Optical properties of anisotropic exciton: Hyperspherical theory

E. A. Muljarov,* A. L. Yablonskii, and S. G. Tikhodeev
General Physics Institute, RAS, Moscow 117942, Vavilov Street, 38, Russia

A. E. Bulatov and Joseph L. Birman
Physics Department, City College of New York, New York, New York 10031
(Received 27 February 1998; revised manuscript received 1 October 1998)

A perturbation Brillouin-Wigner approach to anisotropic exciton problem, based on a hyperspherical formalism, is developed. The binding energies and oscillator strengths of elongated as well as flattened excitons are calculated numerically. It is shown that with an increase of the anisotropy degree the oscillator strengths are markedly redistributing between optically active and formerly inactive states, making the latter optically active. [S0163-1829(99)03307-X]

The progress in the physics of semiconductor heterostructures has revived the interest to the anisotropic exciton problem.[1,2] In bulk semiconductors, the anisotropy of carrier effective masses and/or of dielectric susceptibilities is induced by the band and/or polarization anisotropy of the crystal. In layered semiconductors such as $A^{II}B^{VI}$, $A^{II}B^{VII}$, and $A^{IV}B^{VII}$ compounds the effective-mass anisotropy is due to localization of carriers inside layers and tunneling between layers. This effect becomes very important in artificial layered structures such as semiconductor superlattices (SL's), where the miniband formation is responsible for a strong mass anisotropy.[3] The dielectric constant becomes anisotropic too, if the SL's constituent layers have different dielectric susceptibilities. Thus, the exciton in the SL's becomes significantly anisotropic. Recently, the formalism of the anisotropic exciton in a variational form was used in the theory of excitons in short-period SL's (see, e.g., Refs. 4 and 5).

In spite of a long history of theoretical study,[1,2,6–16] the investigation of the optical properties of the anisotropic exciton is not complete. For example, the behavior of exciton oscillator strengths is very important for understanding the experimental absorption spectra. However, the evolution of the oscillator strengths of the anisotropic exciton with the increase of the anisotropy has not been investigated, to our knowledge, with two exceptions: calculations for slightly anisotropic exciton[13] and simulations of optical spectra within an isotropic exciton model.[17] We would like to stress, however, that in both approaches[13,17] it is not possible to describe the reported below drastic changes of the oscillator strengths (due to the level anticrossings[10]) with the increase of the anisotropy.

In this paper the energy spectrum, wave functions, and oscillator strengths of the uniaxial anisotropic exciton are calculated numerically for flattened as well as elongated excitons. We develop a perturbation approach based on the stereographic projection of the momentum space to the unit four-dimensional (4D) sphere, proposed by Fock.[18] We utilize the additional hidden symmetry of the Coulomb potential for an expansion of the anisotropic exciton wave function over a complete basis of hyperspherical harmonics. This expansion depends explicitly on the exciton energy through scaling parameters that follow adiabatically the changes in anisotropy. As a result, the hyperspherical functions turn out to be the most effective basis for numerical calculations.

In addition, recently an elegant model of fractional-dimensional space has been developed (see Refs. 19 and 20, and references therein), allowing us to treat self-consistently the bound as well as continuum states in hydrogen problem of noninteger dimension. However, its direct applicability to the anisotropic exciton problem is troublesome. For example, the fractional-dimensional hydrogen problem conserves the Coulomb degeneracy of levels (so that the binding energies depend on the principal quantum number only), whereas in reality the anisotropy lifts this degeneracy and restores it only in exactly 2D and 3D cases.

Let us describe briefly the perturbation method based on a hyperspherical formalism.[21] First, we transform the Hamiltonian of the uniaxial anisotropic exciton,

$$\hat{H} = -\frac{\hbar^2}{2\mu_\perp}\left(\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial y^2}\right) - \frac{\hbar^2}{2\mu_\parallel}\frac{\partial^2}{\partial z^2} - \frac{e^2}{\sqrt{\varepsilon_\parallel\varepsilon_\perp(x^2+y^2)+\varepsilon_\perp^2 z^2}}, \tag{1}$$

to a dimensionless effective Hamiltonian with isotropic potential energy, introducing the effective Rydberg $\mathrm{Ry}^* = \mu_\perp e^4/2\varepsilon_0^2\hbar^2$ and Bohr radius $a_B^* = \hbar^2\varepsilon_0/\mu_\perp e^2$, where $\varepsilon_0 = \sqrt{\varepsilon_\perp\varepsilon_\parallel}$, and using a dilatation $z \to z\sqrt{\varepsilon_\parallel/\varepsilon_\perp}$. Here μ is the reduced exciton mass, ε is the semiconductor dielectric constant, and subscripts $\parallel$ and $\perp$ refer, respectively, to the quantities along and normal to the axis of symmetry OZ. Next, we calculate the Fourier transform and following Fock[18] perform a stereographic projection of 3D momentum space to the 4D unit sphere $\mathbf{p}/p_\nu \to \vec{u}$ where the 4D vector $\vec{u}$ on the sphere is defined as

$$\vec{u} = \{\mathbf{u}, u_n\} = \left\{\frac{2p_\nu \mathbf{p}}{p^2+p_\nu^2}, \frac{p^2-p_\nu^2}{p^2+p_\nu^2}\right\}, \tag{2}$$

$p = |\mathbf{p}|$, and p_ν is the transformation parameter. If we assume that the transformation Eq. (2) depends on the energy E_ν

<0 of the anisotropic exciton bound state ν via $p_\nu = \sqrt{-E_\nu}$, then, after the wave-function renormalization, the anisotropic exciton Schrödinger equation takes the form

$$\left[1+\frac{\gamma-1}{2}\frac{u_z^2}{1-u_n}\right]\Psi_\nu(\vec{u})=\frac{1}{p_\nu}\frac{1}{2\pi^2}\int\frac{\Psi_\nu(\vec{u}')}{|\vec{u}-\vec{u}'|^2}d^4\Omega', \tag{3}$$

where the integral on the right-hand side is carried out over the unit sphere in 4D space. Here $\gamma=\varepsilon_\perp\mu_\perp/\varepsilon_\parallel\mu_\parallel$ is the anisotropy parameter ($0<\gamma<1$ and $1<\gamma<\infty$ for, respectively, flattened and elongated exciton). For the isotropic exciton $\gamma=1$ and the hyperspherical functions $\Psi^{(0)}_{nlm}(\vec{u})$ become the solutions[18] of Eq. (3) with $p_\nu=1/n$, where $n=1,2,\ldots$ is the principal quantum number ($l=0,1,\ldots,n-1$ and $m=-l, -l+1,\ldots,l$ are, respectively, the orbital and magnetic quantum numbers). The hyperspherical functions afford the irreducible representation of the full symmetry group O(4) of the hydrogenlike system.[22] These functions constitute a convenient complete basis for applying Brillouin-Wigner perturbation method to the bound states[23] of the anisotropic exciton for any $\gamma>0$. Expanding the perturbed wave function Ψ_ν over the hyperspherical harmonics, Eq. (3) takes the matrix form

$$\sum_{s'}\left[n\delta_{ss'}+\frac{\gamma-1}{2}V_{ss'}\right]\mathcal{C}^\nu_{s'}=\lambda_\nu\mathcal{C}^\nu_s, \tag{4}$$

with ''Fock eigenvalues''

$$\lambda_\nu=\frac{1}{p_\nu}=\frac{1}{\sqrt{-E_\nu}} \tag{5}$$

and the perturbation matrix

$$V_{ss'}=\sqrt{nn'}\int\Psi^{(0)*}_s(\alpha,\theta,\varphi)(1+\cos\alpha)\cos^2\theta\Psi^{(0)}_{s'}\times(\alpha,\theta,\varphi)d^4\Omega, \tag{6}$$

where $s=(n,l,m)$ and (α,θ,φ) are the hyperspherical coordinates.[18]

Note that in spite of the energy-dependent transformation Eq. (2), the effective Hamiltonian matrix of Eq. (4) is energy independent, thus allowing its direct diagonalization. The unperturbed spectrum in the Fock representation $\lambda^{(0)}_{nlm}=n$ is equidistant with respect to the hydrogen principal quantum number and does not have a series limit. In the coordinate representation $\mathcal{C}^\nu_s$ are the coefficients in the expansion of the wave function

$$\phi_\nu(\mathbf{r})=\frac{p_\nu^{3/2}}{S_\nu}\sum_s n^2\mathcal{C}^\nu_s\phi^{(0)}_s(\mathbf{r}p_\nu n) \tag{7}$$

over the standard hydrogen wave functions $\phi^{(0)}_{nlm}(\mathbf{r})$ (see, e.g., Ref. 24). It follows from Eq. (7) that the wave function of the anisotropic exciton takes the form of an infinite superposition of spherical harmonics with radially-dependent coefficients. The scaling factors p_ν in the wave functions Eq. (7), which are different for different perturbed states and change adiabatically with γ, play the role of adiabatic scaling parameters in the perturbation theory. Due to the symmetry properties of uniaxial anisotropic exciton Hamiltonian and the full hydrogen symmetry of the basis functions, the matrix elements of the perturbation take the algebraic form,[21] following the selection rules

$$V_{ss'}\neq 0, \quad \text{if } m'=m,\ l'=\begin{cases} l, & n'=n,n\pm1\\ l-2, & n'\leqslant n+1\\ l+2, & n'\geqslant n-1,\end{cases} \tag{8}$$

and $V_{ss'}=0$ for all other s,s'. Thus, matrices with even and odd l as well as with different m can be diagonalized independently.

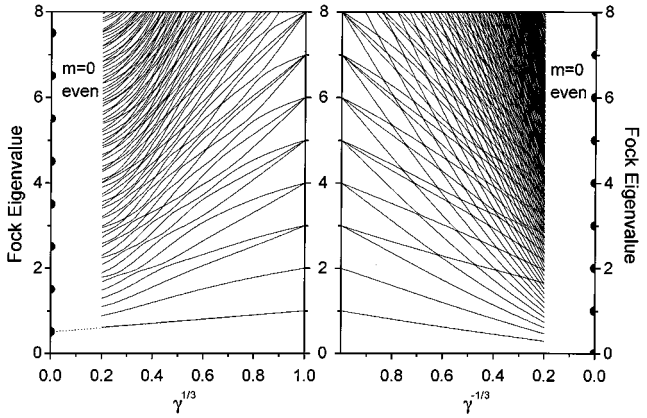

FIG. 1. Fock eigenvalues λ_ν of $m=0$ even-parity states as functions of the anisotropy parameter $\gamma^{1/3}$, $\gamma\leqslant 1$ (left panel), and $\gamma^{-1/2}\lambda_\nu$ as functions of $\gamma^{-1/3}$, $\gamma\geqslant 1$ (right panel). A linear approximation of the ground-state eigenvalue is plotted by dots. Solid curves never intersect each other due to a small anticrossing between the levels. The eigenvalues of purely 2D exciton (left panel) and 1D exciton (right panel) are shown by semicircles.

In numerical calculations the direct diagonalization of a truncated Hamiltonian matrix has been carried out with a relative energy precision of 10^{-4}. For example, to calculate with this precision the ground-state energy for $0.6\leqslant\gamma^{1/3}\leqslant 2$ hydrogen states with the principal quantum number up to 15 and orbital quantum number up to 6 must be taken into account. The numerical procedure becomes unstable for $\gamma\to 0$ and $\gamma\to\infty$. This nonconvergency is caused by the fact that these points, where the symmetry changes (to 2D and 1D, respectively), are peculiar for the perturbation theory. The dimension change causes the levels' degeneration, when a very large (divergent) number of levels are mixed due to perturbation and have to be taken into account.

In Fig. 1 the calculated eigenvalues λ_ν of Eq. (4) [related to the eigenenergies via Eq. (5)] are shown for $m=0$ even-parity states as functions of $\gamma^{1/3}$ for flattened excitons $\gamma\leqslant 1$ (left panel); for elongated excitons $\gamma\geqslant 1$ (right panel) dependences $\gamma^{-1/2}\lambda_\nu$ are shown as functions of $\gamma^{-1/3}$. The multiplier $\gamma^{-1/2}$ in the latter case makes the effective Rydberg finite when $\mu_\perp\to\infty$. Starting at $\gamma=1$ from isotropic case $\lambda_\nu=\lambda^{(0)}_{nml}=n$, all the eigenvalues (with the same m and parity) do not intersect with the anisotropy change (multiple anticrossings occur due to the interaction between states) and at $\gamma\to\infty$ approach the ground-state eigenvalue of 1D exciton[25] $\gamma^{-1/2}\lambda_\nu\to\gamma^{-1/2}\lambda^{1D}_0\to 0$ (right panel). In the opposite case of $\gamma\to 0$ all shown eigenvalues approach the

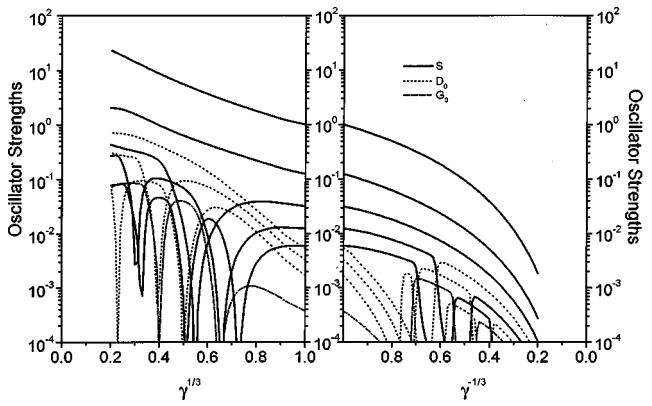

FIG. 2. The anisotropic exciton oscillator strengths of lower S-,D_0-, and G_0-like states as functions of anisotropy parameter $\gamma^{1/3}$ (in units of the ground-state oscillator strength at $\gamma=1$).

ground-state eigenvalue $\lambda_0^{2D}=1/2$ of 2D exciton (left panel). As it can be seen from Fig. 1, the ground-state eigenvalue dependence is almost linear over $\gamma^{1/3}$ for $\gamma \leqslant 1$, and

$$E_0 \approx -\frac{4}{(1+\gamma^{1/3})^2}. \tag{9}$$

Let us now turn to the calculation of the oscillator strengths. In the envelope function approximation the relative oscillator strengths of dipole-allowed transitions f_ν are proportional to $|\phi_\nu(0)|^2$ (see, e.g., Ref. 26). Taking into account that for the unperturbed states $\phi_\nu(0)\neq 0$ only for $l=m=0$, from Eq. (7) we obtain

$$f_\nu \propto |\phi_\nu(0)|^2 = \frac{p_\nu^3}{\pi S_\nu^2}\left|\sum_n \mathcal{C}_{n,0,0}^{\nu}\sqrt{n}\right|^2. \tag{10}$$

Figure 2 shows the numerically calculated oscillator strengths of lower S-,D_0-, and G_0-like states as functions of the anisotropy parameter. We use here the standard hydrogenlike notations that can be done only approximately. Note that when the states are split off due to perturbation, we always label the states with larger oscillator strength at $\gamma \approx 1$ as S state, e.g., the pair of $3S$ and $3D_0$ states—the third and the fourth levels in Fig. 1. Thus, at $\gamma<1$ the $3D_0$ level lies lower than $3S$, contrary to the classification by Faulkner.[10]

It is seen in Fig. 2 that the oscillator strengths of all shown states do not vanish at $\gamma=1$. Originated from the degenerated states of isotropic 3D exciton, the perturbed states become fixed linear combinations of the former even when the perturbation tends to zero. At $\gamma\approx 1$ the S-like state is optically more intensive than the D_0-like state. The picture changes drastically with the increase of anisotropy. Near $\gamma^{1/3}=0.8$ the oscillator strength of the $3D_0$ state overcomes

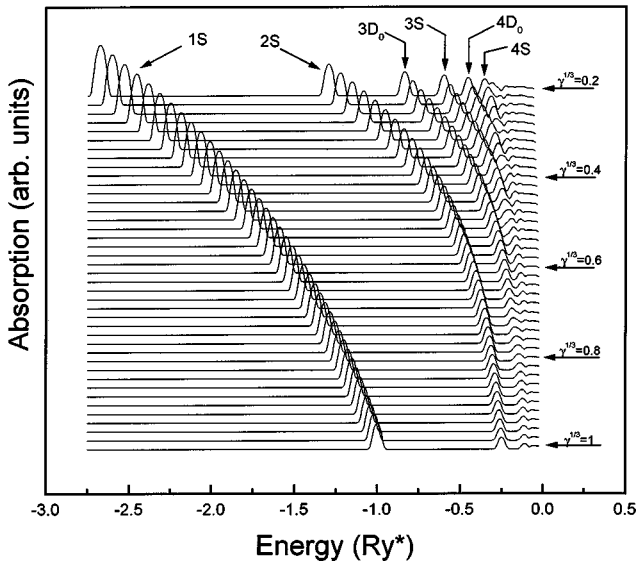

FIG. 3. Anisotropic exciton absorption spectra (calculated with inhomogeneous broadening of 0.02 Ry*) for different values of anisotropy parameter $\gamma \leqslant 1$. The spectral intensities are logarithmically scaled and, being normalized to the intensity in the isotropic case, are measured in arbitrary units.

that of the $3S$ one. For $\gamma^{1/3}<0.8$ the intensity of the $3S$ state collapses and then revives due to the interaction with $4D_0$ state. Moreover, the anisotropy increase leads to substantial growth of the oscillator strengths of higher excited states, such as $4S$ and $4D_0$, making them optically significant. A similar situation takes place if $\gamma>1$ (when a transition from 3D to 1D exciton occurs). Such a redistribution of the oscillator strengths between different states can be clearly seen in the absorption spectra, Fig. 3, calculated for different values of $\gamma^{1/3}$.

To summarize, the perturbation theory of the anisotropic exciton is developed with the help of the Fock transform. The eigenvalues and eigenvectors are found by a numerical diagonalization of the effective Hamiltonian matrix. The energies and oscillator strengths of anisotropic exciton states are calculated for $0<\gamma<1$ (flattened excitons) and $1<\gamma<\infty$ (elongated excitons). It is found that with the increase of the anisotropy a strong redistribution of oscillator strengths between optically active and formerly inactive states occurs: the oscillations in optical intensities of higher excited states take place, and the switching on of formerly weak optical transitions is predicted.

The authors are thankful to Professor R. Zimmermann for helpful discussions. This work was supported by the Russian Basic Research Foundation, Russian Ministry of Science (program ''Nanostructures'') and INTAS. A.E.B. was supported by CUNY.

*Electronic address: muljarov@gpi.ru

[1] C. Kittel and A. Mitchell, Phys. Rev. **96**, 1488 (1954).
[2] W. Kohn and J. M. Luttinger, Phys. Rev. **98**, 915 (1955).
[3] G. Bastard, *Wave Mechanics Applied to Semiconductor Heterostructures* (Les Editions de Physique, Les Ulis, France, 1988), p. 26.
[4] M. F. Pereira, Jr., I. Galbraith, S. W. Koch, and G. Duggan, Phys. Rev. B **42**, 7084 (1990).
[5] Partha Ray and P. K. Basu, Phys. Rev. B **47**, 15 958 (1993).
[6] J. J. Hopfield and D. G. Thomas, Phys. Rev. **122**, 35 (1961).
[7] R. G. Wheeler and J. O. Dimmock, Phys. Rev. **125**, 1805 (1962).
[8] J. A. Deverin, Nuovo Cimento B **63**, 1 (1969).

[9] B. Segal, Phys. Rev. **163**, 769 (1967).
[10] R. A. Faulkner, Phys. Rev. **184**, 713 (1969).
[11] O. Akimoto and H. Hasegawa, J. Phys. Soc. Jpn. **22**, 181 (1967).
[12] E. O. Kane, Phys. Rev. **180**, 852 (1969).
[13] A. Baldereschi and M. G. Diaz, Nuovo Cimento B **68**, 217 (1970).
[14] R. Zimmermann, Phys. Status Solidi B **46**, K111 (1971).
[15] Jian-Bai Xia, Phys. Rev. B **39**, 5386 (1989).
[16] J. Deppe, M. Balcanski, R. F. Wallis, and K. P. Jain, Solid State Commun. **84**, 67 (1992).
[17] M. F. Pereira, Jr., Phys. Rev. B **52**, 1978 (1995).
[18] V. A. Fock, Z. Phys. **98**, 145 (1935).
[19] X. F. He, Phys. Rev. B **42**, 11 751 (1990); **43**, 2063 (1991).
[20] Ch. Tanguy, P. Lefebvre, H. Mathieu, and R. J. Elliot, Phys. Status Solidi A **164**, 159 (1997).
[21] Details of the hyperspherical approach to the anisotropic exciton problem will be published elsewhere.
[22] M. Bander and C. Itzykson, Rev. Mod. Phys. **38**, 330 (1966); **38**, 346 (1966).
[23] The problem of anisotropic exciton scattering states can be approached analogously using hyperspherical harmonics on a two-sheeted 4D hyperboloid as a basis for the perturbation problem.
[24] L. D. Landau and E. M. Lifchitz, *Quantum Mechanics. Nonrelativistic Theory* (Pergamon Press, New York, 1976).
[25] It is well known that in exactly 1D case the ground-state exciton energy is infinite (logarithmically diverges). See, e.g., Ref. 24.
[26] G. Dresselhaus, Phys. Rev. **106**, 76 (1957).