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View online: http://dx.doi.org/10.1063/1.4869152
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/104/11?ver=pdfcov
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Temperature dependence of the voltage-controlled perpendicular anisotropy in nanoscale MgO/CoFeBiTa magnetic tunnel junctions

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(Received 5 February 2014; accepted 10 March 2014; published online 19 March 2014)

In this work, we experimentally study the temperature dependence of the perpendicular magnetic anisotropy (PMA) and of the voltage-controlled magnetic anisotropy (VCMA) in nanoscale MgO/CoFeBiTa-based magnetic tunnel junctions. We demonstrate that the temperature dependence of both the PMA and the VCMA coefficient follow power laws of the saturation magnetization, but with different exponents. We also find that the linear dependence of the PMA on electric field is maintained over a wide temperature range, although the VCMA strength decreases faster as a function of temperature as compared to the PMA. Possible mechanisms leading to the different exponents are discussed.

Magnetic Tunnel Junctions (MTJs) have become one of the main building blocks in spintronic circuits due to their large tunneling magnetoresistance (TMR) ratio, possibility to electrically control the magnetization, and compatibility with conventional semiconductor fabrication processes.1,2 More importantly, the observation of a large perpendicular magnetic anisotropy (PMA) in the interface between CoFeB and MgO3,4 allows for constructing MTJs with stable perpendicular magnetization states, a key requirement for the scalability of magnetic random access memory (MRAM) bits.5 Furthermore, the demonstration of the sensitivity of this PMA to electric fields in the MgO layer,6 via the voltage-controlled magnetic anisotropy (VCMA) effect, and the application of VCMA to realize voltage-induced switching in nanoscale junctions,7–9 has created the possibility for a highly energy-efficient magnetoelectric random access memory (MeRAM) architecture,5,10 where electric fields, rather than currents, are used to write the information into the memory bits.

The magnetic anisotropy in ferromagnetic materials has been shown to have a strong dependence on temperature in previous theoretical and experimental works.11 In particular, Callen and Callen’s theory12 predicts that uniaxial anisotropies (e.g., PMA in our case) decrease with temperature as $M_s(T)$, while cubic anisotropies decrease up to $M_s^{10}(T)$, where $M_s(T)$ is the temperature-dependent saturation magnetization of the material. On the other hand, the temperature dependence of the VCMA effect has not been studied yet either theoretically or experimentally. Therefore, quantifying the temperature dependence of both PMA and VCMA is an important requirement for the design of emerging nanoscale memory and logic devices that exploit such effects. Beyond practical considerations, the study of the temperature dependence of the PMA and VCMA effects is also of interest as it may contribute to a better understanding of the underlying physics behind both of these phenomena.

In this Letter, we report on the experimental study of the temperature dependence of the interfacial PMA and the VCMA in nanoscale MTJ devices based on MgO/CoFeBiTa material stacks. We demonstrate that the temperature dependence of PMA and VCMA both follow power laws of $M_s(T)$, but with different exponents for the two quantities, where the strength of the VCMA effect is shown to decrease at a faster rate as a function of temperature compared to PMA. Additionally, the VCMA effect is observed to remain linear (i.e., the PMA varies linearly with the applied electric field) across the considered temperature range for this work (100 K to 400 K). The different power law exponents for the two quantities under study may indicate that only some of the mechanisms involved in the PMA of these structures are sensitive to electric fields, hence also contributing to the VCMA effect.

For this work, a multilayer stack with the composition of substrate | bottom electrode | PtMn (20) | Co$_{70}$Fe$_{30}$ (2.3) | Ru (0.85) | Co$_{60}$Fe$_{20}$B$_{20}$ (3) [fixed layer] | MgO (1.3) | Co$_{20}$Fe$_{60}$B$_{20}$ (1.5) [free layer] | Ta (5) | top electrode (thickness in nm) was deposited in a Singulus TIMARIS physical vapor deposition (PVD) system, and subsequently annealed at 300 °C for 2 h in an in-plane magnetic field of 1 T. The stack was then patterned into 125 nm x 50 nm elliptical nanopillars for electrical measurements [See Fig. 1(a)] using electron-beam lithography and ion milling techniques. The MgO tunneling barrier was designed to be thick enough (resistance-area (RA) product ~ 750 Ω·μm²) to make current-induced spin-torque effects negligible. Also, the thicknesses in the Co$_{70}$Fe$_{30}$Ru|Co$_{60}$Fe$_{20}$B$_{20}$ synthetic antiferromagnet (SAF) were tuned to cancel the offset field from the SAF into the free layer, while the PtMn provides an exchange bias strong enough to make the fixed layer insensitive to in-plane ($H_x$) magnetic fields in the range of study (± 2 kOe). Finally, the thickness of the Co$_{20}$Fe$_{60}$B$_{20}$ free
layer was chosen such that the stable magnetization state of the free layer is in the perpendicular (z) direction.

The inset in Figure 1(b) shows resistance (R) versus in-plane magnetic field (Hx) curves measured at three different temperatures. The hard-axis-like curves confirm that the free layer has a stable perpendicular magnetization, while the small size of the nanopillars ensures a largely single-domain rotation of the magnetization with in-plane fields. In order to quantify the dependence of the effective anisotropy field $H_{k\text{eff}}(T, V)$ (i.e., the field required to saturate the perpendicular free layer into the in-plane direction) on temperature $T$ and, due to the VCMA effect, on voltage $V$ applied to the MTJ, the measured resistance values are translated into conductance $G = 1/R$. Considering the linear relationship between MTJ conductance and the component of the free layer magnetization along the direction of the fixed layer in MTJs\textsuperscript{13,14} (i.e., $m_z = M_z/M_s$ in our case), the measured $R$-$H_z$ curves are translated into $m_z$-$H_z$ curves. Finally, $H_{k\text{eff}}$ can be calculated from the highlighted area in Figure 1(b), i.e.,\textsuperscript{14}

$$H_{k\text{eff}}(T, V) = \frac{1}{0} H_x(m_z) dm_z.$$  \hspace{1cm} (1)

For this work, $H_{k\text{eff}}(T, V)$ was measured for temperatures from 100 to 400 K in steps of 20 K, and for each temperature, the voltage dependence was measured for voltages between $-0.8$ and $+0.8$ V in steps of 0.2 V.

The dependence of the saturation magnetization on temperature $M_s(T)$ was independently measured in a 5 mm × 5 mm sample with a composition of substrate | MgO (2) | Co$_{20}$Fe$_{60}$B$_{20}$ (1.5) | Ta (5) | cap, which corresponds to the structure of the free layer in the nanopillar devices. The sample was annealed in the same conditions as the stacks used to fabricate the MTJs. Figure 2(a) shows measurements of the magnetic moment $M$ as a function of out-of-plane magnetic field ($H_z$) for four different temperatures using a superconducting quantum interference device (SQUID) magnetometer, confirming that the Co$_{20}$Fe$_{60}$B$_{20}$ layer has a perpendicular easy-axis over a wide range of temperatures for a thickness of $t_{CoFeB} = 1.5$nm. A strong dependence on temperature of the coercivity (dependent also on the perpendicular anisotropy) is observed, where the coercivity decreases from $\sim$250 Oe to $\sim$15 Oe when increasing the temperature from 10 to 300 K. At the same time, a weaker dependence of the saturation magnetization $M_s$ on temperature $T$ is measured. Figure 2(b) shows the extracted data for $M_s$ versus $T$ in the range from 10 K to 400 K. The data are found to fit well to Bloch’s law\textsuperscript{15}.

FIG. 1. (a) The devices used in this work were 125 nm × 50 nm elliptical magnetic tunnel junctions, where the Fe-rich CoFeB free layer has a perpendicular easy-axis. The MgO thickness was designed to be thick enough to make current-induced torques negligible, while the synthetic antiferromagnet was designed to cancel stray fields into the free layer. Magnetic fields in the x-direction ($H_x$) are used to saturate the magnetization parallel (P) or antiparallel (AP) to the fixed layer. (b) The scatter plot data in the inset show the experimental resistance $R$ versus $H_x$ curves measured for three different temperatures, which are translated to the x-component of the magnetization $m_x = M_x/M_s$ taking into account the linear relation between $m_x$ and the conductance $G = 1/R$. The effective anisotropy field $H_{k\text{eff}}$ is then calculated for different temperatures and voltages by calculating the highlighted area. The solid lines show the results obtained from micromagnetic simulations, which are found to accurately reproduce the experimental results (scatter data), with an error smaller than 1% in the estimation of the PMA.

FIG. 2. (a) SQUID measurements of the magnetization $M$ as a function of the perpendicular magnetic field $H_z$. The magnetization is observed to have a perpendicular easy-axis for the whole temperature range under study in this work. While a weak dependence of the saturation magnetization $M_s$ on temperature $T$ is measured, the strong dependence of the coercivity on temperature is evidence for the strong dependence of the PMA on temperature. (b) The extracted dependence of $M_s$ as a function of temperature is found to fit well to Bloch’s law with fitting parameters $T^* = 1120$ K and $M_s(0) = 1457$ emu/cm$^3$.\hspace{1cm}
\[ M_s(T) = M_s(0) \left(1 - \left(\frac{T}{T_C}\right)^{3/2}\right) \]  

(2) due to the large Curie temperature \( T_C \) of CoFe(B) compared to the range of temperatures under consideration. From the fit, we estimate that \( T' = 1120 \text{ K} \), while a value of \( M_s(0) = 1457 \text{ emu/cm}^2 \) is obtained for the spontaneous magnetization at absolute zero.

We first quantify the dependence of the interfacial PMA energy \( K_i \) on temperature at equilibrium, i.e., without applying any voltages to the MTJ.\(^{16}\) The inset in Figure 3 shows the extracted values for the effective anisotropy field at equilibrium \( H_{k,eff}(T, V = 0) \), where we observe a decrease in \( H_{k,eff} \) from ~1610 Oe to ~720 Oe when increasing the temperature from 100 to 400 K. \( K_i \) can be directly related to the effective perpendicular anisotropy \( K_{eff} \), which is proportional to \( H_{k,eff} \).

Specifically,

\[ K_{eff}(T, V) = \frac{M_s(T)H_{k,eff}(T, V)}{2} \]

\[ = K_i(T, V) \frac{2\pi(N_z - N_x)M_s^2(T)}{t_{\text{CoFeB}}} \]

(3)

where the first term accounts for the PMA, while the second term corresponds to the demagnetization energy. The geometrical demagnetization factors in the \( z \) and \( x \) directions can be estimated to be \( N_z = 0.9343 \) and \( N_x = 0.0150 \), respectively, by using the elliptical cylinder approximation.\(^{17}\)

Figure 3 shows the extracted values for \( K_i(T, V = 0) \), calculated using Eq. (3), where we obtain a value of 1.45 erg/cm\(^2\) = 1.45 mJ/m\(^2\) for the PMA at room temperature \((T = 300 \text{ K})\), in good agreement with previous reports for the MgO insulating layer.\(^{3,4}\) The dependence of \( K_i \) on temperature is found to fit well to a power law of \( M_s(T) \), i.e.,

\[ K_i(T) = K_i(0) \left(\frac{M_s(T)}{M_s(0)}\right)^{\gamma} = 2.18 \pm 0.04, \]

(4) where the PMA at zero temperature is \( K_i(0) = 2.02 \text{ erg/cm}^2 \). It is worth noting that a power law for \( K_i(T) \) with an exponent \( \gamma = 2.3 \pm 0.22 \) was also found in the 5 mm \( \times \) 5 mm sample used to extract \( M_s(T) \), consistent inside experimental errors with the result obtained from the MTJ stack.

In order to rule out possible contributions from micromagnetic effects or a strong influence from second order PMA in the obtained power law exponent for \( K_i \), we performed micromagnetic simulations to reproduce the hard-axis loops at different temperatures. The solid lines in Fig. 1(b) show the results from these simulations for three different temperatures, where we obtain a very good match to the experimental data assuming negligible second order PMA and obtaining an error smaller than 1% in the estimation of \( K_i \).\(^{18}\)

Next, we quantify the temperature dependence of the VCMA effect, in particular, by looking at the temperature dependence of the VCMA coefficient \( \xi \), which describes the change of interfacial anisotropy energy per unit electric field, i.e., \( \xi = \Delta K_i/(\Delta V/d_{\text{MgO}}) \), where \( d_{\text{MgO}} \) is the thickness of the MgO insulating layer. Figure 4(a) shows the change of the effective anisotropy field \( \Delta H_{k,eff} \) as a function of the voltage \( V \) for four different temperatures between 100 K and 380 K, where \( \Delta H_{k,eff} = H_{k,eff}(V) - H_{k,eff}(V = 0) \). Similar to previous studies at room temperature for the MgO/CoFeB/Ta system,\(^{14,19}\) our results show a linear dependence of \( \Delta H_{k,eff} \) with voltage, where a positive voltage (more precisely, the depletion of electrons from the CoFeB|MgO interface)

\[ \frac{\Delta H_{k,eff}}{V} \approx 1.2 \pm 0.05 \text{ Oe/V} \]

FIG. 3. The inset shows the extracted dependence of the effective anisotropy field \( H_{k,eff} \) (i.e., the field required to saturate the perpendicular free layer into the in-plane direction) as a function of temperature at zero bias voltage. Using the extracted values, the interface anisotropy is calculated and the temperature dependence is found to fit well to a power law of \( M_s(T) \) with an abnormal power law exponent \( \gamma = 2.18 \pm 0.04 \). The power law exponent combines contributions from the MgO/CoFeB and CoFeB/Ta interfaces.

FIG. 4. (a) Change of the effective anisotropy field \( \Delta H_{k,eff} = H_{k,eff}(V) - H_{k,eff}(V = 0) \) as a function of voltage for different temperatures. The VCMA effect is observed to remain linear over the temperature range of study, but the change of the anisotropy field per unit voltage \( \Delta H_{k,eff}/\Delta V \) (the fitted slopes, in Oe/V) is reduced when temperature increases. (b) The inset shows the extracted values of \( \Delta H_{k,eff}/\Delta V \) as a function of temperature. By using these values, the VCMA coefficient \( \xi \) (i.e., the change of interface anisotropy per unit electric field) is calculated and also found to fit well to a power law of \( M_s(T) \), but with an exponent \( \gamma' = 2.83 \pm 0.2 \) that is closer to Callen-Callen’s law.\(^{12}\)
increases the effective perpendicular anisotropy. More importantly, we observe that the linearity of the VCMA effect is maintained over the wide temperature range under study; however, the relative change of the effective field per unit voltage $\Delta H_{\text{eff}}(T)/\Delta V$ (i.e., the slope of the $\Delta H_{\text{eff}}$ versus $V$ curves, in units of Oe/V) is observed to decrease as the temperature increases. The inset in Figure 4(b) shows the extracted values for $\Delta H_{\text{eff}}/\Delta V$ as a function of temperature during the experiment. Consequently, the temperature-dependent VCMA coefficient is calculated as $\xi(T) = M_s(T) (\Delta H_{\text{eff}}(T)/\Delta V)_{\text{MgO|CoFeB}}/2$, where the computed values are plotted in Figure 4(b). The VCMA coefficient at room temperature ($T = 300$ K) is found to be $3.1 \times 10^{-9}$ erg/V-cm = 31 fJ/V-m, in good correspondence with our previous work, and the coefficient is observed to decrease from $\sim 46$ to $\sim 25$ fJ/V-m when the temperature is increased from 100 to 400 K. Further, the temperature dependence of the VCMA coefficient is also found to fit well to a power law of $M_s(T)$, but notably with a different exponent compared to $K_i(T)$, i.e.,

$$\xi(T) = \xi(0) \frac{M_s(T)}{M_s(0)} \gamma \quad \gamma = 2.83 \pm 0.2, \quad (5)$$

with $\xi(0) = 48.9$ fJ/V-m as the VCMA coefficient at zero temperature.

Our results indicate that, while both $K_i(T)$ and $\xi(T)$ follow power laws of $M_s(T)$, they are described by different exponents, where the exponent for the VCMA coefficient $\xi(T)$ ($\gamma = 2.83 \pm 0.2$) is found to be close to Callen-Callen’s $M_s^2(T)$ law at small reduced temperatures $T/T_C$ (a condition met by the temperature range considered in this work), while an abnormal exponent $\gamma = 2.18 \pm 0.04$ describes the temperature dependence of the interfacial perpendicular anisotropy $K_i(T)$. Although Callen-Callen’s law has been found to be accurate in describing the temperature dependence of uniaxial anisotropies in simple ferromagnetic systems (for example, uncapped ultrathin Fe grown on a GaAs substrate, where a $M_s^2(T)^{2.9 \pm 0.2}$ dependence of the uniaxial anisotropy was measured experimentally), deviations from such a power law have been previously observed in more complicated systems. In particular, this has been reported in the case of alloys, where the material is constructed by more than a single sublattice and/or where the exchange interaction is anisotropic, as well as in cases where non-magnetic materials with large spin-orbit coupling are present which strongly contribute to the anisotropy, but do not have a pronounced effect on the other magnetization properties (e.g., the Curie temperature or the saturation magnetization). Both of these scenarios violate two basic assumptions of Callen-Callen’s theory: First, single-ions with localized magnetic moments are considered the origin of the magnetization in the material; and second, the anisotropy is affected by temperature by the same mechanisms as the magnetization (i.e., average deviations in the magnetic moments due to temperature fluctuations giving rise to a decrease in $M_s$). Hence, recent theoretical works have predicted a leading $M_s^2(T)$ dependence for the uniaxial anisotropy when considering two-ion itinerant or localized contributions, especially in the presence of high spin-orbit coupling materials that contribute to the PMA. The latter conditions give rise to contributions to the temperature dependence of the PMA which are mutually exclusive to those coming from Callen-Callen-like behavior. A clear example of the interplay between different contributions has been observed in experiments performed on FePt, where the anisotropy shows a $M_s^2(T)^{1.1}$ dependence and such a power law exponent has been also reproduced via ab-initio calculations, indicating only a 10% contribution from Callen-Callen-like behavior.

For the system in the current work, the measured PMA of the CoFeB free layer is an overall quantity that includes contributions from both the MgO|CoFeB and CoFeB|Ta interfaces, i.e., $K_i = K_i_{\text{MgO|CoFeB}} + K_i_{\text{CoFeB|Ta}}$. However, in principle only the MgO|CoFeB interface should be sensitive to the applied electric field, and therefore the VCMA coefficient is solely related to this interface. The perpendicular anisotropy contribution from the MgO|CoFeB interface has been attributed to hybridization of Fe d-orbitals with O p-orbitals, whereas the VCMA effect is due to the modification of the occupancy between different hybridized orbitals. Therefore, the fact that the localized orbitals (moments) modified by voltage follow more closely the conditions assumed in Callen-Callen’s law might explain why the VCMA coefficient power law exponent is dominated by a $M_s^2(T)$ contribution. On the other hand, the non-integer power law exponent of $K_i(T)$ obtained in this work has a leading $M_s^3(T)$ contribution, which may be related to the influence of the high spin-orbit coupling Ta, and a smaller $M_s(T)$ contribution, which could be accounted for by the MgO|CoFeB interface. In fact, a previous work by Worledge et al. demonstrated that the CoFeBiTa interface plays a key role in the strong PMA of the MgO|CoFeB|Ta stack, albeit the exact role of Ta (and in general, of the high spin-orbit coupling metal interfacing with a ferromagnetic material showing a large PMA), is more complicated, involving its role both as a sink of boron atoms during post-deposition annealing, as well as promotion of (001) crystalline orientation in the resulting CoFe(B) layer. In addition, a recent work by Liu et al. proposed that the Ta increases the effective spin-orbit coupling of the CoFe(B), and at the same time, the small electronegativity of Ta promotes the hybridization of Fe-O bonds, both of these effects resulting in a larger PMA. However, a more detailed understanding of the exact contribution of each mechanism to explain the different power law exponents obtained in this work will require ab-initio calculations.

Finally, in terms of device applications where the VCMA effect is used to switch MTJ devices, a key figure of merit in the electric-field-driven switching process is the ratio between the VCMA coefficient and the PMA, i.e., $\xi(T)/K_i(T)$. Given that these two quantities are not proportional, but rather $\xi(T)$ decreases faster as a function of temperature compared to $K_i(T)$, as the temperature increases, the figure of merit $\xi(T)/K_i(T)$ decreases, and therefore, larger voltages will be needed to achieve the electric-field-driven switching for a constant value of the interfacial anisotropy (i.e., for a constant retention time when used as memory). For example, we estimate that an increase of 10% in the switching voltage will be required to compensate for
the loss in the $\zeta(T)/K_i(T)$ figure of merit, when increasing the temperature between the typical limits of operation for a memory device (240 to 400 K).

In conclusion, the temperature dependence of the interfacial perpendicular magnetic anisotropy and its sensitivity to electric fields in the MgO layer have been measured over a wide range of temperatures for nanoscale MgOCoFeB/Ta MTJ devices. It was demonstrated that the temperature dependence of these quantities follow power laws of the saturation magnetization, but with different power law exponents. Our results support previous reports indicating that the high power law dependence of these quantities follow power laws of the saturation magnetization, but with different power law exponents.

A small voltage of 10 mV is applied to the MTJ in order to measure the resistance state of the device.

For the micromagnetic simulations, the values for $M_s$ at different temperatures were extracted from Fig. 2(b), while the exchange stiffness $A$ was used as a fitting parameter.

This work was partially supported by the DARPA program on Non-Volatile Logic, and in part by the NSF Nanosystems Engineering Research Center for Translational Applications of Nanoscale Multiferroic Systems (TANMS). J.G.A. and P.U. would like to acknowledge partial support through the Qualcomm Innovation Fellowship.

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