Two-dimensional electron gas in Zn-polar ZnMgO/ZnO heterostructure
grown by metal-organic vapor phase epitaxy

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We report the formation of two-dimensional electron gas (2DEG) at the Zn1−xMgO/ZnO interface
grown by metal-organic vapor phase epitaxy on sapphire substrates. The existence of the 2DEG
is confirmed by the observation of Shubnikov–de Haas oscillations and the integer quantum
Hall effect. In particular, the Zn0.8Mg0.2O/ZnO heterostructure shows a high Hall mobility of
2138 cm2/V s with a carrier sheet density of 3.51 × 1012 cm−2 at 1.4 K. We attribute the origin of
2DEG to be the donor states on ZnMgO surface. The dependence of carrier sheet density of 2DEG
on ZnMgO layer thickness and Mg composition (x) are also investigated. © 2010 American
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Since the observation of Shubnikov–de Haas (SdH) oscillations and the quantum Hall effect (QHE) in high
mobility two-dimensional electron gas (2DEG) in polar ZnO/Zn1−xMgO heterostructures,1 there is a considerable
interest in the utilization of polarization field in polar oxide based heterostructure epitaxy. So far, the formation of 2DEG
at the Zn1−xMgO/ZnO interface has been observed only by a few groups using either molecular beam epitaxy (MBE)
or pulse laser deposition techniques.2–7 On the other hand, metal-organic vapor phase epitaxy (MOVPE)
is well known to be one of the most important deposition processes for making high-quality epilayers and well-defined heterostructure
with homogeneous properties over a large deposition area. In this work, we report the formation of 2DEG confined
in Zn-polar Zn1−xMgO/ZnO heterostructure which can be realized by the MOVPE technique. The formation mechanism and transport properties of 2DEG are also discussed on the basis of a simple charge control model that utilizes the effect of polarization-induced charge density.

Zn1−xMgO/ZnO heterostructures were grown on sapphire (0001) substrates using Aixtron MOVPE system with
close-coupled showerhead (CCS) configuration. Dimethylzinc (DMZn) and bis(methylcyclopentadienyl) magnesium
(MeCp2Mg) were used as Zn and Mg sources, respectively. We used the nitrous oxide (N2O) as the oxidizing source
and nitrogen gas as the carrier gas. After annealing the substrate in N2 at 1000 °C, a 0.4-μm-thick ZnO buffer layer was deposited at 450 °C, followed by a 3.3 μm undoped ZnO layer and an undoped Zn1−xMgO layer, both grown at 920 °C. We note that the deposited ZnO layer exhibits a Zn-face polarity with excellent crystal quality and layer-by-layer surface morphology.8 A series of Zn1−xMgO layer with different thickness ranging from 27 to 120 nm was also grown under same conditions. The Mg composition (x) was determined from the reflectance measurement of the band

gap energy of Zn1−xMgO and using the equation

\[ E_g(x) = E_g(0) + 2.145x \]

The capacitance-voltage (C-V) measurement was performed using mercury contacts at frequency of
1 MHz. The Hall effect and magnetotransport were carried out using a van der Pauw configuration with a magnetic field
up to 10 T at 1.4 K.

Figure 1 shows a typical atomic force microscopy (AFM) image of Zn0.83Mg0.17O epilayer with thickness of
27 nm. The growth condition is optimized in order to obtain a two-dimensional layer-by-layer epitaxial mode for the Zn-polar Zn1−xMgO layer. We observe the surface morphology is dominated by wide terrace features with surface rms roughness of ~0.23 nm. Figure 2 shows the asymmetric reciprocal space map of (105) plane measured by high-resolution x-ray diffraction. Both the ZnO and Zn0.83Mg0.17O peaks are located at the same position of Qs, the reciprocal space vector in the plane of the layers. This feature suggested

\[ \text{FIG. 1. AFM image of Zn0.83Mg0.17O epilayer with thickness of 27 nm on ZnO template.} \]
that the Zn$_{0.83}$Mg$_{0.17}$O layer is coherently strained to the underneath ZnO layer.

Figure 3 displays the typical C-V concentration profile as a function of distance from the top surface for the Zn$_{0.83}$Mg$_{0.17}$O/ZnO heterostructure. The high electron concentration observed is the confined 2DEG at the interface. As we move away from the interface, the net donor density falls off sharply, reaching a value of $1.5 \times 10^{16}$ cm$^{-3}$ at the ZnO layer. The dip of carrier concentration in the depth-profiling at 0.125 μm is due to the undoped ZnO grown under the conditions of lower rate and higher V/I ratio. The integration of area under the accumulation peak of the Zn$_{0.83}$Mg$_{0.17}$O/ZnO interface yields a sheet concentration of $1.23 \times 10^{12}$ cm$^{-2}$ for the 2DEG. We found that the sheet carrier concentration increases with the ZnMgO layer thickness as well as the x composition. This behavior is similar to the case in 2DEG at AlGaN/GaN heterostructure, in which strong electric polarization effects lead to the formation of high carrier concentration.\textsuperscript{10–13} The formation mechanism of 2DEG will be discussed from the viewpoint of polarization effects later.

Figures 4 show the magnetoresistance ($R_{xx}$) and Hall resistance ($R_{yx}$) as a function of magnetic field (B) up to 10 T at 1.4 K for the Zn$_{0.83}$Mg$_{0.17}$O/ZnO heterostructure. In the magnetoresistance, strong SdH oscillations are clearly seen and the 2DEG density is estimated by the oscillation period to be $1.91 \times 10^{12}$ cm$^{-2}$. The observation of the QHE effect gives direct evidence of the existence of a 2DEG at the ZnMgO/ZnO heterostructure interface. The onset of SdH oscillations occurs at a B field of 3 T and the spin splitting of the Landau levels is visible at filling factors as high as $\nu=37$. The electron density was determined to be $n_{1}=3.51 \times 10^{12}$ cm$^{-2}$ from the low-field Hall resistivity and its corresponding mobility is 2138 cm$^2$/Vs. The discrepancy in the sheet density determination from the magnetoresistance and Hall effect could be due to the significant parallel conduction in our sample. Additionally, the minima in $R_{xx}$ do not approach zero in the integer QHE states. We believe that the parallel conduction path is originated from the interfacial layer of ZnO and sapphire, which suffered from a large dislocation density and Al donor diffusion from sapphire substrate during the high temperature epilaxial growth. This parallel conduction results in the underestimation of Hall mobility of 2DEG at the ZnMgO/ZnO interface.

In order to understand the behavior of 2DEG density as a function of Zn$_{1-x}$Mg$_x$O layer thickness and x composition, we need to consider the mechanisms that control the transfer of electrons in the Zn$_{1-x}$Mg$_x$O/ZnO system. We make use of a simple charge control model to calculate the 2DEG sheet carrier concentration and then compare with our work and those reported in the literature. In the absence of the external field, the total polarization of ZnO or Zn$_{1-x}$Mg$_x$O layers is the sum of the spontaneous polarization ($P_{SP}$) and the strain-induced piezoelectric polarization ($P_{PE}$), and their directions are indicated in the inset of Fig. 3.

We assume that the Zn$_{1-x}$Mg$_x$O layer is fully strained on ZnO and polarization constants vary linearly with x composition. Thus, the dependence of total polarizations induced charges on the x composition in Zn$_{1-x}$Mg$_x$O barrier can be expressed as $\sigma(x)=0.029x$ (C/m$^2$).\textsuperscript{6} Since the surface of the grown Zn$_{1-x}$Mg$_x$O/ZnO is Zn-polar, the positive polarization-induced charges are located at the interface (see inset of Fig. 3). These positive charges tend to be compensated by the electrons that formed a 2DEG at the Zn$_{1-x}$Mg$_x$O/ZnO interface. The maximum sheet carrier concentration ($n_S$) located at this interface of the nominally undoped structure can be expressed as\textsuperscript{11,12,15}

$$n_S(x) = \frac{+\sigma(x)}{e} + \frac{\varepsilon_0 \varepsilon_r(x)}{e^2} \left[ e \phi_0(x) + E_F(x) - \Delta E_c(x) \right],$$

where $d$ is the width of the ZnMgO layer thickness and x is the Mg content. The surface potential $e \phi_0(x)$ is assumed to
The effective mass is taken to be \( m^* \approx 0.26m_e \) and the dielectric constant is approximated to be linearly dependent on \( x \); \( \varepsilon(x) = 8.75 + 1.08x \). By substituting the required values in Eq. (1), the maximum \( n_C \) can be determined.

Figure 5(a) shows the sheet carrier concentrations \( n(x) \) as a function of \( x \) composition for different Zn\(_{1-x}\)Mg\(_x\)O layer thickness. The solid lines correspond to the calculated results based on Eq. (1) for \( d = 15, 50, \) and 300 nm. The 2DEG density is seen to increase approximately linearly with \( x \) and the slope shows a very slight variation of \((1.83 - 1.98) \times 10^{13} \) cm\(^{-2}\) indicating that the dependence on the Zn\(_{1-x}\)Mg\(_x\)O layer thickness is small. This trend is very similar to those observed in O-polar or Zn-polar Zn\(_{1-x}\)Mg\(_x\)O/ZnO structures grown by MBE.\(^{5,14}\) The calculated 2DEG density agrees well with our experimental results and also those reported in the literature obtained either by C-V profiling or low temperature Hall measurements.\(^{1-4,9}\) Figure 5(b) shows the dependence of sheet carrier concentration \( n(x) \) as a function of Zn\(_{1-x}\)Mg\(_x\)O thickness for different \( x \). The solid lines correspond to the calculated results based on Eq. (1) for various \( x \). As the Zn\(_{1-x}\)Mg\(_x\)O layer thickness increases, the 2DEG density approaches the maximum value of polarization-induced sheet charge density \( \sigma(x) \) as indicated by dashed lines in Fig. 5(b).

The charge control model of Eq. (1) sets a cut-off critical thickness \( t_{CR} \) of the Zn\(_{1-x}\)Mg\(_x\)O layer below which the 2DEG is not formed. The \( t_{CR} \) decreases from 38 to 1 nm as the \( x \) increase from 0.05 to 0.45. This behavior is similar to that of AlGaN/GaN heterostructure.\(^{11}\)

The good agreement between the theoretical prediction and experimental results allow us to infer the source of electrons in 2DEG. If ZnO layer was the origin of the 2DEG, then the carrier concentration would be independent of Zn\(_{1-x}\)Mg\(_x\)O layer thickness as well as \( x \) composition. The 2DEG density is also not proportional to the Zn\(_{1-x}\)Mg\(_x\)O layer thickness, which suggests that the effect of donors in ZnMgO layer is not so important. The same conclusion is drawn by Tampo et al.\(^{16}\) that the mobile 2DEG charges originate from the donorlike surface states and oxygen vacancies are expected to be most possible candidates, which has a deep level of 0.7 eV below the conduction band of ZnO.\(^{18}\)

We note that there is somewhat a large discrepancy in the sheet carrier concentration values obtained by C-V profiling (e.g., \( 1.23 \times 10^{12} \) cm\(^{-2}\) for Zn\(_{0.83}\)Mg\(_{0.17}\)O/ZnO) and the calculated value (\( 2.02 \times 10^{12} \) cm\(^{-2}\)) using Eq. (1). This could be due to the depletion of a thinner ZnMgO barrier layer that led to an underestimation of the integrated carrier concentration of the C-V profiling. Additionally, the unintentional composition gradient during the growth of Zn\(_{1-x}\)Mg\(_x\)O layer will cause a slight increase in the band gap energy and the \( x \) composition with the Zn\(_{1-x}\)Mg\(_x\)O in our samples. This is possibly due to the composition-pulling effect, which has been also observed in the MBE growth for rich-Mg Zn\(_{1-x}\)Mg\(_x\)O and also in In\(_{1-x}\)Ga\(_x\)N growth.\(^{19,20}\)

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