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Electric field effect on magnetism in metallic ultra-thin films

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Recent experimental developments on the electric field effect on magnetism in metallic magnetic materials are reviewed. The change in the electron density at the surface of metallic ultra-thin magnets by the application of an electric field results in modulations of the Curie temperature, magnetic moment, magnetic anisotropy, and domain wall velocity. The study focused on this paper is the electric field effect on the Curie temperature (magnetic phase transition) in Pt/Co ultra-thin film systems. Electric field modifications of the magnetic moment induced by ferromagnetic proximity effects in Pd, which is usually a nonmagnetic element, are also discussed.

Keywords: electric field effect, metallic ultra-thin films, ferromagnetism, phase transition, ferromagnetic proximity effect

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INTRODUCTION

Magnets are often needed in modern industry because they have spontaneous magnetization. The direction of the magnetization is utilized as an information bit in magnetic recording media, and the strength of the magnetization is one factor that determines the performance of magnetic devices, e.g., a motor. If the direction or strength of the magnetization can be effectively controlled, the functions and applications of magnetic devices could be expanded, and the dissipation power of these devices will be drastically reduced.

The mechanism for controlling magnetism by the application of an electric field through an insulator is one strong candidate which can fulfill the above expectation [1, 2]. The electric field control of the Curie temperature [3], coercivity [4], magnetization direction [5, 6], domain wall motion [Yamanouchi_JJAP], and magnetic moment [7] has been experimentally demonstrated using ferromagnetic semiconductors, in which the magnetic property is a function of a carrier concentration that can be modulated by the application of an electric field. These works have defied the previously held belief that the magnetic properties of a material cannot be changed without changing the temperature once the material has been prepared.

Recently, in ferromagnetic metals (FMs), similar results based on the electric field effect have been reported at room temperature [8–11]. Because of the screening effect, the electric field cannot penetrate into the bulk in such metallic systems. Thus, the effect obtained in FMs could be attributed to the change in the electron density at the surface of the material [12, 13]. In particular, at the surface of the 3*d* transition FMs, the shift of the Fermi level due to the electric field application changes the relative occupation of the 3*d* orbitals, resulting in the change in the interface magnetic anisotropy [9, 13, 14]. This electrical control of magnetic anisotropy is useful for magnetic recording technology.

Although magnetization switching in magnetic tunnel junctions using spin transfer torque [15–17] is existing technology for writing information in magnetic random access memories, it requires current with very high density; thus, further reduction of unwanted energy dissipation by Joule heating is required. Magnetization switching using the electric field modulation of magnetic

anisotropy [6, 18–21] is expected to reduce the power consumption by more than one or two orders of magnitude because the power required for switching is attributed to charging or discharging of the electrons.

To apply an electric field to the surface of an FM, voltage is applied to a capacitor structure containing an FM electrode. In this structure, the change in the sheet electron density Δn_s in the FM electrode by gating corresponds to CV_G/e, where C is the capacitance per unit area, V_G is the gate voltage, and e is the electron charge. Therefore, using a device with larger C and applying a larger V_G is the key for realizing a larger Δn_s. The capacitance is determined by the relative permittivity k and the thickness of the insulator layer d as $C = k\varepsilon_0/d$, where ε_0 is the vacuum permittivity. To obtain a larger C, a higher k and smaller d are required for the dielectric layer.

A conventional approach for obtaining a large C is the adoption of high-k materials (e.g., Al_2O_3, HfO_2, or ZrO_2). Another important consideration is that the capacitor should have a high breakdown field E_{BD} ($= V_{BD}/d$, where V_{BD} is the breakdown gate voltage), allowing for the application of a large V_G across the capacitor to obtain a large Δn_s. Dielectric layers obtained by atomic layer deposition (ALD) are known to have a high E_{BD} [22]. In particular, if 50-nm-thick HfO_2 ($k \sim 20$) dielectric layer is adopted and 10 V of V_G (electric field of 0.2 V/nm) is applied, $\Delta n_s \sim 2\times10^{13}$ cm^{-2}. If only the electrons in the topmost mono-layer (ML) of the FM surface can be modulated because of the screening effect, the modulated electron number per FM atom is on the order of 0.01 [11].

Another way to realize a large C is to use an ionic liquid (or a liquid electrolyte). When a gate voltage is applied between a FM and counter gate electrodes through the ionic liquid, ions are attracted electrostatically to both electrodes; thus, electric double layers (EDLs), which are pairs of sheets of negative and positive charges consisting of the ions in the liquid electrolyte and the induced charges in the surface of the FM electrode (or the gate electrode), respectively, are formed. The EDL capacitor has a very large C because the gap of the two charged sheets, which corresponds to d, is ∼1 nm [23]; thus, a large Δn_s is realized. In this case, the modulated electron number per FM atom can reach the order of 0.1 by the application of a few volts of V_G.

The change in the electron density at the FM surface is a reasonable explanation for the electric field control of magnetism. As another candidate, the electric field modulation of the uniaxial perpendicular magnetic anisotropy (PMA) caused by the Rashba effect at the interface between the two different materials, e.g., a FM metal and nonmagnetic metal or insulator, has been theoretically proposed [24]. Furthermore, the voltage-driven oxidation-reduction reaction or O^{2-} migration in the FM/oxide bilayer is known to be another source for altering the magnetic properties of a FM [25, 26]; however, this effect is much slower than the electron charging effect.

In this paper, recent experimental advances on the electric field control of the ferromagnetic phase transition in Pt/Co systems are reviewed. In addition, electric field modifications of the magnetic moment in ferromagnetic Pd, which is usually a nonmagnetic element, deposited on Pt/Co system are also discussed.

ELECTRIC FIELD CONTROL OF THE MAGNETIC PHASE TRANSITION

The author found that not only the magnetic anisotropy but also the ferromagnetism itself is electrically switchable around room temperature in a perpendicularly magnetized ∼2 ML of Co ultra-thin film deposited on a Pt underlayer. The layer structure used in the experiment was Pt(∼1 nm)/Co(0.4 nm)/MgO(2.0 nm) from the bottom side. These layers were deposited on an intrinsic Si substrate by dc or rf-sputtering. The Pt layer was confirmed to have a fcc(111) texture by scanning transmission electron microscopy (STEM) (see Supplementary Online Material of Koyama et al. [27]).

The first experiment was performed using a solid-state capacitor structure with a 50-nm-thick ALD-HfO_2 dielectric insulator layer [11]. In this experiment, the anomalous Hall effect was used to detect the magnetization state; thus, the HfO_2 insulator and the gate electrode were formed on top of the Hall-bar-mesa structure made of Pt/Co layers. Here, the anomalous Hall resistance R_{Hall} is proportional to the perpendicular component of the magnetization. An optical microscopy image is shown in the inset of **Figure 1A**. A clear difference was observed in the magnetization curves detected by the anomalous Hall effect for positive and negative V_G (±10 V) applications, as shown in **Figure 1A**. (Note that positive V_G is defined as the direction of increasing electron density at the Co surface). When the positive V_G was applied, a clear rectangular hysteresis loop was observed, whereas a linear response was obtained under the application of negative V_G. This V_G-dependent change in the magnetization curve was observed in a reversible way. From the detailed measurements, e.g., temperature dependence of magnetic susceptibility, this dramatic effect was not attributed to the switching of the magnetic easy axis from the perpendicular to the in-plane direction. This effect was attributed to the phase transition from the ferromagnetic to paramagnetic state. From the Arrott-plot [28] (or the Arrott-Norkes plot [29]), the temperature T dependence of the spontaneous R_{Hall} (R^s_{Hall}) for the application of positive and negative V_G, which is proportional to the spontaneous magnetization, was obtained (see **Figure 1B**). The Curie temperature T_C between them was determined to be ∼12 K. Because of the two dimensionality of the Co film, T_C in this sample was around room temperature [30].

Ionic-liquid gating was also adopted for modulating the magnetic properties of the similar Pt/Co sample [31]. In this experiment, a direct magnetization measurement using a superconducting quantum interference (SQUID) magnetometer was used to detect the magnetization under gating. A polymer film containing the ionic liquid (EMI^+-$TFSI^-$) was placed on the sample, and a Pt foil was placed on top to serve as a gate electrode (see the inset of **Figure 1C**). This film-shaped ionic liquid simplifies the measurement in the SQUID chamber. By applying V_G between the Co layer and the gate electrode,

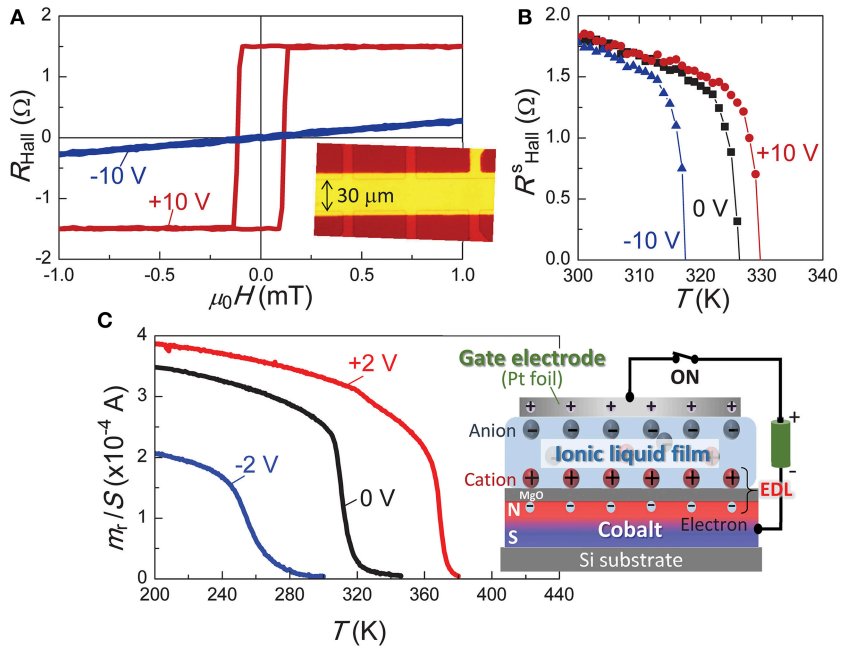

FIGURE 1 | Electric field effect on ferromagnetism in the Pt/Co system. (A) Magnetization curves detected in the Hall resistance R_{Hall} for $V_G = -10$ and 10 V at 321 K. The inset shows the top view of the Hall bar structure with the gate electrode on top (yellow region) taken by an optical microscope. Solid-state HfO_2 was used as a gate insulator in this device. **(B)** Temperature T dependence of the spontaneous Hall resistance R^S_{Hall} which is proportional to the spontaneous magnetization obtained with the device in **(A)**. The Curie temperature increases (decreases) with positive (negative) V_G. **(C)** The T dependence of the remanent magnetic moment per unit area (m_r/S) for $V_G = +2$, 0, and -2 V. The inset shows the cross-sectional image of the device structure. An ionic liquid was used to gate the sample.

the EDL is formed through the MgO capping layer, and the electron density at the Co surface can be modulated. **Figure 1C** shows the T dependence of the remanent magnetic moment per unit area (m_r/S) for three different V_G values ($V_G = +2$, 0, and -2 V). In this device, a giant change in T_C up to ~100 K around room temperature was achieved. This is believed to be the result of the larger change in the electron density in the EDL capacitor compared to that in the HfO_2 capacitor structure.

In both devices (the ALD-HfO_2 and the ionic-liquid-EDL capacitors), the direction of the change in T_C was the same: positive (negative) V_G, i.e., the increase (decrease) in the electron density, resulted in higher (lower) T_C. Thus, the T_C had a positive slope with respect to the electron density. According to previous reports or *ab-initio* calculations for a bulk CoNi alloy [32], an increase in the Ni composition, i.e., the decrease in the electron number per atom, results in the reduction of T_C, which is similar to the case of the Slater-Pauling curve (atomic magnetic moment with respect to the electron number for 3*d* transition metal alloys). The direction of the change in T_C under gating opposed the Salter-Pauling type behavior (negative slope). Recently, the electric field effect on T_C for a similar Pt/Co structure was calculated by *ab-initio* calculations via Monte Carlo simulations that consider the magnetic anisotropy, and the same direction of the T_C change with the gating experiment was reported [33]. In the paper, it has been pointed out that, because the *sp* electrons predominantly contribute to the screening of the electric field, the change in the electron number for *d* electrons opposes that in the total electrons, i.e., the sign of the slope of T_C with respect to the number of *d* electrons is the same with the Slater-Pauling curve. This can be one reasonable explanation for the experiment. Another possibility is the oxidation-reduction reaction by the application of the electric field [25, 26]. The negative V_G is in the direction of the oxidation reaction, which should reduce T_C and the magnetic moment of the Co layer. Moreover, the reduction reaction may increase T_C if the Co layer was slightly oxidized after deposition. The evaluation of these effects will be considered in future study.

ELECTRIC FIELD CONTROL OF MAGNETIC MOMENT IN Pd

The results shown in the previous section show that ferromagnetism is switchable in ferromagnets using an electric field. It is natural to consider whether the electric field can make non-magnets ferromagnetic. Moreover, if one electron is removed from a Cu atom by an electric field, it is not known if Cu will show ferromagnetism similar to Ni. This is an extreme example, and it may be impossible to remove one electron from a metallic atom using an electric field. However, this concept may be realized with metals that are similar to ferromagnets. Among these metals, Pd is a nonmagnetic metal that nearly satisfies the Stoner criterion [34–37].

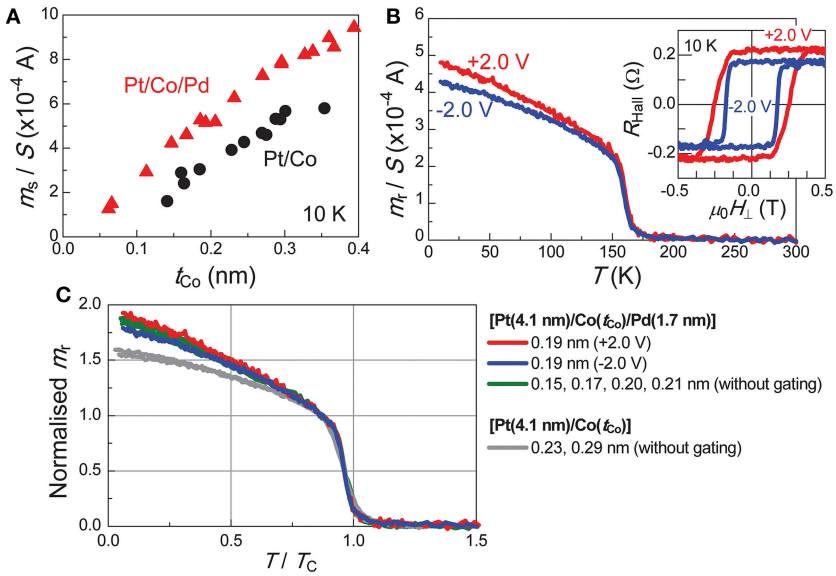

FIGURE 2 | Electric field effect on the induced magnetic moment in Pd. (A) The Co thickness t_{Co} dependence of the saturation magnetic moment per unit area (m_s/S) at 10 K for the Pt(4.1 nm)/Co(t_{Co}) and Pt(4.1 nm)/Co(t_{Co})/Pd(1.7 nm) samples. **(B)** The T dependence of the remanent magnetic moment per unit area (m_r/S) for the Pt(4.1 nm)/Co(0.19 nm)/Pd(1.7 nm) sample under positive (+2 V) and negative (−2 V) V_G. The inset shows the magnetic hysteresis loops for both V_G at 10 K. For the hysteresis measurement, the anomalous Hall effect was used. **(C)** The T/T_C dependence of the normalized m_r for the Pt/Co and Pt/Co/Pd samples with various t_{Co}. The m_r was normalized at $T/T_C = 0.87$. All data points without gating were sandwiched by the positively and negatively gated data points, indicating that the induced magnetic moment at the surface of the Pd layer was modulated by the application of an electric field.

Ab-initio calculations have shown that the peak of the density of states of bulk nonmagnetic Pd is located at an energy near the Fermi level [34], suggesting that an applied electric field may affect the magnetic state in Pd [37]. Although the modulation of paramagnetic properties was observed in Pt thin films using the EDL capacitor [38], electric-field-induced ferromagnetism has not been reported in nonmagnetic metals. Thus, as a first step, the tenability of the induced magnetic moment in Pd by the ferromagnetic proximity effect has been investigated [39].

A magnetic moment is induced by the ferromagnetic proximity effect in a Pd or Pt layer deposited on a ferromagnetic metal layer [40–45]. **Figure 2A** shows the Co thickness t_{Co} dependence of the saturation magnetic moment per unit area (m_s/S) at 10 K for the Pt(4.1 nm)/Co(t_{Co}) and Pt(4.1 nm)/Co(t_{Co})/Pd(1.7 nm) samples. The Pd and Pt layers were confirmed to have an fcc [**?**] texture using X-ray diffraction. Both samples were capped by 2-nm-thick MgO layer. For both series, m_s/S increases with t_{Co}. However, the samples with the Pd layer showed larger m_s/S than the Pt/Co samples, indicating that a magnetic moment was induced in the Pd layer. Assuming that the magnetic moment is uniformly induced in the entire Pd layer, the induced magnetic moment per Pd atom is calculated to be $\sim 0.1\mu_B$ (μ_B is the Bohr magneton), the order of which is in good agreement with previous studies [40, 42, 43]. The Pd thickness dependence of the magnetic moment (not shown) indicated that the magnetic moment was induced up to 2 nm from the Pd/Co interface. Thus, the Pd surface of the samples shown in **Figure 2A** is expected to be ferromagnetic. Note that no superparamagnetic behavior was observed in Pt/Co/Pd samples even when the thickness of the Co layer was about half monolayer (∼0.1 nm). In addition, the decrease in T_C and perpendicular magnetic anisotropy were confirmed after annealing of several Pt/Co/Pd samples at 200 or 300°C, suggesting that there was much less intermixing of Pd or Pt with Co before annealing.

Figure 2B shows the T dependence of the remanent magnetic moment per unit area (m_r/S) for one of the Pt/Co/Pd samples with $t_{Co} = 0.19$ nm under positive and negative V_G. Though T_C was not visibly changed, a clear difference was observed in m_r/S, and the difference increased with decreasing temperature. The squareness ratio was almost one for both V_G values at low T as shown in the inset; thus, this change was attributed to the change in the induced magnetic moment at the surface of the Pd layer. Two samples with different t_{Co} values were investigated. The modulated atomic magnetic moment for V_G = 1 V was calculated to be 0.05–0.08μ_B, and the modulated electron number per Pd atom was estimated to be 0.03–0.05 from the measured capacitance of each device.

Figure 2C shows the T/T_C dependence of the normalized m_r for Pt/Co and Pt/Co/Pd samples with various t_{Co}. The m_r was normalized at $T/T_C = 0.87$. In general, a larger increase in the magnetic moment was observed in the Pt/Co/Pd samples as T decreased. This behavior is probably due to the larger magnetic susceptibility of the Pd layer at lower T [43, 45]. All data points for samples without gating were sandwiched by the data points from the positively and negatively gated ones in Pt/Co/Pd samples. This is one example showing that the induced

magnetic moment at the surface of the Pd layer was modulated by the application of the electric field. Inducing ferromagnetism in non-magnetic materials is the next challenge.

CONCLUSIONS

Many experimental results reveal that various magnetic properties are tunable using an electrical gating. Although control of the phase transition and the magnetic moment was emphasized in this paper, the speed of the magnetic domain wall is also tunable using an electrical gating [46–48]. Electric-field-induced magnetization switching [20, 21] based on the control of the magnetic anisotropy [5, 9, 10] should significantly impact future magnetic recording applications. These facts mean that use of the electric field effect has already been established in magnetic materials as well as semiconductors. The electrical control window of the magnetic properties is expected to continue to increase.

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