Giant-magnetoresistance anomaly associated with a magnetization process in UFe_4Al_8

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Magnetoresistance and magnetization measurements on a UFe_4Al_8 single crystal are reported. A strong anisotropic magnetoresistance is found in this compound and is used to prove the ferromagnetic order of the U lattice. A strong anomaly in the parallel and perpendicular magnetoresistance is interpreted as an unusual magnetization process in which the magnetization remains blocked perpendicularly to the applied magnetic field. This interpretation is confirmed by the magnetization measurements.

The giant-magnetoresistance effects and the intermetallic compounds based on actinides are hot topics in the field of magnetism. At the frontier between these two topics, we report in this paper an anomalous behavior in the magnetization process of $UFeAl_8$ which leads to a strong and unusual anomaly in the magnetoresistance. Although studied for a long time,[1,2] the magnetic structure of UFe_4Al_8 remains an enigma. This compound crystallizes in the tetragonal $ThMn_{12}$-type structure with the U atoms in the center of a cage formed by eight Fe atoms.[3] Mössbauer[4] and neutron[4–6] experiments showed that the Fe atoms order below 150 K and an antiferromagnetic (AF) arrangement is most frequently proposed,[5,6] the magnetic cell remaining identical to the crystallographic one. Due to this high symmetry, neutron experiments are difficult to interpret unambiguously, and up to now failed to determine the magnetic structure of the U lattice. However, magnetization measurements provide some clear results: on a powder sample, they indicate (a, b) as an easy magnetization plane[7] and recent magnetization measurements on a single crystal[8] showed that a and b are the easy axes. The remanent magnetization deduced from these measurements combined with the AF order of the Fe atoms suggest a ferromagnetic (FM) state for U atoms with a magnetic moment of $1.6\mu_B$/U at 4.2 K. At low temperatures the magnetization curves presented a step which had not been interpreted.[8] This paper focuses on this last feature. A strong magnetoresistance anomaly suggests that this step involves an unusual magnetization process in which the magnetization remains blocked in a direction perpendicular to the applied field; this interpretation is confirmed by the magnetization measurements.

The resistivity was measured up to 160 kOe on a parallelipipedic single crystal, with the current flowing along the largest dimension (b axis). Due to the crystallographic equivalence of the a and b axes, a 90° rotation of the sample holder around the c axis of the crystal allowed the measurement of the so-called perpendicular ($\mathbf{H}\perp\mathbf{I}$, $\rho_\perp$) and parallel ($\mathbf{H}\|\mathbf{I}$, $\rho_\|$) magnetoresistivity with the magnetic field $\mathbf{H}$ always parallel to one of the easy axis. The longitudinal ($M_\| : M\|H$) and transverse ($M_\perp : M\perp H$) magnetization measurements were performed in a superconducting quantum interference device magnetometer equipped with two sets of pickup coils, one with the axis coil parallel to the external field and the other one perpendicular to it. The single crystal was introduced in the sample space with the a axis parallel to the direction of the magnetic field and the b axis parallel to the axis of the transverse coils. With the experimental arrangement, the magnetization along the two easy axes was measured during the inversion magnetization process.

Figure 1 shows the behavior of the magnetoresistance (MR) for the two configurations. Above 30 kOe, the MR is always negative and monotonous up to 160 kOe.[9] The different values for the resistivity obtained for the perpendicular ($\rho_\perp$) and the parallel ($\rho_\|$) configurations are due to the so-called anisotropic magnetoresistance (AMR) effect. For bulk materials, this effect is observed in ferromagnetic materials and is due to a scattering cross section for the conduction electron that depends on the angle between their spins and the localized magnetic moment[10] or to the Lorentz force acting on the conduction electrons, eventually enhanced by the intense internal magnetization due to the FM alignment.[11] A discussion on the microscopic origin of this AMR in this compound is beyond the scope of this paper.

Upon an increase of the applied magnetic field from high negative values up to zero (from the left to the middle of Fig. 1), the AMR ($\rho_\perp > \rho_\|$) subsists. The existence of this AMR in zero applied field during a hysteresis cycle indicates that the cause of this effect is still present. Using the ferromagnetism as the source for this AMR, the nonzero AMR at $H=0$ denotes a remanent magnetization as already observed.[8] In the case of an antiferromagnetic order of the iron sublattice, this AMR at zero field can be ascribed to a

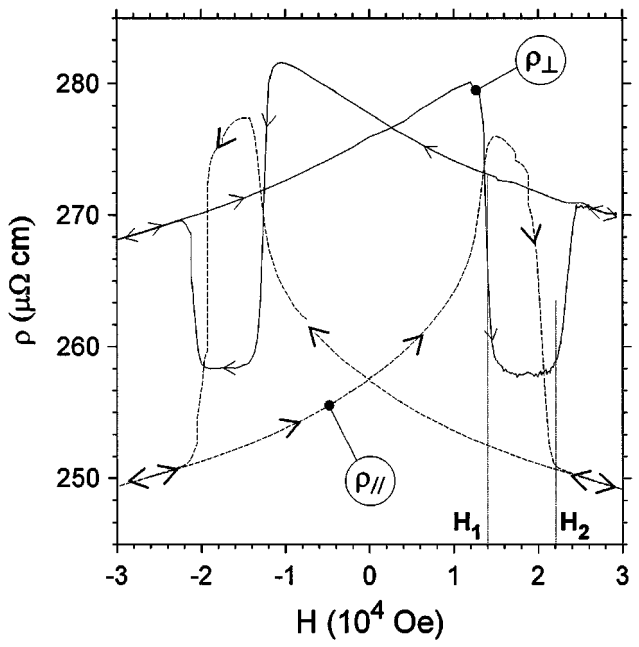

FIG. 1. Magnetoresistance of a UFe_4Al_8 single crystal in the perpendicular ($\rho_\perp$: $H\perp I$, $H\|a$) and parallel ($\rho_\|$: $H\|I$,$H\|b$) configurations at 2 K. The complete hysteresis cycles, starting from negative magnetic field values, are shown. The magnetoresistance curves obtained from the virgin state (zero-field cooled) are not shown. H_1 and H_2 denote the two characteristic fields. The small asymmetries in respect to $H=0$ are due to temperature drifts during the field sweeps.

FM order of the uranium atoms. Reversing the field direction and increasing it (from the middle to the left of Fig. 1), a huge anomaly shows up for both configurations between two characteristic fields H_1 and H_2 (Fig. 1). Two major features characterize the anomalies.

(i) The amplitude of the anomaly, both for perpendicular and parallel configurations, is close to the difference $(\rho_\perp-\rho_\|)$ far away from H_1 and H_2, this feature existing up to the highest temperatures where the anomaly is still clearly seen ($T\approx100$ K).

(ii) The anomaly has an opposite sign in the two configurations.

These two facts can be summarized by describing the anomaly as an interchange of $\rho_\perp$ and $\rho_\|$ for fields between H_1 and H_2. Such an interchange can be explained by two 90° rotations of the magnetization in the (a,b) plane: At the first one, for $H=H_1$, **M** rotates from antiparallel (**M**$\|a$, **M**$\|-$**H**) to perpendicular to the magnetic field (**M**$\|b$, **M**$\perp$**H**) and at the second one, for $H=H_2$, from perpendicular to parallel (**M**$\|a$, **M**$\|$**H**) (Fig. 2). As previously explained, the relevant angle for the AMR is the angle between the electrical current I and the magnetization M instead of the angle between I and the applied field H, therefore, due to the 90° change of the angle between the magnetization and the electrical current, the two rotations previously described lead to the interchange of $\rho_\perp$ and $\rho_\|$ (Fig. 2). During a field decrease (Fig. 1, from right to left), symmetrical curves are obtained. Measurements with the magnetic field applied along the c axis did not show any anomaly, proving that the rotation occurs in the (a, b) plane.

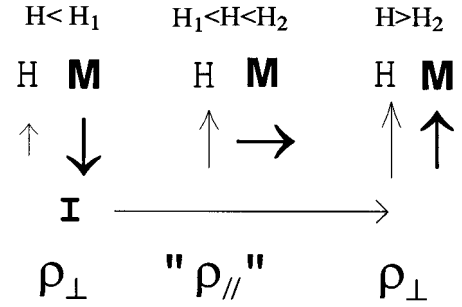

FIG. 2. Sketch of the angles between the magnetic field H, the magnetization M, and the intensity I for the perpendicular configuration ($I\perp H$) during the two-steps inversion magnetization process. For the parallel configuration ($I\|H$)—not shown in the figure—instead of [$\rho_\perp$, "$\rho_\|$", $\rho_\perp$] for increasing H, [$\rho_\|$, "$\rho_\perp$", $\rho_\|$] is obtained.

The magnetization inversion process is displayed in Fig. 3(a) which shows the longitudinal (**M**$\|a\|$**H**) and transverse (**M**$\|b$, **M**$\perp$**H**) magnetization for a hysteresis cycle. The equivalence of the characteristic fields H_1 and H_2 detected by MR and magnetization is clearly seen in Fig. 3(b). Figure 3(a) shows that between these two fields the longitudinal magnetization is close to zero, whereas the transverse one approaches the longitudinal magnetization value for fields higher than the upper characteristic field (M_{H_2}). This figure strongly confirms the two 90° rotations of the magnetization proposed to explain the anomaly detected in transport measurements. Such a behavior, in which the total magnetization

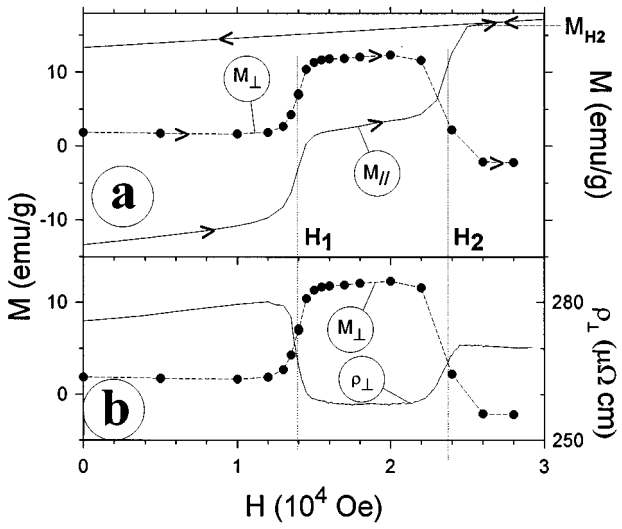

FIG. 3. (a) Longitudinal ($M_\|$:$M\|H$,$M\|a$) and transverse ($M_\perp$:$M\perp H$,$M\|b$) magnetization at 2 K as a function of the applied field H. For the sake of clarity, only the results for positive fields are shown. The sample has been magnetized on a field of -50 kOe and measurements are taken increasing the field up to 50 kOe and then decreasing it. The virgin magnetization curves are not shown. (b) perpendicular magnetoresistance ($\rho_\perp$) and transverse magnetization ($M_\perp$) in the same conditions of Fig. 2(a). The magnetoresistive anomaly occurs between H_1 and H_2, when the magnetization has rotated 90°.

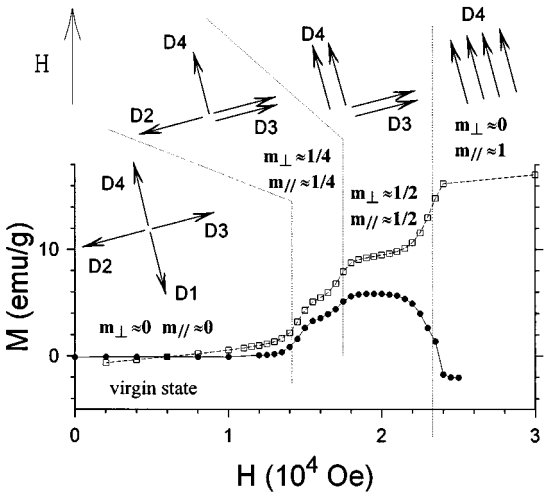

FIG. 4. Virgin magnetization curves at 2 K (open squares: $M_{\parallel}$; closed circles: $M_{\perp}$). The measurements are performed increasing the field from 0 up to 25 kOe, after cooling under zero magnetic field. The relative population of the domains, when H increases, is sketched in the upper part of the figure as proposed in the text.

remains blocked perpendicularly to the magnetic field ($H_1<H<H_2$) before its alignment along the field ($H>H_2$), is unusual and contrasts with the standard FM materials, where the global magnetization rotates directly from the positive to the negative direction at the coercive field (hysteresis square cycle) or progressively reverses direction upon domain formation.[12] The fact that, between H_1 and H_2, $M_{\perp}$ does not reach exactly the value M_{H_2} can be explained by a small misalignment of the crystal axes in respect to the coil axes; the negative signal obtained for $M_{\perp}$ at fields higher than H_2 is probably also caused by this misalignment. On the other hand, the contribution to the magnetization of the Fe sublattice under magnetic field is probably not negligible and should also be taken into account for an exact quantitative description of the magnetization results.

The virgin magnetization curve (Fig. 4) shows three different steps which strongly support our interpretation. Due to the symmetry of the magnetic cell, the virgin state (sample cooled under zero magnetic field) consists of four equally populated domains ($D1$, $D2$, $D3$, and $D4$), each with a magnetization pointing in one of the two directions of the two easy axes a and b (Fig. 4). Increasing the field up to a value close to H_1, the less energetically favorable domain D_1 rotates 90°. However, in the case of a small crystal misalignment, the directions 2 and 3 are not equivalent due to the Zeeman energy, and the rotation of the $D1$ magnetization populates the D_3 domain: the relative magnetizations $m_{\parallel,\perp}$, both in the direction of the field ($m_{\parallel}=M_{\parallel}/M_{H_2}$) and perpendicular to it ($m_{\perp}=M_{\perp}/M_{H_2}$) are equal to $\frac{1}{4}$ (Fig. 4). A further increase of the field will lead to a reorientation of D_2 along the direction of D_4 ($m_{\parallel}=m_{\perp}=\frac{1}{2}$, Fig. 4), and finally, at H_2, the domain D_3 disappears leading to a single domain state ($m_{\parallel}=1$, $m_{\perp}=0$, Fig. 4). Following this reasoning, in the nonvirgin curve, the domain D_2 is never populated, leading to the simpler magnetization curves of Fig. 3(a). As in the case of Fig. 3(a), a misalignment of the crystal and the contribution of the Fe atoms are probably the reasons for the nonexact expected value ($\frac{1}{4}$, $\frac{1}{2}$, 1) for the relative magnetizations $m_{\parallel}$ and $m_{\perp}$.

The virgin curve for the longitudinal magnetization with steps at $m_{\parallel}=\frac{1}{4}$ and $m_{\parallel}=\frac{1}{2}$ is reminiscent of the magnetization curve obtained in the so-called multi-k structure (or multisteps) compounds like CeSb where the magnetization jumps correspond to a change in the antiferromagnetic magnetic structure.[13,14] In these types of compounds, no hysteresis, or at least a small one, occurs. In our case, the important hysteresis and the appearance of a transverse magnetization together with the features of the resistive anomaly unambiguously exclude such a type of explanation.

The blockage of the magnetization in such an *a priori* unfavorable energetic position is not obvious to understand. The standard model, which takes into account only the global magnetic moment, the applied field, and classical anisotropy considerations to evaluate the energy of the different magnetization orientations,[15] is insufficient to describe this phenomenon. As a matter of fact, when the Zeeman energy ($E=-\mathbf{M}\cdot\mathbf{H}$) is higher than the energy barrier between a and b due to the magnetocrystalline anisotropy ($E=K_1\sin^2 2\Theta$ where K_1 is the anisotropy constant of the crystal and Θ the angle between the magnetization and an easy axis), a magnetic moment antiparallel to the magnetic field rotates. In this simple model, the magnetization cannot stay perpendicular to the magnetic field because the torque ($\mathbf{M}\times\mathbf{H}$) on the magnetic moment is maximum and forces an alignment of the magnetization along the field. Actually, the role of the Fe atoms, the magnetic coupling between these and the U atoms, as well as the anisotropy energies for both species have to be considered for a complete understanding of this behavior: for instance, a change in the angle between the alignment axes of the Fe and U atoms or a new order for the Fe sublattice could drastically change the equilibrium configuration and might lead to such an unusual magnetic behavior.

As a conclusion, the magnetoresistance results obtained on a UFe_4Al_8 single crystal show a strong anisotropic magnetoresistance effect. This effect has been used to confirm the ferromagnetic alignment of the U moments, a result which would be hardly obtained by neutron diffraction experiments. The magnetoresistance curves display a very peculiar anomaly in the parallel and perpendicular configurations which is interpreted as a two-steps magnetization process due to two 90° rotations. Magnetoresistance measurements confirmed this interpretation. Due to their important technological applications, the magnetization processes for the FM compounds have been widely studied.[15,16] However, as far as we know, a blockage of the global magnetization perpendicularly to the applied field during the magnetization process, as it occurs in UFe_4AL_8, has never been previously described. This unusual state, coupled to the highly anisotropic magnetoresistance, leads to the spectacular magnetoresistance behavior of Fig. 1 that places UFe_4Al_8 in a new family of giant-magnetoresistance compounds.

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