Confined excitons in semiconductors: Correlation between binding energy and spectral absorption shape

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We calculate excitonic absorption spectra in several types of semiconductor nanostructures by using an original formalism involving fractional-dimensional spaces. This compact, analytical formulation predicts a surprising one-to-one mapping between the enhanced exciton Rydberg and the shape of the spectrum, whatever the physical origin of the confinement. By all-numerical calculation of the optical susceptibility we check that different systems with identical exciton Rydbergs exhibit nearly identical spectra, strikingly close to those given by the fractional-dimensional approach.

The concept of "reduced dimensionality" has been regularly encountered in many fields of physics in recent decades. It is mostly connected with the change—or the suppression—of one or several degrees of freedom for a given system. Thus is usually obtained by the reduction of one or several geometrical dimensions of an object, or by introducing anisotropy through some perturbation. In general, the mathematical description of ideal problems in one, two, or three dimensions is well established.

Now, in most cases, the experimental reality would normally correspond to some intermediate dimension, as evidenced by the frequency of locutions such as "quasione-" or "quasi-two-dimensional," for example. In particular, the recent progress in crystal growth permits the fabrication of very small semiconductor structures like quantum wells, superlattices, quantum wires, and dots, with a one-atomic-layer accuracy. This makes it possible to observe the effects of spatial restrictions on the wave functions of electrons, holes, or Wannier excitons-i.e., electron-hole pairs bound by a screened Coulomb interaction. The first of these effects is a dramatic change in the energetic spectrum of quantum states. Consequently, the optical properties of these artificial materials, mostly governed by the physics of excitons, are very different from those of bulk three-dimensional (3D) semiconductors, which they are made of. However, none of the integer-dimensional modelings of transition energies and oscillator strengths perfectly describes the experimental reality.

In recent years, numerical methods of increasing accuracy have been proposed for calculating the optical spectra of such "low-dimensional" systems. So far, the most accurate of these modelings are all-numerical resolutions¹⁻⁵ of the excitonic Schrödinger equation, which is particularly tedious in the case of the so-called scattering (unbound) states. The resulting wave functions are then used to calculate the spectral optical susceptibility of the medium. Such methods are rather complicated from both the theoretical and numerical points of view. Moreover, they have to be properly adapted to each different physical situation, since quantum wells or quantum wires, for instance, cannot be treated in the same way.

Alternatively, the effects of spatial restriction on excitons can be faced by using metric spaces with fractional dimensions:⁶⁻⁹ the quenched exciton is treated exactly as a hydrogenic system with a noninteger dimensionality α . The absorption spectrum is then obtained from a single general equation,⁸⁻⁹ where α appears as a characteristic parameter of the system. To date, the most reliable way to determine α consists in first calculating the binding energy of the squeezed exciton by a variational model; then α is derived from the elementary relationship between the binding energy and the corresponding dimensionality. Indeed,⁶⁻⁸ the binding energy of the α D exciton is given by $E_{1s} = 4\Re / (\alpha - 1)^2$, where \Re is the effective Rydberg (the binding energy) of the 3D exciton. Defining a more mathematical way to determine exactly the value of the dimensionality remains an open problem.

Once α is known, and taking the zero of photon energies at the interband gap of the structure, the whole optical cross-section spectrum is readily obtained, versus the photon energy $\hbar\omega$, by using the exact generalization of Elliott's formula^{8,9} for arbitrary dimensions,

$$\sigma(\hbar\omega) = \sigma_0 \left\{ \sum_{n=1}^{\infty} \frac{4\Re \Gamma(n+\alpha-2)}{(n-1)! \left[n+\frac{\alpha-3}{2}\right]^{\alpha+1}} \delta(\hbar\omega - E_n) + \left|\Gamma\left[\frac{\alpha-1}{2}+i\gamma\right]\right|^2 e^{\pi\gamma} \frac{\gamma^{2-\alpha}}{\pi} U(\hbar\omega) \right\}.$$
(1)

U(x) represents the Heaviside step function, $\Gamma(x)$ is Euler's gamma function, and $\gamma = \sqrt{\mathcal{R}/\hbar\omega}$ is the reduced parameter introduced by Elliott. We shall not discuss here the value of the amplitude factor σ_0 , which has been given in a previous work, and shall concentrate on the spectral term within the curly brackets in Eq. (1). The first part of this term is a series of Dirac distributions,

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bound centered on the energy levels E_n $=\mathcal{R}/[n+(\alpha-3)/2]^2$. The amplitude of these peaks rapidly decreases with increasing n, while they tend to constitute a pseudocontinuum, the absorption of which has a finite value. The second part of the spectral term corresponds to the so-called scattering states of the exciton, which constitute a real continuum. Of course, by letting $\alpha = 3$ or 2 in Eq. (1), one gets the well-known expressions of Elliott¹⁰ and of Shinada and Sugano.¹¹ Broadening effects can be included by convolution with a Lorentzian function of half width at half maximum β .

Obviously, the most surprising conjecture of the fractional-dimensional formalism is that the physical mechanism causing the confinement, the geometrical shape, and the dimensions of the confining system are entirely described by a single dimensionality parameter. As a corollary, this approach predicts that, as far as the shape of the absorption spectrum is concerned, completely different physical systems should show identical spectra, provided that they have the same dimensionality, i.e., the same exciton binding energy. Up to now, this question had not been investigated in such terms, because the common intuition suggests that the details of the system strongly influence optical properties. This explains the fact that very few attempts have been made to propose a simple and universal modeling of optical spectra in quantum-confined structures. So, the fundamental question is the following: Is there really a one-to-one relationship between the binding energy of the perturbed exciton and the shape of the absorption spectrum, independent of the origin of the spatial restriction?

In this paper, we address this question by calculating linear absorption spectra near the fundamental band-gap energy of semiconductor systems such as quantum wells, quantum wires—of various geometries—and bulk materials in magnetic fields. The prevision of the fractional-dimensional Elliott equation is checked by carrying out an advanced all-numerical calculation of the *n*dimensional linear optical susceptibility (*n* is an integer). Using the density matrix formalism in the framework of the envelope functions, the linear optical susceptibility can be traced back to a quantum-mechanical two-particle problem for the electron-hole motion, which contains the confinement potentials in the case of a microstructure,^{3,4} or the magnetic field:⁵

$$\chi^{(nD)}(\omega) = \frac{|\mu|^2}{\epsilon_0} \frac{1}{\Omega^{(nD)}} \int d^3r \int d^3r' \times \sum_{\Lambda} \frac{\Phi_{\Lambda}(\mathbf{r}, \mathbf{r}) \Phi_{\Lambda}^*(\mathbf{r}', \mathbf{r}')}{E_{\Lambda} - \hbar(\omega + i\beta)} , \qquad (2)$$

where

$$\left[\frac{1}{2m_e}\left|\frac{\hbar}{i}\nabla_e + e\,\mathbf{A}(\mathbf{r}_e)\right|^2 + W_e(\mathbf{r}_e) + \frac{1}{2m_h}\left|\frac{\hbar}{i}\nabla_h - e\,\mathbf{A}(\mathbf{r}_h)\right|^2 + W_h(\mathbf{r}_h) - \frac{e^2}{4\pi\epsilon_0\epsilon|\mathbf{r}_e - \mathbf{r}_h|}\right]\Phi_{\Lambda}(\mathbf{r}_e,\mathbf{r}_h) = E_{\Lambda}\Phi_{\Lambda}(\mathbf{r}_e,\mathbf{r}_h) .$$
(3)



FIG. 1. (a) Calculated absorption spectra of an ideal quantum-well wire obtained from a vanishingly thin quantum well by adding a parabolic potential well along one in-plane direction (the total additional electron-hole confinement energy is $\hbar\Omega = 6\Re$). (b) and (c) The same for two ideal quantum wells of respective widths 0.4 and 1.6 a_B . Dots correspond to numerical calculations of the dielectric susceptibility. Solid curves result from the fractional-dimensional Elliott equation. The inset gives a magnified view of the high-energy parts of the spectra.

Here, $m_{e/h}$ are the masses and $W_{e/h}$ the confinement potentials for the electron and hole, respectively, and **A** is the vector potential of the magnetic field. The quantity $\Omega^{(nD)}$ denotes the *n*-dimensional normalization volume. The Schrödinger equation is numerically solved in real space using a representation in real space with 5000-50 000 base functions, in order to model the continuous spectrum. For simplicity, we consider ideal systems with equal electron and hole effective masses, infinitely high square and/or parabolic potential wells, and we express all energies and lengths in units of the 3D exciton effective Rydberg (\mathcal{R}) and Bohr radius a_B , respectively.

For each situation, the numerical calculation provides the binding energy of the 1s exciton state. Using the above relationship between E_{1s} and α , we derive the corresponding dimensionality, which allows us to calculate analytically the spectral-shape term in Eq. (1). This spectrum is finally compared with the result of the numerical calculation.

Figures 1 and 2 display examples of such calculations for different physical systems. The absorption spectra have only been calculated near the fundamental valenceto-conduction band gap. In other words, we have not considered here the absorption due to possible excited confinement subbands. In fact, bound exciton states related to such excited critical points are coupled with scattering states originating from lower-lying singularities. This leads to the well-known Fano resonances,^{4,5} which strongly affect the shape of absorption spectra, in particular when broadening effects are small. Such phenomena fall outside the scope of this study.

In all cases, the intensities of the 1s absorption peaks, calculated by both analytical and numerical methods,



FIG. 2. Calculated absorption spectra of two pairs of physical systems with identical exciton binding energies. (a) corresponds to a square $1.134a_B$ -wide quantum well (solid line) as well as to a quantum-well wire (dashed line), built from a $0.8a_B$ -wide well, with an additional in-plane parabolic confinement. The total additional electron-hole confinement is $2\Re$. (b) is the spectrum of a $0.091a_B$ -wide square well (solid line), or that of a cylindrical quantum wire (dashed line) with completely parabolic confinement (electron-hole confinement energy: $6\Re$). Dotted lines show the results of the α -dimensional Elliott equation. In both cases (a) and (b), it is almost impossible to distinguish between the three lines.

have been matched to each other, and very small broadening parameters ($\beta = 0.2 \Re$) have been assumed, so as to emphasize as many details as possible. Figure 1 shows the spectra obtained for ideal quantum wells with infinite potential barriers and two different widths, and for a quantum wire obtained from a vanishingly thin square well by adding a parabolic potential well along one direction, in the layer plane (quantum-well wire). The sharper the curvature of this potential profile, the stronger the confinement.

In Fig. 2, we present the spectra obtained for two square quantum wells, compared, respectively, to that of a quantum-well wire, built upon a quantum well of finite thickness by adding a parabolic in-plane potential well, and to that of an ideal cylindrical quantum wire with a parabolic confinement potential. Note that the latter situation is essentially similar to that obtained by applying a magnetic field onto a bulk semiconductor. In this last case, the strength of the field-which acts as the curvature of the quadratic potential of the cylindrical wirecontrols the degree of confinement of the electron-hole relative motion, around the field axis. The numerical parameters in Fig. 2 have been taken such that the two pairs of systems exhibit exactly the same exciton binding The principal effects of increasing the energy. confinement-of reducing the dimensionality-are the shift of the 1s peak towards lower energy, and the simultaneous enhancement of its intensity, relative to the rest of the spectrum. A slight change in the variation of the absorption by scattering states, above the band gap, can also be noticed.

Whatever the situation, a striking agreement is ob-

tained between both numerical and analytical methods on all important points: both the energy and relative intensity of the secondary absorption peak, mainly due to the contribution of the 2s state, are given as almost identical by both approaches. The same remark is valid for the absorbance due to excitonic unbound states, above the freeelectron band gap, and for its spectral variation. Even more striking is the fact that excitonic absorption spectra of systems with very different shapes and physical nature seem to follow some scaling law, of which the fractionaldimensional approach gives an excellent picture. This can be observed in Fig. 2, where, for each value of the dimensionality, all curves are very close to each other, which is an interesting result in itself.

We have achieved a systematic verification of this point by calculating the absorption spectra of a variety of structures, namely, (i) quantum wells of various widths; (ii) quantum-well wires based on square wells of thicknesses 0 and 0.8 Bohr radii, with several curvatures of the additional in-plane quadratic potential; (iii) quantum wires obtained by parabolic potential wells with a cylindrical symmetry around the wire axis; and (iv) bulk semiconductors submitted to magnetic fields of various strengths. We have chosen to characterize the spectral shape of the absorption by the value of the ratio, R = O(E = 0) / O(1s), of the optical density at the band gap (E=0) to the optical density at the 1s peak. Now, the fractional-dimensional formalism yields an explicit variation of R versus E_{1s} ; in other words, any reference to the α -dimensional parametrization can be eliminated: Lorentzian for а broadening, we obtain R $= \pi \beta x^{2x+2}/2\Gamma(2x), \text{ where } x = \sqrt{\mathcal{R}/E_{1s}}.$

In Fig. 3 we have plotted \mathcal{R} versus E_{1s} , not only from the above equation (solid curve), but also from the results of numerical calculations (circles, squares, triangles). Figure 3 proves, first, that there *is* effectively a very good correlation between the exciton binding energy and the general shape of the absorption spectrum, regardless of the physical mechanism of the confinement. Second, Fig. 3 demonstrates that this correlation is very well described by the fractional-dimensional approach.



FIG. 3. Plot of the dimensionless ratio between optical densities at the free-electron band gap and at the top of the 1s peak vs the corresponding exciton binding energy. Circles, squares, and triangles correspond to different physical systems, while the curve shows the result of the fractional-dimensional formalism.

Practically, this result constitutes a warranty of reliability for the analytical modeling which can be safely used for all types of semiconductor heterostructures. The simplicity of its formulation is a decisive advantage over costly numerical procedures.

From the fundamental viewpoint, the above result can be explained by the fact that the optical response is essentially a picture of a spatially averaged electron-hole interaction: it is obtained as the integrated probability for creating an electron-hole pair by absorption of a photon. Boundary effects are thus washed out, which explains their small contribution. Reducing the dimensionality then forces the spectrum to follow a kind of general pattern, of which the above generalized Elliot formula is a remarkable approximation. Besides, it is most likely that the same formula can describe cases where the electronhole interaction is weakened (namely, type-II or separate-confinement structures), by considering dimensionalities larger than 3. We have shown an example of the considerable simplification brought by the use of fractional dimensions. Now, the above type of scaling behavior is probably met in many other fields of physics: experimentalists often obtain results which lie "somewhere in between" the usual integer-dimensional cases. We believe that a fractional-dimensional approach should be serviceable in all cases where the measured quantity gives an averaged picture of the system, wiping out microscopic features. The mathematical determination of the effective dimensionality remains an open question, but "seminumerical" methods, comparable to those described above, could be used.

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