Magnetic order of UPt_3 in high magnetic fields

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The weak magnetic order of the heavy-fermion superconductor UPt_3 has been investigated by elastic neutron-scattering measurements in magnetic fields up to 12 T along the *a* and *c* axes of the hexagonal crystal structure. The small antiferromagnetically ordered moment of $0.02\mu_B/(\mathrm{U\ atom})$ shows only a weak dependence on the applied magnetic field and no sign of a domain repopulation for $B\|a$. In high magnetic fields an increase in the magnetic correlation length is observed for magnetic fields along the *c* axis.
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I. INTRODUCTION

One of the central problems in the understanding of the heavy-fermion superconductors is the role of the antiferromagnetic order. So far, no systematics have been observed in the nature of the magnetism in these systems; UPd_2Al_3 (Ref. 1) shows a relatively large ordered moment of $0.85\mu_B/(\mathrm{U\ atom})$, UNi_2Al_3 (Ref. 2) a reduced moment of $0.12\mu_B/(\mathrm{U\ atom})$ with an incommensurate magnetic structure, URu_2Si_2 (Ref. 3) a strongly reduced moment of $0.03\mu_B/(\mathrm{U\ atom})$, while for UBe_{13} (Ref. 4) no magnetic order has been observed. The magnetic interactions in heavy-fermion systems are governed by a hybridization of the *f* electrons and the conduction electrons that lead to a competition between the Kondo screening and the indirect exchange (Ruderman-Kittel-Kasuya-Yosida) interaction of the *f* electrons. As a consequence, a strong enhancement of the quasiparticle mass at low temperatures, and a rich variety of magnetic structures, is observed. The relation of the antiferromagnetic order to the superconductivity, and their coexistence at low temperatures, is one of the major issues yet to be resolved experimentally and theoretically.

For UPt_3, elastic neutron-scattering measurements[5] revealed the onset of antiferromagnetic order below $T_N=6\,\mathrm{K}$ with an unusually small ordered moment of $0.02\mu_B/(\mathrm{U\ atom})$. The antiferromagnetic order has a propagation vector of $\mathbf{q}=(\frac{1}{2},0,0)$ with the ordered moment along the propagation vector in the basal plane of the hexagonal close-packed crystal structure (space group $P6_3/mmc$). The magnetic Bragg peaks are not resolution limited but show a Lorentzian broadening that corresponds to a finite correlation length of the order of $\xi\approx 250$ Å. The weak antiferromagnetic order, observed by neutron-scattering measurements, has not been observed by any other technique. Careful specific-heat measurements[6] failed to detect any anomaly around T_N with a precision of 0.1%. The proximity to a magnetic instability is nicely demonstrated by the effect of doping with small concentrations of Pd,[7] which causes a substantial increase in the size of the ordered moment with a maximum value of $0.6\mu_B/(\mathrm{U\ atom})$ for 5% Pd doping. Magnetization measurements[8] in high magnetic fields revealed a metamagnetic transition at $B^*=20$ T for $B\|a$. Above the metamagnetic transition the heavy-fermion state is suppressed, and the induced magnetization corresponds to a relatively large value of $0.7\mu_B/(\mathrm{U\ atom})$.

Due to the small size of the ordered moment, the magnetic phase diagram for high magnetic fields is difficult to measure and has only partly been determined in previous works. For magnetic fields along the *c* axis, Bruls *et al.*[9] performed elastic neutron-scattering measurements and found that the antiferromagnetic transition temperature shows a small suppression from $T_N=5.5$ K in zero field to $T_N=4.1$ K at a field of 10 T. In addition, sound-velocity measurements as a function of magnetic field[9,10] showed a pronounced minimum at a field that decreases with increasing temperature and vanished around T_N. The minimum was interpreted as the critical field for the antiferromagnetic order and also exists for $B\|a$, where it occurs at 5 T at $T=0$ K, compared to 10.5 T for $B\|c$.

At low temperatures, UPt_3 shows unique unconventional superconducting properties. The superconducting transition at $T_c^+=0.50$ K is followed by a second superconducting transition at $T_c^-=0.44$ K. As a function of magnetic field and temperature, an exotic superconducting phase diagram[11–13] is observed with three different superconducting phases that meet at a tetracritical point. Neutron-scattering[14,15] and magnetic x-ray-diffraction[15] measurements at low temperatures revealed that the superconductivity coexists with the antiferromagnetic order. Combined elastic neutron-scattering and specific-heat measurements[16] under hydrostatic pressure showed a direct relation between the size of the weak ordered moment and the splitting of the superconducting transition temperatures T_c^+ and T_c^-. By applying a pressure of 3–4 kbar, the antiferromagnetic order is fully suppressed and the two superconducting transition temperatures merge.

In order to describe the exotic superconducting phase diagram, two main scenarios have been proposed. In the first scenario,[17] the small splitting of the superconducting transi-

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tion temperatures T_c^+ and T_c^- is considered accidental and due to a near degeneracy of two different representations of the order parameter. In the second scenario,[18,19] the double transition is caused by a symmetry-breaking field that lifts the degeneracy of the components of a vector order parameter within a single representation. The most likely candidate for the symmetry-breaking field is the weak antiferromagnetic order, as the magnetic order lowers the symmetry of the system. In this case, the superconducting transition temperatures are sensitive to the orientation of the applied magnetic field with respect to the ordered moment. For magnetic fields within the basal plane, the three magnetic domains may each have a slightly different superconducting phase diagram.

In view of the important implications to the understanding of the superconductivity, Lussier *et al.*[20] studied the effect of an applied magnetic field within the basal plane on the antiferromagnetic order. Their elastic neutron-scattering measurements at $T=1.8$ K showed no change in the antiferromagnetic order and, in particular, no significant domain repopulation for applied magnetic fields up to 3.2 T $(B\perp c)$, which includes the entire field range of interest for the superconductivity $(B_{c2}=2.2\ \mathrm{T})$.[11–13] In order to explain the absence of domain repopulation for magnetic fields within the basal plane $(B\perp c)$, Lussier *et al.*[20] proposed the existence of a triple-*q* structure. The question whether the magnetic order corresponds to a single-*q* or a triple-*q* structure is crucial for the understanding of the unconventional superconductivity because the magnetic order determines the symmetry of the system. For a single-*q* structure, the magnetic unit cell has twice the volume of the structural unit cell and a D_{2h} symmetry, while the magnetic unit cell for a triple-*q* structure is four times bigger than the structural unit cell, but does not affect the D_{6h} symmetry of the lattice.

Recently, Fomin and Flouquet[21] proposed an alternative scenario that ascribes the weak magnetic contribution, observed in elastic neutron-scattering measurements below 6 K, to the development of magnetic fluctuations. These fluctuations are sufficiently slow to appear static on the time scale of a neutron-scattering experiment. This is compatible with zero-field muon-spin relaxation measurements[22,23] that fail to see the antiferromagnetic order on much longer time scales.

In order to study the field dependence of the weak antiferromagnetic order, we performed elastic neutron-scattering measurements in applied magnetic fields up to 12 T for $B\parallel a$ and $B\parallel c$. Earlier measurements were performed in magnetic fields up to 3.2 T (Ref. 20) for $B\parallel a$ and, respectively, 2.5 T (Ref. 14) and 10 T (Ref. 9) for $B\parallel c$. Our extended field range allows us to test the interpretation of the sound-velocity measurements[9,10] that predict a critical field of 5 T for $B\parallel a$ and 10.5 T for $B\parallel c$. In addition, it allows us to study a possible domain repopulation at high magnetic fields $(B\parallel a)$. If a sizable domain repopulation is indeed observed for applied magnetic fields along the *a* axis, a triple-*q* structure can be excluded for the antiferromagnetic order with important implications for the unconventional superconductivity.

II. EXPERIMENTAL PROCEDURE

The neutron-scattering experiments were performed on two high-quality single crystals of masses 1.5 g and 8 g, prepared under ultrahigh vacuum by the Czochralski technique and annealed for 6 days at a temperature of 950 °C. Resistivity measurements confirmed the good crystal quality, with a residual resistance ratio of RRR=586 for the 1.5 g sample and RRR=671 for the 8 g sample for an electrical current along the *a* axis. The superconducting transition temperature of both samples was $T_c^+=0.53$ K. The antiferromagnetic order in UPt_3 was studied by elastic neutron-scattering measurements on the cold triple axis spectrometer IN 14 of the ILL. We used an initial wave vector of $k_i=1.48\ \mathrm{\AA}^{-1}$, a collimation of 37′-40′-60′-60′, with a beryllium filter before, and a graphite filter after, the sample. The crystals were mounted in a 12 T vertical cryomagnet and aligned with, respectively, the *a* axis (1.5 g sample) and the *c* axis (8 g sample) along the applied magnetic field. An applied magnetic field along the *c* axis would not be expected to influence the population of the magnetic domains while an applied magnetic field along the *a* axis would normally favor one of the three domains. For a vertical magnetic field along the *a* axis, only the energetically most favorable domain is located in the scattering plane of the spectrometer.

III. RESULTS

Measurements of the magnetic Bragg-peak intensity were performed for applied magnetic fields up to 12 T with $B\parallel a$ and $B\parallel c$. In Figs. 1 and 2, scans through the magnetic Bragg peak at $\mathbf{Q}=(\frac{1}{2},0,1)$ for $B\parallel a$ and $\mathbf{Q}=(\frac{1}{2},-\frac{3}{2},0)$ for $B\parallel c$ are shown as a function of the crystal rotation angle ω. In an applied magnetic field of 10 T, the magnetic Bragg-peak intensity slightly increases for $B\parallel a$ and slightly decreases for $B\parallel c$. The field dependence of the integrated Bragg-peak intensity is shown in Fig. 3. The integrated intensity is normalized to its zero-field value and compared to earlier low-field measurements of Lussier *et al.*[20] $(B\parallel a)$ and Bruls *et al.*[9] $(B\parallel c)$. The nearly constant Bragg-peak intensity for $B\parallel a$ indicates the absence of a substantial domain repopulation. In case of a complete domain repopulation, the intensity of the magnetic Bragg peak at $\mathbf{Q}=(\frac{1}{2},0,1)$ is expected to increase by a factor of 3 due to the depopulation of the other two domains.

It is known from earlier work that the magnetic Bragg peaks are not resolution limited, but show a Lorentzian broadening that corresponds to a finite magnetic correlation length of the order of $\xi\approx250$ Å.[5,14] In Table I the transverse $(\perp\mathbf{Q})$ magnetic correlation length $\xi_\perp$, deduced from the Lorentzian broadening of our ω scans, is shown for the magnetic Bragg peaks at $\mathbf{Q}=(\frac{1}{2},0,1)$ for $B\parallel a$ and at $\mathbf{Q}=(\frac{1}{2},-\frac{3}{2},0)$ for $B\parallel c$. In zero field the transverse correlation lengths for $\mathbf{Q}=(\frac{1}{2},0,1)$ and $\mathbf{Q}=(\frac{1}{2},-\frac{3}{2},0)$ show a sizable difference that may indicate an anisotropic magnetic correlation length. However, one cannot exclude that this difference is due to a sample dependence, as the measurements have been performed on two different samples. In high magnetic fields the correlation length in the basal plane shows an increase for $B\parallel c$. The origin of the observed finite magnetic correlation length is still unknown.

In Fig. 4, the integrated intensity of the magnetic Bragg peaks is shown as a function of temperature. The antiferromagnetic transition temperatures of, respectively, 6.5 K (1.5

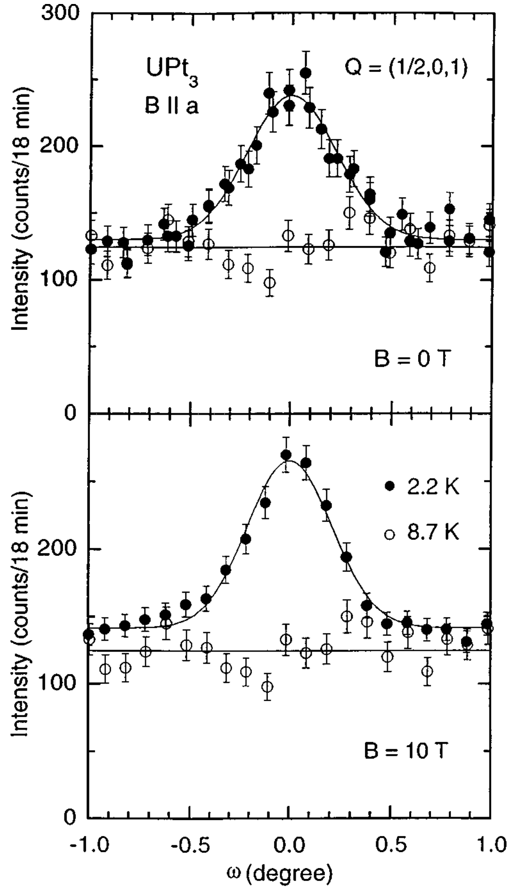

FIG. 1. Magnetic Bragg peaks at $\mathbf{Q}=(\frac{1}{2},0,1)$ as a function of the crystal rotation angle ω in a magnetic field of $B=0$ and 10 T for $B\|a$. For comparison, the high-temperature scan ($T>T_N$) in zero field is included.

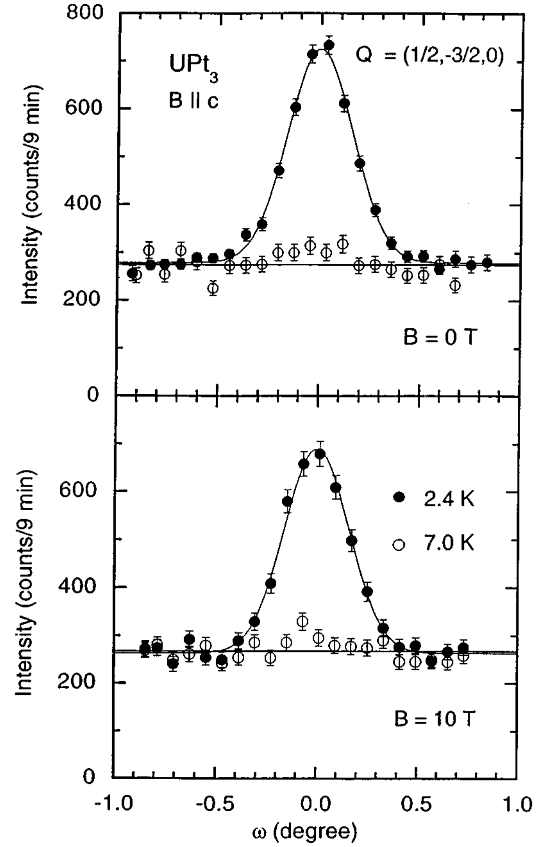

FIG. 2. Magnetic Bragg peaks at $\mathbf{Q}=(\frac{1}{2},-\frac{3}{2},0)$ as a function of the crystal rotation angle ω in a magnetic field of $B=0$ and 10 T for $B\|c$. For comparison, the high-temperature scans ($T>T_N$) are included.

g sample) and 6.6 K (8 g sample) in zero field show a suppression of 0.7 K ($B\|a$) and 0.4 K ($B\|c$) in a magnetic field of 10 T. Although the transition temperature decreases in a magnetic field, the integrated Bragg-peak intensity increases at low temperatures for magnetic fields along the a axis.

IV. DISCUSSION

Our measurements of the magnetic Bragg-peak intensity as a function of applied magnetic fields for $B\|a$ and $B\|c$ show that a field of 12 T is not sufficient to suppress the weak antiferromagnetic order. This rules out a previous interpretation of ultrasound data by Bruls *et al.*[9,10] Their measurements of the sound velocity as function of magnetic field show a pronounced minimum at 5 T for $B\|a$ and 10.5 T for $B\|c$, which they ascribe to the critical field for the antiferromagnetic order. Although a similar minimum has been observed in the magnetostriction,[24] our present elastic neutron-scattering measurements indicate that it is not related to the critical field of the antiferromagnetic order.

If we only consider the effect of an applied magnetic field on the small antiferromagnetically ordered moments, the domain repopulation for $B\|a$ can be estimated in terms of a molecular-field interaction $\lambda=T_N/C$, where $C=ng^2\mu_{\rm eff}{}^2/3k_B$ is the effective Curie constant with a magnetic ion density $n=N/V$. The energy gain (per volume) due to the magnetic torque of the magnetic field on the small antiferromagnetically ordered moment $\mu_{\rm eff}$ is given by $E(B)-E(0)=-B_\perp^2/2\lambda$, where $B_\perp$ is the component of the applied magnetic field perpendicular to the propagation vector. For $B\|a$, the energy difference between the energetically most favorable domain ($B_\perp=B$) and the two remaining domains ($B_\perp=B/2$) is $\Delta E=3B^2/8\lambda$. The population of the energetically most favorable domain is $a_0=1/[1+2\exp(-\Delta E/nk_BT)]$. For the relevant parameters of our system, the estimated domain repopulation turns out to be negligible (10^{-4}), which is consistent with the observed weak-field dependence of the Bragg-peak intensity for $B\|a$. In order to obtain an increase of 10% in population of the most favorable domain at $T=1$ K, a magnetic field of $B=200$ T is needed in the absence of pinning for the magnetic domain walls. The absence of a significant domain repopulation for $B\|a$ does not allow a discrimination between a single-q or a triple-q structure. The weak field dependence of the magnetic Bragg-peak intensity is expected for a triple-q structure[25] but not inconsistent with a single-q structure due to the small magnetic torque of the applied magnetic field on the ordered moment. The critical field for a transition to a spin aligned paramagnetic phase can be estimated by $B_{\rm cr}=2M_0\lambda$, where $M_0=n\mu_{\rm eff}/2$ is the sublattice magnetization. For a field-independent effective moment, the critical field would correspond to a value of $B_{\rm cr}\approx1250$ T.

The effect of an applied magnetic field is not limited to the antiferromagnetically ordered moment. The ordered mo-

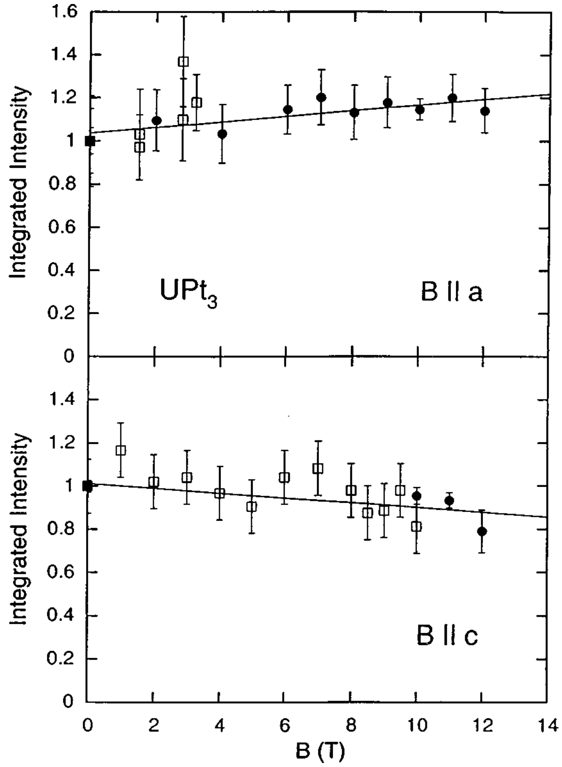

FIG. 3. Relative integrated intensity of the magnetic Bragg peak as a function of applied magnetic field up to 12 T ($B\|a$ and $B\|c$). The measured data points (solid circles) are normalized to the zero field value and compared with earlier low-field measurements (open squares) of Lussier *et al.* (Ref. 20) ($B\|a$) and Bruls *et al.* (Ref. 9) ($B\|c$).

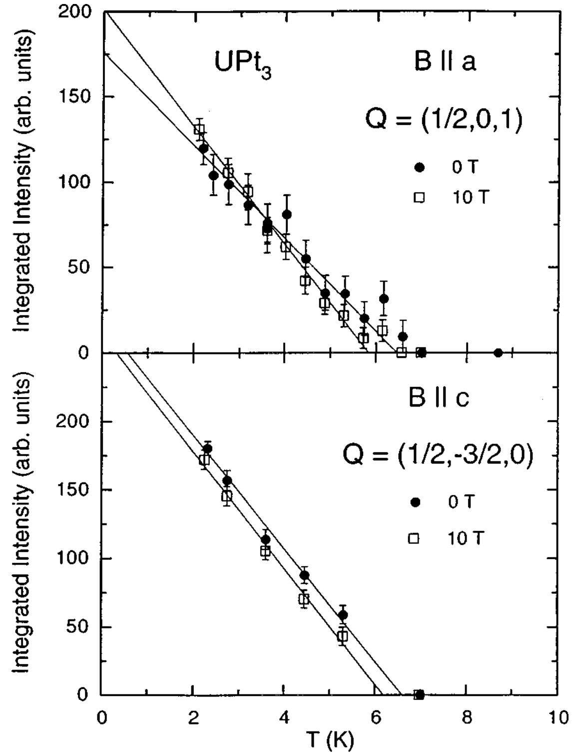

FIG. 4. Temperature dependence of the integrated intensity of the magnetic Bragg peak in a magnetic field of $B=0$ and 10 T ($B\|a$ and $B\|c$).

ment of $0.02\mu_B$/(U atom) is only a fraction of the moment on the uranium ions at low temperatures [$\approx 1\mu_B$/(U atom)]. The hybridization with the conduction electrons causes strong Kondo fluctuations that screen the moments and lead to the formation of heavy quasiparticles. High-field magnetization measurements show that an applied magnetic field of 12 T causes an induced magnetic moment of 0.2 (0.1) μ_B on the uranium ions for $B\|a$ ($B\|c$).[8] This induced magnetic moment is not expected to play a significant role in the heavy-fermion properties, as specific-heat measurements in high magnetic fields[26] show that the effective mass of the heavy quasiparticles is not affected by an applied magnetic field until the metamagnetic transition at $B^*=20$ T ($B\|a$) is approached. Apparently, an applied magnetic field up to 12 T does not change the balance between the indirect magnetic exchange and the Kondo screening of the uranium moments, and does not significantly change the size of the antiferromagnetically ordered moment. The small increase in the ordered moment, observed for applied magnetic fields along the *a* axis, suggests a weak change in anisotropy of the magnetic correlations. In combination with the reduction of the antiferromagnetic ordering temperature, this can lead to an unusual crossing of the magnetic intensity curves as a function of temperature, as observed in Fig. 4 for applied magnetic fields of 0 and 10 T along the *a* axis.

TABLE I. Transverse magnetic correlation length $\xi_\perp$ at $\mathbf{Q}=(\frac{1}{2},0,1)$ for $B\|a$ and at $\mathbf{Q}=(\frac{1}{2},-\frac{3}{2},0)$ for $B\|c$ as a function of the applied magnetic field.

B	$B\|a$	$B\|c$
T	$\xi_\perp$ Å	$\xi_\perp$ Å
0	216(35)	388(35)
10	236(26)	393(54)
11		465(66)
12		549(112)

If the magnetic signal below 6 K is in fact not static, but corresponds to slow magnetic fluctuations as recently proposed by Fomin and Flouquet,[21] a weak field dependence is expected. The magnetic field in this scenario only modifies the excitation spectrum of the fluctuations. Unfortunately, the model still has a qualitative nature and does not give quantitative predictions for the field dependence of these slow magnetic fluctuations. In order to test whether the magnetic signal that develops below 6 K corresponds to static magnetic order or slow fluctuations, experiments on longer time scales are required. Muon-spin relaxation measurements[22,23] indeed fail to observe the ordered moment, although this can also be related to a canceling of the dipolar magnetic field at the stopping site of the muon. An attempt to clarify this question by high-resolution neutron spin-echo measurements on the IN11 spectrometer at the ILL was unsuccessful because of the low intensity of the magnetic Bragg peaks.

V. CONCLUSIONS

In conclusion, we have measured the weak ordered moment of UPt_3 with elastic neutron-scattering measurements in magnetic fields up to 12 T for $B \| a$ and $B \| c$. The applied magnetic field was not sufficient to suppress the magnetic field or observe a repopulation of magnetic domains. The integrated magnetic Bragg-peak intensity showed a small increase for $B \| a$ and a small decrease for $B \| c$. These results do not allow us to make a distinction between a single-q structure or a triple-q structure due to the weak magnetic torque of the applied magnetic field on the ordered moment. In high magnetic fields, a small increase in the magnetic correlation length is observed for $B \| c$.

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[1] A. Krimmel, P. Fischer, B. Roessli, H. Maletta, C. Geibel, C. Schank, A. Loidl, and F. Steglich, Z. Phys. B **86**, 161 (1992).
[2] A. Schröder, J. G. Lussier, B. D. Gaulin, J. D. Garrett, W. J. L. Buyers, L. Rebelsky, and S. M. Shapiro, Phys. Rev. Lett. **72**, 136 (1994).
[3] C. Broholm, J. K. Kjems, W. J. L. Buyers, P. Matthews, T. T. M. Palstra, A. A. Menovsky, and J. A. Mydosh, Phys. Rev. Lett. **58**, 1467 (1987).
[4] A. de Visser, N. H. van Dijk, K. Bakker, J. J. M. Franse, A. Lacerda, J. Flouquet, Z. Fisk, and J. L. Smith, Phys. Rev. B **45**, 2962 (1992).
[5] G. Aeppli, E. Bucher, C. Broholm, J. K. Kjems, J. Baumann, and J. Hufnagl, Phys. Rev. Lett. **60**, 615 (1988).
[6] R. A. Fisher, B. F. Woodfield, S. Kim, N. E. Phillips, L. Taillefer, A. L. Giorgi, and J. L. Smith, Solid State Commun. **80**, 263 (1991).
[7] A. de Visser, R. J. Keizer, A. A. Menovsky, M. Mihalik, F. S. Tautz, J. J. M. Franse, B. Fåk, N. H. van Dijk, J. Flouquet, J. Bossy, and S. Pujol, Physica B **230–232**, 49 (1997).
[8] P. H. Frings, J. J. M. Franse, F. R. de Boer, and A. Menovsky, J. Magn. Magn. Mater. **31–34**, 240 (1983).
[9] G. Bruls, D. Weber, I. Kououdis, H. Weinfurter, K. Clausen, M. Yoshizawa, B. Lüthi, and E. Bucher, Physica B **186–188**, 258 (1993).
[10] G. Bruls, D. Finsterbush, I. Kouroudis, B. Wolf, D. Wichert, B. Lüthi, L. Jansen, and L. Taillefer, Physica B **223–224**, 36 (1996).
[11] G. Bruls, D. Weber, B. Wolf, P. Thalmeier, B. Lüthi, A. de Visser, and A. Menovsky, Phys. Rev. Lett. **65**, 2294 (1990).
[12] S. Adenwalla, S. W. Lin, Q. Z. Ran, Z. Zhao, J. B. Ketterson, J. A. Sauls, L. Taillefer, D. G. Hinks, M. Levy, and B. K. Sarma, Phys. Rev. Lett. **65**, 2298 (1990).
[13] N. H. van Dijk, A. de Visser, J. J. M. Franse, and L. Taillefer, J. Low Temp. Phys. **93**, 101 (1993).
[14] G. Aeppli, D. Bishop, C. Broholm, E. Bucher, K. Siemesmeyer, M. Steiner, and N. Stüsser, Phys. Rev. Lett. **63**, 676 (1989).
[15] E. D. Isaacs, P. Zschack, C. L. Broholm, C. Burns, G. Aeppli, A. P. Ramirez, T. T. M. Palstra, R. W. Erwin, N. Stücheli, and E. Bucher, Phys. Rev. Lett. **75**, 1178 (1995).
[16] S. M. Hayden, L. Taillefer, C. Vettier, and J. Flouquet, Phys. Rev. B **46**, 8675 (1992); T. Trappmann, H. v. Löhneysen, and L. Taillefer, *ibid.* **43**, 13 714 (1991).
[17] D.-C. Chen and A. Garg, Phys. Rev. Lett. **70**, 1689 (1993); A. Garg and D.-C. Chen, Phys. Rev. B **49**, 479 (1994).
[18] R. Joynt, Phys. Rev. Lett. **71**, 3015 (1993); J. Sauls, Adv. Phys. **43**, 113 (1994).
[19] K. Machida and M. Ozaki, Phys. Rev. Lett. **66**, 3293 (1991); K. Machida, T. Ohmi, and M. Ozaki, J. Phys. Soc. Jpn. **62**, 3216 (1993).
[20] B. Lussier, L. Taillefer, W. J. L. Buyers, T. E. Mason, and T. Petersen, Phys. Rev. B **54**, R6873 (1996).
[21] I. A. Fomin and J. Flouquet, Solid State Commun. **98**, 795 (1996).
[22] P. Dalmas de Réotier, A. Huxley, A. Yaouanc, J. Flouquet, P. Bonville, P. Imbert, P. Pari, P. C. M. Gubbens, and A. M. Mulders, Phys. Lett. A **205**, 239 (1995).
[23] A. de Visser, R. J. Keizer, R. van Harrevelt, A. A. Menovsky, J. J. M. Franse, A. Amato, F. N. Gygax, M. Pinkpank, and A. Schenck, Physica B **230–232**, 53 (1997).
[24] N. H. van Dijk, Ph.D. thesis, University of Amsterdam, 1994.
[25] J. Rossat-Mignod, G. H. Lander, and P. Burlet, in *Handbook on the Physics and Chemistry of Actinides*, edited by A. J. Freeman and G. J. Lander (Elsevier, Amsterdam, 1984), Vol. 1, p. 415.
[26] T. Müller, W. Joss, and L. Taillefer, Phys. Rev. B **40**, 2614 (1989).