Non-trivial magnetic order in URu_2Si_2?

1995 J. Phys.: Condens. Matter 7 5089

(http://iopscience.iop.org/0953-8984/7/26/014)

J. Phys.: Condens. Matter 7 (1995) 5089–5096. Printed in the UK

Non-trivial magnetic order in URu_2Si_2?

T E Mason†‡, W J L Buyers§‖, T Petersen†‡, A A Menovsky¶ and J D Garrett+

† Department of Physics, University of Toronto, Toronto, Ontario, Canada M5S 1A7
‡ Department of Solid State Physics, Risø National Laboratory, 4000 Roskilde, Denmark
§ Chalk River Laboratories, AECL Research, Chalk River, Ontario, Canada K0J 1J0
‖ Physics Department, University of British Columbia, Vancouver, BC, Canada V6T 1Z1
¶ Natuurkundig Laboratorium Der Universiteit van Amsterdam, NL-1018XE Amsterdam, The Netherlands
+ Institute for Materials Research, McMaster University, Hamilton, Ontario, Canada L8S 4M1

Received 17 January 1995

Abstract. We have searched for the hidden multispin order parameters that have been proposed to explain the weak ordered moment observed in the heavy-fermion superconductor URu_2Si_2. Since the exotic spin order is predicted to cause broken-symmetry Bragg peaks that develop rapidly with applied magnetic field, we have measured their strength in fields up to 8 T, for which a large easily observable moment is predicted. The Bragg neutron scattering is found to decrease rather than increase when a field is applied along the easy tetragonal c-axis, while there is no change for a field at right angles. The results rule out the proposed symmetric multispin order parameters. The zero-field transition is found to exhibit critical softening of the spin gap and a cusp in the susceptibility at T_N, as expected for the ordering of magnetic dipoles.

Among heavy-fermion materials that become superconducting at low temperatures URu_2Si_2 and UPt_3 are particularly interesting because the superconducting state coexists with an antiferromagnetically ordered state. This state has a sublattice magnetization two orders of magnitude smaller than that expected for f electrons of uranium [1, 2, 3]. UNi_2Al_3 and UPd_2Al_3 also have superconducting states coexisting with antiferromagnetically ordered moments of ~ 0.1 and $1\mu_B$ respectively [4, 5] although, in the latter case, there is evidence the superconducting and magnetic degrees of freedom are decoupled [6]. In URu_2Si_2 the development of antiferromagnetic order is accompanied by a substantial change in the magnetic dynamics from an overdamped response above T_N to a propagating spin-wave mode with anisotropic damping below T_N. The magnetic dynamics of the ordered state are well described qualitatively as a transition between two singlets [1, 2]. The ordered moment would then be induced by intersite interactions [7] and could be quite small. However, it is difficult to reconcile the large specific heat jump at $T_N = 17.5$ K [8] with the observed ordered moment of only $(0.04 \pm 0.01)\mu_B$. Furthermore, measurements of the nonlinear susceptibility, χ_3, of URu_2Si_2 have found that it is positive with an anomaly very similar to that exhibited by the specific heat [9], a result not expected for simple magnetic dipole ordering.

The difficulties of explaining the weak antiferromagnetic ordering in heavy-fermion superconductors as conventional dipolar ordering have led to suggestions that the real order parameter may be of a tensor or non-local nature [9, 10] (e.g. quadrupolar, octupolar etc) or possibly involve the more exotic multispin correlators described by Gor'kov and Sokol

0953-8984/95/265089+08$19.50

[11]. Polarized neutron measurements, together with symmetry considerations, have shown that the weak antiferromagnetic peaks observed at low fields in URu_2Si_2 can only arise from local order that is purely dipolar and do not correspond to higher-angular-momentum symmetry-breaking fields or linear combinations of those fields with a magnetic dipole [12]. The observation of broken time-reversal symmetry [12] by polarized neutron scattering rules out the possibility of quadrupolar ordering of the type discussed by Santini and Amoretti [13] if the weak Bragg peaks are an intrinsic property of URu_2Si_2. The existing measurements do not rule out the possibility that the real order parameter is of the more exotic kind proposed in [11] which would be hidden at zero magnetic field to a dipolar probe such as neutron scattering. In this case the observed dipole moment would have to be regarded as a parasitic effect brought on by local defects in response to a symmetry-breaking field not visible in a neutron scattering experiment. However, a large observable ($\sim 1\mu_B$) staggered dipole moment would appear if a large uniform magnetic field [14] of order 10 T were applied. This occurs because the applied magnetic field induces a different magnetization on each of the two sublattices.

In order to test for the presence of all the spin nematic and triple-spin correlator order parameters discussed by Barzykin and Gor'kov [14] it is necessary to study the magnetic field dependence of the staggered magnetization in three different geometries. This is due to the differences in the direction of the induced spin polarization for each state together with the experimental constraint that magnetic neutron scattering is only sensitive to the component of spin polarization perpendicular to the momentum transfer. As an example consider the case for a magnetic field applied along the (00l) direction. The anisotropy of the order parameter would be determined by the crystalline anisotropy, which, in the case of body-centred tetragonal URu_2Si_2, has an easy magnetic c-axis. The spin polarization for the n-type spin nematic (using the notation of [14] where L^i are the components of the spin polarization and n is the uniaxial direction) is proportional to:

$$L^i \sim n^i(n \cdot H) - \frac{1}{3}H^i \tag{1}$$

and with $n = \hat{c}$ and $H = H\hat{c}$ one obtains $L \sim \frac{2}{3}H\hat{c}$. The neutron scattering cross section is proportional to the square of the staggered magnetization and would therefore be expected to rise quadratically with applied field. Measurements at the (100) antiferromagnetic position would be sensitive to this c-axis-polarized staggered magnetization from this two-spin correlator. For ordering of the triple-spin correlator discussed in [14] the induced spin polarization is proportional to

$$L^i \sim a^i(b \cdot H)(c \cdot H) + b^i(a \cdot H)(c \cdot H) + c^i(a \cdot H)(b \cdot H). \tag{2}$$

In this case a magnetic field along (00l) would not induce a staggered spin polarization, while a magnetic field along $(h, \bar{h}, 0)$ would result in a c-axis-polarized staggered magnetization proportional to the square of the magnetic field. The neutron scattering cross section would therefore increase as the applied field to the fourth power. Measurements of the staggered magnetization at the (111) position would detect this signal.

We present the results of elastic neutron scattering measurements of the staggered magnetization of URu_2Si_2 in a uniform applied magnetic field for the three different field directions required to test predictions of [14]. The measurements were carried out on the DUALSPEC triple-axis spectrometer at Chalk River Laboratories. In order to remove all higher-order contamination of the neutron beam (which can give rise to a spurious peak at the antiferromagnetic position) we employed a Si111 monochromator and analyser and a neutron energy of 3.52 THz together with two pyrolytic graphite filters. Collimation before and after the sample was 0.8°. The three samples employed were the same crystals as used in previous

studies [1, 2, 15]. They were mounted in a split-coil superconducting solenoid with magnetic fields of up to 8 T along the (00l), (h00), and ($h\bar{h}$0) directions. The sample used in the study with $\boldsymbol{H}$ parallel to ($h\bar{h}$0) had a somewhat higher superconducting transition temperature and longer antiferromagnetic correlation length [15] but the same ordered moment, Néel temperature, and magnetic excitation spectrum as the others, indicating the same magnetic ground state for all three. This experimental configuration is able to detect the very weak antiferromagnetic peak present in zero magnetic field [1, 2, 15]. The magnitude of the moment expected for the exotic order parameters ($\sim 1\mu_B$) [14] exceeds the sensitivity of the experiment by more than two orders of magnitude.

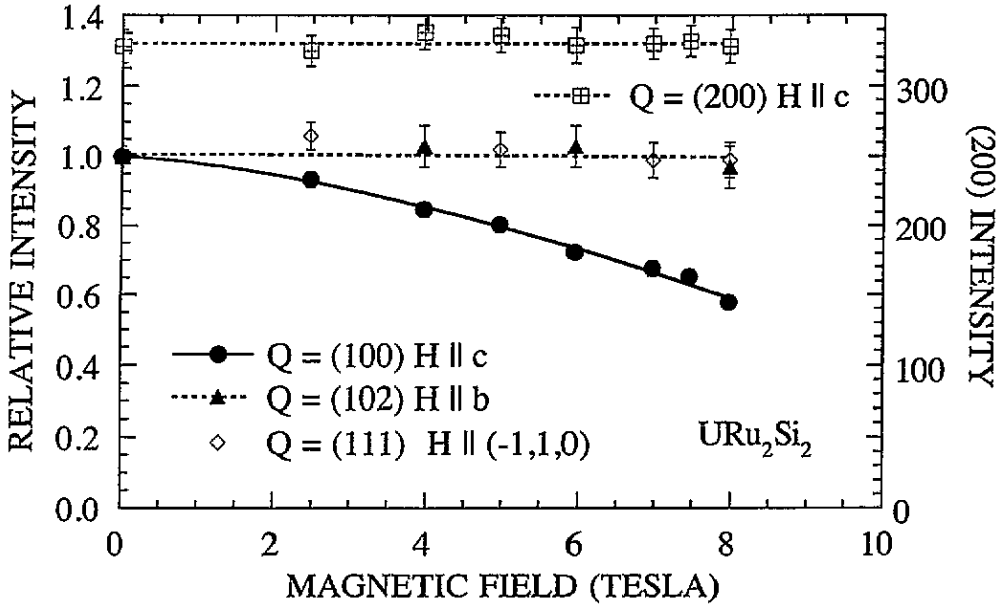

Figure 1. The magnetic field dependence of the magnetic Bragg peak intensity for fields applied parallel to the (001), (100), and ($\bar{1}$10) directions along with the structural Bragg peak intensity measured at (200). If there were an exotic order parameter of the type proposed in [11, 14] the magnetic intensity would be expected to increase with field attaining a value approximately two orders of magnitude greater than that observed in zero field.

The results of our measurements are summarized in figure 1 which shows the magnetic field dependence of the integrated intensity of the (100), (102), and (111) antiferromagnetic Bragg peaks for magnetic fields up to 8 T applied along the (00l), (h00), and ($h\bar{h}$0) directions respectively at a temperature of 4.3 K. The intensity is plotted relative to the zero-field value which corresponds to an ordered moment of $0.04\mu_B$. Also shown is the intensity of a nuclear reflection which was monitored to ensure that the sample was not moving (data are shown only for (200) with $\boldsymbol{H} \parallel \mathbf{c}$, but were also verified for the other two geometries).

All three field directions fail to show the substantial increase in the staggered magnetization predicted for symmetric exotic spin-tensor ordering. For a magnetic field applied in the tetragonal basal plane, perpendicular to the weak ordered moment and the spin excitations, there is no detectable change in the Bragg peak intensity up to 8 T. This is consistent with the large anisotropy in the magnetic susceptibility [8] which favours the easy c-axis. Application of the magnetic field along the c-axis causes a gradual suppression of

the (100) Bragg peak intensity consistent with earlier measurements at lower fields [2, 16]. The field dependence is well described by a power law in reduced field:

$$I \sim 1 - \left(\frac{H}{H_0}\right)^{\alpha} \tag{3}$$

with $\alpha = 3/2$ and a characteristic field $H_0 = 14.5(3)$ T (this is the field at which the staggered magnetization would vanish in the absence of corrections to the power law).

The field dependence of the staggered dipole moment in a magnetic field rules out all five candidate non-trivial order parameters proposed for URu_2Si_2 [14]. The prediction of multispin order was deduced only for an exchange-only scenario, in contrast to the strong crystalline anisotropy of this and other heavy-fermion systems.

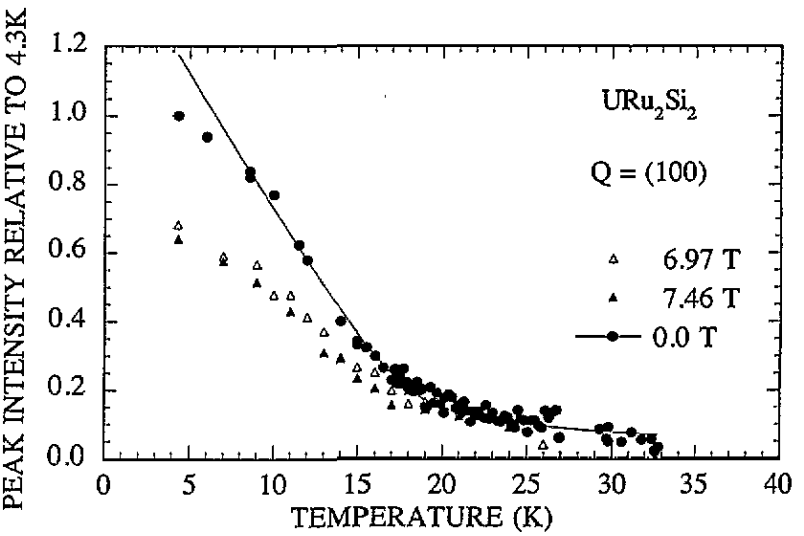

Figure 2. The temperature dependence of the (100) magnetic Bragg peak in zero field and for $H = 6.97$ T and 7.46 T magnetic fields along the tetragonal c-axis. Although the application of a magnetic field along the moment direction significantly suppresses the ordered moment, the Néel temperature is not substantially reduced. The line is the sum of a fit to the critical scattering and to the Bragg peak at zero field using mean-field critical exponents.

The field dependence is qualitatively consistent, however, with that expected for a singlet ground-state system with antiferromagnetic induced moment ordering [1, 2]. In this case the field along the c-axis acts to enhance the moment induced on the sites where the spin is parallel to the field while acting to suppress the moment on the antiparallel sites. When the ordering is via induced moments the net effect is a suppression of the staggered dipole moment. Somewhat more perplexing is the temperature dependence of the ordered moment in a field, shown in figure 2. Although a 7.5 T field reduces the Bragg peak intensity by a factor of 0.6 (corresponding to 77% of the zero-field moment) T_N is not significantly reduced. One would expect these two quantities to track one another; the fact that they do not implies that the ordered moment does not set the energy scale for the phase transition, a result also implied by the discrepancy between the size of the moment and the magnitude of the specific heat jump. A similar effect is observed in UPt_3 which shows a reduction of the ordered moment but not a significant reduction in T_N upon applying hydrostatic pressure [17]. Specific heat [18] and transport [19] also show little variation with magnetic field.

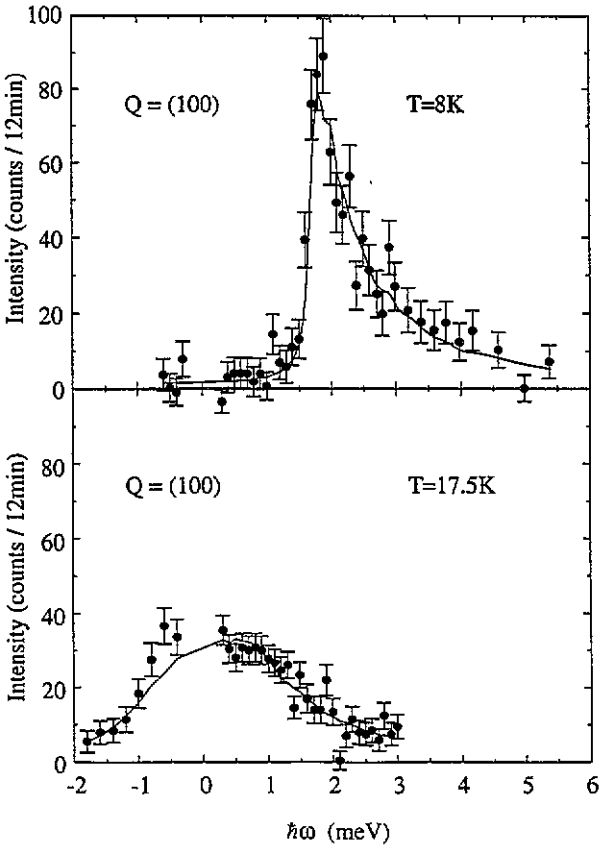

Figure 3. The spin excitation at $Q = (100)$ for two different temperatures. At low temperatures the excitation is sharply defined, but at the Néel temperature the energy has decreased and the damping increased. The lines are fits to the function described in the text.

To illuminate further the nature of the phase transition we performed a high-resolution neutron scattering study on the TAS7 spectrometer at Risø National Laboratory of the temperature dependence of the inelastic scattering at the antiferromagnetic zone centre. If the order parameter is dipolar or contains a dipolar component, as suggested by the results described above and those of [12], the real part of the generalized susceptibility at the ordering wavevector should diverge at the phase transition. To obtain optimal resolution and remove higher-order reflections we used a fixed final neutron energy of 5.0 meV with a beryllium filter after the sample. The (002) reflection from pyrolytic graphite was used to monochromate and analyse the beam. The collimation from reactor to detector was 36′–30′–60′–120′. The crystal was the same as the one mounted with the magnetic field along $(00l)$ in the previously described experiment. It was oriented with $(hk0)$ in the scattering plane. The resultant energy resolution was 0.15 meV allowing us considerable precision in determining the behaviour of the spin waves. Scans of the inelastic response at the ordering wavevector (100) taken at and well below the Néel temperature are shown in figure 3. It is clear that the excitation energy, Δ, decreases as $T \rightarrow T_N$ while the damping increases. To extract values for energy and damping of the excitations we convoluted the resolution

function of the spectrometer configuration with the harmonic oscillator lineshape

$$S(q,\omega) = (n(\omega)+1)\frac{A\omega\Delta_q\Gamma}{(\omega^2-\Delta_q^2)^2+\omega^2\Gamma^2} \tag{4}$$

where q is the wavevector relative to $Q_0 = (100)$, ω is the energy transfer, Δ_q and Γ are the energy and damping of the excitation respectively, A is the temperature-dependent intensity of the excitation and $n(\omega)$ is the Bose–Einstein population factor. We took $\Delta_q^2 = \Delta^2 + v^2q^2$ after having found from measurements at wavevectors displaced from Q_0 that v was temperature independent and isotropic. The high-energy tail in figure 3 arises from the velocity, v, through the the out-of-plane resolution.

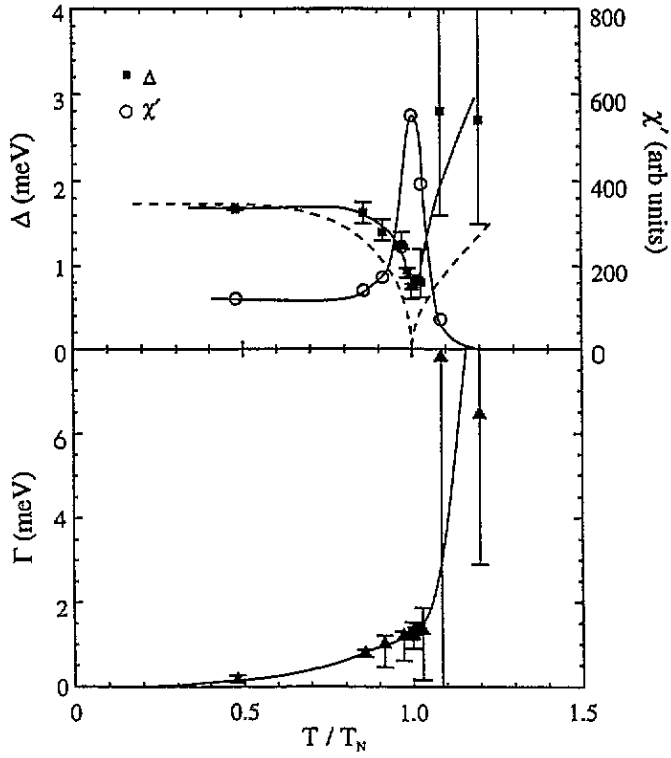

Figure 4. Temperature dependence of the spin-wave damping, Γ, and energy gap, Δ, and the susceptibility, χ', at the antiferromagnetic ordering wavevector (100) determined by the fits described in the text. The solid lines are guides to the eye, and the broken line is the gap calculated for the singlet ground-state model.

The temperature dependences of the gap, $\Delta = \Delta_0$, damping, Γ, and staggered susceptibility, $\chi' = \chi'(q=0,\omega=0) = A/\Delta$, are shown in figure 4. Qualitatively the results agree with what is expected for a singlet ground-state system with antiferromagnetic ordering [1, 2, 20]; the spin-wave frequency softens as the transition is approached and the damping increases. We show our calculations of the mean-field temperature dependence of the gap in figure 4 by the broken line; near T_N the data exhibit a similar rapid rise with reduced temperature, suggesting that the gap is associated with the primary order parameter. Although the spin-wave energy extracted from the fits does not actually vanish at T_N the evidence suggests that the real gap likely does collapse. We know that the fraction of the resolution volume sampling the lowest energies becomes a vanishingly small fraction of the

resolution volume as $T \to T_N$ since there $\Delta_q \to vq$ with phase space vanishing as q^2; thus no real experiment contains information from the truly zero-energy modes right at T_N. Also the model lineshape depends only on the combination Δ^2/Γ for $\Delta \ll \Gamma$, so the gap energy becomes difficult to refine near T_N. Thus our results are consistent with a true vanishing of the gap at the phase transition. The amplitude, A, remains robust near T_N. The strongest evidence for critical behaviour in the dipolar channel is the staggered susceptibility, χ'. It shows a sharp peak at T_N consistent with the cusp expected at a critical transition after some rounding by residual resolution effects.

The cusp in the magnetic susceptibility at the ordering wavevector as T approaches T_N is evidence that the transition involves ordering of the magnetic dipoles. The critical behaviour of the susceptibility and the gap are consistent with a magnetic transition. If the magnetic dipole were only a secondary or parasitic order parameter varying as a power of a hidden order parameter we would expect a smooth non-critical behaviour near T_N [21] rather than the critical gap and susceptibility. Since the matrix element associated with this excitations is large ($\sim 1\mu_B$) [2], the observation of a peak in the susceptibility cannot be associated with parasitic dipolar ordering in a small fraction of the sample as required to explain the antiferromagnetic Bragg peaks if the order parameter were quadrupolar [13] or tensor [14].

In conclusion, we have measured the magnetic field dependence of the sublattice dipole magnetization of URu_2Si_2 up to 8 T in the three geometries required to test for the symmetric tensor order parameters proposed by Barzykin and Gor'kov [14]. We do not find the substantial increase with field in the staggered moment expected for these non-trivial magnetic states. The measurements are not sensitive to double- and triple-spin correlator states for which the tensor is not totally symmetric (such as the p-nematic or chiral spin state) but those states would not explain the observed zero-field peak or its field dependence. The suppression of the ordered moment with application of a magnetic field along the c-axis with a characteristic field of 14.5 T is qualitatively consistent with the expectations for a singlet ground-state induced moment system, as is the cusp in the staggered susceptibility at T_N. This confirms that the order parameter contains a dipolar component, in agreement with polarized neutron measurements and symmetry considerations [12]. Since the magnetic dipole does indeed seem to be the order parameter for the antiferromagnetic transition, the question of how the large energy scale for the phase transition and the specific heat is compatible with the extremely weak ordered moment remains a mystery.

Acknowledgments

We thank M Gauthier for technical assistance, L P Gor'kov for helpful discussions, and CIAR, NSERC, and AECL for provision of neutron scattering facilities and support.

References

[1] Broholm C, Kjems J K, Buyers W J L, Matthews P, Palstra T T M, Menovsky A A and Mydosh J A 1987 *Phys. Rev. Lett.* **58** 1467

[2] Broholm C, Lin H, Matthews P T, Mason T E, Buyers W J L, Collins M F, Menovsky A A, Mydosh J A and Kjems J K 1991 *Phys. Rev.* B **43** 12 809

[3] Aeppli G, Bucher E, Broholm C, Kjems J K, Baumann J and Hufnagl J 1988 *Phys. Rev. Lett.* **60** 615

[4] Geibel C, Thies S, Kaczorowski D, Mehner A, Grauel A, Seidel B, Ahlheim U, Helfrich R, Petersen K, Bredl C D and Steglich F 1991 *Z. Phys.* B **83** 305

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

[6] Caspary R, Hellmann P, Keller M, Sparn G, Wassilew C, Köhler R, Geibel C, Schank C, Steglich F and Phillips N E 1993 *Phys. Rev. Lett.* **71** 2146
[7] Wang Y-L and Cooper B R 1969 *Phys. Rev.* **185** 696
[8] Palstra T T M, Menovsky A A, van den Berg J, Dirkmaat A J, Kes P H, Nieuwenhuys G J and Mydosh J A 1985 *Phys. Rev. Lett.* **55** 2727
[9] Ramirez A P, Coleman P, Chandra P, Brück E, Menovsky A A, Fisk Z and Bucher E 1992 *Phys. Rev. Lett.* **68** 2680
[10] Chandra P, Ramirez A P, Coleman P, Brück E, Menovsky A A, Fisk Z and Bucher E 1993 *Preprint*
[11] Gor'kov L P and Sokol A 1992 *Phys. Rev. Lett.* **69** 2586
[12] Walker M B, Buyers W J L, Tun Z, Que W, Menovsky A A and Garrett J D 1993 *Phys. Rev. Lett.* **71** 2630
[13] Santini P and Amoretti G 1994 *Phys. Rev. Lett.* **73** 1027
[14] Barzykin V and Gor'kov L P 1993 *Phys. Rev. Lett.* **70** 2479
[15] Mason T E, Gaulin B D, Garrett J D, Tun Z, Buyers W J L and Isaacs E D 1990 *Phys. Rev. Lett.* **65** 3189
[16] Mason T E, Lin H, Collins M F, Buyers W J L, Menovsky A A and Mydosh J A 1990 *Physica* B **163** 45
[17] Hayden S M, Taillefer L, Vettier C and Flouquet J 1992 *Phys. Rev.* B **46** 8675
[18] de Visser A, Kayzel F E, Menovsky A A, Franse J J M, van den Berg J and Nieuwenhuys G J 1986 *Phys. Rev.* B **34** 8168
[19] Sugiyama K, Fuke H, Kindo K, Shimohata K, Menovsky A A, Mydosh J A and Date M 1990 *J. Phys. Soc. Japan* **59** 3311
[20] Als-Nielsen J, Kjems J K, Buyers W J L and Birgeneau R J 1977 *J. Phys. C: Solid State Phys.* **10** 2673
[21] Collins M F 1989 *Magnetic Critical Scattering* (Oxford: Oxford University Press)