Linear-Polarization Optical Property of CdSe Quantum Rods*

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Abstract: The linear-polarization optical property of CdSe quantum rods is studied in the framework of effective-mass envelope function theory. The effects of shape and magnetic field on the linear polarization factors are investigated. It is found that CdSe quantum spheres have negative polarization factors (*xy*-polarized emission) and quantum long rods with small radius have positive linear polarization factors (*z*-polarized emission). The *z*-direction is the direction of the *c* axis. Quantum long rods with large radius have negative linear polarization factors, due to the hexagonal crystal symmetry and the crystal field splitting energy. The linear polarization factors decrease and may change from a positive value to a negative value; i.e., the *z*-polarized emissions decrease relative to *xy*-polarized emissions as the magnetic field applied along the *z* direction increases.

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1 Introduction

The method to synthesize CdSe nanostructures has improved. Single CdSe dots have been achieved[1], and CdSe quantum rods have also been synthesized[2], whose shape can be controlled[3,4]. Recently much attention has been paid to the linear-polarization optical property of these nanostructures[1,2,5~8]. These nanostructures have become a major subject of attention because of their prospective applications in devices. *xy*-polarized emissions from CdSe spheres were observed and used by Empedocles *et al*.[6] to monitor the rotationalmotion of quantum dots in various host matrices. A two-dimensional transition dipole was introduced to investigate the *xy*-polarized emissions[1,5], which are actually due to the hexagonal-crystal symmetry and the crystal field splitting energy. *z*-polarized emissions from CdSe rods were observed[2], and a one-dimensional transition dipole was introduced to investigate it[7].

Motivated by the experimental progress, we study the effects of the shape and magnetic field on the linear-polarization optical property of wurtzite quantum rods using the former model[9~13,15].

2 Model and calculation

If we take the basic functions of the valence-band top as

$$|1,1\rangle = (1/\sqrt{2})(X + \mathrm{i}Y)$$
$$|1,1\rangle = Z$$
$$\text{and } |1,-1\rangle = (1/\sqrt{2})(X - \mathrm{i}Y) \tag{1}$$

the effective-mass Hamiltonian[10] of a hole in the zero SOC case is written as

$$H_{\mathrm{h}0} = \frac{1}{2m_0}\begin{pmatrix} P_1 & S & T \\ S^* & P_3 & S \\ T^* & S^* & P_1 \end{pmatrix} \tag{2}$$

where

$$P_1 = \gamma_1 p^2 - \sqrt{\frac{2}{3}}\gamma_2 P_0^{(2)}$$
$$P_3 = \gamma_1' p^2 + 2\sqrt{\frac{2}{3}}\gamma_2' P_0^{(2)} + 2m_0\Delta_c$$
$$T = \eta P_{-2}^{(2)} + \delta P_2^{(2)}$$
$$T^* = \eta P_2^{(2)} + \delta P_{-2}^{(2)}$$
$$S = Ap_0 P_{-1}^{(1)} + \sqrt{2}\gamma_3' P_{-1}^{(2)}$$
$$S^* = -Ap_0 P_1^{(1)} - \sqrt{2}\gamma_3' P_1^{(2)} \tag{3}$$

$P^{(2)}$ and $P^{(1)}$ are the second and first-order tensors of the momentum operator, respectively, $p_0 =$

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$\sqrt{2m_0\Delta}$, and Δ = 40meV. Hereafter we take the negative hole energy to be positive. The effective-mass parameters[10] for CdSe are given in Table 1. The matrix elements of the tensors of the operators are given by Zhang *et al*.[15]. The SOC Hamiltonian is written as

$$H_{so} = \begin{pmatrix} -\lambda & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & \sqrt{2}\lambda & 0 & 0 \\ 0 & 0 & \lambda & 0 & -\sqrt{2}\lambda & 0 \\ 0 & \sqrt{2}\lambda & 0 & \lambda & 0 & 0 \\ 0 & 0 & -\sqrt{2}\lambda & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & -\lambda \end{pmatrix} \quad (4)$$

Table 1　Parameters for CdSe in the actual calculation

m_x	m_z	γ_1	γ_2	γ_2'	η	γ_1'	γ_3'	A	λ/meV	Δ_c/meV
0.1756	0.1728	1.7985	0.7135	0.7970	1.4492	2.166	0.3779	0.6532	139.3	25

Here, we take the basis functions to be $|1,1\rangle\uparrow$, $|1,0\rangle\uparrow$, $|1,-1\rangle\uparrow$, $|1,1\rangle\downarrow$, $|1,0\rangle\downarrow$ and $|1,-1\rangle\downarrow$. The envelope functions are

$$\Psi_h = \sum_{M=m+1/2}\sum_{l,n}\begin{pmatrix} a_{l,n,m-1,\uparrow}C_{l,n}j_l(k_n^l r)Y_{l,m-1}(\theta,\phi) \\ b_{l,n,m,\uparrow}C_{l,n}j_l(k_n^l r)Y_{l,m}(\theta,\phi) \\ d_{l,n,m+1,\uparrow}C_{l,n}j_l(k_n^l r)Y_{l,m+1}(\theta,\phi) \\ a_{l,n,m,\downarrow}C_{l,n}j_l(k_n^l r)Y_{l,m}(\theta,\phi) \\ b_{l,n,m+1,\downarrow}C_{l,n}j_l(k_n^l r)Y_{l,m+1}(\theta,\phi) \\ d_{l,n,m+2,\downarrow}C_{l,n}j_l(k_n^l r)Y_{l,m+2}(\theta,\phi) \end{pmatrix} \quad (5)$$

M is the z component of the total angular momentum. The effective-mass Hamiltonian of an electron is written as

$$H_{e0} = \frac{p^2}{2m_a} - \frac{1}{2m_b}\sqrt{\frac{2}{3}}P_0^{(2)} \quad (6)$$

where

$$\frac{1}{m_a} = \frac{1}{3}\left(\frac{2}{m_x} + \frac{1}{m_z}\right)$$
$$\frac{1}{m_b} = \frac{1}{3}\left(\frac{1}{m_x} - \frac{1}{m_z}\right) \quad (7)$$

We take the basic functions as $S\uparrow$ and $S\downarrow$, where S is the Bloch state of the conduction-band bottom. The envelope functions are

$$\Psi_e = \sum_m\sum_{l,n}\begin{pmatrix} e_{l,n,m,\uparrow}C_{l,n}j_l(k_n^l r)Y_{l,m}(\theta,\phi) \\ e_{l,n,m,\downarrow}C_{l,n}j_l(k_n^l r)Y_{l,m}(\theta,\phi) \end{pmatrix} \quad (8)$$

For quantum rods, we introduce a coordinate transformation[13]

$$x = x'$$
$$y = y'$$
$$z = ez'$$
$$p_z = p_{z'}/e \quad (9)$$

where e is the aspect ratio; i.e., $e = \frac{L}{2R}$, L is the length, and R is the transverse radius of the quantum rods.

We assume that the external magnetic field is applied along the z axis of the crystal structure. We choose the symmetric gauge, so that the vector potential is written as

$$\boldsymbol{A} = \left(-\frac{1}{2}B_z y, \frac{1}{2}B_z x, 0\right) \quad (10)$$

In the presence of an external magnetic field, the momentum operator becomes $\boldsymbol{p}+e\boldsymbol{A}$. Because the different components of $\boldsymbol{p}$ do not commute, then the $p_\alpha p_\beta$ terms in the Luttinger Hamiltonian are not symmetric. Luttinger[14] introduced the symmetrized product

$$\{p_\alpha p_\beta\} = \frac{1}{2}(p_\alpha p_\beta + p_\beta p_\alpha) \quad (11)$$

He divided the Luttinger Hamiltonian into two parts, the symmetric part and the antisymmetric part. The antisymmetric part is simply written as

$$H_{asym} = K\mu_B \boldsymbol{I}\cdot\boldsymbol{B} \quad (12)$$

The whole Hamiltonians of electron and hole, respectively, are

$$H_e = H_{e0} + H_{mm\_e} + H_{Zeeman\_e} \quad (13)$$
$$H_h = H_{h0} + H_{so} + H_{mm\_h} - H_{asym} - H_{Zeeman\_h} \quad (14)$$

The terms H_{mm_e}, H_{mm_h}, H_{Zeeman_e}, and H_{Zeeman_h} are given by Zhang *et al*.[15].

If the wave propagation is along the x axis of the crystal structure, the linear polarization factor is written as

$$P = (I_z - I_y)/(I_z + I_y) \quad (15)$$

I_z and I_y are the intensities of the z and y polarized transitions. With the optical transition between a given electron state and a given hole state, they are proportional to

$$I_z = \left\{\sum_{l,n,m,s} b_{l,n,m,s}e_{l,n,m,s}\right\}^2 \quad (16)$$

$$I_y = \left\{\sum_{l,n,m,s}(a_{l,n,m,s}e_{l,n,m,s} - d_{l,n,m,s}e_{l,n,m,s})/\sqrt{2}\right\}^2 \quad (17)$$

$a_{l,n,m,s}$, $b_{l,n,m,s}$, $d_{l,n,m,s}$ and $e_{l,n,m,s}$ are given in Eq.(5) and Eq.(8).

3　Results and discussion

The effective-mass parameters[10] for CdSe are given in Table 1. We use the energy unit ε_0 =

$\frac{1}{2m_0}\left(\frac{\hbar}{R}\right)^2$ and the dimensionless magnetic field strength unit $b=\frac{\hbar eB}{m_0\varepsilon_0}$.

3.1 Structure and shape effects

The effect of the crystal field splitting energy Δ_c on the linear-polarization optical property of CdSe quantum rods is shown in Fig. 1. From Fig. 1 (a), we see that the linear polarization factors of optical transitions of CdSe quantum spheres are negative. This is due to the hexagonal crystal symmetry and the crystal field splitting energy Δ_c, which lift the energies of the state with Z Bloch state and make the state with XY Bloch states the lowest state of the valence band, with the result that the linear polarization factors are negative, in agreement with the experiment results[1,5,6]. With bigger Δ_c, the energies of the state with Z-Bloch state are lifted more, and the linear polarization factors become even more negative. We also see that with bigger Δ_c, the linear polarization factors of optical transitions of CdSe quantum long rods with small radius are less positive and have smaller saturation values. From Fig. 1(b), we see that as e increases, the normalized intensity of σ_x transitions decreases, and the normalized intensity of σ_z transitions increases at first, then decreases slowly due to the decrease of the overlap of the electron and hole wave functions when e is sufficiently large. From Fig. 1(c), we see that as Δ_c increases,

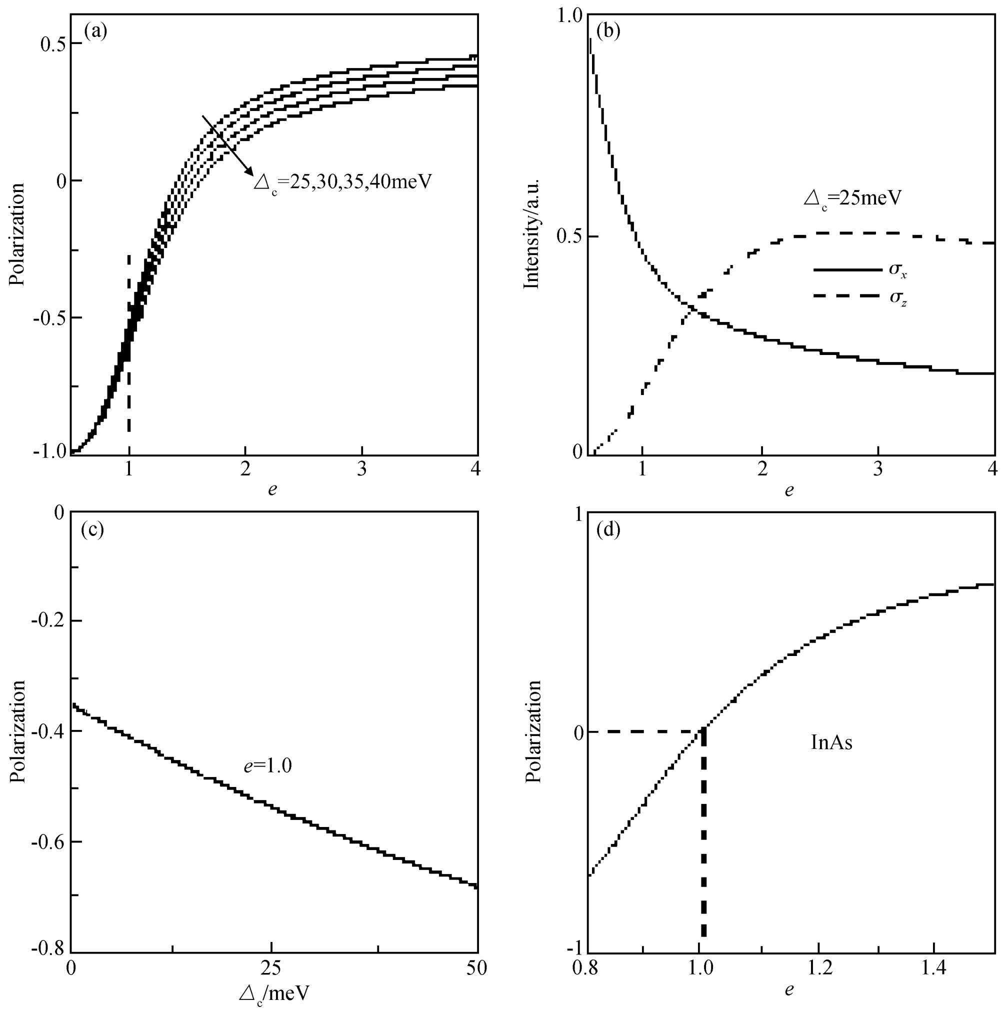

Fig. 1 (a) Linear polarization factors of optical transitions of CdSe quantum rods with $R=2$nm at $T=300$K as functions of e; (b) Normalized intensities of σ_x and σ_z transitions of CdSe quantum rods with $R=2$nm and $\Delta_c=25$meV at $T=300$K as functions of e; (c) Linear polarization factors of optical transitions of CdSe quantum spheres with $R=2$nm at $T=300$ K as functions of Δ_c; (d) Linear polarization factors of optical transitions of InAs quantum rods with $R=2$nm at $T=300$K as functions of e

the negative linear polarization factor decreases. With $\Delta_c = 0$, the linear polarization factor is not zero, but negative. This is due to the hexagonal crystal symmetry as we use the effective-mass parameters in Table 1. For example, γ_1 and γ_1' are not equal, while they are equal in zinc-blende structure semiconductor material. The linear polarization factor of a zinc-blende quantum sphere should be zero, as shown in Fig. 1(d).

The effect of the lateral dimension on the linear-polarization optical property of CdSe quantum rods is shown in Fig. 2. From Fig. 2(a), in which e is always greater than 1.66, we see that the linear polarization factors decrease from positive values to negative values as R increases. The linear polarization factors of optical transitions of CdSe quantum long rods with large radius are negative. This is because when R is sufficiently large, the quantum confinement effect that makes the state with the Z Bloch state the lowest state of the valence band is so weak that it cannot compensate the effect of the hexagonal crystal symmetry and the crystal field splitting energy Δ_c, which makes the state with XY Bloch states the lowest state of the valence-band. At higher temperatures, the linear polarization factors decrease more slowly. It is explicitly shown in Fig. 2(c) that CdSe quantum long rods with small radius have positive linear polarization factors, and CdSe quantum long rods with large radius have negative linear polarization factors. The normalized intensities are shown in Fig. 2(b) and Fig. 2(d), respectively.

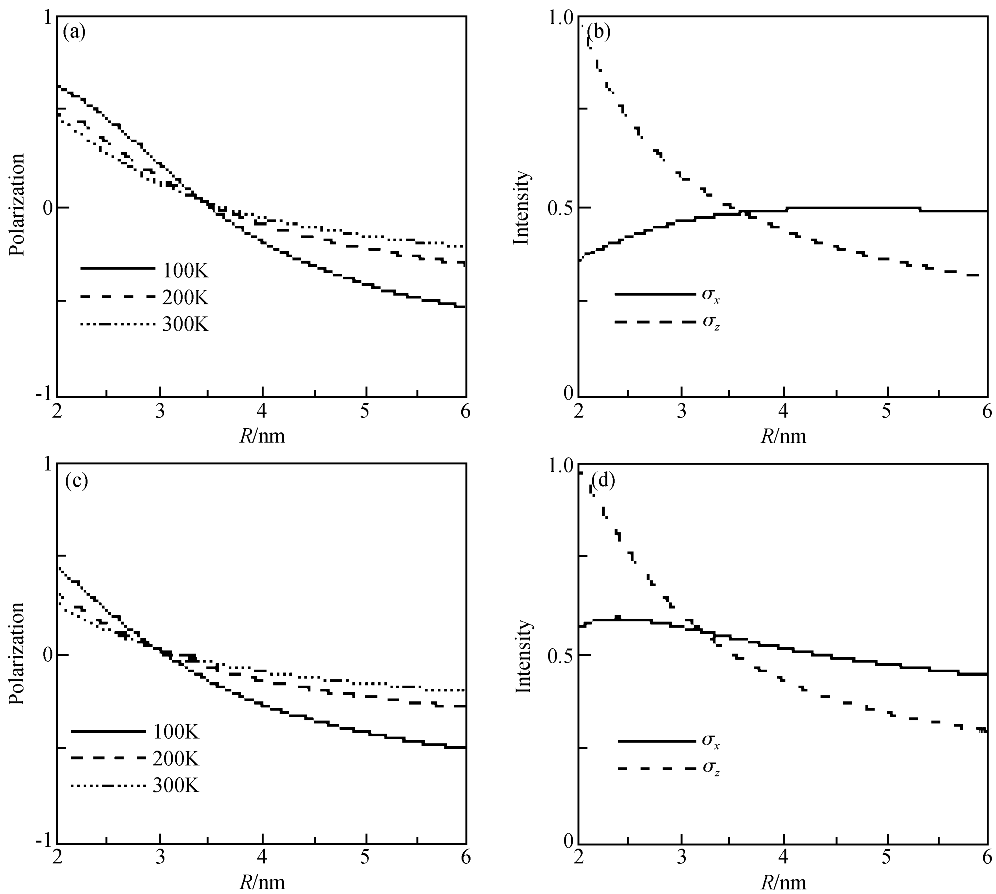

Fig. 2　(a) Linear polarization factors of optical transitions of CdSe quantum rods with $L = 20$nm and $\Delta_c = 25$meV as functions of R; (b) Normalized intensities of σ_x and σ_z transitions of CdSe quantum rods with $L = 20$nm and $\Delta_c = 25$meV at $T = 300$K as functions of R; (c) Linear polarization factors of optical transitions of CdSe quantum rods with $e = 2$ and $\Delta_c = 25$meV as functions of R; (d) Normalized intensities of σ_x and σ_z transitions of CdSe quantum rods with $e = 2$ and $\Delta_c = 25$meV at $T = 300$K as functions of R

3.2 Magnetic field effect

The linear polarization factors of optical transitions of CdSe quantum spheres with R = 2nm and Δ_c = 25meV as functions of b are shown in Fig.3(a), and fitted in Fig.3(b). From Fig.3(a), we see that the negative linear polarization factor decreases as b increases. This is mainly due to the antisymmetric splitting. At higher temperatures, the negative linear polarization factors are bigger, and decrease more slowly with increasing b. We can use a simple model that takes into account only the antisymmetric splitting to explain this phenomenon. In the simple model, as b increases, the energy of the $|1,1\rangle$ state goes down, the energy of the $|1,-1\rangle$ state goes up, and the energy of the $|1,0\rangle$ state does not change. Considering the Boltzmann distribution, the linear polarization factor in the simple model is a function of b, whose form is

$$y = \frac{c - e^{dx} - e^{-dx}}{c + e^{dx} + e^{-dx}} \tag{18}$$

where y and x represent the linear polarization factor and the magnetic field strength b, respectively. We fit the three curves in Fig.3(a) at different temperatures with different parameters c and d in Eq.(18) in Fig.3(b). The parameter d in the Boltzmann factor is proportional to the reciprocal of the temperature. The simple model fits very well. The linear polarization factors of optical transitions of CdSe quantum rods with R = 2nm and e = 2 and Δ_c = 25meV as functions of b are shown in Fig.4(a), and fitted in Fig.4(b). We see that the linear polarization factor decreases as b increases, the same as Fig.3(a). At higher temperatures, the negative linear polarization factors have bigger values at b = 0, and decrease more slowly with increasing b. At T = 100K the linear polarization factor changes from a positive value to a negative value as b increases. We call the magnetic field, under which the linear polarization factor is zero, the crossing magnetic field, as shown in Fig. 4(a). At lower temperatures, the linear polarization factor has bigger value at b = 0, and decreases more quickly as b increases, so the crossing magnetic field may be smaller. The linear polarization factor of optical transitions of CdSe quantum rods with R = 3nm and e = 2 and Δ_c = 25meV at T = 10K as a function of b is shown in Fig.5. We see that the linear polarization factor changes from a positive value to a negative value around B = 17.55T as the magnetic field increases.

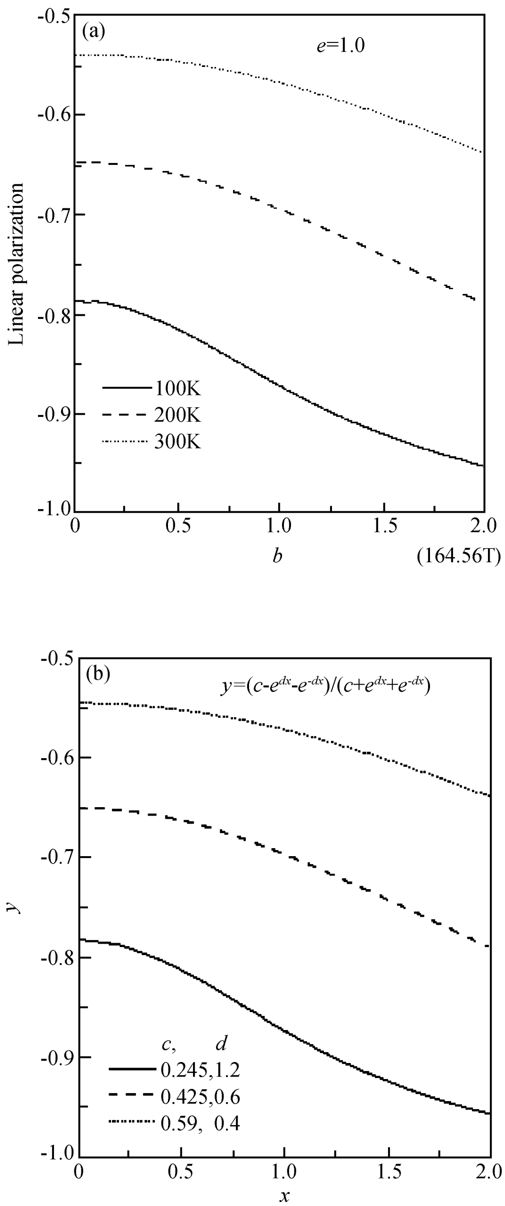

Fig. 3 (a) Linear polarization factors of optical transitions of CdSe quantum spheres with R = 2nm and Δ_c = 25meV as functions of b; (b) Function $y = \frac{c - e^{dx} - e^{-dx}}{c + e^{dx} + e^{-dx}}$

4 Conclusion

The linear-polarization optical property of CdSe quantum rods is studied in detail. It is found that CdSe quantum spheres have negative polarization factors (xy-polarized emission). The CdSe

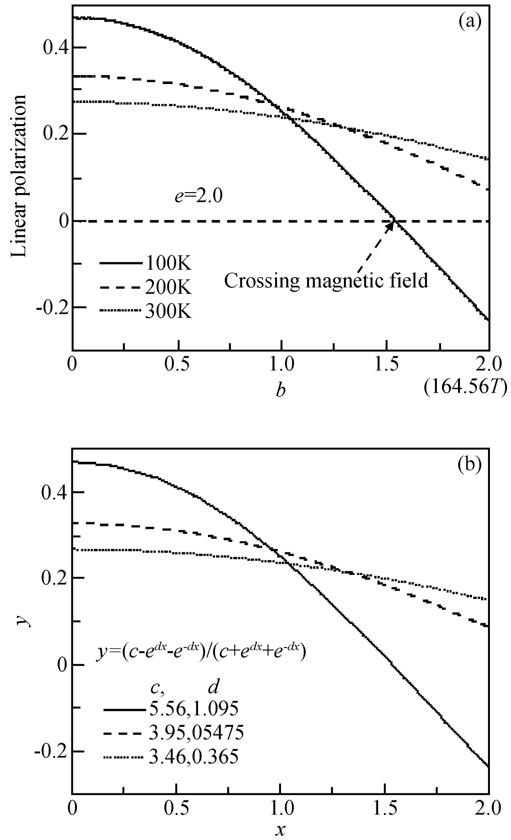

Fig. 4 (a) Linear polarization factors of optical transitions of CdSe quantum rods with $R = 2\text{nm}$ and $e = 2$ and $\Delta_c = 25\text{meV}$ as functions of b; (b) Function $y = \frac{c - e^{dx} - e^{-dx}}{c + e^{dx} + e^{-dx}}$

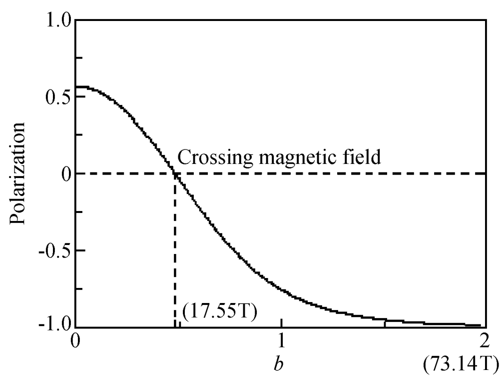

Fig. 5 Linear polarization factor of optical transitions of CdSe quantum rods with $R = 3\text{nm}$ and $e = 2$ and $\Delta_c = 25\text{meV}$ at $T = 10\text{K}$ as a function of b

quantum long rods with small radius have positive linear polarization factors (z-polarized emission). The CdSe quantum long rods with large radius have negative linear polarization factors. Under a magnetic field applied along the z axis of the crystal structure, the linear polarization factors decrease. That is to say, the z-polarized emission decreases relative to xy-polarized emission as the magnetic field increases in the sphere and rod cases. The linear polarization factor of CdSe quantum rods with small radius may change from a positive value to a negative value as the magnetic field increases.

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CdSe 量子棒的线偏振光学性质*

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摘要： 用有效质量包络函数理论研究了 CdSe 量子棒的线偏振光学性质，考虑了形状和磁场的影响. 发现 CdSe 量子球具有负的线偏振因子（*xy*-平面内的线偏振发射），而小半径长量子棒具有正的线偏振因子（*z*-方向的线偏振发射）. *z*-方向就是晶格 *c*-轴方向. 因六角晶格对称性和晶格场劈裂能的影响，大半径长量子棒具有负的线偏振因子. 线偏振因子随着 *z*-方向磁场的增加而减小，可能由正变负，即 *z*-方向的线偏振发射相对 *xy*-平面内的线偏振发射减小了.

关键词： 量子棒；线偏振；CdSe

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