

Förster energy transfer from a semiconductor quantum well to an organic material overlayer

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Abstract. We predict an efficient electronic energy transfer from an excited semiconductor quantum well to optically active organic molecules of the nearby medium (substrate and/or overlayer). The energy transfer mechanism is of the Förster type and, at semiconductor-organic distances of about 50 Å, can easily be as fast as 10–100 ps, which is about an order of magnitude shorter than the effective exciton lifetime in an isolated quantum well. In such conditions, the Wannier-Mott exciton luminescence is quenched and the organic luminescence is efficiently turned on. We consider both free as well as localized quantum well excitons discussing the dependence of the energy transfer rate on temperature and localization length. A similar mechanism for the non-radiative energy transfer to the organic overlayer molecules from unbound electron-hole pairs excited in the 2D continuum is shown to be much less competitive with respect to other relaxation channels inside the inorganic quantum well (in particular, 2D exciton formation).

PACS. 71.35.Aa Frenkel excitons and self-trapped excitons – 78.20.Bh Theory, models, and numerical simulation – 78.66.Qn Polymers; organic compounds

1 Introduction

A large effort has recently been devoted to the study of organic light emitting diodes [1] and lasers [2]. Förster-like energy transfer between different dye molecules in solid solutions has already been used to achieve light amplification in optically pumped organic thin films [3]. However, light emitting organic materials have poor transport properties compared to inorganic semiconductors and to achieve efficient electrical pumping of such devices is a challenging problem. Prompted by the rapid advances of epitaxial growth techniques for crystalline molecular materials on different organic or inorganic substrates, we consider here a novel hybrid configuration in which both inorganic semiconductors and organic materials are present: the basic idea is to pump the optically active organic molecules via electronic energy transfer from the two-dimensional (2D) Wannier-Mott excitons of a semiconductor quantum well (QW). Hopefully, in this hybrid system, it would be possible to use the efficient electrical pumping of the inorganic semiconductor material to produce luminescence in the organic one.

In organic-inorganic heterostructures, in fact, it is possible to take advantage of the complementary functional properties of both types of materials. This has been

shown to be the case, for instance, in the coherent hybrid Wannier-Mott/Frenkel excitons at a covalent-semiconductor/molecular-solid interface [4]. Such novel excitons have both the large radius of a Wannier-Mott exciton and the large oscillator strength of a Frenkel exciton. They have also been shown to possess large optical nonlinearities [5]. The focus of the present paper is on the basic physics governing the resonant electronic energy transfer from the electronic excited states of the semiconductor quantum well to the excited states of the organic material. While the former have a spatially coherent wavefunction extending over many unit cells (both in the case of free excitons and electron-hole pairs characterized by a center of mass 2D wavevector and in the case of localized excitons described by a slowly varying envelope function), the latter are in general strongly scattered by phonons (incoherent excitons). Usually, the relaxation of the final excited states in the organics is much faster than the back transfer rate (of course, the interesting case is when the luminescence quantum yield of the organics is high); in this respect, our considerations correspond to the Förster picture of energy transfer. Our model calculations, combining a microscopic quantum well model with a macroscopic electro-dynamical description of the organic medium, take into account the dielectric constant discontinuities and can be applied to any multilayer structure. Employing realistic material parameters, the energy transfer mechanism we consider is shown to be fast enough to efficiently quench

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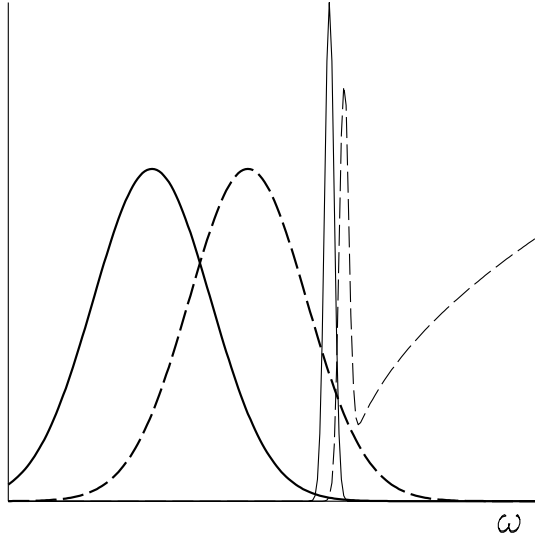


Fig. 1. Schematic inorganic semiconductor QW (thin lines) and organic medium (thick lines) absorption (dashed lines) and luminescence (solid lines) spectra. Notice the overlap of the QW exciton luminescence peak and the broad organic medium absorption.

the Wannier-Mott exciton luminescence and to turn on the organic molecule light emission.

The configuration we consider consists of a semiconductor quantum well sandwiched between two semiconductor barriers, the whole semiconductor structure embedded in a bulk-like organic material (for the sake of simplicity, we choose a symmetric configuration and consider the organic material to be isotropic). The background dielectric constant of the semiconductor material is taken to be real, whereas the total dielectric constant of the organic material has both a real and an imaginary part in the frequency region of interest. In fact, we are interested in an organic material having a broad absorption band in the optical range overlapping with the two-dimensional Wannier-Mott exciton sharp resonance, as sketched in Figure 1. The Förster-like energy transfer rate due to the dipole-dipole interaction can be calculated simply from the Joule losses [6] in the organic material (for the details see Sect. 2 of the present work). These are proportional to the imaginary part of the dielectric function and are produced by the penetration into the organics of the electric field generated by the semiconductor exciton polarization (explicitly taken into account as a source term in the macroscopic electrostatic equations). Neglecting retardation is a valid approximation as the typical exciton center of mass in-plane wavevector is much larger than the wavevector of the corresponding resonant light. We also assume to be in the linear regime in which excitons can be described in the bosonic approximation.

In the present work after the general discussion of the approach used (Sect. 2), we consider three cases: a free exciton in a perfect QW, translationally invariant in two dimensions (Sect. 3) (this case for the L-exciton has already been considered in the short publication [7]);

an exciton, localized due to the QW width fluctuations (Sect. 4); quasi-thermalized free electron-hole pairs in a perfect QW (Sect. 5).

2 Dipole-dipole mechanism of energy transfer: general relations

In this section we justify the macroscopic approach to the problem of the dipole-dipole energy transfer from a 2D semiconductor subsystem to a bulk-like organic subsystem.

We restrict ourselves to the linear approximation, therefore, we only need to consider the behaviour of a single electron-hole pair in the semiconductor QW. Consequently, we consider only two states in the semiconductor subsystem, $|0\rangle$ – the ground state and $|e, h\rangle$ – the excited state, a single electron-hole pair described by the envelope function $\psi(\mathbf{r}_e, \mathbf{r}_h)$, and the conduction and valence band extrema Bloch functions $u_c(\mathbf{r}_e)$ and $u_v(\mathbf{r}_h)$, where $\mathbf{r}_e, \mathbf{r}_h$ are the positions of the electron and the hole. The spin part is not explicitly considered and can be introduced if needed. The normalization is assumed to be

$$\int d^3\mathbf{r}_e d^3\mathbf{r}_h |\psi(\mathbf{r}_e, \mathbf{r}_h)|^2 = 1. \quad (1)$$

We do not specify the form of ψ , it may be the wave function of a 1s-exciton, moving as a whole with the center-of-mass momentum $\hbar\mathbf{K}$, that of a localized exciton, or just a direct product of two 2D plane waves, corresponding to the free carriers (if needed, the in-plane normalization area S may be introduced). The Hamiltonian describing such a pair is:

$$\hat{H}_{e,h} = |e, h\rangle \hbar\omega \langle e, h| + |0\rangle 0 \langle 0|, \quad (2)$$

$\hbar\omega$ being the energy of the e-h pair in the excited state under consideration.

The operator of the QW polarization is

$$\hat{\mathbf{P}}^{QW}(\mathbf{r}) = \mathbf{d}^{vc} \psi(\mathbf{r}_e, \mathbf{r}_h) \Big|_{\mathbf{r}_e=\mathbf{r}_h=\mathbf{r}} |0\rangle \langle e, h| + \text{h.c.}, \quad (3)$$

where

$$\mathbf{d}^{vc} = \int_{u.c.} u_v^*(\mathbf{r})(-e\mathbf{r})u_c(\mathbf{r}) d^3\mathbf{r} \quad (4)$$

is the matrix element of the electric dipole moment between the conduction and valence bands (the integration is performed over the unit cell). Its Cartesian components d_i^{vc} ($i = x, y, z$) may be expressed in terms of the Kane's energy E_0 [8]:

$$|d_i^{vc}|^2 = \frac{e^2 \hbar^2 E_0 c_i^2}{2m_0 E_g^2}, \quad (5)$$

where m_0 is the free electron mass, E_g is the energy gap and c_i is the appropriate symmetry coefficient. In semiconductors of the zinc-blende structure $c_x^{hh} = c_y^{hh} = 1/\sqrt{2}$,

$c_z^{hh} = 0$ (heavy holes) and $c_x^{lh} = c_y^{lh} = 1/\sqrt{6}$, $c_z^{lh} = \sqrt{2/3}$ (light holes).

We assume that the excitations in the organic medium (crystalline or amorphous) because of strong dissipation or spatial disorder cannot be characterized by a wave vector. In this case, the excitations in the organics are localized, thus corresponding to the excited states of a molecule or a group of strongly coupled molecules. Thus, the organic subsystem may be described by the ground state $|g\rangle$, and the excited states $|\mathbf{R}, \nu\rangle$, where \mathbf{R} is the position of the excited state and ν is a quantum number (or a set of quantum numbers), labeling the excited states at the point \mathbf{R} . As we restrict ourselves to the linear regime, only “one-particle” excited states will be considered, which means that two excitations $|\mathbf{R}, \nu\rangle$ and $|\mathbf{R}', \nu'\rangle$ are not allowed to exist simultaneously. For isolated molecules ν is discrete (an integer, enumerating the energy levels of a molecule). However, since we are going to describe a dissipative process, we need to consider a continuum of states (corresponding to the broadening of the molecular levels, which is always present in real media). Therefore, ν must be some continuous quantum number, describing a continuous spectrum, determined by the particular dissipation mechanism, which is not necessary to specify here. We use the following normalization of the states

$$\langle g|g\rangle = 1, \quad (6)$$

$$\langle g|\mathbf{R}, \nu\rangle = 0, \quad (7)$$

$$\langle \mathbf{R}, \nu|\mathbf{R}', \nu'\rangle = \delta(\nu - \nu') \delta(\mathbf{R} - \mathbf{R}'), \quad (8)$$

$$\hat{1}_{org} = |g\rangle\langle g| + \int d^3\mathbf{R} \int d\nu |\mathbf{R}, \nu\rangle\langle \mathbf{R}, \nu|, \quad (9)$$

where $\hat{1}_{org}$ is the unit operator for the organic subsystem. In general, the excited states ν may be arranged in different ways at different points \mathbf{R} , and the set of parameters ν as well as the integration range may vary from point to point. We will not introduce this dependence in an explicit way, but we should keep in mind that the order of integration in (9) cannot be changed. The Hamiltonian of the organic medium is written as

$$\hat{H}_{org} = |g\rangle\langle g| + \int d^3\mathbf{R} \int d\nu |\mathbf{R}, \nu\rangle E_\nu(\mathbf{R}) \langle \mathbf{R}, \nu|. \quad (10)$$

The operator of the organic medium polarization (dipole moment per unit volume) is

$$\hat{\mathbf{P}}^{org}(\mathbf{R}) = \int d\nu |\mathbf{R}, \nu\rangle \mathbf{d}^\nu(\mathbf{R}) \langle g| + \text{h.c.}, \quad (11)$$

where $\mathbf{d}^\nu(\mathbf{R})$ is the matrix element of the dipole moment between the excited and the ground state, generally speaking, including the contributions of both electrons and nuclei:

$$\mathbf{d}^\nu(\mathbf{R}) = \langle \mathbf{R}, \nu| \sum_i (-e_i \hat{\mathbf{r}}_i) |g\rangle, \quad (12)$$

where e_i and $\hat{\mathbf{r}}_i$ are the charge and the position operator of the i th charge in the medium and the sum is taken over all charges, constituting the medium.

We neglect the retarded interaction, since the typical length scales of our problem are of the order of 100 Å, which is much less than the resonant light wavelength (or, equivalently, the typical value of the exciton wave vector is much larger than that of the resonant light). The Hamiltonian of the Coulomb dipole-dipole interaction between the QW and the organic medium, responsible for the energy transfer, is

$$\hat{H}_{int} = - \int d^3\mathbf{R} (\hat{\mathbf{P}}^{org}(\mathbf{R}) \cdot \hat{\mathcal{E}}(\mathbf{R})), \quad (13)$$

where $\hat{\mathcal{E}}(\mathbf{R})$ is the operator of the electric field, produced by the QW polarization $\hat{\mathbf{P}}^{QW}(\mathbf{r})$. The relation between the latter two operators is the same as between the corresponding classical quantities in the electrostatics of continuous media (namely, the electric field is obtained from the polarization via the appropriate Green’s function).

The decay rate (inverse lifetime) of the electron-hole pair is given by the Fermi Golden Rule [9]:

$$\frac{1}{\tau} = \frac{2\pi}{\hbar} \int d^3\mathbf{R} \int d\nu |\mathbf{d}^\nu(\mathbf{R}) \cdot \mathcal{E}^{vc}(\mathbf{R})|^2 \delta(E_\nu - \hbar\omega), \quad (14)$$

where $\mathcal{E}^{vc}(\mathbf{R}) = \langle 0|\hat{\mathcal{E}}(\mathbf{R})|e, h\rangle$. Considering the general expression for the dielectric function [10], which in our normalization of states may be written as

$$\varepsilon_{ij}(\mathbf{R}, \omega) = \delta_{ij} - 8\pi \int d\nu \frac{E_\nu(\mathbf{R}) d_i^\nu(\mathbf{R}) (d_j^\nu(\mathbf{R}))^*}{(\hbar\omega)^2 - (E_\nu(\mathbf{R}))^2 + i\eta\omega}, \quad (15)$$

where the infinitesimal $\eta \rightarrow +0$ prescribes the position of the poles in the lower complex half-plane of ω , the expression above may be identically rewritten as

$$\frac{1}{\tau} = \frac{1}{2\pi\hbar} \int d^3\mathbf{R} \text{Im} \varepsilon_{ij}(\mathbf{R}, \omega) \mathcal{E}_i^{vc}(\mathbf{R}) (\mathcal{E}_j^{vc}(\mathbf{R}))^*, \quad (16)$$

where $\varepsilon_{ij}(\mathbf{R}, \omega)$ is the dielectric function of the absorbing organic medium. Note that the equation (16) multiplied by the energy of the excitation $\hbar\omega$ coincides with the expression for the power dissipated in the medium in the presence of classical external electric field of frequency ω and the amplitude, numerically equal to $\mathcal{E}^{vc}(\mathbf{R})$ [11]. In this respect the quantum mechanical problem is equivalent to that of the classical oscillating polarization inside the QW.

Summarizing, we give the following recipe for calculating the energy transfer rate. Suppose that inside the QW we have the classical macroscopic quasistationary polarization, oscillating with the frequency ω :

$$\mathbf{P}(\mathbf{r}, t) = \mathbf{d}^{vc} \psi(\mathbf{r}, \mathbf{r}) e^{-i\omega t} + \text{c.c.} \quad (17)$$

Then, solve the electrostatical problem (*i.e.*, neglecting retardation) and find the corresponding electric field

$$\mathcal{E}(\mathbf{R}, t) = \mathcal{E}(\mathbf{R}) e^{-i\omega t} + \text{c.c.} \quad (18)$$

The latter, substituted into (16), will give the correct quantum mechanical decay rate if the complex dielectric function of the organics is known (independently of its microscopic structure). Both the microscopic dipole approximation and the macroscopic description of the organic medium are valid as long as the obtained electric field is slowly varying in space on the molecular scale. This condition is fulfilled in all cases considered below, since the typical wave vectors of excitons as well as e-h pairs in the QW are small compared to the inverse lattice constant (and the localization length is larger than the lattice constant). The expression (16) will be the starting point for our further considerations (see also [7]).

3 Free excitons

The energy transfer from free excitons was briefly considered in reference [7]. Here we perform a more detailed study of this case.

First, we specify the geometry of the problem, which is the same for all subsequent sections. We consider a symmetric structure, consisting of a semiconductor QW of thickness L_w between two barriers of thickness L_b each, the whole semiconductor structure being surrounded by thick slabs of an organic material (actually, we assume each slab to be semi-infinite). We assume that in the frequency region here considered the semiconductor background dielectric constant ϵ_b is real and the same for the well and the barrier, while that of the organic material $\tilde{\epsilon}$ is complex. For simplicity we assume the organic material to be isotropic (generalization to the anisotropic case is straightforward). So, the dielectric constant to be used in equation (16) as well as in the Poisson equation below, is

$$\epsilon_{ij}(\mathbf{r}) = \begin{cases} \epsilon_b \delta_{ij}, & |z| < L_w/2 + L_b, \\ \tilde{\epsilon} \delta_{ij}, & |z| > L_w/2 + L_b, \end{cases} \quad (19)$$

where the z -axis is chosen to be along the growth direction, $z = 0$ corresponding to the center of the QW.

We adopt a simplified microscopic quantum mechanical model of a 2D Wannier-Mott exciton, in which the polarization (Eq. (17)) can be taken to vanish for $|z| > L_w/2$ and inside the well to be given by the product of the 1s-wave function of the relative motion of the electron and hole at the origin, with the the lowest subband envelope functions for the electron and hole in the approximation of the infinitely deep well and finally with the wave function of the center-of-mass motion, all of them normalized according to equation (1). Thus, we have

$$\mathbf{P}(\mathbf{r}) = \mathbf{d}^{vc} \sqrt{\frac{2}{\pi a_B^2}} \frac{2}{L_w} \cos^2\left(\frac{\pi z}{L_w}\right) \frac{e^{i\mathbf{K}\mathbf{r}_{\parallel}}}{\sqrt{S}}, \quad (20)$$

where S is the in-plane normalization area, \mathbf{K} is the in-plane wave vector of the center-of-mass motion, $\mathbf{r}_{\parallel} \equiv (x, y)$ – the in-plane component of \mathbf{r} and a_B is the 2D 1s-exciton Bohr radius [8]. We choose as x the direction of the in-plane component of the exciton dipole moment \mathbf{d}^{vc} , preferring to consider the polarization not with respect to the

wave vector, but to some fixed frame. This little complication is justified since next to the free exciton we intend to study the case of the localized exciton, *i.e.*, a system with broken 2D translational symmetry. Evidently, we need to consider two cases: \mathbf{d}^{vc} being parallel and perpendicular to the QW plane. We will refer to them as X and Z polarizations respectively. When dealing only with free excitons in a single well, three modes of different symmetry would be identified: longitudinal (L), transverse (T) and perpendicular (Z). The L and Z modes correspond to the X and Z polarizations above, (their energies are split by the depolarization shift, but this is immaterial for the following). For the T mode the dipole-dipole interaction here considered vanishes [4].

The corresponding electric field $\mathcal{E}(\mathbf{r}) \equiv -\nabla\varphi(\mathbf{r})$ can be obtained from the solution of the Poisson equation (the charge density being $\rho(\mathbf{r}) \equiv -\nabla \cdot \mathbf{P}(\mathbf{r})$)

$$\epsilon(z)\nabla^2\varphi(\mathbf{r}) = 4\pi \nabla \cdot \mathbf{P}(\mathbf{r}), \quad (21)$$

with the appropriate boundary conditions at $z = \pm L_w/2$ and at $z = \pm(L_w/2 + L_b)$, *i.e.*, continuity of the tangential component of the electric field $\mathcal{E}(\mathbf{r})$ and of the normal component of the electric displacement $\mathbf{D}(\mathbf{r}) = \epsilon(z)\mathcal{E}(\mathbf{r})$. Writing $\varphi(\mathbf{r}) = \phi(z) e^{i\mathbf{K}\mathbf{r}_{\parallel}}$, we have the equation for $\phi(z)$:

$$\left[\frac{d^2}{dz^2} - K^2 \right] \phi(z) = \begin{cases} 4\pi\rho(z)/\epsilon_b, & |z| < L_w/2, \\ 0, & |z| > L_w/2, \end{cases} \quad (22)$$

where

$$\rho^{(X)}(z) = iK_x L_w \rho_0 (1 + \cos qz), \quad (23)$$

$$\rho^{(Z)}(z) = -q L_w \rho_0 \sin qz, \quad (24)$$

$$\rho_0 = \sqrt{\frac{2}{\pi a_B^2}} \frac{d^{vc}}{\sqrt{S} L_w}, \quad q \equiv 2\pi/L_w \quad (25)$$

with the boundary conditions that $\phi(z)$ and $\epsilon(z) d\phi(z)/dz$ should be continuous at the four interfaces. The corresponding solution in the organic material (for $z > L_w/2 + L_b$) is given by

$$\phi(z) = \rho_0 C_{\mathbf{K}} e^{-K(z-L_b-L_w/2)}, \quad (26)$$

$$C_{\mathbf{K}}^{(X)} = -\frac{iK_x}{K} \frac{8\pi^2 q}{K(K^2 + q^2)} \times \frac{\sinh(KL_w/2)}{\epsilon_b \sinh(KL_b + KL_w/2) + \tilde{\epsilon} \cosh(KL_b + KL_w/2)}, \quad (27)$$

$$C_{\mathbf{K}}^{(Z)} = \frac{8\pi^2 q}{K(K^2 + q^2)} \times \frac{\sinh(KL_w/2)}{\epsilon_b \cosh(KL_b + KL_w/2) + \tilde{\epsilon} \sinh(KL_b + KL_w/2)}. \quad (28)$$

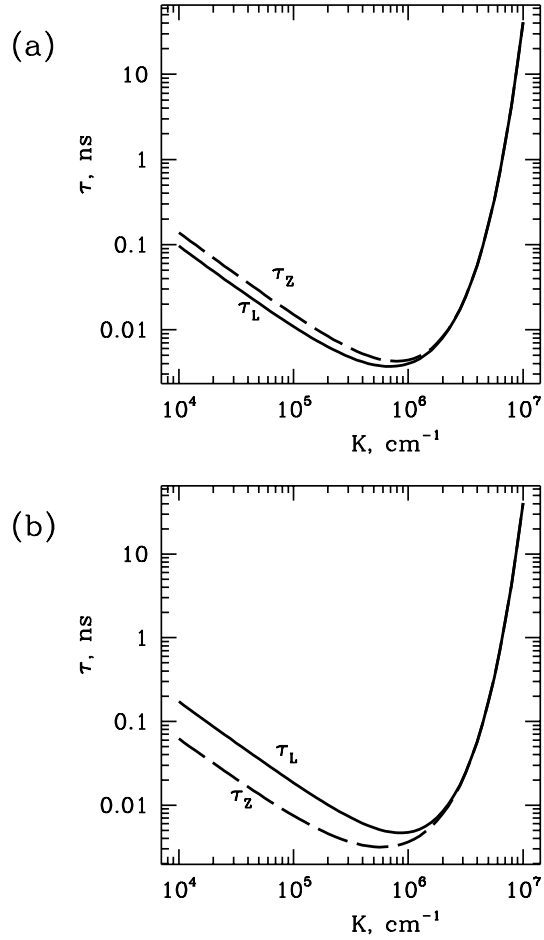


Fig. 2. Free L-exciton (solid line) and Z-exciton (dashed line) lifetime τ (ns) versus the in-plane wave vector K (cm^{-1}). $d^{vc} = 0.1 e a_B$, $L_w = 60 \text{ \AA}$, $L_b = 40 \text{ \AA}$, $\epsilon_b = 6$, $\tilde{\epsilon} = 4 + 3i$ (a); the same, but $\epsilon_b = 4$, $\tilde{\epsilon} = 6 + 3i$ (b).

Thus, the electric field penetrating the organic material is given by

$$\mathcal{E}(\mathbf{r}) = [-i\mathbf{K} + K\mathbf{e}_z]\phi(z) e^{i\mathbf{K}\mathbf{r}_\parallel}. \quad (29)$$

Now we simply substitute this electric field into (16) and get the decay rate:

$$\begin{aligned} \frac{1}{\tau} &= \frac{S}{2\pi\hbar} \text{Im} \tilde{\epsilon} \int_{L_b+L_w/2}^{+\infty} 2K^2 |\phi(z)|^2 dz \\ &= \frac{\text{Im} \tilde{\epsilon}}{\pi^2\hbar} \frac{|d^{vc}|^2}{a_B^2} \frac{K |C_{\mathbf{K}}|^2}{L_w^4}, \end{aligned} \quad (30)$$

where we have considered the absorption only at $z > L_w/2 + L_b$ (considering also the organic material in $z < -L_w/2 - L_b$, τ would be twice shorter).

We evaluate τ from equation (30) for parameters representative of II-VI semiconductor (*e.g.*, ZnSe/ZnCdSe) quantum wells [12]: $\epsilon_b \approx 6$, $d^{vc} \approx 0.1 e a_B$ (about 12 Debye, the Bohr radius is taken to be 25 \AA) and of organic condensed media such as metallophthalocyanines [13] or

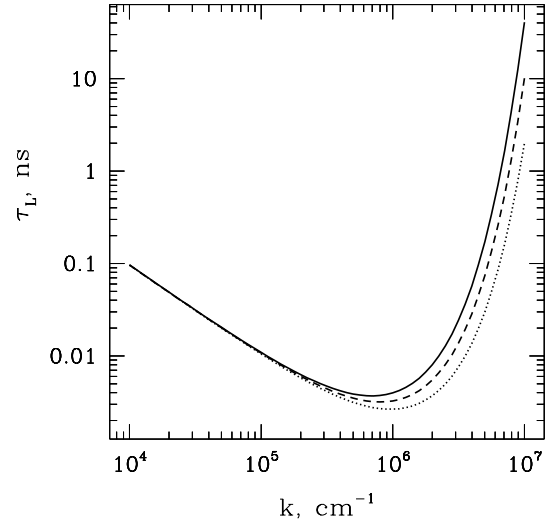


Fig. 3. Free L-exciton lifetime τ (ns) versus the in-plane wave vector K (cm^{-1}) for three well widths: $L_w = 20 \text{ \AA}$ (dotted line), $L_w = 40 \text{ \AA}$ (dashed line), $L_w = 60 \text{ \AA}$ (solid line). Other parameters are $L_b = 40 \text{ \AA}$, $\epsilon_b = 6$, $\tilde{\epsilon} = 4 + 3i$.

fullerenes [14] ($\tilde{\epsilon} \approx 4 + 3i$). We consider two cases: \mathbf{d}^{vc} lying in the QW plane, $\mathbf{K} \parallel \mathbf{d}^{vc}$ (L-exciton) and \mathbf{d}^{vc} perpendicular to the QW plane (Z-excitons). Taking $L_w = 60 \text{ \AA}$, $L_b = 40 \text{ \AA}$, we plot τ_L and τ_Z as functions of K for $\epsilon_b = 6$, $\tilde{\epsilon} = 4 + 3i$ and $\epsilon_b = 4$, $\tilde{\epsilon} = 6 + 3i$ in Figures 2a and 2b. It is seen from the plot, that the lifetime does not depend drastically on the polarization and the real parts of dielectric constants. Figure 3 shows that the dependence on L_w is also weak, while L_b (Fig. 4), when grows, gives an obvious exponential factor (clearly seen from the hyperbolic functions in the denominators of (27, 28)). The most interesting dependence is that on K . We see, that τ exhibits a minimum at $K_{min} \sim 1/L_b$. This dependence may be easily understood if one recalls that the dipole-dipole interaction between two planes behaves like

$$V(\mathbf{K}, z) \sim K e^{-Kz}, \quad (31)$$

which, when substituted into the Fermi Golden Rule gives the correct asymptotics $\tau \sim 1/K$ at $K \rightarrow 0$ and exponential growth at $K \rightarrow \infty$. The dipole-dipole interaction in organic-inorganic heterostructures in a similar planar configuration has been studied previously in reference [4], where the dipole-dipole interaction was responsible for the hybridization of Frenkel and Wannier excitons with the same wave vector.

Considering at first a quasi-thermalized exciton distribution, typical values of K at a temperature $\sim 100 \text{ K}$ are $\sim 3 \times 10^6 \text{ cm}^{-1}$. The corresponding energy transfer lifetime (tens of picoseconds) is much less than the exciton recombination lifetime which is about 200 ps in II-VI semiconductor QWs, as reported by different authors (Ref. [12] and references therein, Ref. [15]). We remark that for the case of free excitons in a quantum well, the effective radiative lifetime (which, assuming a thermal distribution, increases linearly with temperature)

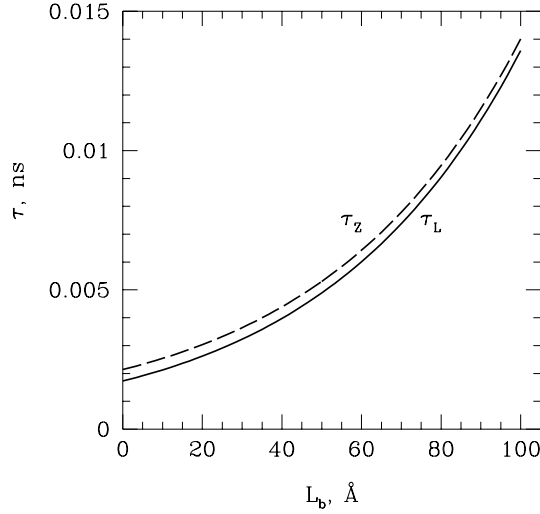


Fig. 4. Free L-exciton (solid line) and Z-exciton (dashed line) lifetime τ (ns) versus the barrier width L_b (Å). $K = 10^6 \text{ cm}^{-1}$, $L_w = 60 \text{ Å}$, $\epsilon_b = 6$, $\tilde{\epsilon} = 4 + 3i$.

is determined by the population transfer from non-radiative excitons with large K to small K excitons undergoing a fast radiative decay [16]. Thus, the dipole-dipole energy transfer mechanism considered here proves to be efficient enough to quench a large fraction of the semiconductor excitons, thereby activating the organic medium luminescence. Moreover, the intraband relaxation of excitons due to the acoustic phonon scattering occurs at time scales of the order of 20–30 ps at 10 K [15], which is larger than the minimal transfer lifetime, obtained here (less than 10 ps for $K_{min} \sim 10^6 \text{ cm}^{-1}$). This makes it possible to excite the QW in a way to produce the initial nonequilibrium distribution of excitons with $K = K_{min}$, tuning the frequency of the excitation pulse to exceed the energy $\hbar\omega_{exc}(K_{min})$ of the exciton with $K = K_{min}$ by one LO-phonon frequency Ω_{LO} (since in II-VI semiconductors the free-carrier-to-exciton relaxation is governed mainly by LO-phonon scattering and happens at times of about 1 ps [15,17–19]), or an integer multiple of Ω_{LO} , if the exciton binding energy is larger than $\hbar\Omega_{LO}$. A numerical estimate for ZnSe gives $\hbar\omega_{exc}(K_{min}) - \hbar\omega_{exc}(K = 0) \sim 1 \text{ meV}$, while $\hbar\Omega_{LO} \approx 31 \text{ meV}$ [15], so that the following kinetics of excitons at $K \sim K_{min}$ is governed mainly by the acoustic phonons. Finally, another possibility would be to resonantly pump excitons with the appropriate K by using a coupling grating configuration [20].

Analogous calculations may be performed for the case of III-V semiconductor materials. We take $\epsilon_b \approx 11$, $d^{vc} \approx 0.05 ea_B$ and plot the L-exciton lifetime versus the wave vector K for several values of L_w (Fig. 5, analogous to Fig. 3 for II-VI materials). All other parameters are the same as in Figure 3. We see that the lifetime is longer compared to that in Figure 3 by about an order of magnitude, which is due to the larger values of a_B and ϵ_b . However, the energy transfer discussed here is still efficient enough because the effective exciton recombination time in III-V materials is also larger (about 1 ns [21]),

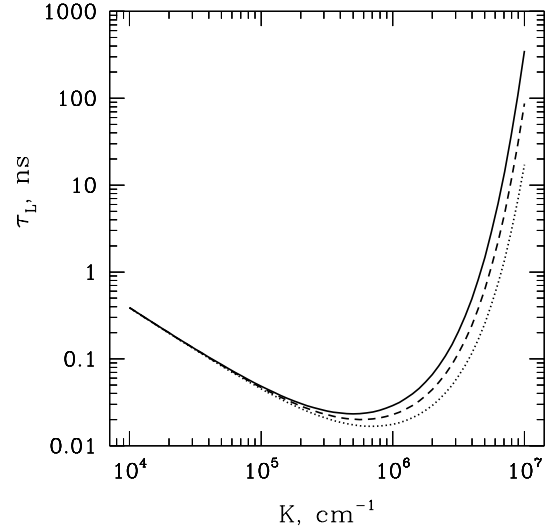


Fig. 5. The same as in Figure 3, but for the III-V-semiconductor compounds ($\epsilon_b = 11$, $d^{vc} = 0.05 ea_B$), all other parameters being the same as in Figure 3.

4 Localized excitons

Now we turn to the situation when the QW width fluctuations, alloy disorder or impurities localize the 2D exciton (such a situation is more frequent for II-VI semiconductor quantum wells than for III-V ones). Then, the wave function of the center-of-mass exciton motion $\Phi(\mathbf{r}_{\parallel})$ is no longer just a plane wave, and the corresponding polarization is given by

$$\mathbf{P}(\mathbf{r}) = \mathbf{d}^{vc} \sqrt{\frac{2}{\pi a_B^2}} \frac{2}{L_w} \cos^2\left(\frac{\pi z}{L_w}\right) \Phi(\mathbf{r}_{\parallel}), \quad (32)$$

which implies that $\Phi(\mathbf{r}_{\parallel})$ is normalized according to

$$\int d^2\mathbf{r}_{\parallel} |\Phi(\mathbf{r}_{\parallel})|^2 = 1. \quad (33)$$

The solution of the Schrödinger equation for a particle in the random potential, caused by the QW width fluctuations and the alloy disorder is beyond the scope of the present paper (much work has been done in this field, *e.g.* see [22] and references therein). We can mention only some general properties that $\Phi(\mathbf{r}_{\parallel})$ should have: (i) it should be localized within some distance $L \gtrsim L_w$, (ii) it should be smooth and without nodes. As a consequence, its spatial Fourier expansion should contain mainly the components with wave vectors $K \lesssim 1/L$.

Expanding the wave function $\Phi(\mathbf{r}_{\parallel})$, the charge density $\rho(\mathbf{r})$ and the potential $\varphi(\mathbf{r})$ into plane waves

$$\begin{aligned} \Phi(\mathbf{r}_{\parallel}) &= \int \frac{d^2\mathbf{K}}{(2\pi)^2} \Phi_{\mathbf{K}} e^{i\mathbf{K}\mathbf{r}_{\parallel}}, \\ \rho(\mathbf{r}) &= \int \frac{d^2\mathbf{K}}{(2\pi)^2} \rho_{\mathbf{K}}(z) e^{i\mathbf{K}\mathbf{r}_{\parallel}}, \\ \varphi(\mathbf{r}) &= \int \frac{d^2\mathbf{K}}{(2\pi)^2} \varphi_{\mathbf{K}}(z) e^{i\mathbf{K}\mathbf{r}_{\parallel}}, \end{aligned} \quad (34)$$

we again obtain equation (22), but the charge density is now given by

$$\rho_{\mathbf{K}}^{(X)}(z) = iK_x L_w \tilde{\rho}_0 L \Phi_{\mathbf{K}} (1 + \cos qz), \quad (35)$$

$$\rho_{\mathbf{K}}^{(Z)}(z) = -q L_w \tilde{\rho}_0 L \Phi_{\mathbf{K}} \sin qz, \quad (36)$$

$$\tilde{\rho}_0 = \sqrt{\frac{2}{\pi a_B^2} \frac{d^{vc}}{L L_w^2}}. \quad (37)$$

The solution is

$$\varphi_{\mathbf{K}}(z) = \tilde{\rho}_0 L \Phi_{\mathbf{K}} C_{\mathbf{K}} e^{-K(z-L_b-L_w/2)}, \quad (38)$$

with the same $C_{\mathbf{K}}$, given by (27, 28). For the decay rate we obtain:

$$\frac{1}{\tau} = \frac{\text{Im } \tilde{\epsilon}}{2\pi\hbar} \int_{L_b+L_w/2}^{+\infty} dz \int \frac{d^2\mathbf{K}}{(2\pi)^2} 2K^2 |\varphi_{\mathbf{K}}(z)|^2 \quad (39)$$

$$= \frac{\text{Im } \tilde{\epsilon}}{\pi^2\hbar} \frac{|d^{vc}|^2}{a_B^2} \frac{1}{L_w^4} \int \frac{d^2\mathbf{K}}{(2\pi)^2} K |\Phi_{\mathbf{K}}|^2 |C_{\mathbf{K}}|^2. \quad (40)$$

It is possible to get some information about the decay rate (40) based only on general properties of the wave function, mentioned above. We have three length scales in our problem: L_w , L_b and L . First, we have the condition $L_w \lesssim L$. Since wave vectors with $KL \gtrsim 1$, being cut off by $|\Phi_{\mathbf{K}}|^2$, do not contribute to the integral, we may set $KL_w \rightarrow 0$. The subsequent analysis depends on the relation between L_b and L .

If $L_b \ll L$, we may put $KL_b \rightarrow 0$ as well. Then we have

$$\frac{C_{\mathbf{K}}^{(X)}}{L_w^2} \rightarrow -\frac{2\pi i}{\tilde{\epsilon}} \frac{K_x}{K}, \quad \frac{C_{\mathbf{K}}^{(Z)}}{L_w^2} \rightarrow \frac{2\pi}{\epsilon_b} \quad (41)$$

and the integral may be estimated as

$$\frac{1}{\tau_X} = A \frac{2}{\hbar} \frac{\text{Im } \tilde{\epsilon}}{|\tilde{\epsilon}|^2} \frac{|d^{vc}|^2}{a_B^2} \frac{1}{L}, \quad \frac{1}{\tau_Z} = A \frac{4}{\hbar} \frac{\text{Im } \tilde{\epsilon}}{\epsilon_b^2} \frac{|d^{vc}|^2}{a_B^2} \frac{1}{L} \quad (42)$$

up to a numerical factor $A \sim 1$, determined by the detailed shape of $\Phi_{\mathbf{K}}$. We have set the average value of K over the wave function $\Phi_{\mathbf{K}}$ to be A/L and, if for the X polarization we assume $\Phi_{\mathbf{K}}$ to be cylindrically symmetric (which may be considered as the average over the realizations of disorder), then the numerical factor A is the same for both cases.

In the opposite limit, $L_b \gg L$ (which also implies $L_b \gg L_w$), we may set $\Phi_{\mathbf{K}} = \Phi_{\mathbf{K}=0}$ since the values of \mathbf{K} , contributing to the integral, are determined by $C_{\mathbf{K}}$, (namely, $K \lesssim 1/L_b$) which in this limit takes the form

$$\frac{C_{\mathbf{K}}^{(X)}}{L_w^2} \rightarrow -\frac{iK_x}{K} \frac{4\pi}{(\tilde{\epsilon} + \epsilon_b) e^{KL_b} + (\tilde{\epsilon} - \epsilon_b) e^{-KL_b}}, \quad (43)$$

$$\frac{C_{\mathbf{K}}^{(Z)}}{L_w^2} \rightarrow \frac{4\pi}{(\tilde{\epsilon} + \epsilon_b) e^{KL_b} - (\tilde{\epsilon} - \epsilon_b) e^{-KL_b}}. \quad (44)$$

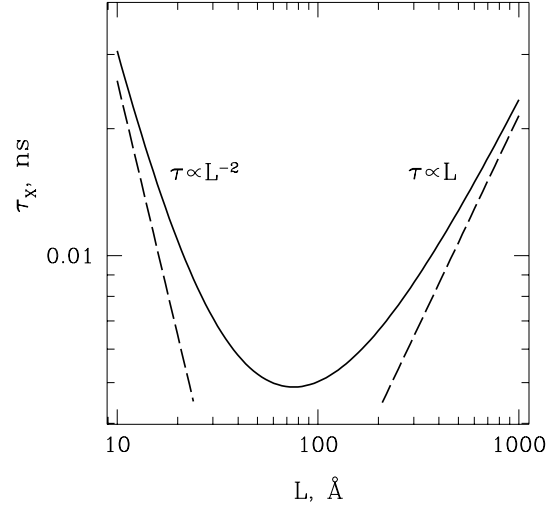


Fig. 6. Localized X-exciton lifetime τ (ns) versus the localization length L (\AA) (solid line) along with the limiting cases $L \ll L_b$ and $L \gg L_b$ (dashed lines). $L_w \ll L$, $L_b = 40 \text{ \AA}$, $\epsilon_b = 6$, $\tilde{\epsilon} = 4 + 3i$.

Estimating the integral, we have

$$\frac{1}{\tau_X} = A \frac{1}{\pi\hbar} \frac{\text{Im } \tilde{\epsilon}}{|\tilde{\epsilon} + \epsilon_b|^2} \frac{|d^{vc}|^2}{a_B^2} \frac{|\Phi_{\mathbf{K}=0}|^2}{L_b^3}, \quad (45)$$

where $|\Phi_{\mathbf{K}=0}|^2 \sim L^2$, which follows from the normalization condition. The expression for $1/\tau_Z$ differs from this by an additional factor of 2 and the factor A may be different in the two cases. It is determined by the values of $\tilde{\epsilon}$, ϵ_b

$$A = \int_0^\infty \frac{4\xi^2 d\xi}{\left| \frac{\tilde{\epsilon} + \epsilon_b}{|\tilde{\epsilon} + \epsilon_b|} e^\xi \pm \frac{\tilde{\epsilon} - \epsilon_b}{|\tilde{\epsilon} + \epsilon_b|} e^{-\xi} \right|^2}, \quad (46)$$

and is bounded by

$$\frac{\pi^2}{12} = \int_0^\infty \frac{\xi^2 d\xi}{\cosh^2 \xi} < A < \int_0^\infty \frac{\xi^2 d\xi}{\sinh^2 \xi} = \frac{\pi^2}{6}. \quad (47)$$

So we see that at $L \ll L_b$ the decay rate is proportional to L^2 , at $L \gg L_b$ – to L^{-1} , therefore it has a maximum at some $L \sim L_b$. This is in agreement with the results of the previous section, since the plane waves, giving the largest contribution to the wave function $\Phi(\mathbf{r}_{\parallel})$ and thus determining the decay rate, have the values of wave vector of the order of $K \sim 1/L$ and we have seen that wave vectors, corresponding to the shortest lifetimes were $K_{min} \sim 1/L_b$.

To illustrate these considerations, we choose a specific example of the localized wave function – that of the ground state in the isotropic parabolic potential:

$$\Phi_{\mathbf{K}} = \sqrt{4\pi} L e^{-K^2 L^2/2}, \quad (48)$$

which obviously has all the necessary features mentioned in the beginning of this section. For this wave function

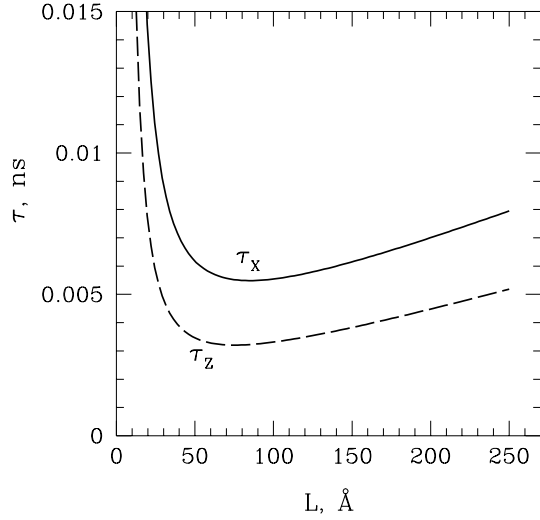


Fig. 7. Localized X-exciton (solid line) and Z-exciton (dashed line) lifetime τ (ns) versus the localization length L (Å). $L_w = 10$ Å, $L_b = 40$ Å, $\epsilon_b = 6$, $\tilde{\epsilon} = 4 + 3i$.

the integral in (40) may be evaluated numerically for arbitrary parameters L , L_w , L_b (we remind that only physically relevant are $L \gtrsim L_w$). The results of the calculation (τ versus L) are plotted in Figures 6 and 7 along with the asymptotic dependencies for $L_b = 40$ Å, $\epsilon_b = 6$, $\tilde{\epsilon} = 4 + 3i$ (we have set $L_w \rightarrow 0$ for the plots in Fig. 6, but a more specific value $L_w = 10$ Å was chosen for Fig. 7). In the limit $L \gg L_b$ the coefficient $A = \sqrt{\pi}/2$, the coefficient for $L \ll L_b$, given by (46) was calculated numerically.

We also plot the dependence, analogous to that in Figure 7, for parameters typical of III-V-semiconductors: $d^{vc}/ea_B = 0.05$ and $\epsilon_b = 11$ (Fig. 8); analogously to the previous section, we obtain larger lifetimes, than those for II-VI-semiconductors.

5 Free carriers

Finally, we consider the situation, when the carriers are not bound into excitons, thus forming a 2D plasma. We assume electrons and holes to be quasi-thermalized within corresponding bands (considering only one subband for each band), distributed according to the ideal Fermi gas law with the temperature T and the chemical potentials μ^e and μ^h for electrons and holes respectively (we set the Boltzmann constant equal to unity everywhere in this section, measuring the temperature in the units of energy). For an ideal 2D Fermi gas the chemical potential μ is related to the concentration n via

$$e^{\mu(n,T)/T} = e^{\mu_0(n)/T} - 1, \quad \mu_0(n) = \frac{2\pi\hbar^2 n}{m}, \quad (49)$$

where μ_0 is the chemical potential at the zero temperature, m is the corresponding (electron or hole) mass, spin degeneracy is not taken into account (*i.e.*, all concentrations are those of particles with a given spin).

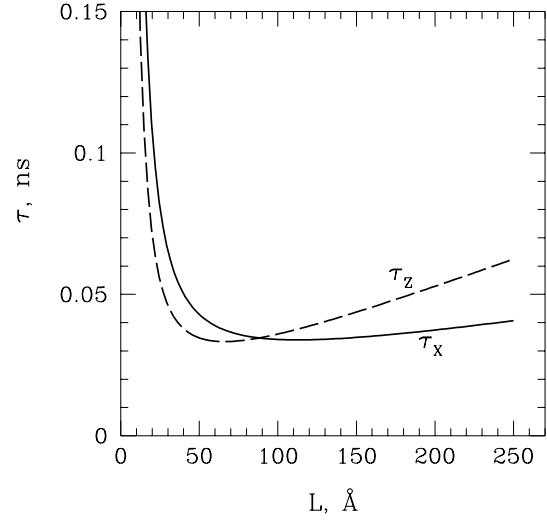


Fig. 8. The same as in Figure 7, but for the III-V-semiconductor compounds, other parameters being the same as in Figure 7.

If N_e (N_h) is the total number of electrons (holes) in the system, then the total recombination rate due to the energy transfer here considered can be written as

$$-\frac{dN_e}{dt} = -\frac{dN_h}{dt} = \sum_{\mathbf{k}, \mathbf{k}'} \frac{f_{\mathbf{k}}^e f_{\mathbf{k}'}^h}{\tau(\mathbf{k} + \mathbf{k}')}, \quad (50)$$

where $f_{\mathbf{k}}^e$, $f_{\mathbf{k}'}^h$ are the Fermi occupation numbers and $\tau(\mathbf{k} + \mathbf{k}')$ is the corresponding recombination time for a single electron-hole pair with 2D wave vectors \mathbf{k} and \mathbf{k}' . It depends only on the total momentum $\mathbf{K} = \mathbf{k} + \mathbf{k}'$ since the wave function of the pair is simply

$$\begin{aligned} \psi(\mathbf{r}, \mathbf{r}) &= \frac{2}{L_w} \cos^2\left(\frac{\pi z}{L_w}\right) \frac{e^{i(\mathbf{k} + \mathbf{k}')\mathbf{r}_{\parallel}}}{S} \\ &= \frac{2}{L_w} \cos^2\left(\frac{\pi z}{L_w}\right) \frac{e^{i\mathbf{K}\mathbf{r}_{\parallel}}}{S}, \end{aligned} \quad (51)$$

the in-plane motion being described by a direct product of two plane waves. The corresponding polarization is given by (20), where the factor $\sqrt{2/(\pi a_B^2)}$ must be replaced by $1/\sqrt{S}$. For the pair recombination rate we obtain the expression (30), multiplied by the factor $\pi a_B^2/2S$:

$$\frac{1}{\tau(\mathbf{K})} = \frac{1}{2\pi\hbar} \frac{|d^{vc}|^2}{SL_w^4} \text{Im} \tilde{\epsilon} K |C_{\mathbf{K}}|^2, \quad (52)$$

which depends on the normalization area. Such a dependence is however immaterial: rewriting (50) for the 2D densities $n_{e,h} = N_{e,h}/S$ and transforming the sum into an integral, the total recombination rate becomes:

$$-\frac{dn_e}{dt} = -\frac{dn_h}{dt} = \int \frac{d^2\mathbf{k} d^2\mathbf{k}'}{(2\pi)^4} \frac{S f_{\mathbf{k}}^e f_{\mathbf{k}'}^h}{\tau(\mathbf{k} + \mathbf{k}')}. \quad (53)$$

In the general case this integral cannot be calculated analytically, but the situation simplifies significantly in

the classical limit ($T \gg \mu_0^e, \mu_0^h$). The occupation numbers then reduce to the Boltzmann distribution:

$$f_{\mathbf{k}}^{e,h} = \exp\left(-\frac{\hbar^2 k^2}{2m_{e,h}T} + \frac{\mu^{e,h}(n_{e,h}, T)}{T}\right) \quad (54)$$

and, if we perform the change of the integration variables

$$\mathbf{k}, \mathbf{k}' \longrightarrow \mathbf{K} = \mathbf{k} + \mathbf{k}', \tilde{\mathbf{k}} = \frac{m_h \mathbf{k} - m_e \mathbf{k}'}{m_e + m_h} \quad (55)$$

($\tilde{\mathbf{k}}$ being the momentum of the relative motion) and use the fact that τ does not depend on $\tilde{\mathbf{k}}$, the integral is factorized. The Gaussian integral over $\tilde{\mathbf{k}}$ gives a normalization factor and finally we obtain

$$\begin{aligned} -\frac{dn_{e,h}}{dt} &= \frac{\hbar n_e n_h \text{Im} \tilde{\varepsilon} |d^{vc}|^2}{(m_e + m_h)T L_w^4} \\ &\times \int \frac{d^2 \mathbf{K}}{(2\pi)^2} K |C_{\mathbf{K}}|^2 \exp\left(-\frac{\hbar^2 K^2}{2(m_e + m_h)T}\right). \end{aligned} \quad (56)$$

If we introduce the “thermal length” L_T defined by

$$L_T^2 \equiv \frac{\hbar^2}{2(m_e + m_h)T}, \quad (57)$$

the integral in (56) formally coincides with that in (40) for the Gaussian wave function. Then the recombination rate may be written as

$$\begin{aligned} -\frac{dn_{e,h}}{dt} &= n_e n_h S \int \frac{d^2 \mathbf{K}}{(2\pi)^2} \frac{4\pi L_T^2}{\tau(\mathbf{K})} e^{-K^2 L_T^2} \\ &= \frac{\pi a_B^2}{2} \frac{n_e n_h}{\tau_{loc}}, \end{aligned} \quad (58)$$

where τ_{loc} is the lifetime of the exciton from the previous section with the Gaussian wave function (48), localized at the length L_T . Of course, the recombination rate of free carriers cannot depend on the exciton Bohr radius and a_B^2 in the numerator (58) just cancels the analogous factor in τ_{loc} .

The process under consideration is a bimolecular decay rather than a monomolecular one. To determine the appropriate characteristic time we write down the kinetic equations (53) in the form:

$$\frac{dn_e}{dt} = \frac{dn_h}{dt} = -n_e n_h g(n_e, n_h, T). \quad (59)$$

In the classical limit $g = g(T)$ does not depend on the concentrations at all. Suppose for a while that this situation takes place. We assume that the concentrations are equal $n_e = n_h = n$ (*e.g.*, this is the case for an optically pumped undoped well). Then the solution for $n(t)$ is

$$n(t) = \frac{n(0)}{1 + gn(0)t}. \quad (60)$$

So, the transfer rate may be defined as $gn(0)$ in the classical case and in the situation when the degeneracy of the

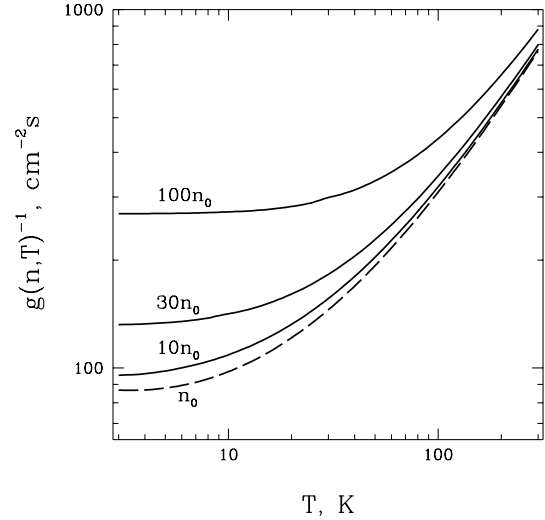


Fig. 9. The inverse recombination rate constant $g(n, T)^{-1}$ (cm^{-2}s) versus temperature T (K) for different carrier concentrations $n_e = n_h = n = 10n_0, 30n_0, 100n_0$ (solid lines, $n_0 \equiv 10^{10} \text{ cm}^{-2}$). The dashed line ($n = n_0$) also represents the classical limit, which is concentration independent. The parameters are $\epsilon_b = 6$, $m_e = 0.16 m_0$, $m_h = 0.6 m_0$, $L_w = 60 \text{ \AA}$, $L_b = 40 \text{ \AA}$, $\tilde{\varepsilon} = 4 + 3i$.

plasma becomes significant, $ng(n)$ has the meaning of the instantaneous transfer rate. In the following we analyze the behaviour of the inverse recombination rate constant $g(n, T)^{-1}$ whose dependence on n is then a measure of degeneracy of the electron-hole gas, and which, divided by n , gives the effective lifetime.

The results of the numerical integration of (53) for the II-VI parameters ($\epsilon_b = 6$, $m_e = 0.16 m_0$, heavy holes with $m_h = 0.6 m_0$, X-polarization), 60 \AA well, 40 \AA barrier, $\tilde{\varepsilon} = 4 + 3i$, are shown in Figure 9. We plot $g(n, T)^{-1}$ as a function of temperature in the range $3 \text{ K} \leq T \leq 300 \text{ K}$ for several values of the concentrations 10^{11} cm^{-2} , $3 \times 10^{11} \text{ cm}^{-2}$, 10^{12} cm^{-2} along with the limiting case (58), which is reached even at low temperatures for concentrations lower than $n_0 = 10^{10} \text{ cm}^{-2}$ (actually, the curve for $n = n_0$ is indistinguishable from the classical curve in all the temperature range). This is in agreement with the fact that the corresponding “Fermi energies” for electrons and holes are $\mu_0^e(n_0) = 3.5 \text{ K}$, $\mu_0^h(n_0) = 0.93 \text{ K}$. The “thermal length” at $T = 3 \text{ K}$ is $L_T = 140 \text{ \AA}$ (correspondingly, for $T = 300 \text{ K}$ $L_T = 14 \text{ \AA}$), which explains the monotony of the classical dependence on T : it is nothing different, but the *left* part of the plot in Figure 6, properly renormalized to the case of free carriers.

We see that the corresponding effective lifetime ($\tau = 1/(ng)$, according to the above considerations) is of the order of 1 ns. The relaxation to excitons by means of LO-phonon emission, as already has been mentioned, takes about 1 ps in II-VI materials [19], if the density is not too high ($n \ll 1/a_B^2$). This means that the carriers actually do not even reach the thermal quasi-equilibrium, quickly binding into excitons; the results of the two previous sections are thus much more relevant. However, at higher

densities, $n \gtrsim 1/a_B^2$, when the plasma phase is favoured with respect to the excitonic phase [23,24] and, on the other hand, the effective energy transfer rate itself is increased ($1/\tau \propto n$, for arbitrary momentum distribution, not necessarily equilibrium), the rate of the dipole energy transfer from the free carriers may be comparable with rates of other processes, giving some information on the carrier kinetics as well.

Calculations were also performed for the case of III-V-type materials. We do not present them on a separate figure since they have no significantly different features with respect to what has already been mentioned in the previous sections.

6 Conclusions

We have studied the dipole-dipole energy transfer from an excited semiconductor quantum well to a light-emitting organic medium, considering different excitation regimes and experimental conditions. According to our results, the kinetics of the initial free carrier population (produced, *e.g.*, by the electrical pumping) is not significantly changed by the presence of the organic medium, since the energy transfer from free carriers turns out to be slower than the process of exciton formation (unless excitation density and temperature are very high). On the other hand, the subsequent evolution of free or localized excitons is strongly affected by the presence of the organic medium. In an isolated QW the effective lifetime of the exciton distribution may be of several hundred ps. However, excitons coupled to the organic medium efficiently transfer their energy to the organic molecules before they can recombine inside the QW. For quantum wells based on the II-VI semiconductors and in a realistic configuration, such transfer may occur at time scales of the order of 10 ps. This simple physical picture shows that the system studied here may be promising for optical devices based on hybrid organic-inorganic structures, combining the good transport properties of inorganic semiconductors and the large oscillator strengths of organic materials.

Finally, we wish to stress that our model calculations can be easily applied to any hybrid multilayer structure and could be generalized to the case in which the structure is embedded in a microcavity.

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