Neutron and magnetization studies of the UP-US system*

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Solid solutions exist for $0 \leq x \leq 1$ in the system $UP_{1-x}S_x$. All compositions are cubic at room temperature with the NaCl crystal structure. The results of neutron-diffraction and bulk-magnetization measurements on a number of compositions are reported, with particular emphasis on the region $0.10 \leq x \leq 0.20$. For $x = 0.1$ the initial magnetic structure ($T_N = 98\,^\circ$K) is type $1A$; at $\sim 85\,^\circ$K this transforms to the type-I structure, but at $\sim 50\,^\circ$K the structure reverts to the $1A$. Bulk-magnetization experiments show that the material is metamagnetic during the high-temperature $1A$-I transition. The induced ferromagnetic moment has been examined by measuring the depolarization of polarized neutrons transmitted through the sample. For $x = 0.15$ the magnetic structure is type $1A$ at all temperatures below T_N. For $x = 0.20$ an additional phase, with a 3 + 3,— configuration, coexists with the type-$1A$ phase between 40 and 80 °K. Large peaks in the magnetization are also observed just below T_N for the $x = 0.15$ and 0.20 compositions.

I. INTRODUCTION

The UP-US solid-solution system has been studied extensively over the last few years. Uranium phosphide is antiferromagnetic ($T_N = 125\,^\circ$K) with ordering of the first kind,[1] and exhibits an interesting "moment jump" at 22 °K, which has been the subject of experimental[2] and theoretical[3] studies. Uranium sulfide is a highly anisotropic ferromagnet ($T_C = 178\,^\circ$K).[4] The complete solubility of US in UP indicates that a series of compounds, characterized by the notation $UP_{1-x}S_x$, where $0 \leq x \leq 1$, may be obtained. In common with UP and US, the solid solutions all have the simple NaCl crystal structure. Previous studies on this system include magnetization,[5,6] specific-heat,[7] nuclear-magnetic-resonance,[8] x-ray-diffraction,[6,9] electrical-resistivity,[6] and neutron-diffraction[10] measurements.

The neutron-diffraction results show that for a substitution of 5% US in UP, i.e., $x = 0.05$ in the notation $UP_{1-x}S_x$, the magnetic structure transforms from the type I to the type $1A$ at low temperature.[11] The type-I structure consists of ferromagnetic (001) sheets stacked in an alternating + − sequence. The spin direction is parallel to the [001] axis, i.e., perpendicular to the ferromagnetic sheets. In the $1A$ structure the spin direction remains the same but the ferromagnetic sheets are stacked in the + + − − sequence. With increasing values of x the I-$1A$ transition occurs at higher temperatures until by $x = 0.15$ the magnetic structure is the type $1A$ at T_N(~95 °K). For $x = 0.25$ the neutron[12] and magnetization[13] experiments indicate that the antiferromagnetic (which dominate in UP) and ferromagnetic (which dominate in US) exchange interactions are comparable. This, in turn, leads to long-range magnetic structures, such as the 5+, 4− configuration.[12] A similar situation exists in the UAs-US system.[14] For $0.25 < x < 0.34$ the detailed phase diagram has not been determined. Although the ferromagnetic interactions are the most important in this region, on the basis of similar studies in the UAs-US system[14] we would anticipate the presence of antiphase or longitudinal-wave modulations. One major difficulty in this composition range is that the individual phases may be stable over only a small range of x values. Since the variations in x over a given sample may be as much as $0.1x$,[9] more than one magnetic phase is often seen in the neutron experiments. Some of the magnetic properties of the UP-US system are summarized in Table I.

The present paper is concerned primarily with the region $0.10 \leq x \leq 0.20$ in the UP-US system. First, by improving the instrumental resolution in the neutron experiments we have clarified the magnetic phase diagram given for this composition range in Ref. 10. Second, we report magnetization measurements for $0.0 \leq x \leq 0.50$. For the samples with $x = 0.10$, 0.15, and 0.20 large peaks in the magnetization are observed at, or just below, the Néel temperature. In the case of $x = 0.10$ the susceptibility at 92 °K is 360×10^{-6} emu/g, whereas at both low and high (300 °K) temperatures

TABLE I. Magnetic properties of $UP_{1-x}S_x$ solid solutions. The paramagnetic Curie temperatures Θ_p, the effective magnetic moment μ_{eff}, the initial ordering temperature, and the value of σ_{00} were obtained from the magnetization experiments. The types of antiferromagnetic order and μ_{sat}, the fully ordered moment at 5 °K, were obtained from neutron experiments.

Nominal x	Chem. annal. x	a_0 (Å)	Θ_p (±3) (°K)	μ_{eff} (±0.05) (μ_B)	Initial ordering (°K)			Subsequent transitions			μ_{sat} (±0.05) (μ_B)	σ_{00} (μ_B)
0	···	5.589	43.5	3.14	125	I		22 °K		I	1.95	
0.02	0.022	5.586	48.1	3.22	122	I		22 °K		I	2.00	
0.05	0.055	5.584	50.4	3.28	116	I		27–35 °K		IA	1.9	
0.075	0.083	5.582	70.3	3.10	110	I		32–42 °K		IA	1.9	
0.10	0.103	5.579	67.4	3.16	98	IA	→	I	→	IA	1.9	
0.15	0.148	5.576	87.8	2.86	95	IA					1.8	
0.20	0.203	5.570	101.6	2.96	92	IA	→	3+, 3−	→	IA	1.8	
0.25	0.235	5.567	113.6	2.90	98	F	→	5+, 4−	→	IA	1.7	1.28
0.28	0.287	5.564	114.7	3.06	109	F	→	LW?	→	IA + F	~1.7	1.18
0.33	0.342	5.561	126.4	2.82	112	F	→	LW?	→	F	~1.7	1.35
0.50	0.486	5.543	148.2	2.79	150	F					1.7	1.50

the value is $\sim 25\times10^{-6}$ emu/g. Third, in an effort to understand the precise origin of this induced magnetic moment, we have performed neutron experiments in an applied field, and measured the depolarization of polarized neutrons transmitted through the sample as a function of both temperature and field.

II. EXPERIMENTAL

We have used the same samples as in the earlier studies.[9,10] Details of the sample preparation and characterization are described in those references and by Baskin.[15] The compositions as determined by chemical analysis are given in Table I. We shall refer to nominal compositions throughout.

A. Magnetic structure

The elastic neutron-diffraction experiments on the samples $x = 0.10$, 0.15, and 0.20 have been repeated with improved instrumental resolution.[14] The results are presented in Figs. 1 and 2. In Fig. 1(a) the intensities of the (110) and (11 $\frac{1}{2}$) magnetic peaks are given as a function of temperature for the $x = 0.10$ sample. The (110) peak is characteristic of the type-I magnetic structure,[1] in which ferromagnetic (001) planes are arranged in the $+-+-$ sequence. The (11 $\frac{1}{2}$) magnetic peak is characteristic of the type-I*A* magnetic structure,[11] in which the ferromagnetic sheets are stacked in the $++--$ sequence. In both antiferromagnetic structures the magnetic moments are aligned parallel to the [001] axis. From Fig. 1(a) we see that the structure at T_N is the type I*A*, but at ~85 °K (on cooling) the structure transforms to the type I, and then *returns* to the type I*A* at ~50 °K. [Considerable hysteresis, indicated in Fig. 1(b), was observed at both the 50 and 85 °K transitions. No hysteresis was observed at the ordering temperature. To avoid confusion over this point, we shall refer to temperatures as observed in cooling the samples.] In all three phases the intensities derived from complete neutron scans have confirmed that the structures are correctly described as type I or type I*A*.

In Fig. 1(b) the square of the magnetic moment is presented as a function of temperature. We note that, as with UAs[14] and $UP_{0.75}S_{0.25}$,[12]

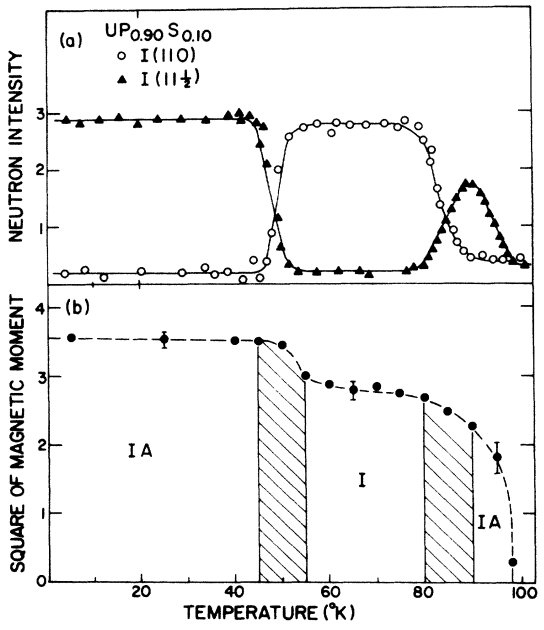

FIG. 1. (a) Intensity of the (110) and (11$\frac{1}{2}$) diffraction peaks as a function of temperature. (b) Magnetic phase diagram derived from the intensities in (a). I and I*A* refer to the antiferromagnetic structures (see text) and the shaded areas indicate mixed-phase regions.

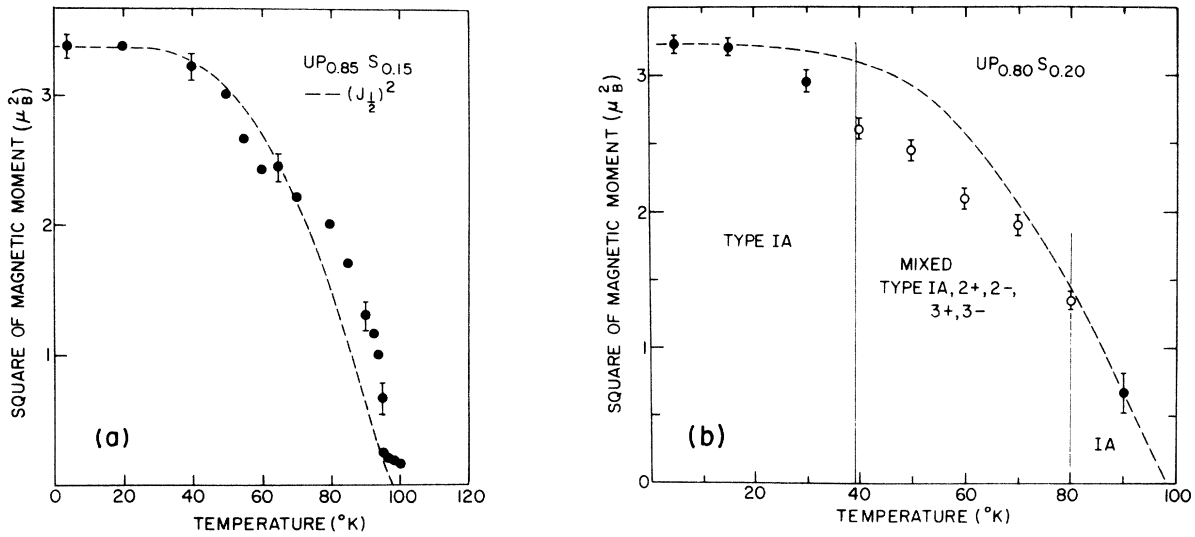

FIG. 2. (a) Square of the magnetic moment as a function of temperature for $UP_{0.85}S_{0.15}$. The magnetic structure is type IA. (b) Magnetic phase diagram as a function of temperature for $UP_{0.80}S_{0.20}$.

the magnetic moment increases abruptly when the low-temperature transition to the type-IA structure occurs. This is also very reminiscent of the "moment jump" in UP,[2] although no structural transition occurs in the latter.

In Fig. 2(a) the square of the magnetic moment as a function of temperature is plotted for $x = 0.15$. The magnetic structure is type IA at all temperatures below T_N, but a rather unusual "hesitation" occurs in the μ^2-vs-T curve at ~60 °K. (This unusual temperature dependence was briefly noted in Ref. 10). No simple explanation for this behavior is apparent. However, the temperature is sufficiently close to that of the I-IA transition in the $x = 0.10$ sample to suggest a relationship between the two. For $x = 0.15$ the exchange interactions may be too weak to force a structural change.

For $x = 0.20$ the situation is more complicated, as illustrated in Fig. 2(b). With the improved resolution as compared to the original experiments,[10] we have seen additional satellites around the (111) nuclear peaks. These satellites are at the positions $(111)^{-\tau}$ and $(111)^{-3\tau}$, where $\tau = \frac{1}{3}c^*$. These peaks, which are in addition to the $(11\,\frac{1}{2})$ peak observed at all temperatures, indicate that a 3+, 3− modulation exists between 40 and 80 °K. A complete description of this type of structure, together with the resulting neutron intensities, is given in Refs. 12 and 14. The presence of a long-range magnetic structure in the $x = 0.20$ composition is not surprising since the 5+, 4− structure ($\tau = 0.222c^*$) appears in the $x = 0.25$ sample.[12] The simultaneous existence of two magnetic phases in this system has been attributed to inhomogeneities in the samples.[9]

Throughout the discussion of Figs. 1 and 2 we have assumed that the magnetic form factor of uranium is known. This is not strictly true, but the magnetic moments have been determined from low-angle peaks ($\sin\theta/\lambda \leq 0.25$ Å^{-1}), where the form factor will be independent of the electronic configuration of the uranium ion. We have used the form factor given in Refs. 12 and 14.

B. Magnetization

The magnetization of a set of 11 samples with compositions in the range $x = 0.0$ to $x = 0.5$ has been measured as a function of temperature and applied magnetic field strength. The measurements were made on one of two force balances, depending on whether the samples were weakly (paramagnetic) or strongly (ferromagnetic) magnetized.[16,17] The paramagnetic measurements were made at a few field strengths up to a maximum of about 12 kOe, with a view to confirming that the magnetization was linearly dependent on the field (susceptibility independent of field). The ferromagnetic measurements were made at closely spaced intervals of field, up to a maximum of 28.5 kOe. The over-all results of the measurements are shown in Figs. 3–6. Where susceptibilities are quoted, it has been established that the susceptibility is independent of field (except for the sharp peak in the $x = 0.10$ sample); otherwise magnetization values are given. Where appropriate, ferromagnetic Curie temperatures were estimated by the Arrott[18]-Belov[19] procedure. From measurements of magnetization (σ) in different fields (H) and at different temperatures (T), graphs were constructed of σ/H against σ^2 at constant T. These were extrap-

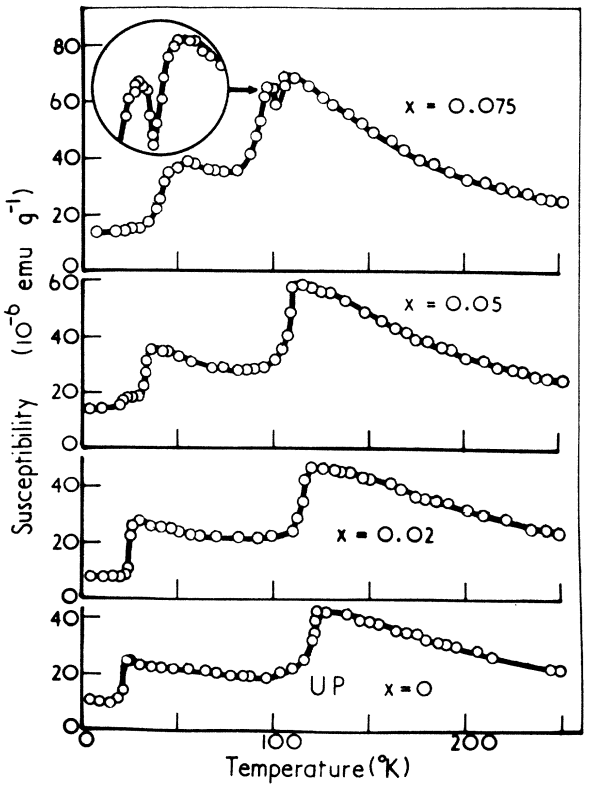

FIG. 3. Magnetic susceptibility of the $UP_{1-x}S_x$ samples with $x = 0$, 0.02, 0.05, and 0.075.

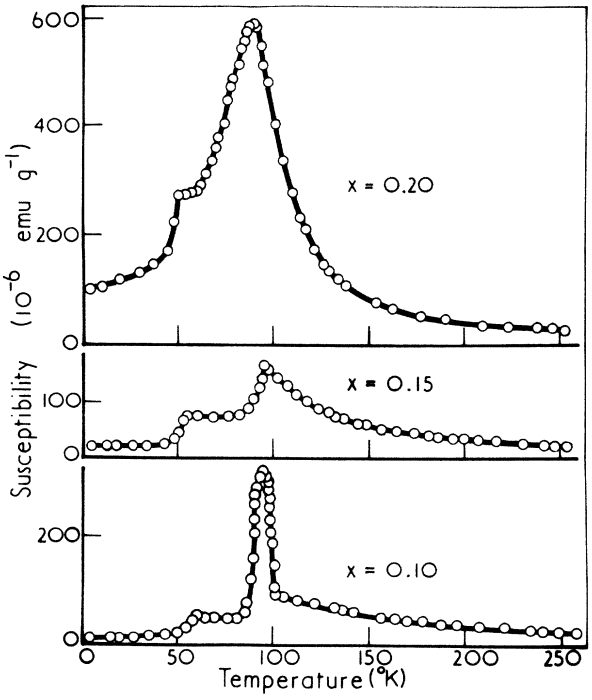

FIG. 4. Magnetic susceptibility of the $UP_{1-x}S_x$ samples with $x = 0.10$, 0.15, and 0.20, measured in a field of 12 kOe.

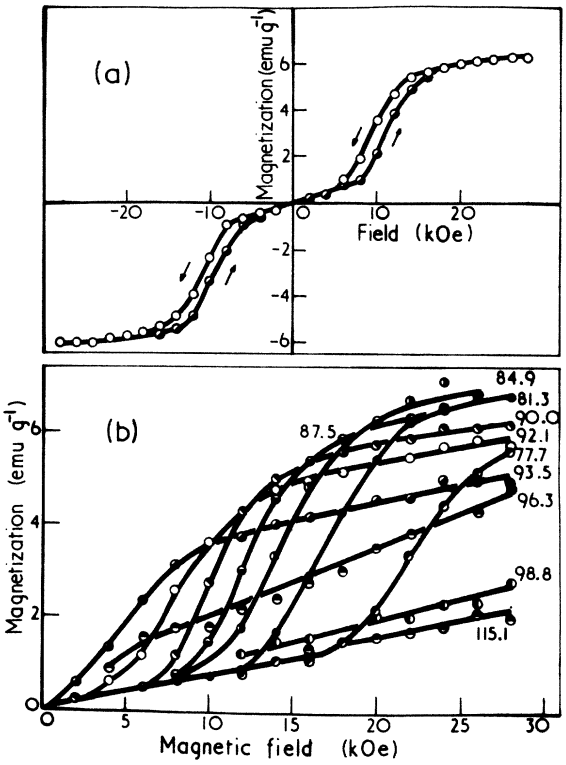

FIG. 5. Metamagnetic behavior in $x = 0.10$ sample. (a) Hysteresis loop measured at 90 °K. (b) Isothermal magnetization-field curves measured at the temperature indicated on the various curves.

olated to $(\sigma/H)_{T,0}$, where $\sigma^2 = 0$. Interpolation of a further graph of $(\sigma/H)_{T,0}$ against T to that temperature at which $(\sigma/H)_{T,0} = 0$ gave the Curie temperature.

For $x < 0.1$ the susceptibility (Fig. 3) shows two anomalies, one at T_N and other other at a lower temperature, which is the same as the "moment jump" temperature in the $x = 0.0$ and 0.02 samples and the I-IA transition in the $x = 0.05$ and 0.075 samples. The results for UP ($x = 0.0$) are very similar to those reported by Gulick and Moulton.[2] For $x = 0.075$ a small peak appears in the susceptibility at 97 °K. At 92.5 °K the graph of the magnetization plotted against field shows a slight tendency to turn upwards at 16 kOe. This sample seems to lie on the edge of the range of compositions that exhibit metamagnetic behavior (see below). The peak at 97 °K is distinct from that at the Néel temperature of 109 °K.

For $x = 0.10$, 0.15, and 0.20 very large peaks are observed in the susceptibilities at temperatures close to T_N (Fig. 4). A close examination of the 0.10 sample revealed a metamagnetic transition (Fig. 5), in which the application of an external magnetic field leads to a transition into an appar-

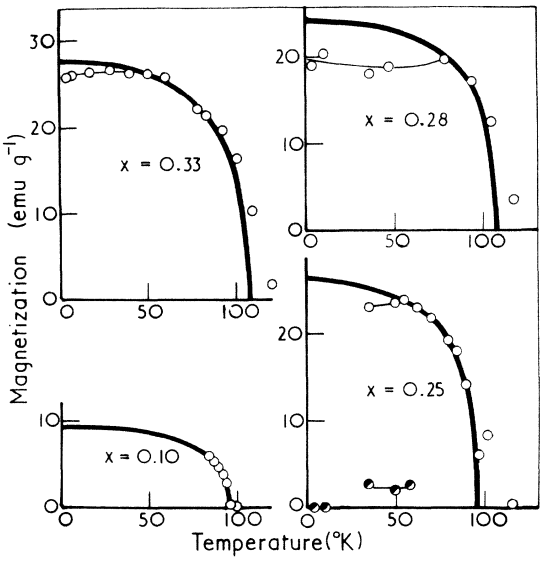

FIG. 6. Spontaneous magnetization plotted against temperature for the ferromagnetic states of the samples with $x = 0.10$, 0.25, 0.28, and 0.33. The solid lines represent Brillouin curves ($J = \frac{1}{2}$) fitted to the experimental points. For $x = 0.25$ the shaded points are for low-field extrapolations to the spontaneous moment.

ently ferromagnetic state. The critical field for the start of this process decreases sharply, and apparently linearly with temperature, from 16.5 kOe at 78 °K to 0 at 95 °K. In sufficiently strong fields (depending on the temperature) what seems to be a normal ferromagnetic state is reached which possesses an intrinsic magnetization σ_{HT} that increases with increasing field like a conventional ferromagnet. Extrapolating this back to $H = 0$ gives the spontaneous magnetization σ_{0T} for the induced ferromagnetic state. Fitting a Brillouin function (for example, $J = \frac{1}{2}$) to the variation of σ_{0T} with temperature suggests that the Curie temperature of the induced ferromagnetic state is about 96 °K and that its fully aligned moment σ_{00} would be 9.5 emu/g, i.e., $0.46\mu_B$/(U atom). To test whether the metamagnetic behavior was possibly due to domain wall pinning within a ferromagnetic state, the specimen was taken around a hysteresis loop at 90 °K, between field limits of ±28.5 kOe. The form of the curve [Fig. 5(b)] shows a narrow loop, but the main effect is characteristic of a flipping and not of a wall-pinning mechanism. The indications are that the sharp peak in the susceptibility graph (Fig. 4) is wholly due to the flipping mechanism. This peak does not appear in the initial susceptibility.

The maximum moment per uranium atom in the fully ordered antiferromagnetic (AF)1*A* state at low temperature is $1.87\mu_B$ as indicated by neutron diffraction. A maximum induced ferromagnetic moment of $0.46\mu_B$ suggests, in the simplest interpretation, that 0.46/1.87 = 25% of the mass of the material is involved in the flipping reaction. The critical field H_c for initiating the metamagnetic transition decreases rapidly with increasing temperature, becoming zero at between 92 and 94 °K. H_c is roughly proportional to $(\Theta/T)^{1/2}$, but over the limited range of temperature it is not possible to be sure of the relationship.

For $x = 0.15$ anomalies occur in the susceptibility at 55 °K and at 95 °K (Fig. 4). The upper one is clearly at the Néel temperature, but the reason for the lower one is not obvious. It coincides with the unusual hesitation of μ^2 vs T seen in the neutron results [Fig. 2(a)], but no change in the magnetic structure is observed in this sample. For $x = 0.20$ the main peak at 90 °K (Fig. 4) is at the Néel temperature. The shoulder at ~50 °K is probably associated with the onset of the 3+, 3− modulation that exists between 40 and 80 °K [Fig. 2(b)]. The unusually high value of the susceptibility at the top of the main peak (590×10^{-6} emu/g) seems to be a consequence of the close proximity of T_N and the paramagnetic Curie temperature (102 °K).

For $x = 0.25$ the neutron[12] and magnetization[13] results have been published. Metamagnetic behavior is observed with a field-induced transition from a magnetically compensated state (i.e., the 5+, 4− magnetic structure observed by neutron diffraction) to ferromagnetism. The magnetization-vs-field curves are similar to those in Fig. 5(a), except that the transition temperatures are different. The full ferromagnetic state is reached in the field used in the present work at temperatures between 55 °K and $T_C = 98$ °K. As for the $x = 0.10$ sample, the spontaneous magnetization σ_{0T} has been estimated at each temperature by extrapolating linear parts of the graphs of σ_{0T} against H to $H = 0$. This has been done both for the high-field and, where appropriate, for the low-field parts of the (σ, H) curves. To estimate the saturation magnetic moment σ_{00} a Brillouin curve was fitted to the graph of σ_{0T} against temperature (Fig. 6). The value obtained was $\sigma_{00} = 26.5$ emu/g, corresponding to $1.28\mu_B$/(U atom). The measured Curie temperature was used as a fixed point in fitting the curve and in the absence of better information the Brillouin curve for $J = \frac{1}{2}$ was fitted. The result is relatively insensitive to the value of J assumed.

For temperatures between 35 and 59 °K and fields below the metamagnetic transition the corresponding approximate extrapolation gives $0.12\mu_B$/(U atom), which is about $\frac{1}{9}$ of $1.28\mu_B$. This is consistent with the observation by neutron diffraction of the 5+, 4− magnetic structure, which would be expected to have a moment of $(5-4)/(5+4) = \frac{1}{9}$ of the full moment. AT 4 °K and 10 °K the low-field extrapolation is towards a moment of zero,

corresponding to a fully compensated antiferromagnetic state. Saturation was not reached in fields of up to 30 kOe at temperatures between 35 and 55 °K.

For the $x = 0.28$ and 0.33 samples ferromagnetic behavior was observed. Graphs of magnetization against temperature are included in Fig. 6. Brillouin curves have been fitted as before, taking the measured Curie temperatures as fixed points. The values for σ_{00} for the $x = 0.28$ and 0.33 samples are 24.4 and 27.8 emu/g, respectively. An unusual aspect of the magnetization of these samples, as well as of the 0.25 sample, is the apparent drop of σ_{0T} below the Brillouin curve at the lower temperatures. Although the effects of anisotropy are important in actinide ferromagnets, this drop in σ_{0T} may indicate the presence of a longitudinal-wave modulation of the magnetic moments at lower temperatures in these samples. For $x = 0.50$ conventional ferromagnetic behavior was observed and $\sigma_{00} = 31.0$ emu/g $= 1.5\mu_B$ /(U atom).

The saturation magnetization of the samples with $0.25 \leq x \leq 0.50$ are in the range 1.2 to $1.5\mu_B$/(U atom), which is significantly below the moments of $\sim 1.7\mu_B$ determined by neutron diffraction (Table I). This difference may be due to the very high anisotropy in actinide ferromagnets,[20] although with the exceptions already mentioned the specimens did appear to be saturated in fields of 30 kOe.

C. Neutron diffraction in a magnetic field

To investigate whether the magnetic structure of the $x = 0.10$ sample changes in a magnetic field we have performed two neutron experiments with the sample in a field. First, neutron scans were taken with and without an applied field of 8.5 kOe and with the sample at 91 °K. Within experimental error the two scans were identical, indicating that the antiferromagnetic nature of the type-IA ordering is unaffected by such a field. A small ferromagnetic moment of $0.18\mu_B$ would not have been observed in this experiment. Second, the induced magnetic moment in the polycrystalline sample has been measured with polarized neutrons. With a field of 10.4 kOe applied to the sample, the induced moment at 91 °K is $0.18\mu_B$/(U atom) (see Fig. 5). Such a magnetic moment corresponds to a magnetic scattering amplitude of $p = 0.043 \times 10^{-12}$ cm at the (200) reflection with $\sin\theta/\lambda = 0.179$ Å^{-1}. The nuclear scattering amplitude at the same reflection is $b = 1.342 \times 10^{-12}$ cm. The polarized-beam technique[21] for elastic scattering at a Bragg reflection consists of measuring the Bragg intensity as a function of the two neutron spin states. The ratio of these intensities is the so-called "flipping ratio." For one incident neutron spin the intensity of the reflection is proportional to $(b + p)^2$, and for the reverse spin state is $(b - p)^2$. The expected flipping ratio for the (200) reflection is therefore 1.137. This value has to be lowered slightly if the neutron beam is depolarized when passing through the sample. The experimental value for the flipping ratio at 91 °K and with an applied field of 10.4 kOe is 1.12 ± 0.03, which confirms the magnetization results.

D. Neutron-beam depolarization

A highly polarized beam of neutrons transmitted through a magnetic sample will be reduced in polarization if any component of magnetization perpendicular to the neutron-spin polarization direction exists within the sample. The neutron can be treated classically if the dimensions of its wave packet are small compared with the dimensions of regions of constant magnetization in the sample. Under these conditions the change in the polarization depends on the magnitude of the magnetic inductance within a domain and on the average domain size.[22] If, on the other hand, the magnetization fluctuates over atomic dimensions, as in a para- or antiferromagnet, the dimensions of the neutron wave packet are larger than the fluctuations and the neutron depolarization must be averaged over the packet. In this case the depolarization is much smaller and usually cannot be detected. In general, therefore, we expect to observe depolarization for ferromagnetic samples (unless they are fully saturated), but not for para- or antiferromagnets.

The depolarization experiments were performed on a polarized-neutron diffractometer at the CP-5 Research Reactor. Polarized neutrons are produced by a $Co_{0.92}Fe_{0.08}$ monochromator crystal by transmission from the (200) planes.[21] The neutron polarization can be reversed by a radio-frequency oscillator tuned to the neutron Larmor precession frequency. The neutrons are then transmitted through the polycrystalline sample, thickness of 0.7 cm, and Bragg reflected from the (200) planes of a matching (with the monochromator) CoFe analyzer crystal. Both the sample and analyzing crystal were contained in a cryostat and variable magnetic field ($H_{max} = 10.4$ kOe) assembly. The temperature gradient in the sample was estimated to be less than 2 °K. The temperature of the sample was measured with a carbon resistor for $T < 30$ °K and with a platinum resistor for higher temperatures. With no sample in the beam the flipping ratio ($I_{rf\,off}/I_{rf\,on}$) of the analyzer crystal was 80. By inserting the sample between the rf coil and the analyzer the flipping ratio will be reduced if depolarization occurs. This method is very sensitive since the flipping ratio can be measured easily to 0.5%. A change in the flipping ratio R from 80 to 75 corresponds to a depolarization of 0.17%.

Initially experiments on ferromagnetic US illus-

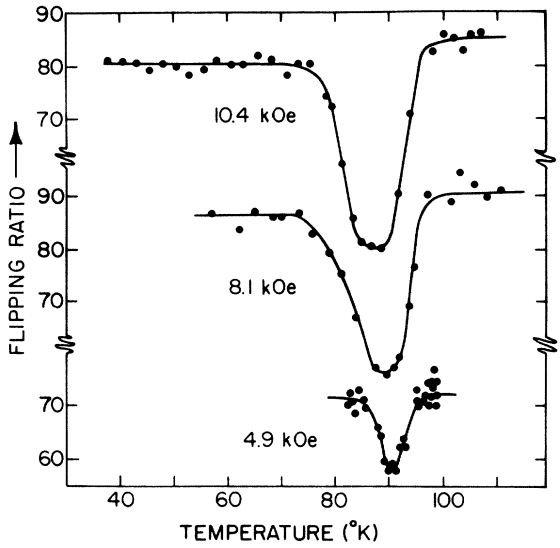

FIG. 7. Flipping ratio of the CoFe analyzer crystal behind the $x = 0.10$ sample as a function of sample temperature and applied magnetic field.

trated that severe depolarization occurs for temperatures below T_C. A small depolarization ($< 0.2\%$) was observed with the sample in the paramagnetic state because a small fraction of the incident neutrons will be scattered magnetically and thus experience a spin reversal.[23] No depolarization was observed in an antiferromagnetic sample of UP. The results for the $x = 0.10$ sample are shown in Fig. 7. The depolarization is essentially zero in both the paramagnetic and low-temperature ($T < 70$ °K) regions, but exhibits maxima (minima in R) very similar to those observed in the magnetization experiments at intermediate temperatures. The neutrons are depolarized in this experiment because of the ferromagnetism induced in the sample by applying a magnetic field. The magnetization experiments (Fig. 4) indicate that at the I-IA transition the antiferromagnetic configurations are unstable in the presence of a small applied field, the critical field being essentially zero at ~90 °K. In addition, the anisotropy of these systems guarantees that the magnetization directions of the ferromagnetic domains will not all be parallel to the applied field. Under these conditions some depolarization of the transmitted neutron beam will occur. With an increase in the applied magnetic field, both the amount of depolarization as well as the temperature range over which it occurs should increase. Figure 7 indicates that both these effects are observed to be a function of field. According to the magnetization experiments, at $H = 10$ kOe, approximately 10% of the sample is ferromagnetic. Applying the Halpern-Holstein formalism[22] the ferromagnetic domain size in the powder particles is ~1 μm, whereas the Larmor precession distance is 50 μm. The size of the powder particles is estimated at 40 μm. The depolarization measurements therefore reinforce the concept of the metamagnetic transition, since they have detected regions of ferromagnetism in a sample that is predominately antiferromagnetic.

III. DISCUSSION

Magnetic investigations of $UP_{1-x}S_x$ for $0.1 \leq x \leq 0.2$ reveal a delicate balance between the interactions stabilizing the type-I and type-IA magnetic structures. In this study we have concentrated on the $x = 0.10$ composition, in which the magnetic transitions in zero field are well characterized (Fig. 1), and a metamagnetic state is observed for relatively low fields in the temperature region in which the structure transforms from the IA to I structure. Neutron experiments have shown that no structural modification of the type-IA configuration exists. At ~90 °K the material is either a type IA antiferromagnet or ferromagnetic. In a single-crystal experiment on such a system the response to the magnetic field would presumably be first order, the critical field depending on the magnitude of the anisotropy and exchange fields. In a polycrystalline material the random orientation of the antiferromagnetic moments with respect to the field direction leads to a finite slope of σ versus H in the critical-field region [Fig. 5(a)]. A similar situation occurs in the observation of spin-flop transitions in polycrystalline samples.[24] Neutron experiments with a polarized-neutron diffractometer demonstrate that the concept of an induced ferromagnetic moment in a sample that is predominantly antiferromagnetic is correct. The sensitivity of the depolarization experiments is a direct consequence of the polycrystalline nature of the specimen and this technique should be of wider application, for example, in investigations of amorphous magnetic systems.

A simple phenomological explanation for the induced moment in the antiferromagnetic system takes note of the nature of the exchange coupling between the (001) sheets in the type-I (+ − + −) and type-IA (+ + − −) structures. At the transition between the two, the interlayer coupling in some cases changes sign (i.e., goes from anti- to ferromagnetic), and is, therefore, essentially zero at some intermediate temperature. The magnetic spins are then free to follow the applied magnetic field, resulting in a metamagnetic configuration. However, this metamagnetic state is not associated simply with the I-IA transition because no metamagnetic behavior (up to the fields of 28 kOe) is observed at the second IA-I transition ~50 °K. Without detailed neutron experiments in higher magnetic fields (preferably with single crystals)

the exact magnetization process cannot be determined unambiguously. Clearly the temperature dependence of the anisotropy and exchange interactions will determine the critical field necessary for metamagnetism in a given composition.

Finally, we should point out the similarities between the magnetic structure and magnetization results[25] for CeBi and $UP_{0.90}S_{0.10}$. The type-I*A* magnetic structure, which is common to a number of cerium and uranium compounds, is a particularly intriguing one, in that it cannot be predicted by nearest-neighbor interactions, nor can it be stabilized by bilinear exchange terms. Further work, especially on single crystals if they could be obtained, would be most interesting.

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