

## Modulation of ferromagnetism in (In,Fe)As quantum wells via electrically controlled deformation of the electron wave functions

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We demonstrate electrical control of ferromagnetism in field-effect transistors with a trilayer quantum well (QW) channel containing an ultrathin *n*-type ferromagnetic semiconductor (In,Fe)As layer. A gate voltage is applied to control the electron wave functions  $\varphi_i$  in the QW, such that the overlap of  $\varphi_i$  and the (In,Fe)As layer is modified. The Curie temperature is largely changed by 42%, whereas the change in sheet carrier concentration is two to three orders of magnitude smaller than that of previous gating experiments. This result provides an approach for versatile, low power, and ultrafast manipulation of magnetization.

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Recently, the change of magnetic properties using an external gate voltage in field-effect transistor (FET) structure was demonstrated in carrier-induced ferromagnetic semiconductors (FMSs) [1–8] and ferromagnetic ultrathin metal films [9–14], which is expected to reduce the power consumption of spin devices. Commonly in these experiments, although the ferromagnetic channels are very thin (<5 nm), the two-dimensional (2D) quantization is smeared out because of the carriers' low coherency. The Curie temperature ( $T_C$ ) of these thin films monotonically depends on the carrier density  $n$ , which is varied by the effect of electrical gating, as predicted for three-dimensional (3D) ferromagnets [1–3]. A very large change in the sheet carrier density  $n_{\text{sheet}}$  ( $\Delta n_{\text{sheet}} = 10^{13}$ – $10^{14}$  cm<sup>-2</sup>) is typically required to generate a pronounced change in the magnetic properties, which in practice is difficult, and consumes a lot of energy ( $E \propto \Delta n_{\text{sheet}}^2$ ). Furthermore, the response time is limited by the device's capacitance and the carriers' transit time from outside electrodes to the channel, which is at shortest picoseconds.

In 2D ferromagnetic systems such as FMS quantum wells (QWs), however, the physics is different, as illustrated in the top panel of Fig. 1(a): Due to the quantization of the density of states,  $T_C$  does not change with the carrier density unless the Fermi level has reached the next subband [15]. Instead, theoretical studies suggested that  $T_C$  is very sensitive to the shape of the carrier wave function  $\varphi_i(z)$  of the occupied subband  $i$ , as described by the equation  $T_C \propto \sum_i \int_{\text{FMS}} |\varphi_i(z)|^4 dz$  [16–19], where  $z$  is the direction normal to the interface and the integral is performed inside the ferromagnetic layer in the QW (the fourth power is of great importance because it does *not* represent the carrier density, which is the square of the wave function, but represents the distribution of carriers in the ferromagnetic layer). Therefore, as illustrated in the bottom panel of Fig. 1(a), by using the gate voltage ( $V_G$ ) to deform the shape of the wave functions  $\varphi_i$  of the QW, one can effectively control the magnetic properties *without the need of changing the carrier concentration*. This method has two important advantages over the conventional method:

First, since no extra charge is needed, the charging energy of the FET's capacitor, which determines the power consumption for modulating magnetization, can be greatly reduced. Second, instead of traveling hundreds of nanometer-long paths as in the conventional method, electron carriers are redistributed over only a few nanometers in the QW. Therefore, the modulation speed can be as fast as  $\sim 100$  fs [20,21]. Another feature of this method is the ability to tune the relation between  $T_C$  and  $V_G$  at will by specifying the appropriate position and thickness of the ferromagnetic layer inside the QW [16–18]. However, experimental demonstration of such functionality has never been reported due to the lack of a good ferromagnetic QW so far.

Here we demonstrate such a control of ferromagnetism of 2D ferromagnetic QW via deforming the carrier wave function, using electrical gating in a FET structure. The system used in this study is a semiconductor InAs-based QW that contains a thin *n*-type FMS (In,Fe)As layer. Among the many types of FMSs, (In,Fe)As is unique and promising because it is the only reliable *n*-type FMS among III-V based semiconductors [22]. In this material, Fe atoms in the trivalent state Fe<sup>3+</sup> replace In atoms and thus play only the role of local magnetic moments. Electron carriers, which are independently supplied by defects or donors, interact strongly with these local Fe magnetic moments through the *s-d* exchange interaction to induce ferromagnetism with a  $T_C$  of up to several tens of Kelvin with a modest electron density ( $\sim 6 \times 10^{18}$  cm<sup>-3</sup>), and there is plenty of room for improvement [19,22–26]. We have observed that the electron carriers in (In,Fe)As reside in the conduction band and exhibit high coherency [19,24]. In fact, magnetic circular dichroism (MCD) spectroscopy has confirmed the quantum size effect (QSE) in QWs consisting of InAs/(In,Fe)As/InAs trilayers with a total thickness as large as 40 nm, in which the electron wave functions smoothly extend throughout the trilayers [19,25]. These features of the *n*-type FMS (In,Fe)As make it possible to demonstrate the electrical control of ferromagnetism via wave function manipulation.

We prepared a QW structure (sample A) consisting of InAs (2 nm)/(In<sub>0.94</sub>,Fe<sub>0.06</sub>)As (8 nm)/InAs (5 nm) on an AlSb buffer, all grown on a semi-insulating GaAs (001) substrate by molecular beam epitaxy [left panel in Fig. 1(b)] [27]. Because of the large conduction-band offset at the bottom

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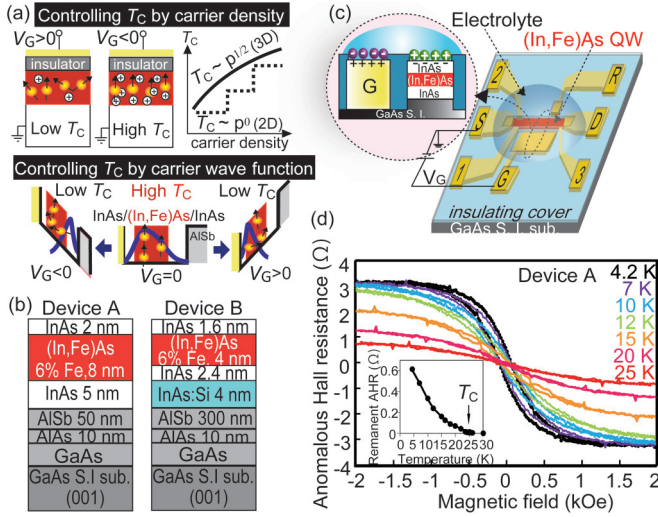


FIG. 1. (Color) (a) Schematic illustration of the two methods: Controlling  $T_C$  by carrier density as in previous works (top panel) and by carrier wave function as in the present study (bottom panel). Unlike 3D cases, in ferromagnetic QW,  $T_C$  does not change monotonically with the carrier density, which is due to the steplike change of the density of states. (b) Sample structures of device A (left panel) and device B (right panel). In both samples, the QW is the top InAs/(In<sub>0.94</sub>,Fe<sub>0.06</sub>)As/InAs trilayer structure. The Fe concentration is fixed at 6%. In device B, the bottom 5-nm InAs layer in the QW is doped with Si ( $5 \times 10^{18} \text{cm}^{-3}$ ). (c) Schematic structure of the FET device with electrolyte (DEME-TFSI) between the gate (G), the reference electrode (R), and the trilayer QW. The source (S) and drain (D) electrodes and the electrodes numbered 1, 2, and 3 are used for transport measurements. (d) Anomalous Hall resistance (AHR) of device A at various temperatures. The inset shows the temperature dependence of the remanent AHR, which indicates a  $T_C$  of 24 K.

InAs/AlSb interface (1.35 eV) [28], the electrons are confined to the top InAs/(In,Fe)As/InAs trilayer and form quantized electronic states. The QSE is evident in the blue shift of the MCD spectrum of sample A [27]. We used an electric-double-layer-type FET structure to control the carrier wave functions in the magnetic trilayer QWs, as illustrated in Fig. 1(c). The sample was etched into a  $50 \times 200 \mu\text{m}^2$  Hall bar using standard photolithography and ion milling. A side-gate electrode (G), a reference electrode (R), and several electrodes (source S, drain D, and electrodes numbered 1–3) for transport measurements were formed via the electron-beam evaporation and lift-off of an Au(50 nm)/Cr(5 nm) film. The side-gate pad (G), the reference pad (R), and the (In,Fe)As channel were covered with electrolyte [N,N-diethyl-N-methyl-N-(2-methoxyethyl)ammonium bis(trifluoromethylsulfonyl) imide, DEME-TFSI] to form the FET structure. Other regions of the device were separated from the electrolyte by an insulating resist. As illustrated in the inset of Fig. 1(c), when a positive  $V_G$  is applied, ions in the electrolyte accumulate at the surface of the semiconductor channel and form an electric-double-layer capacitor, which changes the potential and electron density in the InAs/(In,Fe)As/InAs trilayer QW.

The transport and magnetic properties of the trilayer QW were characterized by Hall measurements [27]. The Hall resistance of (In,Fe)As always consists of a large  $n$ -type normal

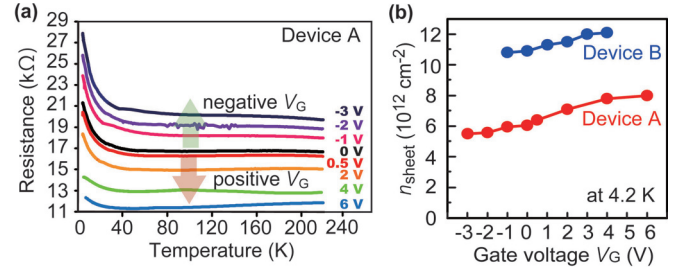


FIG. 2. (Color online) (a) Temperature dependence of the  $R_{13}$  of device A at various gate voltage  $V_G$  values. (b)  $n_{\text{sheet}}$  values of device A (red circles) and device B (blue circles) at 4.2 K as functions of  $V_G$ .

Hall resistance (NHR) and a much smaller anomalous Hall resistance (AHR) that is proportional to the magnetization. The small AHR ( $\sim 3\%$  of the total Hall resistance) can be obtained by subtracting the negative slope of the NHR at a high magnetic field (1 T) from the raw Hall-resistance data. The large contribution of the NHR in the Hall-resistance data allows us to accurately estimate the electron density from the Hall measurements.  $T_C$  was estimated by monitoring the temperature dependence of the remanent AHR. Figure 1(d) presents the AHR of the trilayer QW in device A at  $V_G = 0$  V at various temperatures, in which clear ferromagnetic hysteresis is observed at low temperatures. The inset presents the remanent AHR vs temperature  $T$ , indicating a  $T_C$  of 24 K.

Figure 2(a) shows the temperature dependence of the resistance ( $R$ - $T$ ) measured between terminals 1 and 3 of device A ( $R_{13}$ ) using a four-terminal method.  $R_{13}$  systematically decreases (increases) when a positive (negative)  $V_G$  is applied, as expected. Figure 2(b) shows  $n_{\text{sheet}}$  in the InAs/(In,Fe)As/InAs trilayer QW at various  $V_G$  values estimated by Hall measurements at 4.2 K. The  $n_{\text{sheet}}$  of device A (red circles) changes from  $6.06 \times 10^{12} \text{cm}^{-2}$  (at  $V_G = 0$  V) to  $8 \times 10^{12} \text{cm}^{-2}$  (at  $V_G = 6$  V) or to  $5.52 \times 10^{12} \text{cm}^{-2}$  (at  $V_G = -3$  V). The  $\Delta n_{\text{sheet}}$  is small, probably because of the existence of a large number of surface states at the top InAs layer, which tends to pin the Fermi level.

Figures 3(a) and 3(c) show the evolution of the AHR of device A at 15 K when  $V_G$  is adjusted as follows:  $0 \text{ V} \rightarrow 0.5 \text{ V} \rightarrow 6 \text{ V} \rightarrow 0 \text{ V}$  and  $0 \text{ V} \rightarrow -3 \text{ V} \rightarrow 0 \text{ V}$ , respectively. At  $V_G = 6$  and  $-3$  V, the hysteresis in the AHR- $H$  characteristics almost disappears, whereas it recovers after  $V_G$  is returned to 0. These results demonstrate that the ferromagnetism of the (In,Fe)As thin film can be reversibly controlled by electrical gating. Figures 3(b) and 3(d) show the remanent AHR vs  $T$  of device A at various positive and negative  $V_G$  values, respectively, and illustrate the evolution of  $T_C$ . As summarized in Fig. 4(a), the  $T_C$  of the (In,Fe)As layer of device A (black circles) decreases from its initial value of 24 K ( $V_G = 0$  V) to 17 K ( $V_G = 6$  V) or 14 K ( $V_G = -3$  V).

Compared with previous electrical-gating experiments [1–14], the present results for device A exhibit two distinct features. First, very effective control of  $T_C$  is realized (the largest change in  $T_C$ ,  $\Delta T_C/T_C = [T_C(V_G) - T_C(0)]/T_C(0)$ , is  $-42\%$  at  $V_G = -3$  V) despite a very small change in  $n_{\text{sheet}}$  (the corresponding  $\Delta n_{\text{sheet}} = -5.4 \times 10^{11} \text{cm}^{-2}$ ). This  $\Delta n_{\text{sheet}}$  is two to three orders of magnitude smaller than the change of the  $n_{\text{sheet}}$  that was required in almost all the previous gating

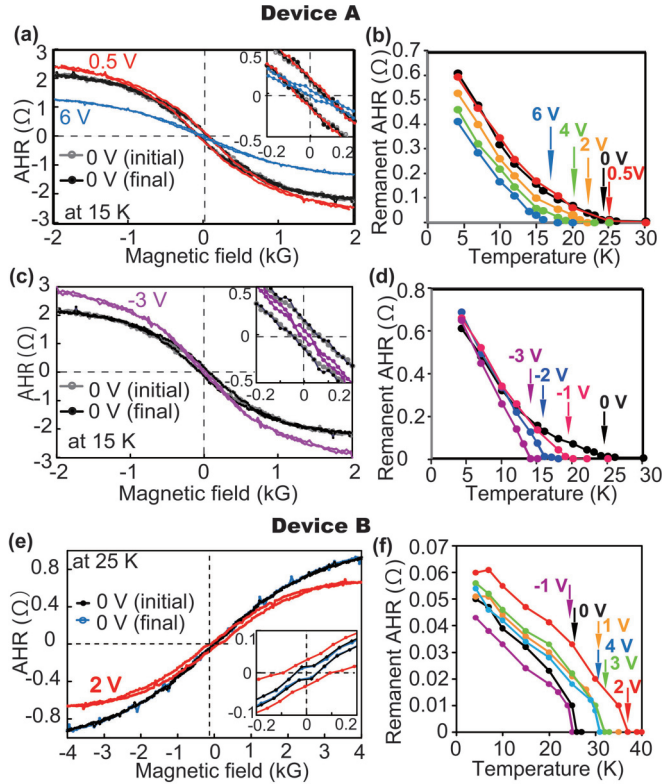


FIG. 3. (Color) (a), (c) Evolution of the AHR of device A measured at 15 K when adjusting  $V_G$  as follows:  $0\text{ V} \rightarrow 0.5\text{ V} \rightarrow 6\text{ V} \rightarrow 0\text{ V}$  and  $0\text{ V} \rightarrow -3\text{ V} \rightarrow 0\text{ V}$ , respectively. The insets present the AHR data near the origin, which exhibit clear changes in hysteresis characteristics. (b), (d) Temperature dependence of the remnant AHR of device A at various  $V_G$  values. The colored arrows indicate the  $T_C$  values of device A at each  $V_G$ . (e) Evolution of the AHR of device B measured at 25 K when adjusting  $V_G$ :  $0\text{ V} \rightarrow 2\text{ V} \rightarrow 0\text{ V}$ . The inset presents the AHR data near the origin. (f) Temperature dependence of the remnant AHR of device B at various  $V_G$  values. The colored arrows indicate the  $T_C$  values of device B at each  $V_G$ .

experiments (the only exceptions whose  $\Delta n_{\text{sheet}}$  are comparable with the present study are CdMnTe QW [6] and GeMn quantum dots [7], in which, however, the ferromagnetism was turned off by depleting carriers in the ferromagnetic channels). This means that the energy for charging the ferromagnetic channel is dramatically reduced by a factor of  $10^{-4}$ – $10^{-6}$  while maintaining the same effect of modulating the ferromagnetism. Second,  $T_C$  varies nonmonotonically with  $n_{\text{sheet}}$ , reaching a maximum near  $V_G = 0.5\text{ V}$  and decreasing at both positive and negative  $V_G$ . This is characteristic of 2D FMS QWs, in which the overlap of the electron wave functions and the (In,Fe)As layer determines the single  $T_C$  value of the entire system. Figure 4(b) presents the changes in the potential profile (blue curves) and the electron wave functions (yellow shapes) of the QW in device A at  $V_G = 0, 6$ , and  $-3\text{ V}$ , which were obtained through self-consistent calculations [27]. The calculated electron-density distribution in the QW versus  $V_G$  is summarized in Fig. 4(c). The integral of the electron-density distribution over the entire QW yields  $n_{\text{sheet}}$ . The overlap of the wave functions and the local Fe magnetic moments in the (In,Fe)As layer is largest at  $V_G = 0.5\text{ V}$  and decreases as the

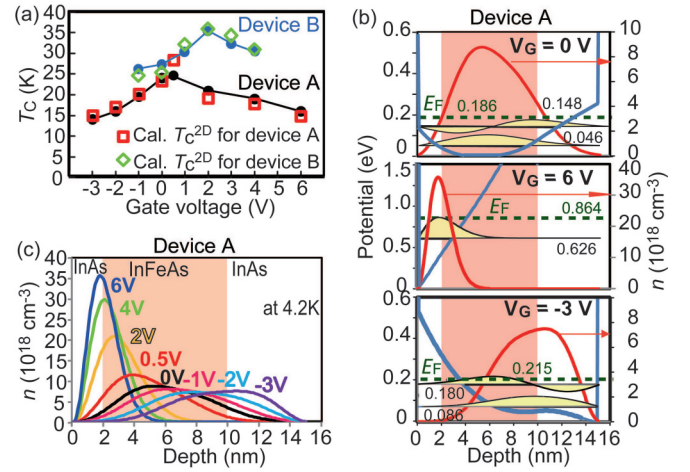


FIG. 4. (Color online) (a) Experimental  $T_C$  values of device A (black circles) and device B (blue circles), respectively, as functions of  $V_G$ . Red squares and green diamonds represent the  $T_C^{2D}$  values calculated using the 2D mean-field Zener model for device A and B, respectively, which exhibit good agreement with the experimental results. (b) Calculated potential profiles (blue curves), electron wave functions (yellow shapes), and electron-density distributions (red curves, with arrows pointing to the corresponding right-hand vertical axes) of the QW of device A at  $V_G = 0, 6$ , and  $-3\text{ V}$ , from top to bottom. The Fermi levels (green dashed lines) and quantized energy levels (black lines, in the eV unit) in the trilayer QWs are shown. (c) Calculated evolution of the electron-density distribution in the trilayer QW of device A at 4.2 K when  $V_G$  is varied from  $-3\text{ V}$  to  $6\text{ V}$ . The reddish regions correspond to the (In,Fe)As layer.

wave functions move towards the ends of the trilayer QW at both positive and negative  $V_G$ , leading to the observed behavior of  $T_C$ . These results clearly indicate the feasibility of controlling the ferromagnetism via wave function manipulation and confirm the intrinsic electron-induced ferromagnetism of n-type FMS (In,Fe)As.

In a 2D FMS structure,  $T_C$  is given by [16,17,19]

$$T_C^{2D} = \frac{S(S+1)}{12} \frac{A_F^{2D} J_{sd}^2}{k_B \pi \hbar^2} N_{\text{Fe}} \sum_{E_i < E_F} \int_{(\text{In,Fe)As}} |\varphi_i(z)|^4 dz. \quad (1)$$

Here,  $z$  is the growth direction,  $S$  is the spin angular momentum of an Fe atom ( $=5/2$ ),  $m^*$  is the effective electron mass at the  $\Gamma$  point,  $k_B$  is the Boltzmann constant,  $J_{sd}$  is the  $s$ - $d$  exchange interaction,  $A_F^{2D}$  is the Stoner enhancement factor in 2D structures [29], and  $N_{\text{Fe}}$  is the Fe atom density. The calculated  $T_C^{2D}$  values [red squares in Fig. 4(a)] exhibit good agreement with the experimental results. It is worth noting that a semiclassical approach based on a modified 3D mean-field Zener model, in which the QSE is ignored, cannot explain the behavior of  $T_C$  observed in the present experiment [27]. From the calculations of  $T_C^{2D}$ , the effective  $s$ - $d$  exchange interaction energy  $N_0\alpha$  of the (In,Fe)As QW was estimated to be 3.6 eV. This effective  $N_0\alpha$  value was calculated from the value of  $(A_F^{2D})^{1/2} J_{sd}$ , which implicitly includes the enhancement effect associated with electron-electron interactions in 2D structures.

Finally, we demonstrate the ability to control the relation between  $T_C$  and  $V_G$  by modifying the QW structure.



Another sample and its corresponding FET device were prepared (device B) using the same procedure but a modified QW structure: InAs(1.6 nm)/(In<sub>0.94</sub>,Fe<sub>0.06</sub>)As (4 nm)/InAs (2.4 nm)/Si-doped InAs(4 nm) [right panel of Fig. 1(b)]. Si donors ( $5 \times 10^{18} \text{ cm}^{-3}$ ) in the bottom 4-nm InAs layer supply electrons to the QW and attract the 2D electron wave functions towards the bottom AISb side. This feature, together with the reduced thickness (4 nm) of the (In,Fe)As layer, creates a situation in which the wave functions and Fe magnetic moments are separated when  $V_G = 0$ ; however, their overlap increases as a positive  $V_G$  is applied. As shown in Fig. 2(b), device B exhibits a higher  $n_{\text{sheet}}$  ( $1.08 \times 10^{13} \text{ cm}^{-2}$  at  $V_G = 0$ ) than device A but a weak dependence of  $n_{\text{sheet}}$  on  $V_G$ , similar to device A. Figure 3(e) shows the evolution of the AHR- $H$  characteristics of device B at 25 K for  $V_G = 0 \text{ V} \rightarrow 2 \text{ V} \rightarrow 0 \text{ V}$ , and Fig. 3(f) summarizes the remanent AHR vs  $T$  curves at various  $V_G$  values. The sign of AHR in device B is positive, which is opposite to that of device A. The QWs in devices A and B are different in the inversion asymmetry (due to the space charge potential of activated Si donors in device B) and  $n_{\text{sheet}}$ , both of which could be the origin of the different AHR sign [30,31]. Positive AHR has also been observed in bulklike (In,Fe)As samples doped with Be that have high electron density [22]. Thus, at this stage we suggest that the sign change of AHR is probably due to the difference in  $n_{\text{sheet}}$ , although a more elaborate study is needed to clarify the mechanism. As can be seen in Fig. 3(e), unlike device A, the ferromagnetism of the (In,Fe)As layer in device B is enhanced by applying positive  $V_G$ , as expected, and  $T_C$  increases from 27 K (at  $V_G = 0 \text{ V}$ ) to a maximum of 35 K (at  $V_G = 2 \text{ V}$ ) or to 30 K (at  $V_G = 4 \text{ V}$ ), as illustrated in Fig. 4(a) (blue circles). The  $T_C$  values calculated by the 2D mean-field Zener model for device B are plotted as green diamonds, which also show

good agreement with the experimental values (see Ref. [27] for details of the calculations). Thus, using the appropriate QW structure, one can either increase or decrease the  $T_C$  of the (In,Fe)As layer in QWs with the same device configuration. This high degree of freedom in controlling the ferromagnetism is an important advantage, which is attractive for device applications.

In summary, we have demonstrated the electrical control of ferromagnetism in (In,Fe)As QWs by manipulating the overlap between the 2D wave functions and the (In,Fe)As layer. The  $T_C$  was largely changed by 10 K ( $\Delta T_C/T_C = 42\%$ ), whereas the change in  $n_{\text{sheet}}$  is two to three orders of magnitude smaller than that of previous gating experiments. The behavior of  $T_C$  was quantitatively explained by the mean-field Zener model for FMS QWs. We have also demonstrated the ability to customize the  $T_C$ - $V_G$  relation by modifying the QW structure. These results confirm the intrinsic electron-induced ferromagnetism of n-type FMS (In,Fe)As and open up possibilities of using this material in spintronic devices, as well as provide an approach for versatile, low power, and ultrafast manipulation of magnetization.

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