Ferromagnetism and Half Metallicity in Two-dimensional MO (M = Ga, In) Monolayers Induced by Hole Doping

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ABSTRACT: Identification of new two-dimensional (2D) materials with magnetic properties has received strong research attention in the development of advanced spin-based devices. By means of first-principles calculations, we investigate the stability, electronic properties and the hole-doping-induced magnetic properties of novel metal oxide (MO, M = Ga, In) monolayers. They are intrinsically non-magnetic stable semiconductors, with high energetic, vibrational and thermal stability. Hole doping can switch them from non-magnetic to ferromagnetic and turn them into half-metals over a wide range of hole densities. Monte Carlo simulations predict that the highest Curie temperature of the GaO monolayer can reach ~125 K. Our results indicate that monolayer MO could be eligible candidate materials for novel 2D spintronic devices.

KEYWORDS: 2D materials, GaO, InO, ferromagnetic, hole doping, first-principles calculation

1. INTRODUCTION

Two-dimensional (2D) crystals have been attracting unparalleled research attention ever since the successful exfoliation of graphene.¹⁻⁴ Extensive research efforts have been devoted to the continuous discovery of alternative 2D materials, in pursuit of diverse and exceptional properties.^{5,6} The potential applications of 2D materials, such as solar cells, electrocatalysts, photocatalysts, and field-effect transistors (FETs), etc., have been investigated intensively.^{7,8} Nevertheless, it is still a challenge to control or induce magnetism in these 2D materials for spintronic applications,^{9, 10} because the majority of 2D materials are intrinsically nonmagnetic.

It is worth noting that there are some 2D materials exhibiting peculiar “Mexican-hat” valence band dispersion, or a flat band edge in their valence band structure, which would result in a sharp van Hove singularity in their electronic density of states (DOS). Once their Fermi level is shifted towards the valence band by hole doping, it would lead to electronic instability and could result in magnetism, superconductivity and other physical phenomena.¹¹⁻¹³ This would provide a feasible way to introduce controllable magnetism by manipulating the carrier doping concentration, which can be realized experimentally by gating techniques. Compared to the approaches like introducing defects and adatoms, carrier doping is more appropriate to control. Most importantly, magnetism induced by carrier doping is mainly contributed by p state in these materials, which enable a higher Fermi velocity, and thus higher speed spin-polarized transport, due to the more delocalization nature of p orbitals (as compared to d or f -states). Previous first principles calculations indicate that group-III monochalcogenides (e.g., GaS, GaSe and InSe),^{11, 14, 15} [12, 15, 16] group-IV monoxides (SnO and PbO),^{12, 16} InP₃,¹⁷ SnX₂ (X=S and Se),¹⁸ MoN₂,¹⁹ blue phosphorene and gray arsenene,²⁰ etc. show ferromagnetic characteristics with hole doping. However, either they can be ferromagnetic over a limited hole doping range, or their magnetism can only be triggered at a relatively high doping density. Therefore, it is of significance to search

for more novel 2D crystals that are ferromagnetic with high Curie temperature (T_c) for spintronic devices.

In this research, we propose two novel metal oxide (MO, $M = \text{Ga, In}$) monolayers, on the basis of first principles calculations. The stability of these 2D materials is verified by their formation energies, phonon dispersions as well as by molecular dynamic simulations. Additionally, their electronic and magnetic properties under hole doping are also calculated. It turns out that MO monolayers are intrinsically non-magnetic semiconductors that can become ferromagnetic over a wide range of hole concentrations. Their T_c are further evaluated using Monte Carlo simulations.

2. COMPUTATIONAL METHODS

In our density functional theory (DFT) calculations, the electron-ion interaction was described by projector augmented wave (PAW) pseudopotentials,²¹ and the generalized gradient approximation (GGA), parameterized by the Perdew-Burke-Ernzerhof (PBE) approach,²² was used as the exchange correlation functional. The initial geometry optimizations, the non-spin-polarized electronic properties as well as the phonon dispersion calculations using the linear displacement method were carried out by means of Quantum-ESPRESSO code.²³ The energy cutoff of 80 Ry k-point meshes of $8 \times 8 \times 1$ and $16 \times 16 \times 1$, automatically generated by the Monkhorst-Packs scheme, were used for structural optimizations and self-consistent calculations. The structural optimizations were carried out until the residual forces converged within 10^{-5} Ry/bohr. The Heyd-Scuseria-Ernzerhof functional (HSE06),²⁴ which mixes 25% nonlocal exchange with the PBE functional, was used for more accurate electronic density of states calculations. The spin polarized calculations for the electronic and magnetic properties under hole doping simulation were further performed using the Vienna ab initio simulation package (VASP).²⁵ The energy cutoff of 550 eV and a dense k point grid of $32 \times 32 \times 1$ were employed to obtain accurate spin polarization energies and

magnetic moments. Vacuum layers of thickness larger than 15 Å were used to avoid artificial interaction between neighboring images for the MO monolayers.

The ab-initio molecular dynamics (AIMD) simulations were carried out using 5×5×1 supercells (with 100 atoms) were equilibrated at 300 K for 10 ps with a time step of 2 fs. The constant-temperature and volume canonical ensemble (NVT) was selected, and the temperature was controlled by means of a Nose thermostat.

3. RESULTS AND DISCUSSION

The crystal structure of MO monolayer resembles the one of GaSe/InSe monolayer, which is shown in Figure 1(a). Each O atom is bonded to three neighboring M atoms, while every M atom is 4-fold coordinated by three adjacent O atoms and one M atom, forming the O-M-M-O stacking in the vertical direction. We also propose another phase of MO, which is obtained by shifting one of the surface O atoms to the hexagonal center, as can be seen in Figure S1 (a) in the supporting information. For simplicity, they are referred to as H and T phase, respectively. Their optimized structural parameters are summarized in Table S1. From the phonon dispersion spectra, one can see that there are only very limited range of negative frequencies for both phases of MO, which means that both phases are vibrationally stable. Nevertheless, their total energies indicate that the H phases are more energetically favorable than the T phases, although the total energy differences are as small as ~6 and ~4 meV/atom, respectively, for GaO and InO monolayers. In the following part, we will mainly focus on this more stable H phase for MO, and if not specifically indicated, MO refers to the monolayer in the H phase.

To further verify the stability of the MO monolayers, we calculated their formation energies per atom (E_{form}), which is defined as:

$$E_{\text{form}} = (E_{\text{MO}} - 2E_{\text{M}} - E_{\text{O}_2})/4 \quad (1)$$

Figure 1. (a): Top and sides views of a single-layer MO in the H phase. The unit cell is highlighted by red dashed lines; (b) and (c): phonon dispersion spectra of H-GaO and H-InO monolayers, respectively.

Here, E_{MO} is the total energy of a MO monolayer, E_{M} is the energy of a metal atom in its stable bulk structure and E_{O_2} corresponds to the energy of an isolated oxygen molecule in its paramagnetic ground state. Negative E_{form} means that the monolayers are more energetically favorable than their elementary substances. The calculated E_{form} of GaO and InO are -1.67 and -1.35 eV/atom, respectively. As a comparison, the corresponding E_{form} of single-layer GaSe and InSe are calculated to be -0.59 and -0.53 eV/atom using the same method. Besides, the E_{form} of MoS_2 and MoSe_2 have been reported to be -0.83 and -0.75 eV/atom, respectively.²⁶ Therefore, we can consider that MO monolayers have a high energetic stability.

AIMD simulations are also carried out on $5 \times 5 \times 1$ supercells at temperatures of 300 K to evaluate the stability of MO monolayers at room temperature. Figure S2 depicts the time-dependent energy fluctuations as well as the snapshots of the final atomic configurations in the AIMD simulations. The total energies of both H-GaO and H-InO oscillate within narrow ranges, without abrupt variation. At 300 K, all the atoms of the MO monolayers are slightly vibrating around the equilibrium positions in the cells, with no broken bond observed, and the crystal structures are still retained integrally, attesting to their room temperature thermal stability.

Figure 2. Orbital projected band structures and total density of states of (a) H-GaO and (b) H-InO monolayers.

After having confirmed the stability of MO monolayers, we now focus on their electronic properties. The orbital projected band structures and total density of states (DOS) of MO monolayers are displayed in Figure 2. Band structures of the T phases are given in Figure S3. The monolayer MO in both the H and T phases are non-magnetic semiconductors, with indirect band gaps in their ground states. The band gaps of H-GaO and H-InO are calculated to be ~ 1.45 eV and ~ 0.33 eV, respectively at PBE level. The conduction band minima (CBM) of both H-GaO and H-InO monolayers are located at the Γ point and there are two valence band maximums (VBM) of H-GaO lying

between Γ -M and Γ -K points, whereas the VBM of H-InO is at the K point of the first Brillouin zone. The situation is similar for the T phase MO monolayers. All these materials exhibit inverted Mexican-hat-like band dispersion in the top valence bands. From the projected band structures, one can see that the top valence bands of H-GaO and H-InO are mainly dominated by the p_z orbitals of M atoms and O atoms (denoted as M- p_z and O- p_z), with a small contribution from the O- p_x and M- s orbitals, whereas the conduction band minima are mainly derived from the M- s orbitals and partially from the O- s , O- p_z and M- p_z orbitals. The Mexican-hat-like band dispersions in 2D should give rise to a sharp peak in the DOS. As shown in the rightmost panel of Figure 2, there are noticeable Van Hove singularities in the vicinity of the valence band edge for H-GaO and H-InO. This is confirmed by both PBE and HSE results. Note that the HSE calculations predict larger band gaps of ~ 3.4 eV and ~ 1.7 eV for H-GaO and H-InO, respectively, compared to the PBE results. Given these high DOS peaks, it is expected that the phase transitions, like a paramagnetic to ferromagnetic one, would appear when the Fermi level is shifted closer to the DOS peaks, due to the electronic instability.

Therefore, we investigate the possible magnetic properties of hole-doped MO monolayers using spin polarization calculations, since hole doping would lead to the shift of the Fermi level towards the valence band. The influence of hole doping on the atomic structures is first studied, by relaxing the charged cells. The atomic displacements occurring in the MO monolayers upon hole doping, as observed during the relaxation of the systems, are summarized in Figure S4. Obviously, hole doping leads to the separation of the MO upper and bottom layers. Specifically, the M-M bond lengths expand upon increasing the doping concentration, while the M-O bond lengths and the O-M-M bond angles will shrink, making the atomic planes flatter.

Figure 3. Magnetic moment and spin polarization energy as a function of hole doping density, and spin-polarized orbital projected density of states under different hole doping concentrations of (a) H-GaO and (b) H-InO monolayers

The computed magnetic moment and spin polarization energy (defined as the total energy difference between the ferromagnetic and non-magnetic states of the system) of H-GaO and H-InO monolayers are shown in the left panel of Figure 3. A positive spin polarization energy indicates the preference for the ferromagnetic state. The H-GaO and H-InO monolayers become ferromagnetic when the hole doping concentration is higher than $\sim 0.15 \times 10^{14} \text{ cm}^{-2}$ and $\sim 0.2 \times 10^{14} \text{ cm}^{-2}$, respectively; in this case, the magnetic moment is $\sim 1.0 \mu_B/\text{hole}$ (where μ_B is the Bohn magneton), i.e. the injected holes can be fully spin polarized. The spin polarization energy increases monotonously with the hole density and reaches the maximum of 66.8 and 26.3 meV/hole at 6.3×10^{14}

cm^{-2} and $4.4 \times 10^{14} \text{ cm}^{-2}$ for H-GaO and H-InO, respectively. Thereafter, it declines gradually when the hole density is further increased.

The calculated spin-polarized DOS under different doping levels are given in the right panel of Figure 3, where the energy splitting between spin up and spin down states can be observed. The Fermi level crosses the spin down states, while the up-spins are fully occupied at the Fermi level, making these two materials half-metallic under hole doping. Clearly, the energy splitting between the two spin states enhances with increasing hole concentration, i.e., from $\sim 0.1 \text{ eV}$ at $1 \times 10^{14} \text{ cm}^{-2}$, to $\sim 0.6 \text{ eV}/0.5 \text{ eV}$ at $6.3 \times 10^{14} \text{ cm}^{-2}/4.4 \times 10^{14} \text{ cm}^{-2}$ for H-GaO/H-InO monolayer, respectively. Besides, the increase in hole doping concentration will also enhance the weight of the contribution of Ga/In- s and Ga/In- p_z orbitals to the valence band edges, with respect to the O- p_z one.

In general, the T_c of a ferromagnetic material depends on its spin polarization energy. Notably, the maximum spin polarization energies of MO monolayers are higher than the ones of GaSe (3 meV/hole),¹¹ PtSe₂ (2.5 meV/hole),²⁷ PdSe₂ (7 meV/hole)²⁸. In particular, the one of H-GaO is comparable to those of SnS₂/SnSe₂ (50-100 meV/hole),²⁹ MoN₂¹⁹ (75 meV/hole) and NSb (150 meV/hole),³⁰. The reported highest T_c for GaSe, PdSe₂, MoN₂ and NSb (computed using mean field theory or Monte Carlo simulations) are about 90 K, 700 K, 1100 K, and 615 K, respectively. Consequently, we expect that MO monolayers (especially H-GaO) could present a moderate T_c , being of importance for their potential application in spintronic devices. Their T_c was computed using Monte Carlo simulations. Because both the M and O atoms contribute to the magnetization after hole doping, based on the Heisenberg model, the spin Hamiltonian of the MO monolayers can be given by

$$H = E_0 - \sum_{\langle ij \rangle} J_1 \vec{S}_i \cdot \vec{S}_j - \sum_{\langle\langle ij \rangle\rangle} J_2 \vec{S}_i \cdot \vec{S}_j - \sum_{[ij]} J_3 \vec{S}_i \cdot \vec{S}_j - \sum_{\langle\langle ij \rangle\rangle} J_4 \vec{S}_i \cdot \vec{S}_j - \sum_{\langle\langle\langle ij \rangle\rangle\rangle} J_5 \vec{S}_i \cdot \vec{S}_j \quad (2)$$

where $\langle ij \rangle$ and $\langle\langle ij \rangle\rangle$ are the summation over the pairs of nearest, second nearest M (or O) atoms, and $[ij]$ is the summation over the nearest M and O atoms. $\vec{S}_i$ (or $\vec{S}_j$) is a unit vector. J_1 (J_2) is the first nearest neighbor exchange parameter, which is derived

from the two O (metal) atoms in upper and lower planes. J_3 is the neighbor exchange parameter originating from the O atoms and metal atoms in the same plane. J_4 (J_5) is the neighbor exchange parameter derived from the two O (metal) atoms in the same plane. They can be obtained by mapping the energy obtained from first-principles calculations. Details about the calculation of the exchange parameters are given in Figure S5 and their values are summarized in Table S2.

Figure 4. Monte Carlo simulation of the normalized magnetization of H-MO monolayers as a function of temperature, for various hole doping densities, as indicated in the figure.

The T_c can then be estimated precisely using Monte Carlo simulations, as implemented in the VAMPIRE software package.³¹ Since only orthogonal lattice vectors are supported, we constructed each simulated system by using $50 \times 30 \sqrt{3}$ supercells, with the spins being thermalized for 10000 equilibrium steps, followed by 20000 averaging steps for the calculation of the thermal equilibrium magnetization at every temperature. The calculated temperature-dependent normalized magnetization of

MO monolayers under different hole doping densities are displayed in Figure 4. The magnetization remains high in the low temperature range, and then drops to near zero at a certain critical temperature. The temperature dependent magnetization is then fitted using the Curie-Bloch equation in the classical limit:

$$m(T) = (1 - \frac{T}{T_c})^\beta \quad (3)$$

where T is the temperature and β is a critical exponent. At a hole concentration of $4.4 \times 10^{14} \text{ cm}^{-2}$, the fitted T_c is about 107 K for H-InO. While for H-GaO, the T_c is estimated to be about 125 K at hole doping density of $6.3 \times 10^{14} \text{ cm}^{-2}$. Importantly, these calculated T_c are higher than the ones of 2D CrI_3 (45 K),³² and $\text{Cr}_2\text{Ge}_2\text{Te}_6$ (30 K),³³ comparable to the one of Fe_3GeTe_2 monolayer (130 K),³⁴ which makes them promising candidates for spintronic devices. Finally, we also point out that carrier accumulation of up to 10^{15} cm^{-2} can be experimentally realized, by using ionic liquids or polymer electrolytes as gate dielectrics.^{35,36} Thus, a hole concentration of $\sim 6 \times 10^{14} \text{ cm}^{-2}$ will still be reached in MO monolayers.

4. CONCLUSIONS

In summary, we have investigated the stability, electronic and hole-doping induced magnetic properties of novel MO (M=Ga, In) monolayers, using first principles calculations based on density functional theory. The energetic stability of these two monolayers has been confirmed by their relatively high formation energies, and their vibrational and thermal stabilities have also been verified by phonon dispersion calculations and molecular dynamic simulations, respectively. Both GaO and InO monolayers are intrinsically non-magnetic semiconductors, with Mexican-hat-like band edges in their valence band near the Γ point, resulting in sharp peaks in their electronic density of states. Hole doping leads to a spontaneous ferromagnetic phase transition and turns these two monolayers into half-metals over a wide range of hole densities. Their estimated Curie temperature, using Monte Carlo simulations can reach $\sim 125 \text{ K}/107 \text{ K}$ reached for the H-GaO/H-InO monolayer, respectively. Our study indicates

that these novel two-dimensional MO phases can be potential candidates for 2D-based spintronic devices.

■ ASSOCIATED CONTENT

Supporting Information.

Structural parameters of MO monolayers, atomic structure, phonon dispersion and band structures of MO monolayers in the T phase, AIMD results, Variation rates of the bond lengths and bond angles of the charged system after relaxation, Schematics of the various magnetic structures, the details of exchange parameters calculations and their values.

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Notes

The authors declare no competing financial interest.

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