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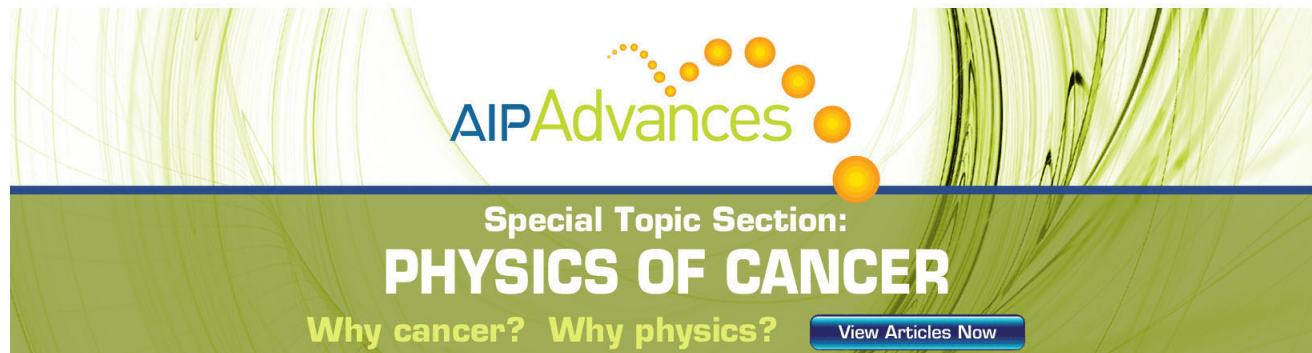
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# Spin-polarized two-dimensional electron gas in undoped $\text{Mg}_x\text{Zn}_{1-x}\text{O}/\text{ZnO}$ heterostructures

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Quantum transport properties of two-dimensional electron gas (2DEG) in undoped  $\text{Mg}_x\text{Zn}_{1-x}\text{O}/\text{ZnO}$  heterostructures grown by metal organic vapor phase epitaxy have been investigated. A large zero-field spin-splitting energy more than 15 meV in the 2DEG is determined at 1.6 K. Meanwhile, ferromagnetism is observed in the heterostructures. The findings reveal that the 2DEG is spin polarized at zero magnetic fields. It is believed that the exchange interaction between the itinerant electrons in the two-dimensional channel and the magnetic polarons in the  $\text{Mg}_x\text{Zn}_{1-x}\text{O}$  barrier around the interface results in the spin polarization of the 2DEG. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4711775>]

Owing to their advantages of wide direct-band gap, high electric breakdown field, and high saturation velocity, ZnO-based materials have great potential application in future electronic and photonic devices.<sup>1,2</sup> The recent realization of high-mobility two-dimensional electron gas (2DEG) in  $\text{Mg}_x\text{Zn}_{1-x}\text{O}/\text{ZnO}$  heterostructures grown by molecular beam epitaxy (MBE) reported by Tsukazaki *et al.*<sup>3</sup> opened up promising opportunities for studying quantum transport properties of ZnO-based heterostructures. Tunable 2DEG has been exhibited via a polymer-oxide Schottky barrier in metal-semiconductor field-effect transistor (MESFET) structure,<sup>4</sup> and the fractional quantum Hall effect<sup>5</sup> has also been observed in ZnO heterostructures.

In addition, ZnO is also thought to be a promising host material to achieve room temperature ferromagnetism for potential applications in spintronic devices.<sup>6</sup> Following the observations of  $d^0$  magnetism in undoped wide-gap oxide  $\text{HfO}_2$  films,<sup>7</sup> unexpected magnetic order has been discovered in undoped ZnO thin films<sup>8,9</sup> and nanoparticles.<sup>10</sup> It has been reported that defects in ZnO-based structures play an important role in creating  $d^0$  magnetism.<sup>11</sup> It is thus assumed that spin-polarized 2DEG, combining the magnetic properties with quantum transport properties, may exist in ZnO-based heterostructures.

In this study, high-mobility 2DEG in undoped  $\text{Mg}_x\text{Zn}_{1-x}\text{O}/\text{ZnO}$  heterostructures has been realized by metal organic vapor phase epitaxy (MOVPE). We carried out a detailed study on the sub-band occupation and zero-field spin splitting of the 2DEG, utilizing quantum transport measurements. We also investigated the magnetization properties of the  $\text{Mg}_x\text{Zn}_{1-x}\text{O}/\text{ZnO}$  heterostructures. The results indicate the existence of the  $d^0$  ferromagnetism in the heterostructures. This ferromagnetic character determines the electron

transport properties, revealing spin-polarization of the 2DEG.

Zn-faced  $\text{Mg}_x\text{Zn}_{1-x}\text{O}/\text{ZnO}$  heterostructures were grown on sapphire (0001) substrates using Aixtron MOVPE system with close-coupled showerhead (CCS) configuration.<sup>12,13</sup> The samples have same structure consisting of a 400 nm-ZnO buffer layer grown at 480 °C and a 1500 nm-ZnO epilayer grown at 900 °C, and then 60 nm- $\text{Mg}_{0.2}\text{Zn}_{0.8}\text{O}$  barrier layer was deposited on ZnO at 900 °C. Quantum transport properties of  $\text{Mg}_{0.2}\text{Zn}_{0.8}\text{O}/\text{ZnO}$  heterostructures were measured in van der Pauw geometry in a rotator-equipped Oxford refrigerator at low temperatures and high magnetic fields up to 14 T. The Hall electron density  $n_H = 4.56 \times 10^{12} \text{ cm}^{-2}$  and mobility  $\mu = 1845 \text{ cm}^2/\text{Vs}$  at 1.6 K are obtained from the low-field Hall measurements. Magnetic properties were investigated by a SQUID (superconducting quantum interference device) magnetometer.

From the samples, well-resolved Shubnikov-de Haas (SdH) oscillations and clear quantum Hall plateaus were observed at 1.6 K, as shown in Fig. 1(a), indicating the formation of high-mobility 2DEG in the  $\text{Mg}_{0.2}\text{Zn}_{0.8}\text{O}/\text{ZnO}$  heterostructures. The slope of Hall resistivity decreases as increasing the magnetic field, and becomes constant when field is large enough, as shown in Fig. 1(a) by the guiding lines, which is the anomalous Hall effect (AHE). The AHE is not that strong, since it depends on a variety of material specific parameters besides the magnetism. The SdH oscillations measured at various angles further identify the two dimensional character of the electron gas. The observed anomalous periodicity in the SdH oscillations seems to suggest the existence of Zeeman splitting of the 2DEG in a single sub-band. For clarifying the origin of this anomalous periodicity, we further performed the fast Fourier transform (FFT) analysis on the SdH oscillations. Interestingly, the FFT frequency spectrum exhibits three peaks as shown in Fig. 1(b). As the Zeeman splitting only affects the phase of the oscillations,<sup>14</sup> therefore the three frequencies must correspond to

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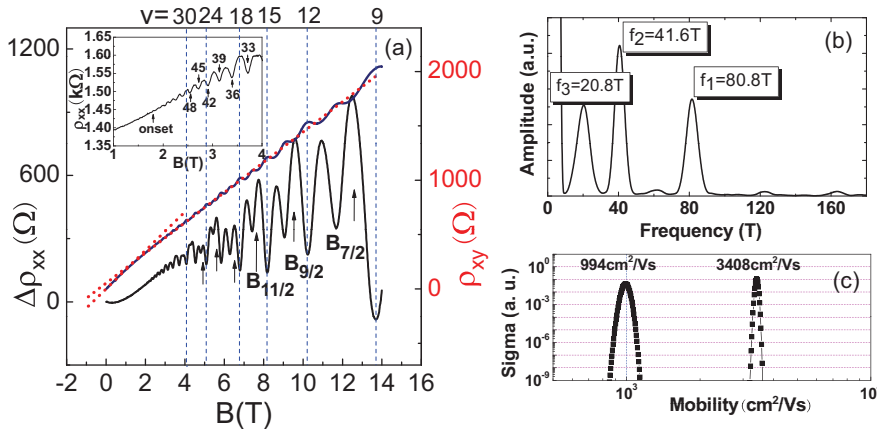


FIG. 1. (a) SdH oscillations and quantum Hall plateaus of the 2DEG in a  $\text{Mg}_x\text{Zn}_{1-x}\text{O}/\text{ZnO}$  heterostructure at 1.6 K. The dotted lines coherent to the Hall resistivity line are guiding lines used to indicate the AHE. The SdH minima correspond to both odd- and even- integer filling factors of the first sub-band. (b) FFT spectrum of the SdH oscillations at 1.6 K. (c) Mobility spectrum of the electrons in the heterostructure at 9 K.

multi-sub-band occupation or zero-field spin splitting rather than Zeeman effect.

In order to investigate the sub-band occupation, the mobility spectrum analysis<sup>15</sup> was conducted on the magneto-resistivity at 9 K when the SdH oscillations were extremely weak. Two peaks are observed in the mobility spectrum as shown in Fig. 1(c), strongly indicating two kinds of electrons with high mobilities. In view of the fact that the electron mobility in the ZnO buffer layer is extremely small, i.e., below  $60 \text{ cm}^2/\text{Vs}$ , the two electron mobility peaks should correspond to two sub-bands in the 2DEG. As a matter of fact, the first sub-band in the 2DEG has larger amplitude of wavefunction at the interface and hence suffers stronger scattering from the scattering centers around the interface,<sup>16</sup> leading to a lower mobility. By calculating the electron densities related to the mobility peaks, the ratio  $n_1/n_2 = 3.4$  is obtained, where  $n_1$  and  $n_2$  are the electron density of the first and second sub-bands, respectively.

We further deduced the 2DEG density from the frequencies of the SdH oscillations. If we assume that the three peaks in the FFT spectrum correspond to three-sub-band occupation, the total 2DEG density calculated will be  $6.91 \times 10^{12} \text{ cm}^{-2}$ , much larger than the Hall electron density. With the help of the mobility spectrum analysis, it is concluded that the three frequencies in the FFT spectrum belong to two sub-bands, namely  $f_3$  belongs to the second sub-band, while  $f_1$  and  $f_2$  belong to the first sub-band with spin non-degeneracy. As a result, the actual total density of the 2DEG should be  $3.96 \times 10^{12} \text{ cm}^{-2}$  ( $n_1 = e(f_1 + f_2)/h = 2.96 \times 10^{12} \text{ cm}^{-2}$ ,  $n_2 = 2ef_3/h = 1.00 \times 10^{12} \text{ cm}^{-2}$ ). The ratio  $n_1/n_2 \approx 3$  is in excellent agreement with the corresponding value obtained from the mobility spectrum within the fitting error-bar. A large zero-field spin-splitting energy of 15.16 meV of the first sub-band is then obtained by using  $E_{\uparrow} - E_{\downarrow} = 2\pi\hbar^2(n_{\uparrow} - n_{\downarrow})/m^*$ , where  $m^*$  is taken to be  $0.3m_0$ .<sup>3</sup> The filling factor  $\nu = n_1h/eB$  counts for the number of spin-resolved Landau levels (LLs) in the first sub-band below Fermi level, as shown in the inset of Fig. 1(a). When the spin-down LL crosses the Fermi level once with increasing magnetic field, the spin-up LL will cross the Fermi level twice due to the large difference between  $f_1$  and  $f_2$ . The clear observation of the co-existence of odd- and even- filling factors down to very low fields indicates the large spin splitting at zero fields.

In order to probe the origin of the large spin splitting, the effect of the spin-orbit coupling (SOC) interaction must be considered. The dominating mechanism of SOC in semiconductor heterostructures is Rashba SOC interaction<sup>17</sup> caused by the structure inversion asymmetry. We note that the Rashba SOC coefficient in the conduction band is smaller in ZnO in comparison to that in GaN.<sup>18</sup> The zero-field spin-splitting energies in  $\text{Al}_x\text{Ga}_{1-x}\text{N}/\text{GaN}$  heterostructures are 0.2 meV–2.5 meV at the 2DEG densities ranging from  $5 \times 10^{11} \text{ cm}^{-2}$  to  $1.1 \times 10^{13} \text{ cm}^{-2}$ .<sup>19–21</sup> Therefore, the SOC interaction itself cannot produce such a large spin-splitting energy observed in our experiments. Some other factors must be involved, in addition to the SOC interaction.

Actually, the observation of AHE in the undoped  $\text{Mg}_{0.2}\text{Zn}_{0.8}\text{O}/\text{ZnO}$  heterostructure is strong evidence of carrier mediated ferromagnetism in the 2DEG. Figure 2(a) presents the  $M$ - $H$  curve measured at 2 K by SQUID with applied field perpendicular or parallel to the  $\text{Mg}_{0.2}\text{Zn}_{0.8}\text{O}/\text{ZnO}$  heterointerface. The contribution from the diamagnetic background from the substrate has been subtracted. Clear hysteresis loops for both configurations show strong magnetization anisotropy. The largest magnetization is obtained in the perpendicular configuration. All these features verify the existence of intrinsic ferromagnetism in the heterostructures. Figure 2(b) exhibits the temperature dependence of the remnant magnetization curve  $M(T)$  of the heterostructure up to 390 K. The shape of the  $M(T)$  curve is more of concave type, which deviates from the Weiss mean-field theory. This is supposed to be due to the disorder in the sample.

Although nontrivial issues related to the “ $d^0$  magnetism” are still under debate, significant contribution of defects to  $d^0$  magnetism, such as zinc vacancies ( $V_{\text{Zn}}$ ) in ZnO, point defects in  $\text{HfO}_2$  films, and carbon vacancies ( $V_{\text{C}}$ ) or point defects in graphene-like systems, has been proven relevant, both theoretically and experimentally.<sup>9,22–27</sup> Since there is no doping of magnetic ions in our sample, the ferromagnetism can only be resulted from various defects, which could provide localized magnetic moments. We have measured the controlled samples with only ZnO epilayer grown on sapphire, and no distinct magnetism anisotropy and remnant magnetization were found in the sapphire layer and the ZnO epilayer. We therefore suggest that it is the  $\text{Mg}_x\text{Zn}_{1-x}\text{O}$  barrier layer and the ZnO buffer layer which make the major contribution to the ferromagnetism, considering the relative



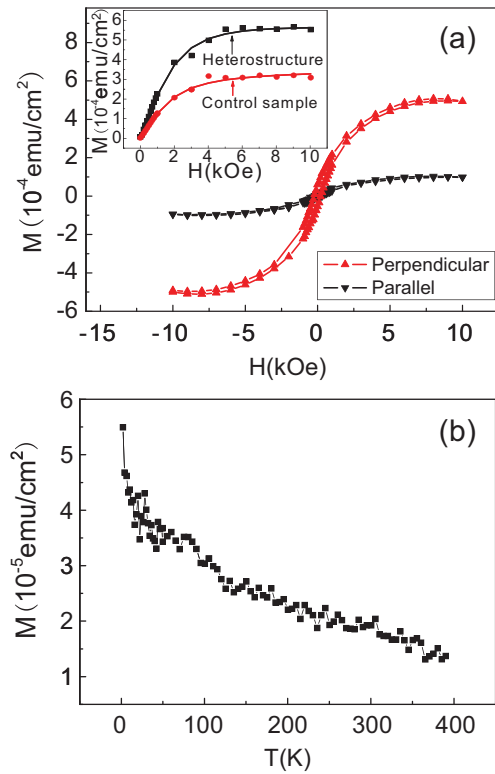


FIG. 2. (a) M-H curves of the  $\text{Mg}_x\text{Zn}_{1-x}\text{O}/\text{ZnO}$  heterostructure. The magnetic field is applied perpendicular (red) and parallel (black) to the heterointerface. The inset shows the fitting of the M-H curves, both the heterostructure and the control sample, by the Brillouin function. (b) Temperature dependence of the remnant magnetization curve from 2 to 390 K.

high defects density due to the lattice mismatch between  $\text{Mg}_x\text{Zn}_{1-x}\text{O}$  and ZnO layers, and ZnO buffer layer and sapphire, respectively.

The Brillouin function theory is used to fit the M-B curve. The fitting function we used is

$$M(B) \sim \left[ \frac{2S+1}{2S} \coth\left(\frac{2S+1}{2S}y\right) - \frac{1}{2S} \coth\left(\frac{1}{2S}y\right) \right] \times \left( y = \frac{gS\mu_B}{k_B T} B \right), \quad (1)$$

where  $B$  is the applied magnetic field,  $S$  is the spin quantum number,  $T$  is the temperature, and  $g$  is the Landé  $g$ -factor, respectively. In the fitting, we set  $T = 2$  K,  $g = 2$ . As shown in the inset of Fig. 2(a), the best-fit parameter  $S$  for both heterostructures and control samples is 16.5, indicating that the same origin of the magnetism in both the barrier and buffer layer. The spin quantum number  $S$  is unusually high, like in Ref. 28, which suggests the formation of magnetic polarons by several defects around the interface. Actually, a simple calculation has been done in Ref. 29, which shows that small magnetic clusters can be formed by several defects.

Given the above analysis, we infer that the large spin-splitting energy can be interpreted based mainly on the magnetism from the magnetic polarons. For a MOVPE grown sample, the interface is usually not sharp due to the “composition pulling effect” of Mg composition. This has been proven recently by capacitance-voltage measurements in MOVPE grown  $\text{Mg}_x\text{Zn}_{1-x}\text{O}/\text{ZnO}$  heterostructures.<sup>13</sup> In

this case, electrons wavefunction could penetrate into the barrier, and has large amplitude around the interface. The overlap between the mobile electrons wavefunction and magnetic polarons is thus strongly enhanced. Due to the effective exchange interaction between electrons and magnetic polarons in the  $\text{Mg}_x\text{Zn}_{1-x}\text{O}$  layer around the interface, the magnetism there should be enhanced and large spin splitting of sub-band arises. Therefore, the 2DEG is spin polarized. In view of the fact that the 2DEG in the first sub-band has much larger amplitude of wavefunction around the interface than that in the second sub-band, we expect that the magnetic exchange interaction for electrons in the first sub-band is stronger than that in the second sub-band. This explains why the spin splitting can only be observed in the first sub-band.

In conclusion, the quantum transport measurements of undoped  $\text{Mg}_x\text{Zn}_{1-x}\text{O}/\text{ZnO}$  heterostructure grown by MOVPE have been performed at low temperatures and high magnetic fields. A large zero-field spin-splitting energy of the 2DEG more than 15 meV is obtained at 1.6 K. Meanwhile, the defect-induced ferromagnetism is observed in the heterostructures. The fitting of magnetism according to Brillouin function indicates the formation of magnetic polarons around the interface. It is believed that the magnetic exchange interaction between itinerant electrons in the two-dimensional channel and the magnetic polarons in  $\text{Mg}_x\text{Zn}_{1-x}\text{O}$  barrier near the interface plays a major role in the spin polarization of the 2DEG. We hope that our results pave a new way for new spintronic devices based on ZnO semiconductor systems.

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