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FERMI RESONANCE INTERFACE SOLITONS

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We discuss the nonlinear dynamics of interface between two layers of organic semiconductors in the case of Fermi resonance, which occurs when the energy $\hbar\omega^c$ of excitation on one side of the interface is approximately equal to $2\hbar\omega^b$, where $\hbar\omega^b$ is the excitation energy on the other side. We demonstrate that Fermi resonance interaction across the interface gives rise to nonlinear plane waves propagating along the interface and also to localized at interface $2D$ solitons.

1. It is well known that the epitaxial growth of inorganic semiconductors is limited to materials with small lattice mismatch. The organic materials, on the contrary, are bound by weak van der Waals forces what allows the layering of materials with different lattice constants. Therefore great efforts have been made recently with the aim to create strongly ordered crystalline organic thin films and multilayer structures [1-5]. The theoretical analysis of linear and nonlinear optical properties of multilayer organic structures became also topical and some problems in this field have been discussed in recent papers [6-14].

One of the mechanisms of a "hand-made" optical nonlinearity of multilayer organic structures pointed out recently [8,9] is based on the Fermi resonance between excitations of neighbouring layers. In Refs [9, 10, 12] such interface Fermi resonance was discussed for the case when the energy of two excitons $2\hbar\omega^b$ in one layer is close to some exciton energy $\hbar\omega^c$ in the neighbouring layer. In these papers the new states - quantum and classical Fermi Resonance Interface Modes (FRIM) were found which appeared due to an intermolecular anharmonic interaction across the interface. This phenomenon extends the usual Fermi resonance in the bulk molecular crystals and can be important, as it was shown in [8-14], in the investigations of linear as well as nonlinear optical properties of multilayer structures.

In the limit of strong pumping, i.e. at large occupation numbers of excitations, it is natural to use a classical approximation suggested for FRIM model in Refs. [8,10]. It has been shown in [10] that the anharmonic interaction under consideration can lead to bistability in the energy transmission through the interface. Using the simplest 1D model, it was demonstrated that there is the close connection between bistability and classical FRIM.

Now we extend this model to $3D$ case with $2D$ interface and consider the energy propagation along the interface.

2. We assume that the bilayered structure consists of two molecular crystals separated by perfect plane interface. The molecules of c type occupy the sites of simple cubic lattice on the right from the interface $(n_x, n_y, n_z;\ n_x = 0, 1, 2, \ldots)$ and the molecules of b type occupy the sites of the lattice on the left from the interface $(n_x, n_y, n_z;\ n_x = -1, -2, \ldots)$. To demonstrate the appearence of the Fermi resonance interface solitons we consider here the simplest case of Fermi resonance between c and b harmonic vibrations, assuming that $\hbar\omega^c \cong 2\hbar\omega^b$. For this case

the main anharmonic interaction between c and b molecules corresponds to the cubic anharmonicity and has the form

$$\hat{H}_{int} = \Gamma[c_{0,n_y,n_z}(b^{\dagger}_{-1,n_y,n_z})^2 + h.c.],$$

where $b^{\dagger}(b)$ and $c^{\dagger}(c)$ are the creation (annihilation) operators for b and c excitations.

In the classical approximation we replace all operators by their mean values $\langle b_{n_x n_y n_z}\rangle = B_{n_x n_y n_z}$ and $\langle c_{n_x n_y n_z}\rangle = C_{n_x n_y n_z}$, where B and C are classical amplitudes of vibrations. These variables in our model satisfy the following equations:

$$i\partial B_{n_x n_y n_z}/\partial t - \omega^b B_{n_x n_y n_z} - V^b(B_{n_x-1,n_y,n_z} + B_{n_x+1,n_y,n_z} + B_{n_x,n_y-1,n_z} + \\ +B_{n_x,n_y+1,n_z} + B_{n_x,n_y,n_z-1} + B_{n_x,n_y,n_z+1}) = 0 \qquad (1)$$

for molecules in the bulk of the b-crystal,

$$i\partial B_{-1,n_y,n_z}/\partial t - \omega^b B_{-1,n_y,n_z} - V^b(B_{-2,n_y,n_z} + B_{-1,n_y-1,n_z} + \\ +B_{-1,n_y+1,n_z} + B_{-1,n_y,n_z-1} + B_{-1,n_y,n_z+1}) - 2\Gamma B^*_{-1,n_y,n_z} C_{0 n_y n_z} = 0 \qquad (2)$$

for b-molecules near to the interface $(n_z = -1)$,

$$i\partial C_{0,n_y,n_z}/\partial t - \omega^c C_{0,n_y,n_z} - V^c(C_{1,n_y,n_z} + C_{0,n_y-1,n_z} + C_{0,n_y+1,n_z} + \\ +C_{0,n_y,n_z-1} + C_{0,n_y,n_z+1}) - \Gamma B^2_{-1,n_y,n_z} = 0 \qquad (3)$$

for c-molecules near to the interface $(n_z = 0)$, and

$$i\partial C_{n_x n_y n_z}/\partial t - \omega^c C_{n_x n_y n_z} - V^c(C_{n_x-1,n_y,n_z} + C_{n_x+1,n_y,n_z} + C_{n_x,n_y-1,n_z} + \\ +C_{n_x,n_y+1,n_z} + C_{n_x,n_y,n_z-1} + C_{n_x,n_y,n_z+1}) = 0 \qquad (4)$$

for molecules in the bulk of the c-crystal.

We shall look for the localized near the interface solution in the form of plane wave

$$B_{n_x n_y n_z} = Be^{-\frac{i\omega}{2}t} e^{\kappa_b n_x} e^{\frac{i}{2}(k_y n_y + k_z n_z)}, \qquad (5)$$

$$C_{n_x n_y n_z} = Ce^{-i\omega t} e^{-\kappa_c n_x} e^{i(k_y n_y + k_z n_z)}.$$

with $\kappa_b > 0$ and $\kappa_c > 0$. From the equations (1) and (4) we get

$$\omega^b - \omega/2 + 2V^b(\cosh\kappa_b + \cos(k_y/2) + \cos(k_z/2)) = 0, \qquad (6)$$

$$\omega^c - \omega + 2V^c(\cosh\kappa_c + \cos k_y + \cos k_z) = 0.$$

These equations give us the values of κ_b and κ_c as functions of ω and (k_y, k_z). Eqs. (2) and (3) give the relations between variables B and C which can be written with the help of (6) in the form

$$2\Gamma B^* C = V^b B e^{\kappa_b}, \qquad \Gamma B^2 = V^c C e^{\kappa_c}. \qquad (7)$$

These relations yield immediatelly

$$I \equiv |B|^2 = \frac{V^b V^c}{2\Gamma^2} e^{\kappa_b + \kappa_c}, \qquad (8)$$

As follows from (8), the quantities κ_b and κ_c are real in case of real V^b, V^c and Γ, and they increase with increase of intensity I. The quantities $V^b e^{\kappa_b}$ and $V^c e^{\kappa_c}$ can be found easily form (6) what leads to the dispersion relation in an implicit form

$$8\Gamma^2 I = \{\frac{\omega}{2} - \omega^b - 2V^b(\cos(k_y/2) + \cos(k_z/2)) + [(\frac{\omega}{2} - \omega^b - 2V^b(\cos(k_y/2) + \\ + \cos(k_z/2)))^2 - 4(V^b)^2]^{1/2}\}\{\omega - \omega^c - 2V^c(\cos k_y + \cos k_z) + [(\omega - \omega^c - \\ -2V^c(\cos k_y + \cos k_z))^2 - 4(V^c)^2]^{1/2}\}. \tag{9}$$

If V^b and V^c vanish, we return to the two-molecule model discussed in Ref. [10]. Signs before roots correspond to positive values of $(\omega/2 - \omega^b - 2V^b(\cos(k_y/2) + \cos(k_z/2))$ and $(\omega - \omega^c - 2V^c(\cos k_y + \cos k_z))$, otherwise they should be reversed. Such a choice leads to correct limit as $V^b, V^c \to 0$.

3. In a long wave limit $|k_y|, |k_z| \ll 1$ eq. (9) leads to the quadratical dependence of ω on wave numbers k_y and k_z. If we consider nonlinear nonuniform wave propagating along the interface in z-direction, then the variables B and C will depend on t and z and their equations of motion will have in a long wave limit the form

$$i\frac{\partial B}{\partial t} - \tilde{\omega}^b B - \tilde{V}^b \frac{\partial^2 B}{\partial z^2} - 2\Gamma B^* C = 0, \tag{10}$$

$$i\frac{\partial C}{\partial t} - \tilde{\omega}^c C - \tilde{V}^c \frac{\partial^2 C}{\partial z^2} - \Gamma B^2 = 0.$$

where $\tilde{\omega}^b, \tilde{\omega}^c, \tilde{V}^b, \tilde{V}^c$ are some constants determined by the dispersion relation in the limit of small $|k_z|$ and at $k_y \equiv 0$. These equations may be considered as equations describing two-plane model with renormalized parameters.

We would like to demonstrate that the system under consideration has solitonic excitations. To this end, we consider the simplest case when the solution of eqs. (10) has the form

$$B = F\exp[(-i\Omega t + ikz)/2], \quad C = \beta F \exp(-i\Omega t + ikz), \quad F = F(z - vt), \tag{11}$$

where β is constant. Substitution of these expressions into (10) yields

$$(\Omega/2 - \tilde{\omega}^b + \tilde{V}^b k^2/4)F - i(v + \tilde{V}^b k)F' - \tilde{V}^b F'' - 2\Gamma F^2 \beta = 0, \tag{12}$$

$$(\Omega - \tilde{\omega}^c + \tilde{V}^c k^2)F - i(v + 2\tilde{V}^c k)F' - \tilde{V}^c F'' - \Gamma F^2/\beta = 0.$$

There are two possibilities for vanishing the imaginary parts of these equations:

$$(i) \quad k = 0, \quad v = 0, \tag{13}$$

$$(ii) \quad \tilde{V}^b = 2\tilde{V}^c, \quad v = -\tilde{V}^b k = -2\tilde{V}^c k.$$

At first we consider the case (i). Equations (12) for F are compatible if

$$\frac{\Omega/2 - \tilde{\omega}^b}{\Omega - \tilde{\omega}^c} = \frac{\tilde{V}^b}{\tilde{V}^c} = 2\beta^2 \tag{14}$$

what determines β and Ω:

$$\beta = \pm\sqrt{\frac{\tilde{V}^b}{2\tilde{V}^c}}, \quad \Omega = \frac{2(\tilde{\omega}^b \tilde{V}^c - \tilde{\omega}^c \tilde{V}^b)}{\tilde{V}^c - 2\tilde{V}^b}. \tag{15}$$

In what follows we shall choose positive sign for β. Then F satisfies the equation

$$F'' - \frac{2\tilde{\omega}^b - \tilde{\omega}^c}{\tilde{V}^c - 2\tilde{V}^b}F + \Gamma\sqrt{\frac{2}{\tilde{V}^b\tilde{V}^c}}F^2 = 0 \tag{16}$$

Its integration gives (with the integration constants such that $F' \rightarrow 0$, $F \rightarrow 0$ as $z \rightarrow \infty$)

$$F = \frac{\alpha}{cosh^2(\kappa z)}, \tag{17}$$

where

$$\alpha = \frac{3\sqrt{\tilde{V}^b\tilde{V}^c}}{2\sqrt{2}\Gamma} \cdot \frac{2\tilde{\omega}^b - \tilde{\omega}^c}{\tilde{V}^c - 2\tilde{V}^b}, \quad \kappa = \frac{1}{2}\left(\frac{2\tilde{\omega}^b - \tilde{\omega}^c}{\tilde{V}^c - 2\tilde{V}^b}\right)^{1/2}. \tag{18}$$

Thus, we have found for real κ the soliton solution for the interface wave

$$B = \frac{\alpha e^{-i\Omega t/2}}{cosh^2(\kappa z)}, \quad C = \frac{\alpha\beta e^{-i\Omega t}}{cosh^2(\kappa z)}, \tag{19}$$

where all the parameters are defined above. This solution corresponds to the soliton at rest. Apparently, it is a very special case of more general solitonic solution.

The case (ii) in (13) leads to a particular solution for moving soliton. Now we have

$$\beta = \pm 1, \qquad \Omega = \frac{2}{3}(2\tilde{\omega}^c - \tilde{\omega}^b) - \frac{\tilde{V}^b}{2}k^2. \tag{20}$$

Integration of equation for F leads to the moving soliton solution

$$B = \frac{\alpha\exp(-i\Omega t/2 - ivz/2\tilde{V}^b)}{cosh^2[\kappa(z - vt)]}, \quad C = \frac{\alpha\exp(-i\Omega t - ivz/\tilde{V}^b)}{cosh^2[\kappa(z - vt)]}, \tag{21}$$

where

$$\alpha = \frac{1}{2\Gamma}\left(\tilde{\omega}^c - 2\tilde{\omega}^b\right), \quad \kappa = [\frac{\tilde{\omega}^c - 2\tilde{\omega}^b}{6\tilde{V}^b}]^{1/2}. \tag{22}$$

As v goes to zero we return to the solution (19) for the soliton at rest with $\tilde{V}^b = 2\tilde{V}^c$. One may expect that there are solitonic excitations for more arbitrary choice of parameters describing the system. This possibility will be discussed elsewhere.

Thus, we have found that there are Fermi Resonance Interface Modes propagating along the interface between two crystals provided their vibronic excitations satisfy Fermi resonance condition. In the limit of strong excitations these modes can be described by classical theory and for some parameters or frequencies they can exist as the localized soliton states. Such propagating modes can play an important role in the energy transmission along the interfaces.

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1. F.F.So, S.R.Forrest, Y.Q.Shi and W.H.Steier, Appl. Phys. Lett. **56**, 674 (1990); F.F.So and S.R.Forrest, Phys. Rev. Lett. **66**, 2649 (1991).
2. G.E.Collins, V.S.Williams, L.-K.Chau et al., Synth. Metals **54**, 351 (1993).
3. A.J.Dann, H.Hoshi and Y.Maruyama, J.Appl.Phys. **67**, 1371 (1990).
4. N.Karl and N.Sato, Mol.Cryst.Liquid Cryst. **218**, 79 (1992); M.Möbus and N.Karl, Thin Solid Films **215**, 213 (1992).
5. H.Akimichi,T.Inoshita, S.Hotta et al., Appl. Phys. Lett. **62** (23), 3158 (1993).

6. V.M.Agranovich, Mol. Cryst. Liq. Cryst. **230**, 13 (1993).
7. V.M.Agranovich, R.D.Atanasov, and G.F.Bassani, Chem.Phys.Lett. **199**, 621 (1992).
8. V.M.Agranovich, Phys. Scripta, T**49**, 699 (1993).
9. V.M.Agranovich and O.A.Dubovsky, Chem.Phys.Lett. **210**, 458 (1993).
10. V.M.Agranovich and J.B.Page, Phys.Lett.A **183**, N5/6 (1993).
11. V.M.Agranovich, Nonlinear Optics (1994, in press).
12. Th.Neidlinger, P.Reineker, V.M.Agranovich and V.I.Yudson, Nonlinear Optics (1994, submitted).
13. V.M.Agranovich, P.Reineker, and V.I.Yudson, Syntetic Metals (1994, in press).
14. V.M.Agranovich and J.B.Page, BAPS (March,1994)