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DETERMINATION OF THE EXCITON-FREE CARRIER COUPLING CONSTANT IN SEMICONDUCTORS

Nguyen Ba An
Institute of Theoretical Physics,
P. O. Box 429 Boho, Hanoi 10000 Vietnam

Le Thi Cat Tuong
Institute of Physics, Nghia Do, Tu Liem, Hanoi, Vietnam

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Abstract

Based on theory of intrinsic optical bistability a possible method has been proposed for experimentally derternining the exciton-electron (exciton-hole) coupling constant in a highly excited semiconductor. Both sign and magnitude of the constant can be determined by measuring the exciton density or the output light intensity versus the input light intensity.

IN HIGHLY excited semiconductors the interaction between different kinds of quasiparticles plays a very important role in bringing about various novel phenomena that do not occur at low excitation levels.The exciton-exciton interaction, for example, may under certain conditions results in effects such as optical bistability [1-3], polariton dispersion anomalies [4], self-induced transparency [5], biexciton formation [6,7], ect. The knowledge of both sign and magnitude of the interaction among quasiparticles is needed to guide the technologists in fabricating devices with necessary properties. From a theoretical point of view, quasiparticle-quasiparticle interactions might be analytically calculated. However, their expressions are much dependent on calculation framework to be used and on physical situations to be considered. The interaction among excitons, say, can be derived exactly in the small momentum limit neglecting spins [8,9] or approximately in the whole range of momentum and of quasiparticle masses without [10] and with [11] taking spins into account. In polar semiconductors excitons can couple to each other by exchanging virtual phonons that requires further corrections [6,7]. Besides, there exists also a lot of factors which can by no means be accounted for because of lack of well-developed theoretical tools. So, generally speaking, theory can hardly provide precise values of the interaction between concrete quasiparticles of a given material under certain circumstances. In this respect, methods for experimentally determining interaction constants seem to be quite helpful. In [12] one of us has presented a way to determine the exciton-exciton interaction constant by measuring lower-branches of dispersion curves of the giant polariton at different levels of excitation. This communication attempts to outline another experimental possibility of determining the coupling constant between excitons and free carriers that may coexist in a highly excited semiconductor.

Consider a model of a dense system of interacting electrons, holes and excitons [8]. The latters are assumed to be resonantly driven by an external classical and monochromatic field [1-4] with wave vector $\mathbf{k}$, complex amplitudes $A_{\mathbf{k}}^{(\pm)}$ and frequency ω_k. The Hamiltonian we shall deal with has the form (Throughout we put Planck constant and velocity of light equal to unity and denote the sample volume by V) [2-5,8] :

$$H = \sum_{\mathbf{l}} \left[E_x(l)a_{\mathbf{l}}^+ a_{\mathbf{l}} + E_e(l)e_{\mathbf{l}}^+ e_{\mathbf{l}} + E_h(l)h_{\mathbf{l}}^+ h_{\mathbf{l}}\right] +$$

$$+\sum_{\mathbf{pql}} \left\{\frac{F_l}{2V}\left[e_{\mathbf{p+l}}^+ e_{\mathbf{q-l}}^+ e_{\mathbf{q}} e_{\mathbf{p}} + h_{\mathbf{p+l}}^+ h_{\mathbf{q-l}}^+ h_{\mathbf{q}} h_{\mathbf{p}} - \right.\right.$$

$$-2\, e^{+}_{p+1} h^{+}_{q-1} h_q e_p\Big] + \frac{W}{2V} a^{+}_{p+1} a^{+}_{q-1} a_q a_p +$$

$$+\frac{U}{V}\left[a^{+}_{p+1} e^{+}_{q-1} e_q a_p + a^{+}_{p+1} h^{+}_{q-1} h_q a_p\right]\Big\} +$$

$$+g_k \left[A^{(-)}_{\mathbf{k}} e^{-i\omega_k t} + A^{(+)}_{\mathbf{k}} e^{+i\omega_k t}\right] \quad (1)$$

where $E_{z(e,h)}(l)$ and a_l (e_l, h_l) stand for energy and annihilation operator of an exciton (an electron, a hole) with momentum l; F_l, W and U are the Coulomb potential, the exciton-exciton and exciton-electron (exciton-hole) coupling constants, respectively; g_k labels the light field-exciton transition matrix element. Applying standard techniques of Heisenberg equations of motion within the Hartree-Fock approximation we have arrived at the following characteristic equation :

$$I = n\left[(n - \Delta + C)^2 + 1\right] \quad (2)$$

In Eq.(2) I, n and Δ are respectively the dimensionless normalized input light intensity, exciton density and light frequency detuning :

$$I = \frac{Wg^2}{\gamma^3}\left|A^{(\pm)}\right|^2 , \; n = \frac{W\sigma}{\gamma} , \; \Delta = \frac{\omega - E_z}{\gamma} \quad (3)$$

with σ and γ being the exciton density and damping.

$$C = \frac{U}{\gamma}\rho \quad (4)$$

with ρ denoting the electron-hole pair density is a parameter to be determined as will be described below. Note that in the absence of free carriers $\rho = 0$, or the same $C = 0$, Eq.(2) is exactly reduced to that obtained in [2]. It is easy to prove that the condition for function $n = n(I)$ to be three-valued, i.e. for the appearance of optical bistability, reads

$$\Delta > C + \sqrt{3} \quad (5)$$

If $\Delta < C + \sqrt{3}$, no optical bistability arises since then n grows monotonically with increasing I. Finally, when the equality $\Delta = C + \sqrt{3}$ holds, function $n = n(I)$ will possess one inflection point during the development of n versus I.

Making use of the above-said behavior of function $n = n(I)$ we can suggest the following scheme of experiments to determine sign or/and magnitude of C, i.e., of the exciton-free carrier coupling constant U (γ and ρ are assumed already known from other kinds of experiments).

To get information on the sign of C, i.e., on the qualitative character of the coupling constant U, one first tunes on purpose the light frequency so that $\Delta = \sqrt{3}$ (in typical semiconductors γ^{-1} is of order of some picoseconds yielding ω to be about from some tenths to one meV above the exciton energy level. That does not violate the resonance regime of the problem under consideration). Then one should perform the measurement of the ($n - I$)-dependence. If one observes optical bistability (The ($n - I$)-characteristics should be S-shaped. To witness optical bistability one can alternatively measure the output-input light intensity characteristics whose shapes when the effect occurs may be much topologically distinct [13]), then, in accordance with (5), C is negative, and so is U. In this case one expects the formation of trions [6]. Otherwise, absence of optical bistability means the repulsive character of the exciton-electron (exciton-hole) interaction. It is worth noticing at this moment that the question of the sign of the interaction between quasiparticles is often a contraversial one. The exiton-exciton interaction can serve as an example. Its sign is not yet solidly confirmed up to date. The authors of [8,9,14] claimed that the exciton-exciton interaction is of repulsive nature, while in [15-17] the opposite was reported. In ones and the same materials the interaction might be either attractive or repulsive depending on two-exciton state spin symmetry [5,6] and also on exciton momenta , momentum transfer as well as electron-hole effective mass ratio [10,11]. In this connection, as discussed in [1] (or in [18]), studying optical bistability in the excitonic spectral region (or in the spectral region of the M-zone corresponding to the one-photon transition between the exciton and the biexciton states) can resolve the principal question of the sign of the exciton-excioton interaction (or of the difference between the exciton-exciton and exciton-biexciton interactions). Obviously, same conclusion can also be made here regarding the sign of the exciton-electron (or exciton-hole) interaction.

We now try to propose an experiment to determine the magnitude of the exciton-free carrier coupling constant. For this aim, one has to sweep the input light intensity back and forth simultaneously with tuning its frequency to find the critical value of detuning $\Delta = \Delta_c$ at which the exciton density-light input intensity (or the output-input light intensity) correspondence is one-to-one and, more importantly , undergoes one inflection point. Then, from Eqs.(4) and (5), it immediately follows :

$$U = \frac{\gamma}{\rho}\left(\Delta_c - \sqrt{3}\right) \quad (6)$$

This formula helps us to estimate the coupling constant U.

Before going to conclusion it is worth making a remark concerning the feasibility of the proposed measurement of exciton population n as function of input light intensity I. Since n is an internal parameter, it is difficult to directly measure it. In practice, one measures the output light intensity I_{out} but not the exciton population n in dependence on the input light intensity I. Furtunately enough, as was proved in the latter citation of [3] and in [19], the condition for (see (5)) and the critical value of detuning ($\Delta = \Delta_c$) of both kinds of optical bistability characteristics $n = n(I)$ and $I_{out} = I_{out}(I)$ are exactly coincident though their concrete shapes may differ drastically one from another. As our method of determining U is based only on the condition (5) and the critical situation of $\Delta = \Delta_c$, it can be performed by experimentally studying the usually-measured-in-practice dependence $I_{out} = I_{out}(I)$. As for the precision of the suggested method, it is premature to say anything because there exist many other influencing factors that are still not yet included in our consideration.

In conclusion, we have exploited optical bistability as a phenomenum which can be used to determine the quasiparticle-quasiparticle coupling constant in materials (see also [1,12,16]). Together with its main applicability in future all-optical communication processing,optical bistability proves to be a very interesting phenomenum which will find more and more applications in different areas of science and life. The simplest idea spoken out here can of course serve only as a modest suggestion towards better models.

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