Field-induced transition in UPdSn at 3 T

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A single crystal of the intermetallic compound UPdSn has been studied by means of neutron diffraction on a two-axis spectrometer in magnetic fields up to 7 T and at temperatures between 6 and 46 K. The uranium moments have previously been shown to exhibit two antiferromagnetic phase transitions at approximately 25 and 40 K. The c axis is known to be the hard magnetic axis, and if the field is applied in the hexagonal basal plane, the magnetization measurements show a low-temperature spin-flop transition at approximately 3 T. In the present measurements, the field was applied along the [100] direction of the orthorhombic magnetic unit cell. We have studied the temperature dependence of the (010) antiferromagnetic reflection, which corresponds to the order parameter of the 25-K transition, at fixed fields; the ground state is stable for fields below 3.5 T and temperatures below 25 K, and we have established the magnetic phase boundaries in this regime. We also made a series of measurements in which the sample was zero-field cooled from high temperature and then the field was ramped up and down at fixed temperature. In this configuration, we observe significant irreversible domain repopulation effects below the spin-flop transition as the field is increased, but the curves obtained on reducing the field from above the spin-flop transition seem to be indicative of intrinsic single-domain properties; they are consistent with the field-cooled data. We have also studied the induced (ferromagnetic) moment, and have found that the behavior is very different above and below the 25-K transition. For $T<25$ K, there is a sharp transition at approximately 3 T in agreement with bulk magnetization measurements, while above 25 K, the magnetic intensity rises parabolically with fields as would be expected for a simple antiferromagnet; the induced magnetic moment in this case arises from the transverse magnetic susceptibility.

I. INTRODUCTION

The hexagonal intermetallic compound UPdSn was shown in the survey of Palstra *et al.*[1] to order antiferromagnetically at low temperatures. Subsequent magnetization and neutron powder-diffraction experiments[2] revealed that there are two magnetic transitions at approximately 25 and 40 K, respectively. The low-temperature magnetic structure (phase II) was shown to be as in Fig. 1, with a uranium moment of $2.05\mu_B$ and canting angles of $\theta=45°$ and $\phi=54°$. It was originally thought that $\theta=0°$, when $25<T<40$, and this second intermediate phase was named phase I. However, a careful study of the temperature dependence[3] showed that there is negligible temperature hysteresis and that θ appeared to vary continuously, to the point that there was no clear evidence for the 25K transition in the neutron powder data. However, μ_x, which would be the order parameter for the 25-K transition, does drop precipitously at 25 K and seems to be correlated with the lower feature in the magnetization, as well as a shoulder seen at 25 K in the specific-heat data of Yuen[4] and Nakotte.[5] Indeed, we will show in this article that in our single crystal μ_x drops quickly, and that the 25-K transition does indeed seem to be associated with the ordering of the x component of the moment. Hence in this article, we will continue to refer to three magnetic phases: paramagnetic when $T>40$ K, phase I for 25 K $<T<40$ K, and phase II when $T<25$ K. In other words, we assume that the *ideal* phase I has moments confined to the orthorhombic b-c plane, $\theta=0$ and the 010 reflection would then be absent. While the data reported in Ref. 3 and in this work indicate that this is not absolutely true, this approximation provides a good conceptual framework, within which to understand the field-dependent phenomena in UPdSn.

In addition to the antiferromagnetic order, de Boer *et al.*[6] have reported a spin-flop transition at approximately 3 T, when the magnetic field is applied within the hexagonal basal plane. These high-field single-crystal magnetization measurements were made at 4.2 K only,

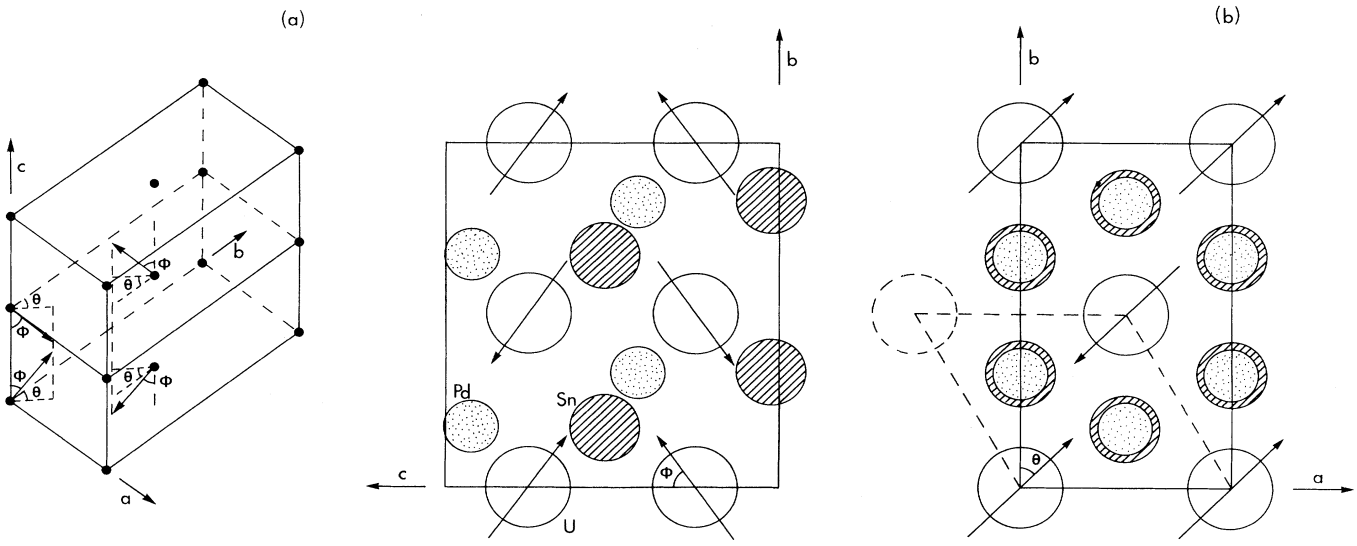

FIG. 1. The magnetic structure of UPdSn at low temperature (magnetic space group P_c2_1), (a) in perspective, and (b) by projection on to the *b-c* and *a-b* planes, respectively. The cell shown is the orthorhombic magnetic unit cell, with two uranium layers per unit cell. In (a), for the sake of clarity, only the uranium atoms are shown and moments are drawn on only the basis of four atoms. Note that this structure exhibits *c*-side anticentering. The canting angles θ and ϕ are spherical polar angles defined with respect to the *c* axis, but with θ and ϕ reversed with respect to the normal convention. Note that in (b), ϕ is the full three-dimensional angle between the moment and the *c* axis, while θ is the angle on the two-dimensional projection. At low temperature (magnetic phase II of Ref. 2), $\phi=54°$ and $\theta=45°$. In magnetic phase I (40 K $>T>$ 25 K), $\theta=0°$ but ϕ retains the value of 54°.

and they found that the magnetization was still not completely saturated at a field of 35 T. Indeed, the hexagonal plane is an easy plane: the *c*-axis susceptibility is much lower. The primary purpose of the present work is to study this field-induced transition, and all of our data have been collected with B in the basal plane. However, if the field is applied along the *c* axis, rather than in-plane, there is evidence of a transition at approximately 13 T, but this transition is out of the range of our present experimental capabilities.

Crystallographically, UPdSn was originally thought to exhibit the $CaIn_2$ structure type (space group $P6_3/mmc$) in which the Pd and Sn atoms are disordered. However, in our previous work[2] we showed that it exhibits the related GaGeLi structure type (space group $P6_3mc$), which allows Pd/Sn ordering. This has subsequently been confirmed by both single-crystal x ray[6] and single-crystal neutron[7] studies. For the present work, it is sufficient to note that the uranium atoms lie on a simple hexagonal lattice, with two uranium layers per crystallographic unit cell. It is rather bizarre that, while the magnetic structures obey a correct magnetic-space-group hierarchy,[8] the anisotropy is not easily explained in any local-moment picture, particularly close to the Néel point where only second-order terms in the Hamiltonian contribute significantly.

In a single-crystal study like the present one, there will in general be six different types of magnetic domain possible. They can be divided into three mirror-image pairs: the three possibilities come from the three choices of hexagonal [100] axis to become the unique orthorhombic [010] axis of the magnetic unit cell. This is illustrated in Fig. 2, which shows all three pairs of domains in both real and reciprocal space. In a single-crystal neutron diffraction study[7] on the SCD diffractometer (essentially a time-of-flight neutron Laue camera) at Los Alamos, we have shown that all three pairs of domains are populated almost equally. This turns out to be quite important in the analysis of the present experiment.

Finally, as an aside, it should be noted that the electronic-specific-heat coefficient γ is remarkably small for a uranium intermetallic[6] and that photoemission studies indicate that the uranium moment in UPdSn is fairly localized: nevertheless there is still some *f*-electron density at the Fermi level.[9] Given these observations and the fact that the uranium moment ($2.05\mu_B$) is fairly large, one might be led to believe that a local-moment approach is appropriate for this material.

II. EXPERIMENTAL METHOD

The crystal is the same one as used previously in the zero-field single-crystal diffraction study at Los Alamos.[7] It was grown by the triarc Czochralski method, and has a cylindrical form with a diameter of approximately 1 mm and a length of 5 mm. The cylinder axis was approximately 18° away from the crystallographic $[110]_{\text{hexagonal}}(=[100]_{\text{orthorhombic}})$ axis. For this experiment, the crystal was mounted within an aluminium block, with a 1-mm hole drilled at 18° to its axis, so that the $[110]_{\text{hexagonal}}$ axis was vertical. The crystal was glued to the aluminium block at its lower end to prohibit move-

ment. This block was shielded with cadmium, sealed in a helium exchange gas and mounted within a variable-temperature helium cryostat. This in turn was inserted within a 7-T vertical-field split-pair superconducting magnet, which was mounted on the BT-9 triple axis spectrometer at the NIST Research Reactor. The spectrometer was used in the two-axis mode with a vertically focusing pyrolytic graphite monochromator and graphite filter. The wavelength used was 2.3505 Å. The analyzer had been removed and the collimation was 40′/30′/40′.

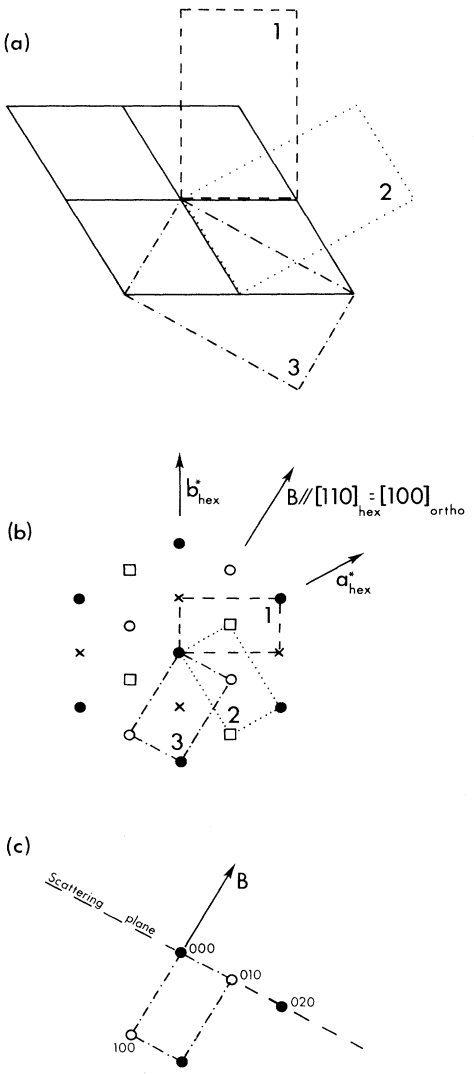

FIG. 2. The real-space and reciprocal-space lattices for UPdSn. (a) shows the hexagonal basal plane of UPdSn (four crystallographic unit cells are shown), while three possible orthorhombic magnetic unit cells are shown by solid lines: (1), (2), and (3), respectively. These rectangles correspond to the *a-b* plane of Fig. 1 and there are three different possible orientations for this cell in the original hexagonal lattice. (b) shows the crystallographic reciprocal lattice (full circles) and the three magnetic reciprocal lattices, one for each domain pair: domain (1) is shown with a dashed line and the magnetic reciprocal lattice points are crosses. (2) is represented by the dotted line and the open squares, while (3) is a dot-dashed line and open circles. The direction of applied magnetic field is also shown. (c) shows the particular domain that was observed in our experiment (3), along with the reciprocal lattice points lying within the experimental scattering plane, the laboratory horizontal plane.

The experiment was set up to ensure that the applied magnetic field lies in the hexagonal basal plane—in fact, it is parallel to the $[100]_{orthorhombic}$ axis of one of the magnetic domain pairs. Furthermore, in addition to the hexagonal c axis which is of minor interest as regards magnetic scattering, the scattering plane contains the $[010]_{orthorhombic}$ axis, and therefore the (010) and (020) reflections for the same domain. The reflections for all the other domains lie out of the horizontal plane and were not accessible in this experiment. Thus we are in a position to monitor the variation of the antiferromagnetic x component of the magnetic moment μ_x^{AF}, which is the order parameter for the 25-K magnetic phase transition, as a function of temperature and magnetic field, by measuring the variation of intensity of the (010) reflection. Likewise, we can monitor the ferromagnetic x component of the magnetic moment μ_x^F by measuring the variation in intensity of the (020) reflection. This information should be the same as that measured in bulk magnetization or susceptibility experiments.

III. RESULTS

A. Temperature scans at zero field

First, we observed the (010) reflection as a function of temperature in zero field, on both cooling and heating. The results for cooling are shown in Fig. 3 and the intensities are directly comparable with the powder results in Figs. 2 and 4(a) of Ref. 3. As in the powder work, there is no discernible hysteresis but the drop in intensity is much sharper in the single crystal. This could be indicative of a real transition at 25 K and the thermal broadening of the transition in the powder sample may be due to poorer sample quality and/or a slightly different stoichiometry. Note, however, that there is still significant intensity in the single crystal all the way up to 40 K. Note also that the peak widths shown in Fig. 3(b) also increase markedly at 25 K. This width is transverse to the reciprocal lattice vector and cannot be observed in a powder measurement. The extra width corresponds to a correlation length of 560 Å, parallel to the c axis, and it seems that there are remnants of phase II with this size above the 25-K transition.

B. Ferromagnetism as a function of field

As stated in Sec. II, we can monitor the ferromagnetic moment by observing the intensity of the (020) reflection. This was done at both 6 and 30 K, below and above the 25-K transition, as shown in Fig. 4. The behaviors are very different. We note that there is a large contribution at zero field from the (020) nuclear Bragg reflection; any magnetic intensity will be added on to this nuclear peak, with the magnetic intensity proportional to the square of the magnetization. Figure 4 shows that there is a sharp increase in the intensity at 3 T in the 6-K data. This is in excellent agreement with the magnetization data taken at

4.2 K by de Boer *et al.*[6] Above 25 K, however, the picture is very different: the magnetic scattering rises almost parabolically with field, which means that the magnetization rises linearly with field and this is exactly what one would expect for the transverse magnetic susceptibility in a simple antiferromagnet. Of course, UPdSn is not a simple antiferromagnet, and there are three domain pairs, but as we show below, the physics can still be understood in this simple way. Note that at 30 K there is no evidence for any phase transition between 0 and 7 T. Note also that while the 30-K data are completely reversible, there is some apparent hysteresis in the 6-K data.

While the 6- and 30-K data are qualitatively different

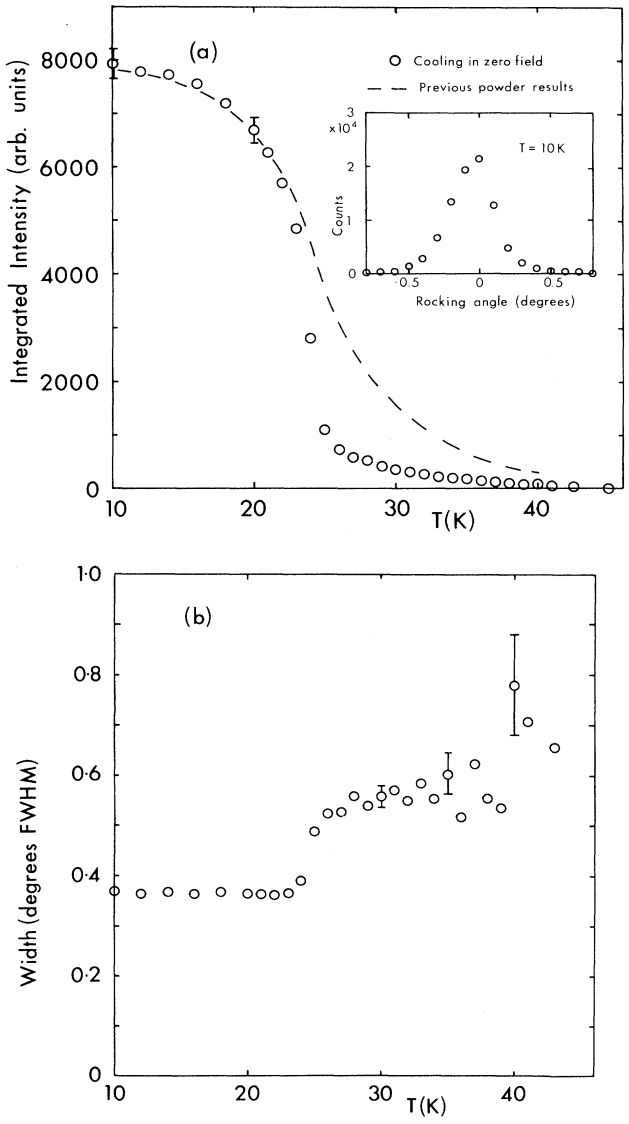

FIG. 3. Variation of the (010) magnetic peak with temperature. $B=0$ T. (a) shows the integrated intensity on cooling, while (b) shows the observed width of the rocking curve. For purposes of comparison, our previous powder results (background subtracted and appropriately scaled) from Ref. 3 are also shown as a dashed line in (a). The inset to (a) shows the rocking curve at 10 K.

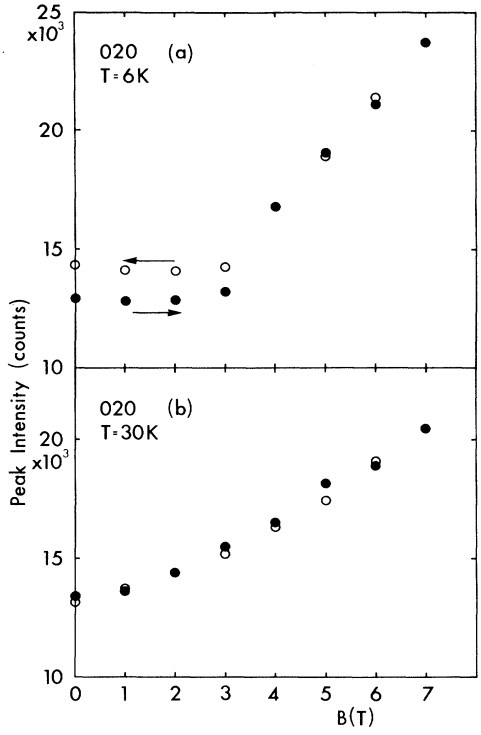

FIG. 4. Variation of the peak intensity of the (020) "nuclear" peak with applied field, (a) at $T=6$ K and (b) at $T=30$ K. In both cases, the crystal was initially cooled from the paramagnetic state (46 K) in zero field. Increasing-field data are indicated by filled circles and decreasing-field data by open circles.

in their temperature dependence, the absolute values of the magnetic scattering at 7 T are rather similar, when one corrects for the temperature variation of the total moment.[3] From the previous powder work,[3] the uranium moment at 30 K is reduced to approximately 75% of its low-temperature value, and μ^2 is therefore reduced to approximately 56% of its low-temperature value, while we observe in this experiment that $(\mu_x^F)^2$ is reduced to 66% of its low-temperature value on warming to 30 K. Given that the samples in the two studies are completely different, one being a powder and the other a single crystal, and that the moment varies quite strongly with temperature in this range, this is excellent agreement. It leads us to suspect that the moment system is in a very similar state in a field of 7 T, at all temperatures below ~40 K.

C. Field scans after cooling in zero field

We made a series of field scans at fixed temperature. In each case the sample was heated to 46 K, cooled in zero field to the required temperature, the field was increased in steps to 7 T, and finally it was reduced in steps back to zero field. We made such measurements at 6, 10, 20, 24, and 30 K and the results are shown in Fig. 5. The results at 6, 10, and 20 K are qualitatively similar with obvious hysteresis effects. There is still some hysteresis at 24 K, which is just below the transition (see Fig. 3). The behav-

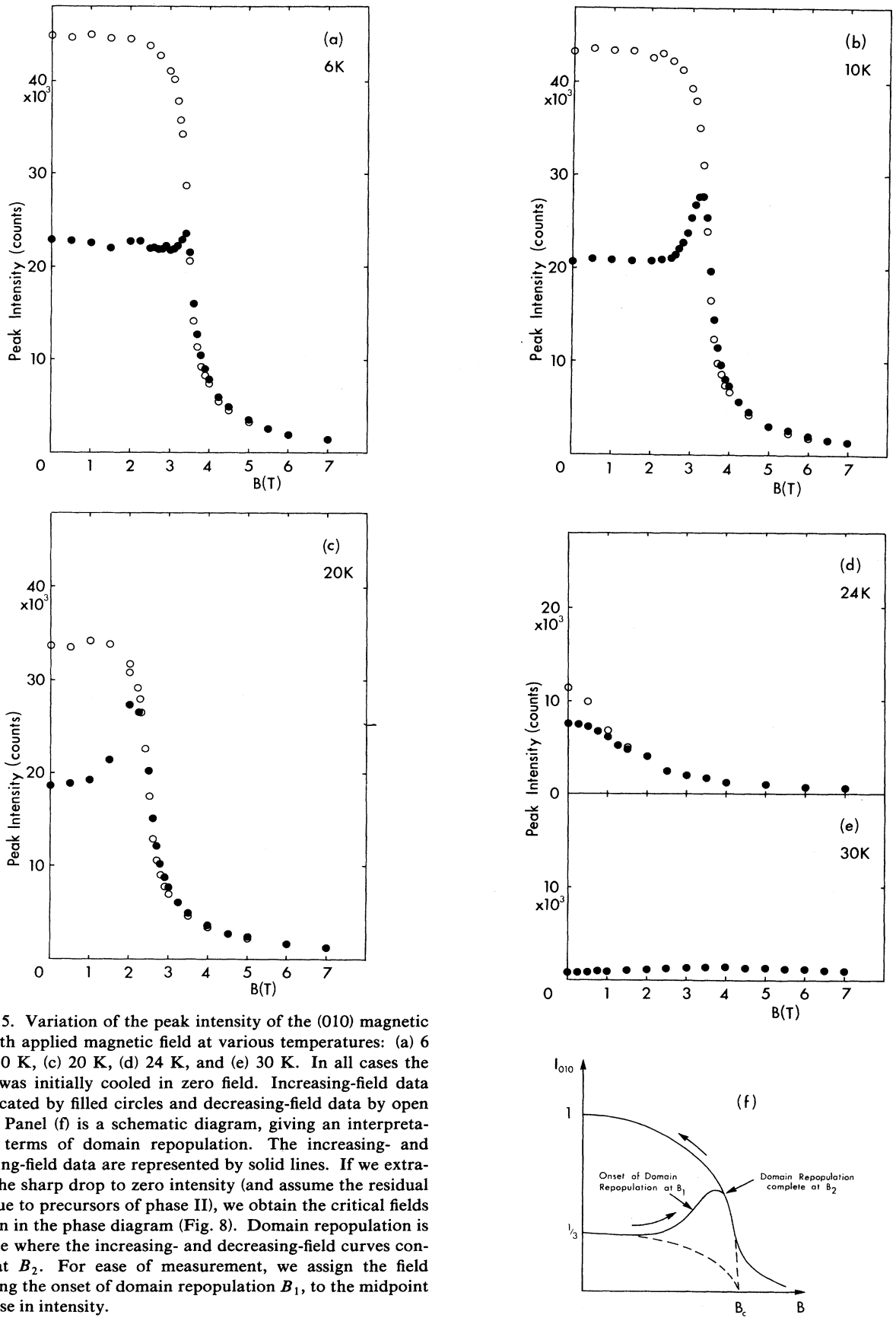

FIG. 5. Variation of the peak intensity of the (010) magnetic peak with applied magnetic field at various temperatures: (a) 6 K, (b) 10 K, (c) 20 K, (d) 24 K, and (e) 30 K. In all cases the crystal was initially cooled in zero field. Increasing-field data are indicated by filled circles and decreasing-field data by open circles. Panel (f) is a schematic diagram, giving an interpretation in terms of domain repopulation. The increasing- and decreasing-field data are represented by solid lines. If we extrapolate the sharp drop to zero intensity (and assume the residual tail is due to precursors of phase II), we obtain the critical fields B_c, given in the phase diagram (Fig. 8). Domain repopulation is complete where the increasing- and decreasing-field curves converge, at B_2. For ease of measurement, we assign the field describing the onset of domain repopulation B_1, to the midpoint of the rise in intensity.

ior at 30 K, above the phase-II boundary, shows no hysteresis. At low temperatures, the peak intensity is initially nearly field independent and then rises rapidly with field before falling precipitously. On reducing the field, the intensity rises monotonically to reach a saturated value at low field. We interpret the difference between the increasing- and decreasing-field curves as being due to domain repopulation effects. In other words, the sharp rise in (010) peak intensity is due to growing that pair of domains at the expense of the others. On the other hand, the intensity then falls rapidly as the phase-II state is destroyed and a state with a ferromagnetic component sets in to replace it. On reducing the field the domain repopulation is frozen in, giving an enhanced signal from the one big domain, rather than one out of three. We would therefore conclude that the data taken on reducing the field is representative of the intrinsic monodomain physics and we can identify the middle of the sharp rise with the phase transition. Indeed, the maximum transition field observed (at 6 K) is 3.5 T. As the temperature is increased, the transition field drops. This behavior is discussed further in Sec. III E below. At 24 K, phase II barely exists and it is as if we are sitting right on the sharp rise. At 30 K, phase II does not exist at all and the behavior is reversible. One would expect the ratio of the repopulated intensity to the random-population intensity to be 3:1, but it is observed to be only slightly greater than 2 times larger.

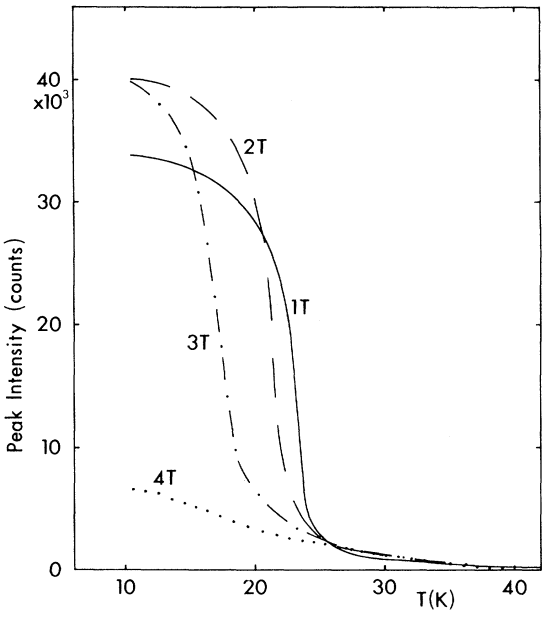

FIG. 6. Variation of the peak intensity of the (010) magnetic peak with temperature at various fixed fields: 1, 2, 3, and 4 T. We show only the cooling data: the heating data are almost indistinguishable on the scale of this figure.

D. Temperature scans in field-cooled state

A series of scans of the (010) intensity, as a function of temperature at fixed field, were also performed. In each case, we took the sample up to 46 K, above the Néel point, and applied a magnetic field. The temperature was lowered in steps to 10 K and then raised in steps back up to 46 K. This procedure was followed in fields of 0 (See Sec. III A and Fig. 3), 1, 2, 2.5, 2.75, 3, 4, and 5 T. Figure 6 shows these results for fields between 1 and 4 T—the results at 5 T are qualitatively similar to those at 4 T and are not displayed here for purposes of brevity. There is no significant hysteresis in any of the curves presented here, but there is a sharp change in intensity in the vicinity of 20 K at all fields below 4 T. Again we identify this sharp change with the transition between phases I and II. The transition temperature is a maximum (25 K) at zero field, but the saturated intensity is greatest at 2.5 T. In fact we plot the field-cooled saturated intensities in Fig. 7, along with the zero-field-cooled random-domain-population intensities and the majority-domain intensities. Clearly, the results are consistent with each other, with field cooling in a field of at least 2.5 T being necessary to produce a majority-domain sample. On the other hand, a majority-domain sample is most effectively produced by ramping the field up to 7 T and back down again, at low temperature.

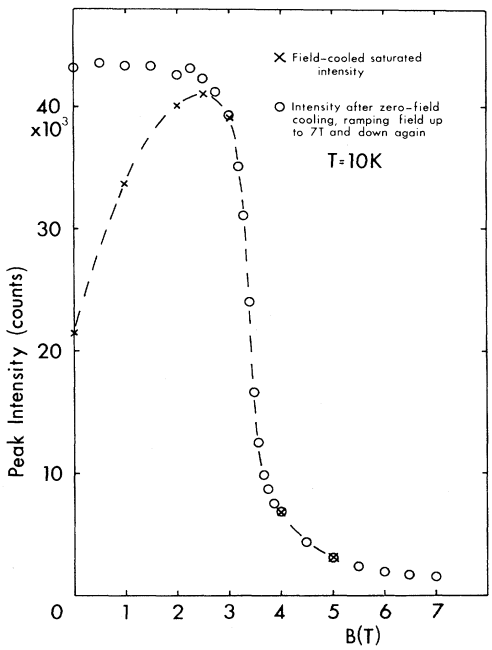

FIG. 7. The variation of field-cooled saturated intensity [for the (010) magnetic reflection] with applied field. This is simply the low-temperature intercept of each of the graphs shown in Fig. 6. For purposes of comparison, the zero-field-cooled intensity, from Fig. 5(b), is also plotted as a set of open circles, while the intensity after ramping the field up and down, from Fig. 6, is shown by the crosses and the dashed line.

E. Phase diagram

In Fig. 8, we compile both the field- and temperature-scan data in a field-temperature phase diagram. Clearly, the boundary of stability for phase II is well defined by

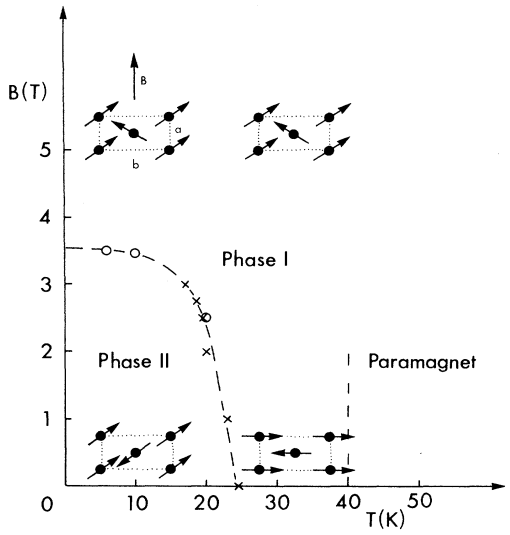

FIG. 8. The magnetic phase diagram of UPdSn, as extracted from the data in Figs. 5 and 6. The dashed line represents the limit of stability of magnetic phase II and is merely a guide to the eye. The open circles represent field scans at fixed temperature and the crosses represent temperature scans at fixed magnetic field.

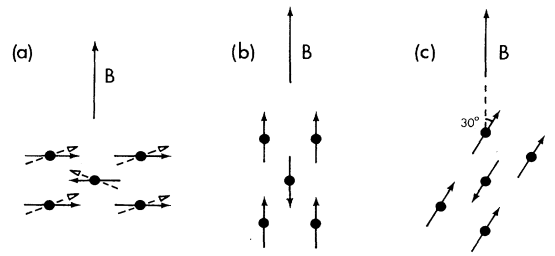

FIG. 9. Schematic diagram of the field conditions required to grow domain (3) at the expense of the domains (1) and (2). (a) shows the projection of the *a-b* plane of phase I, for domain (3), which is in the "transverse" geometry. This moment arrangement is the same as the projection shown in the right-hand part of Fig. 1(b). (b) shows the "parallel" geometry and (c) shows the intermediate configuration corresponding to both domain (1) and (2).

our data. While we have very little information about the nature of the states outside the region of phase II stability (i.e., $B > 3.5$ T or $T > 25$ K), it is plausible that the system has phase-I–type antiferromagnetic order with the magnetic field inducing a small transverse ferromagnetic moment as shown in Fig. 8. This interpretation explains both the appearance of a ferromagnetic component at the phase boundary and the simultaneous disappearance of the (010) antiferromagnetic reflection. The consequences of this model are discussed below.

IV. DISCUSSION

In the rudimentary interpretation of our data given above, we have made some assumptions. First, we assume that the magnetic structure reported previously[2] is indeed the correct structure. Second, we assume that the hysteretic effects in Fig. 5 are due to domain repopulation and that the domain pair to which we are sensitive, (3) in Fig. 2(a), grows at the expense of the other two pairs. In fact if a field greater than 2.5 T is applied in phase I, it appears that we produce a majority-domain sample on subsequently lowering the temperature. This is quite plausible as it is well known[10] that for Heisenberg antiferromagnets, of the two possible configurations shown in Figs. 9(a) and 9(b), the transverse configuration corresponding to domain (3) is energetically favorable because the moments can be canted slightly as shown. Now, domains (1) and (2) correspond to Fig. 9(c), rather than 9(b), but they are still energetically unfavorable compared with domain pair (3). This argument is essentially the same as that showing that the transverse susceptibility of a Heisenberg antiferromagnet is much greater than its longitudinal susceptibility.[10] Indeed, that is probably why we observe the nice parabolic behavior in Fig. 4(b), which is derived from the induced moment due to this transverse magnetic susceptibility.

While it is plausible that domain pair (3) grows at the expense of the others, there is one serious problem with this interpretation: in zero field, one would expect that one third of the crystal is in each of the three domain pairs. If, by applying a field, one produces a monodomain sample, the intensity should increase by a factor of 3. However, the data presented in Fig. 5 give ratios of approximately a factor of 2. Part of this reduction may be due to the effects of extinction, but based on the extinction calculation for the same crystal on a single-crystal Laue diffractometer,[7] we believe the maximum possible effect to be a reduction in intensity by about 18%: this is insufficient to account for the observed discrepancy. However, there is another possible explanation: the factor of 2 is quite striking, because it would imply that μ_x^{AF} would have to increase by a factor of $\sqrt{2}$ in the single domain. There is a very simple way to achieve this: because $\theta = 45°$, μ_x^{AF} would increase by this factor if the moments rotated to lie in the *a-c* plane and $\theta = 90°$. This rotation would have to be irreversible and accompanied by no domain repopulation. A consequence of this realignment would be that the (100) magnetic reflection would disappear completely. Unfortunately, this reflection is not accessible in the present experimental configuration. What one really needs is a horizontal field in which one could study a reciprocal lattice vector, (1,0,0) in this case, which is parallel to the applied field. Even so, there is another problem with this reorientation interpretation: it is energetically unfavorable for exactly the same reason as given in the previous paragraph. We therefore prefer our original interpretation, but believe the crystal is not completely monodomain. The ratio of $\frac{2}{3}$ is then due in part to extinction, but also to incomplete repopulation of the domains.

While the (010) reflection is proportional to the square of the order parameter (μ_x^{AF}) for the magnetic transition at 25 K, and describes phase II, the (100) reflection is proportional to the square of the order parameter (μ_y^{AF})

for the transition at 40 K and describes phase I. This is because the angle ϕ is almost invariant with temperature.[2,3] It would therefore be highly desirable to study this reflection, with field applied parallel to [100]. This can only be done with a horizontal-field apparatus capable of reaching 3 T and more. Such an experiment, in addition to clarifying the question raised in the previous paragraph, would allow us to study the 40-K transition as a function of field and determine more of the phase diagram outside the region of phase-II stability. For instance, do we still have the phase-I component above 3.5 T? Some information can clearly be gained by studying the bulk magnetization as a function of field at various temperatures above 4.2 K, and also as a function of temperature at various fixed fields between 0 and 7 T. But it would be invaluable if the other antiferromagnetic order parameter (μ_y^{AF}) could be studied by means of neutron scattering.

A further objection to our analysis might be that our original structure is incorrect and that the magnetic structure is some kind of triple-q structure. First, we note that our structure is not in fact a single-q structure in the first place: it is modulated along all three of the x-, y-, and z- axes. Presumably any multiple-q structure would have to be similarly complicated. Furthermore, the structure is clearly commensurate with the parent crystallographic structure and therefore, if our orthorhombic cell is incorrect, can be indexed in a quadruple-sized hexagonal cell with $a'=2a$ and $c'=c$. In fact, in our original analysis, we considered all such possibilities allowed by a magnetic space-group analysis like that of Prandl,[8] and the space groups were either incorrect in failing to give the observed systematic absences or gave poor fits to the low-temperature structure. In fact many of them gave zero moment on the central uranium site, with a finite moment on the other sites. It is rather striking that we see exactly the right systematic absences for the side-anticentered orthorhombic cell that we have used. So there is a certain economy in the structure we assume. And finally, any multiple-q structure must account for the two phase transitions that are observed in specific heat, magnetization/susceptibility, and neutron-scattering measurements. This is particularly elegant

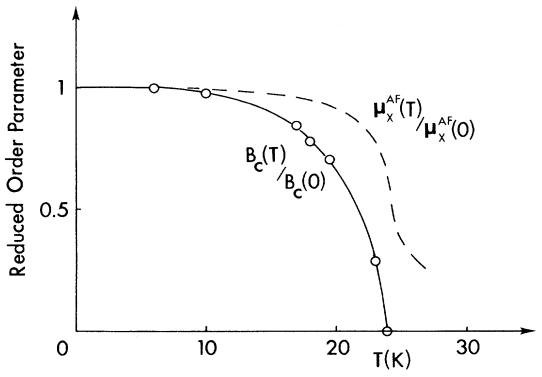

FIG. 10. Variation of reduced critical magnetic field $B_c(T)/B_c(0)$ and reduced x component of the uranium moment $\mu_x^{AF}(T)/\mu_x^{AF}(0)$ with temperature.

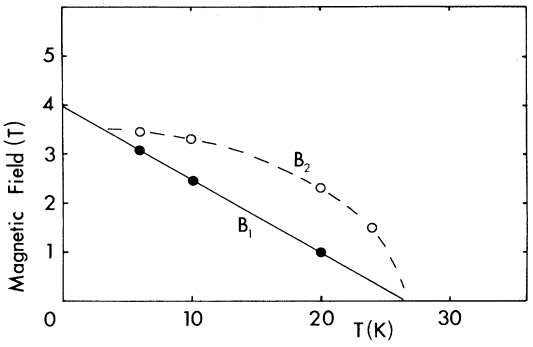

FIG. 11. The variation of critical magnetic fields B_1, necessary to initiated domain repopulation, and B_2, to complete domain repopulation, with temperature. These fields were extracted from the data shown in Fig. 5, and follow the interpretation shown schematically in Fig. 5(f).

with our structure. Of course, the definitive test of our model of domain repopulation is to study the other domains, with the same field configuration. Domain pairs (1) and (2) should decrease in intensity as domain pair (3) grows at their expense. This would involve looking at out-of-plane reflections (see Fig. 2) in similar fields, something that was not possible with the present experimental configuration.

Another consideration is whether only second-order terms, of the type $\mu_i\mu_j$, where i and j signify Cartesian components of the uranium moment, contribute to the free energy. These would include terms like the normal Heisenberg interaction and single-ion anisotropy as discussed previously,[2] as well as terms like the Dzyaloshinskii-Moriya interaction.[11] If only such terms contribute, the critical magnetic field (as shown in the phase diagram in Fig. 8) should take the same functional form with temperature as the x component of the magnetic moment μ_x^{AF}. Both are shown in reduced units in Fig. 10 and the difference is significant. We conclude that higher-order terms are indeed necessary. Of course, $5f$-d hybridization may be very important in this compound, as it is in many other uranium compounds,[12] in which case this type of local-moment picture is merely phenomenology.

Finally, our data contain information about the energetics of domain repopulation. Within our model, we can visualise the zero-field-cooled field-dependent data in Fig. 5 as shown schematically in Fig. 5(f). This figure defines fields for the onset (B_1) and completion (B_2) of domain repopulation. The variation of these fields is shown in Fig. 11. At this point, we have little idea as to how to interpret these data.

V. CONCLUSIONS

We have for the first time mapped out the field-temperature phase diagram for the 25 K/3 T phase transition in UPdSn and have shown that this transition is highly hysteretic. We have also shown that there is no such transition above 25 K. These results are independent of any interpretation. We have offered a plausible

interpretation of the hysteresis in terms of domain repopulation, but this can only be confirmed definitively if the other domain pairs are studied and/or the (100) magnetic reflection is studied with field parallel to the [100] axis. Within this interpretation we have also come up with a recipe for preparing majority-domain samples: either field cool the sample in a magnetic field greater than 2.5 T, or ramp the field up to beyond 3.5 T when cold.

While it was not the main purpose of this study, there are strong indications that, at zero field, the 25-K transition is much sharper than observed in previous powder measurements.[3] It may be that this is because the present single crystal is higher quality than the powder sample. The question still remains as to whether this transition is best thought of as being due to moment rotation, or the condensation of the x component of the moment out of incipient fluctuations. The previous powder work favored the former view, but this question is now reopened, and can only be resolved with a temperature-dependent study on multiple reflections in a single crystal. The crucial test will be whether there is any discontinuity, with temperature, in the variation of the total uranium moment. We intend to perform such experiments just as soon as neutron beam time becomes available.

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