Theory of asymmetric broadening and shift of excitons in quantum structures with rough interfaces

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A quantum-mechanical theory of the influence of disorder on the motion of free particles is presented. Taking into account the dominant pair correlations of the potential fluctuations an analytical formula is derived for the asymmetrically broadened and shifted spectral function. The results are in agreement with a complete numerical treatment of disorder. The theory is applied to excitons in quantum wells with rough interfaces. We show that mainly the center-of-mass motion is influenced. Both the effect of the correlation and the strength of the accompanying potential fluctuations on the optical absorption are discussed. We clarify the relationship between the asymmetry, the shift, and the width of exciton lines. In the limit that these effects are only due to static interface disorder, an analytical relation between potential strength and correlation length is suggested.

I. INTRODUCTION

Crystal-growth techniques together with nanometer lithographic techniques have made it possible to fabricate various semiconductor quantum structures, which have great technological and scientific importance.¹ Among them are quasi-two-dimensional (2D) quantumwell and superlattice structures, but also quantum-wire and quantum-dot systems with quasi-one-dimensional (1D) or quasi-zero-dimensional (0D) character. Despite the progress in atomic-layer epitaxy, as in molecularbeam epitaxy, the fabricated interfaces are far from perfect. Structural disorder on the atomic scale occurs at the growing surface which becomes the interface.² The resulting nominally flat interfaces exhibit islands and terraces, their lateral extent depends on the growth condition. Furthermore, interdiffusion takes place.

Consequently the disorder, or more strictly speaking the interface roughness, can be traced back at least to two origins in the case of III-V compound interfaces, such as $GaAs/Ga_xAl_{1-x}As$ (001) ones. First, the island growth produces steps the heights of which are clearly related to monolayer thickness $a_0/2$ (a_0 is the bulk lattice constant). The process gives rise to fluctuations in the well thickness L_z of quantum-well structures. Since the lateral extent of the resulting islands is larger than $a_0/2$, atomically "rough" heterointerfaces appear. The second type of interface disorder is related to atomic interdiffusion, e.g., the cation exchange in the $GaAs/Ga_xAl_{1-x}As$ case which occurs for thermodynamical reasons. That means, the simple interpretation of interface disorder only in terms of an island size is too naive.³ A completely different situation occurs for the etched surfaces of freestanding wires or dots¹ or buried wires and dots fabricated by laser-induced cation interdiffusion.⁴ However, the influence of the strong geometrical and compositional fluctuations is reduced due to the smoothing action of the band bending.

The spectroscopies of optical interband transitions across the band gap, in particular photoluminescence and optical absorption, are powerful tools to study semiconductor microstructures. The optical processes near the absorption edge are determined by the excitons. The excitons give rise to remarkable probes of the interface disorder. A common feature of the 2D systems is the large inhomogeneous broadening of the exciton transitions, compared with those in pure bulk materials, mainly by the random fluctuations in the well thickness.^{2,5} In 1D wire systems opposite tendencies can be observed depending on the technology of the wire fabrication.^{4,6,7}

The random potentials, arising mainly from local thickness fluctuations in 2D quantum-well structures and affecting the motion of both electrons and holes, give rise not only to an inhomogeneous broadening of the exciton lines. Besides the asymmetric line shape, the interface roughness should result in a shift of the lines depending on the emission or excitation character of the optical process. A remarkable Stokes shift between exciton lines in absorption and luminescence spectra is observed at low temperatures (cf. Ref. 5 and references therein). Obviously, the optical line shape of excitons in semiconductor microstructures is not only determined by static disorder effects, but also by inelastic scattering processes.⁸

Disorder phenomena play an important role in various fields of physics. They not only influence the optical properties but also thermodynamic, acoustic, and transport properties. Despite their random nature accompanied by fluctuations on a microscopic scale, they may cause a quite deterministic behavior in the measurable macroscopic quantities. Hence, a theoretical understanding is of great interest. However, a detailed microscopic or quantum-mechanical description is complicated,⁹ since one has to deal with quantities such as displacement fields and wave functions which can be only calculated for a certain configuration. Moreover, the macroscopic quantity is not simply obtained by a superposition of results of dif-

ferent deterministic microscopic systems. Attempts^{10,11} at a closed theoretical description are restricted to one dimension and limiting conditions with respect to the type of disorder. Recently, such studies have been supplemented by computer simulations of molecular-beam-epitaxy growth of quantum-well wires on a vicinal surface.

A theory of exciton line broadening in terms of monolayer fluctuations has been developed by Weisbuch et al.¹² and, in particular, by Singh, Bajaj, and Chaudhuri¹³ for 2D quantum-well systems. Their picture of weighted interference of luminescence lines is. however, somewhat too naive. A full specification of the interface roughness spectrum is needed for a consistent interpretation of both photoluminescence and structural data.³ Zimmermann and co-workers^{14,15} have treated the effect of the layer-thickness variations in quantum-well structures on the exciton linewidth in more detail. The Schrödinger equation for the center-of-mass motion in a disordered potential is solved and the consequences for the broadening and dephasing are discussed for a Gaussian distribution of the random potential. For decreasing temperature he suggested a transition from a thermal to a topological exciton line shape. The later one is governed by disorder effects. The broadening problem is completely classically attacked by Yang et al.⁵ and Wilkinson et al.¹⁶ It has been shown that the coefficient of proportionality between the Stokes shift and the exciton absorption linewidth is a constant, independent of the material parameters.

A particular problem of the description of exciton spectra in 2D concerns the inhomogeneity of the broadening. The classical treatment of the particle motion is unable to explain the asymmetry of the lines.^{5,16} Generally, there are fundamental difficulties to solve the problem in 2D. Therefore, other authors¹⁷ have taken the results from 1D as a motivation for a heuristic asymmetric line-shape model of quantum-well excitons. The wave-vector conservation is broken and a certain distribution of wave vectors around the zero value is introduced. Similarly the reduction of the linewidth in quantum-wire and quantum-dot systems fabricated by laser-induced interdiffusion is explained in a more phenomenological way.^{18,19}

The major objective of the present paper is to develop a quantum-mechanical theory of asymmetric line shapes that explains the effect of disorder on the line broadening itself, the asymmetry of the broadening, and the line shift on the same level of approximation. In Sec. II, we derive analytical results for the spectral function of a single particle in a random potential, only assuming dominant pair correlations of the potential fluctuations. A successive procedure for the treatment of the particle dispersion and for the inclusion of line-shape asymmetry is given. The validity of the results are checked by comparison with numerical results in 1D and the self-averaging limit. In Sec. III, the theory is applied to the excitons in semiconductor microstructures with rough interfaces. The 2D case of quantum-well structures with strong confinement is explicitly treated. The line shape of optical absorption spectra is computed. Finally, in Sec. IV, a brief summary is given.

II. TREATMENT OF RANDOM POTENTIALS: SINGLE-PARTICLE PROBLEM

A. Random potential and configuration average

We try to find the spectral function for a particle in a volume Ω obeying the Hamiltonian

$$\hat{H} = \hat{H}_0 + V(\mathbf{x}), \qquad (1)$$

where $V(\mathbf{x})$ is a random potential due to, e.g., a more or less random arrangement of the atoms in the sample or certain regions of the sample. \hat{H}_0 denotes the Hamiltonian of the ordered reference system without randomness. In the practical applications of the theory we restrict our considerations to the nearly free motion of a particle with the effective mass m^* . The particle motion may be in three, two, or one dimensions. Correspondingly the space-point vector \mathbf{x} and space elements in the integrations have to be interpreted.

The statistical properties of the random potential are completely characterized by its correlation functions (m = 1, 2, 3, ...)

$$\Psi_{\boldsymbol{m}}(\mathbf{x}_1,\ldots,\mathbf{x}_m) = \langle V(\mathbf{x}_1)\cdots V(\mathbf{x}_m)\rangle , \qquad (2)$$

where $\langle \cdots \rangle$ stands for the configuration average. Without loss of generality, the averaged potential is assumed to be zero, i.e., $\langle V(\mathbf{x}) \rangle \equiv 0$. That means, only fluctuations are taken into account, whereas the average value is already included in the Hamiltonian \hat{H}_0 . In the selfaveraging limit the configuration average $\langle \cdots \rangle$ can be replaced by a spatial average. For large average volumes Ω the macroscopic homogeneity $\Psi_m(\mathbf{x}_1, \ldots, \mathbf{x}_m) =$ $\Psi_m(\mathbf{x}_1 - \mathbf{x}_m, \ldots, \mathbf{x}_{m-1} - \mathbf{x}_m)$ is obvious with the consequence that the correlation functions depend only on (m-1) space-point differences.

The configurationally averaged quantities, which are most important for the characterization of the randomness in the system, are the probability density $P(\omega)$, that in a certain space point **x** a random potential with a certain energy $\hbar\omega$ is realized,

$$P(\omega) = \left\langle \delta \left[\hbar \omega - V(\mathbf{x}) \right] \right\rangle, \tag{3}$$

and the binary correlation function $(\Psi_2 = \Psi)$

$$\Psi(\mathbf{x}_1 - \mathbf{x}_2) = \langle V(\mathbf{x}_1) V(\mathbf{x}_2) \rangle, \qquad (4)$$

respectively. In the case of macroscopic homogeneity the probability density $P(\omega)$ does not depend on the space coordinate.

B. Green's function

The Hermitian Hamiltonian \hat{H} allows the introduction of a left-hand-side resolvent operator for a given complex number z. The corresponding configurationally averaged quantity, the Green's function

$$\hat{G}(\mathbf{x}, \mathbf{x}'; z) = \left\langle (\hbar z - \hat{H})^{-1} \delta(\mathbf{x} - \mathbf{x}') \right\rangle$$
(5)

is represented in terms of eigenstates $|\mathbf{k}\rangle$ of the unperturbed Hamiltonian \hat{H}_0 characterized by sets of quantum numbers \mathbf{k} and energies $\varepsilon(\mathbf{k})$. We neglect off-diagonal elements induced by the random potential and consider only diagonal matrix elements $G(\mathbf{k}, z) = \langle \mathbf{k} | \hat{G}(z) | \mathbf{k} \rangle$ of the Green's function (5). The unperturbed Green's function, i. e., that for vanishing random potentials, is diagonal and has the form $g(\mathbf{k}, z) = [\hbar z - \varepsilon(\mathbf{k})]^{-1}$.

The full Green's function is expanded in a Taylor series with respect to the matrix elements $\tilde{V}(\mathbf{k}-\mathbf{k}') = \langle \mathbf{k} | V | \mathbf{k}' \rangle$, which corresponds to the spatial Fourier transforms of the random potential. It follows

$$G(\mathbf{k}, z) = \sum_{m=0}^{\infty} G^{(m)}(\mathbf{k}, z) , \qquad (6)$$

$$G^{(m)}(\mathbf{k}, z) = \sum_{\mathbf{k}_1 \dots \mathbf{k}_{m-1}} \langle g(\mathbf{k}, z) \, \tilde{V}(\mathbf{k} - \mathbf{k}_1) g(\mathbf{k}_1, z) \cdots \\ \times g(\mathbf{k}_{m-1}, z) \, \tilde{V}(\mathbf{k}_{m-1} - \mathbf{k}) g(\mathbf{k}, z) \rangle$$
(7)

with the Fourier-transformed correlation function (2).

They are calculated approximately. According to Wick's theorem all possible contractions $\tilde{V}(\mathbf{k}_1 - \mathbf{k}_2)\tilde{V}(\mathbf{k}_3 - \mathbf{k}_4)$ of two appearing matrix elements are taken into account and are replaced by the configurationally averaged values. Assuming their statistical independence for different index pairs it holds $\langle \tilde{V}(\mathbf{k}_1 - \mathbf{k}_2)\tilde{V}(\mathbf{k}_3 - \mathbf{k}_4)\rangle = \delta_{\mathbf{k}_1 - \mathbf{k}_2, \mathbf{k}_4 - \mathbf{k}_3}\tilde{\Psi}(\mathbf{k}_1 - \mathbf{k}_2)$. Within the Hartree-Fock approximation the statistical properties of the disordered system are completely traced back onto the pair correlation of the potential fluctuations. As a consequence the even-numbered terms in Eq. (7) can be expressed by the Fourier transform of the binary correlation function $\Psi(\mathbf{x}_1 - \mathbf{x}_2)$,

$$\tilde{\Psi}(\mathbf{k}) = \frac{1}{\Omega} \int_{\Omega} d\mathbf{x} \, e^{-i\mathbf{k}\cdot\mathbf{x}} \Psi(\mathbf{x}) \,, \tag{8}$$

whereas the odd-numbered terms vanish automatically. The expansion coefficients can be calculated in this way for arbitrary powers m. One obtains in each order (m - 1)!! contributions. Here we write down only the first nonvanishing-order terms (m = 0, 2, 4, ...)

$$G^{(0)}(\mathbf{k}, z) = g(\mathbf{k}, z) ,$$

$$G^{(2)}(\mathbf{k}, z) = g^{3}(\mathbf{k}, z) \sum_{\mathbf{k}_{1}} \tilde{\Psi}(\mathbf{k} - \mathbf{k}_{1})g(\mathbf{k}_{1}, z)/g(\mathbf{k}, z) ,$$

$$G^{(4)}(\mathbf{k}, z) = g^{5}(\mathbf{k}, z) \sum_{\mathbf{k}_{1}, \mathbf{k}_{2}} [\tilde{\Psi}(\mathbf{k} - \mathbf{k}_{2}) + \tilde{\Psi}(\mathbf{k}_{1} - \mathbf{k}_{2})g(\mathbf{k} - \mathbf{k}_{1} + \mathbf{k}_{2}, z)/g(\mathbf{k}, z) + \tilde{\Psi}(\mathbf{k}_{1} - \mathbf{k}_{2})g(\mathbf{k}, z)]$$

$$\times \tilde{\Psi}(\mathbf{k} - \mathbf{k}_{1})[g(\mathbf{k}_{1}, z)/g(\mathbf{k}, z)]g(\mathbf{k}_{2}, z)/g(\mathbf{k}, z) .$$
(9)

A consequence of the decoupling of the higher correlation functions concerns the probability density (3). Since only one and the same space coordinate appears the higher even-numbered correlation functions give rise to (m-1)!!equal contributions $[\Psi(0)]^{m/2}$. Summing up one obtains a Gaussian function

$$P(\omega) = \frac{1}{\sqrt{2\pi\sigma^2}} e^{-\frac{(\hbar\omega)^2}{2\sigma^2}}; \quad \sigma^2 = \Psi(0), \qquad (10)$$

the variance of which is governed by the autocorrelation at zero distances.

C. Broadening function

The configurationally averaged Green's function of the random system can be generated by a convolution

$$G(\mathbf{k}, z) = \hbar \int_{-\infty}^{+\infty} d\omega \, A(\mathbf{k}, \omega) \, g(\mathbf{k}, z + \omega) \tag{11}$$

of the unperturbed Green's function $g(\mathbf{k}, z)$ and a spectral function $A(\mathbf{k}, \omega)$. This spectral function represents the full information about the randomness in the system

by a distribution of the spectral weights from that of the Dirac δ function over the whole spectrum. In the limit of vanishing random potentials V, it holds that $A(\mathbf{k},\omega) = \delta(\omega)$. Shifting the argument by the unperturbed one-particle energy $\varepsilon(\mathbf{k})$, one immediately sees that the function $A[\mathbf{k}, \omega - \varepsilon(\mathbf{k})/\hbar]$ also represents the full spectral function of the electron and, hence, contains the full information about the electronic system. Expanding the unperturbed Green's function in expression (11) with respect to the frequency, i. e.,

$$G(\mathbf{k}, z) = \sum_{m=0}^{\infty} A_m(\mathbf{k}) \left[g(\mathbf{k}, z) \right]^{m+1} ,$$
$$A_m(\mathbf{k}) = \hbar^{m+1} \int_{-\infty}^{+\infty} d\omega \, \omega^m A(\mathbf{k}, \omega) , \qquad (12)$$

the moments $A_m(\mathbf{k})$ of the broadening function appear.

Unfortunately an exact analytical representation cannot be generally generated from Eq. (9), neither for the moments nor for the broadening function. In the limit of dispersionless particles with $g(\mathbf{k}_1, z)/g(\mathbf{k}, z) \equiv 1$ such a result follows immediately when comparing Eqs. (9) and (12). One finds

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$$A_m(\mathbf{k}) = \begin{cases} (m-1)!! \left[\Psi_2(0)\right]^{m/2} & \text{for even } m \\ 0 & \text{for odd } m. \end{cases}$$
(13)

Consequently, the spectral function is Gaussian broadened. In the limit of heavy particles with dispersionless energies it is identical with the probability density (3) to find a certain energy value in the potential fluctuations, i. e., $A(\mathbf{k}, \omega) \equiv P(\omega)$. In agreement with the starting approximation the spectral function is independent of the energy of the considered particle. We mention that a Gaussian-broadened function $A(\mathbf{k}, \omega)$ also results for particles with dispersion but in the special case of longrange potential fluctuations, i. e., $\Psi(\mathbf{x}_1 - \mathbf{x}_2) = \Psi(0)$ and $\tilde{\Psi}(\mathbf{k}_1 - \mathbf{k}_2) = \delta_{\mathbf{k}_1\mathbf{k}_2}\Psi(0)$. For the solution of the problem in the opposite limit of white Gaussian noise with $\Psi(\mathbf{x}_1 - \mathbf{x}_2) \sim \delta(\mathbf{x}_1 - \mathbf{x}_2)$ and $\tilde{\Psi}(\mathbf{k}_1 - \mathbf{k}_2) = \text{const}$, the reader is referred to the paper of Halperin.¹¹

The well-known result of Gaussian broadening (see e.g., Ref. 14) can be successively improved, considering the energy dispersion of the particle in all orders. From expression (9) we derive, assuming $\tilde{\Psi}(\mathbf{k}) = \tilde{\Psi}(|\mathbf{k}|)$ and even $m \ (m \ge 2)$,

$$G^{(m)}(\mathbf{k}, z) = [g(\mathbf{k}, z)]^{m+1} \left[(m-1)!! \, \sigma^m + \frac{1}{6} (m-1)!! \, m(m+1) \, \sigma^{m-2} \times \kappa^3 g(\mathbf{k}, z) + \cdots \right], \qquad (14)$$

with the autocorrelation of the field strengths related to the random potentials

$$\epsilon^{3} = \sum_{\mathbf{k}} \varepsilon(\mathbf{k}) \tilde{\Psi}(\mathbf{k}) = \frac{\hbar^{2}}{2m^{*}} \left\langle [\nabla V(\mathbf{x})] \cdot [\nabla V(\mathbf{x})] \right\rangle . \quad (15)$$

After rearrangement of the powers of the unperturbed Green's function in Eq. (14) and comparison of expressions (6) and (12) one finds for the moments in first order in κ^3

$$A_m(\mathbf{k}) = \begin{cases} (m-1)!! \, \sigma^m & \text{for } m = 0, 2, 4, \dots \\ \frac{1}{6}(m-2) \, !! (m-1)m \, \sigma^{m-3} \kappa^3 & \text{for } m = 3, 5, 7, \dots \end{cases}$$
(16)

and $A_1(\mathbf{k}) = 0$. The resulting spectral function representing the inhomogeneous broadening can be written with Eq. (10) as

$$A(\mathbf{k},\omega) = \left[1 - \frac{\kappa^3}{6} \frac{d^3}{d(\hbar\omega)^3}\right] P(\omega).$$
(17)

We emphasize that within the approximations made the first correction to the broadening function remains independent of the considered state $|\mathbf{k}\rangle$. Moreover, its coefficient obeys the same expression (15) for any arbitrary dimensionality of the particle motion. The interesting point of expression (17) is that the inhomogeneous line broadening is explained by the same random mechanism as the homogeneous one, at least, looking at the static topological contribution to the line broadening. In the light of Eq. (15) the inhomogeneous contribution results from the interference of the particle motion with a finite mass and modulation of this motion due to the potential fluctuations. Meanwhile, the same solution (17) has been found independently by Zimmermann.²⁰

From expression (17) we can derive a condition for the validity of the linear approximation with respect to the particle dispersion. It is valid for frequencies close to the energy of the unperturbed particle, that means

$$\frac{\kappa^3 |\hbar \omega - \varepsilon(\mathbf{k})|}{3\sigma^4} \ll 1.$$
(18)

For vanishing dispersion, i.e., $m^* \to \infty$ and $\kappa \to 0$ the correction to the homogeneous Gaussian-type broadening vanishes. The condition (18) also tells us that the perturbative treatment of the particle dispersion is only valid for a randomness of the potentials far from the white

Gaussian noise, i.e., a spatial dependence of the binary correlation function of $\Psi(\mathbf{x}_1 - \mathbf{x}_2) \sim \delta(\mathbf{x}_1 - \mathbf{x}_2)$.

D. Example

In order to verify the analytical results we carry out numerical calculations for Gaussian fluctuations of the random potential. We assume

$$\tilde{\Psi}(\mathbf{k}) = \gamma^2 e^{-\frac{1}{2}\lambda_c^2 \mathbf{k}^2},\tag{19}$$

(20)

with λ_c the correlation length of the potential fluctuations and γ as a parameter characterizing the strength of the potential fluctuations. For expression (19) the variance of the probability density (10) and the asymmetry parameter κ^3 (15) are given for the motion in N dimensions (N = 1, 2, 3) in a volume $\Omega = L^N$ by

$$\sigma^2 = \Psi(0) \;, \;\;\; \kappa^3 = N \Psi(0) \, {\hbar^2 \over 2m^* \lambda_c^2} \;,$$

 \mathbf{and}

$$\Psi(0) = \gamma^2 \left(rac{L}{\lambda_c}rac{1}{\sqrt{2\pi}}
ight)^N.$$

Unfortunately, a random potential is not characterized in a unique way by the binary correlation. Therefore, for the numerical treatment in the self-averaging limit we explicitly apply a Fourier representation of the form

$$V(\mathbf{x}) = \sum_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{x}} c(\mathbf{k}) [\tilde{\Psi}(\mathbf{k})]^{\frac{1}{2}}.$$
 (21)

The coefficients $c(\mathbf{k})$ denote statistically independent random numbers, equally distributed on the unit circle in the complex plane. Since the random potential is real, the restriction $c(\mathbf{k}) = c^*(-\mathbf{k})$ holds. Potentials of the type (21) describe Gaussian noise, because their Fourier amplitudes have a Gaussian distribution with standard deviations independent of the wave vector. The statistical properties of these potentials are completely characterized by the first and second moments, i. e., $\langle c(\mathbf{k}) \rangle = 0$ and $\langle c(\mathbf{k})c(\mathbf{k}') \rangle = \delta_{\mathbf{k},-\mathbf{k}'}$.

In the numerical calculations we have restricted ourselves to the one-dimensional case and used atomic units, Bohr radius a_B and Rydberg energy E_B of the single particle in the corresponding semiconducting material.



FIG. 1. Spectral function of a particle with zero kinetic energy moving in one dimension in a Gaussian random field according to Eq. (19). The length of the normalization interval $L = 4 \times 10^4 a_B$ and the correlation length of the potential fluctuations $\lambda_c = a_B$ are fixed. The strength of the potential fluctuations is varied: $\gamma = 0.5$ (1), 1.0 (2), 1.5 (3), and $2.0 \times 10^{-2} E_{\rm B}$ (4). Solid lines indicate results from the full numerical calculation, whereas dashed lines follow the approximate expressions (10) for dispersionless levels [upper part (a)] or (17) where dispersion is included in the first nonvanishing order [lower part (b)].

Typical values are $a_B = 10 \text{ nm}$ and $E_B = 4 \text{ meV}$, as found for GaAs with the effective reduced mass of a heavy-hole exciton. We calculate the spectral function $A(0,\omega)$ for a particle with vanishing kinetic energy by means of the method of finite elements with a dense real-space sampling and a normalization length L large compared to the characteristic correlation length λ_c of the potential fluctuations. Explicitly we use 2^{17} sample points on a length $L = 4 \times 10^4 a_B$. The correlation length is fixed to be $\lambda_c =$ a_B and the strength of the potential fluctuations is varied using $\gamma = 0.5$, 1.0, 1.5, and $2.0 \times 10^{-2} E_B$. For these parameters we find $\sigma = 0.6 \dots 2.5 E_B$ and $\kappa^3 = \sigma^2 E_B$. The condition for our approximation $\kappa^3/\sigma^3 \ll 1$ is nearly fulfilled, at least for the stronger potential fluctuations. Results are shown in Figs. 1(a) and 1(b). For numerical reasons an additional small Lorentzian broadening of $0.1 E_B$ is introduced into these figures.

The full numerical solutions of the problem presented in Fig. 1(a) as well as Fig. 1(b) as solid lines exhibit the two pronounced influences of the random potential. (i) The spectral function is broadened. Apart from the smaller asymmetric contribution the full linewidth Wat half maximum is essentially given by the variance, $W = \sigma \sqrt{2 \ln 2}$. (ii) The maximum of the spectral function is shifted to lower energies. The redshift follows nearly the law $S = \frac{1}{2}\kappa^3/\sigma^2$. Neglecting the dispersion of the particle energy [cf. Fig. 1(a)], the homogeneous line broadening can be nearly reproduced but not the line positions. However, after inclusion of the particle dispersion within the linear approach [cf. Fig. 1(b)], qualitatively the same features as in the exact solutions are observed. For the considered parameters the agreement of numerical and analytical curves is satisfying. This results from the fact that the first four moments of the two curves $A_m(k)$ (16) (m = 0, 1, 2, 3) are identical. The accuracy of this agreement is better for stronger potential fluctuations. With increasing γ [see expression (19)] the particle dispersion is of minor influence and the shift follows the approximate expression, which itself is independent of γ . The better agreement results from the fact that the condition $\kappa^3/\sigma^3 \ll 1$ for the validity of the perturbational treatment of the particle dispersion is better fulfilled. In fact, in the curves (3) and (4) in Fig. 1(b) it holds $\kappa^3/\sigma^3 = 0.53$ and 0.40, respectively.

III. APPLICATIONS: EXCITONS IN QUANTUM WELLS WITH ROUGH INTERFACES

A. Optical susceptibility

The optical susceptibility of a semiconductor microstructure near the fundamental absorption edge is given by

$$\chi(z) = -\frac{2|\mu|^2}{\varepsilon_0} \frac{1}{\Omega} \int d\mathbf{x} \int d\mathbf{x}' \, \hat{G}_2(\mathbf{x}, \mathbf{x}, \mathbf{x}', \mathbf{x}'; z), \quad (22)$$

with the dipole matrix element μ of the allowed optical

transition in the underlying bulk semiconductor, which is described within a two-band model, i. e., two parabolic bands (conduction band and usually heavy-hole band) with effective masses m_e and m_h separated by a certain energy gap E_g . ε_0 denotes the vacuum dielectric constant. In expression (22) the configurationally averaged resolvent \hat{G}_2 of the corresponding two-particle Hamiltonian \hat{H} ,

$$\hat{G}_{2}\left(\mathbf{x}_{e},\mathbf{x}_{h},\mathbf{x}_{e}',\mathbf{x}_{h}';z\right) = \langle [\hbar z - \hat{H}\left(\mathbf{x}_{e},\mathbf{x}_{h}\right)]^{-1}\delta\left(\mathbf{x}_{e} - \mathbf{x}_{e}'\right)\delta\left(\mathbf{x}_{h} - \mathbf{x}_{h}'\right)\rangle,\tag{23}$$

$$\hat{H}(\mathbf{x}_{e}, \mathbf{x}_{h}) = E_{g} - \frac{\hbar^{2}}{2m_{e}} \Delta_{\mathbf{x}_{e}} - \frac{\hbar^{2}}{2m_{h}} \Delta_{\mathbf{x}_{h}} - v\left(|\mathbf{x}_{e} - \mathbf{x}_{h}|\right) + U_{e}(\mathbf{x}_{e}) + V_{e}(\mathbf{x}_{e}) + U_{h}(\mathbf{x}_{h}) + V_{h}(\mathbf{x}_{h}),$$
(24)

is introduced. Electrons (e) and holes (h) interact via the Coulomb potential $v(|\mathbf{x}|)$ that is simply screened by the static electronic dielectric constant of the bulk material. Apart from the Coulomb attraction the motions of the two particles i = e, h are influenced by additional potentials $U_i(\mathbf{x}_i) + V_i(\mathbf{x}_i)$. For the considered microstructures they are due to the spatial variation of the two band edges near the interfaces. They are split into a systematic part $U_i(\mathbf{x}_i) = \langle U_i(\mathbf{x}_i) \rangle$ and a random part $V_i(\mathbf{x}_i)$ fluctuating around the average value, i. e., $\langle V_i(\mathbf{x}_i) \rangle = 0$.

We restrict our consideration to narrow quantum wells with interface normal vectors in the z direction. The exciton peaks are energetically well separated. Therefore, the two-particle wave functions factorize into confinement wave functions and functions of the remaining in-plane motion in the xy plane with $\mathbf{x}_i = (x_i, y_i)$ (i = e, h). The confinement wave functions are solutions of 1D single-particle Schrödinger equations with steplike confinement potentials $U_i(\mathbf{x}_i) \equiv U_i(z_i)$ describing the averaged spatial variation of the band discontinuities at the interfaces. The form of the Hamiltonian in Eq. (24) can also be used for the in-plane motion. The gap has to be replaced by $E_g^{(2D)}$, i.e., E_g is enlarged by the confinement energies of electron and hole in the considered subbands, e.g., the first heavy-hole and electron subbands. The Coulomb attraction v and the random potentials V_i now represent expectation values, w or W_i , with the confinement wave functions and, hence, depend only on the vectors \mathbf{x}_i in the xy plane. In the strong localization limit the attractive interaction w can be again replaced by a 2D Coulomb potential $v.^{21}$

The treatment seems to be reasonable for the description of atomically rough interfaces interdiffused on a microscopic length scale and/or covered with islands due to the formation of monolayer steps on the interfaces, whose extent is not large compared to the Bohr radius a_B of the exciton. In the case of extended islands the exciton lines feel only an effective well thickness and the exciton lines split via the different confinement effects on $E_g^{(2D)}$ (see Ref. 2 and references therein). This limit can be simply described by a superposition of spectra for different well thicknesses and should not be discussed here.

B. Influence of the random potential

For convenience, for all two-particle problems we introduce 2D center of mass (cm) and relative coordinates in the xy plane

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$$\mathbf{R} = \frac{m_e}{M_0} \mathbf{x}_e + \frac{m_h}{M_0} \mathbf{x}_h \qquad \mathbf{r} = \mathbf{x}_e - \mathbf{x}_h ,$$

$$M_0 = m_e + m_h ; \quad m_0 = m_e m_h / M_0 . \tag{25}$$

Without the random potentials $W_i(\mathbf{x}_i)$ the two motions decouple exactly. Therefore, in the presence of the random fields we expand the two-particle wave functions in terms of the solutions $\varphi_{\lambda}(\mathbf{r})$ and ε_{λ} (λ is the set of quantum numbers) of the 2D hydrogenic problem for the internal motion of the exciton. As a result of this procedure only the expansion coefficients, which depend on the cm coordinates, remain influenced by the random potentials, more strictly speaking by their matrix elements with the wave functions of the internal motion. Unfortunately, the resulting equations of motion are coupled with respect to different stages of the internal motion. Assuming weak coupling we neglect the intermixing of different states φ_{λ} of the internal motion by the disordered interfaces and take into account only diagonal elements

$$V_{\lambda}(\mathbf{R}) = \int d\mathbf{r} \, \left|\varphi_{\lambda}(\mathbf{r})\right|^{2} \left[W_{e}\left(\mathbf{R} + \frac{m_{h}}{M_{0}}\mathbf{r}\right) + W_{h}\left(\mathbf{R} - \frac{m_{e}}{M_{0}}\mathbf{r}\right)\right].$$
(26)

The optical susceptibility (22) can be rewritten in the form

$$\chi(z) = -\frac{2|\mu|^2}{\varepsilon_0} \sum_{\lambda} |\varphi_{\lambda}(0)|^2 \frac{1}{\Omega} \int d\mathbf{R} \int d\mathbf{R}' \, \hat{G}^{(\lambda)}[\mathbf{R}, \mathbf{R}'; z] - (E_g^{(2D)} + \varepsilon_{\lambda})/\hbar], \qquad (27)$$

by means of the single-particle Green's function from Eq. (5) configurationally averaged with respect to the potential in Eq. (26). The prefactor $|\varphi_{\lambda}(0)|^2$ characterizes the Coulomb enhancement of the oscillator strengths. Consequently, expression (27) represents an Elliott formula for 2D (Ref. 22) generalized to the case of the presence of random potentials due to the nonideal interfaces of the quantum-well structures.

The Green's function appearing in Eq. (27) is determined by the single-particle Hamiltonian of the form given in Eq. (1). However, the particle mass is now the total mass M_0 of the electron-hole pair and the random potential depends on the internal motion of the electronhole pair. Explicitly the 2D problem of the cm motion can be described by the Hamiltonian

$$\hat{H}^{(\lambda)} = -\frac{\hbar^2}{2M_0} \Delta_{\mathbf{R}} + V_{\lambda}(\mathbf{R}).$$
(28)

Applying the results of Eqs. (10) and (17) and taking into account the averaged vertical extent L_z of the quantum well, the susceptibility takes the form

$$\chi(z) = \frac{2|\mu|^2}{\varepsilon_0 L_z} \sum_{\lambda} |\varphi_{\lambda}(0)|^2 \hbar$$
$$\times \int_{-\infty}^{+\infty} d\omega \, \frac{A^{(\lambda)}(0,\omega)}{E_{g}^{(2D)} + \varepsilon_{\lambda} - \hbar(\omega + z)} \,, \qquad (29)$$

where the broadening function for vanishing kinetic energy of the cm motion has the form (1).

In the case of smooth random fields as, e.g., due to charged impurities in heavily doped semiconductors, instead of Eqs. (26) and (27) another approximation is more suitable.²³ In this case the potentials, $W_i(\mathbf{x}_i)$, can be linearly expanded with respect to the relative coordinates. An electric-field-induced broadening of the exciton lines due to the Franz-Keldysh effect appears and an Urbach tail of the absorption coefficient results instead of the Gaussian form of Eq. (29).

Expression (29) means that the broadening of each exciton peak, characterized by the quantum numbers λ , is different. Each peak has its own broadening function. The corresponding variance and the characteristic asymmetry parameter depend on the quantum numbers λ . For the explicit representation of these parameters we assume that the spatial dependencies of the random potentials, which influence electrons and holes, are the same, since they have the same geometrical origins. Hence we write $W_e(\mathbf{x}) = \nu W_h(\mathbf{x}) = \frac{1}{2}V(\mathbf{x})$, where $V(\mathbf{x})$ represents the space dependence and ν gives the ratio of the strengths of the random potentials at the same space point for electrons and holes. With the Fourier transform $\Psi(\mathbf{k})$ of the binary correlation function of the potential $V(\mathbf{x})$ from Eq. (8) and the Fourier transforms of the squared wave functions of the internal exciton motion

$$\rho_{\lambda}(\mathbf{k}) = \int d\mathbf{r} \, \left| \varphi_{\lambda}(\mathbf{r}) \right|^2 e^{-i\mathbf{k}\cdot\mathbf{r}} \,, \tag{30}$$

one finds

$$\sigma_{\lambda}^{2} = \sum_{\mathbf{k}} \tilde{\Psi}(\mathbf{k}) S^{(\lambda)} \left(\frac{1}{2}\mathbf{k}\right),$$

$$\kappa_{\lambda}^{3} = \sum_{\mathbf{k}} \frac{\hbar^{2}}{2M_{0}} \mathbf{k}^{2} \tilde{\Psi}(\mathbf{k}) S^{(\lambda)} \left(\frac{1}{2}\mathbf{k}\right),$$

$$S^{(\lambda)}(\mathbf{k}) = \frac{1}{4} \left\{ \left| \rho_{\lambda} \left(\frac{2m_{h}}{M_{0}}\mathbf{k}\right) \right|^{2} + 2\nu \operatorname{Re} \rho_{\lambda}^{*} \left(\frac{2m_{e}}{M_{0}}\mathbf{k}\right) \rho_{\lambda} \left(\frac{2m_{h}}{M_{0}}\mathbf{k}\right) + \nu^{2} \left| \rho_{\lambda} \left(\frac{2m_{e}}{M_{0}}\mathbf{k}\right) \right|^{2} \right\}.$$
(31)

These expressions are quite similar to those in Eqs. (10) and (15) for the single-particle case. However the integrands are now modified by the Fourier transforms of the probability to find an exciton in a certain internal quantum stage in space. During the explicit computations of the properties related to the excitons we have assumed electron-hole symmetry, i.e., $m_e = m_h$ and $\nu = 1$. This assumption considerably reduces the number of independent parameters but hardly influences the effect of randomness on the excitonic spectra.

Apart from the particular form of the Fouriertransformed binary correlation function (8) the line shape parameters σ_{λ} and κ_{λ} are mainly determined by the Fourier-transformed probabilities $\rho_{\lambda}(\mathbf{k})$ (30) of the exciton wave-function square. Because of the prefactor $|\varphi_{\lambda}(0)|^2$ in expression (29) only *s*-like states with $\lambda \equiv n$,s (n = 1, 2, ...) contribute to the optical susceptibility. For *s*-like states the Fourier transforms depend only on the module of the wave vector $\mathbf{k} = |\mathbf{k}|$.

C. Limiting cases

The interface disorder related to the roughness due to steps, island, or interdiffusion should be more or less isotropic in both directions in the 2D space. In this case the potentials $W_i(\mathbf{x}_i)$ and the correlation function $\Psi(\mathbf{R})$ should exhibit a similar dependence on the two Cartesian coordinates. However, for growth on vicinal (001) faces one should expect the appearance of interface domains, i.e., terraces where one direction is preferred. In this case the above-mentioned functions depend only on one coordinate. Here, we restrict ourselves to the first case. The potential fluctuations are mainly due to the varying thickness $L_z(\mathbf{x})$ of the quantum well under consideration. The thickness fluctuates around the average value L_z . For this case, Zimmermann¹⁴ has given an explicit form of the potentials, $W_i(\mathbf{x}_i) = [L_z(\mathbf{x}_i) - L_z] d\varepsilon_i / dL_z$, where ε_i denotes the confinement energy of the electron (i = e) or hole (i = h) due to the confinement in z direction. Here, we apply again the Gaussian form (19) of the binary correlation function. The strength γ is now clearly related to the thickness variation of the confinement energies and the mean-square deviation of the actual well thickness from its averaged value. The correlation length λ_c of the fluctuations defines the characteristic distances, for which the island growth (growth of steps) is correlated. It comes in the range of the atomic diffusion length at the growth temperature or smaller values.

Starting from the binary correlation function (19) it is interesting to study two limiting cases. The first one occurs if the flat interface regions due to the island growth are uncorrelated, more strictly speaking, if their correlation length is small compared to the Bohr radius of the exciton, $\lambda_c \ll a_B$. In this limit of white Gaussian noise it holds $\Psi(\mathbf{x}) = \tilde{\Psi} \Omega \delta(\mathbf{x})$ with Ω as a characteristic area now. The Fourier transform of the binary correlation function is nearly a constant, $\tilde{\Psi}(\mathbf{k}) \equiv \tilde{\Psi}$. With electronhole symmetry one derives from Eq. (31)

$$\sigma_\lambda^2 = 2\, ilde{\Psi}\,\Omega \int d{f r}\, \left|arphi_\lambda({f r})
ight|^4 \, ,$$

$$\kappa_{\lambda}^{3} = 8 \,\tilde{\Psi} \,\Omega \int d\mathbf{r} \, \left|\varphi_{\lambda}(\mathbf{r})\right|^{2} \left(-\frac{\hbar^{2}}{2M_{0}}\Delta_{\mathbf{r}}\right) \left|\varphi_{\lambda}(\mathbf{r})\right|^{2}.$$
 (32)

This φ^4 rule is already known¹⁴ for the squared variance of the Gaussian probability function. Quite different from the situation of single free particles, a finite asymmetry parameter κ_{λ} also occurs in the limit of white Gaussian noise. This is due to the restriction of the space integration by the exciton extent. Apart from the strength $\sim \tilde{\Psi}$ the magnitude of the line-shape parameters is governed by the internal motion of the exciton.

The second limit is characterized by large correlation lengths of the random potentials, more strictly speaking by lengths larger than the spatial extent of the λ th exciton, i.e., $\lambda_c \gg a_B$. Now the correlation function is localized in **k** space, i.e., $\tilde{\Psi}(\mathbf{k}) \sim \delta_{\mathbf{k}0}$. One finds the results presented in the single-particle case in Eqs. (10) and (15), which are independent of the considered exciton state λ , i.e., $\sigma_{\lambda}^2 = \Psi(0)$ and $\kappa_{\lambda}^3 \equiv 0$.

The exciton-line broadening cannot be determined analytically for arbitrary correlation lengths. However, this can be done for binary correlation functions of Gaussian type (19), at least for the most important 1s exciton peak. In this case it holds $\rho_{1s}(\frac{1}{2}k) = [1 + (\frac{1}{8}a_Bk)^2]^{-3/2}$ and the line-shape parameters can be calculated. We find

$$\sigma_{1s}^{2} = \Psi(0)zf(z) ,$$

$$\kappa_{1s}^{3} = \left(\frac{\hbar^{2}}{M_{0}\lambda_{c}^{2}}\right)\Psi(0)z^{2}\left(-\frac{d}{dz}\right)f(z) ,$$

$$z = \frac{1}{2}\left(\frac{8\lambda_{c}}{a_{B}}\right)^{2} , \quad f(z) = \frac{1}{2}\left[1-z-z^{2}e^{z}\mathrm{Ei}(-z)\right] ,$$
(33)

with the integral exponential function $\operatorname{Ei}(-z)$.

The two line-shape parameters in Eq. (33) govern the redshift of the exciton lines $S_{1s} \approx \frac{1}{2} \kappa_{1s}^3 / \sigma_{1s}^2$ and their (homogeneous) width $W_{1s} = \sigma_{1s} (2 \ln 2)^{\frac{1}{2}}$ due to the same type of interface disorder. Generally the ratio of both quantities is a complicated function of the correlation length in units of the exciton radius. However, in the limit of a not too small correlation length, i.e., $\lambda_c \approx a_B$ and $z \gg 1$, which should be fulfilled for the considered situation,^{2,14} the line-shape parameters become independent of the exciton extent.

D. Numerical results

In all practical cases, the limits of strongly correlated or rather uncorrelated potential fluctuations are hardly fulfilled. This fact is indicated in Fig. 2, where the pure exciton influence on the line-shape parameters is indicated. More strictly speaking, in this figure the ratios $\sigma_{ns}^2/\Psi(0)$ and $\kappa_{ns}^3/[\frac{\hbar^2}{M_0\lambda_c^2}\Psi(0)]$ are plotted versus the correlation length λ_c . The figure shows that for small correlation lengths, i.e., for nearly white Gaussian noise, the exciton ground state n = 1 is much more asymmetrically broadened than the higher exciton states. In principle, the disorder influence can be neglected for the excited states. In the limit of long-range correlations the line shapes of the different excitons lines approach each other independent of the exciton radius.

Complete absorption spectra of excitons are shown in Fig. 3 for a correlation length $\lambda_c = 2a_B$ fixed at the radius of the 2D ground state. For numerical reasons an additional homogeneous linewidth $\Gamma = 0.1E_B/\hbar$ due to lifetime broadening effects is assumed. Small (a) and large (b) amplitudes of the potentials fluctuations are considered. More exactly, the quantity Im $\chi(\omega + i\Gamma)$ is plotted versus the frequency ω . That means, the curves represent optical absorption spectra in the region of the exciton bound states. Neglecting temperature effects the low-energy part of the spectra can be also identified with the excitonic luminescence.

In the limit of small disorder effects [Fig. 3(a)] one



FIG. 2. Exciton influence on the parameters of the asymmetrically broadened Gaussian lines. The line-shape parameters $\sigma_{ns}^2/\Psi(0)$ (a) and $\kappa_{ns}^3/\left[\frac{\hbar^2}{M_0\lambda_c^2}\Psi(0)\right]$ (b) are plotted versus the correlation length of the potential fluctuations for the first lowest s-type excitons n = 1, 2, 3, 4, and 5.



FIG. 3. Optical absorption spectra $\text{Im} \chi(\omega + i\Gamma)$ in the region of the exciton bound states. The correlation length of the potential fluctuations is fixed to be $\lambda_c = 2a_B$. A small homogeneous lifetime broadening $\Gamma = 0.1E_B/\hbar$ is taken into account. The limit of small (a) and large (b) amplitudes of the potential fluctuations is considered. In (a) different strengths $\gamma = 0.0$ (solid line), 0.2 (dashed line), and 0.4 E_B (dotted line) are considered. The value in (b) is $\gamma = 2E_B$.

observes mainly the 1s exciton peak. It is slightly broadened and redshifted by the disorder effects. The redshift and the broadening increase with increasing potential fluctuations. Dramatic changes in the line shape of the spectra happen in the case that the broadening effects are in the range of half the exciton binding energy. One observes only one broad peak in the region of the bound states. On the low-energy side it is accompanied by an exponential tail with a quadratic frequency dependence of the exponent. On the high-energy side an additional peak appears. It is due to the Coulomb enhancement of the optical oscillator strengths for scattering states. Without disorder it represents a step at the continuum edge, which is, however, unimportant because of its smallness. It is rather insensitive with respect to disorder in contrast to the line strengths of the bound states. Therefore, it becomes evident in the limit of strong disorder.

IV. SUMMARY

We have developed a method of calculation of the motion of single particles in random fields. The resulting spectral functions are exact for Gaussian-distributed fields and dispersionless levels. They are Gaussian broadened with the square root of the binary correlation function of the random potentials at zero distances as the variance. The results are successively improved taking into account the finite mass of the particles. As a consequence the lines are not only homogeneously broadened but become asymmetric and redshifted. The mechanism that is responsible for the asymmetry and the redshift is in principle the same as that for the homogeneous broadening, the pair correlation of the potential fluctuations. However, it is superimposed with effects of the kinetic energy of the particle.

The method developed for the disorder treatment is applied to excitons in quantum structures with atomically rough interfaces, in particular to narrow quantum wells with slightly interdiffused and stepped interfaces with characteristic lengths of the order of or smaller than the Bohr radius of the excitons. However, it can be also applied to other quantum structures with more complicated interfaces and, hence, other interface disorder of Gaussian type. The influence of the fluctuating potentials can be reduced to that on the motion of the center of mass of the electron-hole pair. The random potentials depending only on the center-of-mass coordinates represent, however, matrix elements between wave functions of the internal motion.

Neglecting the off-diagonal elements, the problem of the exciton in rough quantum wells is traced back to the motion of a single particle, i.e., the center of mass, in an effective random potential. However, because of the appearance of matrix elements, the binary correlation function is everywhere weighted by the square of the probability to find the exciton in a certain space point in a certain quantum stage of the internal motion. Generally, correlation lengths of the potential fluctuations of the order of the exciton radius are assumed. However, the two cases of weak and strong potential fluctuations are discussed in more detail. In the first limit the exciton lines remain well energetically separated but are slightly inhomogeneously broadened and redshifted. In the case of strong disorder a qualitatively new spectrum appears. One observes a wide asymmetric main peak with an exponential tail at the low-energy side. A new structure appears at the continuum edge since the disorder hardly influences the Coulomb enhancement of the scattering states is contrast to the bound states.

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