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Magnetostriction of ytterbium-based Kondo compounds YbXCu₄ (X = In, Ag and Au)

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Abstract. Magnetostrictions were measured for polycrystalline YbXCu₄ (X = In, Ag and Au) by the three-terminal capacitance method up to the magnetic field of 8 T in the temperature range 5–100 K. The volume magnetostrictions of all of the compounds are negative in contrast to the case for Ce-based Kondo systems. The absolute values of the volume magnetostriction coefficient for YbInCu₄ and YbAgCu₄ are large at low temperatures but depressed in the high-temperature localized-moment regime, whereas those of YbAuCu₄ with almost stable Yb³⁺ moments are small. The results are discussed in terms of a modified Kondo-lattice model, where we emphasize that the volume dependence of the effective 4f electron (hole) number plays a dominant role. The anisotropic magnetostrictions of YbAuCu₄ and YbAgCu₄ have a positive sign but that of YbInCu₄ is negative, suggesting a difference in background crystal-field ground states of the Yb³⁺ multiplet.

1. Introduction

Valence fluctuation and the Kondo-lattice formation are observed in intermetallic compounds with Ce and Yb. The compounds YbXCu₄ with X = In, Ag and Au are of particular interest among Yb-based compounds, because they form the same crystal structure with cubic symmetry as MgCu₄Sn (*C*15*b*) type but show a wide variety of magnetic properties depending on the extent of hybridization between the 4f and conduction electrons (c–f interaction) [1].

The temperature-induced first-order valence transition at $T_V \simeq 40$ K discovered in YbInCu₄ [2] is compared to the pressure-driven $\alpha - \gamma$ transition in Ce metal [3], which is one of the fundamental problems in f-electron magnetism. An abrupt drop of the magnetic susceptibility was observed below T_V . Detailed investigations proved this transition to be not magnetic, but of valence type, accompanied by discontinuous and large lattice expansion without change in the crystal symmetry [2,4,5]. At high temperatures the susceptibility follows the Curie–Weiss law with a small negative paramagnetic Curie temperature, θ_p , and an effective magnetic moment, μ_{eff} , close to that of the Yb³⁺ free ion (4.54 μ_B). Below T_V the susceptibility behaves like that of a Pauli paramagnet. The strong hybridization of 4f electrons in Yb with conduction bands leads to a Fermi-liquid behaviour [6–9] due to the Kondo-lattice formation. The Kondo temperature, T_K , determined for the low-temperature state can be as large as $\simeq 500$ K, whereas the high-temperature state is characterized by a modest $T_K \simeq 25$ K [9, 10]. The magnetic field and pressure stabilize the high-temperature state [2, 10], and large magnetic fields result in metamagnetic transitions from the mixed-valence state to the nearly trivalent state of Yb [10, 11]. It is notable that the high-temperature state of YbInCu₄ is characterized

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by a low-carrier-density (semimetallic) nature [12–15], which may be related to the origin of the *first-order* valence transition.

YbAuCu₄ is characterized by the nearly localized moment of Yb³⁺ [16–19]. The susceptibility obeys the Curie–Weiss law except at very low temperatures. The small intersite RKKY interaction induces an antiferromagnetic ordering at a very low temperature below 1 K [1, 16, 20, 21]. The Kondo temperature T_K is $\simeq 2$ K [19, 20, 22]. The high-temperature magnetic properties are dominated by a crystal-field splitting much larger than T_K [19, 26].

YbAgCu₄ is a mixed-valence compound with a nonmagnetic ground state [16, 23]. The temperature variation of the susceptibility is typical for the mixed-valence compound, i.e. exhibits a broad maximum at $\simeq 40$ K. Above about 100 K the susceptibility follows the Curie–Weiss law with $\mu_{eff} = 4.4 \,\mu_B$, suggesting a nearly localized state of Yb 4f electrons. A large linear coefficient of specific heat $\gamma = 240$ mJ K⁻² mol⁻¹ has been obtained [16]. The electrical resistivity shows a Fermi-liquid-like T^2 -dependence at low temperatures [16,24,25]. A number of investigations such as neutron scattering experiments [26], nuclear or electron resonance investigations [27,28], x-ray absorption or photoelectron spectroscopy [18,29] and band calculations [30] confirmed the strong c–f hybridization. The Kondo temperature T_K has been estimated from various experiments and theories as $\simeq 100$ K [16, 18, 24–27, 29, 31–33].

For these YbXCu₄ compounds, magnetostrictions have been measured to monitor the metamagnetic transitions induced by large external fields [11] or in the temperature range close to T_V for YbInCu₄ [34]. No systematic experiments dealing with small magnetostrictions far from the phase boundary have been reported. The purpose of this study is to measure small magnetostrictions with good accuracy, separate isotropic (volume) and anisotropic contributions and discuss the Kondo effect. It should be noted that, although magnetostriction measurements have not been applied extensively to Kondo problems, the volume magnetostriction gives equivalent information on the pressure dependence of the magnetization, which is useful in this field. In addition, an anisotropic magnetostriction is a good probe for investigating the 4f electron state in the crystal, because the 4f shell of Yb³⁺ (J = 7/2) is strongly aspherical, depending on the crystal fields. In this paper we report the results of precise and systematic magnetostriction measurements on the YbXCu₄ (X = In, Ag and Au) compounds.

2. Experimental procedures

Polycrystalline samples of YbXCu₄ (X = In, Ag and Au) were prepared by argon arc melting. The ingots were annealed at 750 °C for seven days. X-ray analyses revealed all the compounds to be of single C15b type. The temperature dependence of the susceptibility was measured for all the samples to check the quality and to obtain the magnetostriction coefficients, C_v and C_a , defined in (3) and (4) below. Since the susceptibility jump at T_V is not completely sharp for polycrystalline YbInCu₄ compared with single-crystalline samples [10, 35], here we will not deal with magnetostrictions in the vicinity of T_V .

Strains both parallel $(\Delta L/L)_{\parallel}$ and perpendicular $(\Delta L/L)_{\perp}$ to the applied field were estimated for the polycrystalline samples by measuring capacitance changes of home-made three-terminal capacitance cells in a slowly sweeping magnetic field up to H = 8 T and in the temperature range of T = 5-100 K. The typical size of specimens was $5 \times 5 \times 5$ mm³. The strain resolution was better than 5×10^{-8} . Volume and anisotropic magnetostrictions were estimated from

$$\omega = \frac{\Delta V}{V} = \left(\frac{\Delta L}{L}\right)_{\parallel} + 2\left(\frac{\Delta L}{L}\right)_{\perp} \tag{1}$$

Magnetostriction of YbXCu₄

$$\lambda = \left(\frac{\Delta L}{L}\right)_{\parallel} - \left(\frac{\Delta L}{L}\right)_{\perp}.$$
(2)

3. Results

Typical results for magnetostrictions are shown in figures 1–3. In each case, the data were fitted with the square of the field, H^2 , which is drawn as a solid curve.

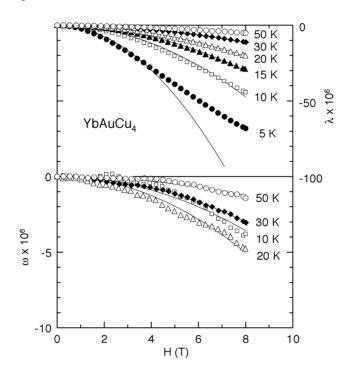


Figure 1. Examples of volume and anisotropic magnetostrictions of YbAuCu₄.

Figure 1 shows magnetostriction curves for YbAuCu₄. The strains, $(\Delta L/L)_{\parallel}$ and $(\Delta L/L)_{\perp}$, of YbAuCu₄ are large and opposite in sign. As a result, the volume magnetostriction ω is very small due to cancellation of these components. The anisotropic magnetostriction λ is, on the other hand, large and positive. The curves at 15 K show the H^2 -dependence. The magnetostrictions at the lower temperatures change as H^2 in the initial fields as well but deviate from the H^2 -dependence at higher fields. This deviation seems to be related to the saturation of the magnetization. It is likely that the magnitude of the magnetostrictions is proportional to the square of the magnetization M^2 . We have estimated the coupling constants, S_v , S_a , C_v and C_a , by making the following definitions:

$$\omega = S_{\rm v}H^2 = C_{\rm v}\chi^2 = C_{\rm v}\chi^2 H^2 \tag{3}$$

$$\lambda = S_{a}H^{2} = C_{a}M^{2} = C_{a}\chi^{2}H^{2}.$$
(4)

The temperature dependences of C_v and C_a are shown in figures 4 and 5, respectively. Both C_v and C_a for YbAuCu₄ are negative with rather small magnitudes. It should be noted that the coupling constants C_v and C_a thus defined are almost constant against temperature regardless of the remarkable temperature dependence of the susceptibility, indicating that the magnetostrictions scale well with M^2 as expected above.

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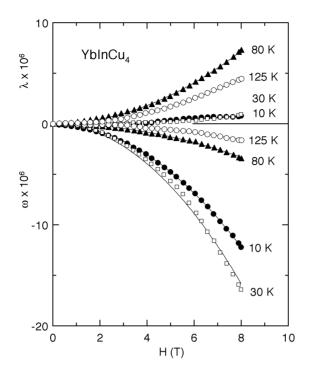


Figure 2. Examples of volume and anisotropic magnetostrictions of YbInCu₄.

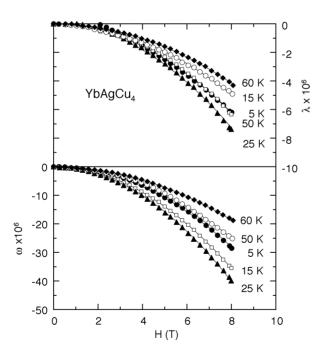


Figure 3. Examples of volume and anisotropic magnetostrictions of YbAgCu₄.

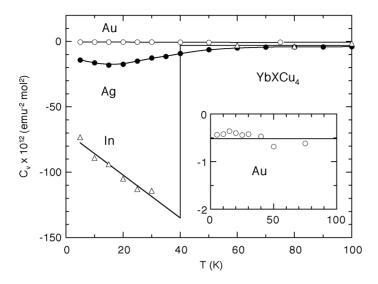


Figure 4. Temperature dependences of the magnetovolume coupling constant, C_v , for YbAuCu₄ (\bigcirc), YbInCu₄ (\triangle) and YbAgCu₄ (\bullet). The inset is a magnification for YbAuCu₄.

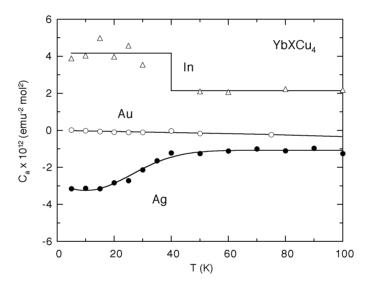


Figure 5. Temperature dependences of the anisotropic magnetostriction coupling constant, C_a , for YbAuCu₄ (\bigcirc), YbInCu₄ (\triangle) and YbAgCu₄ (\spadesuit).

Figure 2 shows magnetostriction curves for YbInCu₄. Here, we do not deal with the giant magnetostriction related to the first-order valence transition at around T_V [11, 34]. The data for YbInCu₄ are qualitatively different from those for YbAuCu₄. Below T_V , the isotropic volume magnetostriction is dominant. The anisotropic contribution is positive in contrast to that for YbAuCu₄. Above T_V the isotropic and anisotropic parts are of approximately the same magnitude. All experimental data except those around T_V vary as H^2 (see the solid curves in figure 2). The temperature dependence of C_V is presented in figure 4. Its absolute magnitude increases with increasing temperature and reaches a maximum just below T_V . Above T_V , C_V

drops drastically and remains nearly constant at the higher temperatures. On the other hand, C_a does not change significantly at T_V as seen in figure 5.

The results for YbAgCu₄ are presented in figure 3. The magnetostriction is substantially of volume type as for YbInCu₄, but has a discernible anisotropic part with negative sign as for YbAuCu₄. As the case of YbInCu₄, C_v is negative over the whole temperature range (figure 4). Its absolute value increases with increasing temperature, then passes through a maximum at around 15 K and decreases to become almost constant at higher temperatures. As seen in figure 5, C_a is negative. Its magnitude decreases with increasing temperature and shows a weak temperature dependence at high temperatures.

4. Discussion

4.1. Volume magnetostriction

The experimental results are summarized as follows. The volume magnetostriction is negligibly small for YbAuCu₄, where the ytterbium is in an almost stable Yb³⁺ state with well defined localized moments interacting weakly with each other. On the other hand, C_v is significantly large for the low-temperature phase of YbInCu₄ and the low-temperature range of YbAgCu₄, both of which are in a mixed-valence or Kondo-lattice state. The high-temperature phase of YbInCu₄, which looks like a nearly localized-moment system, has a fairly small C_v .

4.1.1. The interconfigurational fluctuation model. Qualitatively, these behaviours may be explained in terms of the interconfigurational fluctuation (ICF) model as applied to the YbInCu₄ system in the early stage of the investigation [2]. Generally, ytterbium can be in a mixed-valence state between Yb³⁺ and Yb²⁺. The former state is magnetic with a smaller volume and the latter nonmagnetic with a larger volume. For a mixed-valence state, an applied field stabilizes the magnetic state and then the fraction of Yb³⁺ should increase, giving rise to a volume shrinkage (negative ω). On the other hand, when ytterbium is in the pure trivalent state, a magnetic field does not induce a valence change any longer and hence there is no volume change. It should be noted that the situation in the Ce-based system is just the opposite. Cerium is in a mixed-valence state between Ce³⁺ and Ce⁴⁺. Magnetic Ce³⁺ has a volume larger than nonmagnetic Ce⁴⁺, and therefore, a positive volume magnetostriction is somewhat different, as will be stated in section 4.1.2.

A straightforward approach to explaining the magnetovolume effect of the mixed-valence system based on the ICF scheme was developed by Zieglowski *et al* [36]. They ascribed the volume magnetostriction to the valence change induced by applied field or pressure. According to their model, the volume magnetostriction is given by

$$\omega = \frac{\Delta V}{V} = \left[\pm \frac{\nu_0 (1 - \nu_0)}{2} \frac{(V_{n+1} - V_n)}{V} \frac{\mu_z^2}{k_B^2} \right] \frac{H^2}{(T + T_f)^2}$$
(5)

where v_0 is the fractional occupation of the higher-valence state at T = 0 (tetravalent for Ce and trivalent for Yb), V_n the volume for the *n*-valent state, μ_z the principal-axis component of the relevant magnetic moment and T_f the characteristic temperature describing valence mixing at T = 0. This formula is consistent with the χ^2 -scaling of ω provided that v_0 remains constant, because the susceptibility obeys the Curie–Weiss law for constant v_0 . We have to employ positive and negative signs for cerium and ytterbium, respectively, according to the ICF scheme in which the field always induces the magnetic state (with a larger volume (n = 3) for Ce and a smaller volume (n + 1 = 3) for Yb). Then we always expect positive and negative

volume magnetostrictions for Ce- and Yb-based compounds, respectively. The magnitude of ω is mainly determined by v_0 . It is believed that YbAuCu₄ is in an almost stable trivalent state. This means that $v_0 \simeq 1$ and so $S_v \simeq 0$. The large volume magnetostriction of YbInCu₄ and YbAgCu₄ can be explained in the same context with appropriate values of v_0 and T_f . This interpretation accords well with the intuitive consideration for the mixed-valence state. However, the classical ICF model is only phenomenological, gives parameters which are not justified physically and is in conflict with important characteristics of these compounds such as the Fermi-liquid behaviour.

4.1.2. The conventional Kondo-lattice model. The Kondo lattice model has been employed to analyse quantitatively the magnetostriction of mixed-valence systems [37, 38]. The magnetovolume coupling in the Kondo system is usually ascribed to the volume dependence of $T_{\rm K}$ [39]. At temperatures well above $T_{\rm K}$, the susceptibility of the Kondo compound is approximated by

$$\chi = \frac{C}{T + T_{\rm K}(V)}.\tag{6}$$

Assuming that the Curie constant C is independent of volume, we have

$$\frac{\mathrm{d}\chi}{\mathrm{d}\omega} = -\frac{\chi^2}{C} \frac{\mathrm{d}T_{\mathrm{K}}}{\mathrm{d}\omega}.\tag{7}$$

The thermodynamic relation (the Maxwell relation) for a paramagnet gives

$$\frac{\kappa}{V_{\rm m}}\frac{\mathrm{d}\chi}{\mathrm{d}\omega} = 2S_{\rm v} = 2C_{\rm v}\chi^2 \tag{8}$$

where κ is the compressibility, $V_{\rm m}$ the molar volume, χ the molar susceptibility and $S_{\rm v}$ and $C_{\rm v}$ the magnetovolume coupling constants defined in (3). Then, we have

$$C_{\rm v} = -\frac{\kappa T_{\rm K}}{2V_{\rm m}C} \frac{\mathrm{d}\ln T_{\rm K}}{\mathrm{d}\omega}.$$
(9)

Here, we discuss the magnitude and sign of C_v . Within the simple Kondo-lattice model, it is assumed that

$$\frac{d\ln T_{\rm K}}{d\omega} \simeq \frac{d\ln V_{cf}}{d\omega} \tag{10}$$

where V_{cf} represents the 4f–conduction electron (c–f) hybridization. A volume expansion should depress V_{cf} , leading to a decrease of $T_{\rm K}$ and therefore d ln $T_{\rm K}/d\omega < 0$. Then, equation (9) predicts a positive volume magnetostriction coefficient (and a negative pressure effect on the susceptibility) for the Kondo lattice. This is in accordance with the experimental results for the Ce-based system but conflicts with the present results for YbXCu₄ compounds, indicating that the simple Kondo model is not appropriate for explaining the volume magnetostriction of the Yb-based system.

According to a conventional treatment, equation (9) gives the Grüneisen parameter for the Kondo temperature as

$$\Omega_{\rm K} = -\frac{\mathrm{d}\ln T_{\rm K}}{\mathrm{d}\omega} = \frac{2V_{\rm m}C}{\kappa} \frac{C_{\rm v}}{T_{\rm K}}.\tag{11}$$

Parameters $\Omega_{\rm K}$ for the present compounds estimated from the experimental values of $C_{\rm v}$ are listed in table 1, where common values of $\kappa = 10^{-6}$ bar⁻¹ [19, 24, 34, 40], $V_{\rm m} = 55$ cm³, C = 2.6 emu K mol⁻¹ (the Yb³⁺ free-ion value) and appropriate values of $T_{\rm K}$ (listed in table 1) were used. Similar values have been estimated from other experimental results

[10, 14, 19, 24, 25, 41, 42]. These values are large in magnitude and comparable with those for typical Ce-based Kondo or heavy-fermion compounds although the sign is opposite [37], indicating the strong volume dependence of $T_{\rm K}$. The conventional Kondo-volume-collapse (KVC) model [39] attributes the *first-order* valence transition to the strong coupling between volume and $T_{\rm K}$, i.e. large $|\Omega_{\rm K}|$. The present results do not indicate particularly large $|\Omega_{\rm K}|$ for YbInCu₄ among the YbXCu₄ compounds, suggesting that the simple KVC model does not apply to the *first-order* valence transition of YbInCu₄.

		<i>Т</i> К (К)	n_f	v	$C_{\rm v}^{\rm obs} \times 10^{12}$ (emu ⁻² mol ²)) Ω _K
YbInCu ₄	$\begin{array}{l} T > T_{\rm V} \\ T < T_{\rm V} \end{array}$	25 500	0.96 0.83	-50 -300	$\simeq -3$ $\simeq -100$	$\simeq -30$ $\simeq -50$
YbAgCu ₄ YbAuCu ₄		100 2	$\simeq 0.9$ 0.985	-90 -10	$\simeq -10$ $\simeq -0.5$	$\simeq -30$ $\simeq -70$

Table 1. Magnetovolume parameters for $YbXCu_4$ (X = In, Ag and Au).

4.1.3. The effect of the 4f hole number on the Kondo lattice. Cornelius et al [14] have studied the YbIn_{1-x}Ag_xCu₄ system and pointed out the important role of the effective 4f hole number, n_f , as a factor determining the cell volume and T_K . There has been similar discussion for YbAgCu₄ in [24]. Cornelius et al [14] employed the relation between T_K and n_f derived for the Kondo-impurity limit [43]:

$$T_{\rm K} = \frac{1}{\pi} (2J+1) V_{cf}^2 \rho(\varepsilon_{\rm F}) \frac{1-n_f}{n_f}$$
(12)

where $\rho(\varepsilon_F)$ is the conduction electron density of states at the Fermi level. Assuming that only n_f depends on volume, we have

$$\frac{d\ln T_{\rm K}}{d\omega} = -\frac{1}{n_f(1-n_f)} \frac{dn_f}{d\omega}.$$
(13)

The fractional volume change is related to n_f as

$$\omega = \omega_0 (1 - n_f) \tag{14}$$

where $\omega_0 = (V_2 - V_3)/V$ is the relative volume change between Yb³⁺ and Yb²⁺ states. The value ω_0 is roughly estimated from the volume and valence change at T_V for YbInCu₄ [9, 14] as $\omega_0 = 0.042$, where we employed values of n_f determined from Yb L₃ x-ray absorption spectra. From (14) we have

$$\frac{\mathrm{d}n_f}{\mathrm{d}\omega} = -\frac{1}{\omega_0}.\tag{15}$$

With (13) and (15), equation (9) can be rewritten as

$$C_{\rm v} = -\frac{\kappa}{2V_{\rm m}C} \frac{T_{\rm K}}{n_f (1 - n_f)} \frac{1}{\omega_0}.$$
 (16)

This equation enables us to calculate C_v , if T_K and n_f are given. Using common values of κ , V_m and C already used above and $\omega_0 = 0.042$ and taking the values of n_f from L₃ spectra [14, 18] and appropriate T_K , we calculated crude values of C_v , which are included in table 1. The calculated values are somewhat larger than the experimental values but reproduce the sign and tendency. One should note that the volume dependence of V_{cf} gives a positive contribution

to $T_{\rm K}$ as in (10) and reduces the absolute magnitude of $C_{\rm v}$ in (16), improving the agreement with experiments. This argument shows that the change in the effective number of 4f electrons dominates the magnetovolume coupling and that the direct volume dependence of the c–f hybridization is a secondary effect. There has been similar discussion for the magnetovolume effect of some Ce-based compounds [44].

It should be noted here that equation (6) is applicable only for $T > T_K$ and that equation (12) derived from the single-impurity model may not be fully general. A more comprehensive treatment for the magnetovolume effect of the Kondo system is required.

4.2. Anisotropic magnetostriction

The difference in the temperature dependence of C_a is not so distinctive compared with that for C_v . According to the single-ion model for magnetostriction, the anisotropic magnetostriction originates from realignments of magnetic atoms with aspherical electron clouds induced by the external field [45]. It may not be so sensitive to the valence state. It should be noted, here, that C_a has a positive sign only for YbInCu₄. This suggests that the ground state of the *background* Yb³⁺ multiplet is different from the other two. In other words, crystal fields at Yb sites are different.

The crystal field of cubic symmetry splits the eightfold-degenerate state $({}^{2}F_{7/2})$ of the Yb³⁺ ion into two doublets (Γ_{6} and Γ_{7}) and one quartet (Γ_{8}). For stable-moment YbAuCu₄, a crystalfield-level scheme has been determined by neutron scattering experiments; excited Γ_{8} and Γ_{6} above a ground state Γ_{7} [26], and supported by other measurements [19, 20]. The crystalfield level of YbAgCu₄ is not clear; quasielastic (or inelastic) neutron scattering observed for YbAgCu₄ was interpreted as the result of the Kondo-lattice formation [26], but does not contradict the suggestion of a doublet ground state similar to that of YbAuCu₄, as discussed in [26] and as supported by optical measurements [33]. As for YbInCu₄, Severing *et al* [7] interpreted the inelastic neutron scattering above T_V as excited Γ_7 and Γ_6 doublets above a ground-state Γ_8 quartet, which seems to be consistent with a specific heat analysis [10], whereas Lawrence *et al* [9] interpreted it as due to the Kondo interaction. Although experimental situations are thus complicated, they do not contradict the above speculation based on the signs of C_a . It is likely that the absolute values of C_a relate to the strength of the c–f hybridization (or the magnitude of T_K) in the same way as the volume magnetostriction, but the mechanism has not been made clear.

5. Concluding remarks

We have measured magnetostrictions of Yb-based Kondo compounds YbXCu₄ (X = In, Ag and Au). The volume magnetostrictions are negative for all of the compounds in contrast to the case for Ce-based Kondo systems. We obtained large magnetovolume coupling for large- T_K states. The results were discussed in terms of a modified Kondo-lattice model, where we stressed the important role of the 4f electron (hole) number, n_f (in other words, the conduction electron number) modified by a volume change or an applied field. The volume dependence of n_f results in a much larger effect than the direct contribution from the c–f hybridization; these contributions are opposite in sign in Yb-based systems. There is no reason to doubt that the volume dependence of n_f is also dominant in the Ce-based Kondo system, where both of the contributions are positive in sign. Finally, we would like to emphasize that, if a volume magnetostriction is precisely and appropriately evaluated, it is a particularly useful probe for investigating the Kondo system.

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