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# Theory of the Excitonic Lineshape in Low-Dimensional Structures with Rough Interfaces

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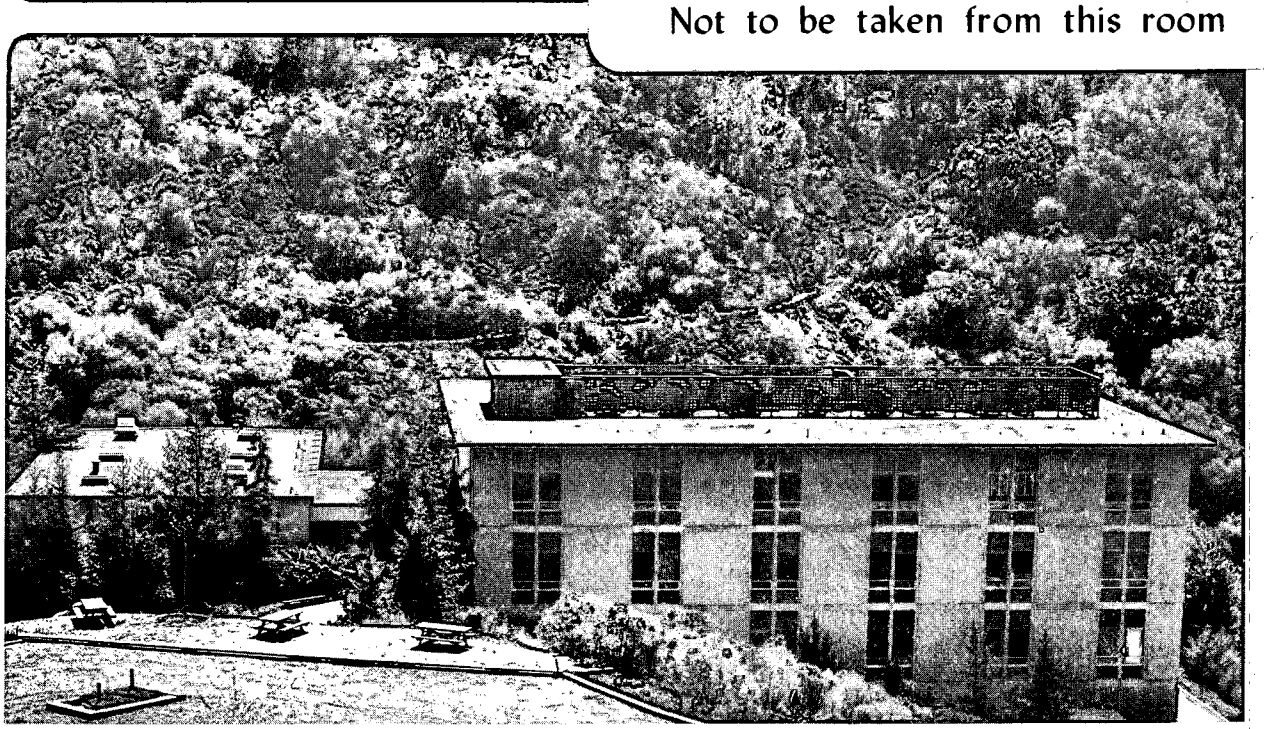

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# THEORY OF THE EXCITONIC LINESHAPE IN LOW-DIMENSIONAL STRUCTURES WITH ROUGH INTERFACES

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# THEORY OF THE EXCITONIC LINESHAPE IN LOW-DIMENSIONAL STRUCTURES WITH ROUGH INTERFACES

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## Abstract

A quantum mechanical theory of single-particle motion under the influence of a stochastic potential is presented. By solving the Dyson equation, the one-particle Green's function can be determined approximately. Assuming a Gaussian distribution, an explicit expression is derived for the spectral function. The analytical results are in good agreement with numerically calculated data. The theory is applied to excitons on rough interfaces. The optical absorption of excitons on rough surfaces can be traced back to the one-particle Green's function for the center-of-mass motion. In contrast to the classical treatment, the asymmetry of the lines and the redshift of the maxima can be explained. The coefficients for linewidth and asymmetry can be expressed in terms of the excitonic wavefunction and the binary correlation function of the stochastic potential.

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## I. INTRODUCTION

The progress in crystal-growth techniques and nanometer lithography allow the production of various semiconductor microstructures which are of great scientific and technological interest [1]. Even in high-quality samples the interfaces are far from perfect [2]. In quantum wells, the fluctuations of the thickness lead to a strong inhomogeneous broadening, compared to bulk semiconductors [2,3]. In quantum-well wires, opposite tendencies can be observed, depending on the method of fabrication [4–6]. Besides a broadening, the absorption lines of the excitons are asymmetric and their maxima are shifted towards lower energies.

Weisbuch *et al.* [7] and Singh *et al.* [8] have developed theories to explain the broadening in terms of weighted interference of luminescence lines. Zimmermann [9] has treated the effect of layer-thickness variations in more detail by solving a Schrödinger equation. The problem of inhomogeneous broadening and Stokes shift is treated classically by Yang *et al.* [3]. There exist earlier works on the quantum mechanical treatment of disorder where exact solutions for certain one-dimensional problems were found [10]. A straightforward generalization to higher dimensions, however, is not possible.

In this paper we develop a quantum mechanical theory for the motion of a single particle in a random potential for arbitrary dimensionality. Explicit expressions are given for Gaussian randomness. The results are applied to excitons in quantum wells and are able to explain the asymmetry and the shift of the absorption profile.

## II. BASIC EQUATIONS

Let us consider a one-particle Hamiltonian $\hat{H}$ of the form

$$\hat{H} = \frac{\hat{p}^2}{2m} + V(\hat{\mathbf{r}}) \tag{1}$$

where $V$ is a random potential, and $\hat{\mathbf{r}}$ is a vector in an $n$-dimensional space.

The random potential is completely characterized by its correlation functions

$$\Psi^{(m)}(\mathbf{r}_1, \ldots, \mathbf{r}_m) = \langle\, V(\mathbf{r}_1) \cdots V(\mathbf{r}_m) \,\rangle \;, \tag{2}$$

where the brackets denote the configuration average. The function $\Psi^{(2)}$ is called autocorrelation function.

An important quantity is the probability density

$$p(E, \mathbf{r}) = \langle\, \delta\,[\, E - V(\mathbf{r})\,]\,\rangle \;. \tag{3}$$

The expression $dE\, p(E, \mathbf{r})$ can be interpreted as the probability of finding $V(\mathbf{r})$ in the interval $[\,E\,,\; E + dE\,]$. For physical reasons we assume macroscopic homogeneity, i. e., the correlation functions (2) are invariant under translation and the probability density (3) is independent of the space coordinate.

The one-particle Green's function in the momentum representation ($\mathbf{p} = \hbar\,\mathbf{k}$) is defined as:

$$G(\mathbf{k}, \mathbf{k}', z) = (H - \hbar z)^{-1}(\mathbf{k}, \mathbf{k}') \,. \tag{4}$$

Due to the assumed spatial homogeneity it holds that $\langle G(\mathbf{k},\mathbf{k}',z)\rangle = \delta_{\mathbf{k}\mathbf{k}'}\, g(\mathbf{k},z)$. If $V(\mathbf{r}) \equiv 0$ is assumed in the definition (1), the Green's function is given by $G^{(0)}(\mathbf{k},\mathbf{k}',z) = \delta_{\mathbf{k}\mathbf{k}'}\, g^{(0)}(\mathbf{k},z)$, where $g^{(0)}(\mathbf{k},z) = [\,\hbar^2 k^2/(2m) - \hbar z\,]^{-1}$.

The Dyson equation for $G$ yields:

$$g(\mathbf{k},z) = \sum_{m=0}^{\infty} (-1)^m\, g^{(m)}(\mathbf{k},z)\,, \tag{5}$$

where

$$g^{(m)}(\mathbf{k},z) = [\,g^{(0)}(\mathbf{k},z)\,]^2 \sum_{\mathbf{k}_1\cdots\mathbf{k}_{m-1}} g^{(0)}(\mathbf{k}_1,z)\cdots g^{(0)}(\mathbf{k}_{m-1},z)\,\times \tag{6}$$

$$\times \quad \langle\, V(\mathbf{k}-\mathbf{k}_1)\cdots V(\mathbf{k}_{m-1}-\mathbf{k})\,\rangle \quad \text{for} \quad m>0\,.$$

On the other hand, the configurationally averaged Green's function $g$ can be generated by a convolution of $g^{(0)}$ with a spectral function, $A$, according to

$$g(\mathbf{k},z) = \int_{-\infty}^{+\infty} d(\hbar\omega)\, A(\mathbf{k},\omega)\, g^{(0)}(\mathbf{k},z-\omega)$$

$$g(\mathbf{k},z) = \sum_{m=0}^{\infty} (-1)^m\, A_m(\mathbf{k})\,[\,g^{(0)}(\mathbf{k},z)\,]^{m+1}\,, \tag{7}$$

$$\text{where} \quad A_m(\mathbf{k}) = \int_{-\infty}^{+\infty} d(\hbar\omega)\,(\hbar\omega)^m\, A(\mathbf{k},\omega)\,.$$

The moments, $A_m$, of the spectral function can be uniquely determined by expanding the functions $g^{(m)}$ (6) in powers of the unperturbed Green's function $g^{(0)}$ and comparing the coefficients. Obviously, it holds that $A_0(\mathbf{k}) \equiv 1$ which means, physically, that the total oscillator strength is not changed by the fluctuations. The spectral function $A$ can be determined by

$$A(\mathbf{k},\omega) = \frac{1}{2\pi\hbar} \int_{-\infty}^{+\infty} dt\, e^{i\omega t} \sum_{m=0}^{\infty} \frac{A_m(\mathbf{k})(-it/\hbar)^m}{m\,!}\,. \tag{8}$$

In the next section we will give an explicit expression for a Gaussian stochastic potential.

## III. SOLUTION FOR GAUSSIAN RANDOMNESS

Now, we assume that the random potential (1) is given by a superposition of plane waves with definite amplitudes but random phases according to

$$V(\mathbf{k}) = \frac{1}{\sqrt{\Omega}}\, C(\mathbf{k})\, s(\mathbf{k})\,, \tag{9}$$

where the $C(\mathbf{k})$ are statistically independent quantities, equally distributed on the unit circle and $\Omega$ denotes the $n$-dimensional normalization volume. The fact that $V(\mathbf{r})$ is a real function requires that $s(\mathbf{k}) = s^*(-\mathbf{k})$ and $C(\mathbf{k}) = C^*(-\mathbf{k})$. The probability density is a Gaussian one, and the binary correlation function is given by the Fourier transform of $|s(\mathbf{k})|^2$.

The functions $g^{(m)}$ defined in Eq. (6) can now be visualized by diagrams, where the nodes mark the points $\mathbf{k}_1, \ldots, \mathbf{k}_{m-1}$, the straight lines stand for the potential $V(\mathbf{k}_i - \mathbf{k}_{i+1})$ and the dashed lines specify couples of vectors where $\mathbf{k}_i - \mathbf{k}_{i+1} = \mathbf{k}_{j+1} - \mathbf{k}_j$. In Fig. 1 all diagrams of the order $m = 4$ are shown.

In the zeroth order of approximation, all $(m-1)!!$ diagrams of $g^{(m)}$ have the same value and it holds that

$$A_m(\mathbf{k}) \equiv \begin{cases} (m-1)\,!!\,\sigma^m & \text{for } m \text{ even} \\ 0 & \text{for } m \text{ odd} \end{cases}$$

$$A(\mathbf{k},\omega) \equiv \frac{1}{\sqrt{2\pi\sigma^2}} \exp\left(\frac{\hbar^2\omega^2}{2\,\sigma^2}\right), \tag{10}$$

$$\text{where } \sigma^2 = \frac{1}{\Omega}\sum_{\mathbf{k}} |s(\mathbf{k})|^2 = \left\langle [V(\mathbf{k})]^2 \right\rangle .$$

As expected, the spectral function coincides with the probability density which can be determined directly from the definition (9). This result holds for arbitrary stochastic potentials.

The first order contributions of $g^{(m)}$ to the moment $A_{m+1}$ are given by the number of dashed lines that cross the nodes in all diagrams of the order $m$. By recursion this number can be found to be $m\,(m+1)\,!!\,/\,6$. Hence, we obtain

$$A_m(\mathbf{k}) \equiv \begin{cases} (m-1)\,!!\,\sigma^m & \text{for } m \text{ even} \\ -\frac{1}{6}\,(m-1)\,m!!\,\sigma^{m-3}\,\tau^3 & \text{for } m \text{ odd} \end{cases}$$

$$A(\mathbf{k},\omega) \equiv \left[1 - \frac{\tau^3}{6}\,\frac{d^3}{d(\hbar\omega)^3}\right] \frac{1}{\sqrt{2\pi\sigma^2}} \exp\left(\frac{\hbar^2\omega^2}{2\,\sigma^2}\right), \tag{11}$$

$$\text{where } \tau^3 = \frac{1}{\Omega}\sum_{\mathbf{k}} \frac{\hbar^2 k^2}{2m}|s(\mathbf{k})|^2 = \frac{\hbar^2}{2m}\left\langle |\nabla V(\mathbf{k})|^2 \right\rangle .$$

In this approximation, which is exact up to the third moment, the spectral function is independent of $\mathbf{k}$ and is determined by only two parameters, $\sigma$ and $\tau$, for the linewidth and asymmetry, respectively. These parameters reflect the probability distribution *and* the spatial correlation of the fluctuations.

In order to verify our analytical results, we have carried out numerical calculations for a Gaussian potential of the form (9) with

$$|s(\mathbf{k})|^2 = \gamma^2 \exp\left(-\frac{1}{2}\lambda^2 k^2\right) \tag{12}$$

in one dimension. The potential is characterized by a strength $\gamma$ and a correlation length $\lambda$. The explicit results are represented in units of $\hbar$, $m$, and $\lambda$.

In Fig. 2 both approximations (10) and (11) for the spectral function are compared for $\gamma = 1.0$ and $2.0$. For numerical reasons a homogeneous linewidth $\hbar\epsilon = 0.1$ is introduced. Whereas the zeroth order approximation can reproduce only the broadening, in the first order the agreement is excellent. The small parameter for the validity of the approximation (11) is $\tau/\sigma$ which is lower than 1 for the parameters used.

## IV. APPLICATION TO EXCITONS ON ROUGH INTERFACES

In this section we will apply our theory to the excitonic absorption of a quantum well with fluctuating thickness. The two-dimensional optical susceptibility of a quantum well is given by

$$\chi(z) = \frac{2|\mu|^2}{\varepsilon_0}\frac{1}{\Omega}\int d^2\mathbf{r}\int d^2\mathbf{r}'\,(H-\hbar z)(\mathbf{r},\mathbf{r},\mathbf{r}',\mathbf{r}')\,, \tag{13}$$

where $\mu$ is the dipole matrix element of the allowed optical transition and $\varepsilon_0$ is the vacuum dielectric constant. The Hamiltonian

$$\hat{H} = \frac{\hat{p}_\mathrm{e}^2}{2m_\mathrm{e}} + \frac{\hat{p}_\mathrm{h}^2}{2m_\mathrm{h}} + V_\mathrm{e}(\hat{\mathbf{r}}_\mathrm{e}) + V_\mathrm{h}(\hat{\mathbf{r}}_\mathrm{h}) - \frac{e^2/(4\pi\varepsilon_0\varepsilon)}{|\hat{\mathbf{r}}_\mathrm{e}-\hat{\mathbf{r}}_\mathrm{h}|}$$

describes the motion of an electron-hole pair in a plane under the influence of the Coulomb potential and stochastic potentials, $V_\mathrm{e}$ and $V_\mathrm{h}$, stemming from the fluctuations of the well thickness.

In the case of fluctuations small compared to the binding energy of the exciton, the eigenstates of the internal motion can be looked upon as decoupled. Then for each internal state with wavefunction $\varphi_n$ a stochastic potential

$$V_n(\mathbf{R}) = \int d^2\mathbf{r}\,|\varphi_n(\mathbf{r})|^2 \times \tag{14}$$

$$\times\left[V_\mathrm{e}(\mathbf{R}+\frac{m_\mathrm{h}}{m_\mathrm{e}+m_\mathrm{h}}\mathbf{r}) + V_\mathrm{h}(\mathbf{R}-\frac{m_\mathrm{e}}{m_\mathrm{e}+m_\mathrm{h}}\mathbf{r})\right]$$

can be defined. With this assumption, the optical susceptibility (13) takes the form

$$\chi(z) = -\frac{|\mu|^2}{\varepsilon_0}\sum_n |\varphi_n(\mathbf{0})|^2\, g_n(\mathbf{0},z)\,. \tag{15}$$

The $g_n$ are the averaged Green's functions (4) of a particle with mass $m_\mathrm{e}+m_\mathrm{h}$ under the influence of the potential $V_n$ (14).

In the remaining treatment we will assume electron-hole symmetry, i.e., $m_\mathrm{e}{=}m_\mathrm{h}$ and $V_\mathrm{e}(\mathbf{r}){=}V_\mathrm{h}(\mathbf{r}){=}\frac{1}{2}V(\mathbf{r})$. For a stochastic potential of the form (9) the coefficients of broadening and asymmetry for each exciton are given by

$$\sigma_n^2 = \frac{1}{\Omega}\sum_\mathbf{k} |s(\mathbf{k})|^2\,\Phi_n(\frac{1}{2}\mathbf{k}) \tag{16}$$

$$\tau_n^3 = \frac{1}{\Omega}\sum_\mathbf{k} \frac{\hbar^2k^2}{2m_\mathrm{e}+2m_\mathrm{h}}\,|s(\mathbf{k})|^2\,\Phi_n(\frac{1}{2}\mathbf{k})\,,$$

where $\Phi_n$ is the Fourier transform of the probability density $|\varphi_n(\mathbf{r})|^2$ of the $n$-th exciton. In the case of white Gaussian noise, i.e., $|s(\mathbf{k})|^2 \equiv |s|^2$, the lineshape is determined by the exciton wavefunction alone, and it holds

$$\sigma_n^2 = 4\,|s|^2\int d^2\mathbf{r}\,|\varphi_n(\mathbf{r})|^4 \tag{17}$$

$$\tau_n^3 = 16\,|s|^2 \int d^2\mathbf{r}\,|\varphi_n(\mathbf{r})|^2\,(-\frac{\hbar^2}{2m_e+2m_h})\,\Delta\,|\varphi_n(\mathbf{r})|^2$$

The $\varphi^4$-rule for $\sigma$ is already known [9].

By means of Eq. (13) and with our findings from the last section we are now able to calculate a whole absorption spectrum that is given by $\Im\,\chi(\omega + i\varepsilon)$. Here $\hbar\omega$ is the photon energy relative to the two-dimensional gap, and $\hbar\epsilon$ is the homogeneous broadening. Again, we assume a Gaussian autocorrelation function (12) All results are represented in excitonic units, the binding energy $E_B$ and the Bohr radius $a_B$ of the three-dimensional exciton.

Fig. 3 shows complete absorption spectra. The correlation length is fixed at $\lambda = 2.0\,a_B$, and a homogeneous broadening $\hbar\epsilon = 0.1 E_B$ is introduced. The values for $\gamma$ are 0.0 (a), 0.2 (b), and $0.4\,E_B\,a_B$ (c). For $\gamma = 0$ (a) the absorption line of the first exciton shows a Lorentzian broadening. The broadening increases for increasing $\gamma$ (b–c) and becomes a Gaussian one, leading to a decrease of the peak height compared to the continuum level that is hardly affected. For $\gamma > 0$ (b – c) the lines are asymmetric and show a redshift of the maximum and slower decay on the high-energy side.

## V. SUMMARY

We have developed a quantum mechanical theory to determine the single-particle Green's function under the influence of a stochastic potential. For Gaussian randomness, an analytical formula is given which approximates the numerical results very well. The theory is applied to excitons in quantum wells with fluctuating thickness. The basic features, the inhomogeneous broadening, the asymmetry of the lines and the shift of the absorption maximum can be explained.

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## FIGURES

FIG. 1: All fourth-order diagrams of the averaged Green's function. The nodes mark the integration variables $\mathbf{k}_1, \ldots, \mathbf{k}_3$. The solid lines correspond to the stochastic potential, and the dashed lines specify opposite arguments of the potentials.

FIG. 2: Comparison of the numerical solution (solid line) for the spectral function with the 0th and 1st order approximation (dashed line) for $\gamma = 2.0$ (a) and 4.0 (b).

FIG. 3: Imaginary part of the optical function vs. frequency in the region of the exciton bound states for $\gamma = 0.0$ (a) 0.2 (b) and $0.4\, E_{\mathrm{B}}\, a_{\mathrm{B}}$ (c). The correlation length of the potential fluctuations is fixed to be $\lambda_{\mathrm{c}} = 2.0\, a_{\mathrm{B}}$. A homogeneous broadening $\hbar\epsilon = 0.1\, E_{\mathrm{B}}$ is introduced.

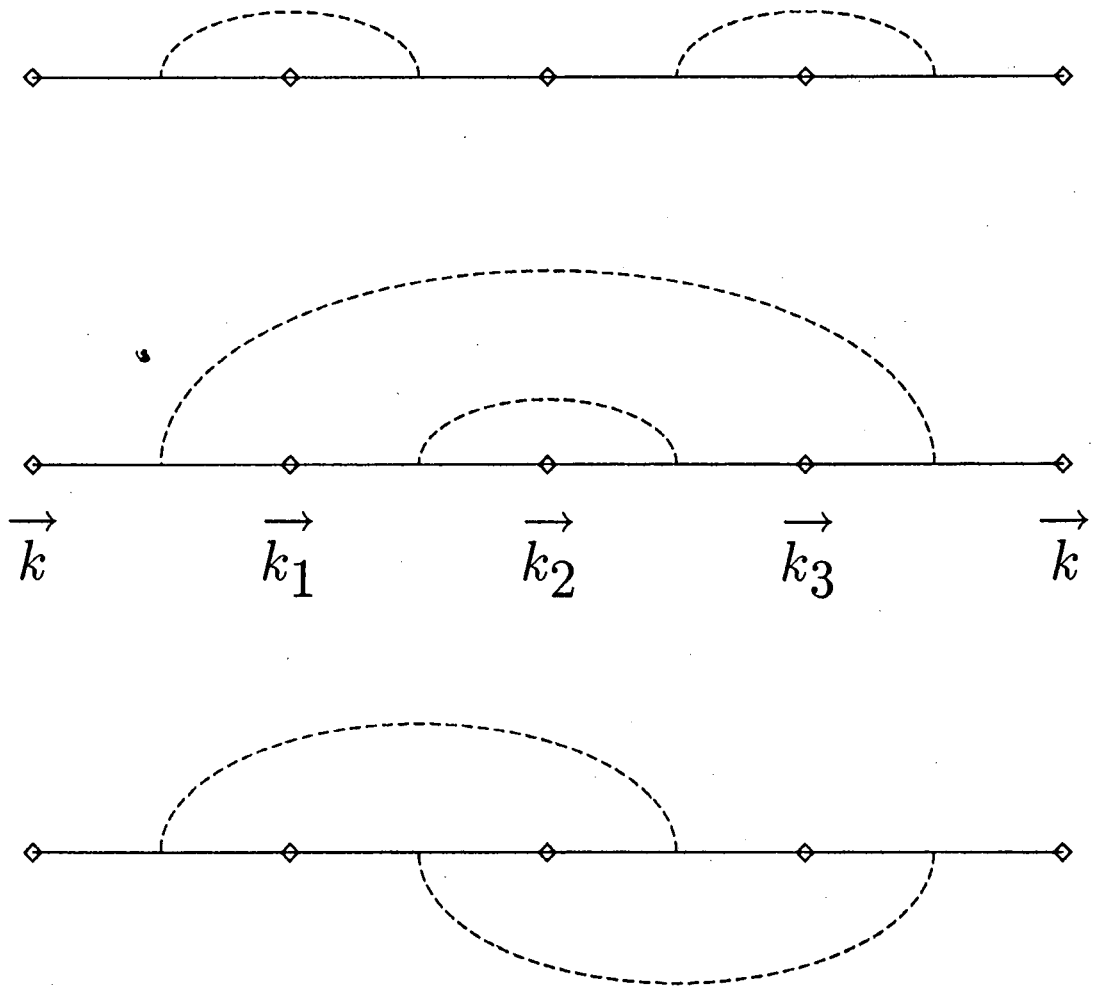


Figure 1

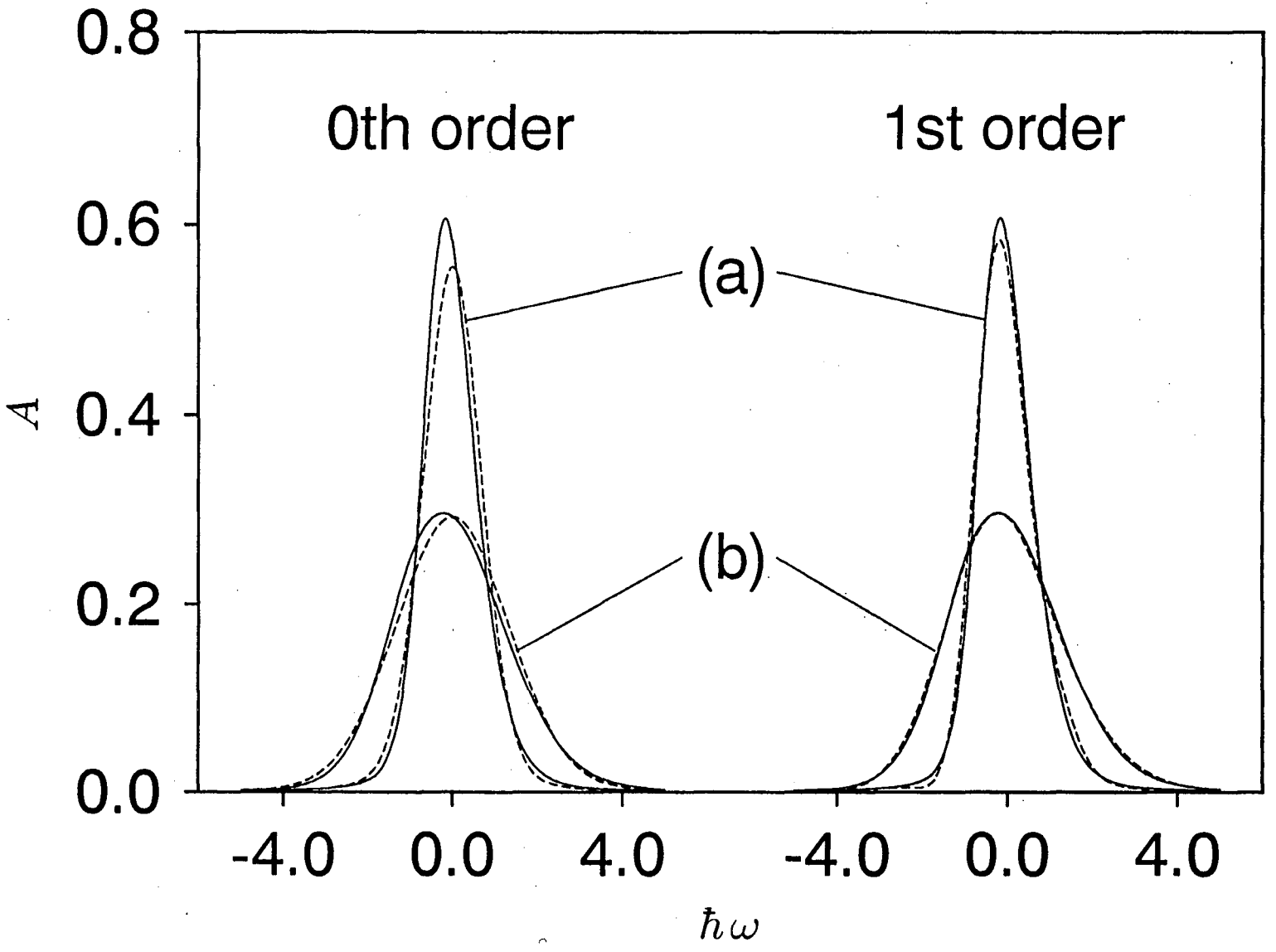


Figure 2

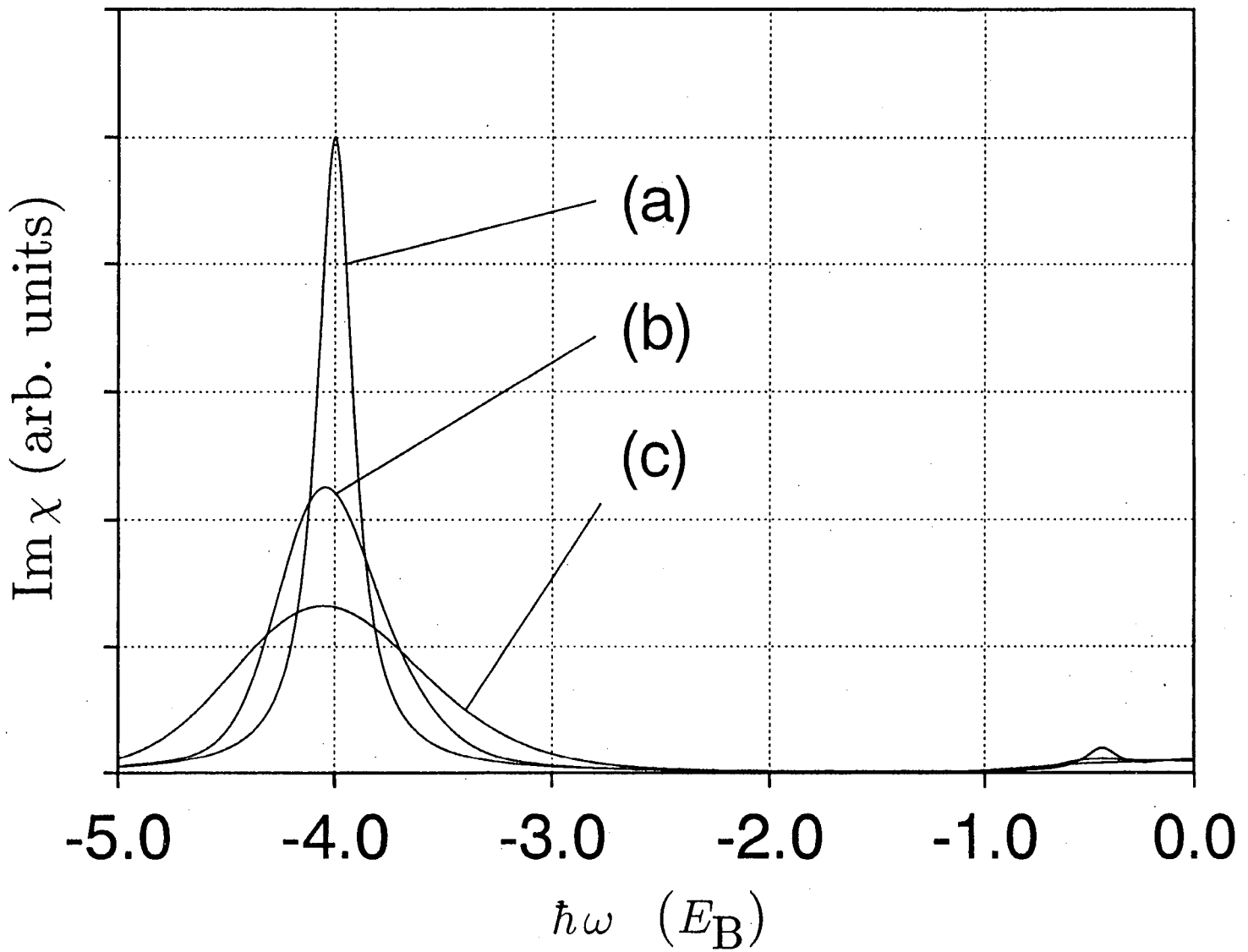


Figure 3

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