_{1-x}Ga_x)_2$ and $Lu(Co_{1-x}Si_x)_2$ have been investigated by several researchers. However, it should be pointed out that metamagnetic transition characteristics and electric properties are remarkably influenced by not only the spin fluctuations but also the compositional inhomogeneity [12, 20]. Therefore, well homogenized specimens are highly necessary for the investigations of the Kadowaki–Woods plot. By using well homogenized Laves phase quasi-binary compounds $Lu(Co_{1-x}Al_x)_2$ and $Lu(Co_{1-x}Ga_x)_2$, we have established the magnetic phase diagrams [21], and demonstrated that these phase diagrams are in good agreement with the theoretical magnetic phase diagram established within the framework of spin fluctuations [22]. In the present paper, the low-temperature electrical resistivity, specific heat and magnetic susceptibility have been investigated for well homogenized Laves phase quasi-binary compounds $Lu(Co_{1-x}M_x)_2$ (M = Al, Ga and Si) in order to discuss the spin fluctuation characteristics and the Kadowaki–Woods plot.

2. Experiment

The quasi-binary specimens were prepared by arc-melting in an argon gas atmosphere. In order to avoid any other ferromagnetic precipitates such as $LuCo_3$ through the loss of Lu during alloying, the Lu composition was kept slightly higher than the stoichiometric composition. The appropriate conditions for homogenization were confirmed as annealing at 1273 K for a week in a similar way as in our previous study [12, 20]. Therefore, the specimens were homogenized under the same conditions in evacuated quartz tubes, followed by quenching into water. The oxidized surface of the annealed specimen was mechanically removed.

The crystal structure was identified by x-ray diffraction as a C15-type Laves phase without any other phases. The metallographic structure was examined by a scanning electron microscope (SEM) in order to confirm the grain boundaries. The compositional homogeneity was confirmed from the composition profiles obtained with an electron probe micro-analyser (EPMA). The analysed compositions were very close to the compositions given by $Lu(Co_{1-x}Al_x)_2$, for example.

The temperature dependence of electrical resistivity was measured by a four-terminal method and the low-temperature specific heat was measured by a relaxation method.

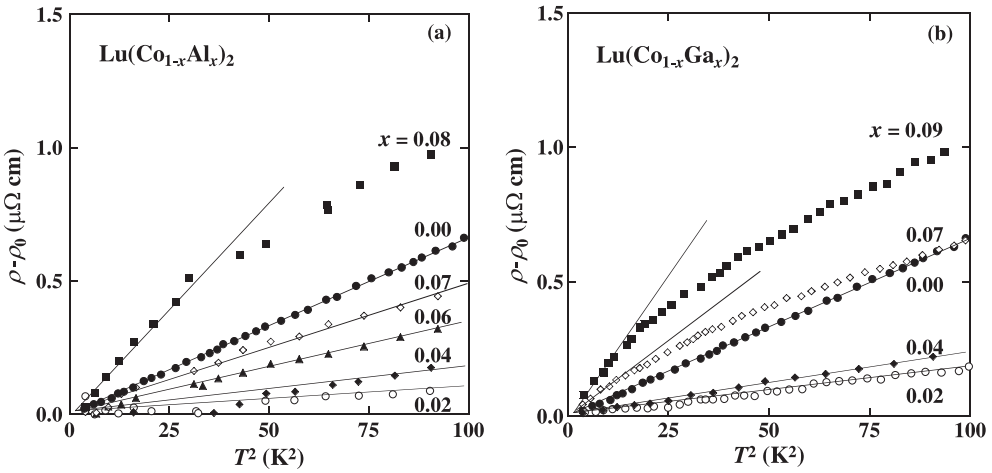

Figure 1. The $(\rho - \rho_0)$ versus T^2 plots as a function of temperature for Laves phase quasi-binary compounds. (a) $Lu(Co_{1-x}Al_x)_2$ and (b) $Lu(Co_{1-x}Ga_x)_2$.

The temperature dependence of the magnetic susceptibility $\chi(T)$ was measured with a superconducting quantum interference device (SQUID) magnetometer.

3. Results and discussion

From our detailed heat-treatments for Laves phase quasi-binary compounds, it is clear that the annealing conditions given in many previous papers are incomplete [12, 20], that is, the single phase identified by x-ray diffractions does not always mean the compositional homogeneity. The compositional inhomogeneity becomes significant around grain boundaries, and the inhomogeneity brings about significant effects on the magnetic properties. For example, the critical field of the metamagnetic transition is sensitively affected by annealing conditions [12, 20]. Therefore, the compositional homogenization is the prerequisite condition to investigate physical properties associated with the spin fluctuations. After performing appropriate heat-treatment for homogenization, we have carried out the measurements of low-temperature electrical resistivity and specific heat.

In the paramagnetic state, the total electrical resistivity $\rho_{\rm tot}(T)$ is given by assuming the Matthiessen rule as [23]

$$\rho_{\rm tot}(T) = \rho_0 + \rho_{\rm ph} + \rho_{\rm sf}, \tag{1}$$

where the subscripts 0, ph and sf denote the contributions due to impurity, phonon and spin fluctuation scatterings, respectively. In a variety of systems, the electrical resistivity is well known to behave as the following expression in the limit $T \to 0$, because the contribution from the $\rho_{\rm ph}$ term is negligibly small at such low temperatures.

$$\rho(T) - \rho_0 = \rho_{\rm sf}(T) \cong AT^2. \tag{2}$$

The coefficient in equation (2) is directly correlated with the spin fluctuation characteristics [19]. Figures 1(a) and (b) show the $(\rho - \rho_0)$ versus T^2 plots for $Lu(Co_{1-x}Al_x)_2$ and $Lu(Co_{1-x}Ga_x)_2$, respectively. The temperature range of the linear portion becomes narrower with increasing x. The value of the coefficient A, that is, the slope of the linear portion, decreases at first and increases with increasing x. The concentration dependence of the

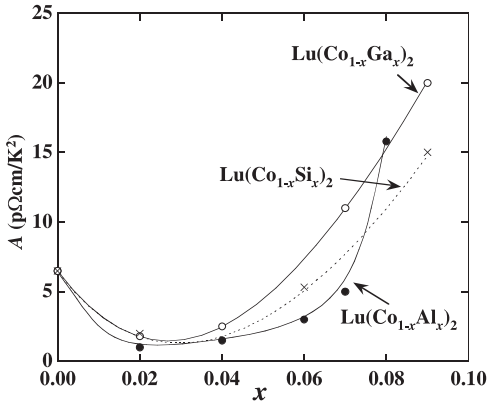

Figure 2. Concentration dependence of the coefficient A for Laves phase quasi-binary compounds $Lu(Co_{1-x}Al_x)_2$, $Lu(Co_{1-x}Ga_x)_2$ and $Lu(Co_{1-x}Si_x)_2$ systems.

coefficient A for Laves phase quasi-binary compounds $Lu(Co_{1-x}Al_x)_2$, $Lu(Co_{1-x}Ga_x)_2$ and $Lu(Co_{1-x}Si_x)_2$ is given in figure 2. The point to observe is that the coefficient A for the present three kinds of quasi-binary system does not show a monotonic increase with increasing x.

Figures 3(a)–(c) show the low-temperature specific heat data for $Lu(Co_{1-x}Al_x)_2$, $Lu(Co_{1-x}Ga_x)_2$ and $Lu(Co_{1-x}Si_x)_2$ systems, respectively. The specific heat coefficient γ_{tot} was estimated by a linear extrapolation of conventional plots in the form C/T versus T^2. As seen from the figure, the magnitude of γ_{tot} sensitively depends on the concentration x. The concentration dependence of the specific heat coefficient γ_{tot} for Laves phase compounds $Lu(Co_{1-x}Al_x)_2$, $Lu(Co_{1-x}Ga_x)_2$ and $Lu(Co_{1-x}Si_x)_2$ is given in figure 4. It should be noted that the present data for $LuCo_2$ are very different from the previous data [24, 25]. The total value of the specific heat coefficient γ_{tot} is given by the following expression:

$$\gamma_{tot} = \gamma_{band} + \gamma_{sf}, \tag{3}$$

where γ_{band} and γ_{sf} are the band and the spin fluctuation terms, respectively.

The term γ_{sf} is given by [26]

$$\gamma_{sf} = \frac{k_B^2}{2h\Gamma_0} \ln(1 + Dq_B^2\chi(0)), \tag{4}$$

where k_B and h are the Boltzmann and the Planck constants, respectively, and $\chi(0)$ is the magnetic susceptibility at 0 K. The parameters q_B and Γ_0 are respectively correlated to the upper limit of wavenumber and the width of the frequency distribution of spin fluctuations. The coefficient D gives the dispersion between the frequency ω and the wavenumber q as $\omega = Dq^2$. Since $\chi(0)$ becomes larger on approaching the ferromagnetic state, the value of γ_{sf} is significantly enhanced. The values of γ_{band} for YCo_2, $LuCo_2$ and $ScCo_2$ are 13.7, 12.7 and 11.4 mJ mol^{-1} K^{-2}, respectively [27]. On the other hand, the experimental values of γ_{tot} for YCo_2 [28], $LuCo_2$ (see figure 4) and $ScCo_2$ [29] are 36.2, 32.6 and 18.4 mJ mol^{-1} K^{-2}, respectively. Therefore, the values of γ_{tot} are much larger than those of the band term γ_{band}. In addition, it should be noted that the sequence of magnitude of γ_{band} is similar to that of γ_{tot}. The values of γ_{tot} for the present compounds are also expected to be large because of a large contribution from spin fluctuations. With increasing x, however, γ_{tot} decreases at first and then increases in analogy with the coefficient A given in figure 2. As for this point, Khmelevski *et al* [30] have recently carried out band-structure calculations for Y $(Co_{1-x}Al_x)_2$ within the density functional theory and the local density approximation (DFT-LDA), and demonstrated similar

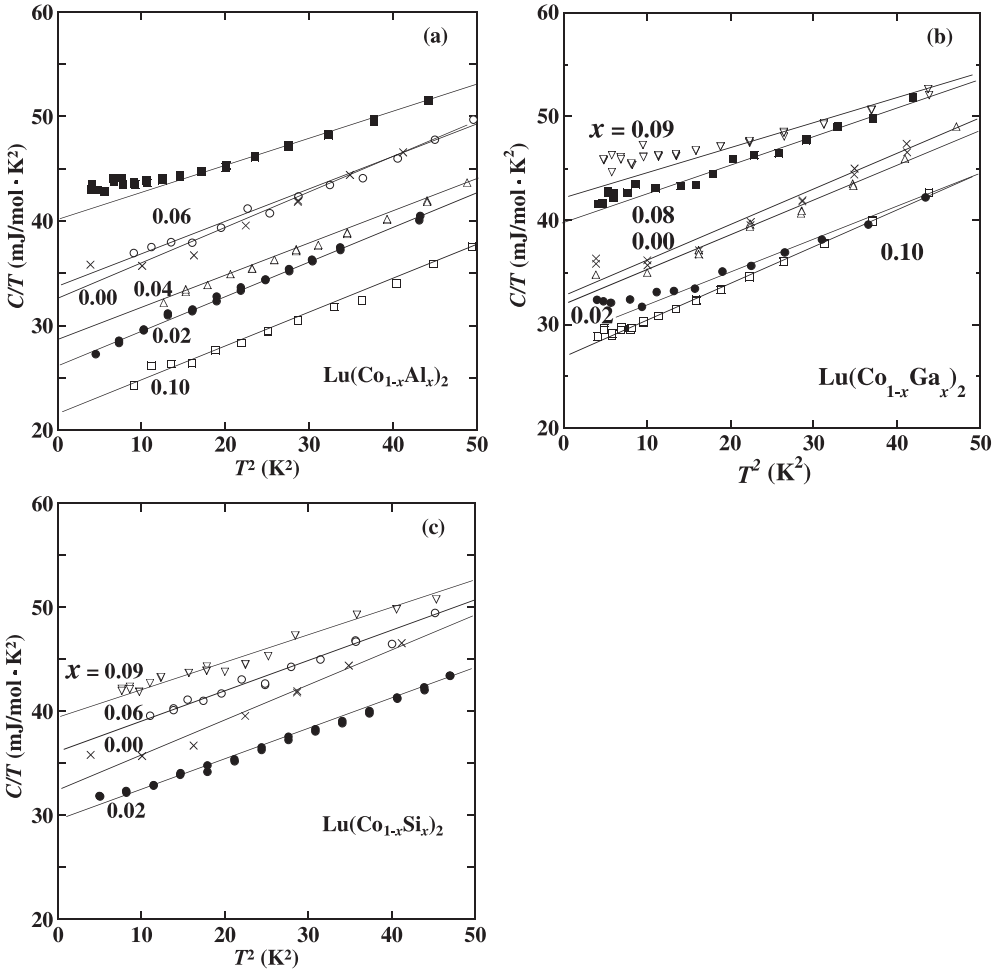

Figure 3. Low-temperature specific heats in the form of C/T versus T^2 for three kinds of Laves phase quasi-binary compound system. (a) $Lu(Co_{1-x}Al_x)_2$, (b) $Lu(Co_{1-x}Ga_x)_2$ and (c) $Lu(Co_{1-x}Si_x)_2$.

concentration dependences of both partial and total densities of states, though the composition where the density of states at the Fermi surface becomes a minimum is slightly different from the results of the present compound systems. Accordingly, the present experimental results and the theoretical calculations mentioned above for the concentration dependences are strikingly different from previous experimental results for $Y(Co_{1-x}Al_x)$ [31, 32] and $Lu(Co_{1-x}Ga_x)$ [6]. Furthermore, $Lu(Co_{1-x}Al_x)_2$ and $Lu(Co_{1-x}Ga_x)_2$ exhibit a pronounced increase in the vicinity of the composition just before the onset of ferromagnetism. The results of $Lu(Co_{1-x}Si_x)_2$ are similar to those mentioned above, except for the data in high concentrations. It should be pointed out that the $Lu(Co_{1-x}Si_x)_2$ compound system exhibits no ferromagnetic behaviours in the whole concentration range investigated [33].

From equation (4), the concentration dependence of $\chi(0)$ is expected to be similar to that of γ_{tot}, regardless of the strength of correlations. Figure 5 shows the concentration dependence of magnetic susceptibility at 0 K, $\chi(0)$, for Laves phase quasi-binary compounds $Lu(Co_{1-x}Al_x)_2$,

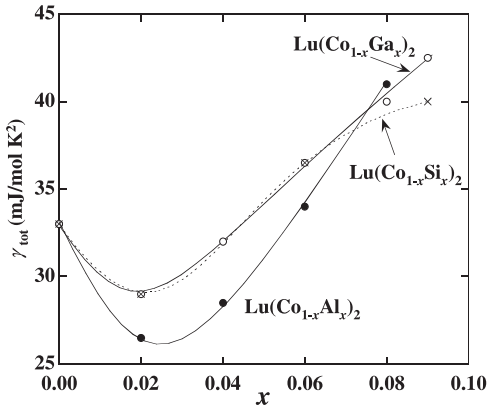

Figure 4. Concentration dependence of the low-temperature specific heat coefficient γ_{tot} for Laves phase quasi-binary compounds $Lu(Co_{1-x}Al_x)_2$, $Lu(Co_{1-x}Ga_x)_2$ and $Lu(Co_{1-x}Si_x)_2$.

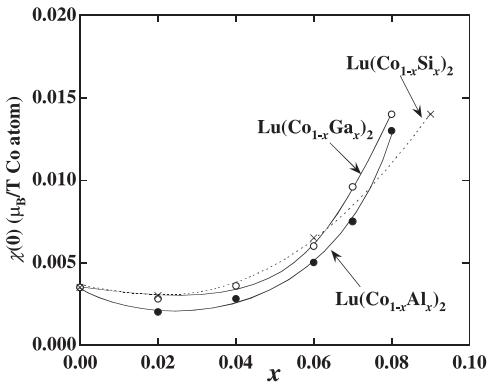

Figure 5. Concentration dependence of magnetic susceptibility at 4.2 K, $\chi(0)$, for Laves phase quasi-binary compounds $Lu(Co_{1-x}Al_x)_2$, $Lu(Co_{1-x}Ga_x)_2$ and $Lu(Co_{1-x}Si_x)_2$.

$Lu(Co_{1-x}Ga_x)_2$ and $Lu(Co_{1-x}Si_x)_2$. The values at 0 K were extrapolated from 4.2 K for the temperature dependence curve of $\chi(T)$. The concentration dependence $\chi(0)$ given in figure 5 is similar to that of γ_{tot} given in figure 4. That is, the concentration dependence of $\chi(0)$ exhibits a minimum at low concentration ranges. Although no detailed discussion has been given, there are glimpses of the same behaviour in available data in published papers. Concretely, similar results at a low concentration range have been confirmed in the temperature dependence curve of magnetic susceptibility for $Y(Co_{1-x}Al_x)_2$ [34] and in the concentration dependence of low-temperature specific heat coefficient for $Lu(Co_{1-x}Al_x)_2$ [26] and $Y(Co_{1-x}Al_x)_2$ [35].

The relation between the magnetic susceptibility and the specific heat coefficient is given by the following Wilson ratio [36]:

$$R_W = \frac{\pi^2 k_B^2}{3\mu_B^2}\left\{\frac{\chi(0)}{\gamma_{tot}}\right\} \tag{5}$$

where μ_B is the Bohr magneton. In the free-electron system, the susceptibility χ_{free} and the electronic coefficient of the specific heat γ_{free} are related to the density of states $N(E)$ around the Fermi energy E_f as $N(E_f)\mu_B^2$ and $(\pi^2/3)k_B^2\rho(E_f)$, respectively, resulting in $R_W = 1$. The ratio R_W is enhanced in heavy-fermion systems due to the electron correlations and

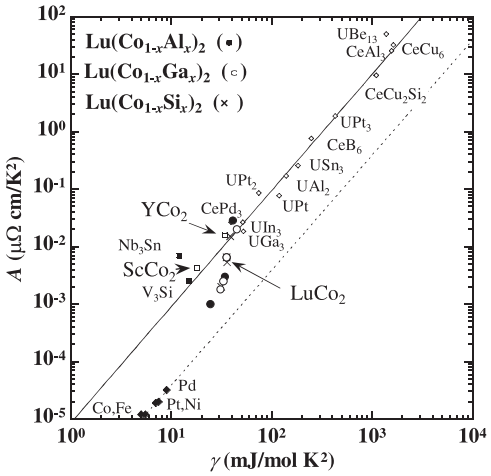

Figure 6. Kawowaki–Woods plot of $Lu(Co_{1-x}Al_x)_2$, $Lu(Co_{1-x}Ga_x)_2$ and $Lu(Co_{1-x}Si_x)_2$ Laves phase quasi-binary compounds, together with those of heavy-fermion compounds [16], A15-type Nb_3Sn, V_3Si compounds [17] and ordinary transition metals [17].

the value of R_W becomes two at the strong-correlation limit in the Anderson model [37]. On the other hand, R_W is about 12 for $LuCo_2$, and R_W becomes larger on coming close to the onset of ferromagnetism for $Lu(Co_{1-x}Al_x)_2$, $Lu(Co_{1-x}Ga_x)_2$ and $Lu(Co_{1-x}Si_x)_2$ compounds. Concretely, R_W for these three kinds of compound with $x = 0.06$ and 0.09 is about 20 and 40, respectively, being much larger than $R_W = 2$. It is well known that the susceptibility $\chi(0)$ is enhanced by exchange interaction in 3d electron systems, and expressed as

$$\chi(0) = \frac{\chi_{\text{free}}}{1 - I}, \tag{6}$$

where $1 - I$ is the Stoner enhancement factor. Since the value of $1 - I$ comes close to zero on approaching the ferromagnetic state, the value of $\chi(0)$ exhibits a divergent increase. On the other hand, the value of γ_{sf} is proportional to $\ln(1 - I)$ because of a relatively large value of $\chi(0)$ in equation (4). Furthermore, the value of Γ_0 obtained from NMR measurements for $Y(Co_{1-x}Al_x)_2$ becomes larger around the critical concentration of onset of the ferromagnetism [38]. Therefore, the increase in γ_{sf} as a function of $1 - I$ is very modest compared with that in $\chi(0)$. Consequently, the Wilson ratio R_W becomes much larger than two, in analogy with the previous discussion for $Y(Co_{1-x}Al_x)_2$ Laves phase compounds [38].

The data positions in the Kadowaki–Woods plot for Laves phase quasi-binary compounds $Lu(Co_{1-x}Al_x)_2$ (•), $Lu(Co_{1-x}Ga_x)_2$ (○) and $Lu(Co_{1-x}Si_x)_2$ (×) are given in figure 6, together with the data of various heavy-fermion compounds, typical A15-type Nb_3Sn and V_3Si compounds [16, 17] and Laves phase binary compounds YCo_2 and $ScCo_2$ [2]. In addition, the positions of several kinds of ordinary transition metal are given in the same figure [17], for comparison. The present results fall into the former case given by the straight solid line with $A/\gamma_{tot}^2 = 1.0 \times 10^{-5}$ $\mu\Omega$ cm $K^{-2}/(mJ\ mol^{-1}\ K^{-2})^2$. In this connection, the value of A/γ_{tot}^2 of the latter case given by the dotted line is contrastively 0.4×10^{-6} $\mu\Omega$ cm $K^{-2}/(mJ\ mol^{-1}\ K^{-2})^2$. Therefore, the values of the former compounds are about 25 times larger than the latter ordinary transition metals. Such large values mentioned above are closely correlated with the spin fluctuations discussed by Takimoto and Moriya [19] as well as a sharp peak structure in their density of states near the Fermi level [17]. Such a sharp peak with a positive curvature of the density of states at the Fermi level [39] under the condition

of a negative coefficient of the fourth-order term in the Landau expansion [40–42] is also necessary for the metamagnetic transition in $Lu(Co_{1-x}Al_x)_2$ [12] and $Lu(Co_{1-x}Ga_x)_2$ [11].

4. Conclusion

Electrical resistivity and low-temperature specific heat have been investigated for well homogenized exchange-enhanced Pauli paramagnetic Laves phase quasi-binary compounds $Lu(Co_{1-x}Al_x)_2$, $Lu(Co_{1-x}Ga_x)_2$ and $Lu(Co_{1-x}Si_x)_2$ in order to discuss the contribution from spin fluctuations. It has been demonstrated that the concentration dependence of physical quantities exhibits a broad minimum at low concentrations in contrast to previous published data. In the Kadowaki–Woods plot, the present data have been compared with available data and it has been confirmed that the present quasi-binary compound systems are classified into the type of heavy-fermion compounds. Main results are summarized as follows.

(1) In the concentration dependence curves, the coefficients of low-temperature electrical resistivity A and the specific heat coefficient γ_{tot} for three kinds of Laves phase quasi-binary compound system exhibits a broad minimum at low concentrations.
(2) The magnetic susceptibility at 0 K, $\chi(0)$, for three kinds of Laves phase quasi-binary compound system also takes a broad minimum in the concentration dependence curves.
(3) The Wilson ratio is extremely large due to the large value of magnetic susceptibility associated with the Stoner enhancement factor.
(4) The Kadowaki–Woods plot is fitted to the same universal straight line for heavy-fermion compounds, implying that there are significant spin fluctuation effects with a sharp peak in the density of states near the Fermi level in the present Laves phase quasi-binary compounds.

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