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Exchange coupling between a magnetoelectric (111)-oriented Cr_2O_3 single crystal and a CoPt multilayer with perpendicular anisotropy exhibits an exchange bias field proportional to the applied axial electric field. Extrapolation from bulk to thin film magnetoelectric pinning system suggests promising spintronic applications due to coupling between the electric field-controlled magnetization and the magnetization of a neighbor ferromagnetic layer. Pure voltage control of magnetic configurations of tunneling magnetoresistance spin valves is an attractive alternative to current-induced magnetization switching. © *2005 American Institute of Physics*.
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Magnetic read-heads and sensors are the trailblazers in the emerging field of spintronics involving giant magnetoresistance (GMR) (Refs. 1–3) and tunnel magnetoresistance (TMR) effects.[4] Evolution beyond passive magnetoelectronic components is envisioned in the next generation of spintronics devices, which should combine memory and logical functions in order to set new standards in future information technology.[5] Recently, there has been growing interest in studying magnetization reversal involving spin transfer from a spin-polarized current injected into the device as an alternative to stray magnetic fields for switching the magnetic configurations in GMR or TMR devices.[6,7] The required huge current densities hamper the technical realization of this attractive concept. We propose to use the electric field as alternative means for controlling the magnetic configuration of magnetoresistive systems. A magnetoelectric (ME) component is the key new component in the device.

In a ME material, an electric field E induces a magnetic moment $\underline{M}=\underline{\underline{\alpha}}\underline{E}$, where $\underline{\underline{\alpha}}$ is the tensor of ME susceptibility. The antiferromagnetic (AF) phase of Cr_2O_3 is a prototypical example of magnetoelectricity.[8–11] An electric field applied along the (111)-direction of Cr_2O_3 lifts the degeneracy of the AF sublattice magnetization and induces a net magnetic moment along the (111)-direction, known as the parallel ME effect. In this paper, we show that the electrically-controlled magnetization in a ME film can be used to modify the magnetization properties of an adjacent ferromagnetic (FM) film. Taking advantage of the antiferromagnetic nature of the ME film, we use the exchange coupling at the AF/FM interface to create an exchange bias proportional to the external electric field. A $Cr_2O_3(111)$/Pt 0.7 nm/[Co 0.3 nm Pt 1.2 nm$]_3$/Pt 3.1 nm heterostructure is used as a model system. The Pt seed layer of 0.7 nm thickness represents our first attempt to mediate between structural and magnetic optimization of the heterostructure. On the one hand, it is known that a Pt layer of 0.5 and 0.2 nm maximizes the EB field in IrMn/(Pt/Co$)_n$ and FeMn/(Pt/Co$)_n$, respectively.[12] On the other hand, one has to avoid the documented formation of CoO at the AF/FM interface.[13] Figure 1 shows a typical perpendicular hysteresis loop m vs H of the heterostructure measured at $T=150$ K and zero electric field ($E=0$) after cooling to below $T_N=307$ K in $\mu_0 H=0.5$ T. Subtraction of the AF background reveals the rectangular FM hysteresis with a small conventional EB, $H_E \approx 0.8$ mT, of the CoPt multilayer (Fig. 1, inset a) indicating perfect perpendicular magnetic anisotropy of the FM film.

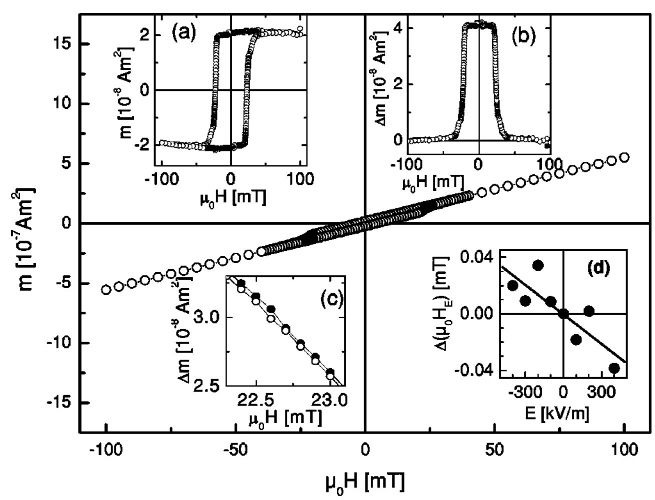

FIG. 1. Hysteresis loop m vs $\mu_0 H$ of the Cr_2O_3/CoPt heterostructure measured at $T=150$ K and $E=0$ after cooling from above $T_N=307$ K in $\mu_0 H=0.5$ T. Inset (a) shows the FM hysteresis m vs $\mu_0 H$ of the CoPt multilayer as measured in $E=0$ after subtracting the AF background. Inset (b) and (c: highly magnified) show the differences Δm vs $\mu_0 H$ of the descending and ascending branches of hysteresis loops obtained in an electric field $E=\pm 300$ kV/m (solid and open circles, respectively). Inset (d) shows the change of the EB field as a function of the electric field at $T=150$ K.

When applying an electric dc field of, say, $E = \pm 300$ kV/m at $T=150$ K virtually the same hysteresis curve is observed. Only closer inspection shows small, but finite shifts due to magnetoelectric EB. In order to determine them quantitatively we proceed as follows. First, we subtract the two branches of each of the hysteresis curves from one another (Fig. 1, inset b) in order to eliminate any nonhysteretic background, in particular the vertical shifts due to the magneto-electric moment, $m = \pm 4 \cdot 10^{-9}$ A m^2, in the bulk of the AF substrate crystal. They risk to mimic an apparent EB in the case of nonperfectly rectangular shaped loops. Information on the real EB shift is now unambiguously contained in the slopes Δm vs $\mu_0 H$ as shown exemplarily in the inset c of Fig. 1. A small shift is clearly observed, but it demands for some data treatment in order to be safely extracted. By assuming a rectangular hysteresis loop the shift $\Delta(\mu_0 H_e)(E)$ is given by the position of the peak of the convolution $f(E,\widetilde{H}) = \int_{-\infty}^{\infty} \Delta m(E=0,H)\Delta m(E,H-\widetilde{H})dH$ vs $\widetilde{H}$,[14] which is determined from an empirical polynomial fit of second order. By this method the entire data set of the loops is involved and a flawed determination of the coercive fields is avoided. This would rely on only two ill-defined points stemming from a field region, where the SQUID signal has its lowest signal-to-noise ratio. Hence, the convolution method is boosting the precision of the determination of the relative shifts to the required level.

An intuitive understanding of the electrically controlled EB is provided in the framework of the Meiklejohn–Bean approach, where the exchange bias field of an AF/FM heterostructure is determined according to $\mu_0 H_e = J S_{AF} S_{FM} / (M_{FM} t_{FM})$.[15] This simple formula describes the dependence of the bias field $\mu_0 H_e$ on a phenomenological coupling J between the FM and AF interface magnetizations S_{FM} and S_{AF}, respectively, where t_{FM} and M_{FM} are the thickness and the saturation magnetization of the FM layer. In the presence of an electric field, the electrically induced interface magnetization in the ME material couples to the adjacent FM film. The positive or negative electric field induced contribution to the total AF interface magnetization S_{AF} modifies the EB-field in accordance with the Meiklejohn–Bean formula.

The electric field induced magnetic moment, its interface contribution, and the corresponding change of the EB effect remains small in heterostructures involving bulk single crystalline AF pinning systems. For Cr_2O_3, with typical applied electric fields of the order of 10^5 V/m on a mm-sized sample, the magnetic moment is only of the order of some ten parts per millions of μ_B per Cr atom. However, if we apply electric fields reaching dielectric breakdown values of thin films (10^9 V/m), the extrapolation of the linear $M(E)$ behavior predicts a magnetic moment of several percent of μ_B per atom. If a ME layer is adjacent to a ferromagnetic (FM) film, exchange coupling allows a shift $\mu_0 H_e$ of the FM hysteresis, with magnitudes becoming relevant for applications as spin valves. Linear extrapolation predicts values of the order of 100 mT if electric field values approaching dielectric breakdown are applied.[16]

The use of a ME thin film as dielectric tunnel junction between two FM layers (Fig. 2) takes maximum advantage of large applied electric field. We expect a significant net

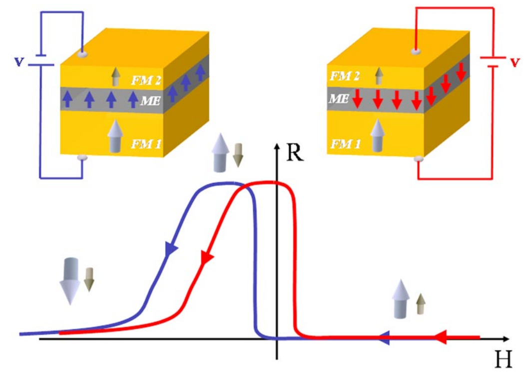

FIG. 2. (Color online) Schematics of the magnetoresistance curve of a TMR device involving a ME film as tunnel barrier. Half-hysteresis curves are shown, after saturation at positive field values. The arrows denote the magnetization directions, with the bottom layer being harder than (or pinned to) the top one. The change of voltage polarity changes the direction of the net magnetization of the ME layer, adding an exchange bias magnetic field to the resistance curve. The two colors indicate a reversal of polarity.

magnetization of the barrier to occur when electric field values typical for TMR devices are used, reaching 1 V/nm, e.g., 10^9 V/m. Applying an electric field creates exchange bias with neighbor ferromagnets FM1 and FM2. Therefore the magnetization curves of both magnetic layers are shifted along the magnetic field axis. The related TMR curves are similarly shifted, as depicted in Fig. 2. An exchange bias field value of the order of the saturation field of the soft magnetic layer, i.e. of several mT, will provide control of the magnetization direction of the soft layer. At low applied magnetic field values, the resistance state of the device will be modified when switching the magnetic orientation of the soft layer, which can be achieved by modifying the electric field in the ME film. The two half-hysteresis magnetoresistance curves of Fig. 2 illustrate how resistance values at low magnetic fields depend on the polarity of the applied voltage bias. In this simplest picture one can switch between two resistance values of the device by switching the applied voltage polarity. The resulting device capabilities are similar to what is expected using current-induced magnetization switching, with the advantages of larger devices and larger resistance values, more suited for applications.

In conclusion, we propose a spin dependent transport device with a resistance state controlled by applied voltages only. By taking advantage of the magnetoelectric effect occurring in an insulating antiferromagnetic film, the electrically-induced magnetization provides control of the magnetic pinning in exchange biased TMR structures. Similar architectures are also possible for GMR type devices involving a ME thin film as a tunable pinning layer. Our estimates show that significant pinning fields should result from exchange coupling between magnetoelectric and ferromagnetic layers, providing a control of the magnetization state of the ferromagnet by the applied voltage through the ME film. In our TMR device, the ME film is used as tunnel barrier and controls the magnetization of the soft magnetic layer. ME tunnel barriers are attractive because of the intrinsic large electric field occurring in these structures.

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