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Frenkel excitons in one-dimensional systems with correlated disorder

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Abstract

We consider two different models to study the effects of correlations in disorder on optical properties of one-dimensional Frenkel excitons. The first model is described by means of pairwise correlated Gaussian diagonal disorder, where two-level molecules form dimers of equal transition energy. In the second model, disorder is produced by uncorrelated fluctuations in molecular positions, yielding nevertheless correlations in the hopping integrals. The peculiarities of the motional narrowing effect for both models of disorder are discussed in detail. In particular, it is shown that for the configurational disorder, this effect sufficiently differs from that with uncorrelated randomness in the nearest-neighbor coupling. © 1999 Elsevier Science B.V. All rights reserved.

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1. Introduction

Discussion of excitons in solids can be traced back more than half a century to the pioneering work by Frenkel [1] and Wannier [2]. After numerous studies on excitons over several decades, the exciton has been well established in bulk insulators and semiconductors [3,4]. In recent years, the exciton concept has gained popularity to explain transport and optical properties in low-dimensional systems, ranging from semiconductor heterostructures [5] to molecular aggregates and

conjugated polymers [6]. Wannier or Frenkel excitons are often involved in the interpretation of experimental data of such low-dimensional systems, although excitons may exhibit an intermediate character between those two extremes [7,8].

Disorder plays an important role in understanding numerous properties of low-dimensional systems. Since the seminal papers by Anderson [9] and by Mott and Twose [10], electronic and transport properties of randomly disordered systems have been the subject of long-lasting interest both from the fundamental and applied viewpoints [11–13]. Effects of disorder in low-dimensional systems are even more dramatic than in bulk materials: Even a small amount of *uncorrelated* disorder induces localization of all electronic states in one-dimensional (1D) systems, as it was originally conjectured by Mott and Twose [10]. Afterwards, a great deal of work has been devoted to examine this conjecture (see, for instance, Ref. [14]).

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However, in the past few years, a considerable amount of work has been devoted to establish that localization may be suppressed and extended states appear whenever disorder exhibits *dimer*-like correlations [15–19]. The notion of correlated disorder implies that certain physical parameters in random systems are not completely independent within a given correlation length, thus leading to a competition between short-range order and the underlying long-range disorder. The interest of this type of correlations is not merely academic since doped polyaniline can be suitably described by the random dimer model, hence explaining its unexpected high conductivity [16]. Recent experiments on transport properties in semiconductor superlattices with intentional correlated disorder [20] present one more validation of these purely theoretical findings. Moreover, it seems that the suppression of localization by correlations is not restricted to *dimer*-like correlated disorder, but it may also appear in the presence of other types of intersite correlations [21].

Several stochastic processes in organic materials are known to generate correlated random sequences [22]. In particular, interactions with phonons can be described by fluctuations of transition energies [23,24] which in the long wavelength limit (at low temperature) become evidently correlated for different times and different sites. Correlations may arise in a *natural* way in random systems with configurational disorder, even if random positions are uncorrelated, as shown below. Within this context, a natural question that arises is about the possible effects due to long- or short-range correlations on exciton properties and how they can be inferred by using optical techniques.

Effects of exponential correlations of the on-site energy disorder on exciton absorption line shape were already investigated some years ago by Knapp [25]. Later, Knoester showed that nonlinear optical response of disordered J-aggregates yields independent information on the size and degree of intersite correlations [26–28]. Dimer-like short-range correlations have already been considered in the literature, and their effects on exciton trapping [29] and optical absorption spectra [30,31] have been studied in detail.

In this paper we focus on two models of correlated disorder to analyze how correlations affects optical properties of 1D Frenkel excitons. First, a simple pairwise model of diagonal disorder is considered, in which energy dimers (two sites of equal energy) are stochastically distributed over linear chains. In the second model, the source of disorder lies in the uncorrelated fluctuations of molecular positions giving rise, in spite of that, to certain correlations in the hopping integrals. We carry out the numerical simulations of the exciton absorption line and use the self-consistent concept of motional narrowing [32,33] to describe theoretically the scaling of the optical observables.

2. Correlated on-site Gaussian disorder

We start with the following 1D Frenkel Hamiltonian for N two-level molecules (N assumed to be even)

$$\mathcal{H} = \sum_{n=1}^N \varepsilon_n |n\rangle \langle n| + \sum_{n,m=1}^N J_{nm} |n\rangle \langle m|, \quad (1)$$

where $|n\rangle$ is the state vector of the n th molecule with transition energy ε_n and $m \neq n$. For our present purposes, we assume the nearest-neighbor interactions and then take $J_{mm} = -J\delta_{|n-m|,1}$ with $J > 0$. Two different systems, referred to as uncorrelated and correlated disordered systems, will be considered. In the first system, on-site energies ε_n are assumed to be statistically independent Gaussian stochastic variables with zero mean $\langle \varepsilon_n \rangle = 0$ and $\langle \varepsilon_m \varepsilon_n \rangle = \sigma^2 \delta_{mm}$, where angular brackets indicate averages over realizations of disorder.

To build up our correlated disordered model, we chose $N/2$ independent variables ε_{2n-1} ($n = 1, \dots, N/2$) according to the same Gaussian distributions as before and then take $\varepsilon_{2n} = \varepsilon_{2n-1}$, so that

$$\begin{aligned} \langle \varepsilon_m^2 \rangle &= \sigma^2, \quad m = 1, 2, \dots, N, \\ \langle \varepsilon_m \varepsilon_{m+1} \rangle &= \sigma^2, \quad m = 1, 3, \dots, N-1, \\ \langle \varepsilon_m \varepsilon_n \rangle &= 0 \quad \text{otherwise.} \end{aligned} \quad (2)$$

2.1. Perturbative treatment

We now calculate the linear absorption spectra for small values of the degree of disorder σ/J [26]. To this end, we rewrite the Hamiltonian (1) as $\mathcal{H} = \mathcal{H}_d + \mathcal{H}_{od}$, where \mathcal{H}_d (\mathcal{H}_{od}) stands for the diagonal (off-diagonal) part in the site representation, respectively. The diagonal contribution \mathcal{H}_d is considered as a perturbation term. The eigenstates and eigenenergies of \mathcal{H}_{od} are given by

$$|k^0\rangle = \sqrt{\frac{2}{N+1}} \sum_{n=1}^N \sin(Kn) |n\rangle, \quad (3a)$$

$$E_k^0 = -2J \cos K, \quad (3b)$$

where $K \equiv \pi k/(N+1)$ and $k = 1, \dots, N$. In this excitonic representation, the matrix elements $(\mathcal{H}_d)_{kk'}$ are expressed through linear combinations of Gaussian variables ε_n with zero mean

$$(\mathcal{H}_d)_{kk'} = \left(\frac{2}{N+1} \right) \sum_{n=1}^N \varepsilon_n \sin(Kn) \sin(K'n). \quad (4)$$

Consequently, they also have a joint Gaussian distribution. Since $\langle (\mathcal{H}_d)_{kk'} \rangle = 0$ for all k and k' , the joint Gaussian distribution is characterized by its covariance super-matrix $\mathcal{B}_{k_1 k'_1, k_2 k'_2} \equiv \langle (\mathcal{H}_d)_{k_1 k'_1} (\mathcal{H}_d)_{k_2 k'_2} \rangle$. Since in what follows we will deal only with one exciton transitions, the matrix elements $B_{kk'} \equiv \mathcal{B}_{kk', kk'} = \langle (\mathcal{H}_d)_{kk'}^2 \rangle$ will consequently be the subject of our analysis.

Since the state $k = 1$ carries almost the entire oscillator strength of the system, in the perturbative limit the optical absorption spectrum is dominated by a Gaussian peak centered at E_1^0 with standard deviation $\sigma_1 \equiv \sqrt{B_{11}}$ [26]. According to this rule, we obtain [32–34]

$$\sigma_1^u = \sqrt{\frac{3}{2} \frac{\sigma^2}{N+1}} \quad (5a)$$

and

$$\sigma_1^c = \sqrt{3 \frac{\sigma^2}{N+1}}, \quad (5b)$$

where subscripts u and c refer to uncorrelated and correlated disorder, respectively.

As it can be seen from Eq. (5), both magnitudes scale as $N^{-1/2}$, showing the so-called motional narrowing effect [25]. In addition, the standard deviation is larger for correlated inhomogeneous broadening and the difference increases on increasing σ . Similar conclusions were drawn by Knapp [25], Knoester [26] and Fidler et al. [35] for chains with long-range correlated disorder as well as in Ref. [31] by solving the microscopic equation of motion for the present problem. The two times increase of the numerical prefactor of σ_1^2 , from $\frac{3}{2}$ to 3, upon introducing intersite correlations simply reflects the fact that for pairwise correlated disorder, a *dimer* (two sites with equal energies) should be considered as a *single unity*. In other words, the effective number of sites involved in the motional narrowing effect is now determined by the chain length N counted in units of the energy correlation length N_c (being two in our case), and thus appears two times smaller compared to the former. The generalization of this result to an arbitrary value of N_c is straightforward and simply consists of replacing the factor $\frac{3}{2}$ by $(\frac{3}{2})N_c$ when passing from Eq. (5a) to Eq. (5b).

Regarding the exciton optical response we are dealing with, mixing of the lowest state ($k = 1$) with the others is of major importance. Hence, one should compare the energy difference $E_2^0 - E_1^0 \simeq 3\pi^2 J/(N+1)^2$ with the typical fluctuation of $(\mathcal{H}_d)_{21}$ represented by $\sqrt{B_{21}}$. The perturbative approach is valid provided $\sqrt{B_{21}} < E_2^0 - E_1^0$ and fails otherwise. Thus, the equality

$$\sqrt{B_{21}} = \frac{3\pi^2 J}{(N+1)^2} \quad (6)$$

determines a value of σ (for a fixed chain length N), which separates the ranges of perturbative and nonperturbative magnitudes of disorder, where B_{21} in the limit $N \gg 1$ is given by

$$B_{21}^u = \frac{\sigma^2}{N+1}, \quad (7a)$$

$$B_{21}^c = \frac{2\sigma^2}{N+1} \quad (7b)$$

for uncorrelated and correlated disorder, respectively. Note that B_{21}^c is also two times larger than

B_{21}^u similar to that found for the corresponding diagonal matrix elements of B . As in the previous case, the generalization of this result to an arbitrary N_c is achieved by replacing the factor 2 by N_c in Eq. (7b).

2.2. Coherently bound molecules

For higher values of disorder ($\sqrt{B_{21}} > E_2^0 - E_1^0$) the perturbative approach fails since not all molecules of the chain contribute to the optical spectrum, but only a portion of them, referred to as the number of *coherently bound molecules*, N^* [25]. This number can be estimated in a self-consistent way by applying the perturbative criterion to a typical localization segment, namely replacing N by N^* in Eq. (6) [32–36]. The number of coherently bound molecules then is given by [32,33]

$$N^* + 1 = \left(\frac{3\pi^2 J}{\sqrt{B_{21}^*}} \right)^{1/2}, \quad (8)$$

where one should take B_{21}^* according to Eq. (7) and replace N by N^* . Finally, one gets

$$\sigma_1^u = \sqrt{\frac{3}{2}} \frac{\sigma^{4/3}}{(3\pi^2 J)^{1/3}} \quad (9a)$$

and

$$\sigma_1^c = (54)^{1/6} \frac{\sigma^{4/3}}{(3\pi^2 J)^{1/3}} \quad (9b)$$

for uncorrelated and correlated disorder, respectively. Notice that $\sigma_1^u < \sigma_1^c$, as occurs in the perturbative limit. The generalization of Eq. (9b) to the case of N_c -correlated on-site energies reads

$$\sigma_1^c = \sqrt{\frac{3}{2}} N_c^{2/3} \frac{\sigma^{4/3}}{(3\pi^2 J)^{1/3}}. \quad (10)$$

2.3. Numerical results and discussion

We have obtained numerically the absorption line shape according to Ref. [35]. We will fix the value $J = 1$ and focus our attention on the standard deviation σ , ranging from 0.05 up to 0.50. We have diagonalized the Hamiltonian (1) for chains of $N = 250$ with free-end boundary conditions. The

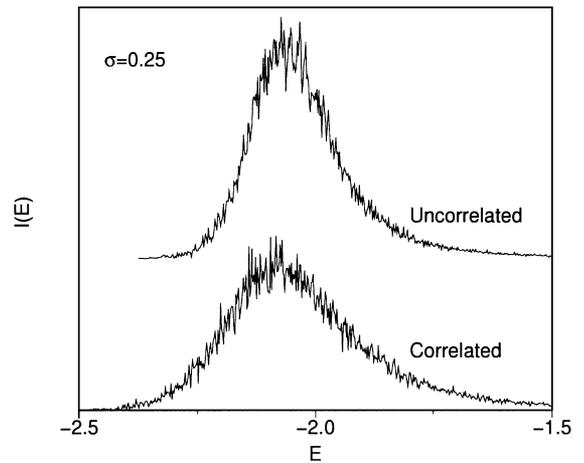


Fig. 1. Absorption spectra for one-dimensional random lattices with Gaussian distribution of uncorrelated (upper curve) and correlated (lower curve) on-site energies with $\sigma = 0.25$.

number of randomly generated systems is 1000 for each value of σ .

We show in Fig. 1 an example of the optical absorption spectra for uncorrelated and correlated disordered systems. The main absorption band shows the characteristic asymmetry discussed in detail for uncorrelated disorder in Refs. [23,24,35,37]. In all the cases we have studied, we found that (i) the standard deviation σ_1 is larger for correlated inhomogeneous broadening and (ii) the difference grows as σ rises. These findings are in agreement with our theoretical estimates.

Fig. 2 shows that σ_1 scales as $\sigma^{4/3}$ for both uncorrelated and correlated disorder, as predicted by our estimates (9a) and (9b). The standard deviation of the spectra can be parameterized as $\sigma_1 = C\sigma^{4/3}/(3\pi^2 J)^{1/3}$, where the constant is $C_u = 1.42$ and $C_c = 2.17$ for uncorrelated and correlated disorder, respectively. These values are slightly larger than those obtained from estimates (9a) and (9b), namely $C_u = 1.22$ and $C_c = 1.94$; nevertheless, the coincidence should be admitted as being highly surprising. Note that the value $C_u = 1.42$ is in agreement with that obtained by the other authors [23,24,35,38–40].

Let us now discuss the problem whether or not the information relative to intersite energy correlations can be revealed from the *linear* exciton optical

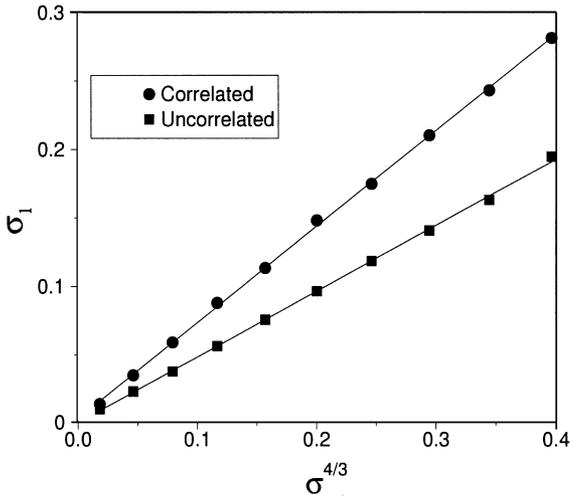


Fig. 2. Standard deviation σ_1 as a function of $\sigma^{4/3}$ for correlated and uncorrelated disordered systems. Solid lines represent the least-squares fits.

response, having in mind the application to linear aggregates of dye molecules (namely, J-aggregates). As it is well known (see, for instance, Ref. [25] and references therein), optical excitations in these systems are Frenkel excitons. The width of the absorption spectrum σ_1 will be the observable providing the data of interest. The numerical factor $C = \sqrt{3/2}N_c^{2/3}$ in the dependence $\sigma_1(\sigma)$ [see Eq. (10)], equal to $C_c = 2.17$ in our particular case of pairwise intersite correlations ($N_c = 2$), differs from that for the case of the absence of correlations ($C_u = 1.42$). Therefore, this coefficient carries the necessary information. To determine C one should measure three parameters: J , σ_1 and σ . The magnitude of intersite coupling J can be measured with appropriate precision from the dimer absorption spectrum of dilute dye solutions, when the aggregation does not take place. The absorption band of J-aggregates usually appears as an isolated peak, so that its width σ_1 is also rather easily measured. With regard to σ , one should assume that its magnitude can be determined from the monomer absorption spectrum. Then, we get all the parameters to be fixed in order to calculate the necessary constant C . Exceeding C over 1.42 means that the correlations are present in the system.

Often, the spectra of monomers are usually structured even at low temperature. In such cases, the

determination of their width σ seems highly questionable. In turn, the analysis of two-exciton transitions, present in the *nonlinear* optical response from the system, can serve as an appropriate method to probe the presence of intersite energy correlations [26,41].

3. Configurational disorder

An example of a *natural* correlated disorder is off-diagonal disorder created by the fluctuations in molecular positions around a regular lattice. For the sake of simplicity, let us consider a circular chain with the intersite interaction of dipole origin, so that $J_{nm} = J/|n - m + \xi_n - \xi_m|^3$ in Eq. (1), where ξ_n is the deviation of the n th molecule from its regular position, which we suppose that it occurs only along the directions toward the two adjacent molecules. This restriction allows us to replace the fluctuations of vector positions by scalars and does not affect the conclusions given below in any way. Despite assuming that ξ_n and ξ_m are uncorrelated, hopping integrals appear to be correlated. Indeed, let us consider for the sake of simplicity these integrals for the nearest neighbors, $J_{n-1,n}$ and $J_{n,n+1}$, and their fluctuations created by the deviation of n th site, linear in ξ_n . Evidently, $\delta J_{n-1,n} = -3J\xi_n$ while $\delta J_{n,n+1} = 3J\xi_n$, just confirming our claim.

In the excitonic representation, the fluctuation of the intersite interaction

$$\delta J_{mn} = \frac{J}{|n - m + \xi_n - \xi_m|^3} - \frac{J}{|n - m|^3}$$

can be written in the form

$$\Delta_{KK'} = \frac{1}{N} \sum_{mn} \delta J_{nm} e^{i(Kn - K'm)}, \quad (11)$$

where $K = 2\pi k/N$ belongs to the first Brillouin zone: $-(N-1)/2 \leq k \leq (N-1)/2$ (N is assumed to be odd). The site index n lies within a symmetric domain, namely $-(N-1)/2 \leq n \leq (N-1)/2$. In what follows, we assume that the standard deviation σ is small, expanding then δJ_{nm} in Eq. (11) in Taylor series with respect to

$$\frac{2(n-m)(\xi_n - \xi_m) + (\xi_n - \xi_m)^2}{(n-m)^2}.$$

To treat the optical properties of the aggregate, the important wave numbers K and K' are the smaller ones, namely $|K|, |K'| \ll 1$. Then, replacing the exponential factors by unity in Eq. (11), one finds that terms linear in ξ_n disappear due to correlations outlined above. Hence, the typical fluctuation of $\Delta_{KK'}$ for $|K|, |K'| \ll 1$ scales in fact as σ^2 . The exact treatment of this problem shows that for the mean-square deviation of $\Delta_{KK'}$, defined as $\sigma_{KK'}^2 = \langle \Delta_{KK'}^2 \rangle - \langle \Delta_{KK'} \rangle^2$, where brackets denote the average over the probability distribution of $\{\xi_n\}$ chosen here to be uncorrelated Gaussians with variance σ^2 , one can arrive at [42]

$$\sigma_{KK'}^2 = \frac{36J^2}{N}(K - K')^2\sigma^2(1 + 40\sigma^2) + \frac{1728J^2}{N}\sigma^4. \quad (12)$$

This result differs from that for uncorrelated diagonal disorder when the corresponding magnitude scales as σ^2/N , with σ^2 being the variance of the site energy distribution [25]. The same behavior (proportional to σ^2/N) appears as well when one simulates off-diagonal disorder by uncorrelated randomness in the nearest-neighbor hopping integrals [39,43] (here, σ^2 stands for the variance of the corresponding distribution).

With respect to the off-diagonal elements of $\sigma_{KK'}^2$, we should note that, in spite of the fact that the first term scales as σ^2 , it has an additional suppression factor proportional to $(K - K')^2 \sim N^{-2}$, much smaller than the fourth order one. Nevertheless, if the first term in Eq. (12) dominates, then $\sigma_{KK'}^2 \propto \sigma^2/N^3$, while in the opposite case one has $\sigma_{KK'}^2 \propto \sigma^4/N$. Both results also differ from the scaling law σ^2/N found for the other types of disorder [25,39,43].

Numerical simulations of optical properties of linear molecular aggregates with off-diagonal disorder generated by Gaussian uncorrelated fluctuations in the molecular positions yielded different behaviors of the optical observables as compared to those for diagonal disorder [35]. The peculiarities found in Ref. [35] can be qualitatively explained from the viewpoint of the modified formula (12). In particular, we will focus on the dependence of the standard deviation of the absorption band, σ_1 , and the radiative rate enhancement factor on

the degree of disorder σ . The radiative rate enhancement factor is approximately given by the number of coherently bound molecules N^* , while σ_1 can be estimated from σ_{KK} replacing N by N^* [25]. Thus, the number N^* is, in fact, the unique quantity determining the observables we are interested in. In our estimate of N^* , we will keep only the second term in Eq. (12) and take the exciton energy spectrum in the nearest-neighbor approximation: $E_K = 2J \cos K \approx 2J - JK^2$. Following the simple rule for estimating N^* discussed in Section 2.2 one can arrive at

$$N^* = \left(\frac{\pi^4}{108} \right)^{1/3} \sigma^{-4/3} \approx \sigma^{-4/3}. \quad (13)$$

Estimating now the standard deviation σ_1 as σ_{KK} replacing N by N^* , we find

$$\sigma_1 = \sqrt{1728J\sigma^{8/3}} \approx 42J\sigma^{8/3}. \quad (14)$$

Notice that for diagonal disorder the corresponding quantities scale as $\sigma^{-2/3}$ [33] and $\sigma^{4/3}$ [23,24,32,35,38,40], respectively.

Using the parameterization σ^α , Fidler et al. [35] found that their numerical data for N^* and σ_1 were fitted by $\alpha = -1.64$ and $\alpha = 2.84$, respectively. Comparison of these data with our results (13) and (14) shows that the numerical σ -scaling of both quantities is reproduced rather well by the theoretical results.

4. Summary

We have considered two different models of Frenkel chains where energy disorder is correlated. In the first model, disorder is a *dimer*-like diagonal disorder, namely site energies always appear at random to pairs, similar to that found in polyaniline [16]. We have shown that both for perturbative as well as for non-perturbative magnitudes of disorder, the width of the main absorption line of the optical spectra increases under the occurrence of correlations. Most important, our results can be easily generalized to larger correlation lengths $N_c > 2$. We have also suggested a possible way to determine whether or not correlations are present in the system from its linear optical response.

On the other hand, the second model of correlated disorder that is presented arises naturally in those systems with configurational disorder, coupled by the dipolar interaction. We have demonstrated that hopping integrals are in fact correlated, even if positions are completely uncorrelated. As a major point, we have shown that motional narrowing effect is essentially different to that previously found for uncorrelated off-diagonal disordered systems.

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