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Experiments and simulations on the magnetization of transparent Co-Fe-Ta-B-O heteroamorphous films

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ABSTRACT
Inducing oxygen into Co-Fe-Ta-B amorphous alloy system makes the transparent amorphous magnetic thin film (Co\textsubscript{20}Fe\textsubscript{47}Ta\textsubscript{20}B\textsubscript{13})\textsubscript{1-x}\textsubscript{O\textsubscript{x}}. Oxygen content plays a major role in the magnetization properties of the films. Samples with x=0.41 have a large coercivity $\sim$1000 Oe, which is 20 times larger than the low oxygen content samples. The forming of a set of single domain magnetic granules rather than continuous magnetic film makes the different magnetization mechanisms. The deviation from Bloch law of saturation magnetization with temperature indicates the Bose-Einstein condensation occurring in magnetic nanostructures. According to the superparamagnetic theory, magnetic phase diagram of the system has been obtained. Monte-Carlo method is used for the simulation of the magnetization of a single domain magnetic granular system at a finite temperature. The simulation results match the experiment to some extent in magnetization-filed curve and field-cooling and zero-field-cooling magnetization curve. This work provides a systematical study of transparent amorphous magnetic thin film system and helps us to know better of this material.

INTRODUCTION
Metallic glass, including amorphous rare-earth and transition-metal alloys, have excellent mechanical properties such as high Young's modulus and fracture strength.\textsuperscript{3,4} Among them, there is a kind of magnetic material with good soft magnetic properties which has applications in surface coating, sensor and transformer core.\textsuperscript{5,6} Co-Fe-Ta-B system is a representative of this material. Recently, researchers made the magnetic thin film transparent by oxygen doping, so that a lot of interesting electric, magnetic and optic properties are generated, which brings it a potential to fabricate a new kind of magnetic semiconductor.\textsuperscript{7,8} Magnetization is the core property of the material both in theory and application, and it is very sensitive to the oxygen content of the films. A few efforts are trying to make the magnetic mechanism clear.\textsuperscript{9,10} However, these works are mostly staying on the surface of the experiment and lacking the theoretical analysis. Here, we systematically investigated the magnetization properties of the (Co\textsubscript{20}Fe\textsubscript{47}Ta\textsubscript{20}B\textsubscript{13})\textsubscript{1-x}\textsubscript{O\textsubscript{x}} system. We found that as the oxygen content changes, there are two kinds of ferromagnetic films. One is the metal-like film, where a continuous metal film exists and a multi-domain structure is formed. The movement of the magnetic domains is the major magnetization mechanism. The other is the granule-like film, where magnetic metal granules with single-domain size are embedded in the weak magnetic matrix. The movement of the magnetic domains is the major magnetization mechanism. The other is the granule-like film, where magnetic metal granules with single-domain size are embedded in the weak magnetic matrix. In this case, a lot of research methods and conclusions can be learned from the earlier works about nano-granular films.\textsuperscript{11,12} The magnetization reversal of single domain granules is the major magnetization mechanism. To further understand the mechanisms, building a proper model and doing a simulation for the system is needed. For the continuous large films with multi-domain structure,
micro-magnetic method can be used because the magnetization is not sensitive to the temperature. However, the magnetic properties with fine granular structure are dramatically related to the temperature due to the thermal fluctuation of spin. In order to simulating magnetic properties in this range, Metropolis algorithm based on Monte Carlo method can be used, which is suitable for simulating a large number of single-domain magnetic granules. Despite the experimental results, this work also provides a model built by a set of single-domain magnetic granules with interactions and gives results matching with the experiments to some extent. Combining with the experimental and simulation results, the magnetization mechanisms of the transparent amorphous magnetic thin films Co-Fe-Ta-B-O is clearer.

EXPERIMENT

\((\text{Co}_{20}\text{Fe}_{47}\text{Ta}_{20}\text{B}_{13})_{1-x}\text{O}_x\) thin films were grown on quartz and SiO\textsubscript{2}/Si wafers by reactive DC magneto sputtering system under a mixture atmosphere with Ar and O\textsubscript{2}. The flow ratio of O\textsubscript{2} and Ar was adjusted from 0 to 0.05 to get different oxygen content in films. Transmission electron microscope (TEM, JEOL 2100) was used to characterize the micro structure and thickness of the films. Element distributions were measured by X-ray photospectroscopy (XPS, Thermo ESCALAB 250Xi). Magnetization properties were measured by the magnetic properties measurement system (MPMS-VSM, Quantum Design).

RESULTS AND DISCUSSION

By doping oxygen into a classical amorphous magnetic alloy system Co-Fe-Ta-B, we can get transparent metal glass which remains its amorphous properties. According to the XPS results in Fig. 1a, as the oxygen content increases, the ratio of the rest elements remains relatively stable. Therefore, samples can be described as \((\text{Co}_{20}\text{Fe}_{47}\text{Ta}_{20}\text{B}_{13})_{1-x}\text{O}_x\). TEM images are shown in Fig. 1b. The thickness of the sample is about 117 nm. As to the microstructures, samples with low \((x=0.14)\) or large \((x=0.52)\) oxygen content have quite uniform amorphous structure. However, percolation patterns have been observed in the sample with \(x=0.41\). This provides an evidence for taking the thin film as a magnetic granular system. The average size of nano-granules is estimated as 2.5 nm and the distance between each granule is \(\sim 1\) nm. The transmittances of the samples are improved by the increasing of oxygen content \(x\) as shown in Fig. 1c. Fig. 1d shows the relationships of the transmittance and the oxygen content under a wavelength of 550 nm. According to the conductivity of the sample, the samples are labeled Metal, Hybrid and Insulator on the optical photos of them, where \(x = 0.14, 0.41, 0.52\), respectively.

\((\text{Co}_{20}\text{Fe}_{47}\text{Ta}_{20}\text{B}_{13})_{1-x}\text{O}_x\) samples have been fabricated from \(x=0.14\) to 0.52 to reveal the evolution of the magnetic properties. First, the \(M-H\) curve at 2 K is measured because it is closer to the ground state of the sample. The results show that there are three types of magnetization curves. As the oxygen content is less than 35%, the samples have low coercivity \(H_c\) around 40 Oe and large squarness ratio \((SQ = M_r/M_s)\), which is close to the properties of bulk magnetic metals. Then, when oxygen content increase to 37% to 45%, \(H_c\) will increase to around 1000 Oe but the SQ is lower. Last, if the oxygen content is larger than 48% the samples will turn into superparamagnetics and the \(M_s\) becomes very low. Fig. 2a shows the different \(M-H\) curves of different samples. The Magnetization of the sample with \(x=0.52\) is very low compared to the samples with lower \(x\). So, the highly oxidized samples are...
considered as the weak magnetic matrix because of the decrease of Fe$^{3+}$ and the increase of Co$^{2+}$ chemical states. Figure 2b is the relationship of $H_c$ and $SQ$ with the oxygen contents at the temperature 2 K. There exists a maximum value of $H_c$ about 1000 Oe which is more than 20 times larger than that of low oxygen content samples but the $SQ$ values decrease monotonically with the oxygen content. Later, the temperature dependence of the magnetization has been measured. Since the magnetic property of the sample with very large oxygen content is poor, the previous two types of samples have been carefully studied. Figure 2c and 2d are the $M$-$H$ curves measured at 2 K, 20 K and 300 K. We found the superparamagnetic appears in the samples with $x$ larger than 0.37, but samples with lower $x$ will remain ferromagnetic even at the room temperature. So that we assume that the magnetic model of the sample is similar as the magnetic granular films. At the scale of a few nanometers, two different amorphous compositions are taken considered in the system. One is the amorphous metal, the other is the amorphous metal oxide. As the oxygen content changes, the samples can be divided into two kinds. One is the metal-like film, when $x<0.35$, the ratio of metal and oxide content is high, amorphous metal is connected as a matrix of the film and amorphous metal oxide granules are embedded into it. The other is the granule-like film, when $x>0.37$, the ratio of metal and oxide is low, amorphous metal are divided into small granules and embedded into the amorphous metal oxide matrix. The magnetization of the metal-like samples is mainly depended by the motion of the domain wall so that the $H_c$ is as low as the bulk Co-Fe-Ta-B metal, while the granule-like samples are formed by a set of single domain granules so that the magnetization is dependent on the anisotropic energy of the nanometer-sized granules.

For ultrafine magnetically granules, there exists a critical size below which the granules can acquire only single magnetic domains even in zero magnetic field. In amorphous material, it is easier for the granules to reach the single domain size. At low temperatures, the magnetization of the granules can hardly overcome the barriers formed by anisotropic energy, thereby the ferromagnetism is achieved. At sufficiently high temperatures, the barriers can be overcome by thermal fluctuation, thereby superparamagnetism occurs. The superparamagnetic relax can be described by the Arrhenius Law. During a measurement time, if the relaxation process is finished, superparamagnetic behavior can be measured. The critical transition temperature of ferromagnetic and superparamagnetic is called the blocking temperature $T_B$. In experiment, the $T_B$ can be obtained by the maximum of magnetization in a zero-field-cooldown (ZFC) curve (Fig. 3a). Samples with $x>0.37$ get a $T_B$ which decreases with the oxygen content due to the smaller size of the amorphous metal granules in higher oxygen content samples, combining with the Curie temperature $T_C$, magnetic phase diagram of the system can be obtained (Fig. 3b). Despite the blocking temperature, thermal fluctuation also affects the magnetization properties such as $M_s$ (Fig. 3c), $H_c$ and $SQ$ (Fig. 3d). The saturated magnetization $M_s$ is decreasing with the temperature, in normal magnetic metals, the relationship of the $M_s$ and the temperature obeys the Bloch law, $M = M_s(1-BT^{3/2})$. However, in this system, there is a deviation from the Bloch law. This have also been observed in other nano-structured magnetic system and explained by the Bose-Einstein condensation of the magnons. The result is a subtle upturn of the magnetization curves of ferromagnetic nano-particles in the 10 to 50 K temperature range. Furthermore, we built a model in order to use Monte-Carlo method to carry out some results. The model is a system with a certain number of spherical granules. Based on the TEM images in Fig. 1c, the distribution of the diameters and distances are taken...
as log-normal distribution and normal distribution with an average value of 2.5 nm and 1 nm as shown in Fig. 4a. Six nearest neighbors are considered when calculating the exchange interaction between granules. Periodic boundary condition is applied for those granules at the boundary of the calculating cell. The total energy ($E$) of the granule system is given as

$$E = -\sum_i m_i \cdot H - \sum_i K V_i (\vec{r}_i \cdot \vec{m}_i)^2 - \frac{1}{2} \sum_{i,j} m_i \cdot m_j + D \sum_{i,j} \frac{m_i \cdot m_j - 3 (m_i \cdot \vec{R}_{ij})(m_j \cdot \vec{R}_{ij})}{R_{ij}^3},$$

(1)

FIG. 3. (Experiment results) (a) FC and ZFC curves of sample $x=0.41$. (b) Magnetic phase diagram where FM is ferromagnetic, SPM is superparamagnetic and PM is paramagnetic. (c) $M_s$ and $T$ relationship of samples $x=0.14$ and $0.41$. (d) $H_c$ and $S_Q$ relationship with temperature.

FIG. 4. (Simulation results) (a) Schematic of amorphous granular system model and the distribution of diameters and distances of the granules. (b) Flow chart of Metropolis algorithm. (c) $M$-$H$ curve under different temperature. (d) ZFC-FC curve under a measurement field 100 Oe.
where $m_i$, $\vec{e}_i$, $V_i$ are the magnetic moment, easy axis direction and the volume of the $i_{th}$ granule. $H$ is the external magnetic field, $R_{ij}$ is the location vector between two granules. The four terms are the Zeeman energy, the anisotropic energy ($K$ is the anisotropic constant), the exchange interaction energy ($J$ is the exchange interaction constant) and the magnetostatic interaction energy ($D$ is the magnetostatic interaction constant). The flow chart of the Metropolis algorithm is shown on Fig. 4b. By adjusting parameters (netostatic interaction constant). The flow chart of the Metropolis algorithm is shown on Fig. 4b. By adjusting parameters (netostatic interaction constant). The simulation result of blocking temperature is 66 K, which is close to the experiment result of 75 K. In this model, we can investigate the work of each energy term and know how they work in the magnetic granular system. The difference of this model with the micro magnetic model is that the micro magnetic program usually just works at zero temperature but Monte-Carlo method can work at finite temperatures, which is suitable for the magnetic granular systems and the amorphous magnetic systems.

CONCLUSION

In conclusion, we have systematically investigated the magnetization process of the transparent amorphous magnetic material system ($\text{Co}_{20}\text{Fe}_{75}\text{Ta}_{20}\text{B}_{15}$)$_{1.3}$O$_x$. There are two kinds of magnetization mechanisms; one is the domain wall motion based magnetization in metal-like films. The other is the magnetization reversal of single domain granules in granule-like films. Based on the analysis of the temperature dependent magnetization, magnetic phase diagram of the system has been obtained. The Bose-Einstein condensation in magnetic nanostructures makes the deviation of the Bloch law. A simulation based on Monte-Carlo method can be achieved by building a proper model of the granular system and the simulation results can match with the experimental results to some extent. This work helps us know better about the transparent amorphous magnetic metal.

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