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Magnetism of superconducting UPt_3

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Abstract. The phase diagram of superconducting UPt_3 in the pressure–temperature plane, together with the neutron scattering data is studied within a two-component superconducting order parameter scenario. In order to give a qualitative explanation to the experimental data a set of two linearly independent antiferromagnetic moments which emerge appropriately at the temperatures $T_N \sim 10T_c$ and $T_m \sim T_c$ and couple to superconductivity is proposed. Several constraints on the fourth-order coefficients in the Ginzburg–Landau free energy are obtained.

1. Introduction

A heavy-fermion superconducting UPt_3 compound is an example of unconventional superconductivity, in which both the gauge and the point group symmetries are broken in the ordered phase. At the temperature $T_N \simeq 5$ K it undergoes an antiferromagnetic transition with the magnetic moments confined to the D_{6h} basal plane; however, the long-range antiferromagnetic correlations have not yet been seen [1, 2]. Far below the Néel temperature, at $T_{c_+} \simeq 0.51$ K ($p = 0$ bar) UPt_3 becomes superconducting [3, 4]. There is another superconducting transition at $T_{c_-} \simeq 0.46$ K ($p = 0$ bar) [3, 4]. This feature and a rich phase diagram in the magnetic field and temperature plane [5] are accepted as evidence of a multicomponent superconducting order parameter. There are also pressure experiments which strongly indicate the coupling between superconductivity and magnetism in UPt_3 [2, 6, 7], namely specific heat measurements under pressure. These show that the two critical temperatures T_{c_+} and T_{c_-} converge into one critical temperature T_c above $p_c \simeq 4$ kbar; see figure 1 [2, 6, 7], which is the pressure that destroys antiferromagnetism in the system. This experiment supports the theory of a two-component order parameter $\psi = (\psi_x, \psi_y)$ in a basal plane of the crystal, belonging to a two-dimensional irreducible representation of the hexagonal point group D_{6h}. In this approach a complex vector ψ couples to the magnetic moment M and the split transition is due to this interaction. The role of magnetism as a symmetry-breaking field coupling to superconductivity is revealed in neutron scattering measurements [1, 9]. In these experiments Aeppli *et al* established that below temperatures of the order of a superconducting transition temperature the neutron scattering intensity of the $(1, 1/2, 0)$ reflection suddenly saturates and is almost constant unless superconductivity occurs. There is a remarkable change in the temperature dependence for a superconducting system. At a temperature of the order of T_c the slope of the neutron scattering intensity changes sign and the intensity becomes an increasing function of temperature; see figure 2 [1, 9]. This is further strong evidence of the coupling between magnetism and superconductivity in UPt_3.

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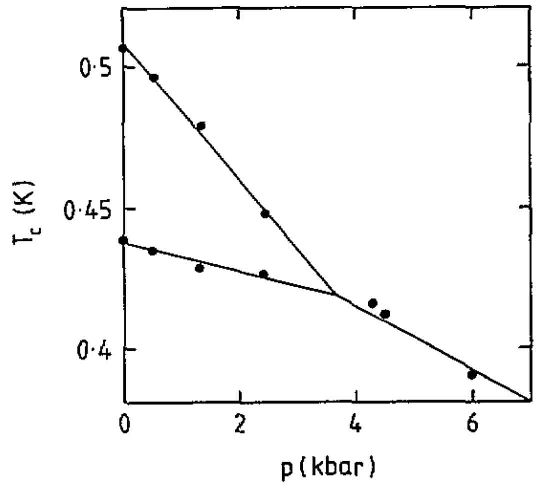

Figure 1. Pressure dependence of the superconducting phase transition temperature [7].

Recently Joynt [10] discussed within a two-component order parameter approach, the phase diagram of UPt_3 in three-dimensional magnetic-field–pressure–temperature space. It agrees qualitatively with measurements [2, 5, 6, 7, 11]. However, the temperature dependence of the magnetic moment observed by Aeppli *et al* [1, 9] was not taken into account. The contradiction here arises as follows. The magnetic Bragg peak observed in neutron scattering [1, 9] which is reproduced in figure 2 shows that the superconductivity is *acting to suppress the magnetism. By thermodynamic reasoning we know that if the onset* of superconductivity reduces the magnetism, then the onset of magnetism must reduce the tendency to superconductivity. The magnetism may be removed by pressure [2, 6, 7]. We observe that as the pressure is reduced below p_c where the magnetism reappears the slope of the transition temperature is increased; see figure 1. In other words the critical temperatures T_{c_+} and T_{c_-} are not suppressed equally by the pressure, which can be expressed quantitatively by an inequality as follows:

$$\frac{T_{c_+}(p=0) - T_c(p=0)}{T_c(p=0) - T_{c_-}(p=0)} > 1. \tag{1}$$

We show that this competition between superconductivity and antiferromagnetism cannot be understood within the simple model of magnetism considered so far.

The plan of the paper is as follows. In section 2 we study the previously mentioned pressure and magnetic field experiments in the frame of a two-dimensional superconducting order parameter scenario. In order to avoid the inconsistencies which follow from this approach we introduce a two-magnetic-moment model in section 3. Within this scenario we analyse the experimental data and obtain several constraints on the Ginzburg–Landau free energy coefficients in sections 3 and 4. Finally we summarize the results in section 5.

2. Two-component superconductivity coupled to magnetism

In this section we review the experimental evidence which supports this model and then construct the free energy. The free energy is used to obtain the coupled order parameters of magnetism and superconductivity. This analysis follows [8, 14, 15]. We reproduce it here because it is important to consider both the temperature and pressure experiments using a

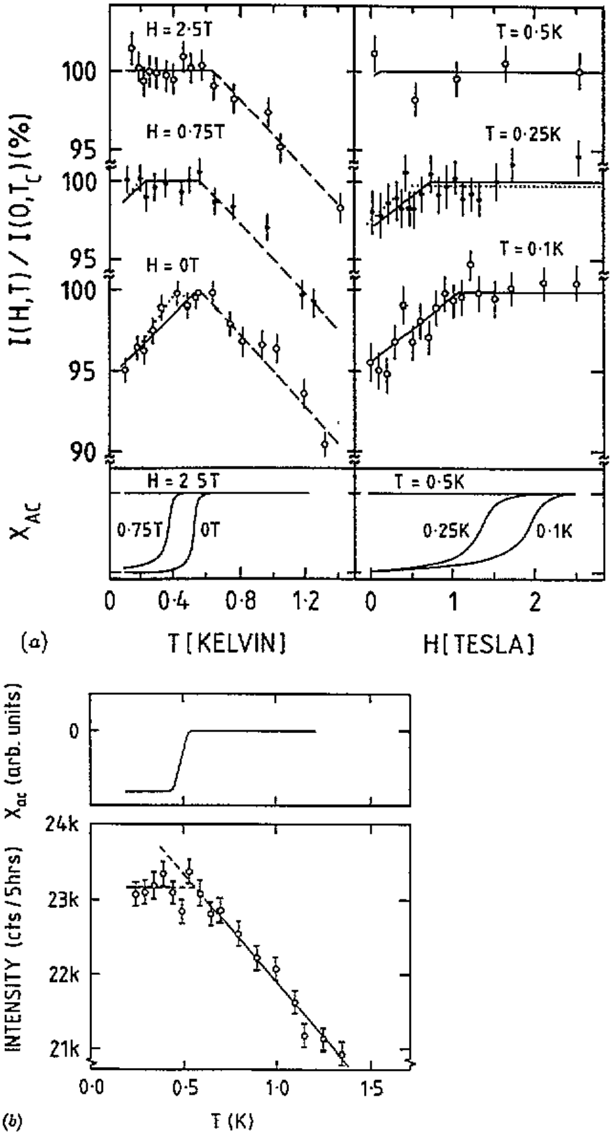

Figure 2. Field and temperature dependence of $(1, \frac{1}{2}, 0)$ [9] (*a*) and $(\frac{1}{2}, 0, 1)$ [16] (*b*) neutron scattering intensities.

unified notation. In this approach we start with a free energy density:

$$F = F_M + F_S + F_{SM} \tag{2}$$

where

$$F_M = \begin{cases} \alpha_M(T^* - T_N)M^2 + \frac{1}{2}\beta_M M^4 & \text{for } T \leqslant T^* \\ \alpha_M(T - T_N)M^2 + \frac{1}{2}\beta_M M^4 & \text{for } T > T^* \end{cases} \tag{3}$$

$$F_S = \alpha_S(T - T_c)|\psi|^2 + \frac{1}{2}\beta_1|\psi|^4 + \frac{1}{2}\beta_2|\psi^2|^2 \tag{4}$$

$$F_{SM} = \gamma|M \cdot \psi|^2 + \alpha M^2|\psi|^2. \tag{5}$$

All the Ginzburg–Landau coefficients are very weakly temperature and pressure dependent, as can be shown in a weak-coupling microscopic theory [12]; hence we choose them to be constant. The magnetic free energy given by equation (3) has been chosen to include the phenomenological saturation of M below T^* [1, 9]. The coefficients in F_M (3) and F_S (4) are positive whereas the γ coefficient in F_{SM} (5) may be chosen to be negative so ψ is the most favourable superconducting order parameter when M is chosen to be parallel to $\hat{x}$. The superconducting order parameter $\psi = (\psi_x, \psi_y)$ is complex and its components ψ_x and ψ_y are written as $\psi_x = |\psi_x|e^{i\varphi_x}$ and $\psi_y = |\psi_y|e^{i\varphi_y}$.

Minimization of the free energy leads to the following equations for the order parameters:

$$0 = \alpha_M(T_M - T_N) + \beta_M M^2 + \gamma|\psi_x|^2 + \alpha|\psi|^2 \qquad \text{or} \qquad M = 0 \tag{6}$$

where

$$T_M = \begin{cases} T & \text{for } T > T^* \\ T^* & \text{for } T \leqslant T^* \end{cases}$$

$$0 = \alpha_S(T - T_c) + \beta_1|\psi|^2 + \beta_2(|\psi_x|^2 + |\psi_y|^2\cos 2(\varphi_x - \varphi_y)) + \gamma M^2 + \alpha M^2 \tag{7a}$$

or

$$|\psi_x| = 0 \tag{7b}$$

$$\begin{cases} 0 = \alpha_S(T - T_c) + \beta_1|\psi|^2 + \beta_2(|\psi_y|^2 + |\psi_x|^2\cos 2(\varphi_x - \varphi_y)) + \alpha M^2 \\ \varphi_x - \varphi_y = \pi/2 \end{cases} \tag{8a}$$

or

$$|\psi_y| = 0. \tag{8b}$$

From these expressions we find the following conditions for M, ψ_x and ψ_y:

$$M = |\psi_x| = |\psi_y| = 0 \qquad \text{for } T > T_N \tag{9}$$

$$M^2 = \frac{\alpha_M}{\beta_M}(T_N - T) \qquad |\psi_x| = |\psi_y| = 0 \qquad \text{for } T^* < T \leqslant T_N \tag{10}$$

$$M^2 = \frac{\alpha_M}{\beta_M}(T_N - T^*) \qquad |\psi_x| = |\psi_y| = 0 \qquad \text{for } T_{c_+} < T \leqslant T^* \tag{11}$$

$$\begin{cases} M^2 = \dfrac{1}{\beta_M}[\alpha_M(T_N - T^*) - (\gamma + \alpha)|\psi_x|^2] \\ |\psi_x|^2 = \dfrac{\alpha_S}{\beta_1 + \beta_2}(T_{c_+} - T) \\ |\psi_y| = 0 \end{cases} \qquad \text{for } T_{c_-} < T \leqslant T_{c_+} \tag{12}$$

$$\begin{cases} M^2 = \dfrac{1}{\beta_M}\left[\alpha_M(T_N - T^*) - (\gamma + \alpha)|\psi_x|^2 - \alpha|\psi_y|^2\right] \\ |\psi_x|^2 = \dfrac{1}{\beta_1 + \beta_2}\left[\alpha_S(T_{c_+} - T) - (\beta_1 - \beta_2)|\psi_y|^2\right] \\ |\psi_y|^2 = \dfrac{\alpha_S}{\beta_1 + \beta_2}(T_{c_-} - T) \end{cases} \qquad \text{for } T \leqslant T_{c_-} \tag{13}$$

where

$$T_{c_+} = T_c - \frac{\gamma + \alpha}{\alpha_S}M^2 \tag{14}$$

$$T_{c_-} = T_c - \frac{1}{\alpha_S}\left[\alpha M^2 + (\beta_1 - \beta_2)|\psi_x|^2\right]. \tag{15}$$

T_c is the superconducting transition temperature in a system without magnetism. The complete solution to equations (9)–(13)—that is the explicit formulae for T_{c_-} and T_{c_+} are given in appendix A (A1) and (A2). The magnetic moment changes as $M^2 = (\alpha_M/\beta_M)(T_N - T)$ for temperatures higher than temperature T^*, then suddenly saturates at T^* ($T^* \sim T_c$) and becomes constant below this temperature: $M^2 = (\alpha_M/\beta_M)(T_N - T^*)$ in a normal (not superconducting) state. This temperature dependence of the magnetic moment is consistent with measurements by Aeppli *et al* [1, 9]. They observed a kink at $T^* \sim T_c$ and an almost constant value of the magnetic Bragg intensity below T^* for magnetic field $H > H_{c_2}$, that is when the system was not superconducting. The T^* temperature is introduced rather artificially into our free energy (3) in order to fit the existing experimental data [1, 9]. We shall comment more on this issue further into the text.

From the free energy density F_S (4) we get the linear pressure dependence of the superconducting transition temperature:

$$T_c = T_c^0 - a_0 p \tag{16}$$

where a_0 is a constant coefficient and T_c^0 a critical temperature T_c at zero pressure ($p = 0$). We also assume the squared magnetic moment to be a linear pressure function:

$$M^2 = M_0^2 \frac{p_N - p}{p_N} \tag{17}$$

where M_0 is the magnetic moment at $p = 0$, $M_0 = M(T, p = 0)$ and p_N ($p_N = p_c \simeq 4$ kbar) is the pressure at which the antiferromagnetism vanishes. In the superconducting system described by the free energy density (2) the magnetic and superconducting terms compete in the coupling term (5). This interaction leads to the splitting of the critical temperature T_c into T_{c_-} and T_{c_+} [8]:

$$T_{c_+} - T_{c_-} = \frac{|\gamma|}{\alpha_S}\frac{\beta_1 + \beta_2}{2\beta_2} M^2. \tag{18}$$

One can establish the pressure dependence of T_{c_+} and T_{c_-} from equations (16) and (17):

$$T_{c_+} = T_{c_+}^0 - a_+ p \tag{19}$$

$$T_{c_-} = T_{c_-}^0 - a_- p \tag{20}$$

where

$$T_{c_+}^0 = T_c^0 + \frac{|\gamma| - \alpha}{\alpha_S} M_0^2 \tag{21}$$

$$T_{c_-}^0 = T_c^0 - \frac{1}{\alpha_S}\left(\alpha + \frac{\beta_1 - \beta_2}{2\beta_2}|\gamma|\right) M_0^2 \tag{22}$$

$$a_+ = a_0 + \left(\frac{|\gamma| - \alpha}{\alpha_S}\right)\frac{M_0^2}{p_N} \tag{23}$$

$$a_- = a_0 - \frac{1}{\alpha_S}\left(\alpha + \frac{\beta_1 - \beta_2}{2\beta_2}|\gamma|\right)\frac{M_0^2}{p_N}. \tag{24}$$

To obtain the proper pressure behaviour (figure 1) the following constraints must be fulfilled:

$$a_+ > a_0 \qquad \text{and} \qquad a_- < a_0. \tag{25}$$

Together with the condition (1) they give the relations between the Ginzburg–Landau coefficients:

$$\frac{1}{2}\left(1-\frac{\beta_1}{\beta_2}\right)|\gamma| < \alpha < \frac{1}{4}\left(3-\frac{\beta_1}{\beta_2}\right)|\gamma|. \tag{26}$$

Now we turn to the magnetic Bragg scattering measurements [1, 9] (figure 2(a)). Since the neutron scattering intensity is proportional to M^2 we look at the magnetic moment and analyse it as a function of temperature. Taking into account that the coupling coefficients α and γ (5) are expected to be much smaller than the other G–L coefficients [12] and therefore neglecting terms which are higher than linear in α and γ from equations (2)–(5) we obtain:

$$M^2 = M_c^2 + a_M T \tag{27}$$

where

$$M_c^2 = \frac{\alpha_M}{\beta_M}(T_N - T^*) - a_M T_c \tag{28}$$

and

$$a_M = \frac{\alpha_S}{\beta_M}\frac{\gamma+\alpha}{\beta_1+\beta_2} \qquad \text{for } T_{c_-} < T \leqslant T_{c_+} \tag{29}$$

$$a_M = \frac{\alpha_S}{\beta_M}\frac{2\alpha+\gamma}{2\beta_1} \qquad \text{for } T \leqslant T_{c_-}. \tag{30}$$

In M^2 given by equations (27)–(30) a discontinuity arises at $T = T_{c_-}$ with a jump which is second order in the coefficients α and γ. Therefore it is negligible in the linear approximation. We present the full formula for M^2 in appendix A equations (A3)–(A7). It can be shown that even within this general description the results of this section still hold.

There are two characteristic temperatures, T_{c_+} and T_{c_-}, distinguished by the superconducting phase transitions; hence the change in the temperature dependence of the magnetic moment due to superconductivity can take place at either one of these temperatures. For M^2 increasing with temperature up to T_{c_+} and then decreasing, i.e. for a kink at $T = T_{c_+}$, the condition

$$a_M > 0 \qquad \text{for } T < T_{c_+} \tag{31}$$

is required, while for a kink at $T = T_{c_-}$ the following constraints are to be fulfilled:

$$a_M < 0 \qquad \text{for } T_{c_-} < T < T_{c_+} \tag{32}$$

and

$$a_M > 0 \qquad \text{for } T < T_{c_-}. \tag{33}$$

Condition (31) leads to the inequality

$$\alpha > |\gamma| \tag{34}$$

whereas from (32) and (33) it follows that

$$\frac{1}{2}|\gamma| < \alpha < |\gamma|. \tag{35}$$

It is evident that condition (34) is inconsistent with the pressure relation (26), while the conditions (26) and (35) yield the relation $\beta_1/\beta_2 < 1$ which contradicts the specific heat measurement data [13]. Put into words: thermodynamics requires that if the magnetic moment is reduced when the sample becomes superconducting then the tendency to become superconducting will be increased if the magnetism is removed. This implies that the continuation of the phase line between normal and superconducting phases for $p > p_c$ should lie above T_{c+} if it is extrapolated back to low pressures, in clear contrast to the data shown in figure 1 and also the more recent data of Boukhny *et al* [18].

We therefore conclude that it is *not possible to explain the pressure and neutron scattering data in the frame of the free energy density (2)–(5).* This paper does not address the alternative possibility that the splitting of T_c is due to the coupling of the superconductivity to the charge density wave [17, 19], except to note that even if the effect of magnetism is only to reduce both T_{c+} and T_{c-} due to a pair breaking mechanism [19], there should still be a break in the slope at T_{c+} at the pressure where magnetism is suppressed.

In the next paragraph we analyse the possibility of the rotation and decrease of the magnetic moment suggested by Blount *et al* [14] and Joynt [15]. The rotation of the magnetic moment can be equivalently described by an additional linearly independent magnetic moment $\boldsymbol{m}$ ($\boldsymbol{m} \perp \boldsymbol{M}$) included.

3. Two-magnetic-moment model

In this section we consider the possibility that the magnetic moment rotates at a temperature of the order of T_c in such a way that the observed Bragg scattering intensity is reduced. This requires two components of magnetization. Therefore we propose a revised G–L free energy density:

$$F = F_S + F_M + F_m + F_{SM} + F_{sm} \tag{36}$$

where

$$F_M = \begin{cases} \alpha_M (T^* - T_N) M^2 + \frac{1}{2}\beta_M M^4 & \text{for } T \leqslant T_m \\ \alpha_M (T - T_N) M^2 + \frac{1}{2}\beta_M M^4 & \text{for } T > T_m \end{cases} \tag{37}$$

$$F_m = \alpha_m (T - T_m) m^2 + \tfrac{1}{2}\beta_m m^4 \tag{38}$$

$$F_{sm} = \gamma' |\boldsymbol{m}\psi|^2 + \alpha' m^2 |\psi|^2 \tag{39}$$

and F_S, F_{SM} are given by equations (4) and (5). T_m is the Néel temperature of the magnetic moment $\boldsymbol{m}$ and $T_m \sim T_c$. The new coefficients α_m and β_m in (38) are positive. This free energy is correct to the fourth-order in the space of $\boldsymbol{M}$, $\boldsymbol{m}$ and ψ. For the sake of simplicity we have neglected the coupling term between the two magnetic moments and the superconducting order parameter ($\boldsymbol{m}\boldsymbol{M}(\psi_x\psi_y^* + \psi_x^*\psi_y)$) assuming it to have a little effect on the results. Another free energy term involving $\boldsymbol{M}$ and $\boldsymbol{m}$ ($\sim m^2 M^2$) is included implicitly in T_m and T^* by a proper diagonalization of the magnetic part of the free energy (see appendix B). As seen from (36) and (37), the magnetic moment M is constant in the absence of superconductivity and equals

$$M^2 = \frac{\alpha_M}{\beta_M}(T_N - T^*). \tag{40}$$

This approximation is correct for temperatures lower than a certain temperature of the order of T_c. We believe that this assumed temperature dependence of M^2 is due to a change in the Fermi surface and is of exclusively microscopic origin. However, in appendix B we present a phenomenological explanation of this fact, when relation (B4) is fulfilled. In this interpretation M^2 becomes constant below the temperature T_m equations (38) and (B7), i.e. the temperature at which the magnetic moment $\boldsymbol{m}$ appears. Although $T_m \sim T_c$, this reasoning is valid only if $T_m > T_{c+}$ which seems to be in agreement with the experimental data [1, 9].

Proceeding in the same way as in section 2, from the pressure requirements (1), (25) and the free energy density (36), we obtain the following conditions:

$$(|\gamma| - \alpha)M_0^2 > \alpha' m_0^2 \tag{41}$$

$$\left[\frac{1}{4}\left(3 - \frac{\beta_1}{\beta_2}\right)|\gamma| - \alpha\right] M_0^2 > \left[\alpha' + \frac{\beta_1 + \beta_2}{4\beta_2}\gamma'\right] m_0^2 \tag{42}$$

where we have assumed that m disappears at the same critical pressure p_N as M does (17):

$$m^2 = m_0^2 \frac{p_N - p}{p_N}. \tag{43}$$

Otherwise a kink in the pressure dependence of T_{c_-} and T_{c_+} should be observed, which is not the case (see figure 1) [2, 6, 7].

Since there are no coupling terms between m and M in the free energy density (36), it yields the same temperature dependence of M^2 as equations (27)–(30). Therefore, in order to obtain the appropriate temperature behaviour of M^2 [1, 9] (figure 2(a)) either (34) or (35) must be satisfied.

Now we are able to give the final conditions for the G–L coefficients in the free energy density which agrees with experiment [1, 2, 6, 7, 9] discussed in this paper. For M^2 increasing with the temperature up to $T = T_{c_-}$ and decreasing above this temperature, the conditions (35) and (41)–(42) are to be held. They lead to a simple constraint on α', which is necessary but not sufficient:

$$|\gamma| M_0^2 > 2\alpha' m_0^2. \tag{44}$$

When M^2 as a function of temperature has a kink at $T = T_{c_+}$, that is increases below this temperature and decreases above it, the conditions (34), (41) and (42) must be fulfilled and they yield the negative value of α':

$$\alpha' < 0. \tag{45}$$

4. $\left(\frac{1}{2}, 0, 1\right)$ neutron scattering intensity

We are going to consider both the magnetic moments M and m more thoroughly now. Here again we restrict the calculations to the terms linear in the coupling coefficients α, β, α' and γ', which yields a negligible M^2 and m^2 discontinuity at T_{c_-} in this approximation. A minimization of the free energy (36) as a function of magnetic moment m leads to the following temperature dependence of m^2:

$$m^2 = m_c^2 + \left(a_m - \frac{\alpha_m}{\beta_m}\right) T \tag{46}$$

where

$$m_c^2 = \frac{\alpha_m}{\beta_m} T_m - a_m T_c \tag{47}$$

and

$$a_m = \frac{\alpha_S}{\beta_m} \frac{\alpha'}{\beta_1 + \beta_2} \qquad \text{for } T_{c_-} < T \leqslant T_{c_+} \tag{48}$$

$$a_m = \frac{\alpha_S}{\beta_m} \frac{2\alpha' + \gamma'}{2\beta_1} \qquad \text{for } T \leqslant T_{c_-}. \tag{49}$$

We assume throughout this paper that the magnetic moments lie in the basal plane since the easy magnetic directions are confined to this plane. In the previous sections we were considering the neutron reflections at the reciprocal-lattice point $q_1 = (1, \frac{1}{2}, 0)$ [1, 9] (figure 2(*a*)). The magnetic Bragg scattering measurements revealed a different temperature dependence of the neutron scattering intensity at $q_2 = (\frac{1}{2}, 0, 1)$ [16] (figure 2(*b*)). Below a temperature of the order of T_c the $(\frac{1}{2}, 0, 1)$ intensity ceases to evolve and becomes constant. Actually, Aeppli *et al* [16] did not go to low enough temperatures to be positive about the independence of T of the measured intensity in the whole temperature range below T_c. Nevertheless, we assume here a constant value of the $(\frac{1}{2}, 0, 1)$ neutron scattering intensity below T_{c_+}, that is, we suggest this effect to be due to superconductivity. The neutron scattering intensity at the reciprocal-lattice point q reflects the magnetic vectors perpendicular to the q vector. For the sake of simplicity we choose a magnetic moment

$$M_1 = M + m \tag{50}$$

perpendicular to $q_2 = (\frac{1}{2}, 0, 1)$ which means that M_1^2 is detected in $(\frac{1}{2}, 0, 1)$ measurements. On this particular magnetic orientation we want to check, without going into the detailed calculation of a general case, whether the two-magnetic-moment model can interpret both neutron scattering experiments. It will yield some additional constraints on the G–L free energy coefficients (36)–(39). One of the possible configurations of the magnetic and reciprocal-lattice vectors considered, where instead of $q_1 = (1, \frac{1}{2}, 0)$ and $q_2 = (\frac{1}{2}, 0, 1)$ their projections on the XY plane, $(1, \frac{1}{2})$ and $(\frac{1}{2}, 0)$, were plotted, is presented in figure 3. M is the magnetic moment seen in $(1, \frac{1}{2}, 0)$ neutron scattering, while M_1 is detected in $(\frac{1}{2}, 0, 1)$ measurements. The temperature dependence of M^2 has been considered in the previous paragraphs of this paper, equations (27)–(30), (40). According to [16] M_1^2 is temperature independent for $T \leqslant T_c \sim T_{c_+}$:

$$M_1^2 = \text{constant} \qquad \text{for } T < T_{c_+}. \tag{51}$$

Assuming the temperature-dependent corrections to M (27)–(30) and m (46)–(49) to be small, experimentally estimated as about 5% of the total magnetic moment values [1, 9], we linearize M and m in T and insert them into equation (50). Then the condition (51) leads to the following constraints on the G–L coefficients:

$$f\left(\frac{\gamma + \alpha}{\beta_1 + \beta_2}, \frac{\alpha'}{\beta_1 + \beta_2}\right) = 0 \tag{52}$$

$$f\left(\frac{\gamma + 2\alpha}{2\beta_1}, \frac{\gamma' + 2\alpha'}{2\beta_1}\right) = 0 \tag{53}$$

where

$$f(x, y) = \frac{\alpha_S}{\beta_M}x + \frac{1}{\beta_m}(\alpha_S y - \alpha_m). \tag{54}$$

We solve equations (52) and (53) and obtain

$$\beta_1 = \frac{\alpha_S}{2\alpha_m}\left[\frac{\beta_m}{\beta_M}(\gamma + 2\alpha) + \gamma' + 2\alpha'\right] \tag{55}$$

$$\beta_2 = \frac{\alpha_S}{2\alpha_m}\left[\frac{\beta_m}{\beta_M}\gamma - \gamma'\right]. \tag{56}$$

According to experiment [13], β_1 and β_2 coefficients should obey the following relation:

$$\beta_1 > \beta_2 > 0. \tag{57}$$

From (55)-(57) we then have:

$$\frac{\beta_m}{\beta_M}\gamma - \gamma' > 0 \tag{58}$$

and

$$\frac{\beta_m}{\beta_M}\alpha + \gamma' + \alpha' > 0. \tag{59}$$

Since γ (5) is negative, inequality (58) leads to a negative γ' value and finally relation (58) is equivalent to:

$$\gamma' = -|\gamma'| \qquad |\gamma'| > \frac{\beta_m}{\beta_M}|\gamma|. \tag{60}$$

Therefore, we have obtained conditions (55), (56) and (59), (60) which are to be fulfilled by G–L free energy coefficients. However, we cannot forget about the constraints which follow from the M^2 temperature evolution requirements (34), (35) and those which are necessary to fit the pressure data (41), (42). One can easily check that the conditions (34) (kink in M^2 at $T = T_{c_+}$) and (59), (60) lead to a negative value of α', while the constraint (35) (kink in M^2 at $T = T_{c_-}$) along with equations (59), (60) yield a positive α' value. From equations (41), (42) we get more information about the magnetic moment values at pressure $p = 0$, that is M_0 (17) and m_0 (43). It is more convenient for this purpose to use the experimentally established β_2/β_1 ratio: $\beta_2/\beta_1 \simeq 0.4$ [13], just to get rid the of β_1 and β_2 coefficients in (42). The relation $\beta_2/\beta_1 = 0.4$ along with the β_1 (55) and β_2 (56) formulae allow the reduction of one of the coupling coefficients through the equation:

$$\alpha' = \frac{7}{4}\left(|\gamma'| - \frac{\beta_m}{\beta_M}|\gamma|\right) + \frac{\beta_m}{\beta_M}(|\gamma| - \alpha) \tag{61}$$

so we can consider γ', γ and α parameters as the only independent ones in all the conditions. It is straightforward to show that α' given by equation (61) obeys equations (34), (35) and (59), (60). Returning to M_0 and m_0 magnitudes, for $\alpha > |\gamma|$ (34), we obtain from (41), (42) that

$$m_0^2 > g_0 M_0^2 \tag{62}$$

where

$$g_0 = \max\left\{\frac{\alpha - |\gamma|}{|\alpha'|}, \frac{\alpha - \frac{1}{8}|\gamma|}{|\alpha'| + \frac{7}{8}|\gamma'|}\right\}.$$

The condition above should be fulfilled when a kink in $\left(1, \frac{1}{2}, 0\right)$ neutron scattering intensity appears at T_{c_+} (34). In order to have m_0, M_0 solutions of (41), (42) when condition (35) is held, that is in the case of the $\left(1, \frac{1}{2}, 0\right)$ neutron scattering peak at T_{c_-}, another constraint has to be fulfilled:

$$\alpha' < \frac{7}{8}|\gamma'|. \tag{63}$$

Inequality (63) is a necessary condition to make sense of relations (41) and (42). Finally, we obtain from (41), (42) the constraint on the relative m_0 and M_0 values:

$$\frac{\alpha - \frac{1}{8}|\gamma|}{\frac{7}{8}|\gamma'| - \alpha'} < \frac{m_0^2}{M_0^2} < \frac{|\gamma| - \alpha}{\alpha'} \tag{64}$$

and another condition which follows directly from (64):

$$|\gamma\gamma'| - |\gamma|\alpha' - |\gamma'|\alpha > 0. \tag{65}$$

We have been looking here at the additional constraints on the fourth-order coefficients in the Ginzburg–Landau free energy that follow from the requirement of a constant magnetic moment detected in $\left(\frac{1}{2}, 0, 1\right)$ neutron scattering measurements [16] (figure 2(b)). We have assumed T_{c_+} to be a characteristic temperature at which the magnetic moment $M_\parallel$ (51) becomes constant. Nevertheless it is straightforward to show that $M_\parallel$ cannot be constant above T_{c_+}. Let us look at the temperatures $T > T_m$ first. Since T_m is the Néel temperature for m (B6), there is only one magnetic moment M left at $T > T_m$. $M_\parallel$ is simply M's projection in a particular direction (figure 3) and shows the same temperature dependence as M does (B9). Therefore $M_\parallel$ is a decreasing function of temperature for $T > T_m$ as M is (B9). In the temperature range $T_{c_+} < T < T_m$, on the other hand, we obtain from the free energy (B8) a constant M^2 value (40) and $m^2 = (T_m - T)\,\alpha_m/\beta_m$. Therefore (50) cannot lead to a constant $M_\parallel$ value, otherwise $\alpha_m = 0$ causing m to vanish, making no sense for this approach.

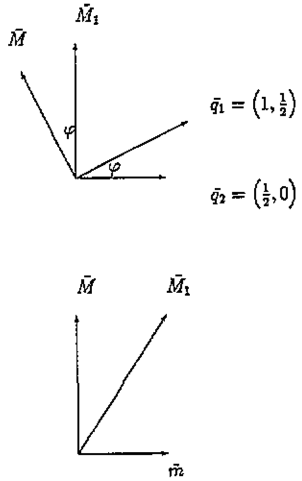

Figure 3. The relative orientation of the magnetic moments M, m and $M_\parallel$ and the neutron scattering vectors q_1 and q_2, where $\tan\varphi = \frac{1}{2}$.

5. Conclusions

We have considered superconducting UPt$_3$ in zero magnetic field. Our interest has been focused on the hydrodynamic pressure [2, 6, 7] and neutron scattering experiments [1, 9, 16]. We have shown that the pressure dependence of the transition temperatures and the abrupt change in the $\left(1, \frac{1}{2}, 0\right)$ neutron scattering intensity at $T \sim T_c$ [1, 9] cannot be explained quantitatively within a simple two-component superconducting order parameter which couples to one-component antiferromagnetism. As one way of reconciling this problem we have suggested the existence of another magnetic moment which emerges at $T \sim T_c$. This generalized approach of the two independent magnetic moments coupling to the superconductivity allowed us to obtain a concise picture of the phenomena discussed and yields several stringent constraints on the fourth-order coefficients in the Ginzburg–Landau free energy density (36). We have concluded that the kink in a $\left(1, \frac{1}{2}, 0\right)$ neutron scattering intensity may exist at T_{c+} when relations (34), (41) and (42) between the G–L coefficients are obeyed, or at T_{c-} under the conditions of (35) and (41), (42). If we interpret the results of $\left(\frac{1}{2}, 0, 1\right)$ Bragg magnetic scattering experiments [16] as characteristic features for all temperatures below T_c and assume that the magnetic moment orientation is as in figure 3, we can express the β_1 and β_2 G–L coefficients in terms of the coupling constants (55) and (56). The requirement $\beta_1 > \beta_2 > 0$ leads to a negative value of the coupling constant γ' (39, 60) and a negative α' (39) coefficient when a peak in $\left(1, \frac{1}{2}, 0\right)$ neutron scattering intensity is at T_{c_+}, or a positive α' value for a peak at T_{c_-}. These considerations also yield some constraints on the zero-pressure magnetic moments values (62), (64) and coupling coefficients (63), (65). We have evaluated constraints (62)–(65) for the experimentally established ratio $\beta_2/\beta_1 \simeq 0.4$ [13]. This given value of β_2/β_1 allows us to express one of the G–L coupling coefficients in terms of the others (61).

We have considered two magnetic moments in a crystal basal plane only. However, we cannot exclude any out-of-plane moments. There is always the possibility of magnetic structure following a recently discovered structural modulation in a crystal [17]. Unfortunately, the resolution of neutron scattering measurements may be too small to be decisive. For completeness it should be added that despite the large amount of experimental evidence the main facts seem not to be established—in particular the phase diagram in the p–T plane measured by Boukhny *et al* [18] differs from [2] because where the slope of the T_{c_-} curve is positive condition (1) does not hold. Moreover, recent x-ray resonant magnetic and neutron magnetic scattering measurements [19] show no correlation between the split superconducting transition and the weak antiferromagnetic order in UPt_3; as they also find no evidence of magnetic moment rotation their results, together with the conclusions of this paper, suggest other possible issues such as symmetry-breaking fields of structural origin [17] and the existence of two one-dimensional superconducting states.

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Appendix A

$$T_{c_+} = T_c - \frac{\alpha + \gamma}{\alpha_S}\frac{\alpha_M}{\beta_M}\left(T_N - T^*\right) \tag{A1}$$

$$T_{c_-} = T_c + \frac{\alpha_M}{\alpha_S}\frac{\gamma\left(\beta_1 - \beta_2\right) - 2\beta_2\alpha}{2\beta_2\beta_M - \gamma\left(\alpha + \gamma\right)}\left(T_N - T^*\right) \tag{A2}$$

$$M^2 = M_c^2 + a_M T \tag{A3}$$

$$M_c^2 = \begin{cases} \dfrac{\beta_1 + \beta_2}{\lambda_+}\left[\alpha_M\left(T_N - T^*\right) - \dfrac{\alpha_S}{\beta_1 + \beta_2}\left(\alpha + \gamma\right)T_c\right] & \text{for } T_{c_-} < T \leqslant T_{c_+} \\ \dfrac{2\beta_2}{\lambda_-}\left[2\alpha_M\beta_1\left(T_N - T^*\right) - \alpha_S\left(2\alpha + \gamma\right)T_c\right] & \text{for } T \leqslant T_{c_-} \end{cases} \tag{A4}$$

$$a_M = \begin{cases} \dfrac{\alpha_S}{\lambda_+}\left(\alpha + \gamma\right) & \text{for } T_{c_-} < T \leqslant T_{c_+} \\ \dfrac{2\alpha_S}{\lambda_-}\beta_2\left(2\alpha + \gamma\right) & \text{for } T \leqslant T_{c_-} \end{cases} \tag{A5}$$

$$\lambda_+ = \beta_M\left(\beta_1 + \beta_2\right) - \left(\alpha + \gamma\right)^2 \tag{A6}$$

$$\lambda_- = 4\beta_1\beta_2\beta_M - 4\beta_2\alpha\left(\alpha + \gamma\right) - \gamma^2\left(\beta_1 + \beta_2\right). \tag{A7}$$

Appendix B

The complete magnetic free energy for the magnetic moments M and m ($m \perp M$) at the temperatures $T < T^*$ is

$$F_{magn} = A_M (T - T_N) M^2 + \frac{1}{2} B_M M^4 + A_m \left(T - T^*\right) m^2 + \frac{1}{2} B_m m^4 + C m^2 M^2 \qquad \text{(B1)}$$

where T_N and T^* are the Néel temperatures for M and m magnetic moments respectively.

We assume $T_N > T^*$. From the minimization of F_{magn} one gets:

$$M^2 = \frac{B_M B_m}{B_M B_m - C^2} \left[\frac{A_M}{B_M} T_N - \frac{A_m}{B_m} \frac{C}{B_M} T^* - \left(\frac{A_M}{B_M} - \frac{A_m}{B_m} \frac{C}{B_M} \right) T \right] \qquad \text{(B2)}$$

and

$$m^2 = \frac{1}{B_M B_m - C^2} \left(A_m B_M T^* - A_M C T_N \right) - \frac{A_m}{B_m} T. \qquad \text{(B3)}$$

For a particular choice of the coupling coefficient

$$C = \frac{A_M}{A_m} B_m \qquad \text{(B4)}$$

M^2 attains a constant value:

$$M^2 = \frac{A_M A_m^2}{A_m^2 B_M - A_M^2 B_m} \left(T_N - T^* \right) \qquad \text{(B5)}$$

and

$$m^2 = \frac{A_m}{B_m} (T_m - T) \qquad \text{(B6)}$$

where

$$T_m = \frac{A_m^2 B_M - A_M^2 B_m T_N / T^*}{A_m^2 B_M - A_M^2 B_m} T^*. \qquad \text{(B7)}$$

The temperature T^* should be of the order of T_N to give a positive value of T_m. From (B5) and (B6) we can see that the magnetic free energy can be written as

$$F_{magn} = \alpha_M \left(T^* - T_N \right) M^2 + \frac{1}{2} \beta_M M^4 + \alpha_m (T - T_m) m^2 + \frac{1}{2} \beta_m m^4 \qquad \text{(B8)}$$

for $T < T_m$, and

$$F_{magn} = \alpha_M (T - T_N) M^2 + \frac{1}{2} \beta_M M^4 \qquad \text{(B9)}$$

for $T > T_m$.

This is the free energy of the two magnetic moments (37)–(38), that we use in this paper. The new G–L coefficients are given in terms of the old ones as

$$\alpha_M = A_M A_m^2 \qquad \text{(B10)}$$

$$\beta_M = A_m^2 B_M - A_M^2 B_m \qquad \text{(B11)}$$

$$\alpha_m = A_m \qquad \text{(B12)}$$

$$\beta_m = B_m. \qquad \text{(B13)}$$

These considerations are relevant only when condition (B4) is fulfilled.

References

[1] Aeppli G, Broholm C, Bucher E and Bishop D J 1991 *Physica* B **171** 278
[2] Hayden S, Taillefer L, Vettier C and Flouquet J 1992 *Phys. Rev.* B **46** 8675
[3] Fisher R, Kim S, Woodfield B, Phillips N, Taillefer L, Hasselbach K, Flouquet J, Giorgi A and Smith J L 1989 *Phys. Rev. Lett.* **62** 1411
[4] Hasselbach K, Taillefer L and Flouquet J 1990 *Phys. Rev. Lett.* **63** 93
[5] Adenwalla S, Lin W, Ran Q Z, Zhao Z, Ketterson J B, Sauls J A, Taillefer L, Hinks D G, Levy M and Sarma B K 1990 *Phys. Rev. Lett.* **65** 2298
[6] Trappmann T, von Löhneysen H and Taillefer L 1991 *Phys. Rev.* B **43** 13714
[7] von Löhneysen H, Trappmann T and Taillefer L 1992 *J. Magn. Magn. Mater.* **108** 49
[8] Joynt R, Volovik G, Mineev V and Zhitomirskii M 1990 *Phys. Rev.* B **42** 2014
[9] Aeppli G, Bishop D, Broholm C, Bucher E, Siemensmeyer K, Steiner M and Stüsser N 1989 *Phys. Rev. Lett.* **63** 676
[10] Joynt R 1993 *Phys. Rev. Lett.* **71** 3015
[11] van Dijk N H, de Visser A and Franse J J M 1993 *J. Low Temp. Phys.* **93** 101
[12] Sauls J A 1993 *Preprint*
Sigrist M and Ueda K 1991 *Rev. Mod. Phys.* **63** 239
[13] Hasselbach K, Lacerda A, Behnia K, Taillefer L, Flouquet J and de Visser A 1990 *J. Low Temp. Phys.* **81** 299
[14] Blount E I, Varma C M and Aeppli G 1990 *Phys. Rev. Lett.* **64** 3074
[15] Joynt R 1990 *J. Phys.: Condens. Matter* **2** 3415
[16] Aeppli G, Bucher E, Broholm C, Kjems J K, Baumann J and Hufnagl J 1988 *Phys. Rev. Lett.* **60** 615
[17] Midgley P A, Hayden S M and Taillefer L 1993 *Phys. Rev. Lett.* **70** 678
[18] Boukhny M, Bullock G L, Shivaram B S and Hinks D G 1994 *Phys. Rev. Lett.* **73** 1707
[19] Isaacs E D, Zschack P, Broholm C L, Burns C, Aeppli G, Ramirez A P, Erwin R W, Stücheli N and Bucher E 1995 *Phys. Rev. Lett.* **75** 1178