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1999 J. Phys.: Condens. Matter 11 3405

(http://iopscience.iop.org/0953-8984/11/16/020)

J. Phys.: Condens. Matter **11** (1999) 3405–3412. Printed in the UK PII: S0953-8984(99)00116-2

Possible negative mode–mode coupling in UPd_2Al_3

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Received 14 December 1998

Abstract. We have measured the magnetization $M(H)$ of the heavy-fermion antiferromagnet UPd_2Al_3, and found that the inverse of the gradient of the linear plot of M^2 against H/M, $g(T)$, is negative at low temperatures. As temperature is raised, $g(T)$ shows a rapid increase and approaches zero at around T_{max}, at which temperature the magnetic susceptibility $\chi(T)$ exhibits a maximum. An analysis of the metamagnetic-like magnetization curves observed for high magnetic fields shows a consistent temperature dependence of the relevant coefficient. We have regarded the negative $g(T)$ as an indication of a negative mode–mode coupling, and on the basis of this assumption we have given a possible explanation for the temperature dependence of $\chi(T)$, showing a crossover from the low-temperature itinerant-electron regime to the high-temperature localized-electron regime.

1. Introduction

UPd_2Al_3 is a well-known heavy-fermion superconductor with a hexagonal crystal structure ($P6/mmm$) in which an antiferromagnetic ordered state ($T_N = 14.3$ K) coexists with superconductivity below a transition temperature $T_c = 2$ K [1]. The following characteristic features have been revealed so far:

(a) A large discontinuity in the heat capacity at T_c suggests that the superconductivity is carried by heavy quasiparticles.
(b) Polarized neutron scattering measurements indicate that magnetic moments reside only on uranium sites [2].
(c) High-resolution photoemission [3] and de Haas–van Alphen (dHvA) effect [4] experiments in conjunction with band-structure calculations [5] show 5f electrons to form a band state.
(d) However, a recent inelastic neutron scattering investigation suggests the coexistence of less-delocalized 5f states with well-delocalized 5f states, i.e., duality of the itinerant and localized natures of 5f electrons [6–8].

It is interesting to note that quasielastic spin fluctuations in a magnetic response in the normal state, which change into an inelastic response below T_c [8, 9], probably mediate an attractive pairing interaction of superconductivity [10].

On the other hand, remarkably anisotropic magnetic susceptibilities $\chi(T)$ seemed to be explained by a crystal-field model such as that of a 4f-electron system [11]. $\chi(T)$ in an external magnetic field applied within an easy hexagonal c-plane increases with rising temperature and

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0953-8984/99/163405+08$19.50

exhibits a maximum at around $T_{max} \simeq 37$ K, and then it obeys a Curie–Weiss law above a characteristic temperature of $T_F \sim 70$–100 K with an effective moment corresponding to that of a free ion of $5f^2$ or $5f^3$ configuration [12]. In this localized-moment model, the appearance of the maximum in $\chi(T)$ is ascribed to the crystal-field singlet ground state. It is noted here that the observation of the Curie–Weiss law at high temperatures is consistent with the temperature dependence of the nuclear spin–lattice relaxation rate T_1 of the ^{27}Al nucleus which is independent of temperature above T_F [13].

It may be accepted that the itinerant- and localized-electron pictures are good approximations at low and high temperatures, respectively. Therefore, the problem to be addressed now is how we describe the crossover between these two states. If we consider a 4f-electron system, then we can ascribe it to the Kondo effect: at low temperatures a localized moment is screened by 'Kondo clouds' to form a singlet ground state, and physical properties such as $\chi(T)$ may be understood on the basis of Fermi liquid theory. As temperature is increased, thermal fluctuations may sweep away the Kondo clouds, and then at temperatures much higher than the Kondo temperature the localized moment may appear. In this case we would usually observe sharp energy levels split by crystal electric fields. For UPd_2Al_3, however, no crystal-field transition was detected by an inelastic neutron scattering investigation—at least for an energy transfer less than about 50 meV [14]. Thus, it may be necessary to propose an alternative model which does not assume the existence of sharp energy levels to describe the temperature dependence of $\chi(T)$. This may be related to a more general problem concerning the difference between cerium and uranium compounds—in other words, it may be appropriate to say that in a uranium system even the origin of the heavy masses is a completely open problem, because they may hardly be ascribed to the Kondo effect, due to the strong hybridizations of 5f wave

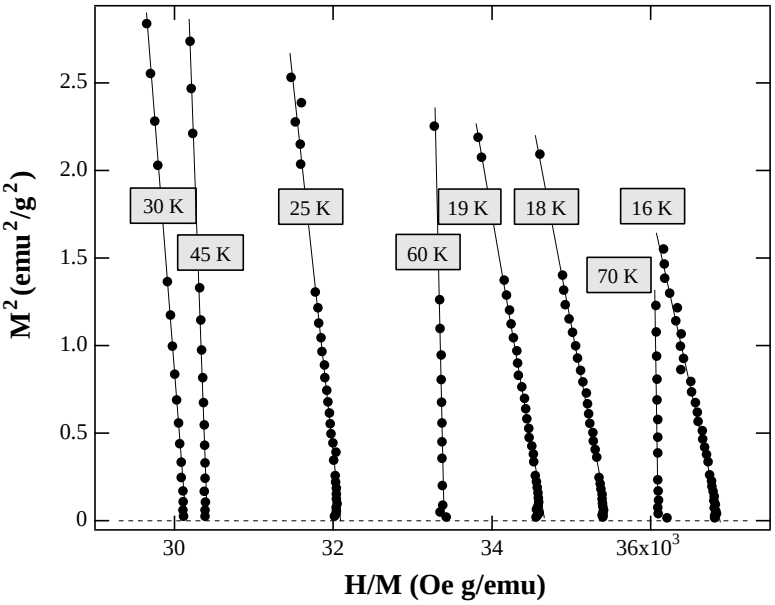

Figure 1. The magnetization for $\boldsymbol{H} \parallel a$-axis in a plot of M^2 against H/M, at a fixed temperature. The intersection of the solid line with the horizontal axis corresponds to $1/\chi(T)$, and the slope is $1/g(T)$, in each case. Note that $g(T)$ is negative at low temperatures.

functions in contrast to the weak hybridizations of 4f wave functions [15].

In the present paper, we will give the experimental results of detailed magnetization measurements up to 230 kOe, and we wish to give a possible interpretation for the crossover based on the results obtained.

2. Experimental results and analysis

Single-crystalline samples were prepared by the Czochralski pulling method using a tri-arc furnace, and a detailed description has been given elsewhere [16, 17]. The magnetization

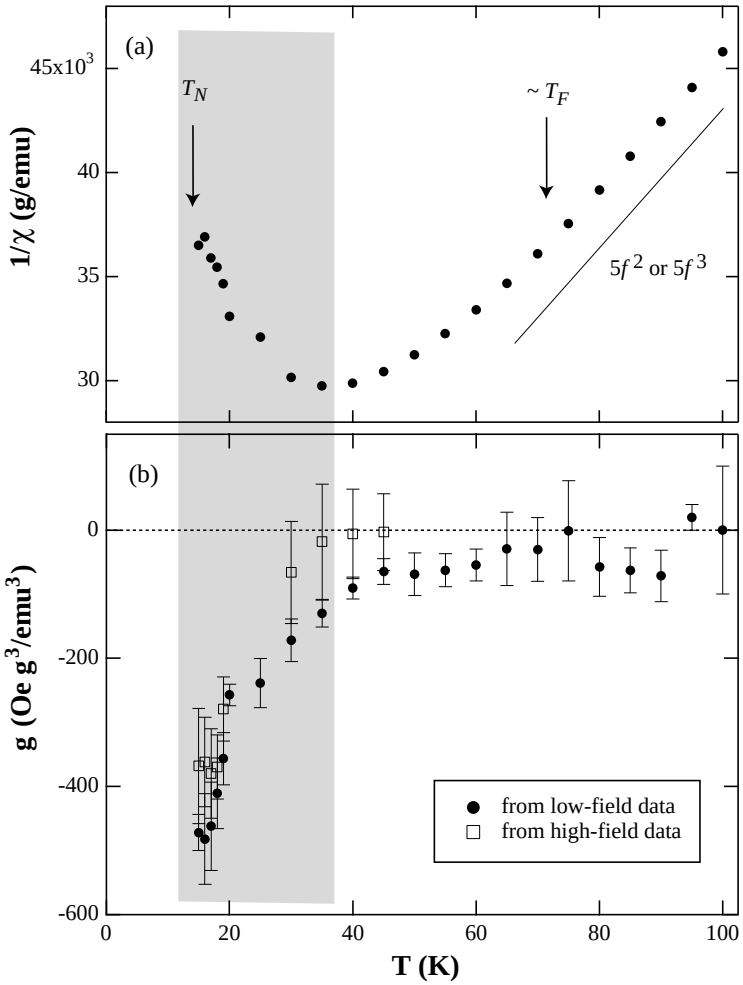

Figure 2. (a) The temperature dependence of the inverse susceptibility, corresponding to $a(T)$ in equation (1). $\chi(T)$ exhibits a maximum at around $T_{max} \simeq 37$ K, and follows a Curie–Weiss law above $T_F \sim 70$ K. (b) Closed circles indicate $g(T)$ obtained from the inverse of the slope of the low-field $M(H)$ given in figure 1, and open squares indicate $g(T)$ obtained from the analysis of the high-field data given in figure 3. We note that $g(T)$ shows a strong temperature dependence below T_{max}.

measurements for the region where $H \leqslant 50$ kOe were made by using a commercial SQUID magnetometer (Cryomagnetics), and those in high fields up to 230 kOe were performed by an induction method in a hybrid magnet at the Institute for Materials Research, Tohoku University. For both cases, external magnetic fields were applied along the crystallographic a-axis.

Figure 1 shows the low-field isothermal magnetization $M(H)$ ($H \leqslant 50$ kOe) in the paramagnetic phase as a plot of M^2 against H/M at a fixed temperature—a so-called Arrott plot. A linear dependence may be observed, as denoted by solid lines, except in a region of very low magnetic fields, where the deviation from the linearity is possibly ascribable to magnetic fields trapped in a superconducting magnet. This proportionality observed at low temperatures (in the paramagnetic state) may suggest that in a free-energy expansion in terms of the magnetization M (see equation (1)) the first two terms alone are sufficient for reproducing the experiment in the magnetic field region concerned:

$$\Delta F(M,T) = \frac{1}{2}a(T)M^2 + \frac{1}{4}g(T)M^4 + \frac{1}{6}c(T)M^6 + \cdots \tag{1}$$

where one may observe from a simple calculation that $a(T)$ is equal to $1/\chi(T)$. In fact, we found it to be consistent with the reported results for $\chi(T)$ [11], as will be shown later. Experimentally, the coefficients $a(T)$ and $g(T)$ are obtained from the intersections of the solid lines with the horizontal axis and the inverse slopes of the lines, respectively. From the measurements of the $M(H)$ curve at each temperature, we get the T-dependences of these parameters, and the results are shown by closed circles in figure 2. One may observe that a minimum appears in the curve representing $a(T) = 1/\chi(T)$ at around $T_{\max} \simeq 37$ K. It is

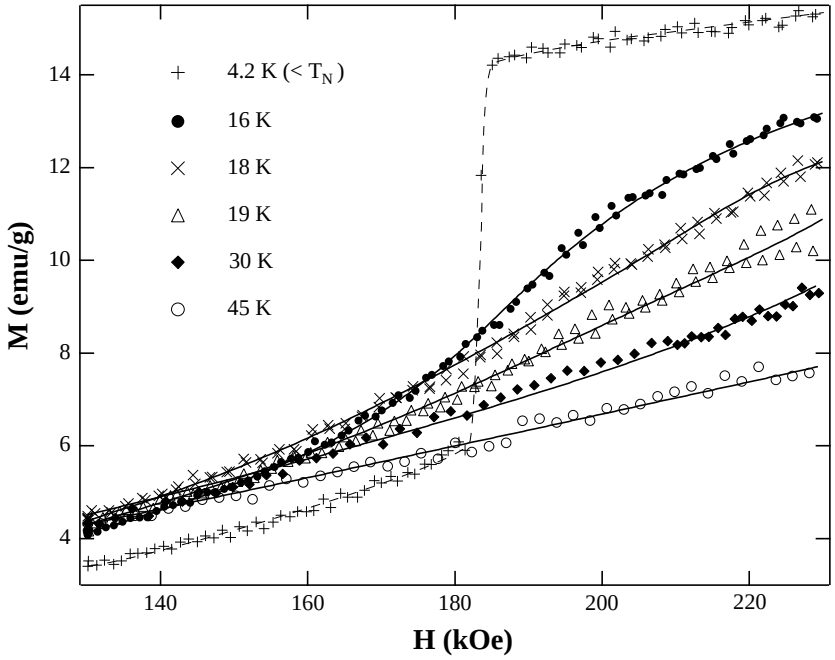

Figure 3. Magnetization curves at several temperatures in the paramagnetic phase near a metamagnetic transition field H_c. Solid curves represent fitted curves, which include contributions up to the M^8-term in equation (2), whereas the broken curve for $T = 4.2$ K (in the antiferromagnetic phase) is a guide to the eye.

quite interesting to note that the coefficient $g(T)$ is negative, increases rapidly with warming, approaches zero at around T_{max}, and remains close to zero above T_{max}. These are new findings and are the most important results in the present paper.

High-field magnetization curves up to 230 kOe at several temperatures are shown in figure 3. At 4.2 K we observe a sharp metamagnetic transition at $H_c \simeq 185$ kOe with a very small hysteresis less than 1 kOe, which is another characteristic of UPd_2Al_3 [18]. As temperature is raised, the transition becomes broadened, and then at $T = 13$–14 K ($<T_N$) only an 'S-shaped' curvature is left (not shown here). We stress that, as can be seen in figure 3, the S-shaped feature apparently survives to temperatures above T_N ($<T_{max}$), as previously pointed out by Oda and co-workers (including some of the present authors) [18].

A least-squares fitting of the high-field magnetization data to equation (2) also yields a negative value for $g(T)$ and $ac/g^2 = 0.78$ at $T = 15$ K:

$$\frac{H}{M} = a(T) + g(T)M^2 + c(T)M^4 + \cdots. \tag{2}$$

As temperature is raised, the absolute value of $g(T)$ is reduced and the value of ac/g^2 approaches unity. This temperature dependence, denoted by open squares, seems to coincide with that deduced from the low-field magnetization data (closed circles), although $g(T)$ estimated from the high-field data seems to show a relatively rapid increase.

The experimental results for $g(T) \sim 0$ above T_{max} imply that there is no trace of the S-shaped feature in $M(H)$ there. Therefore one can see that the appearance of the metamagnetic feature correlates well with T_{max}. In fact, such a correlation has often been suggested, especially for $CeRu_2Si_2$ [19], a prototypical metamagnet, but we consider that the present data incontrovertibly demonstrate the relation between the metamagnetism and the maximum in $\chi(T)$ for UPd_2Al_3.

3. Discussion

In this section, we discuss a possible interpretation for the experimental results obtained in the present investigation. First, let us consider the localized-moment model, which was introduced by Grauel and co-workers (including some of the present authors again) to explain the anisotropic temperature dependence of the magnetic susceptibility. It was pointed out by Sato *et al* that the metamagnetic jump at H_c might be explainable using the crystal-field model [20]. However, the present experimental results shown in figure 2 are qualitatively incompatible with the crystal-field model, which takes no account of any interactions between uranium moments: in the crystal-field model the magnetization at absolute zero temperature is linear in the external magnetic field below H_c, reflecting the H^2-dependence of the magnetic energy of the singlet ground state, and at finite temperatures $M(H)$ may show an S-shaped curvature because the magnetic excited states are mixed into the non-magnetic ground state. This temperature dependence is opposite to that observed for $g(T)$. Furthermore, the inelastic neutron scattering measurements made by Krimmel *et al* did *not* show any sharp crystal-field-level splitting, implying that the crystal-field scheme loses its background. A gap energy of the spin waves was estimated by Süllow *et al* on the basis of the same crystal-field model [21], although a major part of their paper has nothing to do with the crystal-field model, and the estimated gap energy seems not to be in quantitative agreement with that directly obtained by recent inelastic neutron scattering measurements [6–8]. As far as we are aware, there are no experimental results that support the crystal-field model consistently.

One could consider the antiferromagnetic correlations to be responsible for both the appearance of the maximum in $\chi(T)$ and the metamagnetism. In this model, at low T (at low H)

the uniform magnetic susceptibility may be suppressed by the antiferromagnetic correlations, and increasing the temperature (external magnetic fields) may destroy the correlation, yielding an increase of the magnetic susceptibility. However, as long as one confines consideration to within the framework of the localized-electron model, i.e., with a *fixed size* of magnetic moment, one cannot explain the experimental observation that the low-T staggered moment, 0.85 μ_B, is only a quarter of that deduced from the high-T Curie–Weiss law, 3.5 μ_B, or the fact that the staggered moment is nearly half the magnetization just above H_c, 1.5 μ_B/U.

The most important physical implication is, we believe, that the system is in the Fermi liquid regime: in figure 2(a) we assign T_F, below which $\chi(T)$ deviates from the Curie–Weiss law and the Korringa law (T_1T = constant) holds [13], to a value of about 70 K. As mentioned in the introduction, the temperature region $T > T_F$ is characterized by T_1 = constant and the Curie–Weiss law, implying that the system can be regarded as a set of localized moments, while the low-temperature state for $T < T_F$ may be described by Fermi liquid theory. It should be noted that T_{max} is lower than T_F, and that it is in the region $T < T_F$ that the metamagnetism is observed. Therefore, it may be necessary to look for an alternative interpretation of the existence of the maximum in $\chi(T)$ and the metamagnetic behaviour.

Once we work from the basis of an itinerant-electron picture, instead of the fixed-moment model, the above problems may be resolved. The coefficients $a(T)$, $g(T)$, and $c(T)$ can take either positive or negative values, depending on the electronic structures near the Fermi level E_F. The sign and magnitude of $g(T)$, for instance, may be determined from the energy dependence of the (local) density of states (DOS) around E_F, and the negative sign of $g(T)$ may derive from a negative curvature of the DOS at E_F. Such a negative curvature of the DOS at E_F may signify a negative mode–mode coupling, which plays a role in coupling spin-fluctuation modes with different wave vectors characterized by $\boldsymbol{q}$. To understand the implication of this, we consider the following energy functional of spin fluctuations [22]:

$$\Psi = \sum_{q} \frac{1}{2\chi_q} |\boldsymbol{M}_q|^2 + \frac{1}{4} g \sum_{q,q',q''} (\boldsymbol{M}_q \cdot \boldsymbol{M}_{-q'})(\boldsymbol{M}_{q''} \cdot \boldsymbol{M}_{q'-q''-q}) + \cdots \tag{3}$$

where $\boldsymbol{M}_q$ denotes spin fluctuations. In the case where $\boldsymbol{M}_q$ is small, the first term may dominate over higher terms. If we assume that $\boldsymbol{M}_q$ is increased, for instance by an increase of temperature or external magnetic field, the contribution from the higher terms can no longer be neglected. In particular, when the mode–mode coupling is negative, one may see an interesting effect: the negative mode–mode coupling *enhances* the amplitude of the spin fluctuations, because the second term lowers the free energy. Furthermore, since the coefficient of the first term, $1/\chi_q$, is *inversely* proportional to $\langle|\boldsymbol{M}_q|^2\rangle$, the increase of the spin-fluctuation amplitude makes the first-term contribution less important compared to the higher-term contributions, which may accelerate the enhancement of the spin-fluctuation amplitudes. Finally, such an enhancement process may lead to a saturation of the local amplitude of the spin fluctuations at a certain temperature T_{max}. This may explain why the magnetic susceptibility increases rapidly with increasing temperature. When temperature is further increased above T_{max}, where $g(T)$ is nearly zero, the Curie–Weiss-like behaviour may be observed on the high-temperature side, because the amplitude is fixed at its maximum value and thus only the orientation can be changed.

On the basis of the same idea, one is also able to explain easily the appearance of the metamagnetic behaviour. In the case where $a > 0$, $g < 0$, and $c > 0$, it may be seen that $\Delta F(M, T)$ can have two minima, at $M = 0$ and at M_0 ($\neq 0$), at a fixed temperature [24]. The latter state (M_0) can be stable or metastable, depending on the magnitude of ac/g^2: in the case where $9/20 > ac/g^2 > 3/16$, it is metastable and is stabilized by applying external magnetic fields, and at a critical field the metamagnetic transition from the paramagnetic to

the ferromagnetic state is induced. When $ac/g^2 > 9/20$, which is the case now, as mentioned above, $M(H)$ does not show such a metamagnetic transition, and only increases monotonically with increasing H; but as long as $g < 0$, the S-shaped curvature remains observable.

Therefore, we believe that this simple interpretation is compatible with all results reported so far, although a quantitative theoretical investigation is required to confirm it, such as was performed for FeSi [22, 23].

In the introduction, section 1, we mentioned the dual nature of 5f electrons. In the above interpretation, we only consider the itinerant component of the 5f electrons. Theoretical work on the magnetization as a function of temperature and magnetic field in the Fermi liquid regime was performed by Miyake and Kuramoto on the basis of the duality model [25]. According to their theoretical calculation,

(a) a necessary condition for the metamagnetic behaviour to occur is that the second derivative of the DOS of the renormalized f electrons at E_F must be positive,

(b) the larger χ' is, the more readily the metamagnetic behaviour occurs, where χ' parametrizes the extent of suppression of the uniform magnetization due to the antiferromagnetic correlations among adjacent spin pairs (i.e., the localized component of the 5f electrons), and

(c) the positive slope of the χ–T curve observed for $T < T_F$ can be explained by taking into account the mode–mode coupling effect of the spin fluctuations, where the predominant mode–mode coupling term in the limit $T \ll T_F$ is negative, corresponding to a positive second derivative of the DOS.

All of these calculated results are consistent with the present interpretation.

In conclusion, we have found that the coefficient of the M^4-term in the free-energy expansion in terms of M is negative at low temperatures, increases rapidly, and approaches zero with increasing temperature up to T_{max}, at which temperature the magnetic susceptibility exhibits its maximum. We regarded this as a manifestation of the negative mode–mode coupling among spin fluctuations. On the basis of these results, we proposed a possible interpretation to explain the crossover between the low-T itinerant-electron and high-T localized-electron regimes. In the present paper, we disregarded three aspects: the anisotropy of the magnetic susceptibility; the magnetoelastic effect; and the metamagnetic *phase transition* between the antiferromagnetic ordered phase and the unresolved high-magnetic-field one (possibly a ferromagnetic phase with a large amplitude of the local magnetization). As regards the first aspect, we merely refer the reader to the pioneering work on band-structure calculations for UPd_2Al_3 performed by the Darmstadt group, in which the anisotropy of the magnetic susceptibility was calculated within the framework of the band-structure calculation, i.e., in the itinerant-electron picture [5]. The second aspect may be expected to be a secondary effect, as pointed out in reference [25], although it should be taken into account in a thorough quantitative discussion. To discuss the third aspect, we would need to construct a discussion similar to that given in section 4.7 in reference [22]. That is beyond the scope of the present work, and remains a problem for the future.

Acknowledgments

We wish to acknowledge the support of the technical staff at the Institute for Materials Research, Tohoku University, in using the hybrid magnet. One of the authors (NKS) thanks K Miyake and S Süllow for helpful discussions.

References

[1] Geibel C, Schank C, Thies S, Kitazawa H, Bredl C D, Böhm A, Rau M, Grauel M, Caspary R, Helfrich R, Ahlheim U, Weber G and Steglich F 1991 *Z. Phys.* B **84** 1

[2] Paolasini L, Paixao J, Lander G H, Burlet P, Sato N and Komatsubara T 1993 *J. Phys.: Condens. Matter* **5** 8905

[3] Takahashi T, Sato N, Yokoya T, Chainani A, Morimoto T and Komatsubara T 1996 *J. Phys. Soc. Japan* **65** 156

[4] Inada Y, Aono H, Ishiguro A, Kimura J, Sato N, Sawada A and Komatsubara T 1994 *Physica* B **199+200** 119

[5] Knöpfle K, Mavromaras A, Sandratskii L M and Kübler J 1996 *J. Phys.: Condens. Matter* **8** 901 and references therein

[6] Sato N, Aso N, Lander G H, Roessli B, Komatsubara T and Endoh Y 1997 *J. Phys. Soc. Japan* **66** 1884

[7] Sato N, Aso N, Lander G H, Roessli B, Komatsubara T, Endoh Y and Sakai O 1998 *J. Alloys Compounds* **271–273** 433

[8] Bernhoeft N, Sato N, Roessli B, Aso N, Hiess A, Lander G H, Endoh Y and Komatsubara T 1998 *Phys. Rev. Lett.* **81** 4244

[9] Metoki N, Haga Y, Koike Y, Aso N and Onuki Y 1997 *J. Phys. Soc. Japan* **66** 2560

[10] Sato N, Aso N, Lander G H, Roessli B, Komatsubara T and Endoh Y 1997 *J. Phys. Soc. Japan* **66** 2981

[11] Grauel A, Böhm A, Fischer H, Geibel C, Köhler R, Modler R, Schank C, Steglich F, Weber G, Komatsubara T and Sato N 1992 *Phys. Rev.* B **46** 5818

[12] Sato N, Aso N, Tateiwa N, Koga N, Komatsubara T and Metoki N 1997 *Physica* B **230/232** 36

[13] Kyogaku M, Kitaoka Y, Asayama K, Geibel C, Schank C and Steglich F 1992 *J. Phys. Soc. Japan* **61** 2660

[14] Krimmel A, Loidl A, Eccleston R, Geibel C and Steglich F 1996 *J. Phys.: Condens. Matter* **8** 1677

[15] See, for example, Steglich F, Buschinger B, Gegenwart P, Geibel C, Helfrich R, Hellman P, Lang M, Link A and Modler R 1995 *Proc. Int. Conf. on Physical Phenomena at High Magnetic Fields II (Tallahassee, FL)* (Singapore: World Scientific) p 125

[16] Sato N, Sakon T, Takeda N, Komatsubara T, Geibel C and Steglich F 1992 *J. Phys. Soc. Japan* **61** 32

[17] Sato N, Inada Y, Sakon T, Imamura K, Ishiguro A, Kimura J, Sawada A, Komatsubara T, Matsui H and Goto T 1994 *IEEE Trans. Magn.* **30** 1145

[18] Oda K, Kumada T, Sugiyama K, Sato N, Komatsubara T and Date M 1994 *J. Phys. Soc. Japan* **63** 3115 and references therein

[19] For example, Mignot J-M, Flouquet J, Haen P, Lapierre F, Puech L and Voiron J 1988 *J. Magn. Magn. Mater.* B **76+77** 97

[20] Sato N, Sakon T, Imamura K, Inada Y, Aono H and Komatsubara T 1993 *Physica* B **186–188** 195

[21] Süllow S, Janossy B, van Vliet G L E, Nieuwenhuys G J, Menovsky A A and Mydosh J A 1996 *J. Phys.: Condens. Matter* **8** 729

[22] Moriya T 1985 *Spin Fluctuations in Itinerant Electron Magnetism* (Berlin: Springer)

[23] Edwards D M 1982 *J. Phys. F: Met. Phys.* **12** 1789

[24] For example, Yamada H 1993 *Phys. Rev.* B **47** 11 211

[25] Miyake K and Kuramoto Y 1991 *Physica* B **171** 20