Quadrupolar phases in UPd_3

J. Luettmer-Strathmann, C. Kappler,* and M. B. Walker
Department of Physics, University of Toronto, Toronto, Ontario, Canada M5S 1A7
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UPd_3 is a hexagonal crystal that exhibits quadrupolar ordering at low temperatures. The order parameter has three components corresponding to modulations in three directions in the basal plane of the hexagonal crystal. For the case that no symmetry-breaking fields are present the ordered phase has been identified as a trigonal triple-Q phase. Using group theory, we show that under the influence of symmetry-breaking fields up to eight distinct quadrupolar ordered phases are realized in the crystal. We deduce the phase diagram from a Landau free energy that includes magnetic fields and stress and find lines of tetracritical points and one point at which eight phases coexist, an "octacritical" point. By reviewing renormalization-group results we address the effect of critical fluctuations on the phase diagram. Finally, we discuss experimental tests of our model for the quadrupolar phases of UPd_3.

I. INTRODUCTION

Due to the magnetic dipole and electric quadrupole moments of its uranium ions UPd_3, a hexagonal crystal with space group $P6_3/mmc$, exhibits two types of ordered phases at low temperatures. In the absence of external fields, UPd_3 undergoes a transition to a quadrupolar ordered phase at a temperature $T_1 \approx 7$ K and a magnetic transition at $T_2 \approx 4.5$ K.[1–9] In this work we are going to focus on quadrupolar ordering in UPd_3.

Diffraction experiments[3,5] indicate that the ordering is associated with modulations of wave vector $\mathbf{Q}_1 = \frac{1}{2}\mathbf{a}^*$, and that the transition at T_1 is continuous within the experimental uncertainty. In an earlier work, Walker *et al.*[7] determined the symmetry of the order parameter and concluded that the order parameter belongs to the one-dimensional irreducible representation B_{2g} of the little cogroup $D_{2h}(mmm)$ of the wave vector $\mathbf{Q}_1$. Since the star of $\mathbf{Q}_1$ consists of three wave vectors $\mathbf{Q}_i$, $i = 1, 2, 3$, cf. Fig. 1, the order parameter has three components η_i each describing the amplitude of an ordered quadrupole moment and ion-displacement wave corresponding to wave vector $\mathbf{Q}_i$. Depending on the number of nonzero components of the order parameter, we refer to ordered phases as single-, double-, and triple-Q phases, respectively. In the absence of external fields, the quadrupolar ordered phase in UPd_3 is a trigonal triple-Q phase, i.e., $\eta_1 = \eta_2 = \eta_3$.[7]

While a free energy that describes UPd_3 in the absence of external fields is symmetric in the components η_i of the order parameter,[7] this is no longer true for a free energy that includes the coupling of magnetic fields and stress to the order parameter.[9] Including symmetry-breaking terms to lowest order we find that new quadrupolar ordered phases are stabilized, where the type of phase depends on three parameters, namely two field and one temperaturelike variable. For general applied fields, the symmetry between all three components of the order parameter is broken, but the three-dimensional parameter space contains two-dimensional "surfaces of special symmetry" where only one of the components of the order parameter is singled out, while the free energy is symmetric in the other two. In connection with the thermal-expansion measurements of Zochowsky and McEwen,[6] we discussed in our previous work[9] the quadrupolar ordered phases of UPd_3 for external fields on such a surface of special symmetry. In that case, the phase diagram consists of four distinct phases separated by lines of continuous phase transitions that meet at a tetracritical point. In this work we go beyond the surfaces of special symmetry in parameter space and investigate the quadrupolar ordered phases of UPd_3 under the influence of more general symmetry-breaking fields.

The outline of the paper is as follows: In Sec. II we use group-theoretical methods to determine what type of quadrupolar ordered phases can occur in UPd_3. From the Landau free energy presented in Ref. 9 we derive in Sec. III the phase diagram for quadrupolar ordering of UPd_3 in the three-dimensional parameter space of two field and one temperaturelike variable. The phase diagram consists of eight distinct quadrupolar ordered phases, separated by sheets of continuous phase transi-

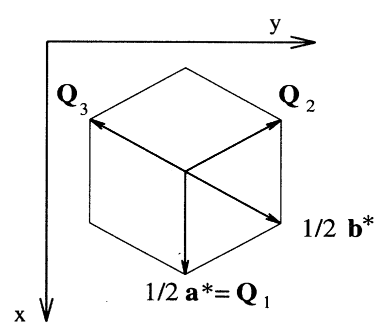

FIG. 1. Basal plane of the hexagonal lattice in reciprocal space with definition of coordinate axes and the elements $\mathbf{Q}_i$ of the star of $\mathbf{Q}_1$.

tions. These critical surfaces meet in tetracritical lines which in turn converge at one point in the diagram. At this point, all eight distinct quadrupolar ordered phases coexist, which makes this point an "octacritical" point. In Sec. IV we review renormalization-group results to discuss the effect of critical fluctuations on our system. Experimental tests of predictions of our model for the quadrupolar phases of UPd_3 are the subject of our concluding section, Sec. V.

II. SYMMETRY OF QUADRUPOLAR ORDERED PHASES

UPd_3 can order in a number of distinct quadrupolar phases that differ from each other by their symmetry properties. In this section we use group-theoretical methods to determine the ordered phases and their space groups.

In the absence of symmetry-breaking fields the space group of UPd_3 is $G = P6_3/mmc$. The symmetry operations of G are irreducibly represented by 3×3 matrices on the space $\mathcal{E}$ spanned by the components η_i of the order parameter. The set of distinct matrices of this irreducible representation forms a group, the image I of G on $\mathcal{E}$, so that the space group G can be written as

$$G = K \times \tilde{I}, \tag{1}$$

where the kernel K contains all those elements of G that are represented on $\mathcal{E}$ by the unit matrix, while $\tilde{I}$ is isomorphic to the image I. We constructed the image of the space group $G = P6_3/mmc$ and found it to be isomorphic to the cubic group O_h.

Since each ordered phase defines a subspace $\mathcal{E}_S$ of $\mathcal{E}$ that is invariant under a subgroup I_S of the image I we can use the analog of Eq. (1) to determine the space group G_S of the phase[10]

$$G_S = K \times \tilde{I}_S, \tag{2}$$

where K is the kernel defined in Eq. (1), and where $\tilde{I}_S$ is isomorphic to I_S.

For a system with three-dimensional order parameter and image O_h the free energy in the absence of symmetry-breaking fields depends only on the following three functions of the components η_i of the order parameter, the so-called entire rational basis of invariants:[10,11]

$$J_1 = \eta_1^2 + \eta_2^2 + \eta_3^2,\ J_2 = \eta_1^4 + \eta_2^4 + \eta_3^4,$$
$$J_3 = \eta_1^2\eta_2^2\eta_3^2. \tag{3}$$

By minimizing the most general expression for a free energy in terms of the invariants J_i, Gufan and Sakhnenko[11] obtained all possible ordered phases of such a system. In the first seven rows of Table I we present a list of these phases along with the space groups obtained through Eq. (2). Terms up to eighth order in the order parameter have to be included in the free energy to stabilize a given phase.

Including only terms to fourth order in the order parameter, the free energy in the absence of symmetry-breaking fields can be written as

$$G_0 = AJ_1 + (B + C)J_1^2 - CJ_2, \tag{4}$$

where $A = \alpha(T - T_1)$ with $\alpha > 0$ gives rise to a continuous phase transition at the temperature $T = T_1$, while thermodynamic stability requires that $B > 0$ and $3B + 2C > 0$. For $T < T_1$ the free energy G_0 is minimized either by a single-Q phase ($C > 0$) or by the trigonal triple-Q phase ($C < 0$). Walker *et al.*[7] determined that $C < 0$ is appropriate for UPd_3. To indicate that the hexagonal normal phase ([hN]) and the trigonal triple-Q phase ([1=2=3]) can be realized in UPd_3 in the absence

TABLE I. Quadrupolar ordered phases of UPd_3 and their space groups. The last column indicates when the phases are expected to occur: (a) in the absence of symmetry-breaking fields, (s) for fields on surfaces of special symmetry, and (n) for fields not on surfaces of special symmetry.

[hN]	$\eta_1=\eta_2=\eta_3=0$	Hexagonal normal	$P6_3/mmc$	(a)
[1=2=3]	$\eta_1=\eta_2=\eta_3$	Trigonal triple-Q	$P\bar{3}m1$	(a)
[oi]	$\eta_i,\ \eta_j=\eta_k=0$	Orthorhombic single-Q	$P\frac{2_1}{n}\frac{2}{m}\frac{2_1}{n}$	(s)
[j=k]	$\eta_i=0,\ \eta_j=\eta_k$	Orthorhombic double-Q	$C\frac{2}{c}\frac{2}{m}\frac{2_1}{b}$	(s)
[i,j=k]	$\eta_i,\ \eta_j=\eta_k$	Monoclinic triple-Q	$C\frac{2}{m}$	(s)
[j,k]	$\eta_i=0,\ \eta_j,\ \eta_k$	Monoclinic double-Q	$P11\frac{2_1}{a}$	(n)
[1,2,3]	$\eta_1,\ \eta_2,\ \eta_3$	Triclinic triple-Q	$P\bar{1}$	(n)
[oN]	$\eta_1=\eta_2=\eta_3=0$	Orthorhombic normal	$C\frac{2}{c}\frac{2}{m}\frac{2_1}{m}$	(s)
[mN]	$\eta_1=\eta_2=\eta_3=0$	Monoclinic normal	$P11\frac{2_1}{m}$	(n)
[mi]	$\eta_i,\ \eta_j=\eta_k=0$	Monoclinic single-Q	$P11\frac{2_1}{a}$	(n)

of symmetry-breaking fields we marked these two phases with an "(a)" in the last column of Table I.

To describe UPd_3 in the presence of stress and magnetic fields the free energy G_0 is generalized to include terms in the applied fields and the order parameter components that are invariant under the operations of $P6_3/mmc$. In Sec. III we discuss this free energy and the resulting ordered phases in detail. At this point we consider changes in symmetry due to the applied fields. Since fields along the z direction preserve the symmetry of the crystal they are not further considered here. Some components of applied fields, e.g., the xz component of the stress tensor, do not couple to the order parameter to lowest order and are therefore not discussed here. Instead, we focus on fields in the basal plane and require $\sigma_{xz} = \sigma_{yz} = 0$ for the components of the stress tensor, and $H_xH_z = H_yH_z = 0$ for the magnetic field.

When UPd_3 is subject to symmetry-breaking fields even its disordered phase is not invariant under all operations of the hexagonal space group $P6_3/mmc$. To be definite, consider a crystal at a temperature well above the transition temperature and subject to stress along the y axis. This system has lost its invariance under rotations through $2\pi/3$ about the z axis (C_3 operation) so that the symmetry of the disordered phase is reduced to the orthorhombic space group $C\frac{2}{c}\frac{2}{m}\frac{2_1}{m}$, where the basis vectors of the unit cell are parallel to the x, y, and z axis. If the stress is applied instead along the axis C_3y or C_3^2y, the resulting normal phases are again orthorhombic, and their unit cells are aligned with C_3x and C_3^2x, respectively. In general, applied fields that break only the C_3 symmetry induce an orthorhombic normal phase ($[oN]$) with space group $C\frac{2}{c}\frac{2}{m}\frac{2_1}{m}$, where the orientation of the unit cell can be one of the three mentioned above, depending on the direction of the applied field.

Symmetry-breaking fields do not only reduce the symmetry of the normal phase but also affect quadrupolar ordering. First of all, the trigonal triple-Q phase cannot be an ordered phase of UPd_3 subject to a C_3-symmetry-breaking field, since the space group of an ordered phase has to be a subgroup of the space group of the normal phase.[12] Furthermore, a field that breaks only the C_3 symmetry singles out one of the components of the order parameter while maintaining the symmetry between the other two components. We refer to such fields as "fields on surfaces of special symmetry" and provide expressions for them in terms of components of the magnetic field and the stress tensor in Sec. III. In our previous work,[9] we determined the quadrupolar ordered phases of UPd_3 subject to applied fields on a surface surface of special symmetry and found them to be the orthorhombic single-Q ($[oi]$), orthorhombic double-Q ($[j{=}k]$), and monoclinic triple-Q ($[i,j{=}k]$) phases. We have marked these ordered phases and the orthorhombic normal phase ($[oN]$) with an "(s)" in the last column of Table I to indicate that they can be realized for applied fields on surfaces of special symmetry.

For an applied field not on a surface of special symmetry, not only the C_3 symmetry, but also the symmetries of rotation by π about the x and y axis are broken. Consequently, the symmetry of the disordered phase is reduced to the monoclinic space group $P11\frac{2_1}{m}$. Due to the reduced symmetry of the normal phase, the ordered phases discussed before and marked with (a) and (s) in Table I are not available in this case. Applied fields not on a surface of special symmetry break the symmetry between all three components of the order parameter. The corresponding free energy, which we discuss in detail in Sec. III, yields single-Q, monoclinic double-Q ($[j,k]$) and triclinic triple-Q ([1,2,3]) phases. Under the influence of general symmetry-breaking fields in the basal plane the single-Q phases are not orthorhombic but monoclinic ($[mi]$). The single-Q phase $[m1]$, for example, has the monoclinic space group $P11\frac{2_1}{a'}$ with basis vectors $\mathbf{a}' = 2\mathbf{a}$, $\mathbf{b}$, and $\mathbf{c}$. The unit cells of the space groups of the other two monoclinic single-Q phases are obtained from this one by rotation through $\pm 2\pi/3$ about the z axis. The space groups of the monoclinic double-Q phases differ from those of the monoclinic single-Q phases by the size of the unit cell. The phase [1,2], for example, has the space group $P11\frac{2_1}{a'}$ with basis vectors $\mathbf{a}' = 2\mathbf{a}$, $\mathbf{b}' = 2\mathbf{b}$, and $\mathbf{c}$.

The quadrupolar phases that occur for a field not on a surface of special symmetry are indicated by an "(n)" in the last column of Table I. Looking at these phases we note, first of all, that there are seven ordered phases in addition to the monoclinic normal phase, namely three monoclinic single-Q, three monoclinic double-Q, and a triclinic triple-Q phase. We thus expect up to eight distinct quadrupolar phases when UPd_3 is exposed to general symmetry-breaking fields in the basal plane. From the group-subgroup relationships between the phases we expect a phase sequence of

$$[mN] \to [mi] \to [i,j] \to [1,2,3]. \tag{5}$$

III. LANDAU MODEL

In this section we determine the phase-diagram for quadrupolar ordered UPd_3 in the presence of symmetry-breaking fields. The appropriate Landau free energy, including coupling between the external fields and the components of the order parameter to lowest order, and invariant under all symmetry operations of the space group $P6_3/mmc$, has been presented in Ref. 9 and is most conveniently written as

$$G = \tilde{A}\sum_{i=1}^{3}\eta_i^2 + B\left(\sum_{i=1}^{3}\eta_i^2\right)^2 + C\sum_{i\neq j}\eta_i^2\eta_j^2 + 2f\eta_1^2 + (-f+g)\eta_2^2 + (-f-g)\eta_3^2, \tag{6}$$

where the coefficients $\tilde{A}$, f, and g depend on external fields as specified below. In zero field this free energy reduces to, cf. Eq. (4),

$$G_0 = A\sum_{i=1}^{3}\eta_i^2 + B\left(\sum_{i=1}^{3}\eta_i^2\right)^2 + C\sum_{i\neq j}\eta_i^2\eta_j^2, \tag{7}$$

where $A = \alpha(T - T_1)$ with $\alpha > 0$, $C < 0$ gives rise to a transition to the trigonal triple-Q phase at $T = T_1$, while $3B + 2C > 0$ for thermodynamic stability.[7]

One effect of external fields is to change the transition temperature T_1. This is reflected in the coefficient $\tilde{A}$ which we write as

$$\begin{aligned}\tilde{A} = A &+ [g_{1r}(\sigma_{xx} + \sigma_{yy}) + g_{1z}\sigma_{zz}] \\ &+ [G_{1r}(H_x^2 + H_y^2) + G_{1z}H_z^2], \qquad (8)\end{aligned}$$

where the σ_{ij} and H_i, $i,j \in \{x,y,z\}$, denote applied stress and magnetic fields, respectively, while $g_{1r}, g_{1z}, G_{1,r}$ and $G_{1,z}$ are coupling coefficients. The symmetry-breaking effect of the applied fields is apparent in the second line of Eq. (6), where the coefficients f and g are a shorthand notation for the following combinations of external fields:

$$f = g_2(\sigma_{xx} - \sigma_{yy}) + G_2(H_x^2 - H_y^2), \qquad (9)$$

$$g = 2\sqrt{3}\,(g_2\sigma_{xy} + G_2H_xH_y)\,, \qquad (10)$$

where g_2 and G_2 are coupling coefficients.

To derive the phase diagram it is convenient to introduce "polar coordinates." We define the length η of the order parameter, and two angles θ and ϕ through

$$\eta_1 = \eta\cos\theta, \qquad (11a)$$

$$\eta_2 = \eta\sin\theta\sin\phi, \qquad (11b)$$

$$\eta_3 = \eta\sin\theta\cos\phi. \qquad (11c)$$

By minimizing the free energy (6) and testing the solutions for stability we arrived at the distribution of phases summarized in Table II. We note, first of all, that eight

TABLE II. Quadrupolar ordered phases of UPd_3 in the presence of symmetry-breaking fields.

Monoclinic single-Q			
η_1, $\eta_2 = \eta_3 = 0$	$\eta^2 = \frac{-\tilde{A} - 2f}{2B}$ $\cos^2\theta = 1$	$\tilde{A} < -2f$ $\tilde{A} > -\frac{3B+2C}{C}f + \frac{B}{C}g$ $\tilde{A} > -\frac{3B+2C}{C}f - \frac{B}{C}g$	$3f + \lvert g\rvert < 0$ $3f + \lvert g\rvert < 0, g < 0$ $3f + \lvert g\rvert < 0, g > 0$
η_2, $\eta_1 = \eta_3 = 0$	$\eta^2 = \frac{-\tilde{A} + f - g}{2B}$ $\cos^2\theta = 0$ $\cos^2\phi = 0$	$\tilde{A} < f - g$ $\tilde{A} > \frac{3B+C}{C}f - \frac{B+C}{C}g$ $\tilde{A} > f - \frac{2B+C}{C}g$	$3f + \lvert g\rvert > 0, g < 0$ $3\lvert f\rvert - \lvert g\rvert < 0, g < 0$ $3f - \lvert g\rvert > 0, g < 0$
η_3, $\eta_1 = \eta_2 = 0$	$\eta^2 = \frac{-\tilde{A} + f + g}{2B}$ $\cos^2\theta = 0$ $\cos^2\phi = 1$	$\tilde{A} < f + g$ $\tilde{A} > \frac{3B+C}{C}f + \frac{B+C}{C}g$ $\tilde{A} > f + \frac{2B+C}{C}g$	$3f + \lvert g\rvert > 0, g > 0$ $3\lvert f\rvert - \lvert g\rvert < 0, g > 0$ $3f - \lvert g\rvert > 0, g > 0$
Monoclinic double-Q			
$\eta_1 = 0$, η_2, η_3	$\eta^2 = \frac{-\tilde{A} - f}{2B + C}$ $\cos^2\theta = 0$ $\cos^2\phi = \frac{1}{2}\left(1 + \frac{2B+C}{C}\frac{g}{\tilde{A} - f}\right)$	$\tilde{A} > 2\frac{3B+2C}{C}f$ $\tilde{A} < f - \frac{2B+C}{C}g$ $\tilde{A} < f + \frac{2B+C}{C}g$	$3f - \lvert g\rvert > 0$ $3f - \lvert g\rvert > 0, g < 0$ $3f - \lvert g\rvert > 0, g > 0$
$\eta_2 = 0$, η_1, η_3	$\eta^2 = \frac{-\tilde{A} - (f - g)/2}{2B + C}$ $\cos^2\theta = \frac{1}{2}\left(1 - \frac{2B+C}{2C}\frac{3f + g}{\tilde{A} + (f - g)/2}\right)$ $\cos^2\phi = 1$	$\tilde{A} > -\frac{3B+2C}{C}(f - g)$ $\tilde{A} < \frac{3B+C}{C}f + \frac{B+C}{C}g$ $\tilde{A} < -\frac{3B+2C}{C}f - \frac{B}{C}g$	$3f - \lvert g\rvert < 0, g > 0$ $3\lvert f\rvert - \lvert g\rvert < 0, g > 0$ $3f + \lvert g\rvert < 0, g > 0$
$\eta_3 = 0$, η_1, η_2	$\eta^2 = \frac{-\tilde{A} - (f + g)/2}{2B + C}$ $\cos^2\theta = \frac{1}{2}\left(1 - \frac{2B+C}{2C}\frac{3f - g}{\tilde{A} + (f + g)/2}\right)$ $\cos^2\phi = 0$	$\tilde{A} > -\frac{3B+2C}{C}(f + g)$ $\tilde{A} < \frac{3B+C}{C}f - \frac{B+C}{C}g$ $\tilde{A} < -\frac{3B+2C}{C}f + \frac{B}{C}g$	$3f - \lvert g\rvert < 0, g < 0$ $3\lvert f\rvert - \lvert g\rvert < 0, g < 0$ $3f + \lvert g\rvert < 0, g < 0$
Triclinic triple-Q			
η_1, η_2, η_3	$\eta^2 = \frac{-3\tilde{A}}{3B + 2C}$ $\cos^2\theta = \frac{1}{3}\left(1 - \frac{3B+2C}{C}\frac{f}{\tilde{A}}\right)$ $\cos^2\phi = \frac{1}{2}\left(1 + g\frac{3B + 2C}{C\tilde{A} + f(3B + 2C)}\right)$	$\tilde{A} < 2\frac{3B+2C}{C}f$ $\tilde{A} < -\frac{3B+2C}{C}(f - g)$ $\tilde{A} < -\frac{3B+2C}{C}(f + g)$	$3f - \lvert g\rvert > 0$ $3f - \lvert g\rvert < 0, g > 0$ $3f - \lvert g\rvert < 0, g < 0$

distinct phases are present, the disordered phase, three monoclinic single-Q phases, three monoclinic double-Q phases, and a triclinic triple-Q phase, as expected from the discussion at the end of Sec. II. The expressions in Table II in connection with the free energy (6) also reveal that the phases are separated by continuous phase transitions. The phase boundaries listed in the right column of Table II define two-dimensional surfaces in the f-g-$\tilde{A}$ parameter space which, with the aid of Eqs. (8)–(10), define surfaces in a space spanned by the temperature and the components of the physical fields. In Fig. 2 we illustrate the phase diagram in the H_x-H_y-$\tilde{A}$ space for applied magnetic fields in the basal plane and vanishing stress tensor. In Fig. 3 we present a cut through the graph of Fig. 2 at constant $H_y > 0$. This graph shows all eight distinct phases separated by lines of continuous phase transitions. As the temperature is lowered at constant H_x three transitions are expected: from the disordered state to a single-Q, then to a double-Q and finally to the triple-Q state. The phase sequences obey indeed the symmetry based rules established in Sec. II, cf. Eq. (5).

There are five points in the graph of Fig. 3 at which four critical lines meet and four phases coexist. These points are tetracritical points and lie on lines of such points, most easily described in terms of the variables $\tilde{A}$, f, and g. Tetracritical points where the disordered phase meets with two single-Q and one double-Q phase are given by

$$f = -\frac{1}{3}|g|, \quad \tilde{A} = -2f \quad \text{for } g \neq 0, \qquad \tilde{A} = f \quad \text{for } g = 0, \quad f > 0, \tag{12}$$

while tetracritical points where the triple-Q phase meets with two double-Q and a single-Q phase are given by

$$f = \tfrac{1}{3}|g|, \quad \tilde{A} = 2f\frac{3B+2C}{C} \quad \text{for } g \neq 0, \qquad \tilde{A} = -f\frac{3B+2C}{C} \quad \text{for } g = 0, f < 0. \tag{13}$$

When the value of H_y in Fig. 3 is lowered, the tetracritical points move closer to the origin and finally, at $H_x = H_y = 0$, the lines of tetracritical points meet, as do the phase transition surfaces in Fig. 2. At that point ($f = g = \tilde{A} = 0$) all eight distinct phases are in coexistence so that it becomes an "octacritical point."

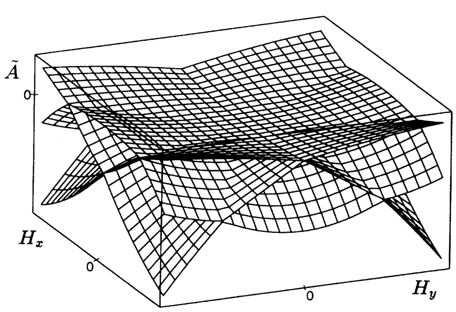

FIG. 2. Phase-transition surfaces in the space of magnetic-field components H_x and H_y and the temperature variable $\tilde{A}$ for applied magnetic fields in the basal plane in the absence of stress.

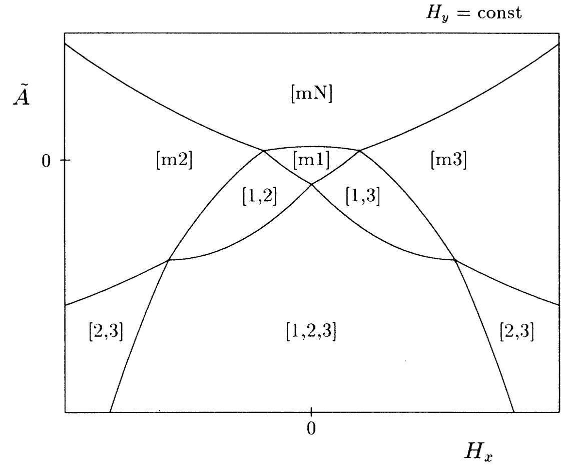

FIG. 3. Intercept of the surfaces in Fig. 2 with the plane H_y =const. The phases are named as in Table I.

Let us now turn to the "surfaces of special symmetry" in the f-g-$\tilde{A}$ parameter space, i.e., surfaces where one of the components of the order parameter is singled out, while the free energy is symmetric in the other two components. From Eq. (6) for the free energy we conclude that for $g = 0$ the component η_1 is singled out, while $f = \frac{1}{3}g$ and $f = -\frac{1}{3}g$, $g \neq 0$, single out η_3 and η_2, respectively. Thus, for $\tilde{A}$ = const, the surfaces of special symmetry can be described by the following straight lines in the f-g plane

$$g = 0, \qquad f = \pm\tfrac{1}{3}g, \quad g \neq 0. \tag{14}$$

We present the lines corresponding to the surfaces of special symmetry in Fig. 4, (a) in the f-g-plane, and (b) in the plane of magnetic fields H_x and H_y for magnetic fields applied in the basal plane and vanishing stress tensor.

In Fig. 5 we present the phase diagram in the $g = 0$ surface of special symmetry, which we compare with the phase diagram in a surface without special symmetry, namely the $f = 0$ surface, in Fig. 6. There are four phases in the surface of special symmetry, Fig. 5, separated by lines of continuous phase transitions that meet at a tetracritical point. When cooled at constant field f the system undergoes two phase transitions, namely from the disordered to either the single-Q or the double-Q phase and finally to the triple-Q phase. For the surface without special symmetry, cf. Fig. 6, six phases are separated by critical lines that meet at a "sexacritical" point, and the system undergoes three phase transitions upon cooling at constant field, namely from the disordered to a single-Q, then to a double-Q, and finally to the triple-Q phase. In the planes of special symmetry the ordered phases are of higher symmetry than in the general case, e.g., orthorhombic double-Q, [2=3], in Fig. 5 compared to monoclinic double-Q, [2,3], in Fig. 6. Since these higher-symmetry phases are described by evaluating the general results, cf. Table II, on surfaces of special symmetry there are no phase transitions between either orthorhombic and monoclinic single-Q phases, or orthorhombic and monoclinic double-Q phases or trigonal, monoclinic and triclinic triple-Q phases.

We note, finally, that the lines of tetracritical points given in Eqs. (12) and (13) lie on surfaces of special symmetry, cf. Eq. (14). A comparison of Eqs. (12) and (13) with Table II reveals that the lines of tetracritical points constitute two of the four critical lines in a surface of special symmetry, namely the lines separating the disordered from the double-Q, and the single-Q from the triple-Q phase. Furthermore, the octacritical point ($f = g = \tilde{A} = 0$) of the three-dimensional parameter space becomes the tetracritical point when we restrict ourselves to a surface of special symmetry as in Fig. 5.

IV. FLUCTUATION EFFECTS

The Landau theory employed in the preceding sections assumes the order parameter to be uniform throughout the system. Sufficiently far from a critical point this is a reasonable approximation since the fluctuations of the order parameter are small, but close to a critical point the fluctuations become large and can no longer be neglected. In this section we use results of renormalization-group and scaling theory to discuss the effects of critical fluctuations on our system.

To construct the Landau-Ginzburg-Wilson (LGW) Hamiltonian for our system we consider the second-order terms of spatial derivatives of the order parameter components η_i that are invariant under the operations of the symmetry group $P6_3/mmc$:

$$\sum_{i=1}^{3}\left[(\partial_x\eta_i)^2+(\partial_y\eta_i)^2\right],\quad \sum_{i=1}^{3}(\partial_z\eta_i)^2,$$
$$2\left[(\partial_x\eta_1)^2-(\partial_y\eta_1)^2\right] - \left[(\partial_x\eta_2)^2-(\partial_y\eta_2)^2\right]-\left[(\partial_x\eta_3)^2-(\partial_y\eta_3)^2\right] - 2\sqrt{3}\left[(\partial_x\eta_2)(\partial_y\eta_2)-(\partial_x\eta_3)(\partial_y\eta_3)\right]. \tag{15}$$

In principle, these terms appear in the LGW Hamiltonian with independent coefficients, i.e., the gradient terms are not isotropic. Since renormalization-group calculations show that this anisotropy does not affect the critical behavior of the system to first order in ϵ,[13,14] we include an isotropic gradient term in our LGW Hamiltonian $\mathcal{H}$, instead:

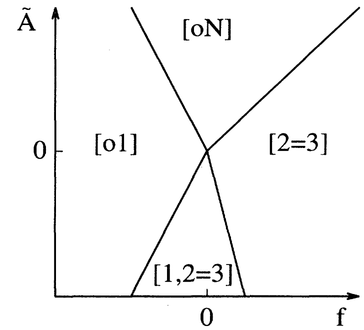

FIG. 5. Phase diagram on a surface of special symmetry. The solid lines indicate continuous transitions between the phases, named as in Table I.

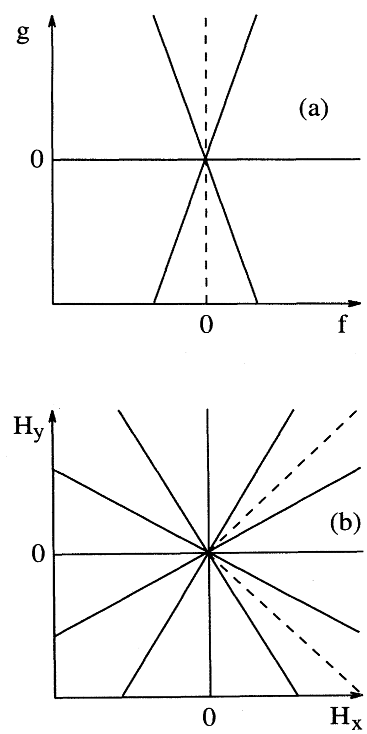

FIG. 4. Intercept of a plane $\tilde{A}$ = const with the surfaces of special symmetry: The solid lines represent Eq. (14) in the f-g plane (a), and in the plane of magnetic fields H_x and H_y for magnetic fields in the basal plane in the absence of stress (b). The dashed lines indicate the intercept with the surface without special symmetry of Fig. 6.

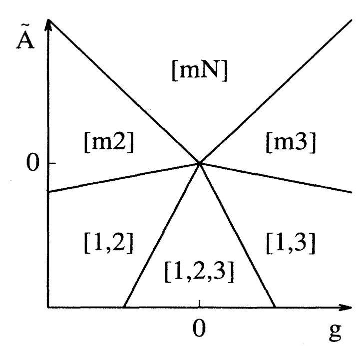

FIG. 6. Phase diagram on a surface without special symmetry. The solid lines indicate continuous transitions between the phases, named as in Table I.

$$\mathcal{H} = \int d^3r \left[\sum_{i=1}^{3} \left\{ \tilde{A}\eta_i^2 + (\nabla\eta_i)^2 \right\} + B\left(\sum_{i=1}^{3}\eta_i^2\right)^2 + C\sum_{i\neq j}\eta_i^2\eta_j^2 + 2f\eta_1^2 + (-f+g)\eta_2^2 + (-f-g)\eta_3^2 \right]. \quad (16)$$

This is a well-known Hamiltonian, see Ref. 15 and references therein, for a system with an $n = 3$ component order parameter. It exhibits cubic anisotropy for $C \neq 0$ and anisotropy in the components of the order parameter due to the symmetry-breaking fields f and g.

Renormalization-group calculations[16] show that the Hamiltonian (16) in the absence of symmetry-breaking fields has one stable fixed point, namely the isotropic or Heisenberg fixed point. This fixed point is attractive if the coefficients B and C are such that $B > 2C$ and $B > -C$.[17] A comparison with the stability condition $3B+2C > 0$ of the Landau theory reveals that for $C < 0$ a Hamiltonian with $-2C/3 < B < -C$ fulfills the Landau stability condition, but is not in the range of attraction of the stable fixed point. In this case the fluctuations drive the system into a region of thermodynamic instability and thus induce a first order phase transition where the Landau theory predicts a continuous one. Hence, in order to assure that the transition to the trigonal triple-Q state is continuous, we have to replace the stability condition $3B + 2C > 0$ by

$$B + C > 0, \quad (17)$$

where $C < 0$. Since this condition is independent of the number of components of the order parameter,[17] the other phase transitions will also be continuous as long as the external fields are sufficiently small. If the applied fields are very large they may change the fourth-order terms in the Hamiltonian and thus change the character of the transitions.[17]

Bruce and Aharony[15] investigated the Hamiltonian (16) for symmetry-breaking fields on a surface of special symmetry, $g = 0$. Their results show that critical fluctuations do not alter the phase diagram Fig. 5 drastically. The critical lines that separate the disordered ([oN]) and the monoclinic triple-Q ([1,2=3]) phase from the partially ordered phases ([o1] and [2=3]) no longer meet at a finite angle at the tetracritical point, but approach each other and the $f = 0$ axis tangentially, where the new critical lines correspond to lower values of the temperature-variable $\tilde{A}$ than the original ones. The shape of the critical lines in the vicinity of the tetracritical point $T = T_c$, $f = 0$ can be expressed in terms of power laws in the external fields

$$t_1 = [T_1(f) - T_c]/T_c \sim |f|^{1/\psi_1}, \quad (18)$$

$$t_2 = [T_2(f) - T_c]/T_c \sim |f|^{1/\psi_2}, \quad (19)$$

where $T_1(f)$ indicates the transition temperature from the disordered phase [oN] to a partially ordered phase, while $T_2(f)$ is the transition temperature from a partially ordered phase to the monoclinic triple-Q phase. The so-called shift exponent $\psi_1 = \phi_f \approx 1.25$ (Ref. 18) is associated with the crossover from the Heisenberg, $n = 3$, critical behavior in the absence of external fields, to the $n = 1$ ($f < 0$) and $n = 2$ ($f > 0$) critical behavior in the presence of the symmetry-breaking field. The critical exponent for the lower phase-transition lines is given by $\psi_2 = \phi_f - \phi_C$, where the exponent ϕ_C is associated with the cubic anisotropy crossover and quite small and negative.[15]

In summary, critical fluctuations affect the condition for the phase transitions to be continuous, cf. Eq. (17), and the shape of the phase transition surfaces in the vicinity of the tetracritical lines and the octacritical point. Furthermore, the values of the critical exponents predicted by the renormalization-group theory differ from those calculated from a Landau model.

V. RELATION TO EXPERIMENT

In this section we discuss experimental tests of predictions from our model for the quadrupolar phases of UPd_3. We start by summarizing some of the main features of the phase diagram which are independent of the as yet undetermined values of the system dependent parameters of the model. We expect the system to pass through the octacritical point when cooled in the absence of symmetry-breaking fields. This corresponds to one phase transition, namely from the disordered to the trigonal triple-Q phase. For symmetry-breaking fields that belong to a surface of special symmetry, cf. Eq. (14) and Fig. 4, two phase transitions are expected upon cooling at constant applied fields: from the disordered to a partially ordered phase, orthorhombic single-Q or double-Q, and then to the monoclinic triple-Q phase, cf. Fig. 5. For fields in the basal plane that do not belong to a surface of special symmetry, finally, three phase transitions are expected: from the disordered to a monoclinic single-Q, from there to a monoclinic double-Q, and then to the triclinic triple-Q phase, cf. Fig. 6.

External fields that break the symmetry of the components of the quadrupolar order parameter can be realized with the aid of applied stress and magnetic fields, cf. Eqs. (9) and (10). In the absence of magnetic fields, for example, the system is confined to a surface of special symmetry when a stress is applied in the basal plane so that $\sigma_{xx} - \sigma_{yy} \neq 0$ but $\sigma_{xy} = 0$. Similarly, magnetic fields along the directions indicated in Fig. 4 (b) correspond to surfaces of special symmetry even in the presence of stress as long as $\sigma_{xy} = 0$. Thus experiments performed under controlled stress and magnetic fields can be used to explore the phase diagrams Figs. 2 and 3, and Figs. 5 and 6.

There are several experimental techniques that can be used to detect these phase transitions but elastic neutron scattering seems to be particularly suitable. First of all, neutron scattering allows us to distinguish between single-Q and multiple (double or triple)-Q phases[7] and, secondly, the use of spin-polarized neutrons makes it possible to detect the onset of magnetic ordering, which is expected at temperatures somewhat below T_1 and whose effects might otherwise be confused with those due to quadrupolar ordering. Neutron-scattering experiments[3,5] have been performed in the absence of external fields and led to the conclusion that the ordered phase is trigonal triple-Q.[7]

In thermal-expansion experiments strains $e_{ij} = -(\partial G/\partial \sigma_{ij})$, $i,j \in \{x,y,z\}$ are measured. Within our Landau model for UPd_3, the strains are related to linear combinations of squares of the components η_i of the order parameter,[9] so that a continuous phase transition appears as a change in slope of the thermal-expansion as a function of temperature. The same, by the way, is true for the temperature dependence of the magnetization of a sample. Zochowski and McEwen[6] performed thermal-expansion measurements in a stressed sample[9] of UPd_3 with and without applied magnetic fields along the x and y axis, i.e., the applied fields correspond to a surface of special symmetry, cf. Fig. 5. Their results which we discussed in detail in our previous work[9] confirm that there are two phase transition upon cooling the sample. Furthermore, the results indicate that the effect of stress is much more pronounced than that of applied magnetic fields. In contrast to our predictions, however, the value of the thermal expansion seems to jump rather than to change continuously at the lower temperature phase transition (to the triple-Q ordered phase). Higher-order terms in the Landau free energy, cf. Sec. II, or fluctuations together with large external fields, cf. Sec. IV, could account for a first-order phase transition. More information about the experimental phase diagram is required, though, before extensions of our model can be investigated. It is also possible that the observed discontinuity is due to a small misalignment of the sample. This would in effect remove the system from a surface of special symmetry, in which case we expect three phase transitions. For a small misalignment two of the transitions would be spaced closely enough to appear as a discontinuity in the curve.

Predictions from the renormalization-group theory concern mostly the asymptotic critical behavior near the surfaces of phase transitions. Due to the fact that the system exhibits multicritical behavior we expect a variety of crossover phenomena to occur. With properly chosen applied fields transitions from the disordered phase to the single-Q, double-Q and triple-Q phase can be observed and are expected to be Ising like ($n = 1$), XY-like ($n = 2$), and Heisenberg like ($n = 3$), respectively. Furthermore, when the octacritical point is approached from below, there is a crossover due to the effects of the cubic anisotropy. Careful measurements of the critical lines for fields in special directions can be used to determine the shift exponents $\psi_1 \approx 1.25$ and ψ_2, defined in Eq. (18) and (19), and thus the crossover exponents ϕ_f and ψ_C. Furthermore, generalized scaling[18] predicts that the strains e_{xx} and e_{yy} obey an asymptotic power law of the form $e_{ii} \sim |T - T_0|^{\beta_f}$ with $\beta_f = 2 - \alpha - \phi_f \approx 0.85$ when the system goes through the octacritical point in the absence of symmetry-breaking fields. Thus, the relevant crossover exponents are in principle accessible, but it will be very difficult to decide in practice whether the asymptotic critical region has indeed been reached, since the Landau values for these exponents, $\psi_1 = \psi_2 = 1$ and $\beta_f = 1$, are quite close to the ones predicted by renormalization-group theory.

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* Present address: Institut für theoretische Physik, Humboldt Universität, Invalidenstr. 110, 10099 Berlin, Germany.

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