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Optical excitation of electron-hole pairs in disordered one-dimensional semiconductors

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We apply the optimal fluctuation method to the calculation of the optical absorption in disordered one-dimensional semiconductors below the fundamental optical gap. We find that a photon energy exists at which the shape of the optimal fluctuation undergoes a dramatic change, resulting in a different energy dependence of the absorption rate above and below this energy. In the limit when the interaction of an electron and a hole with disorder is stronger than their interaction with each other, we obtain an analytical expression for the optical conductivity. We show that to calculate the absorption rate, it is, in general, necessary to consider a manifold of optimal fluctuations, rather than just a single fluctuation. For an arbitrary ratio of the Coulomb interaction and disorder, the optimal fluctuation is found numerically.

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I. INTRODUCTION

The interplay between disorder and interparticle interactions results in a number of remarkable phenomena, e.g., a singularity in the electron density of states at the Fermi energy¹⁻⁴ and an enhanced localization length of pairs of interacting particles.⁵ A familiar situation in which the simultaneous presence of Coulomb interactions and disorder plays an important role, is the process of optical absorption in disordered semiconductors below the fundamental optical gap, i.e., below the band-to-band transition.⁶ With the advent of modern optical materials, this problem is not only of interest in its three-dimensional version, for which it has received most attention, but also in two dimensions (quantum wells^{7,8}) and one dimension (quantum wires^{7,8} and semiconducting polymers⁹). Interestingly, in the absorption spectrum the relative strength of the Coulomb interaction between the electron and hole and their interactions with the disorder, does not only depend on the disorder strength, but also on the photon energy: the lower the photon energy, the larger the amplitude of a disorder fluctuation should be in order to create the corresponding absorbing state below the fundamental optical gap.

The effect of a relatively weak disorder on the Wannier exciton was considered in Refs. 10 and 11. Here, relatively weak refers to the situation where the exciton absorption peak is still visible as a separate peak below the band-to-band transition and is not smeared entirely by the disorder. Moreover, the restriction to weak disorder only applies for photon energies close to the exciton energy in the absence of disorder E_{ex} . In this case, the exciton localization length is much larger than the average electron-hole separation, and the exciton center-of-mass motion decouples from the relative motion of the electron and hole. The Wannier exciton then essentially behaves as a Frenkel exciton in an effective disorder potential and the calculation of the optical absorption spectrum becomes a single-particle problem. This effective approach has been used to study numerically the absorption and luminescence line shapes due to excitons in semiconductor quantum wells with interface roughness.¹²

In Refs. 10 and 11, the low-energy tail of the exciton absorption peak was calculated analytically within the effec-

tive one-particle approach, by using the optimal fluctuation method.^{13,14} This method applies when the dominant contribution to the quantity of interest comes from disorder realizations close to a single large disorder fluctuation. This method can be formulated as a saddle-point calculation of the functional integration over disorder realizations.¹⁵ It was used previously to calculate the optical absorption in disordered Peierls conductors¹⁶ and it is similar to the calculation of the Urbach tails resulting from the interaction of the electron-hole pair with the lattice, treated in the quasistatic approximation.¹⁷

In this paper we apply the optimal fluctuation method to the general case of arbitrary ratio of disorder strength and Coulomb interaction. We show that the shape of the optimal fluctuation crucially depends on the dimensionality of the system and we consider in detail the one-dimensional case. In the strong disorder limit, when the exciton peak is destroyed, and for weak disorder, assuming sufficiently low photon energies, we obtain an analytical expression for the optical conductivity. We show that in these situations, the electron and hole in the optimal fluctuation are localized in two separate potential wells. We also show that in order to calculate the optical absorption rate, one has, in general, to consider a manifold of optimal fluctuations with different values of the electron-hole separation, rather than just one fluctuation.

In the general case of arbitrary ratio of the Coulomb interaction and disorder strength the equation for the optimal fluctuation is solved numerically. We do this for an electron and hole described within the one-dimensional tight-binding model. The equation for the optimal fluctuation in this model is similar to the equation for polaronic excitons and bipolarons obtained in the adiabatic approximation.¹⁸⁻²⁰ We show that, at some energy E_c , lying in the region where the Coulomb interaction and disorder are of the same order, the shape of the optimal fluctuation undergoes a sudden change: while above E_c it has the form of a single potential well, below E_c it consists of a “dip” that localizes the hole and a “bump” that localizes the electron. This transition takes place only in one-dimensional systems.

This paper is organized as follows. In Sec. II we introduce

the continuum model that describes interacting electron-hole pairs in disordered semiconductors. In Sec. III we briefly recall the application of the optimal fluctuation method to the calculation of the one-particle density of states and derive some basic expressions for later use. Then, in Sec. IV, we will discuss the shape of the optimal fluctuation for the electron-hole states, in particular, its dependence on the dimensionality of the system. We obtain and solve perturbatively the nonlinear nonlocal equation for the optimal fluctuation in the one-dimensional case. In Sec. V we obtain an expression for the tails of the optical absorption spectrum, which has a wider range of validity than the optimal fluctuation method. In Sec. VI we present and discuss our numerical results for the optimal fluctuation. Finally, we summarize and conclude in Sec. VII. Some technical details have been moved to the Appendix in order not to disturb the natural flow of the text.

II. CONTINUUM MODEL AND OPTICAL ABSORPTION

We consider direct-gap semiconductors with Wannier excitons, in which case the low-energy electron-hole states can be described in the continuum approximation.²¹ In this approximation, the wave function $\Psi_\alpha(\mathbf{x}_1, \mathbf{x}_2)$ of the electron-hole pair with the energy E_α , counted from the gap value Δ , satisfies the Schrödinger equation,

$$\left[-\frac{\hbar^2}{2m_h}\Delta_1 - \frac{\hbar^2}{2m_e}\Delta_2 + U(\mathbf{x}_1) - \beta U(\mathbf{x}_2) + V(\mathbf{x}_1 - \mathbf{x}_2) \right] \Psi_\alpha = E_\alpha \Psi_\alpha. \quad (1)$$

Here, \mathbf{x}_1 and \mathbf{x}_2 are the coordinates of, respectively, the hole and electron, $m_h(m_e)$ is the effective hole(electron) mass, $V(\mathbf{x}) = -e^2/\epsilon x$ is the Coulomb interaction between the electron and hole, $U(\mathbf{x})$ is the random potential due to impurities acting on the hole, while the disorder potential acting on the electron is $-\beta U(\mathbf{x})$. The dimensionless coefficient β accounts for a different dependence of the energies of the bottom of the conduction band and the top of the valence band on the concentration of impurities.¹¹ We consider here the case of uncorrelated white noise disorder of strength A

$$\langle U(\mathbf{x})U(\mathbf{x}') \rangle = A \delta(\mathbf{x} - \mathbf{x}'). \quad (2)$$

The optical conductivity per unit volume for the electric field, polarized, e.g., in the x direction, is given by

$$\sigma(\omega) = \frac{C}{\omega} F(\hbar\omega - \Delta), \quad (3)$$

where the coefficient C ,

$$C = \frac{2\pi e^2 \hbar^2}{m^2} \left| \int_{V_0} d^3r u_0^*(\mathbf{r}) \frac{\partial}{\partial x} v_0(\mathbf{r}) \right|^2, \quad (4)$$

is expressed through the periodic Bloch waves, $u_0(\mathbf{r})$ and $v_0(\mathbf{r})$, describing, respectively, the electron and hole states with zero wave vector. In Eq. (4) m is the electron mass in

vacuum and the integration goes over one elementary unit cell V_0 , in which the functions $u_0(\mathbf{r})$ and $v_0(\mathbf{r})$ are normalized to unity.

The function F in Eq. (3) is the part of the optical conductivity that has to be calculated within the continuum model

$$F(\hbar\omega - \Delta) = \frac{1}{V} \left\langle \sum_\alpha |D_{\alpha 0}|^2 \delta(\Delta + E_\alpha - \hbar\omega) \right\rangle. \quad (5)$$

In the last equation

$$D_{\alpha 0} = \int d^d x \Psi_\alpha^*(\mathbf{x}, \mathbf{x}) \quad (6)$$

is the ‘‘continuum part’’ of the matrix element of the transition from the ground state to the excited state α [see Eq. (1)]. In Eq. (5) the brackets $\langle \dots \rangle$ denote the disorder average, $V = L^d$ is the total volume, L is the linear size, and d is the dimensionality of the system.

III. THE ONE-PARTICLE CASE

In this section we briefly recall how the optimal fluctuation method can be used to calculate the low-energy tail of the density states (per unit volume)^{13–15}

$$\rho(\varepsilon) = \frac{1}{V} \left\langle \sum_\alpha \delta(\varepsilon_\alpha[U] - \varepsilon) \right\rangle \quad (7)$$

of a single particle moving in a random potential

$$H\psi_\alpha = \left(-\frac{\hbar^2}{2m}\Delta + U(\mathbf{x}) \right) \psi_\alpha = \varepsilon_\alpha \psi_\alpha, \quad \alpha = 0, 1, 2, \dots \quad (8)$$

We are now interested in the density of states with a large negative energy. Such states can only be induced by large negative fluctuations of the disorder potential $U(x)$ (in the absence of disorder the energy of all eigenstates is positive). The density of states is then, essentially, the probability to find such a fluctuation. When this probability is small, it suffices to keep in Eq. (7) only the contribution of the ground state ($\alpha = 0$),

$$\rho(\varepsilon) \approx \frac{1}{V} \langle \delta(\varepsilon_0[U] - \varepsilon) \rangle, \quad (9)$$

because the probability to find a disorder fluctuation that induces an excited state with the same energy is even smaller.

For the white noise potential Eq. (2) the disorder average can be performed by functional integration,

$$\begin{aligned} \rho(\varepsilon) &= \frac{1}{V} \int \mathcal{D}U e^{-S/A} \delta(\varepsilon_0[U] - \varepsilon) \\ &= \frac{1}{V} \int \mathcal{D}U \frac{d\lambda}{2\pi i A} \exp\left(-\frac{1}{A} [S + \lambda(\varepsilon_0[U] - \varepsilon)] \right), \end{aligned} \quad (10)$$

where the ‘‘action’’ S is given by

$$S = \frac{1}{2} \int d^d x U^2(\mathbf{x}). \quad (11)$$

The optimal fluctuation method is the saddle-point calculation of the functional integral in Eq. (10), in which one assumes that the dominant contribution to this integral comes from the vicinity of one “optimal” disorder fluctuation $U(\mathbf{x})$, which has the highest weight among the disorder realizations that induce a state at the energy ε . At the “saddle-point” the variation of

$$S_\lambda = S + \lambda(\varepsilon_0[U] - \varepsilon) \quad (12)$$

with respect to $U(\mathbf{x})$ vanishes. This gives

$$U(\mathbf{x}) = -\lambda \frac{\delta \varepsilon_0[U]}{\delta U(\mathbf{x})} = -\lambda \psi_0^2(\mathbf{x}), \quad (13)$$

where $\psi_0(\mathbf{x})$ is the ground-state wave function, which we can take to be real. Inserting Eq. (13) into the Schrödinger equation (8), we obtain a nonlinear equation for ψ_0 as

$$\frac{\hbar^2}{2m} \Delta \psi_0 + \lambda \psi_0^3 + \varepsilon \psi_0 = 0. \quad (14)$$

It is convenient to introduce the dimensionless coordinate $\mathbf{z} = \kappa \mathbf{x}$, where κ is defined by $\varepsilon = -\hbar^2 \kappa^2 / 2m$ and the dimensionless wave function $\phi(\mathbf{z})$

$$\psi_0(\mathbf{x}) = \frac{\hbar \kappa}{\sqrt{\lambda m}} \phi(\mathbf{z}), \quad (15)$$

which satisfies

$$\Delta_z \phi + 2\phi^3 - \phi = 0. \quad (16)$$

In dimensionless units the spatial extent of the wave function and the optimal fluctuation is of the order of one. For $d=1$, the solution of Eq. (16) is

$$\phi(z) = \frac{1}{\cosh z}, \quad (17)$$

while for $d=2,3$ this equation has to be solved numerically. The dependence of the function ϕ on the radius $z=|\mathbf{z}|$ is shown in Fig. 1.

The solution of Eq. (16) is an extremum of the functional

$$\mathcal{A}[\phi] = \frac{1}{a} \int d^d z \left[(\nabla_z \phi)^2 + \phi^2 - \frac{1}{2a} \phi^4 \right], \quad (18)$$

where $a = \int d^d z \phi^2$. The coefficient λ in Eq. (13), found using the normalization condition $\int d^d x \psi_0^2(\mathbf{x}) = 1$ and Eq. (15), is expressed through a by

$$\lambda = a \frac{\hbar^2 \kappa^{2-d}}{m}$$

and the action S , defined by Eq. (11), for the optimal fluctuation is given by

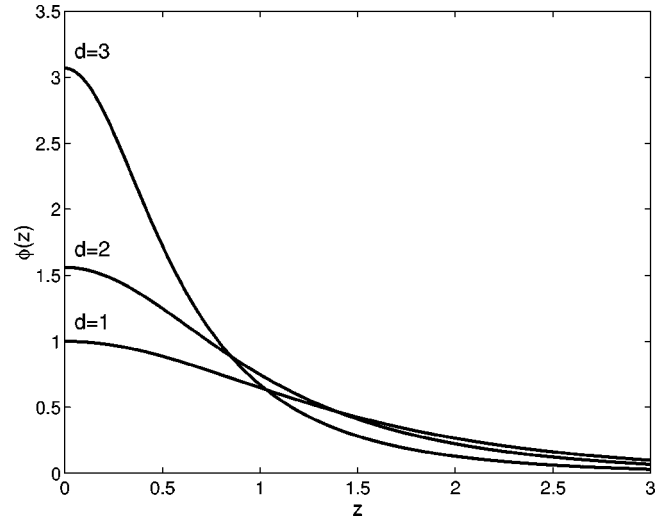


FIG. 1. The shape of the wave function $\phi(z)$ for $d=1,2,3$.

$$S = 2^{1-d/2} b \frac{\hbar^d |\varepsilon|^{2-d/2}}{m^{d/2}}, \quad (19)$$

where

$$b = \int d^d z \phi^4 = \frac{2}{(4-d)} a.$$

The value of the coefficient a (see, e.g., Refs. 22 and 23) is

$$a = \begin{cases} 2 & \text{for } d=1 \\ 5.85 \dots & \text{for } d=2 \\ 6.30 \dots & \text{for } d=3. \end{cases}$$

The result for the single-particle density of states at large negative energies, obtained by the optimal fluctuation method, has the form

$$\rho(\varepsilon) = K(\varepsilon) e^{-S/A}, \quad (20)$$

where the prefactor $K(\varepsilon)$ results from the Gaussian integrations over the small deviations of $\delta U(x)$ from the optimal fluctuation. The calculation of the prefactor also involves the integration over the locations of the optimal fluctuation, which cancels the volume V in the denominator of Eq. (7). For $d=1$, the prefactor $K(\varepsilon)$ is given by¹³

$$K(\varepsilon) = \frac{4|\varepsilon|}{\pi A}. \quad (21)$$

IV. OPTIMAL FLUCTUATION FOR ELECTRON-HOLE STATES

We now return to the problem of an interacting electron-hole pair in the presence of disorder. Similar to the preceding section, we can obtain an equation for the wave function of the typical electron-hole state with large negative energy. However, before turning to a formal consideration [see Eqs. (29) and further], we first give a qualitative discussion of the

properties of the optimal disorder fluctuation for the excitonic states.

From Eq. (13) we see that the spatial extent of the disorder fluctuation, which induces the typical single-particle state with the negative energy ε , equals the spatial extent of this state, given by $\kappa^{-1}\propto|\varepsilon|^{-1/2}$. Similarly, for the electron-hole state with negative energy E , the spatial extent of the optimal fluctuation $r\propto|E|^{-1/2}$. While the magnitude of the disorder potential $\propto|E|$, the magnitude of the Coulomb energy of the electron-hole pair $\propto r^{-1}\propto|E|^{1/2}$. Thus, for large negative E , the Coulomb energy becomes smaller than the energy of the interaction of the electron and hole with disorder and the Coulomb interaction can be treated perturbatively. This situation is very similar to that of the Coulomb gas, which becomes more ideal as its density increases.²⁴

The shape of the typical electron-hole state crucially depends on the dimensionality of the system d . We first neglect the electron-hole interaction completely and consider a localized hole with the energy $\varepsilon_h < 0$ and a localized electron with the energy $\varepsilon_e < 0$, such that $\varepsilon_h + \varepsilon_e = E$. Assuming that the electron and hole are localized far from each other, the action of the corresponding disorder fluctuation is given by the sum of the single-particle actions [see Eq. (19)],

$$S = S_h(\varepsilon_h) + S_e(\varepsilon_e) = 2^{1-d/2} b \hbar^d \left(\frac{|\varepsilon_h|^{2-d/2}}{m_h^{d/2}} + \frac{|\varepsilon_e|^{2-d/2}}{\beta^2 m_e^{d/2}} \right). \quad (22)$$

The factor β^2 in the electron action is due to the fact that the strength of the disorder potential, acting on the electron, is $\beta^2 A$.

For $d=3$ the minimum of the action is reached when only one particle is localized, i.e., at

$$\begin{aligned} \varepsilon_h = E, \quad \varepsilon_e = 0 & \quad \text{for } m_h > m_e \beta^{4/3}, \\ \varepsilon_h = 0, \quad \varepsilon_e = E & \quad \text{for } m_h < m_e \beta^{4/3}. \end{aligned}$$

If we now include the Coulomb interaction between the electron and hole, we obtain the following picture of the typical electron-hole state with a large negative energy: One particle is localized by a disorder fluctuation, while the other particle forms a bound hydrogenlike state with the localized particle. If, e.g., the hole is localized, then the total pair energy is

$$E \approx \varepsilon^{(1)} + \varepsilon^{(2)} + \varepsilon^{(3)}, \quad (23)$$

where $\varepsilon^{(1)} = -\hbar^2 \kappa_h^2 / 2m_h$ is the energy of the localized hole, the second term is the binding energy of the electron, $\varepsilon^{(2)} = -m_e e^4 / 2\epsilon^2 \hbar^2$, and the third term is the energy of the repulsion of the electron from the disorder potential that localizes the hole. This picture is valid when $|\varepsilon^{(2)}| \ll |\varepsilon^{(1)}|$, in which case the size of the hydrogenlike state $r = \epsilon(m/m_e) a_B$ (here a_B is the Bohr radius) is much larger than spatial extent of the disorder fluctuation localizing the hole κ_h^{-1}

$$\kappa_h r = \sqrt{\frac{m_h \varepsilon^{(1)}}{m_e \varepsilon^{(2)}}} \gg 1.$$

In this limit the disorder potential acting on the electron can be approximated by a δ function and the corresponding energy $\varepsilon^{(3)} \approx 50(m_e/m_h)^{3/2} |\varepsilon^{(2)}|^{3/2} / |E|$ is parametrically smaller than $|\varepsilon^{(2)}|$ and can be neglected. Then the absorption rate for $d=3$ is

$$F(E) \propto e^{-S_h/A} = \exp \left\{ -\frac{b \hbar^3}{2^{1/2} m_h^{3/2} A} \left| E + \frac{m_e e^4}{2\epsilon^2 \hbar^2} \right|^{1/2} \right\}. \quad (24)$$

For $d=2$, the situation is, essentially, the same as for $d=3$: the minimization of the action (22) gives that in the optimal fluctuation only one of the two particles is localized by disorder (the hole, for $m_h > \beta^2 m_e$, or the electron, otherwise). For $d=1$ the situation is quite different. In that case the minimum of the action (22) is reached at

$$\frac{\varepsilon_e}{\varepsilon_h} = \beta^4 \frac{m_e}{m_h}, \quad (25)$$

which means that in the one-dimensional electron-hole state with large negative energy both the electron and the hole are likely to be localized. In that case the optimal fluctuation has two parts [cf. Figs. 4(b) and 4(c) below]: a part where the disorder potential is negative (to localize the hole) and a part with positive disorder potential (to localize the electron). The minimal value of the action is then given by

$$S_0 = \frac{4\hbar}{3} \sqrt{\frac{2|E|^3}{M}}, \quad (26)$$

where $M = m_h + \beta^4 m_e$.

There are two kinds of corrections to this action: (i) the correction due to the electron-hole interaction and (ii) the correction due to the interaction of the electron with the disorder fluctuation localizing the hole and vice versa. For large negative E these corrections are relatively small. The regular way to find them is the perturbative solution of the equation for the typical wave function of the electron-hole state of a given energy. This equation can be obtained using the same considerations that led to Eq. (14). The relation between the optimal fluctuation $U(\mathbf{x})$ and the wave function of the electron-hole state $\Psi(\mathbf{x}_1, \mathbf{x}_2)$ it induces, has the form

$$U(\mathbf{x}) = -\lambda \int d^d x' [\Psi^2(\mathbf{x}, \mathbf{x}') - \beta \Psi^2(\mathbf{x}', \mathbf{x})], \quad (27)$$

where we used that Ψ satisfies Eq. (1). Substituting this

optimal fluctuation back into the Schrödinger equation (1) leads to a nonlinear, nonlocal equation for Ψ ,

$$\begin{aligned} & \left(-\frac{\hbar^2}{2m_h} \Delta_1 - \frac{\hbar^2}{2m_e} \Delta_2 + V(\mathbf{x}_2 - \mathbf{x}_1) - E \right) \Psi(\mathbf{x}_1, \mathbf{x}_2) \\ &= \lambda \Psi(\mathbf{x}_1, \mathbf{x}_2) \int d^d x' [\Psi^2(\mathbf{x}_1, \mathbf{x}') - \beta \Psi^2(\mathbf{x}', \mathbf{x}_1) \\ & \quad - \beta \Psi^2(\mathbf{x}_2, \mathbf{x}') + \beta^2 \Psi^2(\mathbf{x}', \mathbf{x}_2)]. \end{aligned} \quad (28)$$

This equation is formally equivalent to the equation for the excitonic polaron and bipolaron wave functions, obtained in the adiabatic treatment of the lattice.^{18–20} It is clearly impossible to solve this equation analytically. In the remainder of this section we obtain the action of the optimal fluctuation for $d=1$ using a perturbative solution of this equation, while in Sec. VI we give the results of a numerical solution, also for $d=1$.

The first term in the perturbative expansion of the wave function

$$\Psi = \Psi_0 + \Psi_1 + \dots$$

is the product of the wave functions of a noninteracting electron-hole pair separated by some distance

$$\begin{aligned} \Psi_0(x_1, x_2) &= \psi_h(x_1 - x_h) \psi_e(x_2 - x_e) \\ &= \frac{\sqrt{\kappa_h \kappa_e}}{2} \phi(\kappa_h[x_1 - x_h]) \phi(\kappa_e[x_2 - x_e]), \end{aligned} \quad (29)$$

where $x_h(x_e)$ are the average hole (electron) positions, the single-particle wave function ϕ is given by Eq. (17), and the wave vectors κ_h and κ_e are defined by $\varepsilon_h = -\hbar^2 \kappa_h^2 / 2m_h$ and $\varepsilon_e = -\hbar^2 \kappa_e^2 / 2m_e$.

In the zeroth order of the expansion the disorder potential in the optimal fluctuation is obtained by substituting Eq. (29) into Eq. (27),

$$\begin{aligned} U_0(x) &= U_h(x - x_h) + U_e(x - x_e) \\ &= -\lambda \psi_h^2(x - x_h) + \beta \lambda \psi_e^2(x - x_e), \end{aligned} \quad (30)$$

where the first term is the negative potential (a dip) that localizes the hole and the second term is the positive potential (a bump) that localizes the electron.

Inserting Eqs. (29) and (30) into Eq. (28) and taking into account that $\varepsilon_h + \varepsilon_e = E$, we obtain that the cancellation of the zeroth-order terms in that equation requires $\lambda = 2\hbar^2 \kappa_h / m_h$ and $\lambda \beta^2 = 2\hbar^2 \kappa_e / m_e$, from which we find

$$\begin{aligned} \varepsilon_h &= \frac{m_h}{M} E, \\ \varepsilon_e &= \frac{\beta^4 m_e}{M} E, \\ \lambda &= 2\hbar \sqrt{\frac{2|E|}{M}}, \end{aligned} \quad (31)$$

which agree with Eq. (25) obtained above. Also, the calculation of the action in the zeroth-order approximation

$$S_0 = \frac{1}{2} \int dx [U_h^2(x) + U_e^2(x)]$$

gives Eq. (26).

Though the calculation of the first-order correction to the wave function is, in general, difficult, the first-order correction to the action can be expressed through the unperturbed wave functions of the electron and hole,

$$\begin{aligned} S_1(r) &= \lambda \int dx \psi_h^2(x) \left[\int dx' \psi_e^2(x') V(x - x' + r) \right. \\ & \quad \left. + \beta \lambda \psi_e^2(x - r) \right], \end{aligned} \quad (32)$$

where $r = x_e - x_h$ is the average electron-hole separation. The somewhat lengthy derivation of this result is given in the Appendix. The first term in the square brackets is the correction due to the electron-hole interaction, while the second term describes the interaction of the hole with the bump, localizing the electron and vice versa [see Eq. (30)].

The optimal electron-hole distance r_* is found by minimizing S_1 with respect to r ,

$$\left. \frac{dS_1}{dr} \right|_{r=r_*} = 0. \quad (33)$$

From the form of Eq. (32) it is clear that the latter condition is just the balance of average forces acting on the hole: the attraction from the electron and repulsion from the disorder potential localizing the electron. Thus the typical electron-hole state of large negative energy can be considered as a kind of “molecule” in which the disorder fluctuations binding the electron and the hole play the role of “nuclei” with, respectively, positive and negative charge.

The result Eq. (32) can be cast into a more transparent form using the fact that for the Coulomb interaction between the electron and hole to be small compared to their interaction with the disorder fluctuations, the optimal electron-hole separation r_* should be large compared to the spatial extent of the disorder fluctuations: $e^{-\kappa_h r_*}, e^{-\kappa_e r_*} \ll 1$. We furthermore assume that $m_h > m_e \beta^4$ and that $e^{-(\kappa_h - \kappa_e) r_*} \ll 1$, i.e., the hole is sufficiently more localized than the electron, which holds unless m_h is very close to $m_e \beta^4$. Then Eq. (32) becomes

$$\frac{S_1(r)}{\lambda} \approx -\frac{e^2}{\epsilon r} + \beta \lambda \psi_e^2(r). \quad (34)$$

For the optimal distance r_* , at which S_1 has its minimum, we obtain

$$\kappa_e r_* e^{-\kappa_e r_*} = \sqrt{\frac{e^2}{4\epsilon \beta \lambda}}, \quad (35)$$

where λ is given by Eq. (31). The dimensionless parameter on the right-hand side is, essentially, the square root of the

ratio of the Coulomb energy to the total energy E , which, by assumption, is small (and thus $\kappa_e r_*$ is logarithmically large).

Furthermore, the second term in Eq. (34) (due to repulsion of the hole from the electron optimal fluctuation) is small compared to the first term (due to the electron-hole interaction),

$$\frac{\beta\lambda\psi_e^2(r_*)}{e^2/(\epsilon r_*)} \approx \frac{1}{2\kappa_e r_*}.$$

Therefore,

$$S_1 \approx -\lambda \frac{e^2}{\epsilon r_*} = \frac{dS_0}{dE} \frac{e^2}{\epsilon r_*},$$

where in the last step we used

$$\lambda = -\frac{dS_0}{dE}, \quad (36)$$

as follows from Eqs. (26) and (31). We thus see that, to the lowest order, the effect of the correction to the action S_1 is to replace $S_0(E)$ by $S_0(E + e^2/\epsilon r_*)$, giving for the absorption rate in one dimension

$$F(E) \propto \exp\left\{-\frac{4\sqrt{2}\hbar}{3AM^{1/2}}\left|E + \frac{e^2}{\epsilon r_*}\right|^{3/2}\right\}, \quad (37)$$

where the electron-hole Coulomb shift depends on E , as according to Eq. (35), $r_* \propto |E|^{-1/2} \ln|E|$.

To end this section about the shape of the optimal fluctuation we note that the numerical solution of Eq. (33) for sufficiently small $|E|$ gives $r_* = 0$, which corresponds to the localization of the electron-hole pair by a symmetric single-well disorder potential. In that case the repulsion of the electron from the disorder potential that localizes the hole is compensated by the Coulomb interaction between the electron and hole. The transition from the symmetric to the asymmetric shape of the optimal fluctuation is studied in detail in Sec. VI, where the results of the numerical solution of Eq. (28) are discussed. We shall show that our analytical approach, in fact, gives a rather accurate description of that transition (see Fig. 5).

V. OPTICAL ABSORPTION TAIL IN ONE DIMENSION

In the precedings section we have shown that for $d=1$ the electron and hole in the typical state with a large negative energy are localized relatively far from each other. This property allowed us to calculate approximately the weight of the optimal disorder fluctuation for the electron-hole pair. It also simplifies the calculation of the preexponential factor in the expression for the absorption rate, which results from the integration over small deviations from the optimal disorder fluctuation. In this section we obtain an analytical expression for the tail of the optical absorption spectrum that actually has a wider range of validity than the standard optimal fluctuation method.

As we have shown in the preceding section, the dominant contribution to the optical absorption at large negative E

comes from the disorder realizations that are close to the sum of the single-particle optimal fluctuations, $U_0(x) = U_h(x - x_h) + U_e(x - x_e)$ [see Eq. (30)], where $U_h(x - x_h)$ localizes the hole with energy ϵ_h near $x = x_h$ and $U_e(x - x_e)$ localizes the electron with the energy ϵ_e near $x = x_e$. In the preceding section we have calculated the optimal electron-hole separation, for which the weight of the disorder fluctuation reaches its maximum. In this section we shall treat x_h and x_e , as well as the single-particle energies ϵ_h and ϵ_e , as the collective variables of the functional integration over disorder. Performing the Gaussian integration over all the small deviations $\delta U(x) = U(x) - U_0(x)$ in Eq. (5), which are orthogonal to the deviations corresponding to the four collective coordinates, we obtain

$$\begin{aligned} F(E) \approx & \frac{1}{L} \int d\epsilon_h d\epsilon_e dx_h dx_e K_h(\epsilon_h) K_e(\epsilon_e) D^2(x_e - x_h) \\ & \times \exp\left\{-\frac{1}{A}[S_h(\epsilon_h) + S_e(\epsilon_e) + \delta S]\right\} \\ & \times \delta(\epsilon_h + \epsilon_e + \delta\epsilon - E). \end{aligned} \quad (38)$$

Here, L is the chain length, D is the transition matrix element [see Eq. (6)], $S_h(\epsilon_h)$ and $S_e(\epsilon_e)$ are the single-particle actions, given by Eq. (22), and $\delta S = \delta S(r)$, where $r = x_e - x_h$, is the correction to the action of the electron-hole pair due to the overlap between the electron and hole optimal fluctuation

$$\delta S(r) = \int dx U_h(x - x_h) U_e(x - x_e).$$

Furthermore, $\delta\epsilon = \delta\epsilon(r)$ is the energy correction due to the electron-hole interaction and the interaction of the electron with the hole optimal fluctuation and vice versa. The first-order correction, calculated using the unperturbed electron-hole wave function Eq. (29), is

$$\begin{aligned} \delta\epsilon(r) = & \int dx [\psi_h^2(x - x_h) U_e(x - x_e) \\ & + \psi_e^2(x - x_e) U_h(x - x_h)] \\ & + \int dx dx' \psi_h^2(x - x_h) V(x - x') \psi_e^2(x' - x_e), \end{aligned} \quad (39)$$

where $r = x_e - x_h$ is the electron-hole separation. In the same approximation the transition matrix element is given by

$$D(r) = \int dx \psi_h(x - x_h) \psi_e(x - x_e).$$

Finally, in Eq. (38) we used the fact that for small overlap between the electron and hole optimal fluctuations the prefactor, resulting from the Gaussian integration over $\delta U(x)$, is the product of prefactors for isolated the hole and electron, $K_h(\epsilon_h) K_e(\epsilon_e)$.²⁵ [We note, that in $K_e(\epsilon_e)$ the disorder strength A has to be substituted by $\beta^2 A$.]

The integration over the center-of-mass coordinate

$$R = \frac{m_h x_h + m_e x_e}{m_h + m_e},$$

which is a zero mode, gives the chain length L . Due to the δ function in Eq. (38) $\varepsilon_e = E - \delta\varepsilon - \varepsilon_h$ and the remaining integration over the hole energy ε_h can be performed in the saddle-point approximation. The condition of the minimum of $S_h(\varepsilon_h) + S_e(\varepsilon_e)$ gives Eq. (25), which determines the saddle-point values of ε_h and ε_e . Then Eq. (38) can be written as follows:

$$F(E) \approx \sqrt{\frac{2\pi A}{B}} \int dr K_h(\varepsilon_h) K_e(\varepsilon_e) D^2(r) \times \exp\left\{-\frac{1}{A}[S_0(E - \delta\varepsilon) + \delta S]\right\}, \quad (40)$$

where

$$B = \frac{d^2}{d\varepsilon_h^2} [S_h(\varepsilon_h) + S_e(E - \varepsilon_h)] = \frac{\hbar}{\mu} \sqrt{\frac{2M}{|E|}},$$

$\mu = \beta^4 m_e m_h / (m_h + \beta^4 m_e)$, and $S_0(E)$ is given by Eq. (26).

Expanding $S_0(E - \delta\varepsilon(r)) \approx S_0(E) - (dS_0/dE)\delta\varepsilon(r)$ and using Eq. (36) and the relation between the single-particle optimal fluctuations and the wave functions: $U_h(x - x_h) = -\lambda \psi_h^2(x - x_h)$ and $U_e(x - x_e) = \lambda \beta \psi_e^2(x - x_e)$, we find that the r -dependent part of the action coincides with the first-order correction to the action of the optimal fluctuation Eq. (32),

$$\delta S - \frac{dS_0}{dE} \delta\varepsilon = S_1(r).$$

Thus, we can write Eq. (40) in the form

$$F(E) \approx \sqrt{\frac{2\pi A}{B}} \int dr K_h(\varepsilon_h) K_e(\varepsilon_e) D^2(r) \times \exp\left\{-\frac{1}{A}[S_0(E) + S_1(r)]\right\}. \quad (41)$$

The typical energy dependence of the absorption rate Eq. (41) is shown in Fig. 2, where we plot $\ln F$ as a function of $(E/E_0)^{3/2}$, with $E_0 < 0$ being the exciton binding energy. One can see, that the energy dependence of $\ln F$ very quickly becomes linear for energies below E_0 . The linear dependence reflects the relative weakness of the Coulomb interaction between the electron and hole [see Eq. (26)], which was the main assumption of this analytical calculation.

In the case when the integral over the electron-hole separation r comes from a small vicinity of the saddle-point r_* , determined by Eq. (33), we find that the saddle-point action coincides with the action for the optimal fluctuation, $S_0 + S_1(r_*)$, obtained in the Sec. IV. The result of the saddle-point integration over r is

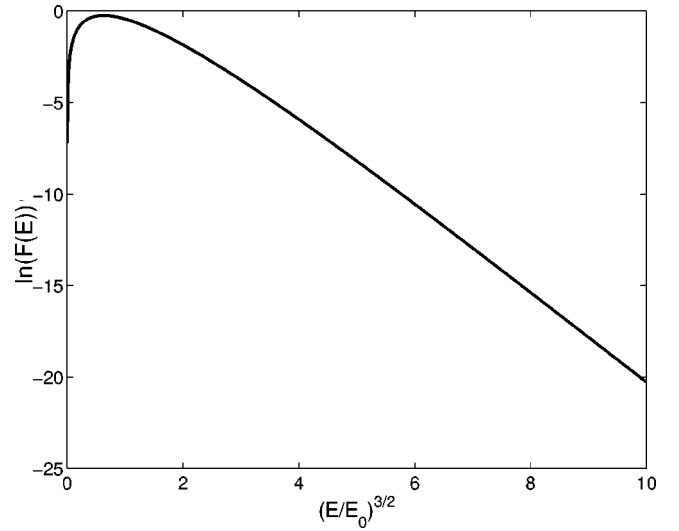


FIG. 2. The energy dependence of the logarithm of the absorption rate.

$$F(E) \approx \frac{2\pi A}{\sqrt{B \frac{d^2 S}{dr_*^2}}} \rho_h(\varepsilon_h) \rho_e(\varepsilon_e) D^2(r_*) e^{-S_1(r_*)/A},$$

where

$$\frac{d^2 S}{dr_*^2} \approx \frac{2\lambda e^2}{\epsilon r_*^3} (\kappa_e r_* - 1)$$

and ε_h and ε_e are the unperturbed single-particle energies, given by Eq. (31) [we have used that the factors $K_h(\varepsilon_h)$ and $K_e(\varepsilon_e)$ are slow functions of the energies].

Equation (41) also applies when, instead of one optimal fluctuation, one finds an entire manifold of disorder realizations that significantly contribute to the optical absorption. In particular, for a noninteracting electron-hole pair the minimum of the action S_1 is reached at the largest possible electron-hole separation, since for $V(x_1 - x_2) = 0$ nothing can counteract the “repulsion” between the two disorder fluctuations, described by the second term in Eq. (32). However, these electron-hole states clearly do not contribute to the optical absorption as the transition matrix element D vanishes at infinite r . Since the repulsion decays exponentially with the electron-hole separation, the action S_1 is a very weak function of r , as soon as the latter exceeds the spatial extent of the electron and hole states, κ_h^{-1} and κ_e^{-1} . The contribution of the electron-hole pairs with large r is then suppressed not by the weight of such fluctuations, but by the smallness of the transition matrix element D , which decays exponentially with r . Thus, when the electron-hole interaction is absent or relatively weak, in order to calculate the absorption rate, one has to sum the contributions of many disorder fluctuations with different electron-hole separations, which can be accomplished using Eq. (41). In this case the electron-hole separation is the “soft mode” in the space of disorder realizations, integration over which is non-Gaussian.

The integration in Eq. (41) for the noninteracting electron and hole becomes particularly simple in the case when the hole is localized stronger than the electron, or more precisely, when $e^{-(\kappa_h - \kappa_e)r} \ll 1$ for relevant electron-hole separations r , which we already have used in the Sec. IV. Then the action S_1 for the noninteracting case is

$$S_1(r) = \lambda^2 \beta \int dx \psi_h^2(x) \psi_e^2(x+r) \approx \lambda^2 \beta \psi_e^2(r)$$

and the transition matrix element is given by

$$D(r) \approx \frac{\pi}{\sqrt{2\kappa_h}} \psi_e(r),$$

and

$$\int dr D(r)^2 e^{-S_1(r)/A} = \frac{\pi^2}{4\kappa_h} \int_{-\infty}^{+\infty} \frac{dz}{\cosh^2 z} e^{-\zeta/\cosh^2 z} \approx \frac{\pi^2 \beta M A}{32\mu |E|^2},$$

where we assumed that

$$\zeta = \frac{\lambda^2 \beta \kappa_e}{2A} = \frac{3m_e \beta^3}{M} \frac{S_0}{A} \gg 1. \quad (42)$$

Since for the applicability of the optimal fluctuation method, anyhow, the action S_0 has to be much larger than A , Eq. (42) holds, unless $3m_e \beta^3/M$ is very small.

Thus, finally, for the noninteracting electron and hole in one dimension the expression for the absorption rate (that also includes the preexponential factor) is

$$F(E) \approx \frac{1}{\beta} \left[\frac{\pi\mu}{2A\hbar} \sqrt{\frac{|E|}{2M}} \right]^{1/2} \exp \left\{ -\frac{4\hbar}{3A} \sqrt{\frac{2|E|^3}{M}} \right\}. \quad (43)$$

VI. NUMERICAL RESULTS

The analytical results of Secs. IV and V for the optimal fluctuation and the optical absorption spectrum were obtained by a perturbative treatment of the Coulomb interaction between the electron and hole. When this interaction is of the same order as the magnitude of the disorder potential, the optimal fluctuation has to be solved numerically. In this section we present our numerical results for the optimal fluctuation in one dimension.

Instead of solving Eq. (28) directly, we perform the numerical calculations for a tight-binding model defined on a lattice. The discrete version of the Schrödinger equation (1) reads

$$-t_1(\psi_{n-1,m} + \psi_{n+1,m}) - t_2(\psi_{n,m+1} + \psi_{n,m-1}) + V_{nm}\psi_{nm} + (U_n - \beta U_m)\psi_{nm} = \mathcal{E}\psi_{nm}, \quad (44)$$

where t_1 and t_2 are, respectively, the hole and electron hopping amplitudes, the indices $n, m = 1, 2, \dots, L$ denote the sites of the one-dimensional lattice with the lattice constant a and periodic boundary conditions, U_n is the disorder potential

$$\langle U_n \rangle = 0, \quad \langle U_n U_m \rangle = \frac{A}{a} \delta_{n,m},$$

and V_{nm} is the regularized Coulomb interaction

$$V_{nm} = -g_0 \left(\frac{1}{|n-m| + \delta_{n,m}} + \frac{1}{N - |n-m| + \delta_{|n-m|, N}} \right). \quad (45)$$

Here, $g_0 = e^2/\epsilon a$ and the second term in the brackets is added to satisfy the periodic boundary conditions.

The discrete equation (44) reduces to the Schrödinger equation (1) in the continuum limit, when all relevant electron and hole states have small wave vectors, $k_1 a, k_2 a \ll 1$. In that case, the dispersion of the free hole dispersion is

$$\varepsilon_h(k) = -2t_1 \cos ka \approx -2t_1 + t_1(ka)^2,$$

so that $m_h = \hbar^2/2t_1 a^2$ and, similarly, for the electron we have $m_e = \hbar^2/2t_2 a^2$. In this calculation we put $a = t_1 = t_2 = 1$. Then the continuum limit is reached for small values of the coupling constant g_0 and the energy E , counted from the bottom of the band,

$$E = \mathcal{E} + 2(t_1 + t_2) = \mathcal{E} + 4 \ll 1.$$

The discrete analog of the action (12) is

$$S_\lambda[\psi, U] = \frac{1}{2} \sum_n U_n^2 + \lambda(E[\psi, U] - \mathcal{E}), \quad (46)$$

where

$$E[U, \psi] = \frac{-2 \sum_{nm} [\psi_{nm}(t_1 \psi_{n+1,m} + t_2 \psi_{n,m+1}) + V_{nm} \psi_{nm}^2 + (U_n - \beta U_m) \psi_{nm}^2]}{\sum_{nm} \psi_{nm}^2}. \quad (47)$$

The denominator in the last equation takes care of the wavefunction normalization. It is readily seen that varying S_λ with respect to the wave function ψ_{nm} (that can be chosen real) yields the discrete Schrödinger equation (44), while the minimization of S_λ with respect to the disorder potential U_n gives a relation between the optimal fluctuation and ψ_{nm} ,

$$U_n = -\frac{\lambda}{\mathcal{N}} \sum_m (\psi_{nm}^2 - \beta \psi_{mn}^2), \quad (48)$$

where we have used the notation

$$\mathcal{N} = \sum_{nm} \psi_{nm}^2. \quad (49)$$

We find the optimal fluctuation U_n and the corresponding wave function ψ_{nm} by minimizing the functional

$$\begin{aligned} \mathcal{A}_\lambda[\psi] = & -\frac{2}{\mathcal{N}} \sum_{nm} \psi_{nm} (t_1 \psi_{n+1,m} + t_2 \psi_{n,m+1}) \\ & + \frac{1}{\mathcal{N}} \sum_{nm} V_{nm} \psi_{nm}^2 - \frac{\lambda}{2\mathcal{N}^2} \sum_n \left(\sum_m (\psi_{nm}^2 \right. \\ & \left. - \beta \psi_{mn}^2) \right)^2, \end{aligned} \quad (50)$$

which is the two-particle analog of Eq. (18). While S_λ depends on both ψ_{nm} and U_n , \mathcal{A}_λ is a functional of the wave function only. One can easily check that the condition $\delta \mathcal{A}_\lambda / \delta \psi_{nm} = 0$ is equivalent to Eq. (44) with the disorder potential U_n given by Eq. (48). The minimization of \mathcal{A}_λ with respect to ψ_{nm} was carried out numerically, using the steepest descent algorithm for $L = 50$. The energy of the electron-hole pair and other quantities of interest, e.g., the optimal fluctuation U_n , its weight, and the corresponding electron-hole wave function, are first obtained as functions of the Lagrangian multiplier λ . Then we eliminate λ by replotting these quantities as functions of the energy E .

We first consider the energy dependence of the weight of the optimal fluctuation, which, essentially, determines the energy dependence of the optical absorption rate. This weight is given by $e^{-S/A}$, where $S = \frac{1}{2} \sum_n U_n^2$ is the action of the optimal fluctuation [cf. Eq. (11)]. Motivated by the dominant $|E|^{3/2}$ behavior of S [cf. Eq. (26)], we plot in Fig. 3 $S^{2/3}$ as a function of the energy E for $g_0 = 0.2$ and $\beta = 0.5$. The open circles are obtained by the numerical procedure described above, while the solid line is the result of our approximate analytical calculation of the action: $S = S_0 + S_1$, where S_0 and S_1 are given by Eqs. (26) and (32), respectively. Clearly, apart from a small energy interval near the exciton binding energy in the absence of disorder, $E_0 \approx -0.08$, the energy dependence of $S^{2/3}$ is indeed close to linear and the agreement between our numerical and analytical results is good.

For energies close to E_0 , the assumption that disorder dominates the Coulomb interaction, used in our analytical approach, breaks down, which explains the deviations of the numerical data from the $S \propto |E|^{3/2}$ law and from the analytical curve. On the other hand, the deviations found at relatively

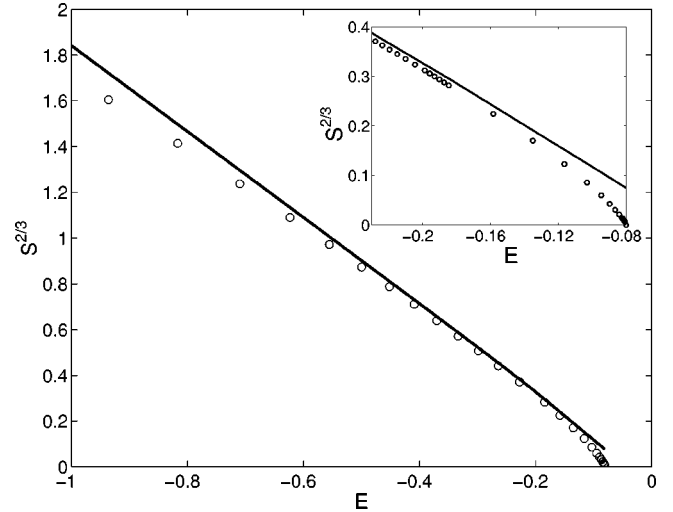


FIG. 3. The action S of the optimal fluctuation to the power $2/3$ plotted as a function of the electron-hole energy E for $g_0 = 0.2$, $\beta = 0.5$, and the chain length $L = 50$. The open circles are results of the numerical calculation and the solid curve was obtained analytically (see explanations in the text).

large energies $|E| \sim 1$ are due to the break down of the continuum approximation, resulting from the fact that the hole becomes localized on a single lattice site.

These changes in the energy dependence of S reflect changes in the shape of the optimal fluctuation and the corresponding electron-hole wave function. In Fig. 4 we plot U_n and the contour plot of ψ_{nm} , calculated numerically, for three different values of the energy: -0.14 , -0.30 ,

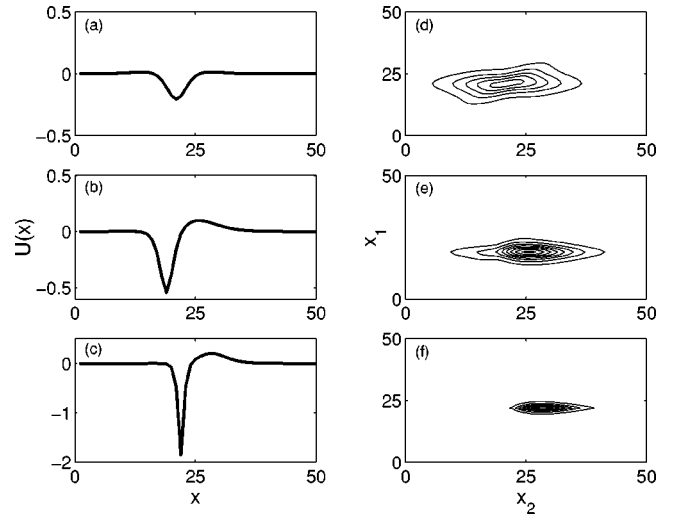


FIG. 4. The shape of the numerically obtained optimal fluctuation for $E = -0.13$ (a), $E = -0.3$ (b), and $E = -0.94$ (c). Panels (d)–(f) show contour plots of the corresponding electron-hole wave function ψ_{nm} . All plots correspond to $g_0 = 0.2$, $\beta = 0.5$, and $L = 50$. The coordinates $x_1 = na$ and $x_2 = ma$, where $n, m = 1, \dots, L$ describe, respectively, the hole and electron positions in the chain in units of $a = 1$. Note, that as E decreases, the shape of the optimal fluctuation undergoes a transition from a single-well to a “dip-bump” structure.

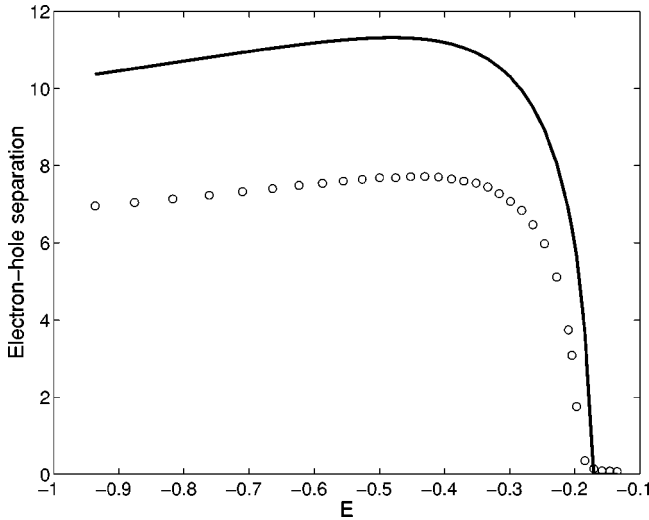


FIG. 5. Numerical (circles) and analytical (solid line) energy dependence of the average electron-hole separation calculated for $g_0=0.2$, $\beta=0.5$, and $L=50$.

and -0.94 . The first value $E = -0.14$ is rather close to the exciton binding energy E_0 . In that case, the optimal fluctuation, shown in Fig. 4(a), is rather shallow and it is symmetric around x_0 , where x_0 is the position of the minimum of this fluctuation. This symmetry implies that no separation exists between the average electron and hole positions. Such an optimal fluctuation was discussed in Ref. 11 for the situation where the Coulomb interaction dominates the disorder and the spatial extent of the optimal fluctuation l_{opt} is much larger than the exciton radius r_{ex} , leading to decoupling of the center-of-mass and the relative motion. That limit is rather difficult to simulate numerically within our discrete model, as the requirement to maintain the validity of the continuum approximation then leads to $1 \ll r_{ex} \ll l_{opt} \ll L$, thus forcing us to consider a very large lattice size L . From Fig. 4 one observes that for $E = -0.14$, l_{opt} is comparable to the exciton radius $r_{ex} \sim 5$. Still, one can see from Fig. 4(d) that the electron-hole wave function, apart from the delocalization along the electron coordinate x_2 , also shows a strong delocalization along the line $x_1 = x_2$, which corresponds to the center-of-mass motion of the exciton. For the second value of the energy, $E = -0.3$, the optimal fluctuation has the asymmetric “dip-bump” shape, which corresponds to the localization of the electron and hole on different sites of the chain [see Fig. 4(b)]. Finally, at $E = -0.94$, the hole is practically localized on one chain site, in which case the discrete model should not be used to simulate the continuum one [see Fig. 4(c)]. From Figs. 4(e),(f) one can see that for $E = -0.3$ and $E = -0.94$ the electron-hole wave function ψ_{nm} is mostly delocalized along the electron coordinate x_2 and it has no delocalization along the center-of-mass direction, $x_1 = x_2$.

The transition from the symmetric to the asymmetric shape of the optimal fluctuation can be most clearly seen from the energy dependence of the average electron-hole separation, defined by

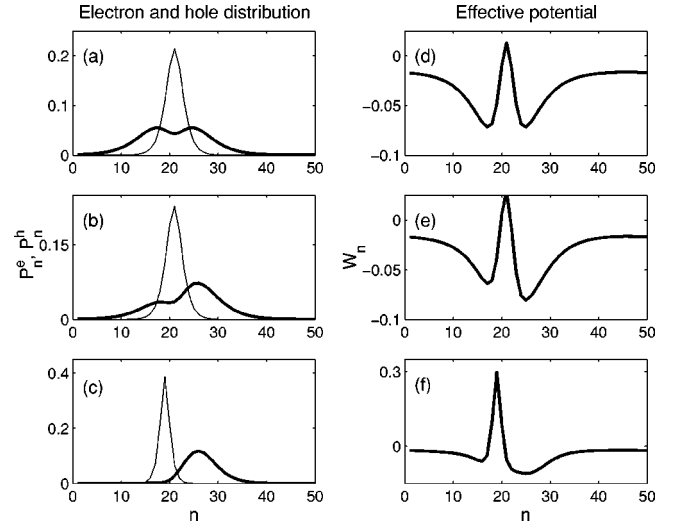


FIG. 6. The left-hand side of the figure shows the electron (thick line) and the hole (thin line) distributions in the typical electron-hole state for $E = -0.18$ (a), $E = -0.20$ (b), and $E = -0.50$ (c). On the right-hand side [panels (d)–(f)] the corresponding effective potentials acting on the electron are plotted. All plots correspond to $g_0=0.2$, $\beta=0.5$, and $L=50$.

$$r = \sum_{nm} (m-n) \psi_{nm}^2.$$

The results of the numerical calculation of r as a function of the energy E are shown in Fig. 5 by circles. In this figure we also plot the optimal electron-hole distance r_* , obtained by our approximate analytical approach of Sec. IV [Eq. (33)]. The minimization of the correction to the action S_1 was performed numerically, as the approximation that was used to obtain Eq. (35) is too crude to describe the changes in the shape of the optimal fluctuation. Figure 5 shows a transition from the optimal fluctuation with zero-average electron-hole separation to one with finite separation. One can also see that the approximate analytical approach provides a good qualitative description of this transition [in particular, the onset of the transition and the shape of the $r(E)$ curve], but it gives a somewhat larger value of the electron-hole separation at low energies.

To clarify the nature of this transition, we plot in Figs. 6(a)–(c) the coordinate distributions of the electron (thick line) and hole (thin line)

$$P_n^h = \sum_m \psi_{nm}^2,$$

$$P_m^e = \sum_n \psi_{nm}^2,$$

for $g_0=0.2$, $\beta=0.5$, and three different values of energy E . In addition, Figs. 6(d)–(f) show the corresponding effective Hartree potential acting on the electron

$$W_m = -\beta U_m + \sum_n P_n^e V_{nm}.$$

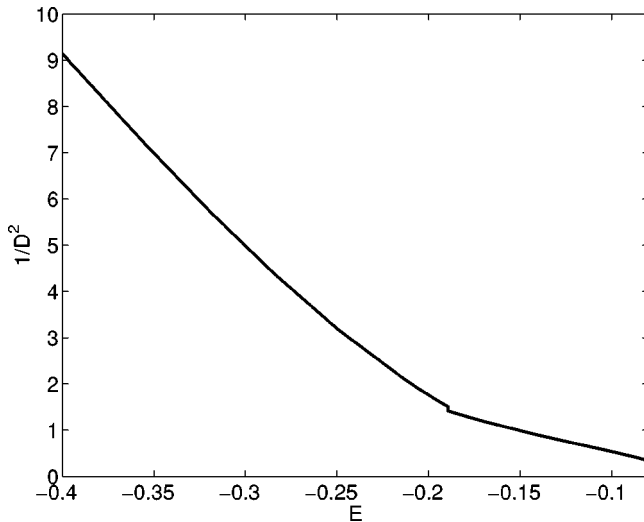


FIG. 7. The energy dependence of D^{-2} for $g_0=0.2$, $\beta=0.5$, and $L=50$.

At $E = -0.18$, just above the transition, the electron wave function has two peaks, in accordance with the double-well structure of the effective Hartree potential [see Figs. 6(a),(d)]. The peak separating the two potential wells is the disorder potential that localizes the hole and repels the electron. As the energy E decreases, the height of the peak grows, which suppresses the electron tunneling between the two wells. For large separations between the wells, the weight of the symmetric optimal fluctuation, in which the electron is delocalized over the two wells, is lower than the weight of the single-well optimal fluctuation with the same electron energy. For $E = -0.20$ in the transition region the two wells become unequal [see Figs. 6(b) and 6(e)], and at $E = -0.50$, well below the transition region, the electron is predominantly located in a single well [see Figs. 6(c) and 6(f)].

We finally note that, though the weight of the optimal fluctuation varies smoothly at the critical energy $E_c \sim 0.19$ [see Fig. 3], the transition matrix element D [see Eq. (6)], being sensitive to the shape of the electron-hole wave function, is singular at the critical energy. This is illustrated in Fig. 7, where we plot the energy dependence of D^{-2} . Clearly the derivative dD/dE is discontinuous at $E = E_c$ (one can see also a small discontinuity of D at the critical energy, which is likely to be a finite-size effect).

VII. CONCLUSIONS

In this paper we studied theoretically the photoexcitation of electron-hole pairs in disordered one-dimensional semiconductors. Using the optimal fluctuation method, we calculated the low-energy tail of the absorption spectrum in these systems. We were, in particular, interested in the effects of the Coulomb interaction between the electron and hole on the energy dependence of the absorption spectrum.

We want to point out, however, that the calculation of the absorption rate is a nontrivial problem even for the noninteracting electron and hole. In particular, it cannot be reduced to

a single-particle calculation, since, on the one hand, the electron and hole move in a common disorder potential, and, on the other hand, the effect of the disorder on the electron is different from the effect of the disorder on the hole. We showed that to calculate the optical absorption tail for relatively weak interactions between electron and hole, one has to go beyond the standard optimal fluctuation method and perform the (non-Gaussian) integration over the “soft mode”—the electron-hole separation [see the derivation of Eq. (43)].

We found that, as the photon energy decreases, the shape of the optimal fluctuation undergoes a crossover. Close to E_0 (the exciton binding energy in the absence of disorder), the Coulomb energy dominates over the disorder and the optimal fluctuation has a symmetric shape. It reflects the fact that at those energies the exciton is not entirely destroyed by disorder. That limit was considered previously in Ref. 11. In the opposite limit, when the disorder dominates over the Coulomb interaction, the optimal fluctuation has two parts: a “dip” that localizes the positively charged hole and a “bump” that localizes the negatively charged electron. This transition to an asymmetric optimal fluctuation is characteristic for motion in one dimension—it does not occur in two and three dimensions. It can, in principle, be observed experimentally, as the absorption rate has a different energy dependence above and below the critical energy (see Figs. 3 and 7).

At photon energies well below the exciton energy, when the Coulomb interaction can be treated as a perturbation, we obtained an analytical expression for the optical conductivity of a disordered one-dimensional semiconductor. We also performed numerical calculations of the optimal fluctuation for a discrete model that allowed us to study the whole region of photon energies, in which the optimal fluctuation method is applicable (i.e., for $E < E_0$, for which the action S is much larger than A). In the continuum limit we found a good agreement between our numerical and analytical results.

Finally, in the region of validity of the optimal fluctuation method, we do not observe a significant delocalization due to interaction discussed in Ref. 5. In particular, as can be seen from Fig. 4, the shape transition has little effect on the localization length of the electron-hole pair. We note, however, that the mechanism of delocalization, proposed in Ref. 5, essentially relies on the high density of excited two-particle states, which enhances the diffusion rate of the two-particle states along the chain. We, however, consider low-energy states that are all strongly localized. The density of such states is relatively small and we do not expect, neither do we see, an enhancement of the localization length. The only exception to this is the case $\beta \sim 1$, when the interaction of a tightly bound electron-hole pair with disorder is relatively weak.

ACKNOWLEDGMENTS

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**APPENDIX: FIRST-ORDER CORRECTION
TO THE WEIGHT OF THE OPTIMAL FLUCTUATION
OF THE ELECTRON-HOLE PAIR**

The calculation of the first-order correction can be performed in two different ways. On the one hand we can use the exact relation between the action of the optimal fluctuation and the disorder potential averaged over the electron-hole wave function:

$$\begin{aligned} & \int d^2x \Psi^2(x_1, x_2) [U(x_1) - \beta U(x_2)] \\ &= -\frac{1}{\lambda} \int dx U^2(x) = -\frac{2}{\lambda} S. \end{aligned}$$

This equation allows us to write the action in the form

$$\begin{aligned} S = \frac{\lambda}{2} \int d^2x \Psi \left(-\frac{\hbar^2}{2m_h} \frac{\partial^2}{\partial x_1^2} - \frac{\hbar^2}{2m_e} \frac{\partial^2}{\partial x_2^2} \right. \\ \left. + V(x_1 - x_2) - E \right) \Psi. \end{aligned}$$

The first-order correction to the action is then given by

$$S_1 = \frac{\lambda}{2} \int d^2x \Psi_0^2 V(x_1 - x_2) + S'_1, \quad (\text{A1})$$

where

$$S'_1 = \lambda \int d^2x \Psi_1 \left(-\frac{\hbar^2}{2m_h} \frac{\partial^2}{\partial x_1^2} - \frac{\hbar^2}{2m_e} \frac{\partial^2}{\partial x_2^2} - E \right) \Psi_0.$$

Using Eq. (29) for the unperturbed electron-hole wave function Ψ_0 and Eq. (16) for the single-particle wave function ϕ , we obtain

$$\begin{aligned} S'_1 = 2\lambda \int d^2x \Psi_0 \Psi_1 \{ |\varepsilon_h| \phi^2(\kappa_h[x_1 - x_h]) \\ + |\varepsilon_e| \phi^2(\kappa_e[x_2 - x_e]) \}. \end{aligned}$$

On the other hand, the direct calculation of the first-order correction to the action, in which one uses Eq. (11), gives

$$\begin{aligned} S_1 = -\frac{4\varepsilon_h\varepsilon_e}{\beta} \int dx \phi^2 \left(\kappa_h[x - x_h] \phi^2(\kappa_e[x - x_e]) \right. \\ \left. + \int dx U_0(x) U_1(x), \right. \end{aligned} \quad (\text{A2})$$

where U_1 is the first-order correction to the disorder potential

$$\begin{aligned} U_1(x) = -2\lambda \int dx' [\Psi_0(x, x') \Psi_1(x, x') \\ - \beta \Psi_0(x', x) \Psi_1(x', x)]. \end{aligned}$$

The second term in Eq. (A2) can be rewritten in the form

$$\begin{aligned} \int dx U_0(x) U_1(x) = 4\lambda \int d^2x \Psi_0(x_1, x_2) \Psi_1(x_1, x_2) \\ \times \{ |\varepsilon_h| \phi^2(\kappa_h[x_1 - x_h]) + |\varepsilon_e| \phi^2(\kappa_e[x_2 \\ - x_e]) - \beta^{-1} |\varepsilon_e| \phi^2(\kappa_e[x_1 - x_e]) \\ - \beta |\varepsilon_h| \phi^2(\kappa_h[x_2 - x_h]) \} \approx 2S'_1, \end{aligned} \quad (\text{A3})$$

where in the last step, we used that the overlap between the unperturbed electron and hole wave functions, $\psi_h(x - x_h)$ and $\psi_e(x - x_e)$ is small, as a result the third and the fourth terms in the curly brackets of Eq. (A3) give a much smaller contribution than the first and the second terms. Thus, we obtain

$$S_1 = -\frac{4\varepsilon_h\varepsilon_e}{\beta} \int dx \phi^2(\kappa_h[x - x_h]) \phi^2(\kappa_e[x - x_e]) + 2S'_1. \quad (\text{A4})$$

Combining Eqs. (A1) and (A4), we obtain

$$\begin{aligned} S_1 = \lambda \int d^2x \Psi_0^2 V(x_1 - x_2) + \frac{4\varepsilon_h\varepsilon_e}{\beta} \int dx \phi^2 \\ \times (\kappa_h[x - x_h]) \phi^2(\kappa_e[x - x_e]), \end{aligned} \quad (\text{A5})$$

which is equivalent to Eq. (32).

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