Quadrupole and dipole orders in URu_2Si_2

1999 J. Phys.: Condens. Matter 11 L519

(http://iopscience.iop.org/0953-8984/11/46/101)

J. Phys.: Condens. Matter **11** (1999) L519–L524. Printed in the UK PII: S0953-8984(99)07964-3

LETTER TO THE EDITOR

Quadrupole and dipole orders in URu_2Si_2

Fusayoshi J Ohkawa and Hirofumi Shimizu

Division of Physics, Graduate School of Science, Hokkaido University, Sapporo 060-0810, Japan

E-mail: fohkawa@phys.sci.hokudai.ac.jp

Received 20 September 1999

Abstract. Exotic magnetism below $T_N \simeq 17.5$ K is studied within the level scheme where the lowest multiplet is a doublet within the $5f^2$ configuration. Effective g-factors of pseudo-spins with $S = \frac{1}{2}$, which describe the degree of freedom of the doublet, are highly anisotropic: $g_x = g_y = 0$ for the xy-components and $g_z \neq 0$ for the z-component. It is proposed that a recently discovered transition of first order at a critical pressure $p_c \simeq 1.5$ GPa is that between an ordered state of quadrupoles, with order parameter $O_{x^2-y^2}$ or O_{xy}, below p_c and an ordered state of dipoles, with order parameter O_z, above p_c; pseudo-spins are ordered within the xy-plane below p_c, and they are along the z-axis above p_c. The proposal of this scenario is followed by many predictions. No static magnetic moments exist below p_c. The anisotropy of Van Vleck's susceptibility within the xy-plane is of twofold symmetry corresponding to $O_{x^2-y^2}$ or O_{xy}. What one observes by means of neutron diffraction and μSR (muon spin resonance) below p_c are dynamically but slowly fluctuating magnetic moments. The softening of magnons occurs with pressures approaching p_c below p_c. Although static magnetic moments exist above p_c, no magnon excitations can be observed there.

It is of interest to know what order parameter is responsible for the exotic magnetism below $T_N \simeq 17.5$ K in URu_2Si_2. According to various results such as specific heat [1], linear [1, 2] and non-linear [3,4] susceptibility, thermal expansion [5], and resistivity [6] measurements, the phase transition at T_N is a typical second-order one. The large specific heat anomaly [1] means that a large entropy is involved in this transition. On the other hand, neutron diffraction [7] suggests that tiny-moment magnetism occurs below T_N. The ordering vector is

$$\boldsymbol{Q} = (0, 0, \pm\pi/c) \tag{1}$$

with c the lattice constant along the c- or z-axis, and the saturated magnetic moments are as small as 0.02–0.04 μ_B per U ion. According to μSR measurements [8], however, they are about 0.003 μ_B per U ion. Furthermore, no internal magnetic fields are observed by means of NMR [9]. One of the most plausible explanations for this inconsistency is that the characteristic timescales or energy scales differ among the three measurements; the energy scale of the NMR is almost static for the relevant spin fluctuations, but that of the neutron diffraction is small but still dynamical; that of the μSR is intermediate. Thus, the absence of internal magnetic fields according to NMR implies that the observed tiny magnetic moments are never static but are fluctuating.

Recently, a first-order transition has been observed as a function of pressure p [10]. Magnetic moments observed by means of neutron diffraction increase with increasing p. However, their temperature dependence is qualitatively different from those for conventional magnets. Magnetic moments show a sharp increase at $p_c \simeq 1.5$ GPa. Above p_c, the temperature dependence of the magnetic moments is conventional. One speculation is that the order parameter is not that of conventional magnetic moments below p_c while it is that of conventional magnetic moments above p_c.

The temperature dependence of static homogeneous susceptibility is unusual [1, 2]. At high temperatures such as $T \gtrsim$ 50–60 K, it increases as temperature decreases. Such a temperature dependence is characteristic of Kondo lattices. However, it shows a rapid decrease below 50–60 K. This rapid decrease suggests that the lowest multiplet is a singlet. Within the level scheme where the lowest multiplet is a singlet, possible order parameters were proposed [11–14]. However, it is difficult to explain the recently discovered first-order transition within this level scheme.

The so-called non-Fermi-liquid behaviour is observed in $U_xTh_{1-x}Ru_2Si_2$ with $x \lesssim 0.1$ [15–17]; various low-temperature properties imply that the lowest multiplet is a doublet. There is no evidence that the level scheme is changed with increasing x. The formation of heavy quasiparticles at low temperatures in URu_2Si_2 also implies that the lowest multiplet is a doublet. Superconductivity occurs below $T_c \simeq 1.5$ K [1, 2, 18]. Although Cooper pairs seem to be anisotropic, they are never exotic. In view of the large specific heat anomaly at T_c, it is certain that heavy quasiparticles are condensed into a superconducting state. Because heavy quasiparticles must be mainly composed of 5f electrons, it is likely that the lowest multiplet is a doublet whose degeneracy is due to time-reversal symmetry. Within this level scheme, two scenarios were proposed, three-spin order [19] and bond order in spin channels [20]. The main purpose of this letter is to propose another scenario, which can explain not only many experimental results but also the recently discovered first-order transition [10].

Because the anisotropy of the g-factors is large, it is likely that the ground configuration is $5f^2$ rather than $5f^1$ or $5f^3$. The multiplet of $5f^2$, which has the total angular moment of $J = 4$, splits into five singlets and two doublets in body-centred tetragonal lattices. The doublets are linear combinations of $|J_z = \pm 3\rangle$ and $|J_z = \pm 1\rangle$. In this letter, it is assumed that one of the doublets which is a pair consisting of

$$|\uparrow\rangle = \cos\alpha |J_z = 3\rangle + \sin\alpha |J_z = -1\rangle \tag{2}$$

and

$$|\downarrow\rangle = \cos\alpha |J_z = -3\rangle + \sin\alpha |J_z = 1\rangle \tag{3}$$

is the lowest multiplet. The spin operators with $S = \frac{1}{2}$ can describe the doublet. The Zeeman term is given by

$$\mathcal{H}_Z = -\frac{1}{2}\sum_i \sum_{\nu=x,y,z} g_\nu \mu_B H_\nu S_{i\nu} \tag{4}$$

with $S_{i\nu}$ one of the spin operators ($\nu = x$, y, and z) and the H_ν magnetic fields; the g-factors are anisotropic, so

$$g_z = g_J(3\cos^2\alpha - \sin^2\alpha) \tag{5}$$

for the z-axis component, with $g_J = 4/5$ being Lande's g-factor, and

$$g_x = g_y = 0 \tag{6}$$

for the xy-plane components. The anisotropy of the g-factors is large, unless

$$|3\cos^2\alpha - \sin^2\alpha| \ll 1.$$

This large anisotropy is closely related to a physical property whereby within the doublet, only dipole transitions of $\Delta J_z = 0$ are possible and those of $\Delta J_z = \pm 1$ are impossible. In general, the interaction between multipoles is given by

$$\mathcal{H}_{\text{multi}} = -\frac{1}{2}\sum_{\Gamma ij} A_{\Gamma,ij} O_{\Gamma,i} O_{\Gamma,j} \tag{7}$$

with the $O_{\Gamma,i}$ being multipole operators. This is equivalent to

$$\mathcal{H}_{\rm ex} = -\frac{1}{2}\sum_{\nu=x,y,z}\sum_{ij} I_{\nu,ij} S_{i\nu} S_{j\nu} \tag{8}$$

within the space of the ground doublet. Note that $I_{z,ij}$ comes from the dipole interaction between O_zs, and that $I_{x,ij}$ and $I_{y,ij}$ come from the quadrupole interactions between $O_{x^2-y^2}$s and O_{xy}s.

When the anisotropy of $\mathcal{H}_{\rm ex}$ is that of an XY-like model, many experimental data below $p_{\rm c}$ can be qualitatively explained by this localized spin model. In this case, pseudo-spins are ordered within the xy-plane such that

$$\langle O_{z,i}\rangle \propto \langle S_{iz}\rangle = 0 \tag{9}$$

and

$$\sqrt{\langle O_{x^2-y^2,i}\rangle^2 + \langle O_{xy,i}\rangle^2} \propto \sqrt{\langle S_{ix}\rangle^2 + \langle S_{iy}\rangle^2} \neq 0. \tag{10}$$

Because $g_x = g_y = 0$, no static magnetic moments can appear. On the other hand, the specific heat anomalies can be as large as those in conventional magnetic phase transitions. Spin waves or magnons must have a gap because of the anisotropy of $\mathcal{H}_{\rm ex}$. These findings are consistent with observations [1, 7, 9]. Magnons can be excited or absorbed only by a $\Delta J_z = 0$ process.

In the presence of the order parameter $\langle O_{x^2-y^2}\rangle \propto \langle S_x\rangle$ or $\langle O_{xy}\rangle \propto \langle S_y\rangle$, Van Vleck's susceptibility is anisotropic. Assume that the ordering vector $\boldsymbol{Q}$ is exactly given by equation (1); there are two sublattices below $T_{\rm N}$. In the presence of magnetic fields within the xy-plane, $\boldsymbol{H} = H(\cos\theta, \sin\theta, 0)$, magnetic moments, $\boldsymbol{m}_\parallel$, parallel to $\boldsymbol{H}$ obey the following θ-dependence: when the order parameter is $\langle O_{x^2-y^2}\rangle$, it follows that

$$m_\parallel = [\pm A_1\cos(2\theta) + B_1]\mu_{\rm B}^2 H. \tag{11}$$

When the order parameter is $\langle O_{xy}\rangle$, on the other hand, it follows that

$$m_\parallel = [\pm A_2\sin(2\theta) + B_2]\mu_{\rm B}^2 H. \tag{12}$$

Here, one of $\pm$ is for one of the sublattices, and the other is for the other sublattice; A_1, B_1, A_2, and B_2 are constants. The total magnetizations are slightly different from those given by equation (11) or (12), because the perpendicular component is generally non-zero. Even if the perpendicular component is considered, however, the twofold symmetry, which corresponds to $O_{x^2-y^2}$ or O_{xy}, is not changed. The anisotropy of Van Vleck's susceptibility can show which quadrupole order parameter appears below $T_{\rm N}$, $\langle O_{x^2-y^2}\rangle$ or $\langle O_{xy}\rangle$.

Because the observed gap of the magnons is rather small [7], $\mathcal{H}_{\rm ex}$ must be almost isotropic. It is reasonable that the multipole interaction depends on pressure. Thus, one of the most likely scenarios for the first-order transition at $p_{\rm c}$ is that in which a pseudo-spin flop occurs; the order parameter above $p_{\rm c}$ is a dipole one, $\langle O_z\rangle \propto \langle S_z\rangle$. Static magnetic moments must appear above $p_{\rm c}$. Because the anisotropy of $\mathcal{H}_{\rm ex}$ becomes smaller with pressures approaching $p_{\rm c}$, magnons with wave vectors close to $\boldsymbol{Q}$ must become soft below $p_{\rm c}$. The pseudo-spins are along the z-axis above $p_{\rm c}$, and the matrix elements of the dipole transitions between $|\uparrow\rangle$ and $|\downarrow\rangle$ are zero. Therefore, no excitations of magnons can be observed above $p_{\rm c}$; Bragg scatterings by magnetic moments, which are statically ordered, can be observed from the $\Delta J_z = 0$ process. The softening of magnons below p_c and the vanishing of magnons above $p_{\rm c}$ have been confirmed by a recent neutron diffraction measurement [21].

Although many observations can be qualitatively explained within the theoretical framework of local moment magnetism, there are still several issues to be discussed. One of the most crucial issues among them is what one observes by means of neutron diffraction and μSR as fluctuating magnetic moments. The Kondo temperature of URu_2Si_2 is about $T_K \simeq$ 50–100 K, and $T_N \simeq 17.5$ K is much lower than T_K. Therefore, magnetism in URu_2Si_2 should be treated within the theoretical framework of itinerant-electron magnetism. Because the degree of freedom of the doublets can be described by pseudo-spins with $S = \frac{1}{2}$, as discussed before, an extended periodic s–d model, which includes magnetic exchange interactions such as the superexchange interaction that are not involved in the simplest version of the periodic s–d model, is expected to be a relevant effective Hamiltonian for URu_2Si_2. The periodic s–d model is mapped to an extended periodic Anderson model (PAM) with strong on-site repulsion. When the PAM is treated, it is straightforward to show that the longitudinal susceptibility can be written in such a way that

$$\chi_z^{\rm obs}(\omega + {\rm i}0, \boldsymbol{q}) = \frac{1}{4} g_z^2 \mu_{\rm B}^2 \frac{\chi_z(0, \boldsymbol{Q})\kappa_z^2}{\kappa_z^2 + q^2 - {\rm i}\omega(1/[\Gamma_z|\boldsymbol{q} - \boldsymbol{Q}|] + 1/\gamma_z)} \qquad (13)$$

for $\boldsymbol{q} \simeq \boldsymbol{Q}$ below T_N or in the presence of the quadrupole order, $\langle O_{x^2-y^2}\rangle$ or $\langle O_{xy}\rangle$, and in such a way that

$$\chi_z^{\rm obs}(\omega + {\rm i}0, \boldsymbol{q}) = \frac{1}{4} g_z^2 \mu_{\rm B}^2 \frac{\chi_z(0, \boldsymbol{Q})\kappa_z^2}{\kappa_z^2 + q^2 - {\rm i}\omega/\gamma_z} \qquad (14)$$

for $\boldsymbol{q} \simeq \boldsymbol{Q}$ above T_N or in the absence of the quadrupole order. Here, the constants, $\chi_z(0, \boldsymbol{Q})$, κ_z, γ_z, and Γ_z, depend on the dispersion relation of the quasiparticles, the intersite magnetic exchange interactions, and the magnitude of the order parameter. For example, κ_z is small when the Fermi surface shows sharp nesting with wave vector $\boldsymbol{Q}$. The transverse susceptibilities vanish because $g_x = g_y = 0$.

It is well known that the ${\rm i}\omega/|\boldsymbol{q}|$ term appears in polarization functions of itinerant-electron systems for small $\boldsymbol{q}$s in the central part of the Brillouin zone. The ${\rm i}\omega/|\boldsymbol{q} - \boldsymbol{Q}|$ term in equation (13) is similar to the ${\rm i}\omega/|\boldsymbol{q}|$ term. When the quadrupole order with wave vector $\boldsymbol{Q}$ appears, the Brillouin zone is reduced; the susceptibility for $\boldsymbol{q} \simeq \boldsymbol{Q}$ in the central part of the reduced Brillouin zone has the similar term. As is shown in figure 1, ${\rm Im}\, \chi_z^{\rm obs}(\omega + {\rm i}0, \boldsymbol{Q} + \boldsymbol{q})$ has a sharp peak around $\omega \simeq 0$ and $|\boldsymbol{q}| \simeq 0$. It is likely that low-energy spectra of ${\rm Im}\, \chi_z^{\rm obs}(\omega + {\rm i}0, \boldsymbol{Q} + \boldsymbol{q})$ are observed as fluctuating magnetic moments. It is interesting to examine whether the $|\boldsymbol{q}|$-dependences of the widths of the neutron diffraction spectra are consistent with equation (13) below T_N and equation (14) above T_N.

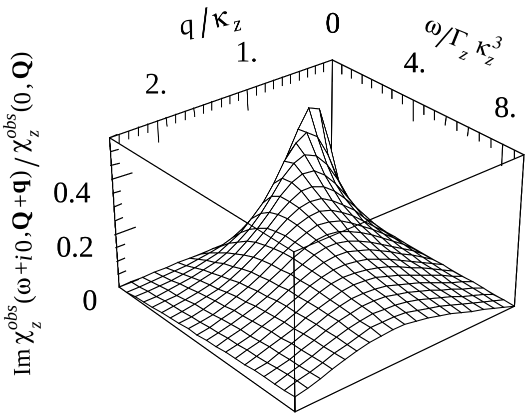

Figure 1. ${\rm Im}\, \chi_z^{\rm obs}(\omega + {\rm i}0, \boldsymbol{Q} + \boldsymbol{q})$ as a function of $|\boldsymbol{q}|$ and ω. Here, $1/\gamma_z = 0$ is assumed.

Denote the dispersion relation of heavy quasiparticles in the presence of the order parameter by $\xi_n(\boldsymbol{k})$. The main contribution to the $\mathrm{i}\omega/|\boldsymbol{q}-\boldsymbol{Q}|$ term in equation (13) comes from the summation over $\boldsymbol{k}$s satisfying

$$|\xi_n(\boldsymbol{k}) - \xi_n(\boldsymbol{k}+\boldsymbol{Q})| \lesssim |\Delta|$$

with $|\Delta|$ being a gap due to the quadrupole order. Then, Γ_z in equation (13) becomes infinitely large as $|\Delta| \to 0$, and it becomes smaller as $|\Delta|$ becomes larger. Therefore, the observed fluctuating magnetic moments must increase with decreasing temperature. When pressures are applied, we assume that the dipole interaction or the z-component of the exchange interaction becomes large and the system is approaching an instability point of O_z. Then, κ_z decreases with increasing pressure. The observed fluctuating magnetic moments must increase with increasing pressure.

In this letter, it is assumed that the lowest multiplet is a doublet within the $5f^2$ configuration. The degree of freedom of the doublet can be described by pseudo-spins with $S = \frac{1}{2}$. The anisotropy of the g-factors is that of the Ising model, so $g_x = g_y = 0$ and $g_z \neq 0$. Many experimental results can be qualitatively explained if the anisotropy of the intersite magnetic exchange interactions is that of an XY-like model below $p_c \simeq 1.5$ GPa while it is that of an Ising-like model above p_c; the order parameter below p_c is a quadrupole one, $O_{x^2-y^2}$ or O_{xy}, while the order parameter above p_c is a dipole one, O_z. No static magnetic moments exist below p_c; magnetic moments observed by means of neutron diffraction and μSR must be dynamically but slowly fluctuating moments. The existence of such low-energy spin fluctuations in the central part of the reduced Brillouin zone is evidence that the magnetism in URu_2Si_2 is to be classified as itinerant-electron magnetism. It is desirable for experiments to be explained quantitatively within the framework of itinerant-electron magnetism.

The authors are grateful to Toshiro Sakakibara and Hiroshi Amitsuka for useful discussions and comments.

References

[1] Palstra T T M, Menovsky A A, van der Berg J, Dirkmaat A J, Kes P H, Nieuwenhuys G J and Mydosh J A 1985 *Phys. Rev. Lett.* **55** 2727
[2] Maple M B, Chen J W, Dalichaouch Y, Kohara T, Rossel C, Torikachvili M S, McElfresh M W and Thompson J D 1986 *Phys. Rev. Lett.* **56** 185
[3] Miyako Y, Kawarazaki S, Amitsuka H, Paulsen C C and Hasselbach K 1991 *J. Appl. Phys.* **76** 5791
[4] Ramirez A P, Coleman P, Chandra P, Brück B, Menovsky A A, Fisk Z and Bucher E 1992 *Phys. Rev. Lett.* **68** 2680
[5] 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
[6] Palstra T T M, Menovsky A A and Mydosh J A 1986 *Phys. Rev.* B **33** 6527
[7] 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
[8] MacLaughlin D E, Cooke D W, Heffner R H, Hutson R L, McElfresh M W, Schillaci M E, Rempp H D, Smith J L, Willis J O, Zirngiebl E, Boekema C, Lichti R L and Oostens J 1988 *Phys. Rev. Lett.* **37** 3153
[9] Kohori Y, Matsuda K and Kohara T 1999 *Physics of Strongly Correlated Electron Systems (Japanese Journal of Applied Physics Series vol 11)* p 65
[10] Amitsuka H, Sato M, Metoki N, Yokoyama M, Kuwahara K, Kawarazaki S, Sakakibara T, Miyako Y and Mydosh J A 1999 *Phys. Rev. Lett.* at press
[11] Santini P and Amoretti G 1994 *Phys. Rev. Lett.* **73** 1027
[12] Sikkema A E, Buyers W J L, Affleck I and Gan J 1996 *Phys. Rev.* B **54** 9322
[13] Okuno Y and Miyake K 1998 *J. Phys. Soc. Japan* **67** 2469
[14] Kasuya T 1997 *J. Phys. Soc. Japan* **66** 3348
[15] Amitsuka H, Hidano T, Honma T, Mitamura H and Sakakibara T 1994 *Physica* B **186–188** 337

[16] Amitsuka H and Sakakibara T 1994 *J. Phys. Soc. Japan* **63** 736

[17] Amitsuka H, Sakakibara T, de Visser A, Kayzel F E and Franse J J M 1997 *Physica* B **230–232** 613

[18] Schlabitz W, Baumann J, Pollit B, Rauchacwalbe U, Mayer H M, Ahlheim U and Bredl C D 1986 *Z. Phys.* B **62** 171

[19] Barzykin V and Gor'kov L P 1993 *Phys. Rev. Lett.* **70** 2479

[20] Ikeda H and Ohashi Y 1998 *Phys. Rev. Lett.* **81** 3723

[21] Amitsuka H 1999 *Int. Conf. on Strongly Correlated Electron Systems (24–28 August 1999, Nagano, Japan); Physica* B at press