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Anomalous magnetic properties of an iron film system deposited on fracture surfaces of α-Al_2O_3 ceramics*

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An iron film percolation system is fabricated by vapour-phase deposition on fracture surfaces of α-Al_2O_3 ceramics. The zero-field-cooled (ZFC) and field-cooled (FC) magnetization measurement reveals that the magnetic phase of the film samples evolve from a high-temperature ferromagnetic state to a low-temperature spin-glass-like state, which is also demonstrated by the temperature-dependent ac susceptibility of the iron films. The temperature dependence of the exchange bias field H_e of the iron film exhibits a minimum peak around the temperature T=5 K, which is independent of the magnitude of the cooling field H_{cf}. However, for $T > 10$ K, (1) H_e is always negative when H_{cf}=2 kOe and (2) for H_{cf}= 20 kOe (1Oe≈80 A/m), H_e changes from negative to positive values as T increases. Our experimental results show that the anomalous hysteresis properties mainly result from the oxide surfaces of the films with spin-glass-like phase.

Keywords: iron film, ceramics, exchange interactions, magnetic measurements
PACC: 7550B, 7560E, 7530C

1. Introduction

It is well known that the surface and interface roughness of the magnetic film system is related to its magnetic properties, such as magnetic anisotropy, coercivity, magnetoresistance and magnetic domain structure, etc.[1−3] Usually, rough film surfaces are fabricated by the ion-bombardment, nonequilibrium growth technique and deposition on rough substrates.[4] Sometimes, rough thin films, in which both the upper and lower surfaces are uneven, can also be prepared by using the fracture substrates of various ceramic materials. The anomalous physical properties of the non-magnetic rough film system deposited on the fracture surfaces of α-Al_2O_3 ceramics, such as the electrical transition properties, $1/f$ noise, etc., have been studied systematically.[5−8] In principle, it is expected that the physical properties of the magnetic thin films may be greatly affected by the rough surface structures of both the films and substrates. For instance, Ye *et al* [9] investigated the Fe film system deposited on liquid substrates by thermal evaporation method recently, they found that both the exchange anisotropy field $H_e(T)$ and the coercivity $H_c(T)$ of the iron films first increase with the temperature, then reach their maximum peaks around the critical temperatures, after that decrease dramatically, and finally they approach the steady values around T=40–50 K, indicating the existence of the spin-glass-like phase in the system below the freezing temperature. It is noted that the surface of the liquid substrate is also very rough.

In this paper, we report the magnetic properties of an iron film percolation system deposited on rough fracture surfaces of α-Al_2O_3 ceramics. Several anomalous behaviours of the exchange bias field and ac susceptibility of the films are observed. The evolution from a high-temperature ferromagnetic state to a low-temperature spin-glass like state is detected. Our experiment shows that the anomalous hysteresis properties of the iron films at low temperatures may result from the spin-glass-like state in the oxide surfaces of the iron films.

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2. Experimental

The samples were fabricated by thermal evaporation of iron (purity 99.99%) in a vacuum of 6.5×10^{-5} Pa at room temperature. The α-Al_2O_3 ceramics (purity 97%) were prepared by the hot-pressed sintering technique. The ceramic substrates, in a thickness of 2–3 mm, were broken with a small hammer. Then the fresh and homogeneous random fracture surfaces of the bulk were used as the film substrates. The deposition rate was f=0.8±0.1 nm·s^{-1} and the nominal film thickness was in the range of d=20–40 nm, which was determined by a quartz-crystal balance. For the sake of comparison, the iron films on both the fracture surfaces of α-Al_2O_3 ceramics and the glass surfaces were fabricated simultaneously.

After deposition, the samples were removed from the vacuum chamber. Then the measurements were taken immediately for the as-prepared samples: the temperature dependence of the magnetic properties of the iron films was measured by a physical property measurement system (PPMS-9, Quantum Design) magnetometer in the temperature range from 2 to 300 K and at low and high magnetic fields (up to 2 T), applied in the film plane. Before the magnetization measurement, the zero magnetic fields were first calibrated. The ac susceptibility of the film samples was measured in an ac magnetic field of 10 Oe (1 Oe≈80A/m) with frequency f=300, 1000, 3000, 10000 Hz after the film was cooled without magnetic field applied. X-ray diffractometer (XRD, Rigaku D/Max 2500/PC with Cu Kα radiation) and field-emission scanning electron microscopy (SEM, SIRION100) were used to study the crystal structure and surface morphology of the as-prepared samples, respectively.

3. Results and discussion

The crystal structure of an as-prepared sample is shown in Fig.1(a). The XRD pattern of the sample exhibits regular α-Al_2O_3 peaks but no iron peaks (see Fig.1(a)), providing evidence that the iron film exhibits microcrystalline or amorphous structure, which is in agreement with that of the non-magnetic films on the fracture surfaces of α-Al_2O_3 ceramics.[5] Figure 1(b) shows the SEM photograph of the surface of the sample, in which a statistical fractal structure is observed. We notice that, since the film is very thin, the morphology in Fig.1(b) is therefore almost the same as that of the original α-Al_2O_3 fracture surface.[6] For this percolation system, the clusters may be separated from each other by the air or by the substrate itself. Therefore, the film structure in our experiment is different from the continuous film.

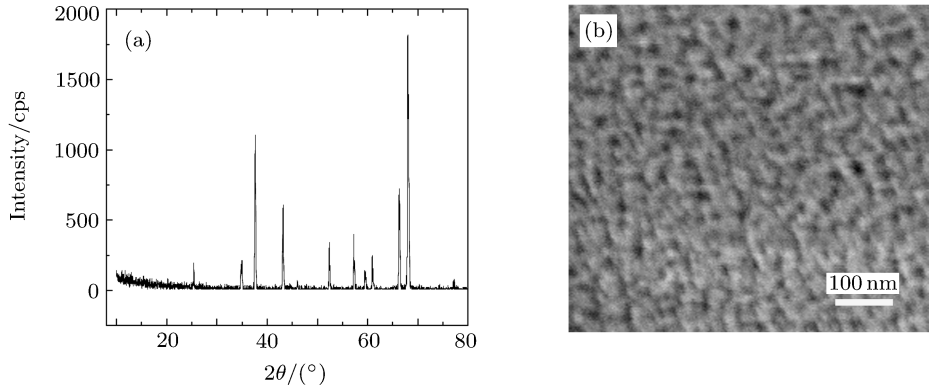


Fig.1. X-ray diffraction pattern (a) and SEM photograph (b) of an as-prepared sample with a thickness d=30 nm.

The hysteresis loops of the iron films with d=40 nm on both the α-Al_2O_3 fracture surfaces and glass surfaces at T=150 K, are shown in Fig.2. We can see that the Fe film is soft magnetized. In comparison with the iron film on the glass surfaces, both the coercivity H_c and the field needed to saturate the sample are enhanced (see the inset of Fig.2). Recently, several studies on the dependence between the surface roughness and the coercivity of the magnetic thin films have been reported.[10,11] It has been shown that the H_c of the films goes up when the surface roughness increases since the domain-wall pinning starts to contribute to

the H_c of the films. In our experiment, the roughness of the film surface is remarkable and so it should lead to the enhancement of the H_c of the film.

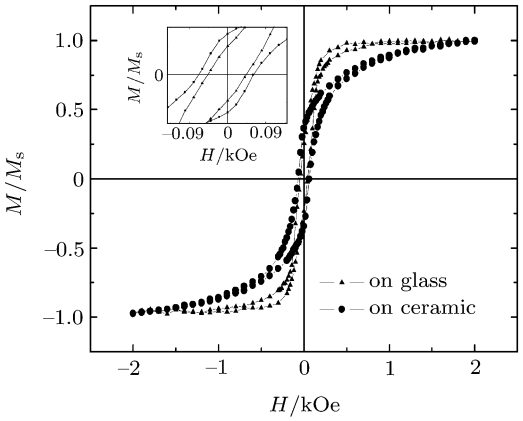


Fig.2. Hysteresis loops of the iron films on both the fracture surface of α-Al_2O_3 ceramics and the glass substrate, for d=40 nm, T=150 K. In the inset: the enlarged curve of the central part of the loops.

In order to investigate the intrinsic magnetic properties of the iron films deposited on the fracture surfaces of α-Al_2O_3 ceramics, the temperature dependence of the zero-field-cooled (ZFC) and field-cooled (FC) magnetization is measured. The ZFC and FC magnetization curves are obtained as follows: after cooling the sample in zero field from 300 to 2 K, a small magnetic field is applied, after that the ZFC magnetic moment M_{ZFC} is recorded at stepwise increasing temperature; subsequently, the sample is cooled to 2 K again in the original field, and the FC magnetic moment M_{FC} is measured in the same way as that of M_{ZFC} measurement. Shown in Fig.3 are the ZFC and FC magnetization curves for an as-prepared sample with a thickness of d=40 nm obtained in different applied fields. It is noteworthy that the M_{ZFC} in Fig.3(a) does not show a maximum and that the ZFC and FC curves remain separated up to 300 K. This M_{ZFC} behaviour in the weak applied field (H=50 Oe) implies that there exists a wide range of size distribution of iron grains in the sample, and some small grains may be superparamagnetic at room temperature.[12] In a higher applied field (H=100 Oe), the ZFC and FC curves diverges around 200 K, as shown in Fig.3(b). In Figs.3(a) and 3(b), there are sharp drops of the M_{ZFC} below a characteristic temperature T_f (e.g., $T_f \approx 52$ K for H=50 Oe and $T_f \approx 33$ K for H=100 Oe), and the difference between the M_{ZFC} and M_{FC} curves increases with reducing temperature. This result indicates that the sample may be in the spin-glass-like state below the characteristic temperature T_f, where a collective freezing of the magnetic moments of the clusters occurs.[13] It also can be seen that the spin-frozen phase at low temperatures is very sensitive to the magnitude of H. The temperature T_f reduces with the increase of H. We will show that this spin-glass-like state may play a crucial role in the hysteresis behaviour of the iron films at low temperatures. For a high field H=500 Oe, however, the abrupt falling behaviour of M_{ZFC} disappears, and the M_{ZFC} and M_{FC} curves are similar, which is in agreement with the behaviour observed in the reentrant spin-glass-like systems.[14,15] It should be pointed out that similar M_{FC} and M_{ZFC} behaviours described above were observed in all the iron film samples with different thickness, however, they were not found in the iron film system on glass surfaces in our experiment.

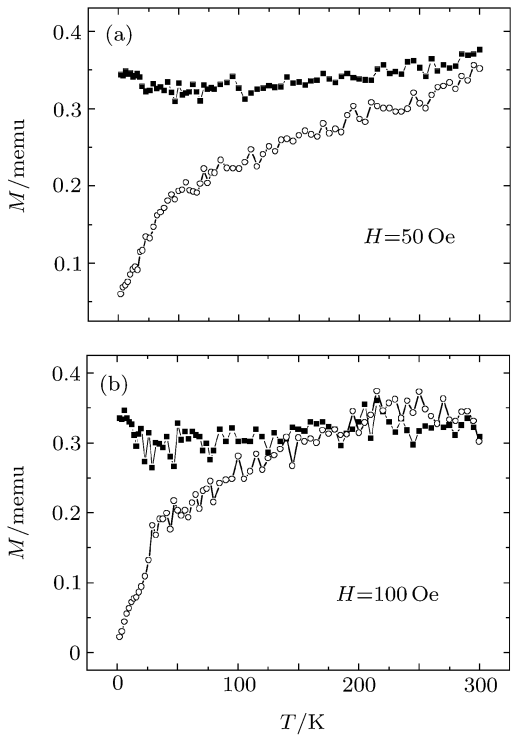


Fig.3. Magnetization as a function of temperature in different magnetic fields after zero-field cooling (open circles) and field cooling (full squares) for an as-prepared sample with a thickness of d=40 nm.

It is well-known that exchange bias phenomena have been observed in the nanoparticle systems,[12,16] the thin ferromagnet (FM)/antiferromagnet (AFM) bilayers [17−19] and the inhomogeneous materials.[20] The defining characteristic is a shift of the hysteresis loop of the FM material from H=0 to a bias field H_e, resulting from the interfacial exchange coupling between the FM phase and AFM phase. The hystere-

sis loops of the iron film samples on the fracture surfaces of α-Al_2O_3 ceramics at different temperatures were measured after cooling from 300 K to a target temperature with a fixed applied field H_{cf}. We observed shifted $M-H$ loops, indicating the presence of oxides on the film surface and also the presence of the exchange interaction between the ferromagnetic iron film and the oxide film surface with spin-glass-like magnetic behaviour at low temperatures.[9] This behaviour is also observed in the iron film system deposited on the liquid surfaces.[9]

Since the cooling field affects strongly the exchange bias phenomena,[21] we then investigate the temperature dependence of the exchange bias field H_e for the sample with d=40 nm in two different cooling fields, i.e., H_{cf}=2 kOe and H_{cf}=20 kOe, as shown in Fig.4(a). Here, the H_e is defined as $H_e=(H_+ + H_-)/2$, where H_+ is the switching field at which M becomes zero as H is swept from negative to positive, and H_- is the switching field as H is swept back to negative. It is noted that the two curves are obviously different: for the low cooling field, H_e is always negative in the whole temperature interval in our experiment, i.e., the loop shifts toward the opposite direction of the cooling field; for the high cooling field, however, H_e changes from negative to positive value as T increases. On the other hand, the behaviours of the two curves are similar at low temperatures, i.e., both are negative and exhibit a valley at $T^* \approx 5$ K, (see Fig.4(a)), indicating that the behaviour of H_e is determined by the coupling between the frozen spins in the oxide film surface and the ferromagnetic spins in the ferromagnetic iron film at low temperatures, and the spins orientation is independent of the cooling field.[12] It should be pointed out that the behaviour of H_e in our experiment is quite different from that of the molecular-beam-epitaxial iron films on GaAs substrates.[22] Therefore, we suggest that the result above may result from the effect of rough substrate surfaces.

A possible explanation for the evolution of the exchange field H_e with temperature at different H_{cf} in Fig.4(a) is the competition between the Zeeman energy of the ferromagnetic iron film in the external magnetic field and the exchange interaction between the ferromagnetic iron film and the oxide film surface.[23] For low H_{cf}, the Zeeman energy cannot overcome the exchange coupling, thus the frozen disordered spins and the ferromagnetic spins are aligned each other, and therefore H_e is negative as temperature T changes. For high H_{cf}, the coupling energy is still larger than the Zeeman energy at low temperatures; however, as T increases and goes beyond a critical value, the exchange coupling energy goes down, and then the Zeeman energy becomes larger than the coupling energy. Therefore, H_e changes from negative to positive as the temperature increases.

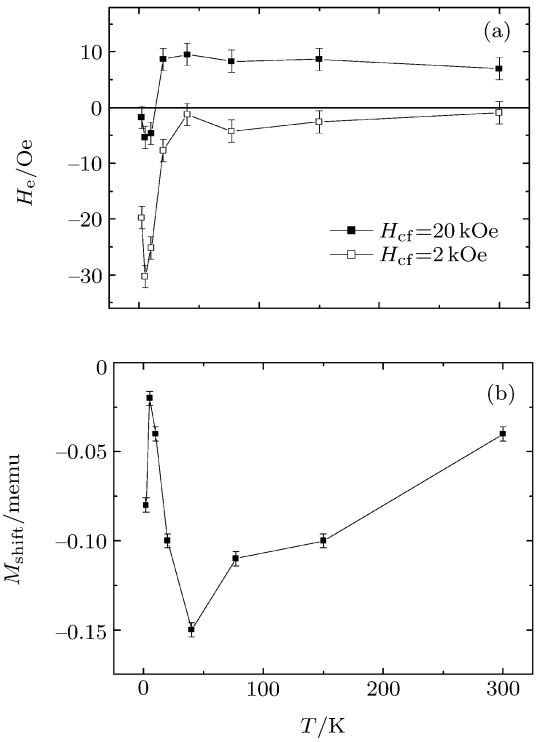


Fig.4. Temperature dependence of (a) the exchange bias field H_e with two cooling fields, i.e., H_{cf}=2 kOe and H_{cf}=20 kOe, and (b) the loop shift M_{shift} with H_{cf}=20 kOe for the iron film sample with d=40 nm.

In addition, the hysteresis loops for H_{cf}=20 kOe shift not only in the field direction, as described above, but in the magnetization axis direction as well. Figure 4(b) shows the temperature dependence of the loop shift in the magnetization axis direction $M_{shift}(T)$, defined as $M_{shift} = (M_+ + M_-)/2$, where M_+ and M_- are the positive and negative magnetizations corresponding to H=0. In Fig.4(b), the saturated moment M_s of the sample is 0.56 memu at 40 K, and the ratio between M_{shift} and M_s is about 26.8%. It is noted that M_{shift} is always negative as the temperature changes, i.e., the hysteresis loop shifts downwards in the whole temperature interval. Nogués *et al* have investigated systematically the dependence of the hysteresis loop shift on the coupling between the FM and the AFM, indicating that if the coupling is antiferromagnetic, a positive exchange bias and a loop shift in the magnetization axis for large cooling fields can be observed.[21−23] Such a phenomenon is also ob-

served in the Fe nanoparticles.[24] Inspired by these results, we suppose that the behaviours of $M_{\rm shift}(T)$ and $H_{\rm e}(T)$ in our experiment may result from the exchange interaction between the ferromagnetic iron film and the oxide film surfaces with spin-glass-like magnetic behaviour at low temperatures. In particular, both $M_{\rm shift}$ and $H_{\rm e}$ reach their maximums at $T \approx$40 K (see Figs.4(a) and 4(b)), suggesting the presence of a strong exchange coupling in the film sample.

Spin-glass behaviour is usually characterized by the ac susceptibility. Figure 5 shows the temperature dependence of the ac magnetic susceptibility of the film sample with d=40 nm, measured at the frequency f=1000 Hz. As the temperature decreases, the real part of the ac magnetic susceptibility, namely χ' in Fig.5(a), drops dramatically around T=52 K, corresponding to the temperature $T_{\rm f}$ in Fig.3(a); then it reaches a minimum value at $T \approx$10K; finally χ' increases continuously. It is noted that the imaginary part of the ac susceptibility, namely χ'', exhibits an obvious maximum peak around $T_{\rm f}$ (Fig.5(b)), then it drops with increasing temperature, and finally it reaches a steady value, i.e., $\chi'' \approx$ 1.6 emu/(Oe·cm^3), which is much larger than the value at low temperatures. This phenomenon is quite different from that of the nanocrystalline iron system and continuous iron films where the steady values of χ'' are almost zero at high temperatures.[25] Here, the values of χ'' should come from both the spin-glass like spins and the ferromagnetic iron spins. The behaviour of the ac susceptibility of the sample is in agreement with the temperature dependence of $M_{\rm ZFC}$ and $M_{\rm FC}$ at low applied field shown in Fig.3(a), implying that the spins are really in the frozen state at low temperatures.[13] In addition, the peak of χ'' shifts to lower temperatures with decreasing frequency, as shown in Fig.5(c). This behaviour demonstrates firmly the presence of the spin-glass-like state.[26]

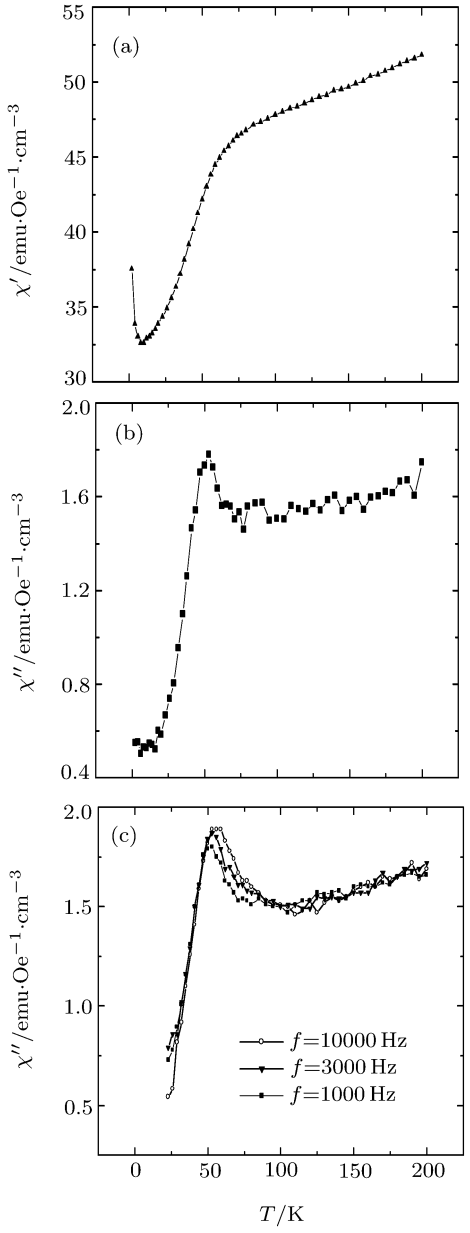


Fig.5. Temperature dependence of the ac susceptibility for the sample with thickness d=40 nm at f=1000 Hz: (a) the real part χ'(T), (b) the imaginary part $\chi''(T)$ and (c) the imaginary part χ'' for different frequencies.

4. Conclusions

In summary, we have fabricated an iron film percolation system on the fracture surfaces of α-Al_2O_3 ceramics successfully. The temperature dependence of the magnetic properties of the film samples is measured.

The hysteresis loop shifts in both the field and magnetization directions are measured: 1) for high cooling field ($H_{\rm cf}$=20 kOe), as the temperature increases, $M_{\rm shift}$ is a negative parameter while $H_{\rm e}$ changes from negative to positive values, indicating the existence of the exchange coupling between the ferromagnetic iron film and the oxide film surface with spin-glass-like magnetic behaviour at low temperatures. 2) For low cooling field ($H_{\rm cf}$=2 kOe), $H_{\rm e}$ is always negative as T changes. While, around T^*=5 K, the curve of $H_{\rm e}$ exhibits a negative valley. The ac magnetic susceptibility of the iron films also exhibits anomalous characteristics: as T decreases, χ' reaches its minimum value at $T_{\rm s} \approx$10 K, and χ'' exhibits a maximum peak around $T_{\rm f}$=52 K. The result corresponds

to the fact that the evolution of the magnetic phase of the samples from a high-temperature ferromagnetic state to a low-temperature spin-glass-like phase has taken place as the temperature decreases.

Our experimental results suggest that the anomalous magnetic properties of the samples may result from the oxide surfaces of the iron film with spin-glass-like spins at low temperature and rough substrate effect. However, here we only present the qualitative study on the relationship between the rough substrate effect and the magnetic properties of the iron films. In order to give the final theoretical conclusion for the physical nature of these experimental phenomena precisely, more quantitative measurements and theoretical studies are still needed.

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