Barkhausen-like antiferromagnetic to ferromagnetic phase transition driven by spin polarized current

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We provide clear evidence for the effect of a spin polarized current on the antiferromagnetic to ferromagnetic phase transition of an FeRh wire at Co/FeRh wire junctions, where the antiferromagnetic ground state of FeRh is suppressed by injecting a spin polarized current. We find a discrete change in the current-voltage characteristics with increasing current density, which we attribute to the Barkhausen-like motion of antiferromagnetic/ferromagnetic interfaces within the FeRh wire. The effect can be understood via spin transfer, which exerts a torque to the antiferromagnetic moments of FeRh, together with non-equilibrium magnetic effective field at the interface. The conclusion is reinforced by the fact that spin unpolarized current injection from a nonmagnetic Cu electrode has no effects on the antiferromagnetic state of FeRh. © *2015 AIP Publishing LLC*.
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Though there has been a tremendous advance in crafting new spintronic devices using ferromagnetic (FM) nanostructures, an opportunity of controlling antiferromagnetic (AFM) states should offer another route for delivering a broad range of new technologies in spintronics. Current way of controlling FM nanostructures relies in essence on spin transfer (ST) torque which impinges on the magnetization orientation of FM elements so as to satisfy the conservation of the total spin angular momentum of electrons and magnetic moments, as spin polarized electrons traverse the interface between two ferromagnetic layers with different orientations.[1–6] The ST has, in fact, been demonstrated, allowing for not only magnetization switching[4,5] and domain wall motions[7] but also generating microwave oscillations.[8] Similar to ferromagnets, ST plays an important role in AFM materials.[9–11] Recent theoretical work has reported that spin flop transitions of the AFM vector, $\boldsymbol{I} = \boldsymbol{M}_1 - \boldsymbol{M}_2$, where $\boldsymbol{M}_1$ and $\boldsymbol{M}_2$ are the sublattice magnetizations, could be induced by precessional instability of the AFM vector even at two orders of magnitude smaller current densities, given ST together with a magnetic field in the FM/AFM heterostructures.[9] Partial or complete alignment of the sublattice magnetizations of an AFM layer was also predicted in a dual FM/AFM/FM spin valve structure,[10] while a phenomenological approach showed that AFM domain walls acquire a net magnetization under spin injection conditions.[12] The flow of an electric current across FM heterostructured interfaces, on the other hand, generates non-equilibrium magnetization, i.e., the spin splitting in the electrochemical potentials of electrons with opposite spins, called spin accumulation, due to the abrupt change in the spin dependent conductivities.[13–16] Such non-equilibrium magnetization acts as an effective magnetic field on a magnetic state, causing the magnetization modulation or even the magnetic phase transitions at FM/metamagnetic interfaces.[17] In spite of such stimulating theoretical predictions, there have been only a few reports *indirectly* observing the effect of a spin polarized current on the AFM states exclusively in exchange biased spin valve structures.[11,18–20]

It is well known that ordered FeRh alloys with the CsCl structure show anomalous magnetic properties including the first-order magnetic phase transition from the AFM to FM states at around 370–380 K,[21–26] accompanied by a large reduction in the resistivity[27] and an isotropic volume expansion of about 1%.[28] The capability to control the staggered moments[29] and the magnetic states via magnetoelastic coupling in FeRh/ferroelectric heterostructures[30,31] also has attracted much interest for its potential applications in spintronics as well as multiferroics. The AFM and FM states are energetically close to each other, and the balance between these states causes the AFM to FM phase transition in thermal heating process.[32] Such a subtle balance between the two magnetic states should likely induce the magnetic transition, if possible ST and non-equilibrium magnetization due to spin polarized current injection could alter the magnetization orientation of the local moments as well as the exchange interactions between local magnetic moments and conduction electrons in the AFM state. Recently, we reported a pioneering finding of the effect of a spin polarized current on the AFM to FM phase transition of FeRh with a Co wire array structure.[33] However, the effect was not clear enough to prove the spin related effect. Here, we demonstrate convincing evidence for the spin polarized current induced AFM-FM phase transition of FeRh in a single Co/FeRh junction structure. Detailed mechanisms of the spin polarized current induced phase transition are discussed.

Since the AFM and FM states of FeRh show a marked difference in the resistivity, our approach for detecting the effect of a spin polarized current is to measure the resistivity under spin injection conditions from a Co wire. For this purpose, Co/FeRh wire junction devices were fabricated by electron beam lithography and Ar ion milling from epitaxial 50–100 nm thick FeRh thin films as shown in Fig. 1(a). FeRh films were grown onto a $SrTiO_3$(001) (STO) substrate at 530 °C using magnetron sputtering from a single alloy target

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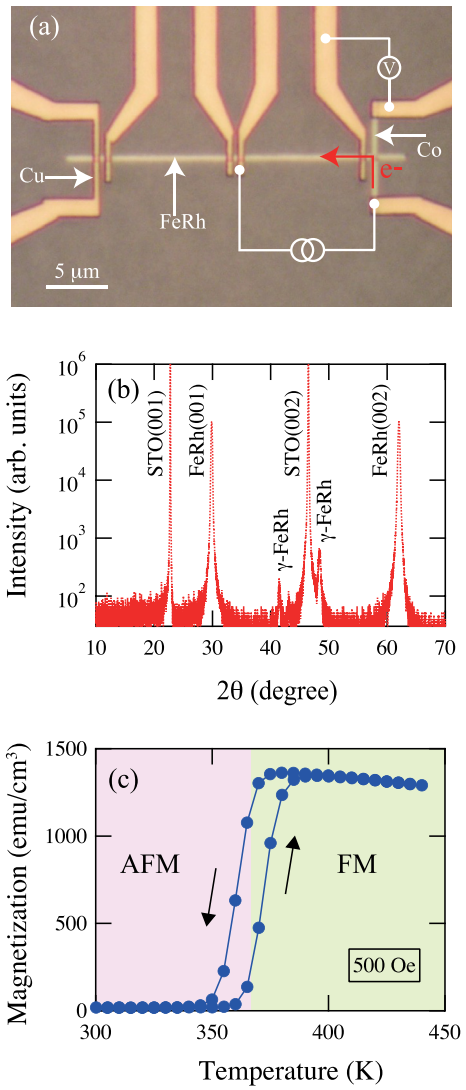

FIG. 1. (a) Optical microscope image of a Co/FeRh and Cu/FeRh wire junction device, where an electron current is injected from Co or Cu into FeRh. The distance between the Co(Cu) source electrode for spin injection and the Cu electrode for voltage detection is 500 nm. (b) X-ray diffraction pattern and (c) temperature dependence of magnetization measured at 500 Oe of an FeRh thin film. A typical AFM-FM phase transition is observed with thermal hysteretic behavior.

in a chamber with a base pressure under 9×10^{-6} Pa. Note that we selected STO as a substrate for growing FeRh films to prevent electrical charging effect in the lithography process although MgO(001) is used, in general, because of the small lattice mismatch between FeRh and MgO. Prior to the fabrication process, the FeRh films were characterized using X-ray diffraction (XRD) in a θ–2θ scanning geometry. (001), (002), and (003) reflections from FeRh were clearly seen in the XRD pattern (Fig. 1(b)), guaranteeing its (001) orientation and the well-ordered CsCl structure. Temperature dependent magnetization measured in a field of 500 Oe ensures the phase transition from the AFM state at room temperature to the FM state at around 370 K (Fig. 1(c)). Thermal hysteresis of the magnetization is typical of a first-order phase transition, compatible with the previous reports,[27] whereas a relatively wider thermal hysteresis in this study should be attributed to lattice distortion induced by a large lattice mismatch of about 8.2% between FeRh and STO.[34] FeRh films characterized this way were patterned into a 500 nm wide wire structure followed by the deposition of a 70 nm thick Co electrode with a width of 500 nm in another molecular beam epitaxy chamber. A Cu/FeRh junction was also made at the other end of the same FeRh wire in order to detect a reference signal from the non-magnetic electrode as shown in Fig. 1(a). The resistivity and current-voltage (I-V) characteristics of FeRh were measured using a four-terminal method at temperatures from 300 to 500 K.

Figure 2 shows the temperature dependence of resistivity measured at different current densities across both Co/FeRh and Cu/FeRh interfaces. To initialize the magnetic domain structure of the Co wire, we apply a magnetic field large enough to saturate the magnetization along the wire axis. A clear drop is seen at around 380 K in the heating process, associated with the AFM-FM phase transition. At a low current density of 8×10^4 A/cm^2, the temperature dependence of the resistivity for both interfaces coincides with each other, showing no significant effects of a spin polarized current on the magnetic state of FeRh. On the other hand, we note that a significant deviation of the resistivity at the Co/FeRh interface from that measured at the non-magnetic Cu/FeRh interface is seen in the AFM state at a higher current density as shown in Figs. 2(b) and 2(c). The reduction in the resistivity of the AFM state for the Co/FeRh interface is a clear manifestation of the instability of the AFM state of FeRh while flowing a current. Special care has to be taken to verify the spin polarized current effect since Joule heating effect near the interface may not be negligible. However, note that current induced Joule heating should be comparable for both interfaces since the resistivities of Co and Cu are 8×10^{-6} and 2×10^{-6} Ω cm, respectively, both of which are two orders of magnitude smaller than that of FeRh

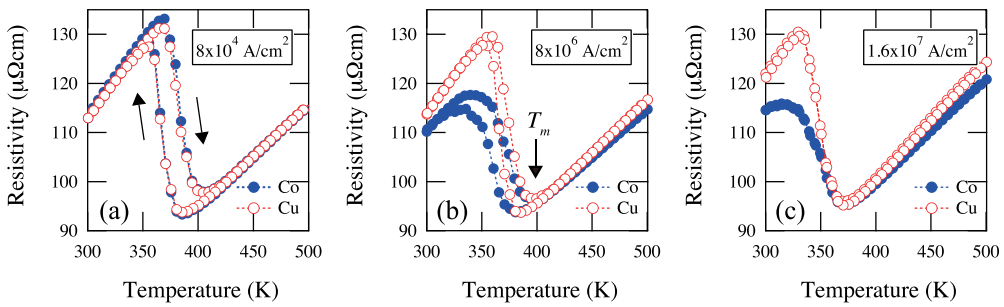

FIG. 2. Bath temperature dependence of the resistivity of an FeRh wire measured with different current densities flowing across Co/FeRh and Cu/FeRh interfaces. The dotted lines are guide of the eyes. The noticeable difference between the resistivities measured at the Co/FeRh and Cu/FeRh interfaces is associated with the AFM-FM transition induced by a spin polarized current.

($6 \times 10^{-4}\,\Omega$ cm). Moreover, the temperatures showing the minimum of the resistivity, T_m, where the AFM-FM transition completes, are the same for both Co/FeRh and Cu/FeRh interfaces, proving that possible heating effects are almost comparable. Therefore, we exclude Joule heating effect as a possible cause for the deviation between the resistivities at both the interfaces and it should be due to spin-related effect occurred at the Co/FeRh interface, although Joule heating effect causes a decrease in T_m with increasing current density.

I-V curves at the Co/FeRh and Cu/FeRh wire interfaces provide another aspect of the spin polarized current induced instability of the AFM state as shown in Figs. 3(a) and 3(b). The voltage increases linearly with increasing current for both the interfaces at low current densities below 10^6 A/cm^2 at 320 K, while a characteristic feature appears at a critical current density of about 2×10^7 A/cm^2. The negative differential resistivity of the non-magnetic Cu/FeRh interface above the critical current density arises from the AFM-FM phase transition due to Joule heating effect. For the Co/FeRh interface, on the other hand, the differential resistivity as a function of current manifests itself in a *discrete* manner at a lower current density I_B. The discrete change in the *I-V* curve is reminiscent of a Barkhausen-like movement of the AFM/FM interface in FeRh. Using the *I-V* curve in Fig. 3(a), we estimate the position of the AFM/FM interface on the assumption that the discrete changes in the *I-V* curves are due to the interface movement as shown in Fig. 3(c). Such features completely disappear and the *I-V* varies smoothly as the bath temperature is raised to the FM state of FeRh at 400 K.

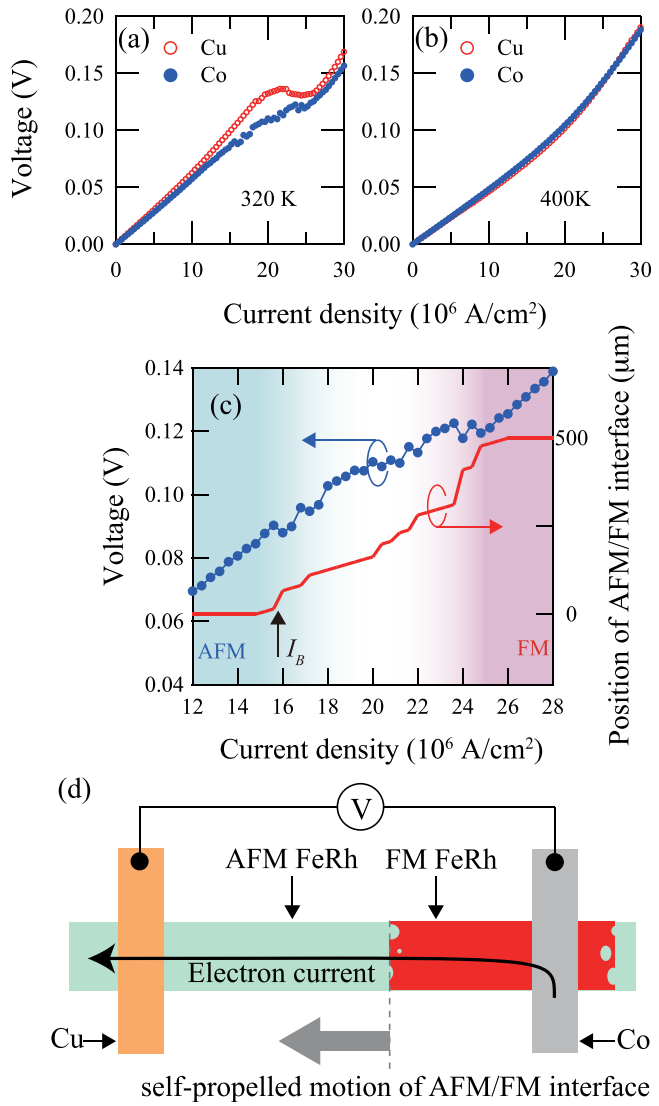

FIG. 3. *I-V* curves at Co/FeRh and Cu/FeRh wire interfaces measured at (a) 320 K and (b) 400 K. A discrete change in the *I-V* curves for the Co/FeRh interface above $I_B = 1.5 \times 10^7$ A/cm^2 is associated with a Barkhausen-like movement of the AFM/FM interface of FeRh. (c) An enlarged view of the *I-V* curve for the Co/FeRh interface and the estimated position of the AFM/FM interface are shown. (d) Schematic diagram of self propelled movement of the AFM/FM interface.

We now discuss the mechanisms explaining how a spin polarized current causes the instability of the AFM state of FeRh. Recent theoretical work has shown that ST greatly influences AFM spin alignment and even a net magnetization can be generated in the AFM state by partially or completely aligning the sublattice magnetizations, as spin polarized electrons are injected across the FM/AFM interfaces.[10,35] Analogous to FMs, ST to AFM sublattice magnetizations affect the dynamics as described in the Landau-Lifshitz-Gilbert equation

$$\frac{\partial \boldsymbol{M}_{1(2)}}{\partial t} = -\frac{\omega}{M_{1(2)} M_{FM}} \boldsymbol{M}_{1(2)} \times \left(\boldsymbol{M}_{1(2)} \times \boldsymbol{M}_{FM}\right) - \gamma \left[\boldsymbol{M}_{1(2)} \times \boldsymbol{H}^{eff}_{1(2)}\right], \quad (1)$$

where γ is the gyromagnetic ratio, $\boldsymbol{H}^{eff}_{1(2)}$ is the effective magnetic field including the external field, the anisotropy field, and the non-equilibrium exchange field, which acts on the sublattice magnetizations $\boldsymbol{M}_{1(2)}$, $\boldsymbol{M}_{FM}$ is the magnetization of an FM injection electrode, and ω is the damping frequency.[3] On the basis of Eq. (1), the ST initially induces oscillatory instability of the strong coupled AFM sublattice magnetizations and spin flop transition occurs with increasing current density as a spin polarized current is injected.[9] More interestingly, relatively low current densities ranging below 10^6 A/cm^2 could generate a magnetization in the AFM layer of an FM/AFM/FM spin valve structure,[10] hence we posit that a similar mechanism induces the AFM-FM transition in FeRh at the interface. Another contribution we need to consider is non-equilibrium exchange interaction between a spin current generated by Co and local moments in FeRh.[3] Within the spin diffusion length in FeRh, non-equilibrium exchange field is induced via a local *s-d* exchange interaction between the spin current and local moments. Since the non-equilibrium exchange field is included in $\boldsymbol{H}^{eff}_{1(2)}$ of Eq. (1) as the leading order contribution, it assists the instability of the AFM state of FeRh or even inducing the AFM-FM transition.

While we have discussed the ST effect so far, the difference in the electrochemical potentials for electrons with opposite spins, i.e., non-equilibrium magnetization $\Delta\mu = \mu_\uparrow - \mu_\downarrow$, can also be responsible for the instability of the AFM state. A theoretical study by Zyuzin and Zyuzin has shown that this effect drives a metamagnetic transition at the FM/metamagnet interface,[17] provided that the effective magnetic field due to a non-equilibrium magnetization at the interface exceeds the critical magnetic field of the metamagnetic transition. We

estimate $\Delta\mu$ for a Co/FeRh interface using the following equation given by van Son *et al.*:[14,16]

$$\Delta\mu = eJ\frac{2(2P-1)(\gamma_{FeRh}L_{FeRh})(\gamma_{Co}L_{Co})}{(\gamma_{Co}L_{Co})+4P(1-P)(\gamma_{FeRh}L_{FeRh})}, \qquad (2)$$

where P is the conductivity polarization of Co, $\gamma_{FeRh(Co)}$ is the conductivity of FeRh(Co), $L_{FeRh(Co)}$ is the spin diffusion length of FeRh(Co), e is the electron charge, and J is the current density. With acceptable parameters $P = 0.37$ for Co, $\gamma_{Co} = 8 \times 10^{-6}\ \Omega$ cm, $\gamma_{FeRh} = 1.1 \times 10^{-4}\ \Omega$ cm, $L_{Co} = 5$ nm, and $L_{FeRh} = 0.5$ nm, we obtain $\Delta\mu = 1.3 \times 10^{-5}$ eV, yielding an effective magnetic field of 1.0 kOe. Because we showed in our previous publication that the magnetic field needed for inducing the instability of the AFM state is larger than 2 T at around 320 K,[36] the estimated value is not large enough to be an alternative substantial mechanism of the spin polarized current induced AFM-FM transition in FeRh.

Although ST in combination with non-equilibrium exchange interaction could induce the AFM-FM transition, the effect is likely limited within the spin diffusion length of FeRh on the order of 1 nm from the Co/FeRh interface; such a short spin diffusion length cannot account for a coherent phase transition over the whole region that we measure the resistivity. However, once the phase transition occurs at the Co/FeRh interface, a *new* FM/AFM interface is formed within the FeRh and non-equilibrium exchange interaction and/or non-equilibrium magnetization appear at the new FM/AFM interface within FeRh, besides at the original Co/FeRh interface. The movement of the FM/AFM interface due to this Barkhausen-like self-propelled transition can spread the region of the FM state of FeRh with increasing current density (see Fig. 3(d)). Such self-propelled effect of metamagnetic transitions due to a non-equilibrium magnetic field at FM/metamagnetic interfaces has been predicted in a theoretical study.[17] From these examination, we consider that the discontinuous Barkhausen-like change in the *I-V* curve of FeRh shown in Fig. 3(a) is in reasonable accord with the self-propelled transition.

It is insightful to comment on another intriguing feature in Fig. 2 that the thermal hysteresis of the resistivity at higher current densities becomes less significant and even disappears at a current density of 1.6×10^7 A/cm^2. No thermal hysteresis indicates that the first-order AFM-FM transition of FeRh changes to the second-order transition without accompanying latent heat. Since the feature occurs irrespective of whether the interface electrode is magnetic or non-magnetic, neither spin transfer with non-equilibrium exchange field nor non-equilibrium magnetization could dominate the feature. One possible cause is momentum transfer to atoms from an electron current, i.e., electron wind force,[37] thereby electrons drive the atom in the direction of the electron current flow as a recoil effect of the scattering of electrons by the atom. Since the phase transition in FeRh is driven by a combination of *s-d* exchange interaction and magneto-volume effect,[32] the first-order phase transition might be smeared by the migration of atoms occurred in a high current density. A similar current induced lattice distortion was reported in epitaxial manganite thin films.[38] While we tentatively attribute this effect to the electron wind force, the details are not yet clear.

In conclusion, we have studied the spin polarized current effect on the AFM to FM phase transition of FeRh in Co/FeRh wire junction device, demonstrating that the spin polarized current can induce instability of the AFM state of FeRh. ST effect with non-equilibrium exchange field via *s-d* exchange interaction contributes to the current induced magnetic phase transition. A Barkhausen-like self-propelled movement of the AFM/FM interface within FeRh has allowed for detecting the AFM/FM transition by electric measurements even if the spin diffusion length in FeRh is on the order of 1 nm. These results appeal that injecting a spin polarized current can be a potential means of controlling the AFM-FM magnetic phase transition of FeRh.

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