Ferromagnetic Interfacial Interaction and the Proximity Effect in a $Co_2FeAl/(Ga,Mn)As$ Bilayer

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The magnetic properties of a $Co_2FeAl/(Ga,Mn)As$ bilayer epitaxied on GaAs (001) are studied both experimentally and theoretically. Unlike the common antiferromagnetic interfacial interaction existing in most ferromagnet-magnetic semiconductor bilayers, a ferromagnetic interfacial interaction in the $Co_2FeAl/(Ga,Mn)As$ bilayer is observed from measurements of magnetic hysteresis and x-ray magnetic circular dichroism. The Mn ions in a 1.36 nm thick (Ga,Mn)As layer remain spin polarized up to 400 K due to the magnetic proximity effect. The minor loops of the $Co_2FeAl/(Ga,Mn)As$ bilayer shift with a small ferromagnetic interaction field of $+24$ Oe and -23 Oe at 15 K. The observed ferromagnetic interfacial coupling is supported by *ab initio* density functional calculations. These findings may provide a viable pathway for designing room-temperature semiconductor spintronic devices through magnetic proximity effect.

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As a model system of diluted magnetic semiconductors (DMSs), (Ga,Mn)As has been investigated extensively in the past decades due to its promising potential in semiconductor spintronic devices [1,2]. However, the low Curie temperature (T_C) of (Ga,Mn)As is the main obstacle for practical applications. The highest T_C reported up to now is only 200 K even after combining heavy Mn doping, postgrowth nanopatterning, and annealing [3]. Recently, the exchange coupled bilayers of ferromagnet-DMS (FM-DMS), e.g. $Fe/(Ga,Mn)As$, have attracted a great deal of attention. These bilayer structures could not only provide a new platform for designing room temperature ferromagnetic DMSs but also offer another pathway to combine semiconductor-based spintronics with metal-based spintronics, providing an entrance to the novel interfacial magnetic coupling phenomena and spintronic application. However, the coupling mechanism at the interface of FM-DMS is still ambiguous. Mark *et al.* reported independent magnetic behavior in $NiFe/(Ga,Mn)As$ bilayers, while Zhu *et al.* observed exchange coupling in $MnAs/(Ga,Mn)As$ bilayers [4,5]. Remarkably, magnetic proximity effect up to room temperature at $Fe/(Ga,Mn)As$ interfaces was reported [6–8]. Recently, a substantial increase of T_C from 40 to 70 K due to the presence of a few monolayers of Fe on top of a 5 nm (Ga,Mn)As layer was also reported [9]. All these results show a promising route to enhance T_C of (Ga,Mn)As by depositing a ferromagnetic metal or alloy epilayer on it.

On the other hand, cobalt-based Heusler alloys are desirable spintronic materials due to their high spin polarization, low Gilbert damping constant, and high T_C [10–12]. High-quality Heusler alloy Co_2FeAl could be well epitaxied on GaAs (001), showing an in-plane uniaxial magnetic anisotropy with an easy axis along the [110] direction [13]. Since (Ga,Mn)As film could also be well epitaxied on GaAs (001) with an easier axis along the $[-110]$ direction at low temperature [14], $Co_2FeAl/(Ga,Mn)As$ bilayers should be an attractive epitaxial system to explore a substantial increase of T_C for the (Ga,Mn)As layer through magnetic proximity effect. These kinds of bilayers may also exhibit other interesting magnetic behaviors due to their different magnetic anisotropy and interfacial magnetic coupling.

In this Letter, the structural characterization and magnetic properties of the high-quality $Co_2FeAl/(Ga,Mn)As$ bilayer grown on a GaAs (001) substrate by molecular-beam epitaxy are investigated. Magnetization and x-ray circular magnetic dichroism (XMCD) measurements reveal a ferromagnetic coupling at the interface between Co_2FeAl and (Ga,Mn)As layers, which is further supported by *ab initio* density functional calculations. Remarkably, Mn ions extending 1.36 nm in the (Ga,Mn)As layer remain spin polarized up to 400 K.

To guarantee high quality of the interface, a $Co_2FeAl/(Ga,Mn)As$ bilayer was grown by a molecular-beam epitaxy system with two growth chambers (VG80) without being exposed to air during the entire growth

process. A 150 nm $Ga_{1-x}Mn_xAs$ layer with $x = 0.07$ was first deposited onto a GaAs (001) buffer layer in the first growth chamber at a substrate temperature $T_s = 250\,^{\circ}C$. The clear (1×1) streaky pattern of reflection high-energy electron diffraction implied high crystalline quality and a smooth surface. No evidence of precipitation of the second phase could be observed during deposition. Subsequently, without breaking vacuum, the sample was transferred via an ultrahigh vacuum channel to the second growth chamber, where a 3 nm Co_2FeAl film was deposited on the (Ga,Mn)As layer at $T_s = 160\,^{\circ}C$. Finally, the structure was capped with a 2 nm Al layer to avoid oxidation. For reference, a single (Ga,Mn)As layer was obtained by etching the metal layer away through dipping a piece of bilayer into diluted hydrochloric acid.

In order to check the crystal structure of the Co_2FeAl/(Ga,Mn)As bilayer, high-resolution double-crystal x-ray diffraction (DCXRD) was performed and the result is shown in Fig. 1. Apart from the (004) and (002) diffraction peaks of GaAs and (Ga,Mn)As, it is difficult to see the XRD peak of the Co_2FeAl layer. This is because the thickness of this layer is too thin, and the position of the XRD peak of this layer is so close to those of (Ga,Mn)As and GaAs that the weak XRD signal of this ultrathin layer is merged into the tails of XRD peaks of (Ga,Mn)As and GaAs [13]. On the other hand, no evidence for a secondary phase, such as MnAs related peaks, can be observed. The full width at half maximum of (Ga,Mn)As is small, indicating high quality of the (Ga,Mn)As layer. To further examine the microstructure of the bilayer, high-resolution cross-sectional transmission electron microscopy (HRTEM) was performed. As shown in the left inset of Fig. 1, both Co_2FeAl and zinc-blende (Ga,Mn)As layers are single crystalline, and a sharp interface exists between them. No detectable MnAs clusters can be seen in the (Ga,Mn)As matrix.

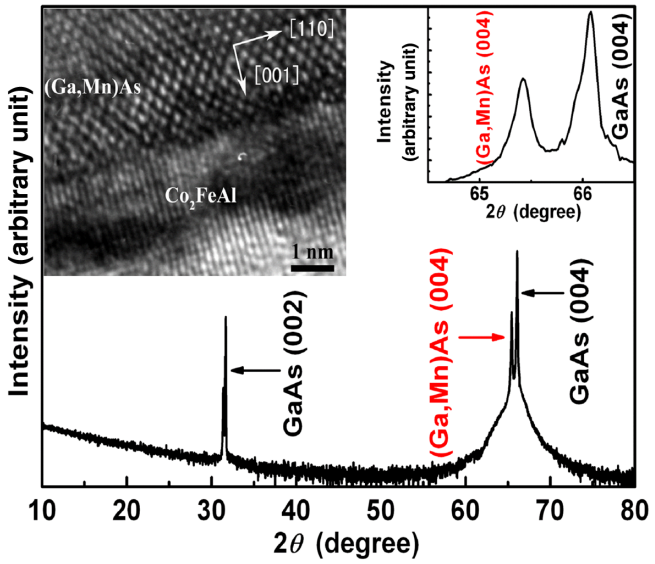

FIG. 1 (color online). A DCXRD curve of a Co_2FeAl/(Ga,Mn)As bilayer. The left inset is the HRTEM image and the right inset is a close-up view around the (Ga,Mn)As (004) peak.

Magnetic measurements were first performed using a superconducting quantum interference device magnetometer. We cooled the samples from 300 to 5 K in a magnetic field of 1 T. Then, we began measurement of the temperature dependence of the remanent magnetization (M_r) of the Co_2FeAl/(Ga,Mn)As bilayer and the reference sample of a single (Ga,Mn)As layer without a magnetic field by heating the sample from 5 to 350 K. As shown in Fig. 2, the M_r of the single (Ga,Mn)As layer becomes zero when temperature is about 50 K, indicating a pure DMS phase. All the $M_r - T$ curves along different directions have an inflexion near 50 K, corresponding to the low T_C of the (Ga,Mn)As layer. The much higher M_r along the [110] direction than that along the [−110] direction from 50 to 350 K is ascribed primarily to the strong uniaxial magnetic anisotropy of the Co_2FeAl layer as the bulk (Ga,Mn)As layer is already in the paramagnetic phase in this temperature range. The magnetic property of a single Co_2FeAl layer grown on GaAs (001) depends on the competition between uniaxial magnetic anisotropy and the fourfold crystalline anisotropy [13]. Uniaxial magnetic anisotropy with an easy axis along the [110] direction plays the main role for the 3 nm Co_2FeAl layer in this study, resulting in a large M_r along the [110] direction and a small M_r along the [−110] direction. This is also supported by the hysteresis loops measured after zero magnetic field cooling (ZFC) along the two directions at different temperatures, as shown in Fig. 3.

Furthermore, one can see from Fig. 3(a) that the hysteresis loop of the bilayer measured at 15 K along the [110] direction exhibits clear symmetrical double-step switching of the Co_2FeAl layer and (Ga,Mn)As layer. The measurement begins at 2 T, with the magnetization of the bilayer pointing along the field direction. As the magnetic field decreases, the magnetization of the (Ga,Mn)As reverses at −40 Oe while the magnetization of the Co_2FeAl reverses at −100 Oe. We also measured the hysteresis loop along the [110] direction at 15 K after cooling in a magnetic field of 1 T; no exchange bias effect was observed (see Supplemental

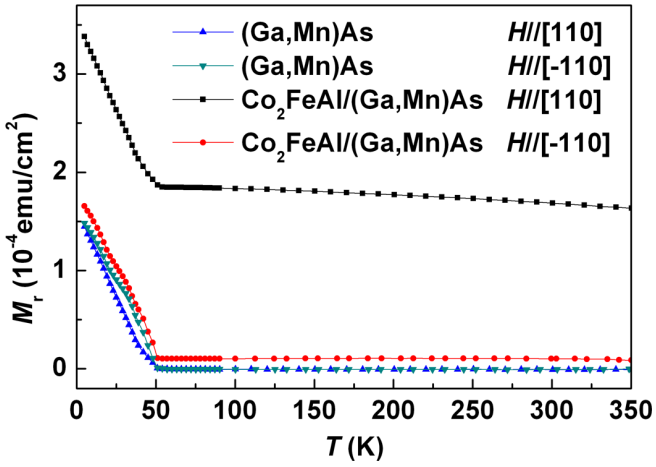

FIG. 2 (color online). Temperature dependence of the remanent magnetization of the Co_2FeAl/(Ga,Mn)As bilayer and the single (Ga,Mn)As layer along the [110] and [−110] directions.

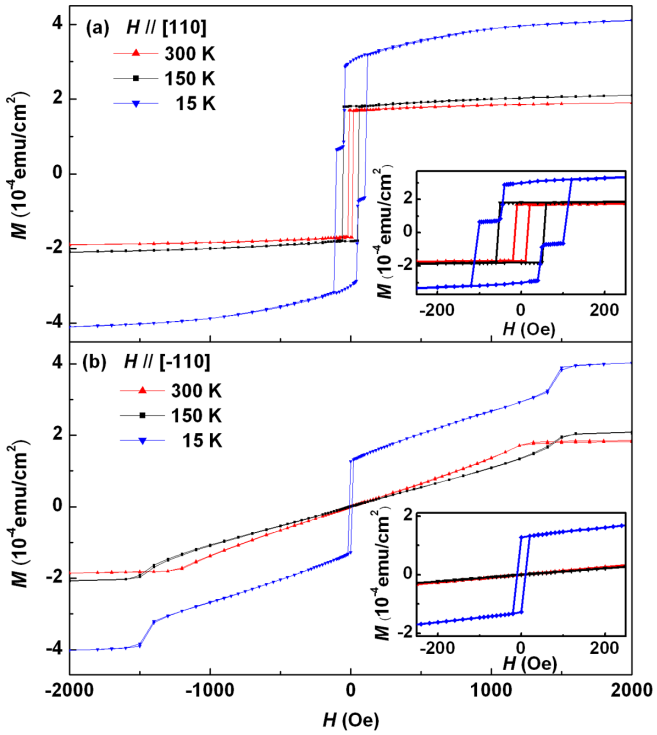

FIG. 3 (color online). Magnetic hysteresis of a Co_2FeAl/(Ga,Mn)As bilayer at 15, 150, and 300 K. The magnetic field was along the [110] (a) and [−110] directions (b). The insets are close-ups around a zero magnetic field.

Material [15]). With increasing temperature to 150 and 300 K, where the (Ga,Mn)As bulk layer is paramagnetic, the hysteresis loops of the bilayer along the [110] direction only show one step, which behave almost the same as that of the single Co_2FeAl layer. Similar to the work in Ref. [13], there is a split field appearing in the hysteresis loop of the bilayer along the [−110] direction at 15 K, but the hysteresis loops become almost linear at 150 K and 300 K.

The minor loops of the Co_2FeAl/(Ga,Mn)As bilayer were measured at 15 K along the [110] direction after cooling from 300 to 15 K with and without an external magnetic field of 1 T, as shown in Fig. 4. The typical minor loop 1 begins at +2 T, decreases to −70 Oe, then sweeps back from −70 Oe to +2 T. The minor loop 2 begins at −2 T, decreases to +70 Oe, then back from +70 Oe to −2 T. The minor loops attributed to the (Ga,Mn)As layer shift with interaction fields of −23 Oe and +24 Oe, indicating that the Co_2FeAl layer induces an interfacial interaction field in the magnetic semiconductor (Ga,Mn)As. The minor loops measured after ZFC and FC almost overlap, which means FC is unnecessary, because both the Co_2FeAl and (Ga,Mn)As layers are reoriented at 2 T. The shape and size of the minor loops are in agreement with the hysteresis loop for the single (Ga,Mn)As layer. This strongly indicates that the interfacial interaction affects the entire (Ga,Mn)As layer in the bilayer sample. Similar results were also reported recently by other groups [5,8]. An interfacial interaction field in (Ga,Mn)As induced by antiferromagnetic coupling between Fe and Mn was

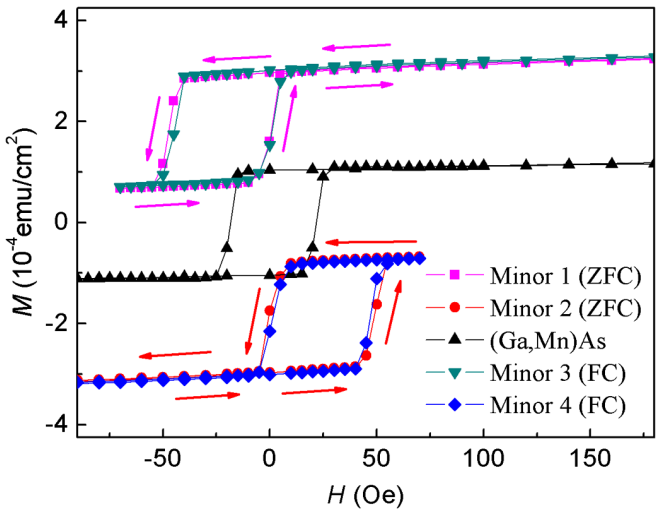

FIG. 4 (color online). Minor loops of a Co_2FeAl/(Ga,Mn)As bilayer and major loop of a (Ga,Mn)As reference sample at 15 K along the [110] direction.

demonstrated by both XMCD and superconducting quantum interference device measurements. The interfacial magnetic coupling plays an important role not only in ferromagnet-antiferromagnet (FM/AFM) exchange bias systems in general [16] but also in the FM-DMS systems here in particular. We will offer further evidence of interfacial magnetic interaction in the bilayer by XMCD measurements below.

We probed the interfacial magnetic coupling of the Co_2FeAl/(Ga,Mn)As bilayer in further detail with the help of element-resolved XMCD measurements. Figures 5(a)–5(c) show the Mn $L_{2,3}$ XMCD spectrum of the single (Ga,Mn)As layer at 300 K and Mn, Fe and Co $L_{2,3}$ XMCD spectra of the Co_2FeAl/(Ga,Mn)As bilayer measured at 22, 100, 300, and 400 K along the [110] direction. All the spectra were acquired in total electron yield mode and normalized by the intensity of the incident beam. Moreover, the signal was detected employing remnant magnetization after turning off a magnetic field of 2000 Oe. Large XMCD signals have been found for Co and Fe elements from the Co_2FeAl layer, which are similar to that of a 30 nm thick Co_2FeAl as reported in Ref. [13], confirming a high remnant magnetization of the 3 nm thick Co_2FeAl along the [110] direction as a result of strong uniaxial magnetic anisotropy of thin Co_2FeAl. A small but unambiguous magnetic signal was obtained in the Mn spectrum of the Co_2FeAl/(Ga,Mn)As bilayer even when the temperature was increased to 400 K, while none was found in the single (Ga,Mn)As layer at 300 K. We can exclude the presence of secondary phases existing in (Ga,Mn)As such as MnAs because T_C of MnAs films is lower than 330 K [17]. We can also exclude the presence of $Co_2Fe_{1-x}Mn_xAl$, because the shape of the Mn x-ray absorption spectroscopy and XMCD curves of $Co_2Fe_{1-x}Mn_xAl$ is different from that of the spectra measured for the bilayer, and the peak position of Mn L_3 for $Co_2Fe_{1-x}Mn_xAl$ is 635 eV in contrast to 639.4 eV for

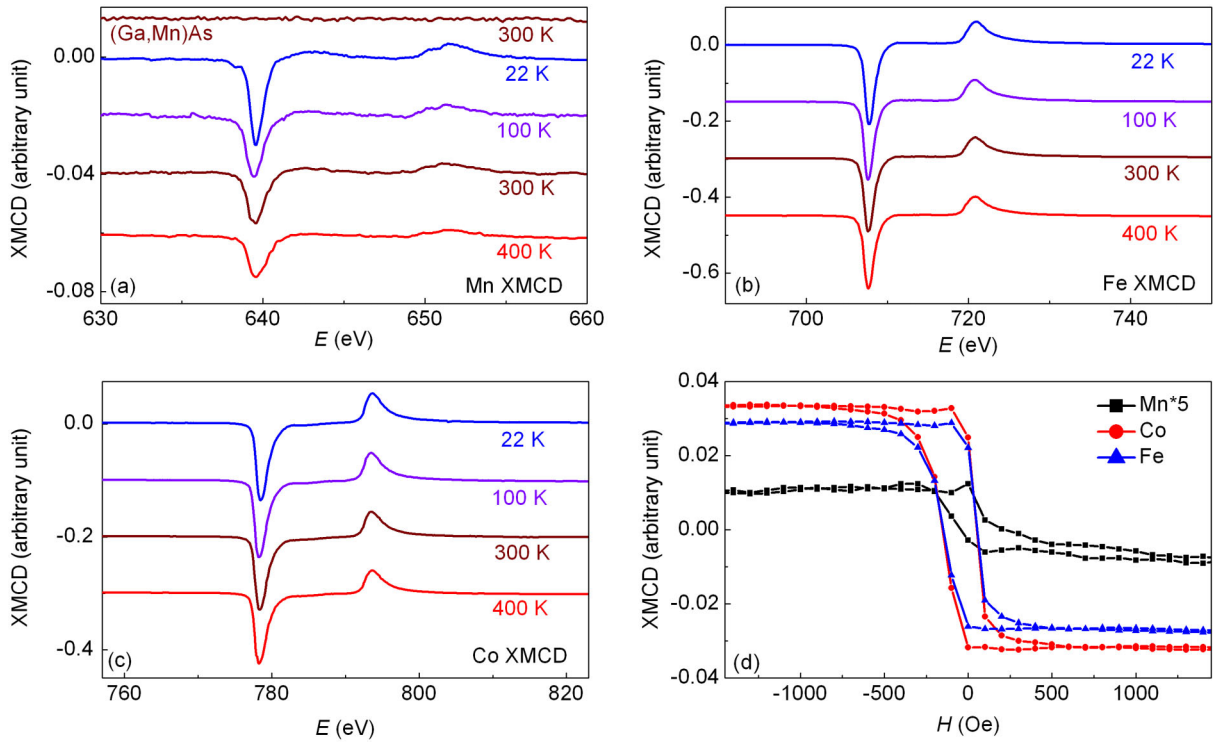

FIG. 5 (color online). XMCD spectra of (a) Mn $L_{2,3}$ of the single (Ga,Mn)As layer and Co_2FeAl/(Ga,Mn)As bilayer; (b) Co $L_{2,3}$ and (c) Fe $L_{2,3}$ of Co_2FeAl/(Ga,Mn)As bilayer at different temperatures. (d) XMCD loops of Mn (square), Co (circle), and Fe (triangle) of the Co_2FeAl/(Ga,Mn)As bilayer at 400 K.

the Co_2FeAl/(Ga,Mn)As bilayer [13]. Considering the paramagnetic phase of the bulk (Ga,Mn)As layer at 400 K [3], we therefore conclude that the spin polarization of Mn is induced by the Co_2FeAl cap layer due to the magnetic proximity effect, consistent with the situation of Fe/(Ga,Mn)As bilayers at room temperature [6,8].

In addition, the XMCD signals of Fe and Mn are opposite in sign from those in Refs. [6,8]. In our study, Fe, Co, and Mn display parallel alignments at different temperatures, confirmed by both the sign of the XMCD spectra and the element specific XMCD hysteresis loops as shown in Fig. 5. In Fig. 5(d), a small Mn magnetic signal is found without an applied magnetic field, with a coercive field identical to that of Co and Fe, proving a strong ferromagnetic interaction in the bilayer. It is similar to the XMCD results of $Co_2Fe_{1-x}Mn_xAl(x = 0.3)$ grown at 160 °C [13]. As x increases to 1, the Mn XMCD signal vanishes due to strong Mn–Mn antiferromagnetic interaction [13]. The Mn concentration is too low to form Mn-Mn, Co-Mn, and Fe-Mn antiferromagnetic coupling in our case. On the contrary, Co-Co, Co-Fe, Fe-Fe, Co-Mn, and Fe-Mn couplings favoring ferromagnetic interactions must be taken into account. As a result, parallel alignment is more favorable than antiparallel alignment at the Co_2FeAl/(Ga,Mn)As interface. Note that a reorientation transition of the magnetic proximity polarization as a function of the (Ga,Mn)As thickness was reported very recently [18], which is, however, not seen here at different temperatures from 22 to 400 K even when we reduced the thickness of the (Ga,Mn)As in the Co_2FeAl/(Ga,Mn)As bilayers from 150 to 5 nm (see Supplemental Material [15]).

We also calculated the thickness of the ferromagnetic (Ga,Mn)As layer induced by magnetic proximity effect based on the model proposed by Maccherozzi *et al.* [6], in which the Co_2FeAl/(Ga,Mn)A bilayer was divided into four parts: (i) the (Ga,Mn)As bulk region, (ii) the interface region with induced ferromagnetic order, (iii) the Co_2FeAl layer, and (iv) the Al overlayer. We found that Mn ions in depths of 2.11 and 1.36 nm into the (Ga,Mn)As layer remain spin polarized up to 300 and 400 K, respectively. For details, see the Supplemental Material [15].

To help in understanding our experimental results, we have also performed detailed *ab initio* density functional calculations with the generalized gradient approximation [19] for the Co_2FeAl/(Ga,Mn)As bilayers. We used the accurate frozen-core full-potential projector-augmented wave method, as implemented in the Vienna *ab initio* simulation package [20,21]. We considered both the ordered $L2_1$ and disordered $B2$ phases for the (001) Co_2FeAl layer of one cubic layer (four formula units) thickness, although the Co_2FeAl layer in our samples was found experimentally to be in the $B2$ phase. The disordered $B2$ Co_2FeAl consists of two simple cubic sublattices, namely, Co sublattice and mixed (Fe,Al) sublattice. We generated many (Fe,Al) atomic configurations for the (Fe,Al) sublattice and chose four low-energy configurations for the $B2$ Co_2FeAl/(Ga,Mn)As bilayers. Two cubic layers (eight formula units) were used to model

the (001) (Ga,Mn)As layer with 1/8 Mn concentration, and both the (Ga,Mn)- and As-terminated interface layer are considered. The bilayer was simulated by the slab-supercell approach with a vacuum layer of ~1.5 nm. The atomic positions were optimized theoretically under the constraint that the in-plane lattice constant was fixed to the experimental GaAs lattice constant of 0.565 nm. The equilibrium structure was obtained when all the forces acting on the atoms were smaller than 0.2 eV/nm. A large plane-wave cutoff energy of 350 eV and a Γ-centered Monkhorst-Pack k mesh of $10 \times 10 \times 1$ were used. The total energy convergence criterion is 10^{-5} eV/atom.

Our total energy calculations show that in all the considered $B2$ phase Co_2FeAl/(Ga,Mn)As bilayers with the (Ga,Mn)-terminated interface, the ferromagnetic coupling across the interface between Fe (Co) and Mn is energetically favored, while in all the $L2_1$ phase Co_2FeAl/(Ga,Mn)As bilayers, the antiferromagnetic coupling between Fe (Co) and Mn is more stable. The discovered FM coupling is rather strong, being ~10 meV per Mn atom or larger. In the $B2$ phase Co_2FeAl/(Ga,Mn)As bilayers with the As-terminated interface, both stable FM and AF couplings across the interface are found, depending on the atomic configuration of the (Fe,Al) sublattice. Furthermore, the local Co (Fe) magnetic moments in the FM-coupled bilayers are large and similar to that in the bulk $L2_1$ phase Co_2FeAl [being $\sim 1.3(2.6)\mu_B$/atom]. The Mn atomic moment is also large and varies from 1.8 to $3.7\mu_B$/atom, depending on the atomic configuration.

In summary, we presented an investigation on the structural and magnetic properties of the Co_2FeAl/(Ga,Mn)As bilayer grown epitaxially on GaAs (001) by molecular-beam epitaxy. Both (Ga,Mn)As and Co_2FeAl layers are single crystalline as proved by DCXRD and HRTEM. Unlike the common antiferromagnetic exchange existing in most ferromagnet/(Ga,Mn)As bilayers, XMCD data reveal that Co_2FeAl and (Ga,Mn)As are ferromagnetic exchange coupled, which is supported by *ab initio* density functional calculations. The Mn ions of about 1.36 nm into the (Ga,Mn)As layer remain spin polarized up to 400 K due to the magnetic proximity effect. Such ferromagnetic interfacial coupling may provide a novel route to manipulating the DMS spin state and offering room temperature operability through the magnetic proximity effect in spintronic devices.

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[15] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.111.027203 for information regarding the calculation of the thickness of ferromagnetic (Ga,Mn)As layer induced by Co_2FeAl; XMCD spectra of the Co_2FeAl/(Ga,Mn)As bilayers with different thickness of (Ga,Mn)As; magnetic hysteresis loops of the Co_2FeAl/(Ga,Mn)As bilayer measured after ZFC and FC.
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