

Excitation-Induced Modification of Polariton Dispersion Curves in Molecular Media Within Different Approaches

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Abstract

Giant polariton dispersions by Frenkel excitons are derived and compared within three approaches: the boson representation, the effective Hamiltonian formalism and that based on the exact commutation relations for non-boson exciton operators. The latter approach resulted in a situation which could serve as a possible method of determining the effective dynamical exciton-exciton coupling constant by measuring dispersion curves of the polariton energy lower-branches under different levels of excitation.

1. Introduction

Due to the strong photon-exciton interaction photons propagating inside a medium must be considered as polaritons [1, 2]. If the medium is illuminated by an intense light source many excitons will be generated which couple not only to photons but also to each other. When the exciton concentration increases excitons are no longer ideal Bose particles. That requires theoretists to handle them very carefully in doing any necessary calculations. Concerning tightly bound pair states in molecular media – Frenkel excitons – there exist at least three different approaches to the problem of the non-boson character of excitons. The first, most usual one (called shortly AP1) is the boson representation [3, 4] which puts excitons into effective interactions among hypothetical ideal bosons. The effective Hamiltonian formalism [5, 6] serves as the second approach (AP2) which tries to construct an effective Hamiltonian in bosonic operators such that from it the physically correct equations of motion of the exciton-photon system could be obtained. The third approach (AP3) attacks excitons as such directly without going to boson description but with using the exact (bilinear and trilinear) commutation relations for non-bosonic exciton operators [7, 8]. If the final physical results are the same within different approaches, what of them will be applied is just the matter of taste. If the results differ somewhat quantitatively, appropriate corrections could be added. But one has to reestimate the validity of the approaches when their results are distinct qualitatively or radically in quantities. This paper would like to discuss the three above-mentioned approaches with the task of deriving the polariton dispersion relations in a highly photo-excited molecular medium. As will be shown, the excitation-induced modifications of the polariton dispersion curves will be distinct when applying different approaches. Our opinions with regard to these distinctions will be given in the Conclusion. We herein use the $\hbar = c = 1$ unit system.

2. Highly Photo-Excited Molecular Media

For simplicity, consider a molecular medium with one molecule in a unit cell and restrict the study to their lowest excited

states when they remain neutral. Further, only one excited state of a molecule is being taken into account so that we can label by P_v and P_v^+ the elementary excitation annihilation and creation operators at a site v ($v = 1, 2, \dots, N$; N the total number of sites or unit cells). We call the medium highly excited if many elementary excitations exist in it at a time (here “highly” without further specification is understood in a relative sense). If the exciting factor is light we shall have a coupled system of many interacting elementary excitations + photons whose Hamiltonian reads

$$H = \Delta \sum_v P_v^+ P_v + \sum_{v'} [M_{v-v'} P_v^+ P_{v'} + F_{v-v'} P_v^+ P_{v'}^+ P_v P_{v'}] + \sum_k \omega_k c_k^+ c_k + \sum_{kv} \frac{R_k}{\sqrt{V}} [P_v^+ c_k \exp(ikr_v) - \exp(-ikr_v) c_k^+ P_v] \quad (1)$$

where Δ is the energy needed for exciting a molecule, $M_{v-v'}$ the resonant integrals characterizing the excitation transition from a molecule in unit cell v to another in unit cell v' , $F_{v-v'}$ the effective dynamical interactions between two excited molecules in unit cells v and v' , $\sum_{v'}$ excludes terms with $v = v'$, c_k and c_k^+ the operators for photons with wave-vector k and renormalized energy $\omega_k = k + A_k$, $A_k = \omega_0^2/2k$ with ω_0 being the plasma frequency, $R_k = -i(2\pi/k)^{1/2} \Delta \Pi$ the photon-elementary excitation coupling, Π the dipole moment operator matrix element, V and r_v the volume of the sample and radius-vector of unit site v . While c and c^+ are bosonic operators P and P^+ belong to paulionic ones

$$\{P_v, P_v^+\} \equiv P_v P_v^+ + P_v^+ P_v = 1, \quad P_v^2 = (P_v^+)^2 = 0 \quad (2)$$

$$n_v = P_v^+ P_v = (n_v)^2 = 0 \text{ or } 1 \quad (3)$$

$$[P_v, P_\mu] \equiv P_v P_\mu - P_\mu P_v = [P_v, P_\mu^+] = [P_v^+, P_\mu^+] = 0, \quad v \neq \mu. \quad (4)$$

Passing to the k -space by new operators a and a^+ referred to as exciton ones and defined as

$$a_k = N^{-1/2} \sum_v \exp(-ikr_v) P_v, \quad a_k^+ = N^{-1/2} \sum_v \exp(ikr_v) P_v^+ \quad (5)$$

we then cast H into

$$H = \sum_k [E_k a_k^+ a_k + \omega_k c_k^+ c_k + \frac{NF_k}{4} \rho_k \rho_{-k} + g_k (a_k^+ c_k - c_k^+ a_k)] \quad (6)$$

$$g_k = (N/V)^{1/2} R_k, \quad \rho_k = 2N^{-1} \sum_q a_{q+k}^+ a_q \quad (7)$$

$$E_k = \Delta + \sum_{v \neq 0} M_v \exp(-ikr_v). \quad (8)$$

F_k is the Fourier components from F_v with $v \neq 0$. In what follows we shall for simplicity neglect the k -dependences of E_k and F_k and write them simply as E and F .

3. Boson representation or approach 1 (AP1)

Since the statistics for paulions is missing, serious difficulties are met in the application of the second-quantization method to the theoretical treatment of phenomena in molecular media. To get rid of paulions a boson representation was developed [3, 4] which expresses paulionic operators through bosonic ones, namely

$$\begin{aligned} P_v &= \left[\sum_{i=0}^{\infty} \alpha_i (B_v^+ B_v)^i \right] B_v, \\ P_v^+ &= B_v^+ \left[\sum_{i=0}^{\infty} \alpha_i (B_v^+ B_v)^i \right] \end{aligned} \quad (9)$$

where B_v and B_v^+ are bosonic operators and the real coefficients α_i are $\alpha_0 = 1$, $\alpha_1 = -1$, $\alpha_2 = (1 + \sqrt{3}/3)/2$, \dots . Denoting by b_k and b_k^+ the bosonic operators in k -space

$$\begin{aligned} B_v &= N^{-1/2} \sum_k \exp(ikr_v) b_k, \\ B_v^+ &= N^{-1/2} \sum_k \exp(-ikr_v) b_k^+ \end{aligned} \quad (10)$$

we can with the aid of eqs. (5), (9) and (10) express a and a^+ in terms of b and b^+ as

$$a_k = \alpha_0 b_k + \alpha_1 N^{-1} \sum_{pq} b_{p+q-k}^+ b_p b_q + \dots \quad (11)$$

$$a_k^+ = \alpha_0 b_k^+ + \alpha_1 N^{-1} \sum_{pq} b_q^+ b_p^+ b_{p+q-k} + \dots \quad (12)$$

" \dots " in eqs. (11) and (12) contain terms of the orders in b (b^+) higher than the third. Substituting eqs. (11) and (12) into eqs. (6) and (7) we have up to the fourth order in b (b^+) the bosonic Hamiltonian H^B

$$\begin{aligned} H^B &= \sum_k \{ \alpha_0^2 E b_k^+ b_k + \alpha_0 \alpha_1 E N^{-1} \sum_{pq} [b_k^+ b_{p+q-k}^+ b_p b_q \\ &+ b_q^+ b_p^+ b_{p+q-k} b_k] + (1/4) \alpha_0^4 F N \rho_k^B \rho_{-k}^B + \omega_k c_k^+ c_k \\ &+ g_k [(\alpha_0 b_k^+ + \alpha_1 N^{-1} \sum_{pq} b_q^+ b_p^+ b_{p+q-k}) c_k \\ &- c_k^+ (\alpha_0 b_k + \alpha_1 N^{-1} \sum_{pq} b_{p+q-k}^+ b_p b_q)] \} \end{aligned} \quad (13)$$

$$\rho_k^B = 2N^{-1} \sum_q b_{q+k}^+ b_q. \quad (14)$$

In the boson representation the non-boson character manifests itself through the appearance of the kinematical boson-boson interaction terms proportional to $\alpha_0 \alpha_1$ and the kinematical collective-excitonic one-photon transition terms proportional to $g_k \alpha_1$. The dynamico-kinematical interaction terms should appear when we go beyond the fourth order terms. Now using H^B we can set up the equations of motion for the averaged values $\langle b_k \rangle$ and $\langle c_k \rangle$. In handling averaged values such as $\langle b_p^+ b_q b_k \rangle$ we resort to the Hartree-Fock approximation splitting $\langle b_p b_q b_k \rangle = \langle b_p^+ b_q \rangle \langle b_k \rangle + \langle b_p^+ b_k \rangle \langle b_q \rangle$ and if there appear operators ρ_k^B we as done in [5] replace them by their averaged values $\langle \rho_k^B \rangle = \delta_{k0} \rho_0^B \equiv \delta_{k0} \rho^B = 2N_B/N$ with N_B being the total number of the boson whose operators are b and b^+ . After some transformation we

have

$$\begin{aligned} d\langle b_k \rangle / dt &= -i[E(1 - 4\rho^B) + F\rho^B] \langle b_k \rangle \\ &- ig_k(1 - \rho^B) \langle c_k \rangle \end{aligned} \quad (15)$$

$$d\langle c_k \rangle / dt = -i\omega_k \langle c_k \rangle + ig_k(1 - \rho^B) \langle b_k \rangle. \quad (16)$$

For a stationary regime the equation system (15) and (16) can easily be solved by seeking possible particular solutions in the forms [6]

$$\langle b_k \rangle = \tilde{b}_k \exp(-i\Omega_k t), \quad \langle c_k \rangle = \tilde{c}_k \exp(-i\Omega_k t) \quad (17)$$

where $\tilde{b}_k, \tilde{c}_k = \text{const}$. Inserting eq. (17) into eqs. (15) and (16) we obtain a system of two homogeneous equations for \tilde{b}_k and \tilde{c}_k whose condition to possess nontrivial roots yields the expression of the two-branch polariton dispersion laws

$$\begin{aligned} \Omega_{\mu k}^B(\rho^B) &= (1/2) \{ \tilde{E}^B(\rho^B) + \omega_k + (-1)^\mu [[\tilde{E}^B(\rho^B) - \omega_k]^2 \\ &+ 4|\tilde{g}_k^B(\rho^B)|^2]^{1/2} \} \end{aligned} \quad (18)$$

where $\mu = 1$ (2) is for the lower (upper) polariton branch, \tilde{E}^B and \tilde{g}_k^B stand for the renormalized energy of the boson and the excitation-induced boson-photon interaction

$$\tilde{E}^B(\rho^B) = E(1 - 4\rho^B) + F\rho^B \quad (19)$$

$$|\tilde{g}_k^B(\rho^B)|^2 = |g_k|^2(1 - \rho^B)^2. \quad (20)$$

4. Effective Hamiltonian formalism or approach 2 (AP2)

The polariton dispersions can also be obtained by diagonalizing the Hamiltonian (6) of the coupled exciton-photon system. However, (6) as such cannot be diagonalized because it is not of the quadratic form and more seriously it is expressed through exciton operators a, a^+ which are of neither Bose nor Fermi type. The (anti-) commutators hold as follows:

$$\{a_p, a_q^+\} = \delta_{pq} + 2N^{-1} \sum_{\nu\mu} \exp[i(\nu r_p - \mu r_\nu)] b_\nu^+ b_\mu \quad (21)$$

$$[a_p, a_q^+] = \delta_{pq} - 2N^{-1} \sum_{\nu} \exp[i(\nu - p)r_\nu] b_\nu^+ b_\nu. \quad (22)$$

The effective Hamiltonian formalism attempts to use the (anti-) commutators (21, 22) to set up the necessary equations of motion of the system which are subject to be simplified within proper approximation and then tries to build an effective bosonic and quadratic Hamiltonian H_{eff} such that from it the above system of equations of motion could be reproduced. If such an H_{eff} is found, its diagonalization will yield the desired polariton dispersions. The described procedure was carried out in Ref. [5] to solve the problem of the giant polariton. Unfortunately, H_{eff} of [5], being hermitian one, is not self-consistent (eq. (16b) in [5] cannot follow from the H_{eff} in it as required). As shown in [6], to be self-consistent H_{eff} must be a non-Hermitian one, which might still be diagonalized numerically by a step-by-step Bogolubov transformation method suggested in Ref. [9]. Since we herein are paying attention to analytic results let us refer the reader who is interested in the effective Hamiltonian formalism and the procedure of diagonalizing a non-Hermitian Hamiltonian to [5, 6, 9].

5. Exact commutation relations for exciton operators or approach 3 (AP3)

The delicate trouble that faces us is the missing of a closed set of commutation relations for exciton operators and hence we

would be unable to use the standard statistical formulae in calculating relevant characteristics of molecular media. Two ways out of such a situation were outlined in Sections 3 and 4. In this Section we remind the idea of Refs. [7, 8] in dealing with the non-boson feature of exciton operators and then apply it to the polariton problem. The worthy thing found in Refs. [7, 8] is that they have been successful in formulating a simple closed set of commutation relations for exciton operators a and a^+ which are of both bilinear and trilinear forms, i.e.

$$[a_p, a_q] = [a_p^+, a_q^+] = 0 \quad (23)$$

$$[[a_k^+, a_p], a_q] = -2N^{-1}a_f, \quad k + f = p + q \quad (24)$$

$$[[a_k^+, a_p], a_q^+] = +2N^{-1}a_f^+, \quad k + q = p + f. \quad (25)$$

Furthermore, it is provable [7] that the ways a_k and a_k^+ act on a state-vector $|L_k\rangle$ containing $L < N$ excitons with momentum k are determined as

$$a_k|L_k\rangle = [L(N - L + 1)/N]^{1/2}|L_k - 1\rangle \quad (26)$$

$$a_k^+|L_k\rangle = [(L + 1)(N - L)/N]^{1/2}|L_k + 1\rangle \quad (27)$$

Equations (26) and (27) give

$$a_k^+ a_k|L_k\rangle = L(N - L + 1)/N|L_k\rangle. \quad (28)$$

Equation (28) means that except for $L = 0$ and 1 the operator-product $a_k^+ a_k$ should not serve as the exciton number operator as in the case of bosons and fermions. However, the total exciton number operator denoted by \hat{N}_x could still be built in terms of a and a^+ in the form [7, 8]

$$\hat{N}_x = \frac{1}{2}N \{[a_k^+, a_k] + 1\}. \quad (29)$$

Such an expression of \hat{N}_x , eq. (29), was used in Ref. [10] to study the density and optical bistability in laser-excited molecular crystals. In what follows we shall use the exact relations (23)–(25) to set up from H of eq. (6) the equations of motion for $\langle a_k \rangle$ and $\langle c_k \rangle$. Again resort to the similar approximations as mentioned in Section 3 we arrive at (for brevity we write ρ instead of $\rho_0 \equiv 2N_x/N$ in all the forthcoming formulae)

$$d\langle a_k \rangle/dt = -i[E(1 - \rho) + F\rho]\langle a_k \rangle - ig_k(1 - \rho)\langle c_k \rangle \quad (30)$$

$$d\langle c_k \rangle/dt = -i\omega_k\langle c_k \rangle + ig_k\langle a_k \rangle. \quad (31)$$

Solving eqs. (30) and (31) for a stationary regime as in Section 3 we obtain the following polariton dispersions:

$$\Omega_{\mu k}(\rho) = (1/2) \{ \tilde{E}(\rho) + \omega_k + (-1)^\mu [[\tilde{E}(\rho) - \omega_k]^2 + 4|\tilde{g}_k(\rho)|^2]^{1/2} \} \quad (32)$$

with

$$\tilde{E}(\rho) = E(1 - \rho) + F\rho \quad (33)$$

$$|\tilde{g}_k(\rho)|^2 = |g_k|^2(1 - \rho). \quad (34)$$

The formal differences between eqs. (15), (16), (18)–(20) and eqs. (30), (31), (32)–(34) are obvious. In the next section we shall show that the differences will not be formal because they may lead to consequences qualitatively distinguished.

6. Results comparison between approaches

For comparison, the analytic results derived in the previous

Sections will be estimated numerically in this Section. As seen from eqs. (18) and (32), two physical factors leading to changes in polariton dispersion curves are the renormalization of the boson-particle (exciton) energy $\tilde{E}^B(\tilde{E})$ caused by both kinematical and dynamical interactions and the quenching of the dipole moment transition $\tilde{g}_k^B(\tilde{g}_k)$. In Ref. [11] the dependence of the polariton dispersion on these two factors was investigated separately within the AP1. Since both $\tilde{E}^B(\tilde{E})$ and $\tilde{g}_k^B(\tilde{g}_k)$ depend on the relative density $\rho^B(\rho)$ which in turn characterizes the level of excitation of the medium it is reasonable and even necessary to follow how the polariton dispersion is modified when one changes the level of excitation. Such a study was carried out [12] in the vicinity of the $k = 0$ point within the AP3. We are now aimed at the excitation-induced modification of polariton dispersion curves in the whole range of the wave-vector k within both AP1 and AP3. For that purpose we take from eqs. (18) and (32) the derivatives of $\Omega_{\mu k}^B(\Omega_{\mu k})$ with respect to $\rho^B(\rho)$. Within the AP1 we have

$$\frac{d\Omega_{\mu k}^B}{d\rho^B} = \frac{\partial\Omega_{\mu k}^B}{\partial\tilde{E}^B} \frac{\partial\tilde{E}^B}{\partial\rho^B} + \frac{\partial\Omega_{\mu k}^B}{\partial|\tilde{g}_k^B|^2} \frac{\partial|\tilde{g}_k^B|^2}{\partial\rho^B} \quad (35)$$

$$\frac{\partial\Omega_{\mu k}^B}{\partial\tilde{E}^B} = \frac{1}{2} \left\{ 1 + (-1)^\mu \frac{\tilde{E}^B - \omega_k}{[(\tilde{E}^B - \omega_k)^2 + 4|\tilde{g}_k^B|^2]^{1/2}} \right\} \quad (36)$$

$$\frac{\partial\Omega_{\mu k}^B}{\partial|\tilde{g}_k^B|^2} = \frac{2(-1)^\mu}{[(\tilde{E}^B - \omega_k)^2 + 4|\tilde{g}_k^B|^2]^{1/2}} \quad (37)$$

$$\frac{\partial\tilde{E}^B}{\partial\rho^B} = F - 4E; \quad \frac{\partial|\tilde{g}_k^B|^2}{\partial\rho^B} = -2|g_k|^2(1 - \rho^B). \quad (38)$$

In the case of the AP3 we formally get the same formulae as from eqs. (35)–(37) if the replacements $\Omega^B, \rho^B, \dots \Rightarrow \Omega, \rho, \dots$ are made. However, instead of eq. (38) we have:

$$\frac{\partial\tilde{E}}{\partial\rho} = F - E; \quad \frac{\partial|\tilde{g}_k|^2}{\partial\rho} = -|g_k|^2. \quad (39)$$

Accounting for the realistic fact that $\rho^B(\rho) < 1$ and $F < E$ one can easily check from the above-written derivatives that the upper-branch ($\mu = 2$) of the polariton energy is always shifted down for any values of k when the level of excitation increases. But for a given increase in $\rho^B(\rho)$ the shift down of the upper-branch is larger within the AP1 than that within the AP3 (see Figs. 1 and 2). As the maximal density of excitons

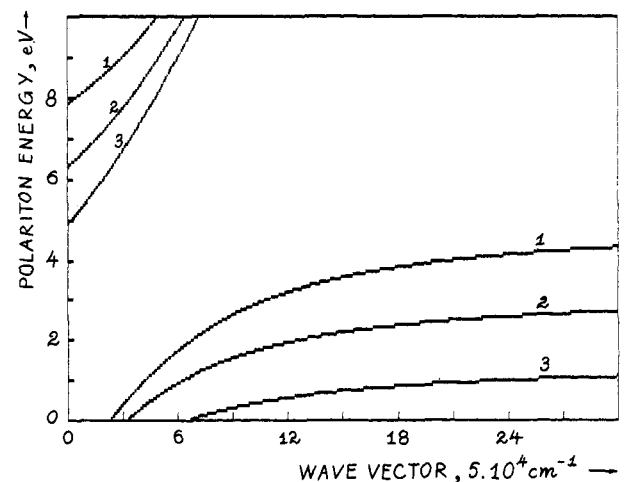


Fig. 1. The two-branch polariton dispersion curves under different excitation-levels $\rho^B = 0, 0.1$ and 0.2 (curves 1, 2 and 3, resp.) within the AP1.

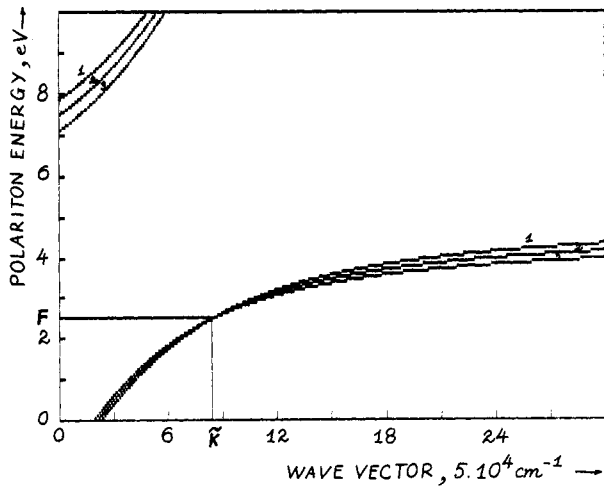


Fig. 2. The same as in Fig. 1 but within the AP3 (note that ρ^B in Fig. 1 is replaced by ρ here).

is determined by the strength of the sample and is of the order of about 10^{18} – 10^{19} cm^{-3} [13], the relative density – spanning the range from 10^{-8} to 10^{-3} – and the excitation-induced energy shift then are very small. In plotting the Figs we have chosen $\rho^B(\rho) = 0.1$ and 0.2 just for the eye. The other used parameters are $N/V = 10^{22}$ cm^{-3} , $E = 5$ eV, $F = 2.5$ eV and $\Pi = 2e \times 10^{-8}$ cm. A worthy thing is that the polariton energy lower-branch ($\mu = 1$) behaves in qualitatively different ways within AP1 and AP3. When $\rho^B(\rho)$ increases both $|\tilde{g}_k^B|^2$ ($|\tilde{g}_k|^2$) and $\tilde{E}^B(\tilde{E})$ decrease [see eqs (38) and (39)]. As Ω_{1k}^B (Ω_{1k}) decreases for decreasing $\tilde{E}^B(\tilde{E})$ and for increasing $|\tilde{g}_k^B|^2$ ($|\tilde{g}_k|^2$) [see eqs. (36) and (37)], there occurs a competition between two tendencies one of which lifts the lower-branch and the other shifts it down when the excitation-level increases. Using eqs. (35)–(39) it is not difficult to verify that within the AP1 the lower-branch Ω_{1k}^B is strongly shifted down in the whole range of k for increasing ρ^B (Fig. 1) whereas within the AP3 there exists, independently of ρ , a certain value of $k = \tilde{k} \equiv E - A + [(F - E)^2 - |g|^2]/(F - E)$ such as $d\Omega_{1k}/d\rho > 0, = 0$ and < 0 for $k < \tilde{k}$, $k = \tilde{k}$ and $k > \tilde{k}$, resp. That means that for increasing ρ the lower-branch Ω_{1k} is lifted in the interval of $k < \tilde{k}$ (in agreement with [12]) and shifted down for $k > \tilde{k}$. At $k = \tilde{k}$ all the lower-branches with different ρ must be intersected (see Fig. 2), i.e. $\tilde{\Omega}_1 \equiv \Omega_{1\tilde{k}}(\rho_1) = \Omega_{1\tilde{k}}(\rho_2) = \dots$; $\rho_1 \neq \rho_2 \neq \dots$. On the other hand, by formally putting $\rho = 1$ and $k = \tilde{k}$ into eq. (32) we immediately obtain the value of $\tilde{\Omega}_1 \equiv \Omega_{1\tilde{k}}(\rho = 1) \equiv F!$ Making the use of this finding we could suggest a possible experimental determination of the empirical effective dynamical interaction constant F in the following way. By measuring (for measurement methods see Ref. [14]) dispersion curves of the polariton energy lower branch Ω_{1k} under different levels of excitation one might experimentally find the intersection point at $k = \tilde{k}$ whose ordinate would give the magnitude of F .

Before going to the conclusion we note that the numerical calculations within the AP2 based on the step-by-step Bogolubov transformation method [9] were also done by the author and the modified-by-excitation dispersion curves of the polariton (not shown) resemble qualitatively those obtained within the AP1.

7. Conclusion

Three different approaches to the giant polariton problem in molecular media were treated in this paper. In our opinion, the AP3 solves the problem most naturally and rigorously because it deals with excitons as they are and by not going to the infinite series in bosonic operators (as done within the AP1) there arise no question such as how to cut the infinite series, up to what order in bosonic operators one has to restrict the expansion, what about the error resulted from the ignorance of higher order terms, etc. Thus, the AP3 seems to be adequate in principle for any exciton density. As to interactions of kinematical type they are automatically involved through the specific exact commutation relations for exciton operators. Since only the AP3 (but neither AP1 nor AP2) predicts a situation under which all lower branches of the polariton energy with different excitation-levels are met at one and the same point (this point can be defined analytically from the theory), its validity could be confirmed if such an intersection point is tested experimentally. We, of course, recognize that the picture outlined here in the framework of the simplest model of a molecular medium with disregarding many physical effects such as the relaxation and broadening ones (they can be considered, e.g. in a phenomenological way as done in [10]), the spatial dispersion, the anti-resonant term contributions, etc., would become more complicated when one accounts for the omitted effects and goes beyond the simplest model and it thus might only serve as an illustration of possible differences between approaches. Nevertheless, it is hopeful, that the method suggested by us for determining the effective interaction constant F would give a sense for both experimentalists and theoretists.

Concerning weakly bound pair states – Wannier–Mott excitons – the polariton theory by them [15, 16] seem to be quite suitable in explaining a lot of resonant optical phenomena (see, e.g. Refs [17–19]). For highly excited semi-conductors a good-working approach like the AP1 was well developed [20, 21] and applied. Such approaches like AP2 and especially AP3, to the author's knowledge, are still missing. We are planning to formulate such expressions like eq. (23) – (29) for Wannier–Mott exciton operators which due to the presence of the hydrogen-like envelope functions seem to be more generalized and might reproduce eqs. (23)–(29) as a particular case.

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