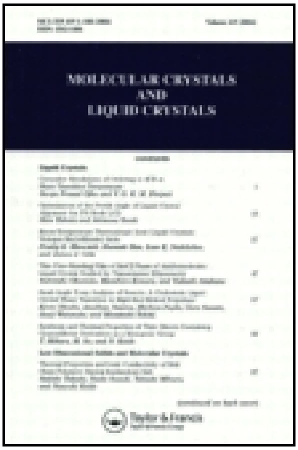

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COUPLING OF EXCITONS IN PARALLEL ORGANIC AND SEMICONDUCTOR QUANTUM WIRES

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Abstract We study the resonance energy transfer between parallel neighboring organic and semiconductor quantum wires which is caused by the resonance dipole-dipole interaction between the wires. Due to this interaction, a new hybrid excited state arises which is a mixture of Frenkel and Wannier-Mott excitons. For the transition dipole moments oriented perpendicular to the wires, the interwire hybridization strength is nonzero even for small exciton wavevectors along the wires and decays rather slowly with increasing interwire spacing. This is in contrast to two-dimensional quantum wells where the dipole-dipole coupling decays fast with the increasing interwell distance and takes place only for nonzero excitonic wavevectors. The hybrid wire excitons possess both a large radius and a relatively large oscillator strength which might be especially interesting for the nonlinear optics.

As noted recently[1], a new type of excitonic states could exist in composite heterostructures fabricated of organic–inorganic semiconductor nanolayers. This hybrid excitonic state could arise due to resonant mixing of Frenkel and Wannier-Mott excitons. The new excitons would possess strong oscillator strengths (typical for Frenkel excitons) and high sensitivity to external perturbations (typical for Wannier-Mott excitons). However, for the system considered[1] (a two-dimensional (2D) organic molecular layer and a neighboring semiconductor 2D quantum well), the electrostatic coupling of Frenkel and Wannier excitons vanishes in the range of small 2D exciton wave vectors. This is because the electric field of a uniformly polarized layer vanishes outside the layer. Thus, the hybridization effects are small just for the most interesting small wave vector excitons which interact actively with light.

Here we consider another composite system – parallel organic and semiconductor quantum wires - and show that in this system the hybridization of Frenkel and Wannier-Mott excitons is especially effective just for excitons with small wave vectors. The resonance coupling of Frenkel and Wannier-Mott excitons is governed by the hybridization parameter

$$\Gamma(k) = \langle F, k|H_{\text{int}}|W, k\rangle, \tag{1}$$

where a one-dimensional (1D) wave vector k characterizes the states $|F, k\rangle$ and $|W, k\rangle$ of Frenkel and Wannier-Mott excitons, respectively; the interaction Hamiltonian H_{int} for a pair of wires is given by

$$H_{\text{int}} = -\int \mathbf{E}(\mathbf{r})\mathbf{P}(\mathbf{r})d\mathbf{r}. \tag{2}$$

Here the integration is performed over the semiconductor wire; $\mathbf{P}(\mathbf{r})$ is the transition polarization operator for Wannier-Mott excitons; and $\mathbf{E}(\mathbf{r})$ is the operator of the electric field created in the semiconductor wire by the exciton in the organic wire.

The resonance coupling of Frenkel and Wannier-Mott excitons results in the appearance of hybrid excited states

$$|\Psi, k\rangle = A(k)|F, k\rangle + B(k)|W, k\rangle. \tag{3}$$

Here

$$\begin{aligned} |A(k)| &= \frac{|\Gamma(k)|}{\sqrt{[E_F(k) - E(k)]^2 + \Gamma^2(k)}}, \\ B(k) &= \frac{\Gamma(k)}{E(k) - E_W(k)}A(k), \end{aligned} \tag{4}$$

where $E_{F(W)}(k)$ are the energies of Frenkel (Wannier-Mott) excitons and the energies $E(k)$ of the hybrid state are given by

$$\begin{aligned} E_\pm(k) &= [E_F(k) + E_W(k)]/2 \pm \\ &\quad \sqrt{[E_F(k) - E_W(k)]^2/4 + \Gamma^2(k)}. \end{aligned} \tag{5}$$

Now we estimate the hybridization parameter $\Gamma(k)$ Eq.(1). The state corresponding to the Frenkel exciton is represented as

$$|F, k\rangle = \frac{1}{\sqrt{N_F}} \sum_{\mathbf{n}} \exp{(ikz_{\mathbf{n}})} A^\dagger_{\mathbf{n}}|0\rangle, \tag{6}$$

where N_F is the total number of molecular sites and $|0\rangle$ is the exciton vacuum state. For the Wannier-Mott exciton state we take the following representation:

$$|W, k\rangle = \frac{1}{\sqrt{L_W}} \int dz_e dz_h \exp{(ik\frac{z_e + z_h}{2})}\Phi_0(z_e - z_h)\Psi^\dagger_{e0}(z_e)\Psi^\dagger_{h0}(z_h)|0\rangle. \tag{7}$$

The operators $\Psi^{\dagger}_{e(h)0}(z)$ correspond to the lowest ($l = 0$) state of the transverse motion in the operator expansion over transverse modes ϕ_l:

$$\Psi^{\dagger}_{e(h)}(\mathbf{r}) = \sum_{l} \Psi^{\dagger}_{e(h)l}(z)\phi^{*}_{e(h)l}(\rho), \tag{8}$$

where $\mathbf{r} = (\rho, z)$. The function $\Phi_0(z_e - z_h)$ in Eq.(7) describes the relative 1D motion of the bound electon and the hole. We consider only the lowest transverse state. Using Eqs. (2-8) we obtain a closed but somewhat complicate expression[2] for the hybridization parameter Γ. As follows from this equation, the interwire coupling is suppressed exponentially for excitons with wave vectors $k > 1/R$, i.e. for most part of the Brillouin zone; here R is the the interwire distance. On the contrary, coupling of excitons with relatively small wave vectors $k < 1/R$ is quite effective. This is in contrast to the case of a 2D system of quantum wells where the coupling at small wave vectors is suppressed because the electric field outside a uniformly polarized layer vanishes. The range of small wave vectors $k \sim 1/\lambda \ll 1/R$ is of special interest as excitations with such wave vectors may be created straightforwardly by light of wavelength λ. In the leading order in $kR \ll 1$, the hybridization parameter $\Gamma(k)$ has the following form

$$\Gamma(k) = \frac{f_{eh}}{\epsilon R^2}\sqrt{\frac{2S_F}{a_{1B}v_F}}(\mu^F_y \mu^W_y - \mu^F_x \mu^W_x), \tag{9}$$

where S_F and v_F are the cross-section and the volume of an elementary cell for the molecular wire; the 1D exciton ground state wave function $\Phi_0(0) = 1/\sqrt{2a_{1B}}$ in the strong-confinement limit has been expressed in terms of the 1D Bohr radius $a_{1B} = (a_0/2)\sqrt{E_0/E_1}$, with a_0 and E_0 being the Bohr radius and the ground state energy of the bulk exciton, respectively; E_1 is the ground state energy of 1D exciton[3,4]; f_{eh} is an overlap integral between the electron and hole wavefunctions $\phi_{e0}(\rho)$ and $\phi_{h0}(\rho)$. We used also the simplifying assumption that the interwire distance R is large in comparison with the thickness of the wires.

Note that the excitonic polarization component along the wires does not contribute to Γ in the leading order in kR. This is due to the obvious fact that a uniform longitudinal polarization is not accompanied by the appearance of an electric charge. To estimate the value of Γ we use the following parameter values: $a_{1B} = 30$Å, $\mu^F = 5$ Debye, $\mu^W = 10$ Debye, $S_F = (50\text{Å})^2$, $v_F = 100$ (Å)^3, $R = 50$ Å, $f_{eh} = 1$, $\epsilon = 3$ and obtain $\Gamma \approx 5.4$ meV.

The resonance coupling of the Frenkel and Wannier-Mott excitons results in the appearance of hybrid excited states $|\Psi, k\rangle = A(k)|F, k\rangle + B(k)|W, k\rangle$. The coupling is strong ($A \sim B$) when the energies $E_F(k), E_W(k)$ of Frenkel and Wannier-Mott excitons are in resonance: $|E_F(k) - E_W(k)| \sim |\Gamma(k)|$. Outside the resonance range, the coupling is governed by the parameter $\Gamma^2/(E_F - E_W)$ and is rather small. The condition of resonance is rather strict for the considered range of parameters and requires a careful choice of materials for both wires. The exciton linewidths should be small as compared to Γ. In general, the excitonic linewidth is determined by the radiative decay and by other dephasing processes. The radiative contribution to the Frenkel exciton linewidth is estimated as $\gamma_{\mathrm{rad}} \approx 0.1$ meV. This quantity is relatively small which means that the main contribution to the excitonic linewidth in quantum wires is of nonradiative nature and due to scattering by structural imperfections, surface corrugation, etc. At low temperatures, the nonradiative linewidth may be ~ 1 meV for Wannier excitons, and even smaller for Frenkel excitons. These linewidths may be thus smaller than the resonant splitting $2\Gamma \approx 11$ meV of the hybrid excitations.

In the resonant case the size of the hybrid state is comparable with that for Wannier-Mott excitons, i.e. it is much larger than the radius of Frenkel excitons. This determines a high sensitivity of the hybrid states to an external field. In addition to the large radius, the hybrid excitons possess also a relatively large oscillator strength which is typical for Frenkel excitons. This might be especially interesting for the nonlinear optics.

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