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Antiferromagnetic domains in UPdSn

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The magnetization of a single crystal of the hexagonal antiferromagnet UPdSn has been studied in fields up to 5 T in order to examine the energetics associated with antiferromagnetic domains. The magnetic unit cell is orthorhombic, so there are three possible domain orientations within the parent lattice. The low-temperature magnetization reflects both spin-flop transition and domain-depopulation effects. Although the interpretation of our results is complicated by the coexistence of these two phenomena, we can conclude that the domain occupancies are history dependent below the spin-reorientation transition which lies at 25 K, but history independent between this transition and $T_N=40$ K. Comparing magnetization results with those from previous neutron-diffraction studies, we propose a magnetic phase diagram for UPdSn.

INTRODUCTION

UPdSn crystallizes in the hexagonal $P6_3mc$ structure and orders below 40 K (Ref. 1) in a noncollinear antiferromagnetic structure (Fig. 1) based on the $Cmcm$ symmetry (the orthorhombic form of $P6_3mc$). This structure is rearranged into another antiferromagnetic phase of orthorhombic symmetry below 25 K.^{1,2} As the crystallographic structure is hexagonal and the magnetic structure orthorhombic, three different domains can occur. Our recent neutron results show that in a zero-field-cooled sample all three domains are present and almost equally populated at 37 K.³ Therefore we may assume this to be the case down to zero temperature. A magnetic field applied along an axis affects each domain in a different way, because the angle between the applied field and the moment direction is different. Recently, we have observed by means of neutron diffraction the irreversible growth of one domain in a magnetic field at the expense of the others.⁴ At 6 K, the intensity of the antiferromagnetic (010) peak is doubled upon application of a magnetic field of 7 T. However, creation of a monodomain sample would have enhanced the intensity by a factor of 3. The question is whether this change of intensity by a factor 2 is purely accidental, possibly caused by extinction, or whether only one domain is depopulated in 7 T. Although bulk magnetization measurements do not provide a microscopic picture, the magnetization must reflect the response of all three domains. In order to extract the contribution of the domains we also have to take into account the spin-flop transition towards a canted antiferromagnetic state, which occurs in the same field region.⁵

EXPERIMENTAL RESULTS AND DISCUSSION

The single crystal used in the present experiments is the same as used before.⁵ Magnetization measurements as a function of temperature and field were performed in fields

up to 5 T applied along the a and b axes (in the orthorhombic notation) in a Faraday balance and a SQUID magnetometer.

First, field scans were performed at fixed temperatures (Fig. 2). Upward curvature of the magnetization is seen below 40 K reflecting the destruction of the antiferromagnetic phase. Below 25 K, hysteresis is developing, presumably connected with changes in the domain occupation, but possibly also with hysteresis in the transition itself. The absence of any remanence in the magnetization, also after application of a magnetic field, confirms a compensated magnetic structure.

Next, temperature scans were done in fixed magnetic fields (Fig. 3). Below 1.5 T, a slight maximum at about 40 K (Ref. 5) (not distinguishable in Fig. 3) is connected with the antiferromagnetic ordering. In higher fields, an enhancement of the magnetization is observed in the same

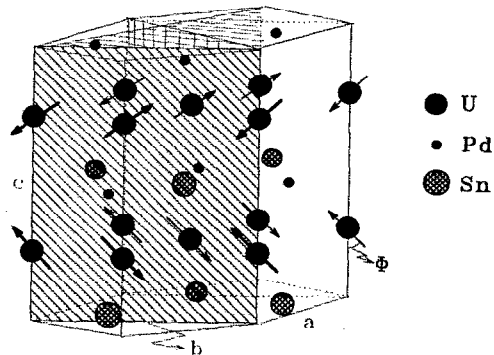


FIG. 1. The magnetic structure of monodomain UPdSn at temperatures between 25 and 40 K. The moments are aligned in the orthorhombic b - c plane indicated by the diagonally hatched rectangle. The canting angle ϕ with the c axis is about 54° . The b - c planes of the other two domains are indicated by vertical and horizontal hatching, respectively. Below 25 K, the moments turn out of the b - c plane by a second canting angle of about 45° . Note, that for reasons of clearness, the c axis is expanded with respect to the b axis.

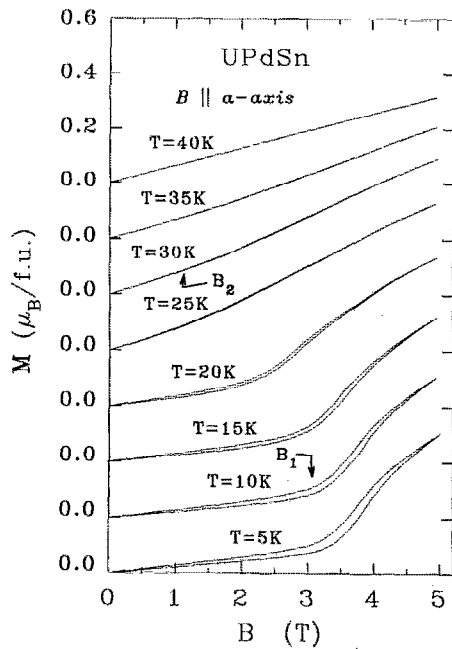


FIG. 2. Magnetization vs field scans at various temperatures for the field applied along the a axis. The critical fields B_i where an upward curvature is observed has been identified with the phase boundaries of the antiferromagnetic phases. The indices of the fields refer to the phase lines in Fig. 5.

temperature region, indicating the boundary between the paramagnetic and the magnetically ordered region. At about 25 K, a second, this time pronounced, maximum is found reflecting the rearrangement of the antiferromagnetic ground state.

The temperature dependence of $M(fc) - M(zfc)$ in magnetic response between a field-cooled sample (fc) and a zero-field-cooled sample (zfc) is shown in Fig. 4. This signal originates mainly from changes in the domain occupancies.

Based on the present results and those obtained in previous neutron experiments⁴ we propose a magnetic phase

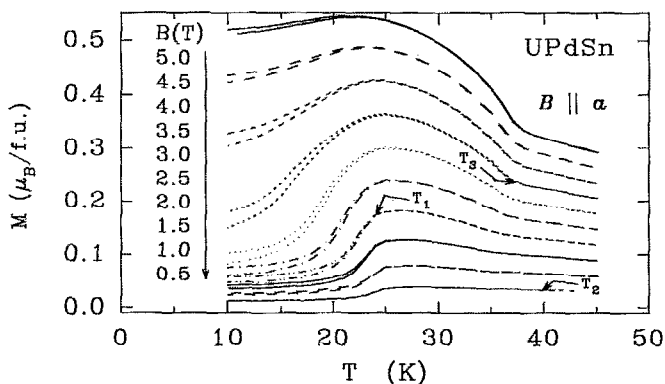


FIG. 3. Magnetization vs temperature scans in various fields applied along the a axis. The critical temperatures T_i of the phase boundaries have been extracted from the maxima. The indices of the temperatures refer to the phase lines in Fig. 5. Note also the difference in the M/B values in the field-cooled (higher values) and the zero-field-cooled data (lower values).

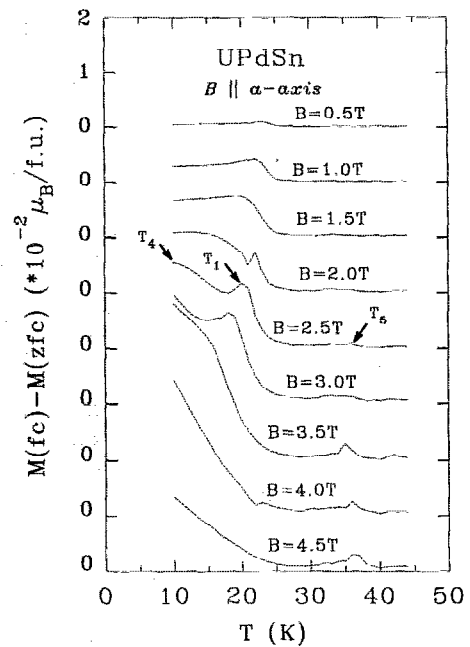


FIG. 4. Temperature dependence of the difference between field-cooled and zero-field-cooled magnetization in various fields, which is thought to originate only from the difference in the occupancies of the three domains. There are three different anomalies at the temperatures T_i visible in these curves. The indices of the temperatures refer to the phase lines in Fig. 5.

diagram for UPdSn as shown in Fig. 5. The critical values obtained from the field scans (Fig. 2) and the temperature scans (Fig. 3) determine thereby the phase boundaries 1-3. The phase diagram contains three different phases

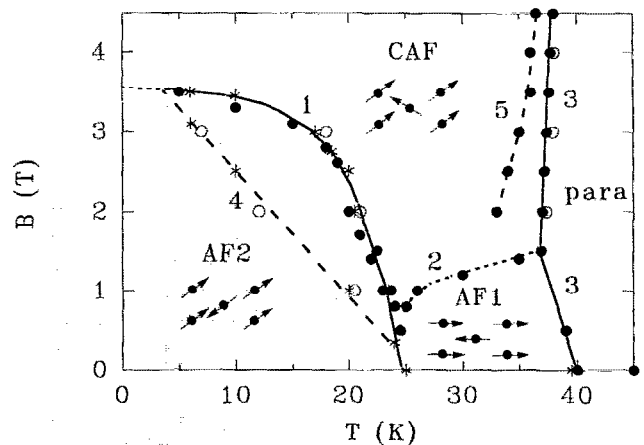


FIG. 5. Magnetic phase diagram of UPdSn. The different symbols used in the plot represent the data obtained from different measurements: *—neutron data taken from Ref. 4; ●—magnetization ($B \parallel a$ axis); ○—magnetization ($B \parallel b$ axis). The moment orientation is represented as a projection on the orthorhombic a - b plane. The numbers indicating the lines represent the following: (1) Phase boundary of the antiferromagnetic phase AF2; (2) presumed phase boundary of the antiferromagnetic phase AF1; (3) phase boundary between the paramagnetic range and the magnetically ordered state; (4) boundary connected with domain effects within the antiferromagnetic phase AF2; (5) boundary connected with domain effects within the CAF phase. Note, that the lines 1-3 are phase boundaries, while the lines 4 and 5 only indicate a change in the domain occupancies.

AF1, AF2, and CAF with moment directions as indicated. There is some uncertainty about phase boundary 2 derived from the critical field for the upturn between 25 and 40 K in Fig. 2. This boundary may be not sharp due to the partial coexistence with phase AF2 surviving above 25 K. Qualitatively different information is given by Fig. 4 as the anomalies in this figure reflect only changes in the domain occupancy. The hump at temperature T_4 is connected with a change of the domain occupancies within the AF2 phase in agreement with previous neutron results.⁴ The second anomaly is a maximum at T_1 , which coincides with the phase boundary between the AF2 and the CAF phases and therefore indicates phase transition to the phase AF2 to coexist with domain effects. The slight anomaly at T_5 , visible only above 1.5 T, may originate from spurious domain effects within the CAF phase.

CONCLUSIONS

From previous neutron-diffraction studies three domains are known to coexist in a *zfc* sample and application of a magnetic field is known to lead to irreversible domain depopulation below 25 K.⁴ Given this, we may speculate about the energetics of the domain effects in UPdSn. The variation in the population of the three domains is reflected in the results in Fig. 4. No significant difference is seen between 25 and 40 K, except for the small anomaly in magnetic fields above 1.5 T, indicating the domain population to be independent of the magnetic history. The enhanced difference between *fc* and *zfc* data at the phase boundary at a temperature T_1 can be interpreted in terms of three domains in the CAF phase. These domains experience slightly different effective fields. Therefore, coming from high temperatures or high fields one domain passes into the AF2 phase, whereas the other two domains remain in the CAF phase until they also reach the boundary. This enhances the response of the magnetization with respect to the field-heated mono-domain sample. In this picture, we suppose that the transformation from AF2 to CAF is accompanied by the evolution of three domains. In absence of anisotropy a mono-domain antiferromagnet prefers perpendicular alignment to the field, whereas a simple ferromagnet prefers parallel alignment. Although our presumed CAF phase is not simply ferromagnetic, we can imagine a situation in which the exchange interaction and the anisotropy energy balance in such a way that the occupancy of all three domains is promoted. Further support for this possibility is found in the observation, that in fields higher than 3.5 T (which is outside of the AF2 phase) domain effects are clearly present (Fig. 3). The situation is more

delicate at the phase boundary between AF2 and AF1, but we believe similar arguments to be valid here also, which is corroborated by the absence of magnetic-history effects in the AF1 phase⁴ and the observed presence of all three domains at 37 K.³

Furthermore, Fig. 4 contains some information concerning the stability of the domain occupancies. Supposing a frozen domain population, $M(\text{fc}) - M(\text{zfc})$ should remain constant all the way down in temperature. Such behavior is only observed for 2 T. With decreasing temperature the difference increases for higher fields and decreases for lower fields. Therefore, changes in the domain occupancies critically dependent on temperature and field are also seen at low temperatures. These effects can be comprised in such a picture: UPdSn prefers a three-domain state, which it gives up domain by domain if a sufficiently strong external field is applied and which it tries to get back in zero field. However, at low temperatures the freedom of the domains to move is strongly restricted. Therefore, the delicate balance of the minimum of the free energy may also allow one domain to depopulate into the second one, whereas the energy difference is too large for the third domain. This, in fact, would explain the factor 2 observed in the neutron experiments. Whether such a picture is correct can be proved only by neutron-diffraction experiments in a horizontal field, which would enable us to control the domain population of all three domains at the same time. Such experiments will be undertaken in the near future.

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