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Anomalous circular photogalvanic effect of the spin-polarized two-dimensional electron gas in $Mg_{0.2}Zn_{0.8}O/ZnO$ heterostructures at room temperature

J. X. Duan,[1] N. Tang,[1,a)] J. D. Ye,[2,3] F. H. Mei,[1] K. L. Teo,[4] Y. H. Chen,[5] W. K. Ge,[1,6] and B. Shen[1,b)]

[1]*State Key Laboratory of Artificial Microstructure and Mesoscopic Physics, School of Physics, Peking University, Beijing 100871, China*
[2]*School of Electronic Science and Engineering, Nanjing University, Nanjing 210093, China*
[3]*Department of Electronic Materials Engineering, Research School of Physics and Engineering, The Australia National University, Canberra 0200, Australia*
[4]*Department of Electrical and Computer Engineering, National University of Singapore, 117576 Singapore*
[5]*Laboratory of Semiconductor Materials Science, Institute of Semiconductors, CAS, Beijing 100083, China*
[6]*Department of Physics, Tsinghua University, Beijing 100084, China*

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A spin-related photocurrent with swirly distribution and anomalous dependence of the total spin-related photocurrent on the incident angle were observed on spin-polarized two-dimensional gas in a $Mg_{0.2}Zn_{0.8}O/ZnO$ heterostructure under illumination of circular polarized light at room temperature. The ferromagnetic two-dimensional Rashba model was adopted to interpret the results. It is demonstrated that a radial spin current induced by the gradient of the spin-polarized electron density is the origin of the anomalousness. This spin current only exists in spin polarized systems. © *2013 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4805079]

Spintronics has attracted much attention for its potential applications in information technology as well as revealing essential questions on the physics of electron spin in condensed matter.[1] Spin generation, manipulation, and detection are the three major aspects in the realization of spin-based devices. Many studies have been done on dilute magnetic semiconductors (DMSs) for their advantages in spin generation, as they can be ferromagnetic at relatively high temperature.[2–4] Spin Hall effect and reciprocal spin Hall effect (RSHE) serve as alternative ways to converting charge current into spin current, and *vice versa*, via spin-orbit coupling (SOC).[5,6] Both of the two effects are extensively studied and found to be useful in spin generation and detection in nonmagnetic systems.[7–9]

ZnO, a wide-bandgap semiconductor, serves as the targeting material for spintronics study, due to a prediction that it is a promising host material to achieve room-temperature ferromagnetic DMS.[10] Theoretical calculations have indicated that the Zn vacancy does lead to magnetism in nonmagnetic doped ZnO.[11] The realization of two-dimensional electron gases (2DEG) in $Mg_xZn_{1-x}O/ZnO$ heterostructures makes ZnO the most possible material to combine 2DEG with magnetism, which will surely be useful in spintronics.[12,13] As a matter of fact, the spin polarization of $Mg_xZn_{1-x}O/ZnO$ heterostructures has been demonstrated in the previous study.[14] Thus, it is quite desirable and fascinating to study the spin properties through RSHE on spin-polarized systems.

In this study, we investigate the spin properties of spin-polarized 2DEG in $Mg_xZn_{1-x}O/ZnO$ heterostructures by the circular photogalvanic effect (CPGE).[15] We will report the observation of anomalous spin-related photocurrent (SRPC) in ZnO-based heterostructures, and propose a model of swirly SRPC generated through RSHE which can exist in spin-polarized electron systems with SOC. To avoid ambiguity, the CPGE current only refers to the one introduced in Ref. 15, in which RSHE is excluded, while SRPC referring to the total circular photogalvanic photocurrent, including CPGE current.

Zn-faced unintentionally doped $Mg_xZn_{1-x}O/ZnO$ heterostructures were grown on sapphire (0001) substrates by metal organic vapor phase epitaxy (MOVPE).[16] The ZnO film consists of a 400 nm-ZnO buffer layer grown at 480 °C and a 1500 nm-ZnO epilayer grown at 900 °C, followed by a $Mg_{0.2}Zn_{0.8}O$ barrier layer deposited on ZnO at 900 °C. The high-mobility 2DEG in the triangular quantum well (QW) is formed in the ZnO epilayer at the heterointerface.

Quantum transport properties of a series of samples with different thickness of barrier layer were investigated in van der Pauw geometry in a rotator-equipped Oxford refrigerator at 1.6 K with magnetic fields up to 14 T. The Hall electron density $n_H = 4.56 \times 10^{12}\,\mathrm{cm}^{-2}$ and mobility $\mu = 1845\,\mathrm{cm}^2/\mathrm{Vs}$ at 1.6 K are obtained from the low-field Hall measurements. A large zero-field spin-splitting energy more than 15 meV was obtained in the 2DEG by Shubnikov-de Haas oscillation. In our previous report, Ref. 14, we demonstrated an observation of unexpected ferromagnetism of the 2DEG in the measurement conducted by a superconducting quantum interference device (SQUID) magnetometer. Room-temperature ferromagnetism was also observed from the temperature dependence in the magnetization measurements. It is demonstrated that the 2DEG is indeed spin-polarized. Details of the measurements and results can be found in the previous report.

The geometry of the sample used in our CPGE measurements is sketched in the inset of Fig. 1. A diode pumped

a)ntang@pku.edu.cn
b)bshen@pku.edu.cn

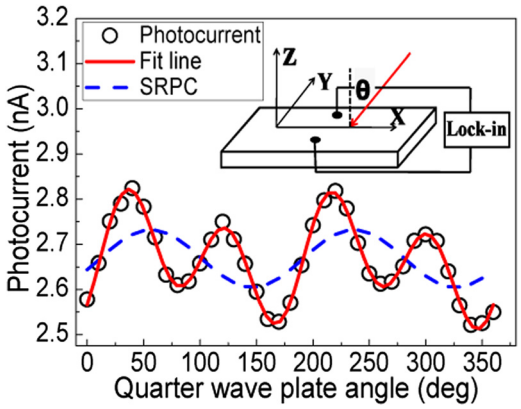

FIG. 1. Total photocurrent measured at normal incidence as the light spot fixed at $x = 0.8$ mm on the right side of the electrodes. Solid line is the fit line, and dashed line denotes the SRPC. The inset sketches the set up of the experiment.

solid state laser with wavelength of 1064 nm and 500±10 mW radiation power provides the excitation. A quarter-wavelength plate is used to shift the polarization degree of the incident light. The total photocurrent is measured by a lock-in amplifier after a preamplifier. The sample, which has a 20 nm thick barrier layer, was cut into a small strip of 12 mm length and 5 mm width, with two Ohmic electrodes made by evaporating Ti/Al/Ni/Au metal multilayer structure and separated by a distance of 3 mm along Y direction. The incident light spot with a Gaussian profile and a diameter of 2 mm is located at the central line X ($y = 0$) between the two electrodes, and can be moved along X, while the incident plane being perpendicular to the connection line between the two electrodes all the time.

The total photocurrent measured can be fitted by the formula,

$$J_{tot} = J_1 \sin 2\varphi + J_2 \sin 4\varphi + J_0, \tag{1}$$

in which J_1 is the amplitude of the SRPC, J_2 the amplitude of the linear photogalvanic effect current, J_0 the background current, and φ the rotation angle between the polarization direction of the incident light and optical axis of the quarter-wave plate.[15] Figure 1 shows the photocurrent we measured with the light spot fixed at $x = 0.8$ mm on the right of the electrodes at normal incidence.

In our experiments, the photon energy is 1.1 eV, which is much smaller than the band gap of ZnO. The photon absorption basically takes place in the 2DEG plane where the electrons in the triangular QW will be excited from sub-bands to higher bulk states.[17] Because of the strong structural inversion asymmetry (SIA), Rashba spin splitting exists in the QW. Theoretically, for a 2DEG in a heterostructure with symmetry of point group C_{6v}, the SRPC only includes the CPGE current, and it can be expressed as $j_y = \gamma_{yx} i (E_y \times E_z^*)_x$, where j_y is the photocurrent density, γ a second-rank pseudo-tensor, $\vec{E}$ the complex amplitude of the electric field of the incident electromagnetic wave, and X, Y the two orthogonal directions in the 2DEG plane as defined previously.[15] The CPGE current shows a symmetric Gaussian spatial distribution as the incident light spot moving from left to right of the electrodes along line X since it depends on the intensity of the incident light. It is also a function of the incident angle which can be expressed as $j_1 \propto t_p t_s \sin\theta/\sqrt{\varepsilon}$, where θ is the incident angle between Z axis and the direction of the light, ε is the dielectric constant of ZnO, and $t_p t_s = \frac{4\cos^2\theta}{(\cos\theta+\sqrt{\varepsilon-\sin^2\theta})(\varepsilon\cos\theta+\sqrt{\varepsilon-\sin^2\theta})}$ is the product of the transition coefficient for the s- and p-polarized light.[15] As such, there should be no CPGE under normal incidence for the lack of Z component of the electric field, and when the incident angle is not that large, the CPGE should become larger with the incident angle increasing. However, a mechanism which can generate SRPC at normal incidence had been reported.[18,19] This effect was named anomalous CPGE (ACPGE) and generated by the radial spin current induced by the gradient of the spin polarization distribution through RSHE. The ACPGE current has an anti-symmetric spatial distribution, which is largest at normal incidence, and decreases with increasing the incident angle. The total SRPC measured can then be separated into two parts. The symmetric part is CPGE, which is a transverse current. The anti-symmetric part is ACPGE, which should be a swirly current.[18]

Figure 2(a) shows the measured SRPC at different incident angles with the light spot moving along line X. As sketched in Fig. 2(b), the symmetric and anti-symmetric parts reverse their sign at large incident angle, which is different from the ACPGE and CPGE predicted and reported previously. The product of the transition coefficient for the s- and p-polarized light does not change sign in the range of the incident angle we used. The index of refraction of $Mg_xZn_{1-x}O$ and ZnO is about 1.9 and 1.94, respectively. The refraction angle is 21.8° when incident angle is 45° for $Mg_{0.2}Zn_{0.8}O$. The difference between the refraction angles of the two layers, $Mg_xZn_{1-x}O$ and ZnO, is less than 0.5°. If the SRPC comes from different layers, the results cannot be interpreted by two layer contributions considering such a small angle difference.

We believe that it is the room-temperature spin-polarized 2DEG in the heterostructures that causes the sign inversion of the ACPGE. We hereby introduce a ferromagnetic two-dimensional Rashba (F2DR) model to explain our experimental results, based on the fact that the 2DEG in our sample is spin-polarized.[20] For the sake of simplicity, we treat the spin polarization as Zeeman spin splitting in a mean magnetic field induced by the room-temperature ferromagnetism. The Hamiltonian of the model is $\hat{H}(\vec{k}) = \frac{\hbar^2\vec{k}^2}{2m^*} - \lambda\vec{k}\cdot(\hat{\sigma}\times\vec{e}_z) - \Delta_0\hat{\sigma}_z$, where m^* corresponds to the effective mass of the electrons in the QW, λ is the coefficient constant representing the SOC, Δ_0 is the mean-field exchange splitting, $\hat{\sigma} = (\hat{\sigma}_x, \hat{\sigma}_y, \hat{\sigma}_z)$ are the Pauli matrices, and $\vec{e}_z$ is the unit vector of the Z direction. This Hamiltonian has the band dispersion $\varepsilon_\sigma(\vec{k}) = \frac{\hbar^2\vec{k}^2}{2m^*} - \sigma\Delta_k$, where $\Delta_k = \sqrt{\lambda^2k^2 + \Delta_0^2}$ and $\sigma = \pm$ labels the two eigenstates $|\vec{k}, \sigma\rangle$ at momentum $\vec{k}$. The spin of the electron in the QW is out of the plane. The light excites electrons in the QW to higher energy levels. Because of the spatial Gaussian profile of the intensity of the incident light spot, the density of the electrons in the QW will also have spatial distribution. Since the electrons in the QW are spin polarized, the gradient of the density distribution can induce a radial spin current, which only exist in spin-

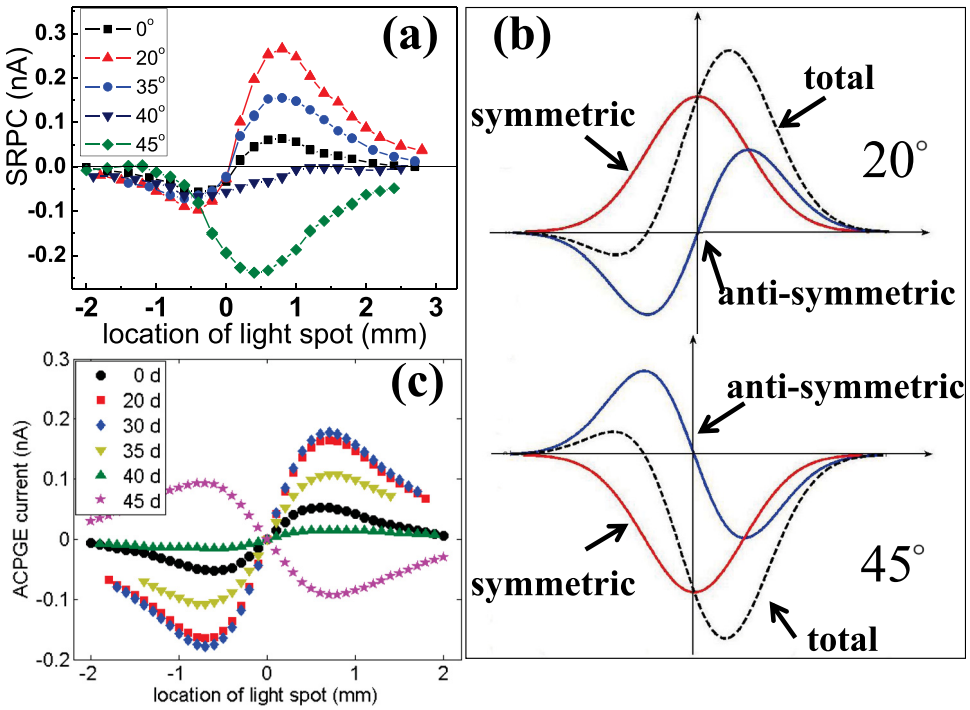

FIG. 2. (a) SRPC current as a function of the light spot location at different incident angle. (b) Sketches of the superposition of the symmetric and the anti-symmetric parts at 20° and 45°. (c) ACPGE currents at different incident angles (being interpolated).

polarized systems. This radial spin current can generate a swirly charge current through RSHE, which contributes to the ACPGE. As the excitation, and so that the spin current, is modulated by the helicity of the incident light, the swirly charge current can be obtained precisely by fitting the data to Eq. (1).

We now discuss the dependence of the ACPGE current on the incident angle. In order to have a semi-quantitative analysis, we will only consider the limitation of the conservation of the angular momentum in the excitation processes. The distribution of the total polarization of the system is $P^z(r) = s^z(r)n(r)$, where $n(r)$ is the density of carriers, and $s^z(r)$ is the spin polarization per carrier at position r. The spin current could thus be written as $q^z(r) = -D \cdot \nabla P^z(r) = -D \cdot s_0^z \frac{\partial n}{\partial r} - D \cdot n_0 \frac{\partial s^z}{\partial r} = q_d^z + q_p^z$, where D is the coefficient of diffusion, and the bottom index 0 denotes the corresponding variables at no incidence of light. The first part comes from the gradient of the density of spin-polarized carriers. The second part is from the gradient of the spin polarization when there is no gradient of density. The intensity of the incident light could be expressed as $I(x, y, \theta) = I(\theta)e^{-\frac{(x\cos\theta)^2+y^2}{r_0^2}}$ at incident angle θ, and $I(\theta) = I(0)\cos\theta$, where $I(0)$ is the intensity at normal incidence. The density and polarization of the electrons are both proportional to the intensity of the incident light, as shown in Fig. 3(a).[18,19] We can write these two terms as $n(r) = n_0(1 - f(r))$ and $s^z(r) = p \cdot f(r) \cdot \cos\theta_r$, respectively, where $f(r)$ is the probability of excitation at r, $p = 1/2$ represents the angular momentum taken by one excited electron, and θ_r is the corresponding refraction angle. The existence of $\cos\theta_r$ in the second term bases on the fact that only the polarization in z direction could contribute to the ACPGE. Therefore, these two radical spin current can be expressed as $q_d^z(r, \theta) = D \cdot \langle s_0^z \rangle n_0(r) \frac{\partial f(r)}{\partial r} \cos\theta$ and $q_p^z(r, \theta) = -D \cdot n_0(r) p \frac{\partial f(r)}{\partial r} \cos\theta \cdot \cos\theta_r$ at oblique incidence, where $\langle s_0^z \rangle$ is the average spin polarization of the 2DEG. These two spin currents have different dependences on the incident angle, so they will cancel each other at a certain angle when they are in opposite directions.

We now try to fit the experimental results by our model. Fig. 2(c) shows the ACPGE at different incident angles. It seems that our model fits the result quite well around the inverse point (Fig. 3(b)). What interests us is the evidence of the existence of the ACPGE induced by gradient of the density of spin-polarized carriers, the inverse of the current indeed is strong evidence. The inverse point now can be

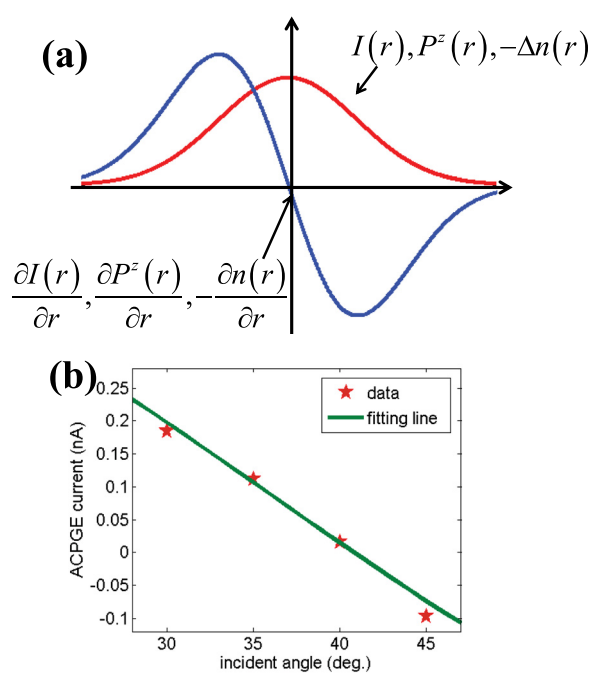

FIG. 3. (a) Sketch of the profile and gradient of intensity of light, spin polarization, and density of carrier. (b) The fitting of the magnitude of ACPGE current to the incident angle.

expressed as $\theta_i = \arccos\left(\frac{\langle s_0^z \rangle}{p}\right)$. Actually, we have not considered the spin relaxation of q_p^z, which will reduce the ACPGE generated by it. This effect could be represented in our model by decreasing the value of p. Thus, $\langle s_0^z \rangle$ will be determined to be much smaller than 1/2, which suggests that our model is valid.

The sign reversion of the symmetric part is quite different. Swirly current comes from the spatial distribution of the total spin polarization. The incident light transfers its angular momentum to the electron gas to generate spin polarization through excitation. The process does not depend on the specific excitations. However, CPGE current is induced by the asymmetric distribution in momentum space. Actually, we had also observed sign reversion of the CPGE current on nonmagnetic $Al_xGa_{1-x}N/GaN$ heterostructures and ZnO bulk samples under illumination of 1064 nm laser. We believe that the sign inversion of CPGE has nothing to do with magnetism, thus the ACPGE current. Further investigation is necessary to understand this interesting phenomenon.

In summary, SRPC has been measured in a $Mg_xZn_{1-x}O$/ZnO heterostructure which is spin-polarized at room temperature. The anti-symmetric part of the total SRPC is generated by two radial spin current through RSHE, as evidenced by the sign reversion at large incident angle. Besides the one had been reported previously, another kind of radial spin current induced by the gradient of the electron density distribution is suggested, which exists only in spin-polarized systems. We believe that the mechanism proposed here is a universal phenomenon in spin-polarized systems and suggest an experimental method to study the spin-polarized materials with strong SOC at room temperature.

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