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Spin-valve magnetoresistance in single-phase $\varepsilon$-Fe$_{2-\varepsilon}$N film

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Dear editor,

$\varepsilon$-Fe$_{2-\varepsilon}$N has been investigated as a potential candidate in spintronics devices [1]. With the changing ratio of Fe:N, the magnetic behaviors and the spin transport properties of $\varepsilon$-Fe$_{2-\varepsilon}$N can be manipulated [2,3]. In particular, the Curie temperature changes from 9 K for $\varepsilon$-Fe$_2$N to 757 K for $\varepsilon$-Fe$_3$N, and the room temperature spin-polarization can be controlled between 0 to 0.5 with different Fe:N ratio [4]. Therefore, $\varepsilon$-Fe$_{2-\varepsilon}$N provides an advanced platform for the design of spintronics devices such as magnetic tunnel junction or spin-based field effect transistor.

$\varepsilon$-Fe$_{2-\varepsilon}$N films are grown on Si(111) by molecular beam epitaxy (MBE) using AlN/3C-SiC intermediate layers, and the magnetic tunnel junction (MTJ) structure of $\alpha$-Fe/AlN/Fe$_3$N is realized [5,6]. Also, nanocrystalline $\varepsilon$-Fe$_3$N films are deposited on glass by magnetron sputtering and an enhancement of extraordinary Hall coefficient is found [7]. Therefore, $\varepsilon$-Fe$_{2-\varepsilon}$N is considered as a promising candidate material for the investigation of intrinsic anomalous Hall effect. Our group has grown $\varepsilon$-Fe$_{2-\varepsilon}$N film on GaN and Al$_2$O$_3$ by metalorganic chemical vapor deposition (MOCVD) or magnetron sputtering. Single-phase $\varepsilon$-oriented $\varepsilon$-Fe$_{2-\varepsilon}$N films have been synthesized and the structural and magnetic properties have been investigated [8,9].

In this study, we report magneto-transport phenomena in $\varepsilon$-Fe$_{2-\varepsilon}$N films. The spin-valve magnetoresistance (MR) effect is observed in ferromagnetic single layer $\varepsilon$-Fe$_{2-\varepsilon}$N film with $c$-oriented single phase. Both of the two $\varepsilon$-Fe$_{2-\varepsilon}$N film samples are grown on $c$-Al$_2$O$_3$ substrate by magnetron sputtering system. The sputtering is performed by applying the DC power (100 W) to a Fe (99.9%) target and changing flow rate ratio of N$_2$/Ar gas. The Fe:N ratio in the achieved $\varepsilon$-Fe$_{2-\varepsilon}$N samples can be estimated according to X-ray diffraction spectra. Detailed experimental procedures are described in our previous study [9]. $\varepsilon$-Fe$_{2.95}$N and $\varepsilon$-Fe$_{2.28}$N films show single-phase $c$-oriented and metallic luster with the thickness of about 45 nm.

Figure 1(a) shows $M-H$ curves of $\varepsilon$-Fe$_{2.95}$N and $\varepsilon$-Fe$_{2.28}$N. Obvious hysteresis loop is observed for $\varepsilon$-Fe$_{2.95}$N while very weak hysteresis behavior is observed for $\varepsilon$-Fe$_{2.28}$N in the enlarged image. This is ascribed to the reducing of ferromagnetic Fe-Fe exchange coupling. Comparing with $\varepsilon$-Fe$_{2.95}$N, more N atoms participate in $\varepsilon$-Fe$_{2.28}$N, and the super-exchange interactions between Fe sites mediated by N atoms could be enhanced which reduces the magnetization of the film.

Figure 1(b) shows MR ratios $(R_H - R_0)/R_0$ of $\varepsilon$-Fe$_{2.28}$N with changing magnetic field from $-20000$ Oe to $20000$ Oe under different temperatures. The magnetic field is perpendicular to the surface of the film and the data are achieved by four probe method. It can be seen that MR ratios increase monotonically with increasing magnetic filed from 0 Oe to $\pm 20000$ Oe. More importantly,
increases to 150 K. The MR ratio almost disappears when the temperature decreases to 3 K. At the meanwhile, the MR ratio can increase up to 0.7% when the temperature increases to 150 K.

Figure 1(c) is MR ratio of ε-Fe$_{2.95}$N with magnetic field ranging from −20000 Oe to 20000 Oe when the temperature is 50 K. As shown in Figure 1(c), spin-valve MR is observed. The maximum values of MR ratio are achieved when the magnetic fields are around ±5000 Oe. We think the appearance of spin-valve MR in single layer ε-Fe$_{2.95}$N film is related to the scattering effect of spin-polarized electrons through the boundaries between ferromagnetic ε-Fe$_{2.95}$N grains. As shown in the inset TEM image of ε-Fe$_{2.95}$N, grain boundaries can be observed and this may be owing to island growth mechanism or misfit dislocations. The MR ratios of ε-Fe$_{2.95}$N under different temperatures are shown in Figure S1. Clear spin-valve MR are observed when the temperature is 3, 12 and 50 K. However, the effect cannot be detected when the temperature increases to 150 K. In order to get further information about magnetic properties of ε-Fe$_{2.28}$N and ε-Fe$_{2.95}$N, field cooled (FC) and zero field cooled (ZFC) temperature dependences of the magnetization are measured from 2 K to 300 K as shown in Figure S2. For ε-Fe$_{2.28}$N, almost no FC-ZFC signal is observed which is owing to the weak magnetic behaviors as shown in Figure 1(a). For ε-Fe$_{2.95}$N, FC magnetization decreases monotonously with the increasing temperature while ZFC curve shows a cusp-like behavior. We can see the transition temperature of magnetic state locates at around 50 K. It is suggested that the magnetic state transition may be ascribed to competition of thermal energy and magnetic exchange interaction, especially the interaction between the adjacent magnetic domains.

Furthermore, the temperature dependences of resistivity are recorded as shown in Figure 1(d). The resistivity of ε-Fe$_{2.28}$N or ε-Fe$_{2.95}$N increases monotonically with increasing temperature from 2 K to 300 K which indicates a similar metallic transport property. Particularly, the resistivity variation with changing temperature from 2 K to 50 K is stronger for ε-Fe$_{2.28}$N comparing to ε-Fe$_{2.95}$N while the resistivity presents the similar behavior when the temperature is above 50 K. It is suggested that the temperature dependences of resistivity are also related to spin-polarized transport because the tendency is inconsistent with the relation of magnetoresistance and temperature.

In conclusion, spin-polarized magneto-transport phenomena in ε-Fe$_{2-n}$N films are observed. More importantly, the spin-valve magnetoresistance effect is observed in ferromagnetic single layer ε-Fe$_{2-3}$N film with c-oriented single phase. We think the spin-valve MR in single layer ε-Fe$_{2-3}$N film is related to the scattering effect of spin-polarized electrons between the adjacent magnetic domains.
film is related to the scattering effect of spin-polarized electrons through the boundaries between ferromagnetic $\varepsilon$-Fe$_2$-$\sim$3N grains. This should be of great help for designing advanced spin-related devices such as magnetic random access memory or spinFET.

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Supporting information Figures S1 and S2. The supporting information is available online at info.scichina.com and link.springer.com. The supporting materials are published as submitted, without typesetting or editing. The responsibility for scientific accuracy and content remains entirely with the authors.

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