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Preparation of uranium-based magnetic amorphous alloys by RF sputtering

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Abstract

The effects of amorphization on ferromagnetic uranium compounds UGe_2 and UGa_2 were investigated. The amorphous alloys a-UGe_2 and a-UGa_2 were successfully produced by radio frequency (RF) sputtering. The ferromagnetic exchange of a-UGe_2 is comparable with the crystalline compound. On the contrary, the ordering temperature and the magnetic moment of a-UGa_2 are strongly reduced. The magnetic responses and the electron-transport properties of the alloys are different from those of the ferromagnetic crystalline counterparts, because the collinear spin arrangement was scattered by the random anisotropy originating from the orbital angular momentum of uranium ions. © 1998 Elsevier Science S.A.

Keywords: Amorphous alloy; UGe_2; UGa_2; Magnetism; Random anisotropy; Sputtering

1. Introduction

In spite of the progress in the study of amorphous magnetism during past 20 years, not many studies have been reported on the actinide amorphous alloys in comparison with the transition-metal and rare-earth amorphous alloys. Nevertheless, the magnetism of the crystalline uranium compounds showing a large variety of ground states is an important field of solid-state physics. It seems that this stagnancy of the studies on the actinide amorphous alloys is caused by a difficulty in sample preparation. Recently our group, however, changed the subject from cerium-based amorphous alloys [1] to uranium-based amorphous alloys [2,3].

Sputter deposition is a particularly efficient technique for preparing amorphous alloys, however, there are some difficulties in cleaning an inner wall of the chamber contaminated by scattered materials. This is a rather serious problem for radioactive materials. Therefore, we made a sputtering machine modified for the preparation of uranium-based amorphous alloys in a glove box.

First of all, we tried to make the two uranium-based amorphous alloys a-UGe_2 and a-UGa_2. The crystalline counterpart UGe_2 is a typical itinerant ferromagnet with a high magnetic anisotropy below $T_C=52$ K [4], whereas the 5f electrons are relatively localized in the ferromagnetic compound UGa_2 having $T_C=125$ K [5]. In this work, the magnetic and electrical properties of a-UGe_2 and a-UGa_2 were investigated.

2. Sample preparation

An RF magnetron system was employed to confine an argon plasma into a narrow space between the alloy target and substrate. A deposition rate of about 10 $Å s^{-1}$ was achieved using alloy targets of 30 mm in diameter and a bulk amorphous sample of 100 μm thickness could be deposited on water-cooled Cu substrate within a few days operation. The alloy targets were prepared by arc melting using 99.9% uranium, 99.9999% germanium and 99.9999% gallium.

Fig. 1 shows X-ray diffraction patterns of the a-UGe_2 and a-UGa_2. The first peaks for both alloys were broadened to over 7°, and are followed by weak oscillations for angles of $2\theta>45°$. Thus, we confirm these samples as amorphous alloys with considerable atomic randomness. The bar diagrams indicate the calculated relative intensities of the UGe_2 and UGa_2 having $ThGe_2$-type (Cmmm) [6] and AlB_2-type (P6/mmm) [7] structure, respectively. The main peak of a-UGe_2 is situated at a slightly lower angle than the dominant (131) reflection of the crystalline compound UGe_2. On the contrary, the dominant peak position of the a-UGa_2 is strongly shifted

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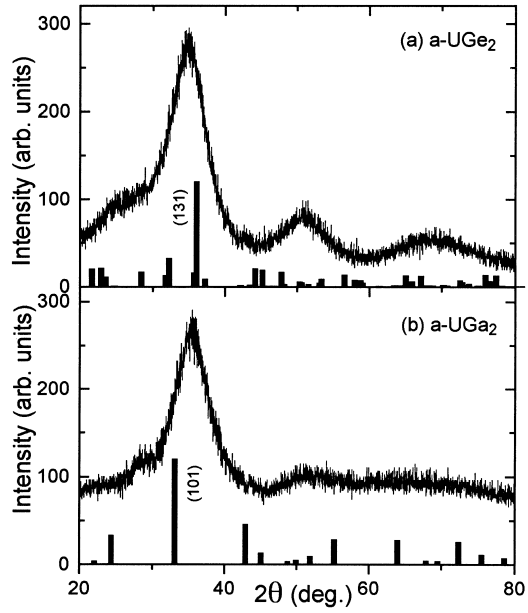

Fig. 1. X-ray diffraction patterns of (a) a-UGe_2 and (b) a-UGa_2.

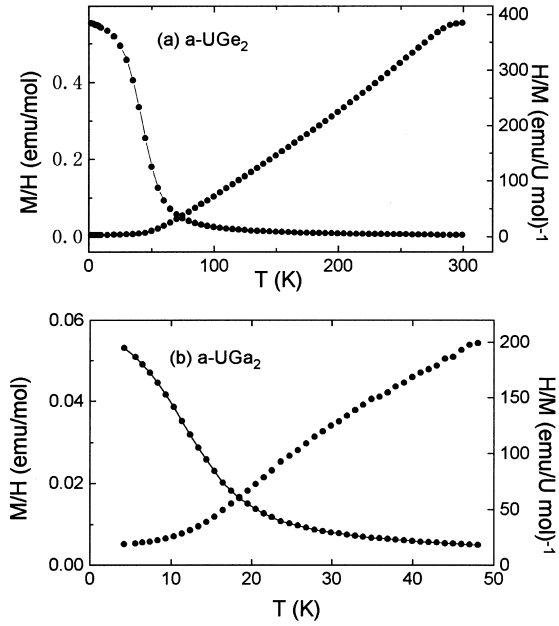

Fig. 2. Temperature dependence of reduced magnetization, M/H, and its inverse values, H/M, for (a) a-UGe_2 and (b) a-UGa_2.

up from the (101) reflection of the UGa_2 crystal. Considering these diffraction patterns as reflecting predominantly the uranium–uranium correlation, the amorphization shrinks the uranium–uranium nearest neighbour distance in the UGa_2 system.

3. Magnetic properties

Temperature dependence of the magnetization was obtained by a vibrating sample magnetometer in a field of 10 kOe. Fig. 2(a) and (b) show the reduced magnetization, M/H, and its inverse, H/M, for a-UGe_2 and a-UGa_2. The M/H of a-UGe_2 shows a strong increase below 80 K similar to the crystalline counterpart. On the other hand, the increase in the M/H of a-UGa_2 is not so large in comparison with the ferromagnetic UGa_2 compound. By an extrapolation from the high-temperature Curie–Weiss behaviour we obtained estimates of the paramagnetic Curie temperature, θ_p, 75 K (for a-UGe_2) and 7 K (for a-UGa_2). The suppression of the ferromagnetic exchange interaction induced by amorphization is more remarkable in a-UGa_2 rather than in a-UGe_2, because the Curie temperature T_C is 52 and 125 K for UGe_2 and UGa_2, respectively. The effective magnetic moments, μ_{eff}, in the paramagnetic state were estimated to be 2.1 and 1.4 μ_B/U for a-UGe_2 and a-UGa_2: both of them are rather smaller than those of the uranium ions (3.62 μ_B/U for U^{3+} and 3.58 μ_B/U for U^{4+}) and of the crystalline counterparts (2.5 μ_B/U for UGe_2 [3] and 3.5 for UGa_2 [4]).

The field dependence of the magnetization for the amorphous alloys was measured at 4.2 K by a sample-extracting magnetometer in the hybrid magnet. As shown in Fig. 3, both curves show strong hysteresis with the remanent magnetization of 0.48 and 0.18 μ_B/U for a-UGe_2 and a-UGa_2, respectively. In particular, for fields below 40 kOe, there are obvious differences between initial and reverse curves. The magnetic moments in the field of 300 kOe of the a-UGe_2 never reach the values of saturated moment for the single crystal sample along the c-axis, which is the easy magnetization direction (1.43 μ_B/U) [3]. Amorphization reduced effectively the magnetic moments, especially for the UGa_2 system: the moment of a-UGa_2 in the field of 300 kOe is smaller than one fourth of the saturated moment for the compound (2.7 μ_B/U) [4]. These non-saturated magnetizations seem to be affected by the random anisotropy around partially localized uranium ions having large orbital angular momentum [8].

An Arrott plot is a powerful analysis to investigate the manner of ferromagnetic phase transition. Mean-field-

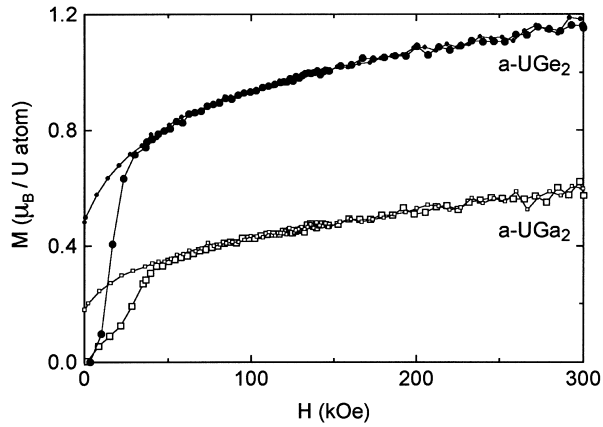

Fig. 3. High-field magnetization curves of a-UGe_2 and a-UGa_2.

theory predicts that H/M should be linear in M^2. Intercepts along the abscissa give the inverse susceptibilities for the paramagnetic phase and intercepts along the positive ordinate represent the squares of the spontaneous magnetization for the ordering phase. The effect on random anisotropy for the Arrott plot can be understood from the following equation [9]:

$$\frac{H}{M}=\left(\frac{T-T_C}{T_C}\right)+M^2\left\{1+A\left(\frac{H}{M}\right)^{-\varepsilon/2}\right\}. \quad (1)$$

T_C is the ordering temperature and $\varepsilon=1-d$ where d is the dimensionality of the system. For the collinear ferromagnets, the ordering temperature T_C means the Curie temperature. Since the factor A is proportional to the square of a ratio of the random anisotropy to the average exchange, $(D/J)^2$, the plots deviate from straight lines when the random anisotropy is strong [10]. As shown in Fig. 4(a), the curves for several temperatures of a-UGe_2 form lines parallel to each other similar to collinear ferromagnets when the applied field is high enough. At low field, however, the curves bend downward for temperatures of 20 and 30 K which are lower than T_C. It seems that the extrapolations of these curves never cross the positive ordinate, which indicates no spontaneous magnetization at any temperature. On the other hand, curvature of the Arrott plot remains positive for all measured temperatures of the a-UGa_2 as shown in Fig. 4(b).

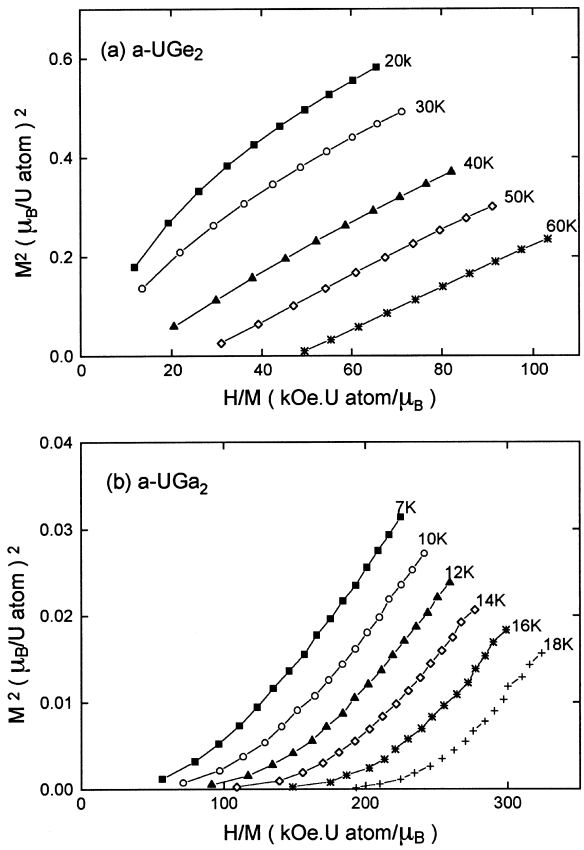

Fig. 4. Arrott plots for (a) a-UGe_2 and (b) a-UGa_2.

4. Electrical properties

Electrical resistivity was measured by a conventional four-probe method. The normalized resistivity at 300 K was plotted in Fig. 5. In this work, we performed a measurement down to 50 mK for a-UGe_2. A resistivity minimum was observed at 88 and 43 K for a-UGe_2 and a-UGa_2, respectively. The same types of behaviours are sometimes found in the rare-earth [11] and uranium-based [12] amorphous alloys containing the non-S state ions with orbital angular momentum. In these cases, the negative temperature coefficient of resistivity is interpreted by the coherent scattering model taking account of the waves scattered by the exchange interaction on neighbour magnetic ions. Fert et al. found the following expression of the magnetic resistivity due to exchange scattering [13]:

$$\rho_m=\rho_M[c_{ion}+c_{ion}^2 m(2k_F)], \quad (2)$$

where c_{ion} is a concentration of non-S state magnetic ions. The ρ_M gives the contribution from the independent scattering by each single ion. Since $m(2k_F)$ is the correlation function between magnetic ions, the second term results from the coherent scattering. Therefore, the fact that the resistivity minima are situated at temperatures higher than T_C in both samples implies the growth of the spin–spin correlation even above T_C. The weak temperature dependence in the resistivity of a-UGe_2 below 1 K is supposed to reflect the frozen moment.

Fig. 6 shows the temperature dependence of the longitudinal magnetoresistance for the same applied magnetic field. The signs of the magnetoresistance changed from positive to negative at around 50 and 20 K for a-UGe_2 and a-UGa_2, respectively. It suggests that interference by the coherent scattering is depressed when the

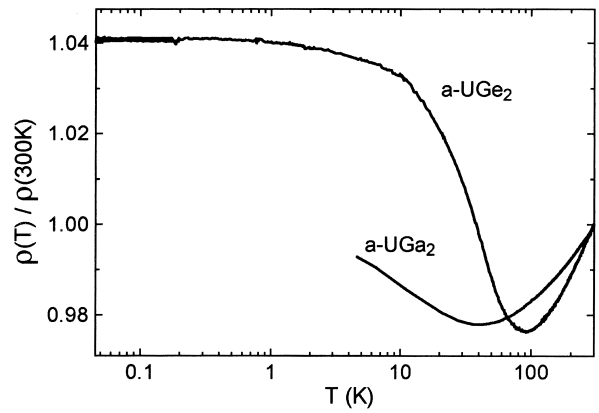

Fig. 5. Temperature dependence of normalized resistivities of a-UGe_2 and a-UGa_2.

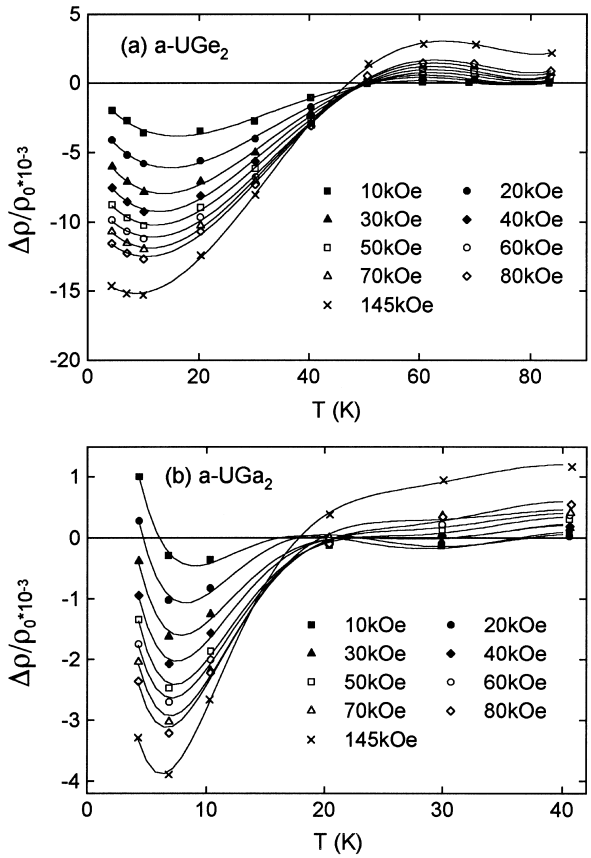

Fig. 6. Temperature dependence of magnetoresistance of (a) a-UGe_2 and (b) a-UGa_2 for several magnetic fields.

magnetic spin–spin correlation was grown below ordering temperature. The same behaviour was observed in rare-earth amorphous alloys such as a-TbAg where the anti-ferromagnetic correlations exist [14].

5. Conclusion

Amorphous alloys a-UGe_2 and a-UGa_2 were successfully produced by the RF sputtering installed in the glove box. a-UGe_2 possesses the ferromagnetic exchange comparable with the crystalline compound. On the contrary, the ordering temperature and the magnetic moments of a-UGa_2 are strongly reduced. In both alloys, the magnetic responses are different from those of the ferromagnetic crystalline counterparts, because the collinear spin arrangement was disturbed by the random anisotropy. Typical resistivity minima in present alloys suggest the existence of the spin–spin correlation between non-S state uranium ions possessing the orbital angular momentum.

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