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1999 J. Phys.: Condens. Matter 11 4381

(http://iopscience.iop.org/0953-8984/11/22/309)

J. Phys.: Condens. Matter **11** (1999) 4381–4390. Printed in the UK PII: S0953-8984(99)98448-5

The effect of pressure on the magnetic susceptibility of $RInCu_4$ (R = Gd, Er and Yb)

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Received 15 October 1998, in final form 16 March 1999

Abstract. The magnetic susceptibility of the intermetallic compounds $RInCu_4$ (R = Gd, Er and Yb) was measured under helium gas pressure up to 2 kbar at the fixed temperatures 78, 150 and 300 K. For $YbInCu_4$, which exhibits a first-order valence phase transition at $T_V \simeq 40$ K, the Grüneisen parameter for the Kondo energy, $\Omega_K \equiv -\mathrm{d}\ln T_K/\mathrm{d}\ln V = -32$, is large and typical for Ce-based heavy-fermion systems in magnitude but opposite in sign. The effect of atomic disorder is discussed on the basis of the data for a chemically disordered sample; the pressure effect at $T = 78$ K is strongly enhanced due to the spatial dispersion of pressure-sensitive T_V, and hence $\mathrm{d}T_V/\mathrm{d}P = -2.0$ K kbar^{-1} is obtained by assuming a Gaussian distribution of T_V. On the basis of an extrapolation of the experimental pressure dependence, a (P, T) phase diagram is proposed for $YbInCu_4$. Reference compounds with stable f moments, $GdInCu_4$ and $ErInCu_4$, show negligible pressure dependences of the susceptibility.

1. Introduction

The integration of local f levels of rare-earth ions into quasi-continuous states of the band spectrum in metallic systems has been of great interest for many years, but the problem is still far from being solved exactly. In some cases, information on electron parameters, which characterize this integration, can be obtained from the temperature dependence of the magnetic susceptibility $\chi(T)$. An effect of pressure on the susceptibility $\mathrm{d}\ln\chi/\mathrm{d}P$ or the magnetovolume effect $\mathrm{d}\ln\chi/\mathrm{d}\ln V$ provides the atomic-volume dependence of the parameters. This dependence is of importance in elucidating both a microscopic origin of the parameters and their role in determining the type of the phase transition between different modes (or phases) with different extents of interaction between f levels and band states.

In particular, with varying thermodynamic variables or chemical composition, the transition from the Kondo or heavy-fermion (HF) state to the intermediate-valence (IV) state may proceed gradually as, for instance, in the $CeIn_{3-x}Sn_x$ system [1], or may be realized by a first-order phase transition accompanied by a considerable change in volume with the lattice symmetry retained. One case for which the latter applies is that of pure cerium, in which the transition associated with a volume change of about $\Delta V/V \simeq 0.15$ [2] is induced by applying pressure. Another is that of $YbInCu_4$, in which the IV–HF transition with $\Delta V/V \simeq 0.005$ is realized with temperature increasing across $T_V \simeq 40$ K [3, 4]. For $YbInCu_4$ the valence transition point, T_V, is very sensitive to pressure and magnetic field [4, 5] and, in addition, substitution of Ag for In results in a change from the discontinuous first-order transition to a smooth one [6]. Hence, $YbInCu_4$ is particularly attractive and convenient for the study of the magnetovolume effect.

0953-8984/99/224381+10$19.50

In this paper, we present results on the effect of pressure (up to 2 kbar at $78 \leqslant T \leqslant 300$ K) on the magnetic susceptibility of unstable-valence $YbInCu_4$ and its isostructural analogues with stable valence, $GdInCu_4$ and $ErInCu_4$, as references. Preliminary results of the present study have already been presented in reference [7]. Up to now, the pressure dependence of χ, $\chi(P)$, has been reported for a non-stoichiometric compound, $Yb_{0.8}In_{1.2}Cu_4$ [5], and for $YbInCu_4$ [8], but both reports are mainly concerned with $T_V(P)$. The magnetostriction measurements for $YbInCu_4$ covering the temperature range around T_V revealed a shift of T_V in high magnetic fields [4]. To our knowledge there are so far no reliable published data on the magnetovolume effect of the high-temperature HF phase.

The second and third sections of this paper describe the experimental procedures and the results on magnetovolume effects, respectively. The fourth deals with discussion of the results; its subsections concern magnetic interactions in $RInCu_4$, the Grüneisen parameter for the Kondo energy of $YbInCu_4$, the role of atomic disorder in the susceptibility of $YbInCu_4$ and a (P, T) phase diagram proposed for $YbInCu_4$. The last section presents a summary and basic conclusions.

2. Experimental procedures

Polycrystalline ingots of $GdInCu_4$, $ErInCu_4$ and $YbInCu_4$ were prepared from a stoichiometric mixture of the elements (Gd, Er, Yb—3N purity; Cu, In—5N purity) in an argon arc furnace. Some ingots were then annealed in evacuated quartz tubes for a week at 750–850 °C. Samples with appropriate sizes were spark-cut from the ingots.

The temperature dependence of the magnetic susceptibility was measured by the Faraday method at $T = 4.2$–300 K in a magnetic field of 0.8 T. The effect of helium-gas-produced pressure (up to 2 kbar) on the susceptibility was studied at fixed temperatures in the range $T = 78$–300 K by using a pendulum magnetometer placed directly in a high-pressure chamber as described in reference [9]. The applied magnetic field was less than 2 T. The relative error of the measurements under pressure was no more than 0.05%. In all cases the susceptibility varies linearly with pressure, and no hysteresis was observed with increasing and decreasing pressure.

3. Experimental results

The susceptibility for stable-valence $GdInCu_4$ and $ErInCu_4$ obeys the Curie–Weiss law

$$\chi(T) = \frac{C}{T - \Theta} \tag{1}$$

with Θ the paramagnetic Curie temperature and C the Curie constant. The values of Θ and the effective paramagnetic moment, μ_{eff}, deduced from C are given in table 1. The latter values correspond well to those of free R^{3+} ions. These quantities are in good agreement with the data presented in references [10, 11], and the $\chi(T)$ curves for $GdInCu_4$ and $ErInCu_4$ are not shown here.

The pressure dependences of the susceptibility of $GdInCu_4$ at $T = 78$ and 300 K are shown in figure 1. The effect of pressure on the susceptibility can be decomposed as

$$\frac{\mathrm{d}\ln\chi}{\mathrm{d}P} = \frac{\mathrm{d}\ln C}{\mathrm{d}P} + \frac{\chi}{C}\frac{\mathrm{d}\Theta}{\mathrm{d}P} \simeq \frac{\chi}{C}\frac{\mathrm{d}\Theta}{\mathrm{d}P} \tag{2}$$

($\mathrm{d}\ln C/\mathrm{d}P = 0$ for the free-ion moment), which appears to be close to the experimental resolution for $GdInCu_4$. The same situation was found for $ErInCu_4$. The estimated values of $\mathrm{d}\Theta/\mathrm{d}P$ for $GdInCu_4$ and $ErInCu_4$ are included in table 1.

Table 1. Magnetic parameters and their pressure derivatives for $RInCu_4$.

Compound	T (K)	χ (10^3 emu mol^{-1})	$\mathrm{d}\ln\chi/\mathrm{d}P$ (Mbar^{-1})	Θ (K)	μ_{eff} (μ_B/f.u.)	$\mathrm{d}\Theta/\mathrm{d}P$ (K Mbar^{-1})
$GdInCu_4$	300	22.2	0.15 ± 0.2	-47	7.86	30 ± 30
	78	61.9	0.25 ± 0.3			
$ErInCu_4$	300	35.8	0.1 ± 0.3	~ 0	9.3	0 ± 30
	78	132.8	0.0 ± 0.3			
$YbInCu_4$	300	6.97	2.2 ± 0.3	-13 ± 3	4.17 ± 0.02	640 ± 50
	150	13.5	4.0 ± 0.3			
	78	21.6[a]	25.5 ± 1.0[a]			
	20	4.5[b]	~ 50[b]			

[a] Data for an imperfect sample with atomic disorder.
[b] Data obtained in a magnetostriction measurement [14].

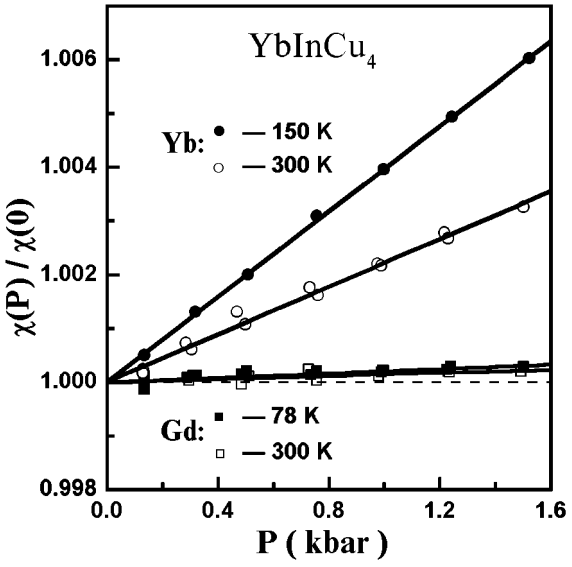

Figure 1. The pressure dependence of the magnetic susceptibility for $YbInCu_4$ at 150 (●) and 300 K (○) and for $GdInCu_4$ at 78 (■) and 300 K (□).

The temperature dependence of the susceptibility for $YbInCu_4$ is also consistent with previous results [6, 12, 13, 15, 16]: a low-temperature plateau χ_0, a sharp rise at $T_V \simeq 40$ K, a maximum, whose amplitude depends on the sample quality, and a subsequent Curie–Weiss-type decay. The reciprocal susceptibilities of our sample at various temperatures are compared in figure 2 with $1/\chi(T)$ for a high-quality single-crystalline sample [6]. The Curie–Weiss parameters obtained for our sample, which are listed in table 1, are close to those reported for other polycrystalline samples.

The pressure effect of $YbInCu_4$ is large, in contrast to those of $GdInCu_4$ and $ErInCu_4$. The results at $T = 150$ and 300 K are shown in figure 1. Measurements were restricted to high temperatures far above T_V to avoid the giant variation around T_V associated with the valence transition. For the high-temperature HF region, we obtain

$$\mathrm{d}\Theta/\mathrm{d}P \simeq 640 \pm 50 \text{ K Mbar}^{-1} \qquad \mathrm{d}\ln C/\mathrm{d}P = 0 \pm 0.3 \text{ Mbar}^{-1} \tag{3}$$

by fitting $\mathrm{d}\ln\chi/\mathrm{d}P$ for this temperature range with equation (2). Reference [8] reported $\mathrm{d}\Theta/\mathrm{d}P = -300$ K Mbar^{-1}, which is opposite in sign, a markedly small $\Theta = -0.2$ K at

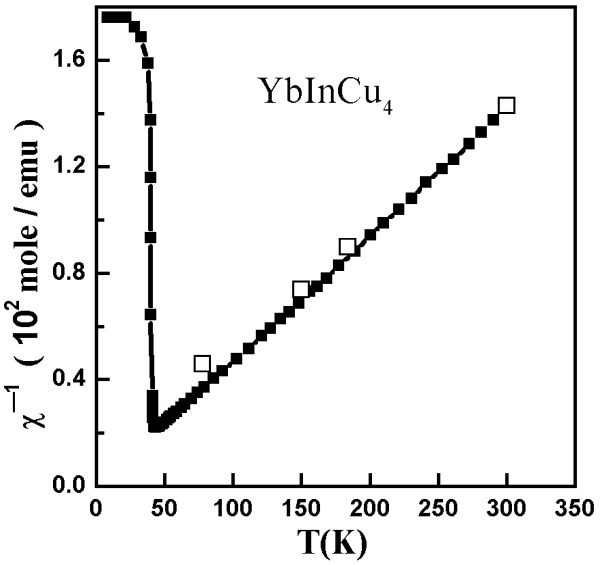

Figure 2. The temperature dependence of the reciprocal susceptibility for $YbInCu_4$. Solid squares represent the data reported in reference [6] and open squares show the data from the present work.

ambient pressure and a pressure-dependent C. These features are different from the ones obtained by us.

At temperatures close to T_V, the magnetovolume effect is very sensitive to the sample quality. Thus a $YbInCu_4$ sample including atomic disorder, which shows a broad peak in the $\chi(T)$ curve around $T = 80$ K, as shown in figure 3(a), was used to underline such a property. The derivatives $\mathrm{d}\ln\chi/\mathrm{d}P$ estimated at various temperatures are plotted against χ in figure 3(b). The value at $T = 78$ K is four times larger than that expected from the linear relation between $\mathrm{d}\ln\chi/\mathrm{d}P$ and χ for a higher-quality sample (the dashed line in figure 3(b)). The non-linear behaviour of $\mathrm{d}\ln\chi/\mathrm{d}P$ versus χ as well as the smeared $\chi(T)$ curve will be discussed in section 4.3 together with other experimental data.

4. Discussion

4.1. Stable-valence $GdInCu_4$ and $ErInCu_4$

In non-magnetic metallic or semimetallic matrices, localized magnetic moments of the rare-earth elements interact with each other via conduction electrons by the Ruderman–Kittel–Kasuya–Yosida (RKKY) type of mechanism. In fact this indirect interaction is far from the classical RKKY exchange, and only in rare cases can it and, hence, Θ be attributed to band-structure peculiarities (see, for example, reference [17]). For $RInCu_4$, we do not have enough knowledge about the band structures and their volume dependence to investigate the details of interactions. Therefore, we will treat stable-valence $GdInCu_4$ and $ErInCu_4$ just as reference materials for the following analysis of $YbInCu_4$.

The contribution of the indirect exchange interaction between f moments to Θ is proportional to the de Gennes factor $G = (g_J - 1)^2 J(J + 1)$, where g_J is Landé's g-factor and J the total angular momentum. As is evident from the inset in figure 4, such an indirect exchange interaction of antiferromagnetic type is dominant in $RInCu_4$ with large G. However, the well defined experimental straight line $\Theta(G)$ does not intercept the origin of the

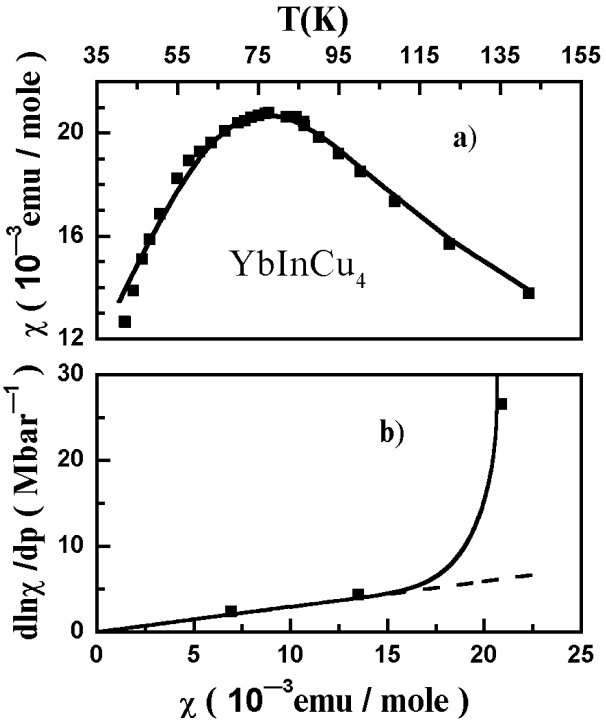

Figure 3. The results for a chemically disordered $YbInCu_4$ sample. (a) The temperature dependence of the susceptibility in the vicinity of the valence transition. (b) The pressure effect of the susceptibility $\mathrm{d}\ln\chi/\mathrm{d}P$ plotted against χ. Solid curves were drawn on the basis of the model described in the text. The dashed straight line is the linear relation for a good-quality sample, i.e. the line expected from the Curie–Weiss law.

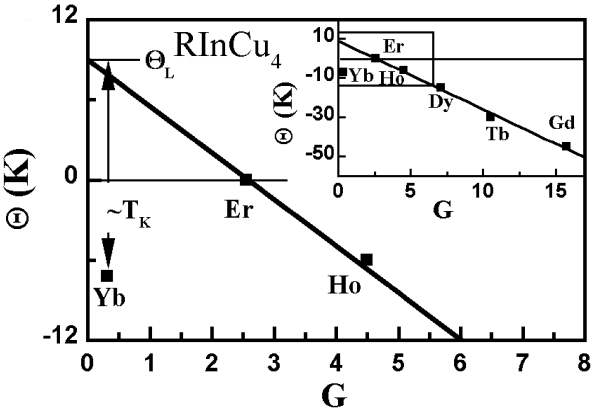

Figure 4. Experimental values of the paramagnetic Curie temperature, Θ [6, 10, 11] plotted against the de Gennes factor, G, for $RInCu_4$.

coordinates. The extrapolated value $\Theta(0) \equiv \Theta_L \simeq 9\,\mathrm{K}$ indicates a *positive*-in-sign background contribution, whose origin remains to be explained. This contribution is, along with the indirect exchange, pressure independent, as inferred from the data obtained for $GdInCu_4$ and $ErInCu_4$.

4.2. The heavy-fermion phase of YbInCu$_4$

The crystal-electric-field (CEF) splitting of the ground-state multiplet $J = \frac{7}{2}$ for Yb^{3+} is modest in $YbInCu_4$ (total splitting $\Delta = 44$ K [18]). Therefore, the effect of the CEF [15, 16], like that of non-f contributions, to the susceptibility is negligible at the temperatures of the present measurements; the non-magnetic analogues $YInCu_4$ and $LuInCu_4$ are weakly diamagnetic [19]. Thus, $\mu_{\rm eff}$ deviating from the trivalent value ($\mu_{\rm eff} = 4.54\ \mu_{\rm B}$) is a consequence of the Kondo effect. An analysis of $\chi(T)$ (figure 2) by means of a theory for the Kondo system with $J = \frac{7}{2}$ [20] gives an estimate of the Kondo temperature $T_{\rm K} \simeq 20$ K [6], which qualifies $YbInCu_4$ as a HF compound. For the HF state, one may expect $T_{\rm K} \sim |\Theta|$ [21]. This relation is satisfied for $YbInCu_4$ if the hypothetical background discussed above is taken into account, i.e. $T_{\rm K} \simeq \Theta_{\rm L} - \Theta$. This fact again suggests the existence of a positive background interaction $\Theta_{\rm L}$ in the HF state of $YbInCu_4$. In any case, the relation $\mathrm{d}T_{\rm K}/\mathrm{d}P \simeq -\mathrm{d}\Theta/\mathrm{d}P$ is believed to be valid. Combining $T_{\rm K} \simeq 20$ K [6] and a bulk modulus $B \simeq 1.0$ Mbar (at 300 K) [4], the Grüneisen parameter for the Kondo energy, $\Omega_{\rm K}$, in the HF state is calculated as

$$\Omega_{\rm K}^{\rm HF} \equiv -\frac{\mathrm{d}\ln T_{\rm K}}{\mathrm{d}\ln V} = \frac{1}{T_{\rm K}}\frac{\mathrm{d}\Theta}{\mathrm{d}\ln V} \simeq -32. \tag{4}$$

For other Yb compounds with unstable valence, the effect of pressure on the susceptibility [22,23] always results in a negative $\Omega_{\rm K}$, in contrast to the positive $\Omega_{\rm K}$ for Ce-based compounds [9,22,24]. The same consequence follows from the effect of pressure on the electrical resistivity in Ce-based compounds [25,26] and $YbAgCu_4$ [27]. This fact suggests a general tendency of f-state depopulation under pressure in Ce- and Yb-based compounds, considering the electron–hole symmetry in the Ce and Yb f shells ($T_{\rm K} \propto (1 - n_{\rm f})/n_{\rm f}$ where $n_{\rm f}$ is the effective number of f electrons or holes [28]). Thus, the pressure effects in both systems can be compared in absolute value, provided that this depopulation is caused mainly by a common mechanism of f-state shift relative to the Fermi energy.

In the Kondo-volume-collapse (KVC) model, it is the pressure-sensitive Kondo energy that is responsible for the first-order valence transition [29–31]. This model was successfully applied to describe phase diagrams of Ce-based compounds. The absolute value of $\Omega_{\rm K}$, which gives an estimate of the effect of the atomic volume on the Kondo energy, is large for $YbInCu_4$ but, in fact, normal for the Kondo systems. It falls just within the data range for the systems with a smooth valence change (for example, $CeInCu_2$, $CeCu_6$ [25], $CeInSn_2$ [9], $CeAl_2$ [24] and $YbAgCu_4$ [27]). Therefore, the large value of $\Omega_{\rm K}$ is not a convincing cause of the first-order valence transition.

Another possible origin is a specific feature of the individual microscopic parameters which determine $T_{\rm K}$. For a structureless conduction band with the width D, the Kondo temperature

$$T_{\rm K} = D\exp\left(-\frac{1}{8|s|}\right) \tag{5}$$

relates through a parameter, $s \equiv N(E_{\rm F})J_{\rm bf}$, to microscopic characteristics such as the density of states at the Fermi level, $N(E_{\rm F})$, and the effective exchange interaction between band and f electrons, $J_{\rm bf}$. Here all CEF levels for the $J = \frac{7}{2}$ octet are assumed to be populated, because $T > \Delta$ in the HF state [18]. Hence the effect of pressure on $T_{\rm K}$, i.e. $\Omega_{\rm K}$ (equation (4)), is decomposed into the pressure dependence of s and D:

$$\frac{\mathrm{d}\ln(T_{\rm K}/D)}{\mathrm{d}\ln s} = -\ln(T_{\rm K}/D) \tag{6}$$

which is derived from equation (5). The expected value of $\mathrm{d}\ln D/\mathrm{d}\ln V$ lies in the range from $-\frac{2}{3}$ for the free-electron-like sp band to $-\frac{5}{3}$ for the d band, and may be negligible in comparison with $\Omega_{\rm K}$ for $YbInCu_4$. The main source of the error in $\mathrm{d}\ln s/\mathrm{d}\ln V$ is the

uncertainty in D itself, which varies widely in Ce-based compounds [32–34]. Assuming the reasonable but arbitrary limits 2×10^3 K $< D < 2 \times 10^4$ K for YbInCu$_4$, we estimate $\mathrm{d}\ln s/\mathrm{d}\ln V \simeq 5.7 \pm 1.0$ from equations (4) and (6). Taking into account the opposite sign, this value agrees with the empirical value -7 ± 1 used in the KVC model [29,31], and does not differ from those for the more-stable-valence Ce-based compounds such as CeCu$_6$ (-6) [35] and CeAl$_2$ (-5.5) [24]. The atomic-volume dependence of the band characteristics is necessary to decompose $\mathrm{d}\ln s/\mathrm{d}\ln V$ further and to identify the dominant contribution. For semimetallic YbInCu$_4$ [36], such a dependence may be much more complicated than the simple bandwidth scaling used above. Unfortunately, *ab initio* calculations have not been performed yet.

It is of interest to estimate Ω_K for the low-temperature IV state. In reference [6] a large value of $T_\mathrm{K}^\mathrm{IV} \simeq 420$ K was estimated, assuming the Fermi-liquid relation $\chi_0 \propto 1/T_\mathrm{K}^\mathrm{IV}$ [20]. Applying the same relation, we have

$$\Omega_\mathrm{K}^\mathrm{IV} \equiv -\frac{\mathrm{d}\ln T_\mathrm{K}^\mathrm{IV}}{\mathrm{d}\ln V} = \frac{\mathrm{d}\ln \chi_0}{\mathrm{d}\ln V} \simeq -40. \tag{7}$$

Here, we used the data obtained in a magnetostriction measurement at $T = 20$ K [14], which are included in table 1, and a bulk modulus $B \simeq 0.83$ Mbar (at $T = 20$ K) [4]. Bearing in mind all uncertainties, this value is nearly the same as $\Omega_\mathrm{K}^\mathrm{HF}$. However, the corresponding derivative, $\mathrm{d}\ln s/\mathrm{d}\ln V \simeq 18 \pm 8$, is three times greater than that for the HF state or those for other Kondo systems, instead tending to support the KVC model [29–31]. A theoretical analysis using the values obtained above may lead to a more definite conclusion.

4.3. Effects of atomic disorder in YbInCu$_4$

A large and abrupt jump of the susceptibility at T_V along with the sensitive pressure dependence of T_V [3–5] results in giant magnetovolume effects in the vicinity of T_V [4]. With a dispersion of T_V, the smearing of the susceptibility jump will be accompanied by the enhancement of the pressure effect far above the ideal T_V (figure 3(b)). To discuss this mechanism quantitatively, we assume that T_V is spatially inhomogeneous in the sample with atomic disorder [37–39] (for a microscopic picture of the disorder in YbInCu$_4$, see reference [38]), and that, for the ith element in the sample, the temperature-independent susceptibility, χ_0, jumps to the Curie–Weiss susceptibility (1) at $T = T_\mathrm{V}^i$ just as in the ideal crystal (figure 1). With a distribution function for T_V^i, $W(T)$, and identical susceptibility parameters throughout the sample, we have

$$\chi(T) = \chi_0 + \left(\frac{C}{T-\Theta} - \chi_0\right) W(T). \tag{8}$$

For perfect single-crystalline YbInCu$_4$, $W(T)$ shows a completely discontinuous switching between 0 and 1 at T_V [6].

A similar equation but with another meaning for the parameters was used in the interconfigurational fluctuation (ICF) model of the valence phase transition [5, 15, 40], which is an alternative to the KVC model. In the model [5, 15], χ_0 stands for the susceptibility of the Yb^{2+} state and W means the probability of the excited Yb^{3+} state. Under certain conditions, the occupation-dependent energy of excitations gives rise to the first-order phase transition from Yb^{2+} to Yb^{3+} with a jump from $W \simeq 0$ to $W \simeq 1$. Some aspects of the susceptibility in YbInCu$_4$ were successfully explained by the ICF model, but this seems to be merely fortuitous, because neither the ground state (Yb^{2+} instead of the IV state), nor the value of the valence jump ($\sim$1 instead of 0.1 [3,4]), nor the overall CEF splitting ($\Delta = 135$ K instead of $\Delta = 44$ K [18]) corresponds to reality.

The subject in which we are interested is taking account of the structural disorder. If we assume that the broadening of $\chi(T)$ is solely due to structural disorder, the main problem is

that of choosing an appropriate $W(T)$. For simplicity, a normal Gaussian distribution function is assumed:

$$W(T) \equiv W(T, \overline{T_V}, \sigma) = \frac{1}{2}\left[1 + \mathrm{erf}\left(\frac{T - \overline{T_V}}{\sqrt{2}\sigma}\right)\right] \tag{9}$$

where erf(z) is a standard error function. An average value of T_V, $\overline{T_V}$, and the dispersion, σ^2, are treated as parameters. The derivative $\mathrm{d}\ln\chi/\mathrm{d}P$ is readily apparent from equations (8) and (9) with $\mathrm{d}T_V/\mathrm{d}P$ as an additional parameter. Thus $\chi(T)$ and $\mathrm{d}\ln\chi(T)/\mathrm{d}P$ can be described simultaneously by the model (8), (9) with the parameters χ_0, μ_{eff}, Θ, $\mathrm{d}\chi_0/\mathrm{d}P$ and $\mathrm{d}\Theta/\mathrm{d}P$ given in table 1. The solid curves in figures 3(a) and 3(b) correspond to the best-fit parameters

$$\overline{T_V} = 57\ \mathrm{K} \qquad \sigma = 22\ \mathrm{K} \qquad \mathrm{d}T_V/\mathrm{d}P = -2.0\ \mathrm{K\ kbar^{-1}}. \tag{10}$$

As seen in figure 3, this model describes $\chi(T, P)$ satisfactorily over the rather wide temperature range around the transition point. The $\overline{T_V}$-value agrees with the known tendency: a rise in the transition point with the degree of atomic disorder in $YbInCu_4$ [38, 39]. The derivative $\mathrm{d}T_V/\mathrm{d}P$ obtained agrees well with the averaged value of reported data [4, 5, 8, 41], which are scattered in the range from -2.3 to -1.7 K kbar^{-1} depending on the property studied and the method used for the identification of T_V. This indicates that the effect of the atomic disorder is properly taken into account in this model. Note that $YbInCu_4$ is a curious case, for which the atomic disorder allows us to derive a reliable value of $\mathrm{d}T_V/\mathrm{d}P$. A further increase in the magnetovolume effect is expected on approaching T_V (figure 3(b)), as is actually observed in the magnetostriction [4]. However, direct $\chi(P)$ measurements at $T < 78$ K were not performed, since a hysteresis may appear. Figure 3(b) also indicates that the non-linearity of the relationship between $\mathrm{d}\ln\chi/\mathrm{d}P$ and χ due to T_V being inhomogeneous is negligible at $T \geqslant 150$ K and has no effect on the $\mathrm{d}\Theta/\mathrm{d}P$ value obtained at higher temperatures even for this imperfect sample.

$YbInCu_4$ is suitable for use in comprehensively studying the role of imperfections and atomic disorder, which affect markedly the features of the phase transition. The model description proposed may be useful in the analysis of the effect of disorder on the various properties of this and related systems.

4.4. The phase diagram of $YbInCu_4$

Assuming a linear extrapolation of the present experimental data to a high-pressure region, a semi-quantitative (P, T) phase diagram for $YbInCu_4$ is proposed as figure 5. Note that the sequence of phases as pressure is varied is opposite to that for Ce-based systems. An external pressure stabilizes the trivalent state with the local moment (LM), which is crowded out from the IV ($P < 20$ kbar) and HF ($P < 35$ kbar) phases. The appearance of a ferromagnetic (FM) phase, which originates in the background interaction with positive sign, Θ_L, in the HF region ($P > 20$ kbar) is predicted. Ferromagnetic ordering in the Kondo lattice is not forbidden [42], but few examples have been found in Ce-based compounds [43]. The growth of the Curie temperature, T_C, is restricted by Θ_L, and so it is expected to be small. It is interesting to note that a ferromagnetic ordering has recently been found in isostructural $YbPdCu_4$ at low temperatures $T < 1$ K [44]. Corrections of the phase diagram to take into account the CEF effects or other interactions such as the quadrupolar one may be important in principle, but they are practically insignificant in this simplified treatment. Anyway, it is of interest to search for a FM phase for $YbInCu_4$ under pressure. Further studies of the phase diagram as well as the band-structure parameters are necessary to understand the real nature and hierarchy of the electron interactions in this class of compounds.

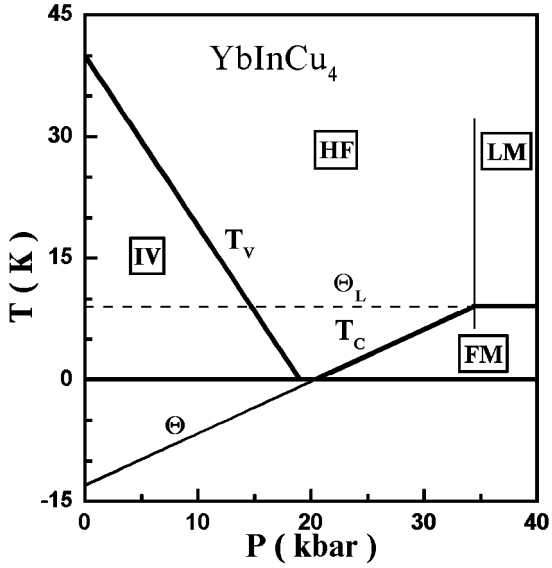

Figure 5. The proposed (P, T) phase diagram for $YbInCu_4$. LM, HF, IV and FM represent localized moment, heavy-fermion, intermediate-valence and ferromagnetic states, respectively.

5. Summary

The present study has allowed us

(a) to show that the indirect exchange interaction between f moments in $RInCu_4$ compounds is independent of pressure,
(b) to conclude that the large value of the Grüneisen parameter for the Kondo temperature, by itself, is not a convincing cause of the first-order valence phase transition,
(c) to propose a (P, T) phase diagram for $YbInCu_4$, in which the phase sequence is opposite to that for Ce-based systems reflecting the same f-state depopulation with pressure,
(d) to predict a possible ferromagnetic ordering in $YbInCu_4$ at high pressure and
(e) to describe the effect of atomic disorder in $YbInCu_4$, by assuming a spatially inhomogeneous T_V, by a conventional Gaussian distribution.

The magnetovolume effect is recommended as a useful tool for studying further phase diagrams and the nature of the electron interactions in $YbInCu_4$ and related materials.

Acknowledgments

The authors thank N Kim for assistance in sample preparation. This work was partially supported by a Grant-in-Aid for Scientific Research given by the Ministry of Education, Science and Culture of Japan.

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