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Metallic Conduction and Ferromagnetism in
MAI$_2$O$_4$/SrTiO$_3$ Spinel/Perovskite Heterostructures
($M = \text{Fe, Co, Ni}$)

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Abstract:

Recently, a high mobility quasi-two-dimensional electron gas (q-2DEG) has been reported for the heterointerface between two insulating and nonmagnetic oxides of spinel $\gamma$-Al$_2$O$_3$ and perovskite SrTiO$_3$ (STO). Herein, we fabricated the epitaxial heterostructure with Al-based magnetic spinel oxide $M$Al$_2$O$_4$ ($M =$ Fe, Co, Ni) on perovskite STO. Remarkably, all the $M$Al$_2$O$_4$ ($M =$ Fe, Co, Ni) films exhibit ferromagnetic behavior up to room temperature. Although the FeAl$_2$O$_4$/STO is insulating, the NiAl$_2$O$_4$/STO and CoAl$_2$O$_4$/STO heterointerfaces are found to be highly metallic and exhibit anomalous Hall effect (AHE) at temperatures below 30 K. Their Hall mobility is as high as $3 \times 10^4$ cm$^2$V$^{-1}$s$^{-1}$, comparable to that of $\gamma$-Al$_2$O$_3$/STO interface. There has been evidence of oxygen-vacancy-related magnetism in $\gamma$-Al$_2$O$_3$/STO at temperatures below 5 K, while the enhanced AHE in NiAl$_2$O$_4$/STO and CoAl$_2$O$_4$/STO likely comes from the magnetic proximity effect induced by the top ferromagnetic $M$Al$_2$O$_4$ spinel films.
The metallic interface between two insulating oxides, where a quasi-two-dimensional electron gas (q-2DEG) resides, provides a promising platform for the exploration of emergent phenomena.\textsuperscript{1,2} Its attractive physical properties, such as superconductivity,\textsuperscript{3} ferromagnetism,\textsuperscript{4} high electron mobility,\textsuperscript{5} strong gating field,\textsuperscript{6,7} quantum Hall effect,\textsuperscript{8} and photo excitation effect,\textsuperscript{9,10} have drawn extensive interest. So far, the isostructural perovskite-type interface, particularly LaAlO\textsubscript{3}/SrTiO\textsubscript{3} (LAO/STO)\textsuperscript{1} has been investigated intensively. However, the high mobility q-2DEG discovered at the non-isostructural interface between spinel $\gamma$-Al\textsubscript{2}O\textsubscript{3} and perovskite STO remains underinvestigated.\textsuperscript{2,11–14} In addition to the remarkably high electron mobility (1.4×10\textsuperscript{5} cm\textsuperscript{2}V\textsuperscript{-1}s\textsuperscript{-1} at 2 K), the spinel structure of $\gamma$-Al\textsubscript{2}O\textsubscript{3} also provides the opportunity to introduce intrinsic ferromagnetism into the heterostructure, which remains unexplored.

Herein, we epitaxially grew three new heterostructures, consisting of $M$Al\textsubscript{2}O\textsubscript{4} ($M$ = Fe, Co, Ni) top films and (001)-oriented TiO\textsubscript{2}-terminated STO substrates, and investigated their interfacial conduction and ferromagnetism. Remarkably, all the $M$Al\textsubscript{2}O\textsubscript{4} ($M$ = Fe, Co, Ni) films exhibit ferromagnetic behavior up to room temperature. We further found that the NiAl\textsubscript{2}O\textsubscript{4}/STO and CoAl\textsubscript{2}O\textsubscript{4}/STO interfaces are metallic and ferromagnetic at low temperatures, as indicated by the appearance of anomalous Hall effect (AHE). The AHE of the $M$Al\textsubscript{2}O\textsubscript{4}/STO interfaces ($M$ = Ni, Co) remains sizable up to 30 K, in contrast to $\gamma$-Al\textsubscript{2}O\textsubscript{3}/STO which shows AHE below 5 K. Moreover, the anomalous Hall resistance ($R_{AHE}$) undergoes a negative to positive sign change when the top film of the heterostructure changes from $\gamma$-Al\textsubscript{2}O\textsubscript{3} to $M$Al\textsubscript{2}O\textsubscript{4}. We proposed that the AHE in $\gamma$-Al\textsubscript{2}O\textsubscript{3}/STO is due to the oxygen vacancies induced ferromagnetism in proximity to the STO surface, while the AHE in NiAl\textsubscript{2}O\textsubscript{4}/STO and CoAl\textsubscript{2}O\textsubscript{4}/STO probably comes from the magnetic proximity effect induced by the ferromagnetic $M$Al\textsubscript{2}O\textsubscript{4} spinel films. Different from NiAl\textsubscript{2}O\textsubscript{4}/STO and CoAl\textsubscript{2}O\textsubscript{4}/STO interfaces, FeAl\textsubscript{2}O\textsubscript{4}/STO is insulating.
Films were grown on TiO$_2$-terminated STO single crystal substrates (5 mm × 5 mm × 0.5 mm in dimensions) by pulsed laser deposition using a KrF laser with a wavelength of 248 nm. During deposition, the substrate temperature was maintained at 650 °C and the oxygen pressure was kept at 1×10$^{-5}$ mbar. The laser fluence was 2 Jcm$^{-2}$ and the repetition rate was 1 Hz. The target-substrate distance was fixed at 5 cm. After deposition, the samples were cooled to room temperature without changing oxygen pressure. For the $\gamma$-Al$_2$O$_3$ deposition, a commercial Al$_2$O$_3$ single crystal target was used. MAl$_2$O$_4$ ($M =$ Fe, Co, Ni) ceramic targets were adopted for other films. These targets were prepared by sintering the mixture of appropriate amounts of Al$_2$O$_3$ with Fe$_2$O$_3$, Co$_3$O$_4$ and NiO powders first, at 1200 °C for 10 h and then, after pressing, at 1350 °C for 36 h. The film growth rate is approximate 0.08 Å/s. The epitaxial growth of the crystalline films was confirmed by both reflection high-energy electron diffraction (RHEED) and high-resolution X-ray diffraction (XRD) measurements. Heterostructures with 4 nm thickness top films are employed for transport and magnetic measurements. While, heterostructures with 40 nm top films are used for XRD measurements. Ultrasonic Al wire bonding was used to get electric connection, and the van der Pauw geometry was adopted.

Fig. 1(a) is a schematic illustration of the spinel/perovskite (MAl$_2$O$_4$/STO) oxide heterostructure. The epitaxial growth of spinel MAl$_2$O$_4$ ($M =$ Fe, Co, Ni) films on perovskite STO substrate is due to their compatible oxygen sub-lattice, as the lattice parameter of MAl$_2$O$_4$ is about twice that of STO.$^2$ Although $\gamma$-Al$_2$O$_3$ and STO show a good lattice match (1%), NiAl$_2$O$_4$, CoAl$_2$O$_4$, and FeAl$_2$O$_4$ exhibit larger lattice mismatch with STO substrate ($> 3\%$) as summarized in Table I. Consequently, the $\gamma$-Al$_2$O$_3$ film can been epitaxially grown on STO (001) substrate with a persistent layer-by-layer two-dimensional growth mode as confirmed by RHEED and high-resolution transmission electron microscopy,$^{13,15}$ while the NiAl$_2$O$_4$, CoAl$_2$O$_4$, and FeAl$_2$O$_4$ films show 3D island growth mode. Despite of this, the epitaxial growth of NiAl$_2$O$_4$, CoAl$_2$O$_4$, and FeAl$_2$O$_4$ films
with the thickness of 40 nm on STO is confirmed by the XRD measurements. As shown in Figs. 1(b), for the $\theta$ - 2$\theta$ scan in the region of 10° - 80°, (004) spinel Bragg peaks is observed on the left side of the corresponding STO peaks of (002). The out-of-line lattice parameters of $\gamma$-Al$_2$O$_3$, NiAl$_2$O$_4$, CoAl$_2$O$_4$, and FeAl$_2$O$_4$ films determined by XRD are 8.02, 8.03, 8.08, and 8.16 Å, respectively. This indicates that all the MAI$_2$O$_4$ films are well strain-relaxed. In addition, for the films of NiAl$_2$O$_4$ and FeAl$_2$O$_4$, impurity phase of MAIO$_2$ (M = Ni, Fe) at 16.6° is detected. For NiAl$_2$O$_4$ film, an extra impurity phase of Ni (200) crystal phase at 51.7° is also observed. These impurity phases could stem from the reduction environment of low oxygen pressure (1×10$^{-5}$ mbar) and high temperature (650 °C) adopted during the film deposition. However, the low diffraction intensity indicates that the amount of these impurities is rather low. Notably, these impurity phases survive after the post annealing at 300°C in 1 bar oxygen for 3 hours, meanwhile, the interface becomes insulating. Therefore, such impurity phases contribute negligibly to the interface conduction as discussed later. This is also consistent with the fact that MAI$_2$O$_4$ (M = Fe, Co, Ni) films grown on (LaAlO$_3$)$_{0.3}$(Sr$_2$TaAlO$_6$)$_{0.7}$ (LSAT) substrates are insulating in nature (see in Supplemental Material S1). Fig. 1(c) shows the Rocking curves of the (004) spinel films grown on STO substrates. The full widths at half maximum (FWHM) of the curves determined by Gaussian fitting are shown in Table I. All MAI$_2$O$_4$ (M = Fe, Co, Ni) films display good crystallinity. In addition, comparing to MAI$_2$O$_4$ spinel films, the relatively larger FWHM (0.21°) of $\gamma$-Al$_2$O$_3$ film might be due to its less ordered crystallographic structure which contains cation vacancies.

Transport measurements show that FeAl$_2$O$_4$/STO is highly insulating. However, the metallic conduction is obtained in $\gamma$-Al$_2$O$_3$/STO, MAI$_2$O$_4$/STO (M = Ni, Co) heterostructures, as shown in Fig. 2(a). Comparing to $\gamma$-Al$_2$O$_3$/STO whose sheet resistance ($R_s$) is 278 Ω/□ at room temperature, the NiAl$_2$O$_4$/STO and CoAl$_2$O$_4$/STO have smaller $R_s$ which are 57.8 Ω/□ and 138 Ω/□, respectively. Figs. 2(b)-(d) display the Hall resistance ($R_{xy}$) of $\gamma$-Al$_2$O$_3$/STO, NiAl$_2$O$_4$/STO and CoAl$_2$O$_4$/STO as
a function of magnetic field \((B)\) in the temperature range from 295 to 2 K. When temperature is high \((T > 100 \text{ K})\), \(R_{xy}\) varies linearly with applied field for all samples. This is the typical behavior of the normal Hall effect (NHE). Cooling the samples to 100 K, \(R_{xy}\) shows nonlinear dependence on magnetic field. In the meanwhile, the magnetic field-dependent magnetoresistance \((MR = (R_{xx}(B)/R_{xx}(B=0)-1)\) traces follow a bell-like shape, where \(MR-B\) displays a U-shape at low field, and shift to a bell-shape at high field. These features suggest that the conductivity comes from two or more carriers as previously reported by Joshua et al.\(^{18}\) and Kim et al.\(^{19}\), and can be fitted by a two-band model (see Supplemental Material S2). However, the \(R_{xy}\) exhibits a stronger curvature in the low-field range when further cooled below a critical temperature of approximately 30 K, which is beyond the capture of the two-band model. To describe the \(R_{xy}-B\) relation at \(T < 30 \text{ K}\), we adopted an extended two-band model that combines the two-band conduction-dominated NHE with an AHE as reported before\(^{20,21}\) (see Supplemental Material S2):

\[
R_{xy} = R_{\text{NHE}} + R_{\text{AHE}} = R_{\text{NHE}} + \alpha L\left(\frac{mB}{k_BT}\right)
\]

where \(R_{\text{NHE}}\) and \(R_{\text{AHE}}\) represent the Hall resistance from two-band conduction and AHE, respectively. The Langevin function \(L\) is introduced to simulate the step-shaped AHE curve in form, \(\alpha\) is a scale factor, and \(m\) is magnetic moment.

Fig. 2(e) displays the determination of the anomalous Hall resistance, \(R_{\text{AHE}}\), from \(R_{xy}\). The results of eq. 1 (thin black line) well reproduce the measured \(R_{xy}\) (thick green line). Basically, the normal Hall resistance \((R_{\text{NHE}})\) varies smoothly with \(B\) in the whole field range, with slightly but identifiable curve bending. In contrast, \(R_{\text{AHE}}\) is constant in high-field range and undergoes a drastic change as \(B\) sweeps through zero field. It also becomes clear that AHE appears below 5 K for \(\gamma\)-Al\(_2\)O\(_3\)/STO, and 30 K for both NiAl\(_2\)O\(_4\)/STO and CoAl\(_2\)O\(_4\)/STO. Based on the \(R_{\text{NHE}}\) in eq. 1, the temperature-dependent density \((n_s)\) and Hall mobility \((\mu)\) of the carriers confined in heterostructures...
can be deduced (Figs. 2(f)-(g)). The $n_s$ of q-2DEGs is nearly constant from 295 K to 2 K. At 2 K, the $\gamma$-Al$_2$O$_3$/STO possesses the lowest $n_s$ with a value of $3 \times 10^{15}$ cm$^{-2}$. The NiAl$_2$O$_4$/STO heterointerface has the highest $n_s$ ($2.2 \times 10^{16}$ cm$^{-2}$) which is higher than $\gamma$-Al$_2$O$_3$/STO by a factor of 7. The $n_s$ of CoAl$_2$O$_4$/STO is $9.8 \times 10^{15}$ cm$^{-2}$. These extremely high carrier densities indicate that 3D STO bulk conduction contributes to the measured conductivity. With regards to the $\mu$, these three heterostructures have comparable values in the range of $2.7 - 3.4 \times 10^4$ cm$^2$V$^{-1}$s$^{-1}$ at 2 K, also consistent with the mobility for bulk STO.$^{1,13}$ It is noteworthy that the FeAl$_2$O$_4$/STO grown under the same condition with NiAl$_2$O$_4$/STO and CoAl$_2$O$_4$/STO is highly insulating. This means that the Al-based spinel/perovskite interface is extremely sensitive to the introduction of the magnetic ions.

The metallic conduction in STO-based heterostructures comes from electrons located on the STO side. The high $n_s$ in $\gamma$-Al$_2$O$_3$/STO and $MA$I$_2$O$_4$/STO ($M = Ni, Co$) could result from the formation of oxygen vacancies in STO due to interfacial redox reactions.$^{2,13,15,22}$ However, the insulating FeAl$_2$O$_4$/STO heterointerface might stem from two reasons. On the one hand, the Fe-based oxide has poor ability to reduce STO substrate during the film deposition.$^{23}$ On the other hand, the band gap of FeAl$_2$O$_4$ (1.78 eV$^{24}$) is much lower than that of STO (3.2 eV), any reconstructed electrons tend to accumulate in the spinel films rather than transferring to the heterointerface.$^{25}$

Figs. 3(a)-(c) show the $R_{AHE}$ variation with respect to $B$ at different temperatures for $\gamma$-Al$_2$O$_3$/STO, $MA$I$_2$O$_4$/STO ($M = Ni, Co$). The $R_{AHE}$ of $\gamma$-Al$_2$O$_3$/STO has the same sign to $B$, while that of $MA$I$_2$O$_4$/STO ($M = Ni, Co$) are opposite to $B$. Similar crossover in the sign of $R_{AHE}$ has also been observed between SrRuO$_3$ and La$_{1-x}$Sr$_x$CoO$_3$ ($x=0.17$) crystals.$^{26}$ But the explanation for such phenomenon remains open, which could result from the intrinsic different origins of the magnetism. Moreover, in the magnetic saturation state, such as under $B = -10$ T at 2 K, $R_{AHE}$ is as large as 0.013 $\Omega$ for $\gamma$-Al$_2$O$_3$/STO, while it is 0.008 $\Omega$ for NiAl$_2$O$_4$/STO and 0.012 $\Omega$ for CoAl$_2$O$_4$/STO. Fig. 3(d)
summarizes the $R_{\text{AHE}}$ for these three heterointerfaces as a function of temperature. Clearly, the AHE appears at $T \leq 30$ K for $\text{MAL}_2\text{O}_4/\text{STO} \ (M = \text{Ni, Co})$, whereas, only below 5 K for $\gamma\text{-Al}_2\text{O}_3/\text{STO}$ as discussed before. Shortly, the AHE of the $\text{MAL}_2\text{O}_4/\text{STO} \ (M = \text{Ni, Co})$ is dramatically different from that of $\gamma\text{-Al}_2\text{O}_3/\text{STO}$.

In order to uncover the origin of AHE in $\gamma\text{-Al}_2\text{O}_3/\text{STO}$ and $\text{MAL}_2\text{O}_4/\text{STO} \ (M = \text{Ni, Co})$ heterointerfaces, magnetic measurements were performed by superconducting quantum interference device (SQUID) magnetometer (as shown in Fig. 3(e)). Remarkably, these three $\text{MAL}_2\text{O}_4/\text{STO} \ (M = \text{Fe, Co, Ni})$ heterostructures exhibit unexpected ferromagnetic properties up to room temperature. Notably, the CoAl$_2$O$_4$ and FeAl$_2$O$_4$ show spin-glass-like ground states in bulk below the Curie-Weiss temperatures (5 K for CoAl$_2$O$_4$ and 12 K for FeAl$_2$O$_4$), and the NiAl$_2$O$_4$ is paramagnetic. In the meantime, the ferromagnetism of $\gamma\text{-Al}_2\text{O}_3/\text{STO}$ is very weak (see Supplementary Material S3). At room temperature (inset of Fig. 3(e)), the magnetizations of FeAl$_2$O$_4$, CoAl$_2$O$_4$ and NiAl$_2$O$_4$ are 135.3, 76.4 and 69.4 emu/cm$^3$ when $B = 6$ T, respectively, which are much lower than the magnetization of Fe$_3$O$_4$ (about 480 emu/cm$^3$ at room temperature). When the temperature is 10 K, FeAl$_2$O$_4$ has the strongest magnetization, which is as large as 291.3 emu/cm$^3$ at $B = 6$ T, while CoAl$_2$O$_4$ and NiAl$_2$O$_4$ have comparable magnetization, which are 196.2 and 177.9 emu/cm$^3$, respectively. As for the interface between $\gamma\text{-Al}_2\text{O}_3$ and STO, the conduction comes from the oxygen vacancies, which could result in ferromagnetism, thus, the AHE as the origin of the ferromagnetism in LAO/STO. These oxygen vacancies not only induce a complex multi-orbital reconstruction thus the mobile q-2DEG, but also result the spin splitting of the electronic states, giving rise to localized Ti 3$d$ electrons thus magnetism. Salluzzo et al. experimentally proved that the oxygen vacancies play a decisive role in the interfacial magnetism in LAO/STO. Whereas, the much higher upper limit temperature (30 K) of AHE observed in our
MAl₂O₄/STO (M = Ni, Co) heterostructures than that of γ-Al₂O₃/STO (5 K) indicates that the oxygen-vacancy-related AHE in MAl₂O₄/STO is relatively weak.

It has also been suggested that interdiffusion of magnetic cations into STO could result in a similar transport behavior in manganite-buffered LAO/STO heterostructure. Since the interdiffusion of cations is also a common phenomenon in the spinel/perovskite heterostructure, such as γ-Al₂O₃/STO. Therefore, we cannot rule out the possibility of the interdiffusion of magnetic ions into STO as a possible origin of AHE at MAI₂O₄/STO heterointerfaces. However, such interdiffusion of magnetic ions into STO could result in Kondo effect, which is absent here. Finally, since the top films are found to be ferromagnetic, we therefore assume that the AHE in MAI₂O₄/STO (M = Ni, Co) comes from the magnetism induced by a magnetic proximity effect as reported for the EuTiO₃-buffered LAO/STO heterostructure.

In summary, we epitaxially grown MAI₂O₄/STO (M = Fe, Co, Ni) heterostructures in comparison to the γ-Al₂O₃/STO heterostructure. Remarkably, all the MAI₂O₄ (M = Fe, Co, Ni) films exhibit ferromagnetic behavior up to room temperature. The heterointerface of FeAl₂O₄/STO is highly insulating. In contrast, the NiAl₂O₄/STO and CoAl₂O₄/STO are metallic conducting. AHE is observed in most of the metallic interfaces of γ-Al₂O₃/STO, NiAl₂O₄/STO and CoAl₂O₄/STO. While the AHE in γ-Al₂O₃/STO is likely due to the magnetism induced by oxygen vacancies, the AHE in NiAl₂O₄/STO and CoAl₂O₄/STO most likely comes from the magnetic proximity effect induced by the top ferromagnetic spinel films.

**Supplementary Material**

See supplementary material for XRD data of NiAl₂O₄ films prepared under different oxygen pressures, on different substrates (STO, LSAT) and after the post oxygen annealing; Two-band model fitting of interface conduction; Magnetization of spinel/perovskite heterostructures.
Acknowledgements:

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References:


Table I. Lattice parameters of $\gamma$-Al$_2$O$_3$, $M$Al$_2$O$_4$ ($M = \text{Fe, Co, Ni}$) in bulk from reports and in film shown in text deduced by XRD data, their mismatches with STO substrate, the full widths at half maximum (FWHM) of Rocking curves for the films, their band gaps, and conductivities of the STO-based heterointerfaces at room temperature:

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<tr>
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<tr>
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<td>138</td>
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Figure captions:

FIG. 1 (a) A schematic sketch of the spinel/perovskite ($MA_2O_4$/STO) oxide heterostructure ($M = \text{Fe, Co, Ni}$). Lattice structures of the spinel and perovskite are shown below. The box represents one unit cell, the lattice parameter of $MA_2O_4$ is about twice that of STO. (b) X-ray diffraction (XRD) $\theta - 2\theta$ scan of the $\gamma$-$Al_2O_3$, NiAl$_2$O$_4$, CoAl$_2$O$_4$, and FeAl$_2$O$_4$ films grown on TiO$_2$-terminated STO substrates. The inset shows the XRD $\theta$-2$\theta$ scan around the STO (002) reflection. (c) Omega Rocking curves of the epitaxial films in spinel/perovskite heterostructures.

FIG. 2 (a) Temperature-dependent sheet resistances ($R_s$) of q-2DEGs in $\gamma$-$Al_2O_3$/STO, NiAl$_2$O$_4$/STO and CoAl$_2$O$_4$/STO heterostructures. (b) - (d) Magnetic dependence of Hall resistances ($R_{xy}$) in the three heterostructures at different temperatures, respectively. (e) Example for the determination of normal Hall effect (NHE) and anomalous Hall effect (AHE) from the total Hall effect for NiAl$_2$O$_4$/STO at 2K. Measured and calculated results are presented as thick green and thin black lines, respectively. Temperature dependence of (f) sheet carrier densities, $n_s$, and (g) Hall mobilities, $\mu$, in these heterostructures.

FIG. 3 (a)-(c) Anomalous Hall resistances, $R_{\text{AHE}}$, in $\gamma$-$Al_2O_3$/STO, NiAl$_2$O$_4$/STO and CoAl$_2$O$_4$/STO heterostructures as a function of magnetic field at different temperatures. (d) Anomalous Hall resistances (at $B = -10$ T) as a function of temperatures for these three samples. (e) Magnetization curves as a function of magnetic field (M-H) for $\gamma$-$Al_2O_3$ and $MA_2O_4$ ($M = \text{Fe, Co, Ni}$) films with a thickness of 4 nm measured at 10 K. The inset is the M-H measured at 300 K.
FIG. 1
FIG. 2
FIG. 3