Propagation of Coulomb-correlated electron-hole pairs in semiconductors with correlated and anticorrelated disorder

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Local ultrafast optical excitation of electron-hole pairs in disordered semiconductors provides the possibility to observe experimentally interaction-assisted propagation of correlated quantum particles in a disordered environment. In addition to the interaction driven delocalization known for the conventional single-band TIP-(two-interacting-particles)-problem the semiconductor model has a richer variety of physical parameters that give rise to new features in the temporal dynamics. These include different masses, correlated vs. anticorrelated disorder for the two particles, and dependence on spectral position of excitation pulse.

PACS numbers: 72.15.Rn, 73.20.Dx, 73.20.Jc, 78.47.+p, 78.66.-w

Introduction. It is by now well established that the center-of-mass (COM) motion of two interacting particles (TIP) in a single tight-binding band of a one-dimensional Hamiltonian possessing diagonal disorder is spatially more extended than both their relative motion and the motion of the particles without interaction [1]. Although this model is a fundamental paradigm for the more general question of the interplay of disorder and interactions in many-body systems, it suffers from not being susceptible to experimental verification. Furthermore, it is not clear whether this finding, relevant for two particles, can be generalized to real transport situations in dense Fermi systems.

On the other hand, low-intensity optical excitation in disordered semiconductor systems produces strongly Coulomb-correlated electron-hole pairs with large mutual separation such that their motion can be considered to be that of an essentially isolated pair. Optical pump-probe experiments are in principle suited to record the coherent dynamics of the pair after pulsed excitation on a ps-time scale before scattering with acoustic phonons becomes relevant.

Following this idea the temporal traces of the participation number of the electron and the hole have been studied in a previous paper [3] by solving the equation of motion for the two-particle amplitude \( p_{ij}(t) = \langle \hat{d}_i \hat{c}_j \rangle \), which is the interband coherence related to the optical polarization. Here \( \hat{d}_i \) (\( \hat{c}_j \)) are hole (electron) operators at site \( i \) (\( j \)). In the low intensity (with respect to the exciting light pulse) limit the sum rules \( n_{ij}^e = \sum_i p_{ij}^e p_{ij}^d \) and \( n_{ij}^h = \sum_i p_{ij}^h p_{ij}^d \) yield the electron (hole) intraband quantities. Their diagonal elements \( n_{ii}^e, n_{ii}^h \) are the time dependent densities of electron and hole. Defining the participation number \( \Lambda(t) = (\sum_i n_{ii}^e)^{-1} \) the evolution of the excited electron and hole wave packet can be plotted as a function of time. Alternatively, the COM-coordinate \( R \) and the relative coordinate \( \rho \) of the electron-hole wave packet can be calculated directly from the pair amplitudes.

It has been demonstrated that the interaction induces an enormous change of the dynamics if compared to the non-interacting case. Instead of an exponential rise of \( \Lambda(t) \) towards a saturation value on the time scale of the excitation pulse, in the interacting case a slow, diffusion-like rise of \( \Lambda \) is seen that does not seem to saturate in the limited time regime accessible to the numerical calculation (and relevant for the real physical situation). For brevity this feature will be called “enhancement” in the following in accordance with the notion of other work in the field, although it is not implied that from the present dynamical calculation in a finite time domain anything like the enhancement of a localization length can be deduced. The dependence of the enhancement on the sign of the interaction has been studied and it was found that the dependence on the sign vanishes only for excitation like rise of \( \Lambda \) is seen that does not seem to saturate in the center of the continuum.

In this contribution the effect of correlated versus anticorrelated disorder and of the interaction strength is studied.

Model. A two-band Hamiltonian \( H_0 \) is considered

\[
H_0 = \sum_{i=1}^{N} \left( \epsilon_i^e \hat{c}_i^+ \hat{c}_i + \epsilon_i^h \hat{d}_i^+ \hat{d}_i \right) - J^e \sum_{i=1}^{N} (\hat{c}_i^+ \hat{c}_{i+1} + \hat{c}_{i+1}^+ \hat{c}_i) - J^h \sum_{i=1}^{N} (\hat{d}_i^+ \hat{d}_{i+1} + \hat{d}_{i+1}^+ \hat{d}_i)
\]

(1)

with nearest neighbour coupling \( J^e \) (\( J^h \)) for electrons (holes) and diagonal disorder given by a box-shaped distribution function of the single-site energies \( \epsilon_i^e \) (\( \epsilon_i^h \)) of total width \( W \). The \( N \) sites form a linear chain with spacing \( a \) and periodic boundary conditions.

The interaction is given by a regularized Coulomb potential in monopole-monopole form

\[
H_C = \frac{1}{2} \sum_{i,j=1}^{N} (\hat{n}_i^e - \hat{n}_i^h)V_{ij}(\hat{n}_j^e - \hat{n}_j^h)
\]

(2)
with
\[ V_{ij} = -\frac{U}{4\pi \varepsilon \varepsilon_0 r_{ij} + \alpha} e^2 \] (3)

where \( \alpha = 5a \) and \( U \) is a dimensionless parameter quantifying both strength and sign of the interaction.

The optical excitation is given by a local dipolar coupling to the light field

\[ H_I = -\sum_{i=1}^{N} (\mu_i E^*(t) \hat{d}_i \hat{c}_i + h.c.). \] (4)

For local excitation at site 0 the optical dipole matrix elements are taken to be \( \mu_i = \mu_0 \delta_{i,0} \). The light field is given by

\[ E(t) = (\pi)^{-1/2} \sigma^{-1} \exp(-t/\sigma^2) \exp(-i\omega_0 t) \] (5)

with central frequency \( \omega_0 \) and temporal width \( \sigma \).

**Equation of Motion.** The equation of motion for the pair amplitude \( p_{ij} \) is derived using the total Hamiltonian \( H = H_0 + H_C + H_I \). In the low intensity limit linear response theory is valid and the equation of motion reads (with \( \hbar = 1 \))

\[ \partial_t p_{ij} = -i(\epsilon_i^e + \epsilon_j^h - V_{ij}) p_{ij} + i \sum_{l=1}^{N} (J^e p_{il} + J^h p_{lj}) + i\mu_j E(t) \delta_{ij}. \] (6)

This equation of motion is solved numerically for \( M \) realizations (typically \( M = 20...40 \)) of the disorder drawn from the distribution of site energies. The observables are then configurationally averaged over these realizations.

**Choice of Parameters.** As the present model describes a situation that is in principle accessible to experiments, the interaction strength is chosen to yield exciton binding energies in the range of values typical for low-dimensional semiconductor heterostructures. The intraband couplings \( J^e,h \) and the strength of disorder \( W^e,h \) are free parameters. However, the tight-binding model is thought to model the conduction and valence band extremities of a disordered semiconductor heterostructure. In this communication we show data for \( J^e = J^h = 20 \text{meV} \) exclusively, i.e. equal electron and hole masses are assumed. The lattice constant, equal to a disorder length scale, is taken to be \( a = 20 \text{Å} \). The external optical pulse resembles typical laser pulses used in ultra-fast optical experiments on coherent phenomena. Its central frequency \( \omega_0 \) is chosen to be situated below the center of the absorption band calculated without interaction, in order to model the dynamics of electron-hole pairs in the continuum in a range of energies that excludes LO-phonon emission. Specifically, the gap of the noninteracting case is the origin of the energy axis, the band center is at \( \Delta = 80 \text{meV} \), and \( \omega_0 = 40 \text{meV} \).

For this excitation condition the sign of the interaction is not irrelevant \[3\]. Here we use attractive interaction throughout. A temporal pulse width \( \sigma = 100 \text{fs} \) is used corresponding to a spectral width of \( 22 \text{meV} \). The number of sites \( N \) is taken large enough (typically \( N = 240 \)) such that for not too small disorder the locally excited wave packet does not reach the boundary of the sample in typically some ten picoseconds. After this time acoustic phonon scattering will lead to effective dephasing and the coherent phenomena studied here will be destroyed. For principal studies, however, the calculation is sometimes performed for much longer times, although the results are no longer relevant to experiments.

**Observables.** The extension of the electron and hole wave packets as a function of time can be characterized by the above mentioned participation number \( \Lambda^{e,h} \) which are single-particle quantities. Using the pair amplitude \( p_{ij} \), the two-particle quantities COM-coordinate \( R = (\sum_{i,j} |p_{ij}|^2 (i + j)^2/2)^{1/2} \) and relative coordinate \( \rho = (\sum_{i,j} |p_{ij}|^2 (i - j)^2/2)^{1/2} \) can be defined. Their ratio \( R/\rho \) is a measure of the interaction induced enhancement.

**Correlated and Anticorrelated Disorder.** While previous results \[4\] have been obtained using uncorrelated disorder, here the influence of correlation is studied. Disorder for electrons and holes is called correlated if the site energies of electrons equal that of the mirror image of the hole energies with respect to the gap center. This kind of disorder is expected in heterostructures due to interface roughness leading to a spatially fluctuating confinement of single-particle wave functions. On the other hand, if the energy separation within an isolated site remains constant, i.e. equal to \( \Delta \), the disorder is called anticorrelated. A spatially fluctuating electrostatic field would induce this kind of disorder. The present model resembles that of the conventional single-band-TIP if electrons and holes have equal masses and if correlated disorder (with \( W^e = W^h \)) is applied. Electron and hole wavefunctions are then pairwise equal, both particles live in the same environment. For \( U = 0 \) all optical matrix elements connecting these pairs of states are equal, all others are zero. Therefore, the optical spectrum for \( U = 0 \) resembles the density of single-particle states, however, with bandwidth being the sum of that of the two bands. As in the last years the conventional TIP-model has been widely studied we here concentrate mainly on the anticorrelated situation.

Figure 1 shows COM-traces for correlated and anticorrelated disorder for \( U = 0 \) and \( U = 3 \). Remarkably, the enhancement for anticorrelated disorder is much more pronounced compared to the correlated case. These traces have been calculated for an optical excitation energy \( \omega_0 = 40 \text{meV} \). Looking at the optical spectra for the anticorrelated case, Fig. 2, one realizes a peak that for \( U = 0 \) lies in the center of the band at \( \Delta = 80 \text{meV} \) and shifts towards lower energy for increasing (attract-
Correlated disorder at time $U$. For the excitation condition leading to the large COM-enhancement at $U = 3$ pairs of states in the vicinity of this peak are excited. The origin of the peak can be traced back to optical transitions connecting states in the tails of the single-particle bands. Even for moderate disorder these pairs of states with the nearly identical transition energy are strongly localized at the same position, and therefore their optical matrix elements are large. Exact eigenvectors for a short sample ($N = 10$) have been calculated for $U = 0$ and $U = 3$, confirming the strong overlap within the contributing pairs. Obviously, these strongly localized tail states are not responsible for the large enhancement.

The fact that the enhancement for anticorrelated disorder is larger than that for correlated disorder is not fully understood yet. A prominent difference exists in the equations of motion for $p_{ij}$ for the two cases. For the anticorrelated case the disorder is absent in the equation for the diagonal elements $p_{ii}$, while it is present in the respective equation for the correlated case.

The behavior of excitons in disordered systems is often discussed in terms of the relative coordinate $r = i - j$ and the COM-coordinate $x = (i + j)/2$ instead of the indices $i$ and $j$ (for equal masses, not to be confused with $R$ and $\rho$). After the transformation $(i, j) \rightarrow (r, x)$ the relative coordinate $r$ can be integrated out in the equation of motion if, e.g., the disorder $W$ is smaller than the exciton binding energy and if discrete excitonic resonances are considered. In our case the equation of motion in terms of $x$ would not contain any disorder in the anticorrelated case, in contrast to the correlated case. Although for the parameters used in Fig. 3 this approach is not strictly valid, it suggests a possible solution for the problem at hand.

Finite-Time Scaling. In order to quantify the enhancement finite-time scaling is applied in the following sense. Although with a dynamical calculation for finite times it is impossible to decide whether the two-particle packet is localized or not a long-time saturation value of the COM-coordinate $R_\infty$ is assumed for practical purposes. While the temporal rise of the interaction-free traces is exponential with time scale given by that of the pulse duration, the short time behavior in the interacting case looks like a diffusive process. So the following interpolation relation is used:

$$R(t) = ((Dt)^{-1/2} + R_\infty^{-1})^{-1}. \quad (7)$$

Figure 3 shows the very good quality of typical fits for times larger than the pulse width. It is found that while the diffusivity $D$ is weakly dependent on disorder and interaction, $R_\infty$ shows a much stronger dependence. In the following $R_\infty$ is taken to characterize the enhancement.

Contour Plots. Figure 3 shows contour plots of the two-particle wave function, $|p_{ij}|^2$, for anticorrelated and correlated disorder at time $t = 165$ ps. All other parameters are equal to that used in Fig. 1. Again, the anticorrelated situation shows much larger enhancement.

Dependence of Enhancement on Interaction Strength. Recently the dependence of the enhancement on interaction $U$ has been discussed and a duality relation describing the behavior for large and small $U$ has been proposed. The present calculations yield similar results, see Fig. 3 for anticorrelated disorder. The ratio $R_\infty/\rho_\infty$ is close to unity for small $U$, while it has a maximum around $U = 3...4$ that increases with increasing disorder. Of course, both $R_\infty$ and $\rho_\infty$ decrease with increasing disorder, however, $\rho_\infty$ faster than $R_\infty$. Remarkably, the position of the maximum does not depend on disorder. This behavior of $R_\infty/\rho_\infty$ is apparently related to the fact that for the chosen excitation energy $\omega_0$ the center of the optical spectrum, indicated by the above mentioned peak, shifts (cf. Fig. 2) through the spectral position of the laser pulse with changing $U$. Around $U = 3...4$ the peak coincides with the excitation energy $\omega_0$, while at larger $U$ the excitation takes place again in a region where states outside the center of the single-particle bands are optically coupled. It is known that the enhancement is more pronounced for less localized single-particle states. These states are situated in the center of the single-particle bands. The Coulomb enhancement is therefore largest if the electron-hole packet is generated in the center of the optical spectrum since the interaction couples these weakly localized states most effectively.

Conclusions. For a situation that is in principle accessible to ultra-fast coherent optical experiments, the delocalizing action of the Coulomb interaction on optically generated electron-hole pairs has been studied. The temporal traces of the center-of-mass coordinate $R$ and the relative coordinate $\rho$ have been fitted by an interpolation scheme describing diffusive and localized behavior at small and large times, respectively. It should be noted, however, that the calculation presented here is unable to yield definite answers about localization or otherwise. Correlated and anticorrelated disorder is studied and it is found that the enhancement of $R_\infty/\rho_\infty$ is much more pronounced for anticorrelated disorder.

Acknowledgements This work is supported by DFG, SFB 383, the Leibniz Prize, OTKA (T029813, T024136, F024135), SNSF (2000-52183.97), and the A. v. Humboldt Foundation.

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FIG. 1. Center-of-mass ($R$) traces for $U = 0$ and $U = 3$ for anticorrelated and correlated disorder. Disorder $W^a = W^h = W = 60$ meV.

FIG. 2. Absorption spectra for $U = 0$ and $U = 3$ for correlated (dotted line) and anticorrelated (solid line) disorder. Other parameters as in Fig. 1.

FIG. 3. Fit of the center-of-mass trace by Eqn. (7) for different interaction strengths $U$. Other parameters are the same as in Fig. 1.

FIG. 4. Contour plots of $|p_{ij}|^2$ at 165 ps for anticorrelated (left) and correlated (right) disorder. $U = 3$. Other parameters are the same as in Fig. 1.

FIG. 5. Dependence of the enhancement $R_\infty/\rho_\infty$ on interaction strength $U$ for three different disorder strengths $W^a = W^h = W$. Inset shows $R_\infty$ (full symbols) and $\rho_\infty$ (open symbols) vs. $U$ separately.
The graph shows the relationship between time (ps) and $R$ (sites) for different values of $U$. The curves are labeled as follows:

- $U=3$, anticorrelated
- $U=3$, correlated
- $U=0$, correlated
- $U=0$, anticorrelated
