Depolarization of backscattered linearly polarized light from ZnO thin film

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The depolarization behavior of backscattered linearly polarized light from ZnO thin film was investigated experimentally. The results show that the characteristics are related to both the polarization orientation and wavelength of linearly polarized incident light. When the incident light is s-polarized, the depolarization behaviors are different for different wavelengths. When the incident light is p-polarized, the depolarization behaviors, on the contrary, are similar for different wavelengths. In addition, there is an optimal incident angle for depolarization of linearly polarized light with different wavelengths, which is equal to their effective Brewster angles, respectively.

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Multiple scattering sequences are found to be the origin of the observed polarization behavior[1]. When polarized light enters a random medium, multiple scattering randomizes the polarization state so that the emergent light is partially depolarized[2,3]. Scalar-wave models for optical fields have been successfully used to explain numerous light scattering phenomena. However, the depolarization of light in a random medium is still not completely understood because of the complexity of vector wave multiple scattering. First order vector perturbation theory[4] for light scattering from interfacial roughness of a dielectric layer is used to study the intensity distribution of backscattered light field, but it cannot reveal the depolarization behavior. Dominique *et al.* have got the relation between entropy production and depolarization[5]: entropy production per scattering due to the irreversible process of depolarization is an exponentially decreasing function of the number of scattering events. And it is essentially an entropy effect arising from the irreversible evolution of the polarization state during scattering. Recent studies[3,4,6,7] have obtained the depolarization characteristics of incident linearly polarized light, but they did not deal with the differences of depolarization arising from incident linearly polarized light with different polarization directions. The wavelength dependence of depolarization behavior was not considered so far. In this paper we investigate the depolarization of linearly polarized incident light with different polarization directions and wavelengths from ZnO thin film experimentally.

ZnO is an important II-VI semiconductor with a room-temperature band gap energy of 3.37 eV and a large exciton binding energy ($\sim$ 60 meV). It has many practical or potential applications, such as random laser[8], transparent conductive films[9]. But the depolarization behavior from ZnO thin film has not been investigated heretofore. In this paper, the study of a detailed experiment is given to characterize the influence of polarization direction and wavelength on depolarization of linearly polarized light scattering from ZnO thin film.

ZnO powder was prepared by hydrolysis pyrogenation method. The typical synthesis procedure is as follows. 2.2 g (10 mmol) of $Zn(CH_3COO)_2{\cdot}2H_2O$ and 2 g (23.8 mmol) of $NaHCO_3$ were mixed at room temperature. The mixture was pyrolyzed at the reaction temperature. The $Zn(CH_3COO)_2{\cdot}2H_2O$ changed into ZnO powders, while the $NaHCO_3$ changed into CH_3COONa and eventually was washed away with the deionized water. ZnO powder was deposited on SiO_2 glass by electrophoresis method and ZnO film was prepared.

Structure of ZnO powders was characterized by a Bruker D8 Advance X-ray diffractometer (XRD) with Cu-$K\alpha$ (λ = 0.15406 nm) radiation. The 2θ range used in the measurement was $10° - 70°$ in steps of 0.05 deg./s. Atomic force microscopy (AFM) images were collected using a CSPM4000 scanning probe microscope system (Benyuan Nano-Instruments Co. Ltd., Beijing). A μMasch NSC21/AlBS cantilever (MicroMasch, Russia) with a force constant of 17.5 N/m was used. The contact scanning mode was used, and the scanning range was set at a size of 2.5×2.5 (μm). The refractive index was measured by a TPY-2 elliptical polarization thickness meter (Tianjin Tuopu Co. Ltd., Tianjin). The depolarization experimental setup is shown in Fig. 1. The output from a semiconductor laser was used as incident source. It passed through a polarizer to produce linearly polarized light. When linearly polarized light was incident on the sample (ZnO thin film), measurements were made to

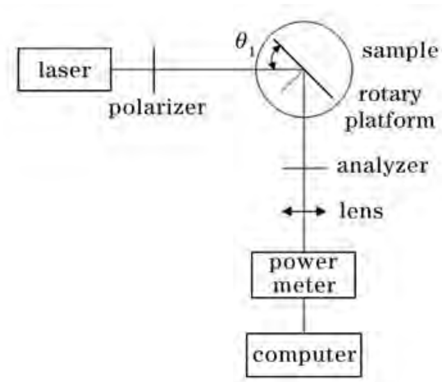

Fig. 1. Experimental setup.

record the backscattered maximal and minimal intensity by rotating the axis of an analyzer. The value for degree of polarization was determined as

$$P = \frac{I_{\max} - I_{\min}}{I_{\max} + I_{\min}}. \tag{1}$$

Figure 2 shows the schematic diagram of the scattering geometry. The linearly polarized light of wavelength λ irradiates the sample at an incident angle θ_i which is alterable in the experiment in the plane defined by x and z axes. We investigate the depolarization under the specular condition that backscattered angle θ_s is equal to incident angle θ_i and the scattered ray locates in x-z plane, i.e., scattering azimuthal angle is equal to zero. Unit vectors $\hat{k}_\mathrm{i}$ and $\hat{k}_\mathrm{s}$ describe the propagation directions of the incident and scattered light, respectively. In this paper, we study the s- and p-polarized incident linearly polarized light, where s- or p-polarized light is with the electric field parallel to the vector $\hat{s}_\mathrm{i}$ or $\hat{p}_\mathrm{i}$. $\hat{s}_\mathrm{i}$ is a unit vector perpendicular to both $\hat{k}_\mathrm{i}$ and z, and $\hat{p}_\mathrm{i} = \hat{k}_\mathrm{i} \times \hat{s}_\mathrm{i}$. Likewise, the polarization directions of the scattered electric field are described by the components of the electric field along $\hat{s}_\mathrm{s}$ and $\hat{p}_\mathrm{s}$ directions.

Figure 3 shows the XRD patterns of ZnO powder. The result indicates that ZnO has a polycrystalline hexagonal wurtzite structure. The major diffraction peak appears at $2\theta = 36.401°$. The full-width at half-maximum (FWHM) is $\beta = 0.554°$. The grain size of the ZnO powders can be estimated by Scherrer formula $D = \frac{0.89\lambda}{\beta\cos\theta}$ with the result of 15.1 nm. Figure 4 shows the AFM images of the surface of the ZnO thin film. The root mean square (RMS) roughness is 3.9 nm.

Figure 5 illustrates the curves of scattering intensities varying with incident angles. Two semiconductor lasers with the wavelengths of 650 and 530 nm were used in our experiments. The experimental setup is almost the same

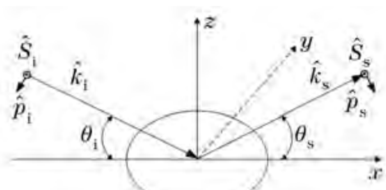

Fig. 2. Schematic diagram of the scattering geometry.

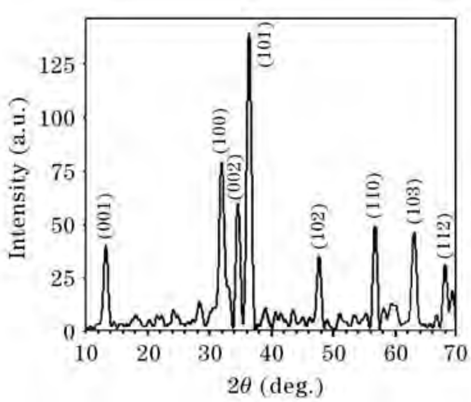

Fig. 3. XRD pattern of ZnO powders.

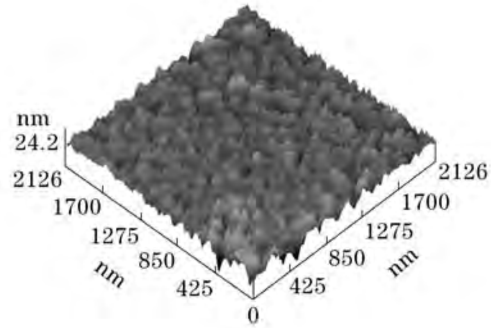

Fig. 4. AFM image of ZnO thin film.

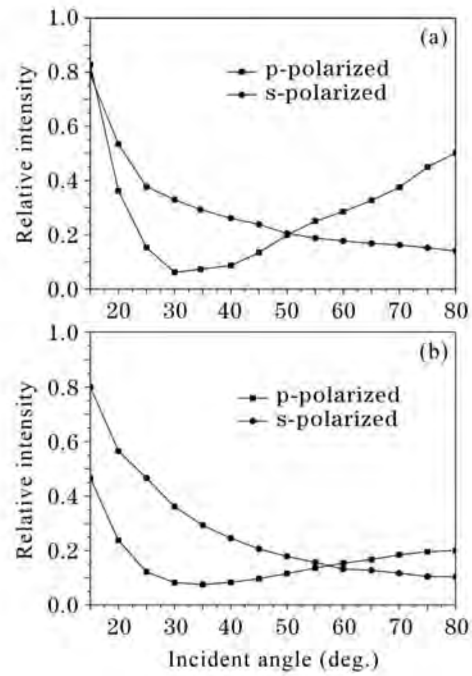

Fig. 5. Relative intensity versus incident angle for ZnO thin film for (a) 650 nm and (b) 530 nm.

as Fig. 1 but the analyzer is taken off. The incident linearly polarized light irradiated on the surface of sample, and scattering light was focused by lens then accepted by power meter at specular direction. The distance between power meter and sample was kept constant when the incident angle changed. When the incident light is s-polarized, the scattering intensity is a monotonically decreasing function of the incident angle. When incident light is p-polarized, the scattering intensity appears a minimum value (we call the incident angle corresponding to this minimum value as effective Brewster angle). The relation between Brewster angle and refractive index is

$$\theta_\mathrm{B} = \arctan(n_1/n_2), \tag{2}$$

where θ_B is the Brewster angle, n_1 is the refractive index of air, n_2 is the refractive index of sample. The effective refractive index of ZnO thin film was calculated through Eq. (2). From Figs. 5(a) and (b), we can obtain that the effective Brewster angles are 30° for 650 nm and 35° for 530 nm. Then the effective refractive indices of ZnO film are 1.732 for 650 nm and 1.428 for 530 nm. Meanwhile, the refractive index of ZnO thin film was obtained for

650-nm light as 1.730 by elliptical polarization thickness meter. So it is valid to measure the effective refractive index of thin film in this method.

In optical band, most natural random surfaces are not very rough, and the scale of local fluctuation is far larger than optical wavelength. When linearly polarized light irradiates such a sample, the scattered light at every angle is basically linearly polarized and the polarization orientation is the same as incident light. That is to say, the spatial distribution of scattered light intensity may be changed by natural random surfaces, but it commonly has no obvious depolarization effect[10]. Obviously, we can see that ZnO thin film has a random rough surface from the AFM photograph in Fig. 4, and the scale of local fluctuation is far smaller than optical wavelength. Under these conditions, we get the results which are entirely different from those in Ref. [10]. Figure 6 shows the relation between degree of polarization of measured scattered light and incident angle. The behavior of depolarization is different for the incident light with various polarization orientation. For the 650-nm light, when the linearly polarized light is s-polarized, the polarization behavior of scattered light is basically changeless, i.e., the scattered light is still linearly polarized. But if the linearly polarized light is p-polarized, the depolarization behavior is obvious. The degree of polarization of scattered light decreases with the increase of incident angle. When the incident angle is approaching to 30°, the scattered light nearly becomes natural light. The degree of polarization increases gradually with the increase of incident angle. The scattered light becomes linearly polarized light finally. At the same time we can find that optimal depolarized incident angle is the same as its effective Brewster angle (Fig. 5(a)). This phenomenon is just contrary with Ref. [11], which stated that the incident light is natural light, and the degree of polarization increases with the increase of incident angle. When the incident angle approaches the Brewster angle, the scattered light becomes linearly polarized light, then becomes partially polarized light by further increasing the incident angle. Figure 6(b) shows the case of 530-nm light. The behaviors of depolarization are approximately similar to those of 650-nm light, but the optimal depolarized incident angle changes. That is to say, the degree of polarization is a monotonically decreasing function of the incident angle when incident light is s-polarized; the optimal depolarized incident angle becomes 35° in the case of p-polarized light, which is equal to the effective Brewster angle (Fig. 5(b)).

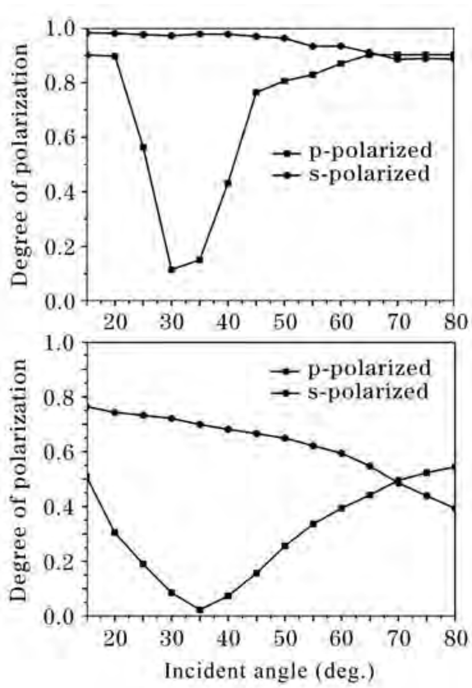

Fig. 6. Measured degree of polarization versus incident angle for p-polarized and s-polarized light with the wavelengths of (a) 650 nm and (b) 530 nm.

Incident linearly polarized light loses its polarization because of random multiple scattering. The Stokes vector S can be used to describe a polarization state with four elements (I, Q, U, V). The first Stokes parameter I is the common intensity, while the other three Stokes parameters specify the state of polarization of the light beam. So Stokes vector $\mathbf{S}$ is defined as

$$\mathbf{S} = \begin{bmatrix} I \\ Q \\ U \\ V \end{bmatrix}. \tag{3}$$

An incident pure state of polarization was considered, of unit intensity normal on the half-space $z > 0$; its Stokes vector is written as

$$\mathbf{S}_{\mathrm{i}} = \begin{bmatrix} \left\langle |E_x|^2 \right\rangle + \left\langle |E_y|^2 \right\rangle \\ \left\langle |E_x|^2 \right\rangle - \left\langle |E_y|^2 \right\rangle \\ \left\langle E_x^* E_y + E_x E_y^* \right\rangle \\ i \left\langle E_x^* E_y - E_x E_y^* \right\rangle \end{bmatrix}, \tag{4}$$

where E_x and E_y denote the x- and y-components of the electric field with respect to the Cartesian coordinate system chosen, the asterisk denotes the complex-conjugate value, and angular brackets denote time averaging. According to Ref. [7], the Stokes vector of outcoming light beam is

$$\mathbf{S}_{\mathrm{o}} = \begin{bmatrix} 1 \\ Qf(n) \\ Uf(n) \\ Vg(n) \end{bmatrix}, \tag{5}$$

with $f(n) = \frac{3(7/10)^n}{2+(7/10)^n}$ and $g(n) = \frac{3(1/2)^n}{2+(7/10)^n}$; $n+1$ is the number of scattering events.

Since the degree of polarization is defined as

$$P = \left(Q^2 + U^2 + V^2\right)^{1/2} \Big/ I, \tag{6}$$

then the output degree of polarization is given by

$$P = f(n) \left\{ Q^2 + U^2 + V^2 (5/7)^{2n} \right\}^{1/2}. \tag{7}$$

In this paper, only input pure linearly polarized light is considered ($E = E_x$ or $E = E_y$), so the output Stokes vector is

$$\mathbf{S}_{\mathrm{o}} = \begin{bmatrix} 1 \\ f(n) \\ 0 \\ 0 \end{bmatrix}, \tag{8}$$

and $P = f(n)$, which is a monotonically decreasing function of the number of scattering events.

The degree of polarization of scattered light is the function of the incident angle in the case of p-polarized incident light. It can be explained by the number of multiple scattering in sample. The energy loss is most serious at Brewster angle, and the number of multiple scattering is the largest. Consequently, from the formula of degree of polarization, $P = f(n)$, we can find that the degree of polarization is the smallest. We can also calculate the corresponding number of multiple scattering at different incident angles by measuring the degree of polarization. For example, the number of multiple scattering is about 16 when the degree of polarization is 0. It is very helpful to study multiple scattering of electric field in random media[12,13] and random laser[14].

In summary, two experimental evidences are presented in this paper. Firstly, the optical effective refractive index of ZnO thin film is obtained as 1.732 for 650-nm light and 1.428 for 530-nm light by measuring the effective Brewster angle. Secondly, the behavior of depolarization of ZnO thin film is different from that of natural random surfaces. Degree of polarization depends on both the polarization orientation and wavelength of incident linearly polarized light. When the incident light is s-polarized, the degree of polarization of scattered 650-nm light keeps constant; while the degree of polarization of scattered 530-nm light is a monotonically decreasing function of the incident angle. When the incident light is p-polarized, the behavior of depolarization of two kinds of incident light is similar. There is an optimal incident angle for depolarization of linearly polarized light with different wavelengths, which is equal to their each effective Brewster angle, respectively.

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