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Off-diagonal disorder model for exciton polaritons in layered media. Disorder-induced origin of radiative widths

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Abstract

Excitons in an infinite layered dipolar medium, such as a layered molecular crystal or a multiple-quantum-well stack, with perfect periodicity exhibit a vanishing radiative width due to the requirement of energy-momentum conservation. Using a polariton model with off-diagonal disorder and a configuration-averaging technique, we show how deviations from the ideal case, specifically in the form of weak departures from perfect periodicity, and the resulting breaking of full translational invariance, give rise to a nonvanishing radiative width.

Shortly after the introduction of the polariton concept, Hopfield recognized that excitons in ideal infinite crystals should not undergo radiative decay [1]. This results from energy-momentum conservation: an exciton with center-of-mass wavevector $\boldsymbol{k}$ couples only to photons with the same wavevector due to momentum conservation. Energy conservation then dictates that radiative decay can only take place when $E_{ex}(\boldsymbol{k}) = \hbar ck$, i.e. when the noninteracting exciton energy is equal to the photon energy, both at $\boldsymbol{k}$. (c is the speed of light in the background dielectric medium.) The exciton–photon interaction, however, gives rise to an anticrossing between the noninteracting exciton and photon dispersions, thus prohibiting radiative decay from occurring. An equivalent view is that at each $\boldsymbol{k}$ the exciton and photon each are described in terms of a harmonic oscillator. The polaritons are then the two coupled modes that result at a given $\boldsymbol{k}$. In particular, regardless of the initial conditions chosen, the dynamics of the system are periodic; irreversible radiative decay does not take place.

The polariton model, in its original form, does not account for scattering, whether by excitons (non-Bose behavior) or phonons, nor for crystal surfaces or heterointerfaces. Other important effects not considered are disorder effects on the electronic states and on the photon modes, or the presence of impurity species with resonances far from $E_{ex}(\boldsymbol{k})$. The effect of all of these is to allow the excitons to access that point in phase space where energy-momentum considerations allow radiative decay. In terms of dipole oscillators, the aforementioned effects disturb the exciton's perfect spatial and temporal coherence which is implied by the two-oscillator model that underlies polariton theory in bulk crystals.

Some of the effects just mentioned are well documented. For example, excitons in optically thick but finite crystals obviously can undergo radiative decay. There has been much interest recently in low-dimensional electronic systems embedded in the three-di-

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mensional vacuum electromagnetic field in which excitons display particularly efficient spatially coherent or *superradiant*, radiative decay [2–8]. There has also been an effort to understand the radiative dynamics of crystaline media in which atoms or molecules with resonances different from the background material are randomly placed [9,10]. Nevertheless, there has been little, if any, work on the effect of static material disorder on the radiative width or other polariton effects.

In this Letter we explore a minimal model of disorder of a specific type. We consider how slight departures from ideal periodicity give rise to a nonvanishing excitonic radiative width in an otherwise infinite medium formed from thin dipolar layers. Thus this model qualitatively describes the origin of the bulk contribution to the radiative width in optically thick layered molecular crystals (molecular crystal slabs) or multiple-quantum-well (MQW) stacks in the absence of other kinds of disorder, scattering, etc. Without loss of generality, we shall employ the language of MQWs in order to allow a straightforward comparison with an existing body of work [6,7,11,12]. Our course is to derive an essentially exact expression (within the polariton model) for the Green function (GF) for the exciton's transition dipole moment in the presence of the interaction with the electromagnetic field. The random displacements of the constituent quantum wells (QW) from their ideal positions show up as a nonadditive, off-diagonal, energy-dependent perturbation of infinite range as viewed in a tight-binding model. We shall then apply a configuration-average technique to extract the disorder-induced radiative width accounting for first-order correlations.

We consider a stack of N identical symmetric QWs (we let $N\to\infty$) each of width L_z much less than the optical wavelength corresponding to the transition of interest, and we take the location of the lth QW in the growth (z) direction to be L_l. The spectral and dynamical properties of an exciton interacting self-consistently with the vacuum electromagnetic field are given by the dipole GF $\vec{\vec{\mathbf{D}}}$ [2], the definition of which is given in Refs. [6,11,12]. In summary, the coupled exciton–electromagnetic modes (polaritons) can be classified as s wave (transverse electric) and p wave (transverse magnetic) [5]. The s-wave mode corresponds to the T-mode polariton in which the transition dipole moment lies in the QW plane but is perpendicular to the in-plane excitation wavevector $\boldsymbol{k}_\parallel$ while the p modes (in a single symmetric QW) can be further classified as the L mode (dipole moment parallel to $\boldsymbol{k}_\parallel$) and the Z mode (dipole moment parallel to z direction). We shall assume that there is no electronic coupling between the T, L, and Z modes, which holds exactly insofar as the heavy-hole and light-hole excitons in zincblende-semiconductor symmetric QWs are concerned. In MQWs, the L and Z modes are optically coupled: this coupling, however, is shown in practice to be negligibly weak for GaAs/AlGaAs MQWs [12]. For the sake of brevity, we shall therefore neglect this coupling. This allows us to drop the dyadic notation and write $\mathbf{D}_\epsilon$ where $\epsilon\in\{\mathrm{T, L, Z}\}$. We find that the GF obeys the Dyson equation $\mathbf{D}_\epsilon^{-1}=\mathbf{D}_\epsilon'^{-1}-\hbar\boldsymbol{\Sigma}_\epsilon^{(1)}$, where $\mathbf{D}_\epsilon'$ is the exciton GF including depolarization effects ((TL)–Z splitting) [5] and $\hbar\boldsymbol{\Sigma}_\epsilon^{(1)}$ is the spatially regular (nonlocal) part of the radiative self-energy (SE) [6,11,12]. $\mathbf{D}_\epsilon'$ is site diagonal, namely $[\mathbf{D}_\epsilon']_{ll'}=2E_{\mathrm{ex}}(\boldsymbol{k}_\parallel)/[(i\varepsilon)^2-E_{\mathrm{ex}}(\boldsymbol{k}_\parallel)^2-2E_{\mathrm{ex}}(\boldsymbol{k}_\parallel)\hbar\Sigma_\epsilon^{(2)}(\boldsymbol{k}_\parallel)]\delta_{ll'}$ where $i\varepsilon$ is the energy variable (in general complex). $\hbar\Sigma_\epsilon^{(2)}(\boldsymbol{k}_\parallel)$ is the spatially singular SE which gives rise to the (TL)–Z splitting. It is nonvanishing only for $\epsilon=\mathrm{Z}$ and is real and energy independent, and therefore only enters the theory as a renormalization of $E_{\mathrm{ex}}(\boldsymbol{k}_\parallel)$ [6,11]. Explicit expressions for all these quantities can be found in Refs. [6,12]. The SE matrix is of the form $\hbar\boldsymbol{\Sigma}_\epsilon^{(1)}=\mathbf{F}\hbar\Sigma_\epsilon^{(1)}(\boldsymbol{k}_\parallel,i\varepsilon)$ where $\hbar\Sigma_\epsilon^{(1)}(\boldsymbol{k}_\parallel,i\varepsilon)$ is the single-QW SE [6,11] and $F_{ll'}=\exp(-\alpha|L_l-L_{l'}|)$ with $\alpha=[k_\parallel^2-(i\kappa)^2]^{1/2}$ and $i\kappa=i\varepsilon/\hbar c$. The terms for $l\neq l'$, therefore, govern the interlayer excitation transfer. They can be regarded as hopping matrix elements in a tight-binding model. They are, however, energy dependent and of infinite range. Moreover, the perturbation is not in any apparent way additive. These complications proclude the straightforward application of known exact results for one-dimensional disordered chains and of many techniques for calculating configuration averaged GFs [13]. The intralayer excitation transfer is built into $\hbar\Sigma_\epsilon^{(1)}(\boldsymbol{k}_\parallel,i\varepsilon)$. Finally, we note that the density of exciton states in each subspace ϵ in the presence of the self-consistent interaction with the elec-

[2] The double arrow signifies a dyadic while the sans serif font signifies a matrix in the site or planewave bases.

tromagnetic field is given by the spectral density $A_\epsilon(E, \boldsymbol{k}_\parallel) = -2\mathrm{Im}\,\mathrm{Tr}\,\mathbf{D}_\epsilon(E+\mathrm{i}0^+, \boldsymbol{k}_\parallel)$. Moreover, $A_\epsilon(E, \boldsymbol{k}_\parallel)$ is an important experimentally accessible quantity because it is proportional to the transmission coefficient and the coherent spontaneous-emission spectrum of an exciton [12]. The widths of features in the spectra, which are governed by the analytic form of the SE, are the radiative widths of the corresponding resonances.

Since we are concerned with MQWs which are almost periodic, the planewave basis $\{|q\rangle\}$ in the z direction is a good zeroth-order starting point. Because $\mathbf{D}_\epsilon$ is diagonal, we need only transform the SE from the site basis $\{|l\rangle\}$ to the planewave basis $\{|q\rangle\}$. In particular, we need only look at $\mathbf{F}$,

$$F_{qq'} = \lim_{N\to\infty} \frac{1}{N} \sum_{l,l'=1}^{N} \exp(\mathrm{i}qL_l)\, F_{ll'} \exp(-\mathrm{i}q'L_{l'}) \quad (1a)$$

$$= \lim_{N\to\infty} \frac{1}{2\pi} \int_{-\infty}^{\infty} \mathrm{d}p \frac{2\alpha}{\alpha^2+p^2}\, \mathscr{S}(p+q)\mathscr{S}^*(p+q')\,, \quad (1b)$$

where $\mathscr{S}(k) = N^{-1/2}\sum_{l=1}^{N}\exp(\mathrm{i}kL_l)$ is the structure factor. The foregoing equations are exact within the polariton model in the absence of LZ coupling (though the inclusion of the latter is trivial). In order to calculate $A_\epsilon(E,\boldsymbol{k}_\parallel)$, it is necessary to diagonalize the infinite matrix $\mathbf{D}_\epsilon$. For the periodic case – $L_l = lL$ with L a constant interwell spacing – the representation $\{|q\rangle\}$ is in fact diagonal since the MQW then possesses full translational invariance. This case was analyzed in Refs. [11,12] where it is shown that all states have vanishing radiative widths as $N\to\infty$. That effects of this type are real is attested to by the measurement of superradiant excitonic emission from QWs in Refs. [14,15], from molecular systems [16], and the observed N dependence of the coherent radiative dynamics in finite MQWs [11,12,17,18]. By adding some randomness – in the form $L_l = lL + \chi_l$ with $|\chi_l| < L$ – we expect to restore, at least in part, the radiative width present in isolated low-dimensional systems.

For $\chi_l \neq 0$, however, we are once again stuck with a problem of infinite size. Fortunately, for many applications it is found that a specific realization of a random medium can be well modeled as an average over all configurations [13]. In diagonal-disorder models the inclusion of only the lowest-order term in the configuration average gives the virtual-crystal approximation, where, for practical purposes, the original random medium is replaced by a uniform one with average parameters. For the type of disorder under consideration, we shall find in addition that the first-order approximation yields the leading modification to the radiative dynamics due to the random interwell spacings. A higher-order treatment is difficult due to the previously mentioned reasons.

Let $\langle \mathscr{O} \rangle$ denote the configuration average of $\mathscr{O}$. In the S-matrix expansion of $\mathbf{D}_\epsilon$ we encounter quantities of the form $\langle \mathscr{S}(p_1+q_1)\mathscr{S}^*(p_1+q'_1)\ldots\mathscr{S}(p_m+q_m)\mathscr{S}^*(p_m+q'_m)\rangle$. If the disorder is uncorrelated between sites, the configuration average of the type above would factor as $\langle \mathscr{S}(p_1+q_1)\rangle\langle\mathscr{S}^*(p_1+q'_1)\rangle\ldots\langle\mathscr{S}(p_m+q_m)\rangle\langle\mathscr{S}^*(p_m+q'_m)\rangle$ were it not for terms which involve multiple occupancy on the same site which is surely correlated. We shall, however, only retain such correlations between products with the same p variable; namely, we shall replace the configuration average of the product of structure factors with $\langle \mathscr{S}(p_1+q_1)\mathscr{S}^*(p_1+q'_1)\rangle\ldots\langle\mathscr{S}(p_m+q_m)\mathscr{S}^*(p_m+q'_m)\rangle$. This should be a good approximation as higher-order correlations will, on average, be weak due to their essentially random phases. Within the first-order approximation, it is easy to show that $\langle \mathbf{D}_\epsilon \rangle$ obeys the following Dyson equation: $[\langle \mathbf{D}_\epsilon\rangle]^{-1} \approx \langle \mathbf{D}_\epsilon^{-1}\rangle = \mathbf{D}_\epsilon'^{-1} - \langle \hbar\boldsymbol{\Sigma}_\epsilon^{(1)}\rangle$. $[\langle\hbar\boldsymbol{\Sigma}_\epsilon^{(1)}\rangle]_{qq'}$ is proportional to $\langle \mathscr{S}(p+q)\mathscr{S}^*(p+q')\rangle$. We have

$$\langle \mathscr{S}(p+q)\mathscr{S}^*(p+q')\rangle$$

$$= \frac{1}{N}\sum_{l,l'=1}^{N} \exp[\mathrm{i}(p+q)lL]\exp[-\mathrm{i}(p+q')l'L]$$

$$\times \langle \exp[\mathrm{i}(p+q)\chi_l]\exp[-\mathrm{i}(p+q')\chi_{l'}]\rangle\,. \quad (2)$$

Our task, thus, reduces to evaluate $C = \langle \exp(\mathrm{i}k\chi_l)\exp(-\mathrm{i}k'\chi_{l'})\rangle$. Let the distribution $P(\chi)$ be defined such that $\langle \mathscr{O}(\chi_l)\rangle = \int_{-\infty}^{\infty}\mathrm{d}\chi\, P(\chi)\mathscr{O}(\chi)$ and moreover assume $\int_{-\infty}^{\infty}\mathrm{d}\chi P(\chi) = 1$. If $\tilde{P}(k-k') = \int_{-\infty}^{\infty}\mathrm{d}\chi P(\chi)\exp[\mathrm{i}(k-k')\chi]$, then $C = [\tilde{P}(k-k') - \tilde{P}(k)\tilde{P}^*(k')]\delta_{ll'} + \tilde{P}(k)\tilde{P}^*(k')$. Assuming $\kappa_{\mathrm{ex}}L \ll 1$ ($\kappa_{\mathrm{ex}} = E_{\mathrm{ex}}(\boldsymbol{k}_\parallel)/\hbar c$), we can neglect Umklapp processes which simplifies the evaluation of the integrals. Combining the previous results gives

$$\langle F_{qq'}\rangle = F_{qq'}^{(0)} + \delta\langle F_{qq'}\rangle\,, \quad (3a)$$

$$\delta\langle F_{qq'}\rangle = \frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{d}p \frac{2\alpha}{\alpha^2+p^2} \times [1-|\tilde{P}(p+q)|^2]\delta_{qq'}, \quad (3b)$$

where $F^{(0)}_{qq'} = F^{(0)}_{qq}\delta_{qq'}$ is $F_{qq'}$ for $\chi_l = 0$ [11]. Therefore, we see that the first-order approximation to $\langle \mathbf{D}_\epsilon \rangle$ is diagonal in the representation $\{|q\rangle\}$, as in the periodic case. Eq. (3) gives sensible results in the limits $P(\chi) = \delta(\chi)$ (no disorder) and $P(\chi) =$ constant (maximum disorder, although here the long-wavelength limit is strictly speaking procluded).

Using Eq. (3) together with the form of the SE and a choice for $P(\chi)$, we can calculate the effect of the weak randomness on the configuration averaged spectral density $\langle A_\epsilon(E, \boldsymbol{k}_\parallel)\rangle$. We find that the randomness induces radiative shifts in addition to non-vanishing radiative widths. We leave the shifts to a future study and concentrate here on the widths. Without calculating $\langle A_\epsilon(E, \boldsymbol{k}_\parallel)\rangle$, we can obtain considerable information on the radiative dynamics from the SE. We employ the exciton-pole approximation [17], whereby the energy argument $i\varepsilon$ in the SE is replaced by $E_{ex}(\boldsymbol{k}_\parallel)$. Provided $|\mathrm{Im}\,\hbar\Sigma^{(1)}_{\epsilon,qq}[E_{ex}(\boldsymbol{k}_\parallel) + i0^+, \boldsymbol{k}_\parallel]| \ll E_{ex}(\boldsymbol{k}_\parallel)$, we can associate $\Gamma_\epsilon(\boldsymbol{k}_\parallel, q) = -\mathrm{Im}\,\Sigma^{(10}_{\epsilon,qq}[E_{ex}(\boldsymbol{k}_\parallel)+i0^+, \boldsymbol{k}_\parallel]$ with the radiative decay rate of the state $(\epsilon, \boldsymbol{k}_\parallel, q)$. Energy-momentum conservation dictates that $\Gamma_\epsilon(\boldsymbol{k}_\parallel, q)$ vanishes for $\hbar c k_\parallel > E_{ex}(\boldsymbol{k}_\parallel)$ [2]. Note that these rates are calculated in the absence of disorder or scattering [6]. $F^{(0)}_{qq'}$ possesses a simple pole, and thus does not contribute directly to the radiative decay. We therefore have

$$\Gamma_\epsilon(\boldsymbol{k}_\parallel, q) = \Gamma^{SQW}_\epsilon(\boldsymbol{k}_\parallel)\,\mathrm{Re}\,\delta\langle F_{qq}\rangle, \quad (4)$$

where $\Gamma^{SQW}_\epsilon(\boldsymbol{k}_\parallel) = -\mathrm{Im}\,\Sigma^{(1)}_\epsilon[E_{ex}(\boldsymbol{k}_\parallel)+i0^+, \boldsymbol{k}_\parallel]$ is the single-QW decay rate [6] which vanishes for $k_\parallel > E_{ex}(\boldsymbol{k}_\parallel)$. In particular, one shows

$$\mathrm{Re}\,\delta\langle F_{qq'}\rangle = \{1-\tfrac{1}{2}[|\tilde{P}(q+\eta)|^2 + |\tilde{P}(q-\eta)|^2]\}\delta_{qq'}, \quad (5)$$

where $\eta = \sqrt{\kappa^2_{ex} - k^2_\parallel}$. Eqs. (4) and (5) represent the principle theoretical results of this Letter. Note that if the scale of variations in $P(\chi)$ is $d \geq 0$, in the long-wavelength limit (qd, $\eta d \ll 1$), then $\mathrm{Re}\,\delta\langle F_{qq}\rangle \sim (q^2+\eta^2)d^2$, and so $\Gamma_\epsilon(\boldsymbol{k}_\parallel, q) \approx (q^2+\eta^2)d^2\Gamma^{SQW}_\epsilon(\boldsymbol{k}_\parallel)$. We shall see this explicitly in the examples below. Thus, we conclude, to first order, the radiative decay rates in the weakly random MQW are given by the single-QW rates modulated by the factor $\mathrm{Re}\,\delta\langle F_{qq}\rangle$; by partially randomizing the spatial phase of the transition dipole moment from QW l to l', external optical coupling is achieved. In the following paragraph we consider specific forms for the distribution function $P(\chi)$.

Let us begin with Gaussian randomness, i.e. $P(\chi) = 1/(d\sqrt{\pi})\exp(-\chi^2/d^2)$ where again d is the characteristic scale of the departure from perfect periodicity. We find

$$\mathrm{Re}\,\delta\langle F_{qq'}\rangle = (1-\tfrac{1}{2}\{\exp[-\tfrac{1}{2}(\eta-q)^2d^2] + \exp[-\tfrac{1}{2}(\eta+q)^2d^2]\})\delta_{qq'}. \quad (6)$$

For qd, $\eta d \ll 1$, we have $\mathrm{Re}\,\delta\langle F_{qq'}\rangle \sim \frac{1}{2}d^2(\eta^2+q^2)$, which is quadratic in ηd and qd, as expected from the general argument given above. Since the crystal grower can choose $P(\chi)$ at will, let us now consider a rather artificial form for the disorder, $P(\chi) = \frac{1}{2}[\delta(\chi+d)+\delta(\chi-d)]$, i.e. each constituent QW resides either to the left or to the right of the ideal position by an amount d. One finds for this case

$$\mathrm{Re}\,\delta\langle F_{qq'}\rangle = [\cos^2(qd)\cos^2(\eta d) + \sin^2(qd)\sin^2(\eta d)]\delta_{qq'}, \quad (7)$$

which again goes as $d^2(\eta^2+q^2)$.

The overall behavior of $\mathrm{Re}\,\delta\langle F_{qq'}\rangle$, and thus of the decay rates $\Gamma_\epsilon(\boldsymbol{k}_\parallel, q)$, depends principally on the scale d of departures from perfect periodicity, rather on the specific form of $P(\chi)$. We can make several conclusions. (I) In order to maximize the enhancement in the coherent optical response of the weakly disordered MQW in the sense of the discussion, this disorder must be maximized subject to such limitations as $|\chi_l| < L$ and $\kappa_{ex}L \ll 1$. An additional limitation is that whatever values of L and χ_l are chosen, the resulting structure must preserve the negligible electronic miniband width due to electronic coupling across the intervening barriers. Otherwise, inhomogeneous broadening may become more severe because the electronic states sample interface disorder (disorder in the plane) more strongly and due to the formation of the minibands themselves. (II) Interface roughness seems to be a nearly ubiquitous feature of QW structures. Therefore, in order to minimize the effects due to this type of disorder, very-high

quality samples are required. Moreover, extreme uniformity from QW to successive QW is also needed. (III) Low temperature is required so that phonons do not provide a significant source of dephasing. Due to the strong coupling of electronic and lattice degrees of freedom in molecular systems, scattering may be inescapable in such materials. (IV) Both static disorder (II) and scattering (III) tend to randomize the macroscopic transition dipole moment associated with the exciton, both within the same QW and from QW to QW. Thus, in order that the effects of the sort discussed in this Letter be measurable, coherent spectroscopic techniques are required. These include the resonant excitation experiments of Deveaud et al. [14] and of Vinattieri et al. [15], and the upconversion and interferometric experiments of Norris et al. [19].

To conclude we have proposed a model for the origin of the radiative width in a system with off-diagonal disorder in the retarded radiative coupling induced by small departures from periodicity. In particular we considered a medium composed of thin identical dipolar layers. We found that even to lowest order the configuration-averaged GF exhibits radiative widths. More realistic models must account for inter- and intra-planar diagonal disorder, the macroscopic crystal surface, nonlinearities associated with the material degrees of freedom, and exciton–phonon interactions. In addition, techniques to successfully carry out higher-order treatments of the configuration average are desired.

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