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Theory of donor electronic Raman scattering in wurtzite-type semiconductors

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A theory is presented for the electronic Raman scattering on donor levels in semiconductors with the wurtzite structure. The analytical expressions of the scattering cross section with different photon polarization configurations are derived in the second order of the perturbation theory. A comparison with the experiment is done for CdS.

I. INTRODUCTION

Since the pioneering works of Elliott and Loudon[1] and Wolff,[2] the electronic Raman scattering from donor and acceptor levels in semiconductors has been investigated both experimentally and theoretically by many authors.[3–14] Recently Yu carried out an experiment on the donor electronic Raman scattering in CdS for linearly polarized incident and scattered lights.[14] He studied the energy dependence of the cross section in an interval of the values of the incident photon energy below the band gap, and observed the enhancement of the cross sections with the increase of the photon energy. To explain his experimental results theoretically Yu has made numerical evaluations of the cross sections, using some rather simple approximate formulas for the scattering amplitudes. The agreement between the experimental data and the theoretically calculated values is fine.

The present work is devoted to the theoretical investigation of the electronic Raman scattering on the donor levels in the semiconductors with the wurtzite structure. Both incident and scattered photons are assumed to be linearly or circularly polarized. The real symmetry properties of the crystal and their implications for the energy bands will be taken into account. We use the unit system with $\hbar=c=1$. The results obtained for CdS are compared with the experimental data and the crude estimations in Ref. 14.

II. CRYSTAL ANISTROPY AND ENERGY BANDS

We are considering the highly anisotropic crystal with the symmetry group C_{6v} and with the energy bands represented in Fig. 1. There are three doubly degenerate valence bands specified by an index $\nu=A$, B, C and a doubly degenerate conduction band. The longitudinal and transverse effective masses of the holes in the corresponding energy band (with respect to the crystal axis) are denoted by $m_\mu^{\parallel}$ and $m_\mu^{\perp}$, and of the electron in the conduction band by $m_e^{\parallel}$ and $m_e^{\perp}$, respectively. The values of the static dielectric constant for the light propagating parallel or perpendicular to the crystal axis are also denoted by $\epsilon_0^{\parallel}$ or $\epsilon_0^{\perp}$, respectively.

At the center of the Brillouin zone the Bloch wave functions of the electrons in the conduction band transform according to the representation Γ_7 of the symmetry group C_{6v} while those for the holes in the valence bands $\nu=A,B,C$ are the basis vectors of the representations $\Gamma_9,\Gamma_7,\Gamma_7$, respectively, of this group. To specify the spin projections of the electrons in the conduction band and the holes in the valence bands we use the notations $\sigma=\uparrow,\downarrow$ and, and $\sigma_\nu=\pm$; $\nu=A,B,C$, respectively.

The Bloch wave functions of the free electrons in the conduction band at the center of the Bril-

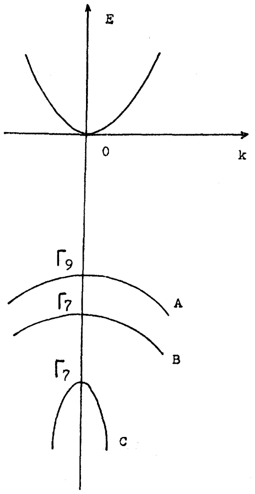

FIG. 1. Energy-band structure of wurtzite-type semiconductors.

louin zone are those of the s type and denoted by $|S\uparrow\rangle$ and $|S\downarrow\rangle$, while the state vectors of the free holes in the band $v=A$, B, C are the linear combinations of the Kane[15] p-type wave functions

$$\left|\frac{X\pm iY}{\sqrt{2}}\uparrow\right\rangle,\ \left|\frac{X\pm iY}{\sqrt{2}}\downarrow\right\rangle,\ |Z\uparrow\rangle,\ |Z\downarrow\rangle\ .$$

In the framework of the $\vec{k}\cdot\vec{p}$ perturbation theory for the free holes with momentum $\vec{k}$ we have the state vectors[16,17]

$$\begin{aligned}
|\vec{k}A+\rangle &= \left|\frac{X+iY}{\sqrt{2}}\uparrow\right\rangle ,\\
|\vec{k}A-\rangle &= -\left|\frac{X-iY}{\sqrt{2}}\downarrow\right\rangle ,\\
|\vec{k}B+\rangle &= \tau\left|\frac{X-iY}{\sqrt{2}}\uparrow\right\rangle+\rho|Z\downarrow\rangle ,\\
|\vec{k}B-\rangle &= -\tau\left|\frac{X+iY}{\sqrt{2}}\downarrow\right\rangle-\rho|Z\uparrow\rangle ,\\
|\vec{k}C+\rangle &= -\rho\left|\frac{X-iY}{\sqrt{2}}\uparrow\right\rangle+\tau|Z\downarrow\rangle ,\\
|\vec{k}C-\rangle &= \rho\left|\frac{X+iY}{\sqrt{2}}\downarrow\right\rangle+\tau|Z\uparrow\rangle ,
\end{aligned} \tag{1}$$

where the coefficients τ, ρ are the functions of the longitudinal $k_{\parallel}$ and transverse $k_{\perp}$ components of the momentum vector $\vec{k}$. These functions depend on the physical parameters of the energy bands and can be written in the following manner:

$$\begin{aligned}
\tau &= (1-q^2)^{1/2}\left[1+\frac{\psi}{\Delta}\right]^{1/2} ,\\
\rho &= \left[q^2-(1-q^2)\frac{\psi}{\Delta}\right]^{1/2} ,
\end{aligned} \tag{2}$$

$$\begin{aligned}
q^2 &= \frac{1}{2}\left[1+\left[1-\frac{8}{9}\frac{\lambda^2}{\Delta^2}\right]^{1/2}\right] ,\\
\psi &= \left[\frac{1}{m_B^{\perp}}-\frac{1}{m_A^{\perp}}\right]k_{\perp}^2+\left[\frac{1}{m_B^{\parallel}}-\frac{1}{m_A^{\perp}}\right]k_{\parallel}^2 ,
\end{aligned} \tag{3}$$

Δ being the difference of the values of the energies at the tops of the valence bands B and C, λ being the matrix element of the spin-orbit coupling.

The electrons in the initial and final states of the Raman scattering process are those in a hydrogenlike system. The state vectors of these electrons are the products of the state vectors $|S\uparrow\rangle$ or $|S\downarrow\rangle$ and the envelope functions $\varphi_n(x,y,z)$ which are the wave functions of an electron with an anisotropic effective mass in a Coulomb field. These envelope functions are the solutions of the Schrödinger equation:

$$\left[-\frac{1}{2m_e^{\perp}}\left[\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial y^2}\right]-\frac{1}{2m_e^{\parallel}}\frac{\partial^2}{\partial z^2}-\frac{e^2}{(\epsilon_0^{\parallel}\epsilon_0^{\perp})^{1/2}[x^2+y^2+(\epsilon_0^{\perp}/\epsilon_0^{\parallel})z^2]^{1/2}}\right]\varphi_n(x,y,z)=E_n\varphi_n(x,y,z)\ . \tag{4}$$

Here we chose the z axis to be the symmetry axis $\vec{C}$ of the crystal. Setting

$$\left[\frac{\epsilon_0^{\perp}}{\epsilon_0^{\parallel}}\right]^{1/2} z=z_1\ ,$$

$$\frac{1}{m_e^{\parallel}}\frac{\partial^2}{\partial z^2}=\frac{1}{m_e^{\perp}}\frac{\partial^2}{\partial z_1^2}+\left[\frac{1}{m_e^{\parallel}}\frac{\epsilon_0^{\perp}}{\epsilon_0^{\parallel}}-\frac{1}{m_e^{\perp}}\right]\frac{\partial^2}{\partial z_1^2}\ ,$$

and considering the last term in the right-hand side (rhs) of the last relation as a perturbation, we can find the approximate wave functions $\varphi_n(x,y,z)$. In the lowest approximation they are obtained from those for a hydrogenlike system after the substitution

$$z\rightarrow\left[\frac{\epsilon_0^{\parallel}}{\epsilon_0^{\perp}}\right]^{1/2} z\ .$$

III. SCATTERING CROSS SECTION

In this section we shall derive the analytical expressions for the scattering cross sections with both linearly and circularly polarized incident and scattered photons. Denote by $|i\rangle$ and $|f\rangle$ the state vectors of the donor electrons in the initial and final states of the scattering process. We start from the following general formula for the cross section in the second-order approximation:

$$\frac{d\sigma}{d\Omega}=2\pi\left[\frac{e^2}{m_0}\right]^2\frac{\omega_s}{\omega_i}\frac{1}{\epsilon_0^i\epsilon_0^s}\sum_{if}\left|\frac{1}{m_0}\sum_{\nu\sigma_\nu}\int\frac{d^3k}{(2\pi)^3}\left[\frac{\langle f\,|\,\vec{\xi}_s^{\,*}\vec{\nabla}\,|\,\vec{\mathrm{k}}\nu\sigma_\nu\rangle\langle\vec{\mathrm{k}}\nu\sigma_\nu\,|\,\vec{\xi}_i\vec{\nabla}\,|\,i\rangle}{E_g+E_i-E_\nu(\vec{\mathrm{k}})+\omega_i}+\frac{\langle f\,|\,\vec{\xi}_i\vec{\nabla}\,|\,\vec{\mathrm{k}}\nu\sigma_\nu\rangle\langle\vec{\mathrm{k}}\nu\sigma_\nu\,|\,\vec{\xi}_s^{\,*}\vec{\nabla}\,|\,i\rangle}{E_g+E_f+E_\nu(\vec{\mathrm{k}})-\omega_i}\right]\right|^2 . \tag{5}$$

Here e and m_0 are the charge and the mass of the free electron, ω_i and ω_s are the energies of the incident and scattered photons, $\vec{\xi}_i$ and $\vec{\xi}_s$ are the unit vectors characterizing their polarizations, E_g denotes the band gap, $E_\nu(\vec{\mathrm{k}})$ denotes the energy of the hole with momentum $\vec{\mathrm{k}}$ in the band ν:

$$E_\nu(\vec{\mathrm{k}})=\Delta_\nu+\frac{k_{\parallel}^2}{2m_\nu^{\parallel}}+\frac{k_{\perp}^2}{2m_\nu^{\perp}} , \tag{6}$$

Δ_ν being the absolute value of the difference of the energies at the top of the bands ν and A, E_i, and E_f are the binding energies of the initial and final donor electrons; ϵ_0^i and ϵ_0^s are the values of the dielectric constant for the incident and scattered lights. We consider the scattering process with the transition of the donor electron from the ground state to the first-excited one. In the lowest-order approximation with the isotropic mass they are the 1s and 2s states. In this case the symbol $\sum_{\text{if}}$ denotes the sum over the spin projections of the final electrons and the average over those of the third initial ones, and we have

$$E_i=-I,\;\; E_f=-\frac{I}{4} , \tag{7}$$

where I is the binding energy of the donor electron

$$I=\frac{e^4m_e^{\perp}}{2}\frac{1}{\epsilon_0^{\perp}\epsilon_0^{\parallel}}\left[1+\frac{\delta}{3}-\frac{8\delta^2}{45}+\cdots\right] ,$$
$$\delta=1-\frac{\epsilon_0^{\perp}}{\epsilon_0^{\parallel}}\frac{m_e^{\perp}}{m_e^{\parallel}} . \tag{8}$$

From the general formula (5) we can derive the analytical expressions of the scattering cross sections for any polarization states of the incident and scattered photons. We obtain the following results:

(A) For linearly polarized photons, at the directions of the incident and scattered light compatible with the given polarization configurations.

(a) $\vec{\xi}_i\,||\,\vec{\xi}_s\perp\vec{\mathrm{C}}$:

$$\frac{d\sigma}{d\Omega}=\tfrac{1}{2}\sigma_0\,|\,A_1^{11}(\omega_i)+A_2^{11}(\omega_i)+B_1^{11}(\omega_i)+B_2^{11}(\omega_i)+C_1^{11}(\omega_i)+C_2^{11}(\omega_i)\,|^2 . \tag{9}$$

(b) $\vec{\xi}_i\,||\,\vec{\xi}_s\,||\,\vec{\mathrm{C}}$:

$$\frac{d\sigma}{d\Omega}=2\sigma_0\,|\,B_1^{22}(\omega_i)+B_2^{22}(\omega_i)+C_1^{22}(\omega_i)+C_2^{22}(\omega_i)\,|^2 . \tag{10}$$

(c) $\vec{\xi}_s\perp\vec{\xi}_i\,||\,\vec{\mathrm{C}}$:

$$\frac{d\sigma}{d\Omega}=\sigma_0\,|\,B_1^{12}(\omega_i)-B_2^{12}(\omega_i)-C_1^{12}(\omega_i)+C_2^{12}(\omega_i)\,|^2 . \tag{11}$$

(B) For circularly polarized incident and scattered photons, with the incident light propagating in the direction of the z axis and with the scattering angle θ.

(a) incident and scattered σ^+ photons:

$$\frac{d\sigma}{d\Omega}=\sigma_0[(1+\cos\theta)^2L(\omega_i)+2\sin^2\theta M(\omega_i)+(1-\cos\theta)^2N(\omega_i)]\ . \qquad (12)$$

(b) incident σ^+ and scattered σ^- photons:

$$\frac{d\sigma}{d\Omega}=\sigma_0[(1-\cos\theta)^2L(\omega_i)+2\sin^2\theta M(\omega_i)+(1+\cos\theta)^2N(\omega_i)]\ . \qquad (13)$$

In the above formulas we used the following notations:

$$\sigma_0=\left[\frac{e^2}{m_0}\right]^2\frac{\omega_s}{\omega_i}\left[\frac{\Pi}{\epsilon_0^{\parallel}}\right]^2\frac{2^7a^4}{\pi}\ , \qquad (14)$$

$$\Pi=\frac{2}{m_0}\,|\langle X|\nabla_x|S\rangle|^2\ , \qquad (15)$$

$$a=\frac{(\epsilon_0^{\parallel}\epsilon_0^{\perp})^{1/2}}{m_e^{\perp}e^2}\left[1-\frac{\delta}{6}\right]\ , \qquad (16)$$

$$\begin{aligned}
L(\omega_i)&=|A_1^{11}(\omega_i)+B_2^{11}(\omega_i)+C_2^{11}(\omega_i)|^2+|A_2^{11}(\omega_i)+B_1^{11}(\omega_i)+C_1^{11}(\omega_i)|^2\ ,\\
M(\omega_i)&=|A_1^{11}(\omega_i)+A_2^{11}(\omega_i)|^2+|B_1^{11}(\omega_i)+B_2^{11}(\omega_i)+C_1^{11}(\omega_i)+C_2^{11}(\omega_i)|^2\ ,\\
N(\omega_i)&=|A_1^{12}(\omega_i)-B_1^{12}(\omega_i)|^2+|A_2^{12}(\omega_i)-B_2^{12}(\omega_i)|^2\ ,
\end{aligned} \qquad (17)$$

$$\begin{aligned}
&A_i^{rs}(\omega_i)=\Phi_A^{rs}(\omega_i)\ ,\quad A_2^{rs}(\omega_i)=\Phi_A^{rs}(\tfrac{3}{4}I-\omega_i)\ ,\\
&B_1^{rs}(\omega_i)=\Phi_B^{rs}(\omega_i)\ ,\quad B_2^{rs}(\omega_i)=\Phi_B^{rs}(\tfrac{3}{4}I-\omega_i)\ ,\\
&C_1^{rs}(\omega_i)=\Phi_C^{rs}(\omega_i)\ ,\quad C_2^{rs}(\omega_i)=\Phi_C^{rs}(\tfrac{3}{4}I-\omega_i)\ ,
\end{aligned} \qquad (18)$$

where $r,s=1,2,$

$$\Phi_v^{rs}(\omega)=\int_0^{\infty}x^2dx\int_0^1dy\frac{4\alpha x^2y^2+4x^2-1}{(4\alpha x^2y^2+4x^2+1)^3(\alpha x^2y^2+x^2+1)}\,\frac{F_v^r(x,y)F_v^s(x,y)}{m_0a^2(E_g+\Delta_v+\omega-I)+\beta_vx^2y^2+\gamma_vx^2}\ , \qquad (19)$$

and

$$\begin{aligned}
&F_A^1(x,y)=F_A^2(x,y)=1\ .\\
&F_B^1(x,y)=F_C^2(x,y)=(1-q^2)^{1/2}(1+\eta x^2y^2-\mu x^2)^{1/2}\ ,\\
&F_B^2(x,y)=F_C^1(x,y)=[q^2+(q^2-1)(\eta_x^2y^2-\mu x^2)]^{1/2}\ ,
\end{aligned} \qquad (20)$$

$$\begin{aligned}
\alpha&=\left[\frac{\epsilon_0^{\perp}}{\epsilon_0^{\parallel}}\right]-1\ ,\\
\beta_v&=\frac{m_0}{2}\left[\frac{1}{m_v^{\parallel}}-\frac{1}{m_v^{\perp}}\right]\ ,\\
\gamma_v&=\frac{m_0}{2}\frac{1}{m_v^{\perp}}\ ,\\
\eta&=\frac{m_0^2a^2}{\Delta_C-\Delta_B}\left[\frac{1}{m_B^{\parallel}}+\frac{1}{m_A^{\perp}}-\frac{1}{m_A^{\parallel}}-\frac{1}{m_B^{\perp}}\right]\ ,\\
\mu&=\frac{m_0^2a^2}{\Delta_C-\Delta_B}\left[\frac{1}{m_A^{\perp}}-\frac{1}{m_B^{\perp}}\right]\ .
\end{aligned} \qquad (21)$$

These formulas are valid for many semiconductors with the wurtzite structure: CdS, CdSe, ZnO.

IV. APPLICATION TO CdS

To obtain the numerical values of the cross sections we consider an example, CdS. For this material we have the following values of the physical parameters of the energy bands[16,17]:

$E_g=2.578$ eV ,

$\Delta_B=15$ meV, $\Delta_C=78$ meV ,

$m_e^{\perp}=0.209m_0$, $m_e^{\parallel}=0.198m_0$,

$m_A^{\perp}=0.69m_0$, $m_A^{\parallel}=5m_0$,

$m_B^{\perp}=m_0$, $m_B^{\parallel}=0.52m_0$,

$m_C^{\perp}=1.25m_0$, $m_C^{\parallel}=0.4m_0$,

$\epsilon_0^{\perp}=9.02$, $\epsilon_0^{\parallel}=9.53$.

The matrix element $\langle X \,|\, \nabla_x \,|\, S \rangle$ in the rhs of Eq. (15) can also be determined from the effective masses of the free charge carriers. From the result of Ref. 18 we have

$$\Pi = 21\text{eV} .$$

Using the above values of the parameters and the analytical expressions derived in the preceding section we can calculate the cross sections. Their values for the three cases (a)—(c) of the processes with linearly polarized photons are plotted in Fig. 2. The energy dependence of these cross sections were investigated experimentally by Yu.[14] There is good agreement between our results and the experimental data of Ref. 14 when the incident photon energy varies in an interval below 20 400 cm^{-1}. At higher energies the polariton effect becomes important and must be taken into account. For the ratios of the cross sections at the incident photon energy $\omega_i = 20\,200\ \text{cm}^{-1}$ we derived the following values:

$$\frac{d\sigma_a^{\text{theor}}}{d\Omega} : \frac{d\sigma_b^{\text{theor}}}{d\Omega} : \frac{d\sigma_c^{\text{theor}}}{d\Omega} = 4.1{:}1{:}0.73 ,$$

while the experimental data equal

$$\frac{d\sigma_a^{\text{expt}}}{d\Omega} : \frac{d\sigma_b^{\text{expt}}}{d\Omega} : \frac{d\sigma_c^{\text{expt}}}{d\Omega} \sim 4.2{:}1{:}1 .$$

Remember also that the crude estimations by Yu in Ref. 14 give nearly the same energy dependence of the cross sections and the values 4:1:0.67 of the above ratios.

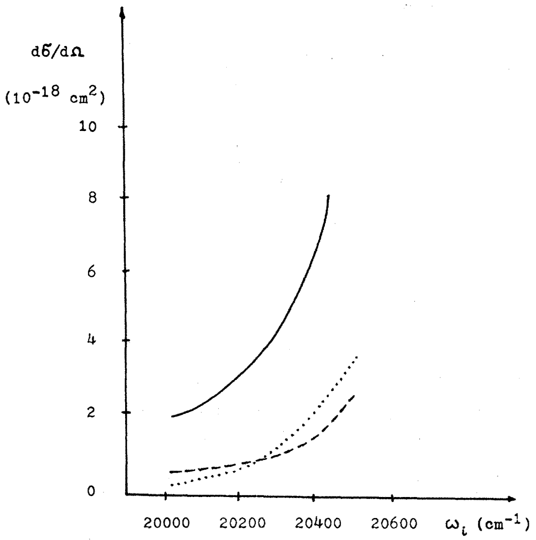

FIG. 2. Dependences of the cross sections on incident photon energy for three different scattering polarization configurations: $\vec{\xi}_i \| \vec{\xi}_s \perp \vec{C}$ (solid curve), $\vec{\xi}_i \| \vec{\xi}_s \| \vec{C}$ (dashed curve), and $\vec{\xi}_s \perp \vec{\xi}_i \| \vec{C}$ (dotted curve).

To establish the relation between the analytical expressions of the cross sections derived in Sec. III and the approximate expressions 5(a)—5(c) in Ref. 14 we neglect the small contributions from the functions $A_1^{rs}(\omega_i)$, $B_1^{rs}(\omega_i)$, $C_1^{rs}(\omega_i)$ in comparison with $A_2^{rs}(\omega_i)$, $B_2^{rs}(\omega_i)$, $C_2^{rs}(\omega_i)$, and suppose that the effective masses are isotropic. In this case from the results in Sec. III we can derive the following formulas.

(a) $\vec{\xi}_i \| \vec{\xi}_s \perp \vec{C}$:

$$\frac{d\sigma}{d\Omega} = \frac{\sigma_0}{\Pi^2} \left| P_{A\perp}^2 \int_0^\infty \frac{F(x)}{H_A(x)} dx + P_{B\perp}^2 \int_0^\infty \frac{F(x)(1-\mu x^2)}{H_B(x)} dx + P_{C\perp}^2 \int_0^\infty \frac{F(x)[1+\mu x^2(1/q^2-1)]}{H_C(x)} dx \right|^2 . \tag{22}$$

(b) $\vec{\xi}_i \| \vec{\xi}_s \| \vec{C}$:

$$\frac{d\sigma}{d\Omega} = \frac{\sigma_0}{\Pi^2} \left| P_{B\|}^2 \int_0^\infty \frac{F(x)[1+\mu x^2(1/q^2-1)]}{H_B(x)} dx + P_{C\|}^2 \int_0^\infty \frac{F(x)(1-\mu x^2)}{H_C(x)} dx \right|^2 . \tag{23}$$

(c) $\vec{\xi}_s \perp \vec{\xi}_i \| \vec{C}$:

$$\frac{d\sigma}{d\Omega} = \frac{\sigma_0}{\Pi^2} \left| P_{B\perp} P_{B\|} \int_0^\infty \frac{F(x)[1-\mu(1-1/2q^2)x^2]}{H_B(x)} dx - P_{C\perp} P_{C\|} \int_0^\infty \frac{F(x)[1-\mu(1-1/2q^2)x^2]}{H_C(x)} dx \right|^2 . \tag{24}$$

In these expressions

$$F(x)=\frac{x^2(4x^2-1)}{(4x^2+1)^3(x^2+1)^2}\,, \tag{25}$$

$$H_v(x)=m_0a^2\left[\omega_v-\omega_i-\frac{I}{4}\right]+\frac{m_0}{2m_v}x^2\,, \tag{26}$$

$$\omega_v=E_g+\Delta_v\,.$$

$P_{v\perp}$ or $P_{v\parallel}$ are the matrix elements of the electrical dipole transitions between the conduction band and the corresponding valence band $v=A,B,C$ for the light polarized parallel or perpendicular to the symmetry axis $\vec{C}$. Formulas (22)–(24) are similar to (but more rigorous than) relations 5(a)–5(c) in Ref. 14.

The results obtained in this paper show that the crude estimations of the energy dependence and the ratios of the cross sections given in Ref. 14 are nearly equal to the values calculated rigorously in the second order of the perturbation theory at the energy interval where the perturbation theory is valid.

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