

Investigation of magnetic properties of monocrystals of the uranium compounds U_3P_4 and U_3As_4 in strong fields

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Measurements of the magnetization of monocrystals of U_3P_4 and U_3As_4 have been made in constant magnetic fields up to 12 kOe and in pulsed magnetic fields up to 300 kOe. It is shown that these compounds possess a very large anisotropy: fields of order 10^5 Oe are insufficient for magnetic saturation along the directions of difficult magnetization (axes of [100] type). The data obtained are in agreement with a noncollinear-magnetic-structure model of U_3P_4 and U_3As_4 .

Magnetic measurements on powders^[1,2] and monocrystals^[3] of U_3P_4 and U_3As_4 (a body-centered cubic structure of Th_3P_4 type, space group $I43d-T_d^6$) have shown that these compounds are ferromagnetic at low temperatures. In^[4], neutron-diffraction investigations of the magnetic structure of U_3P_4 were carried out on powder specimens, and it was shown that the experimental data obtained could be interpreted by supposing that the magnetic structure of this compound was collinear. From magnetization measurements on monocrystals of U_3P_4 and U_3As_4 ^[3] it follows also that they possess a very large magnetic anisotropy: in a field of order 10 kOe, oriented along an axis of difficult magnetization (a direction of [100] type), the magnetization shows practically no deviation from the axis of easy magnetization (a direction of [111] type). This deduction was later confirmed in our measurements of the magnetostriction of U_3P_4 monocrystals in strong magnetic fields^[5].

To obtain more detailed information about the anisotropy of the compounds U_3P_4 and U_3As_4 , we have carried out an investigation of the magnetic properties of monocrystals of these materials in constant magnetic fields up to 20 kOe and in pulsed magnetic fields up to 300 kOe. The U_3P_4 and U_3As_4 specimens were grown by the transport reaction method^[6]. The magnetization in pulsed fields was measured by the induction method on rods of dimensions $5 \times 1.5 \times 1.5$ mm, oriented along the [111] and [100] axes. The measurements of the magnetic properties in constant fields were made on monocrystalline disks of diameter 2 to 3 mm and thickness 1.5 to 2 mm; the plane of the disks was parallel to a (110) plane of the crystal. The magnetization was measured with a vibration magnetometer, the torque with a torque magnetometer of the Akulov type.

The measurements in constant fields showed that the Curie points of the U_3P_4 and U_3As_4 monocrystals, determined by the method of thermodynamic coefficients, were 142 and 199°K, respectively; that is, close to the values obtained on polycrystalline specimens (138 and 198°K^[1,2]). Our results also confirm the data of^[3]: in a field of order 10 kOe, the magnetization shows practically no deviation from the [111] axis. This is clearly evident in Fig. 1, which shows the dependence of the torque $L(\varphi)$ acting on the U_3P_4 and U_3As_4 monocrystals upon the orientation of the field in the (100) plane. The experimental functions $L(\varphi)$ agree well with the theoretical functions

$$L(\varphi) = \sigma_{[111]} H \sin \varphi, \quad (1)$$

constructed on the assumption that the magnetic moment is rigidly attached to axes of [111] type and does not deviate from these axes in a field ($\sigma_{[111]}$ is the magnetization along the [111] axis, φ is the angle between the field direction and the [111] axis nearest to the field direction). The "collapses" on the $L(\varphi)$ curves at field orientations along axes of [110] and [100] types are due to the fact that in fields parallel to these directions, a reorientation occurs from one to another of the axes of [111] type.

The results of the measurements of the magnetic properties of U_3P_4 and U_3As_4 in strong pulsed magnetic fields are shown in Figs. 2 and 3. Figure 2 shows the dependence of the magnetization of these compounds in directions [111] and [100] on the field at various temperatures, and Fig. 3 shows the dependence of the magnetization in these directions on the temperature at various fields. From these figures it is evident that the magnetization of U_3P_4 and U_3As_4 in a [100] direction, even in fields exceeding 100 kOe, is considerably smaller than along a [111] axis. We also direct attention to the

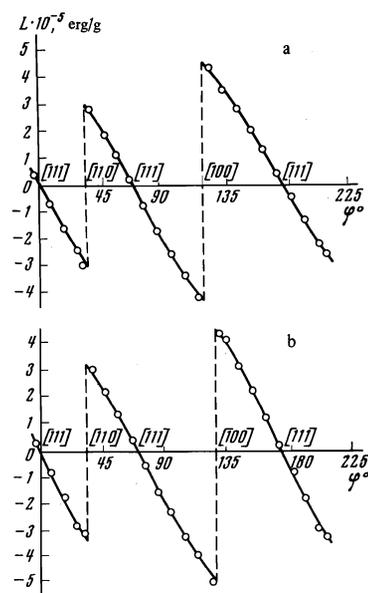


FIG. 1. Dependence of the torque acting on a monocrystal of U_3P_4 (a) or U_3As_4 (b) upon the orientation of the field in the (110) plane: $T = 78^\circ K$, $H = 19.5$ kOe. Points, experimental data; solid lines, theoretical dependence according to formula (1).

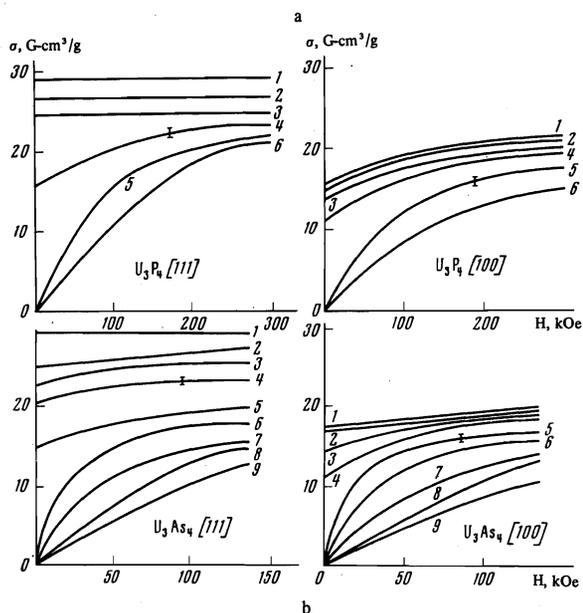


FIG. 2. Dependence of the magnetization of the compounds U_3P_4 and U_3As_4 on the field, at various temperatures, along [111] and [100] axes: a) 1–4, 2; 2–78; 3–100; 4–120; 5–140; 6–160°K; b) 1–78; 2–120; 3–140; 4–160; 5–180; 6–190; 7–200; 8–210; 9–220°K. The vertical lines show the relative error of measurement.

fact that in the magnetically ordered state, the strong-field susceptibilities along the [111] and [100] directions are different. At nitrogen and helium temperatures, the susceptibility of the compounds studied along the [111] axis does not exceed $2 \times 10^{-5} \text{ cm}^3/\text{g}$, whereas the susceptibility along the [100] axis amounts to $(6 \text{ to } 8) \times 10^{-5} \text{ cm}^3/\text{g}$ in a field of order 100 kOe. We note that the difference in magnetization along the [111] and [100] axes in strong fields is observed not only in the magnetically ordered state, but also in the paramagnetic temperature range (Fig. 3). At the same time, our measurements show that the initial paramagnetic susceptibility of U_3P_4 and U_3As_4 is isotropic (Fig. 3).

Thus our results attest to a very large magnetic anisotropy of the compounds U_3P_4 and U_3As_4 . If we suppose that these materials are collinear ferromagnets, then estimates show that at nitrogen and helium temperatures the saturation fields of U_3P_4 and U_3As_4 along a [100] axis exceed 1000 kOe, and the constants of cubic magnetic anisotropy are in order of magnitude $(1 \text{ to } 1.5) \times 10^7 \text{ erg/g}$.

A more complete interpretation of the experimental data is difficult, since the magnetic structure of U_3P_4 and U_3As_4 has not yet been definitely explained. In^[3] it was suggested that in these compounds at low temperatures there occurs a noncollinear magnetic structure, consisting of three sublattices whose magnetic moments are oriented along three different directions of [100] type. The assumption of such a type of magnetic order agrees, as was shown in^[7], with the symmetry of the crystallographic structure of U_3P_4 and U_3As_4 and does not contradict the neutron-diffraction data obtained by U_3P_4 in^[4]. It also enables us to explain some of the peculiarities of the magnetic properties of these compounds: the difference in the value of the magnetic moment of the uranium ion in U_3P_4 and U_3As_4 in the paramagnetic and in the magnetically ordered states^[3] and also the low value of the magnetic entropy in U_3P_4 ^[8].

In the crystal structure of U_3P_4 and U_3As_4 there are

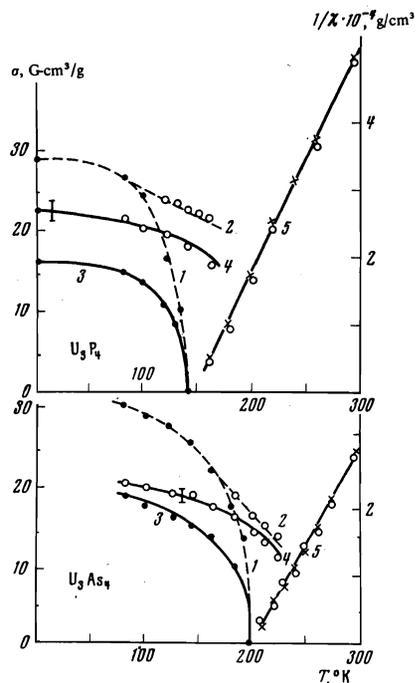


FIG. 3. Temperature dependence of the magnetization and of the paramagnetic susceptibility of the compounds U_3P_4 and U_3As_4 , along different directions: 1—spontaneous magnetization in [111] direction; 2—magnetization in [111] direction in field 300 kOe for U_3P_4 and in field 140 kOe for U_3As_4 ; 3—spontaneous magnetization in [100] direction; 4—magnetization in [100] direction in field 300 kOe for U_3P_4 and in field 140 kOe for U_3As_4 ; 5—reciprocal of paramagnetic susceptibility: X—along [111] axis; O—along [100] axis. Vertical line shows absolute error of measurement.

three types of site occupied by uranium ions, nonequivalent from the point of view of the local symmetry of the crystalline field^[7,9]. This leads to the result that the field of the ligands tends to orient the magnetic moments of the uranium ions on nonequivalent sites along different directions of [100] type. As was shown in^[10], a three-sublattice noncollinear magnetic structure with resultant moment along a [111] axis can be formed as a result of competition between the fields of the ligands and the positive exchange interaction between the sublattices, which tends to orient them parallel to one another. Analogous calculations that we have made show in addition that formation of a noncollinear three-sublattice structure can occur also when the exchange interaction between the sublattices is negative. We note that in such models, a noncollinear magnetic structure with the magnetic moments of the sublattices along three mutually perpendicular directions of [100] type can be formed only if the field of the ligands is considerably larger than the effective field of exchange interaction. If these fields are comparable in magnitude, there occurs a three-sublattice structure in which the magnetic moments of the sublattices are inclined to the [100] axes.

Almost no consideration has been given to magnetization processes in a three-sublattice noncollinear magnetic structure. It is clear, however, that in such a structure the susceptibility along a [111] axis should be smaller than along a [100] axis, since in the first case the application of a field leads only to a deformation of such a structure, whereas in the second there occurs also a rotation of it as a whole in the field. Approximate estimates made in^[10] show that when the exchange interaction between the sublattices is positive, the initial susceptibility of U_3P_4 along the [100] axis should be from

three to four times as large as along the [111] axis. This conclusion does not contradict our experimental data.

One of the peculiarities of a noncollinear magnetic structure is the fact that even at low temperatures, the susceptibility in strong fields along the direction of easy magnetization (a [111] axis) should be different from zero^[10]. Our experiments have shown that in U_3P_4 and U_3As_4 , the susceptibility along the [111] axis at nitrogen and helium temperatures, with our measurement accuracy, is close to zero (it does not exceed 2×10^{-5} cm³/g). This, however, does not contradict the conclusion that a noncollinear magnetic structure is present in the compounds studied; for estimates show^[10] that in U_3P_4 , the susceptibility along the [111] axis caused by noncollinearity should amount to $\sim 2 \times 10^{-6}$ cm³/g, which is considerably smaller than the value that our apparatus enables us to determine.

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