IC/92/320
REFERENCE INTERNAL REPORT
(Limited Distribution)

International Atomic Energy Agency
and
United Nations Educational Scientific and Cultural Organization
INTERNATIONAL CENTRE FOR THEORETICAL PHYSICS

ON THE THEORY OF PHONORITON IN CUBIC SEMICONDUCTORS WITH A DEGENERATE VALENCE BAND

Nguyen Ai Viet *, Nguyen Thi Que Huong *
International Centre for Theoretical Physics, Trieste, Italy

and

Le Qui Thong
Department of Physics, Hue University, 3 Le Loi, Hue, Vietnam.

ABSTRACT

The "phonoriton" is an elementary excitation constructed from an exciton polariton and phonon in semiconductors under intense excitation by an electromagnetic wave near the exciton resonance (L.V. Keldysh and A.L. Ivanov, 1982). In this paper we develop a theory of phonoriton in direct band gap cubic semiconductor with a degenerate valence band using the simple model of J.L. Birman and B.S. Wang (1990). In addition to experimental proofs of the existence of phonoriton we propose an experiment to measure its flight time.

MIRAMARE - TRIESTE

October 1992

* Permanent address: Institute of Theoretical Physics, P.O. Box 429 Bo Ho, Hanoi 10000, Vietnam.

1. Introduction

When a high intensity coherent electromagnetic field with frequency near the band gap irradiates a direct gap semiconductor, it has been predicted that some new elementary excitations may be formed. The existence and properties of these elementary excitations depend not only on the intrinsic properties of the material, but also on the external pump. One of these new elementary excitations is the phonoriton, which was introduced first by L.V. Keldysh and A.L. Ivanov in 1982 [1].

In recent years, the changes of exciton spectra in semiconductors under intense coherent laser pump have been studied intensively, both experimentally and theoretically [2-9]. It is well known that an exciton is a movable elementary excitation. In semiconductors, excitons interact with photons all the time and form exciton polariton (photon plus exciton). Resonant Brillouin Scattering experiment, which was proposed by J.L. Birman and collaborators in 1972 [10], allowed the measurement of the polariton dispersion curves in a large number of semiconductors [11-14] and was the most direct evidence of the existence of these coupled-mode excitations. When a semiconductor with direct band gap is illuminated by high intensity electromagnetic radiation near the exciton resonance, the occupation number for the polariton mode, with frequency equal to that of the incident field, will be very high, and the exciton-phonon interaction becomes effectively a coupling between the scattering polariton modes and corresponding phonons. This coupling leads to the formation of a new elementary excitation, the phonoriton, which is a mixture of polariton and phonon.

Using the closed-time-path Green's function formalism and two-band model, the Keldysh group published series of papers [15-19] and analyzed some aspects of phonoritons. This formalism is useful but rather complicated. In fact, in order to understand the physical picture and many of the basic properties of the phenomenon, much of formalism is unnecessary. In the papers [20-21] J.L. Birman and B.S. Wang proposed a new simple two-level model to explain the origin of phonoriton and discussed its properties. They gave a simple and clear physical picture of the phonoriton and also proposed two experiments: Non-Linear Resonant Brillouin Scattering and Non-Linear Modulation Reflection to detect phonoritons. These non-linear experiments may be direct evidences of the existence of phonoriton, like the case of polariton in the past decade.

The real structure of highest valence band of many cubic semiconductors A_3B_5 and A_2B_6 is rather complicated: the two uppermost valence bands are degenerated at Γ point (k=0) and split into heavy and light hole bands in the neighbourhood of this top. These two types of holes have to be included in the dynamical exciton problem. When holes from these two valence bands are coupled with conduction band electrons, "heavy" and "light" excitons are formed, each of which is able to couple with photon. The result is a three-band exciton-polariton picture [22-23].

In this work, we generalize the results of J.L. Birman and B.S. Wang in the framework of three-band model for studying the problems of phonoriton and by analogy with the exciton-polariton case, we propose the time-of-flight measurements for phonoriton, which

could give another direct experimental test of the existence of phonoritons.

We use the unit system with $\hbar = c = 1$.

2. Three-band approximation

In cubic semiconductors with the highest valence band degenerate at the centre of the Brillouin zone there exist different types of holes and therefore many kinds of excitons. Even though the role of the spin-orbit split subband is neglected, in zinc-blende type cubic semiconductors with direct band gap there are two kinds of excitons involving heavy (H) and light (L) holes. Due to the mutual transition between these excitons and photon there arise three-band exciton polaritons whose dispersion curves have been investigated in detail by means of Resonant Brillouin Scattering experiments.

The theory of three-band exciton polaritons with "heavy" and "light" excitons has been developed long time ago in the interesting works by E.O. Kane [23] and G. Fishman [24], and experimental confirmation of this model has been achieved later by B. Sermage and G. Fishman [14]. Fishman has proposed a simple ansatz : the wave function of each stationary state of the exciton contains the Bloch wave function of the hole in the corresponding state with a definite total angular momentum projection $\boldsymbol{p}$: $M = \pm 3/2$ for the heavy (H) exciton and $M = \pm 1/2$ for the light (L) one. This ansatz has been used widely in the interpretation of the polariton scattering experimental data [11-14]. Following J.L. Birman and B.S. Wang, the truncated Hamiltonian describing the system of interacting photon, excitons, and longitudinal phonon in the three-band model is :

$$\begin{aligned} H &= \sum_{\boldsymbol{p}} \Big\{ \omega^{ex}_{H\boldsymbol{p}} a^{+}_{H\boldsymbol{p}} a_{H\boldsymbol{p}} + \omega^{ex}_{L\boldsymbol{p}} a^{+}_{L\boldsymbol{p}} a_{L\boldsymbol{p}} + \omega_{\boldsymbol{p}} b^{+}_{\boldsymbol{p}} b_{\boldsymbol{p}} + \Omega_{\boldsymbol{p}} c^{+}_{\boldsymbol{p}} c_{\boldsymbol{p}} \; + \\ &+ \frac{\Omega_{Hc}}{2} \left(a^{+}_{H\boldsymbol{p}} b_{\boldsymbol{p}} + b^{+}_{\boldsymbol{p}} a_{H\boldsymbol{p}} \right) + \frac{\Omega_{Lc}}{2} \left(a^{+}_{L\boldsymbol{p}} b_{\boldsymbol{p}} + b^{+}_{\boldsymbol{p}} a_{L\boldsymbol{p}} \right) \; + \\ &+ \sum_{\sigma\sigma' \boldsymbol{q}} \left[M_{\sigma\sigma'}(\boldsymbol{p}-\boldsymbol{q}) a^{+}_{\sigma\boldsymbol{p}} a_{\sigma'\boldsymbol{p}} (c_{\boldsymbol{p}-\boldsymbol{q}} + c^{+}_{-(\boldsymbol{p}-\boldsymbol{q})}) \; + \text{c.c} \right] \Big\} \quad , \end{aligned} \tag{1}$$

where $a^{+}_{\sigma\boldsymbol{p}}, b^{+}_{\boldsymbol{p}}, c^{+}_{\boldsymbol{p}}$ and $a_{\sigma\boldsymbol{p}}, b_{\boldsymbol{p}}, c_{\boldsymbol{p}}$ are creation and annihilation operators for σ-kind excitons ($\sigma = H \leftrightarrow$ heavy, $\sigma = L \leftrightarrow$ light), photons and phonons with momentum $\boldsymbol{p}$, respectively, $\omega_{\boldsymbol{p}}$ is the energy of the photon and $\omega_{\sigma\boldsymbol{p}}$ are the energies of the σ-kind excitons :

$$\begin{aligned} \omega_{\boldsymbol{p}} &= \frac{p}{\sqrt{\epsilon_b}} \quad , \\ \omega_{\sigma\boldsymbol{p}} &= \omega_{o\sigma} + \frac{p^2}{2M_\sigma} \qquad ; \quad \sigma = H, L \quad , \end{aligned} \tag{2}$$

here ϵ_b is the background dielectric constant of the semiconductors, ω_{oz} is the 1s-exciton energy at $\boldsymbol{p} = 0$; M_H and M_L are the effective masses of heavy and light excitons; $\Omega_{\boldsymbol{p}}$ is the energy of the phonon; Ω_{Hc} and Ω_{Lc} are the phonon-heavy-exciton and photon-light-exciton interactions :

$$\begin{aligned} \Omega_{Hc} &= \omega_{o\boldsymbol{p}} \sqrt{4\pi\beta} \quad , \\ \Omega_{Lc} &= \omega_{o\boldsymbol{p}} \sqrt{\frac{4\pi\beta}{3}} \quad , \end{aligned} \tag{3}$$

where β is the oscillation strength of the exciton :

$$4\pi\beta = \frac{1}{\epsilon_b E_g^3} \left(\frac{e\Pi_{cv}}{m_o} \right)^2 \frac{1}{\pi a_x^3} \quad , \tag{4}$$

here Π_{cv} is the usual interband matrix element between S conduction band states and X valence band states :

$$\Pi_{cv} = \langle S | \nabla_x | X \rangle \quad ,$$

a_x is the Bohr radius of exciton, ϵ and m_o are the charge and mass of a free electron, E_g is the band gap of the semiconductor. $M_{\sigma\sigma'}(\boldsymbol{p}-\boldsymbol{q})$ is the matrix element of the exciton-phonon interaction. We can estimate that the photon-exciton interactions is much stronger than the phonon-exciton interaction : $\Omega_{\sigma c} \gg M_{\sigma\sigma'}$.

The first two lines of equation (1) can be diagonalized by means of the well-known Bogoliubov transformation to give three-branch polaritons. The whole Hamiltonian can be written as :

$$\begin{aligned} H &= \sum_{i\boldsymbol{p}} \omega^{pol}_{i\boldsymbol{p}} B^{+}_{i\boldsymbol{p}} B_{i\boldsymbol{p}} \; + \sum_{\boldsymbol{p}} \Omega_{\boldsymbol{p}} c^{+}_{\boldsymbol{p}} c_{\boldsymbol{p}} \\ &+ \sum_{ij,\boldsymbol{pq}} \left[M'_{ij}(\boldsymbol{p}-\boldsymbol{q}) B^{+}_{i\boldsymbol{p}} B_{j\boldsymbol{q}} (c_{\boldsymbol{p}-\boldsymbol{q}} \; + \; c^{+}_{-(\boldsymbol{p}-\boldsymbol{q})}) \; + \text{c.c} \right] \quad , \end{aligned} \tag{5}$$

where $B^{+}_{i\boldsymbol{p}}$ is the creation operator of the ith branch polariton ($i = 1, 2, 3$) with wave vector $\boldsymbol{p}$. The M'_{ij} are renormalized polariton-phonon interaction matrix elements :

$$M'_{ij}(\boldsymbol{p}-\boldsymbol{q}) = \sum_{\sigma\sigma'} \psi^{\sigma}_{i}(\boldsymbol{p})^{*} M_{\sigma\sigma'}(\boldsymbol{p}-\boldsymbol{q}) \psi^{\sigma'}_{j}(\boldsymbol{q}) \quad , \tag{6}$$

$\psi^{H}_{i}(\boldsymbol{p})$, $\psi^{L}_{i}(\boldsymbol{p})$ and $\psi^{Pt}_{i}(\boldsymbol{p})$ are the weights of the heavy exciton, light exciton and photon in the ith branch polariton, respectively :

$$\begin{aligned} \psi^{Pt}_{i}(\boldsymbol{p}) &= \left\{ 1 \; + \; \frac{\Omega^2_{Hc}}{4(\omega^{pol}_{i\boldsymbol{p}} - \omega^{ex}_{H\boldsymbol{p}})^2} \; + \; \frac{\Omega^2_{Lc}}{4(\omega^{pol}_{i\boldsymbol{p}} - \omega^{ex}_{L\boldsymbol{p}})^2} \right\}^{-1/2} , \\ \psi^{H}_{i}(\boldsymbol{p}) &= \frac{\Omega_{Hc}}{2(\omega^{pol}_{i\boldsymbol{p}} - \omega^{ex}_{H\boldsymbol{p}})} \psi^{Pt}_{i}(\boldsymbol{p}) \quad , \\ \psi^{L}_{i}(\boldsymbol{p}) &= \frac{\Omega_{Lc}}{2(\omega^{pol}_{i\boldsymbol{p}} - \omega^{ex}_{L\boldsymbol{p}})} \psi^{Pt}_{i}(\boldsymbol{p}) \quad . \end{aligned} \tag{7}$$

The polariton energies ω_{ip}^{pol} of the ith branchs with wave vector $\boldsymbol{p}$ are determined by the equation :

$$\frac{\omega_{\boldsymbol{p}}}{\omega_{i\boldsymbol{p}}^{pol}} = 1 + \frac{\Omega_{Hc}^2}{4\omega_{i\boldsymbol{p}}^{pol}(\omega_{H\boldsymbol{p}}^{ex} - \omega_{i\boldsymbol{p}}^{pol})} + \frac{\Omega_{Lc}^2}{4\omega_{i\boldsymbol{p}}^{pol}(\omega_{L\boldsymbol{p}}^{ex} - \omega_{i\boldsymbol{p}}^{pol})} \quad , \tag{8}$$

and equal

$$\begin{aligned} \omega_1^{pol}(\boldsymbol{p}) &= r\cos\left(\frac{\varphi}{3}\right) - \frac{A}{3} \quad , \\ \omega_2^{pol}(\boldsymbol{p}) &= -r\cos\left(\frac{\varphi}{3} + \frac{\pi}{3}\right) - \frac{A}{3} \quad , \\ \omega_3^{pol}(\boldsymbol{p}) &= -r\cos\left(\frac{\varphi}{3} - \frac{\pi}{3}\right) - \frac{A}{3} \quad , \end{aligned} \tag{9}$$

where :

$$\begin{aligned} r &= 2\sqrt{\frac{1}{3}\left(\frac{A^2}{3} - B\right)} \quad , \\ \varphi &= \arccos\left[\frac{-\left(\frac{2A^3}{27} - \frac{AB}{3} + C\right)}{2\sqrt{\frac{1}{27}\left(\frac{A^2}{3} - B\right)^3}}\right] \quad , \end{aligned} \tag{10}$$

$$\begin{aligned} A &= -(\omega_{H\boldsymbol{p}} + \omega_{L\boldsymbol{p}} + \omega_{\boldsymbol{p}}) \quad , \\ B &= \omega_{H\boldsymbol{p}}\omega_{\boldsymbol{p}} + \omega_{L\boldsymbol{p}}\omega_{\boldsymbol{p}} + \omega_{H\boldsymbol{p}}\omega_{L\boldsymbol{p}} - \frac{1}{4}(\Omega_{Hc}^2 + \Omega_{Lc}^2) \quad , \\ C &= \frac{1}{4}\Omega_{Hc}^2\omega_{L\boldsymbol{p}} + \frac{1}{4}\Omega_{Lc}^2\omega_{H\boldsymbol{p}} - \omega_{H\boldsymbol{p}}\omega_{L\boldsymbol{p}}\omega_{\boldsymbol{p}} \quad . \end{aligned} \tag{11}$$

Here we denote that $i = 1$ is the lower polariton branch, $i = 3$ is the upper polariton branch and $i = 2$ is the middle one.

The annihilation operators $B_{i\boldsymbol{p}}$ of the ith branch polariton can be written as :

$$B_{i\boldsymbol{p}} = \psi_i^{Pt}(\boldsymbol{p})b_{\boldsymbol{p}} + \psi_i^{H}(\boldsymbol{p})a_{H\boldsymbol{p}} + \psi_i^{L}(\boldsymbol{p})a_{L\boldsymbol{p}} \quad . \tag{12}$$

We can obtain also the inverse relations :

$$\begin{aligned} b_{\boldsymbol{p}} &= \psi_1^{Pt}(\boldsymbol{p})B_{1\boldsymbol{p}} + \psi_2^{Pt}(\boldsymbol{p})B_{2\boldsymbol{p}} + \psi_3^{Pt}(\boldsymbol{p})B_{3\boldsymbol{p}} \quad , \\ a_{H\boldsymbol{p}} &= \psi_1^{H}(\boldsymbol{p})B_{1\boldsymbol{p}} + \psi_2^{H}(\boldsymbol{p})B_{2\boldsymbol{p}} + \psi_3^{H}(\boldsymbol{p})B_{3\boldsymbol{p}} \quad , \\ a_{L\boldsymbol{p}} &= \psi_1^{L}(\boldsymbol{p})B_{1\boldsymbol{p}} + \psi_2^{L}(\boldsymbol{p})B_{2\boldsymbol{p}} + \psi_3^{L}(\boldsymbol{p})B_{3\boldsymbol{p}} \quad . \end{aligned} \tag{13}$$

3. Phonoriton in three-band model

We suppose a intense coherent laser beam with frequency ω_o near the exciton resonance enters the semiconductor and creates a polariton wave. Denote ω_L and ω_T – the longitudinal and tranverse energies of a bare exciton. Below ω_T only one exciton polariton branch

with $i = 1$ exist and far below ω_T it is fundamentally photon-like. Above the ω_L, one external photon can generate three simultaneously propagating exciton-polariton modes of the same polarization in semiconductors. Near the ω_L they are primarily exciton-like, far above ω_L the upper polariton branch arises which quickly takes on predominantly photon character. For simplicity, we choose $\omega_o < \omega_T$, in this case only one $i = 1$ exciton polariton branch was generated by incident photon and we denote this wave as $\omega_o = \omega_1^{pol}(\boldsymbol{k}_o) = \omega_{k_o}$.

To develop the model, we use here the J.L. Birman and B.S. Wang picture [20-21]. When the intensity of the laser beam is high, the polariton mode ω_{k_o} will be "macroscopically occupied". But for all other modes, the occupation number is near zero. Thus the interaction terms related to mode ω_{k_o} should be treated seperately. On the other hand, since the polariton-phonon interaction not related to the mode ω_{k_o} is rather weak; this merely contributes to a nonzero imaginary part of the polariton energy. Therefore, we drop these terms. Then we divide the system into three parts : incident polariton, phonon, and scattered polariton. We concentrate on the the scattered polaritons and phonons. They are coupled by the polariton-phonon interaction $MB_{k_o}^+$ and MB_{k_o}. If the incident laser light is coherent and the intensity high enough, to a good approximation we can replace the operators $B_{k_o}^+$ and B_{k_o} in the interaction term by their number expectation value $< B_{k_o}^+ >$ and $< B_{k_o} >$, respectively :

$$\begin{aligned} B_{k_o}^+ \longrightarrow < B_{k_o}^+ > &= \alpha_{k_o}e^{-i\omega_{k_o}t} = \alpha^*(t) \quad , \\ B_{k_o} \longrightarrow < B_{k_o} > &= \alpha_{k_o}e^{i\omega_{k_o}t} = \alpha(t) \quad , \end{aligned} \tag{14}$$

with :

$$\alpha_{k_o} \approx \sqrt{VN_o} \quad , \tag{15}$$

where N_o is the occupation number of the incident polariton, V is the volume of the crystal. These two numbers $\alpha_{k_o}^*$ and $\alpha(t)$ come from the "eigenvalues" of the annihilation operators B_{k_o} acting on a coherent state of the polariton mode ω_{k_o}, which occurs because of the intensity of pumping. We obtain the Hamiltonian :

$$\begin{aligned} H_{k_o} = &\sum_{i\boldsymbol{p}} \omega_{i\boldsymbol{p}}^{pol}\, B_{i\boldsymbol{p}}^+ B_{i\boldsymbol{p}} + \sum_{\boldsymbol{p}} \Omega_{\boldsymbol{p}}\, c_{\boldsymbol{p}}^+ c_{\boldsymbol{p}} + \\ &+ \sum_{i\boldsymbol{p}} \Big\{ M_{i1}'(\boldsymbol{p} - \boldsymbol{k}_o)\, \alpha_{k_o} \left(e^{i\omega_{k_o}t} B_{i\boldsymbol{p}}^+ c_{\boldsymbol{p}-\boldsymbol{k}_o} + e^{-i\omega_{k_o}t} B_{i\boldsymbol{p}} c_{\boldsymbol{p}-\boldsymbol{k}_o}^+ \right) \\ &+ M_{i1}'(\boldsymbol{p} - \boldsymbol{k}_o)\, \alpha_{k_o} \left(e^{-i\omega_{k_o}t} B_{i\boldsymbol{p}} c_{\boldsymbol{k}_o-\boldsymbol{p}} + e^{i\omega_{k_o}t} B_{i\boldsymbol{p}}^+ c_{\boldsymbol{k}_o-\boldsymbol{p}}^+ \right) \Big\} \quad . \end{aligned} \tag{16}$$

Denote $n_{i\boldsymbol{p}} = B_{i\boldsymbol{p}}^+ B_{i\boldsymbol{p}}$ the number operator of the i th branch exciton polariton with momentum $\boldsymbol{p}$, we have the relations :

$$\begin{aligned} B_{i\boldsymbol{p}}^+ n_{k\boldsymbol{q}} &= (n_{k\boldsymbol{q}} - 1)B_{i\boldsymbol{p}}^+ \quad , \\ B_{i\boldsymbol{p}}\, n_{k\boldsymbol{q}} &= (n_{k\boldsymbol{q}} + 1)B_{i\boldsymbol{p}} \quad . \end{aligned} \tag{17}$$

Using an unitary transformation :

$$u(t) = e^{i\omega_{k_o} t \sum_{kq} n_{kq}} , \tag{18}$$

$$\tilde{H}_{k_o} = u^+ H_{k_o} u + i u^+ \frac{\partial u}{\partial t} , \tag{19}$$

we can eliminate the time-dependent factors $e^{\pm i\omega_{k_o} t}$ in (15) and find the following expression for the Hamiltonian :

$$\tilde{H}_{k_o} = \sum_{ip} \left(\omega_{ip}^{pol} - \omega_{k_o} \right) B_{ip}^+ B_{ip} + \sum_{p} \Omega_p c_p^+ c_p + + H_S + H_{AS} , \tag{20}$$

where H_S is the Stokes scattering Hamiltonian :

$$H_S = \sum_{ip} M'_{i1}(p - k_o)\alpha_{k_o} \left(B_{ip} c_{k_o - p} + B_{ip}^+ c_{k_o - p}^+ \right) , \tag{21}$$

and H_{AS} is the anti-Stokes scattering Hamiltonian :

$$H_{AS} = \sum_{ip} M'_{i1}(p - k_o)\alpha_{k_o} \left(B_{ip}^+ c_{p - k_o} + B_{ip} c_{p - k_o}^+ \right) . \tag{22}$$

Since the interaction between the spectra at anti-Stokes and Stokes frequencies is essentially a high-order effect and because of their separation $2\Omega_p$ is much biger than the binding energy of bare exciton E_{ox}, we may study the spectra around Stokes and anti-Stokes frequencies individually.

For the case of Stokes scattering, we have the same results of [20-21]. The Stokes Hamiltonian can be diagonalized to give phonoriton branches $\omega_i^S(p)$:

$$\tilde{H}_S^{phn} = \sum_{p} \left\{ \omega_1^S(p) d_{1p}^+ d_{1p} + \omega_2^S(p) d_{2p}^+ d_{2p} \right\} , \tag{23}$$

by unitary transformations :

$$\begin{aligned} d_{1p} &= x(p) B_{1p} + y(p) c_{k_o - p}^+ , \\ d_{2p} &= y(p) B_{1p}^+ + x(p) c_{k_o - p} , \end{aligned} \tag{24}$$

where d_{1p} and d_{2p} are the annihilation oterators of Stokes scattered phonoritons.

We have the expression for $\omega_{1,2}^S(p)$ of Stokes scattered phonoritons as follows :

$$\omega_{1,2}^S(p) = \frac{1}{2} \left(\omega_{1p}^{pol} - \Omega_{k_o - p} - \omega_{k_o} \right) \pm \frac{1}{2} \left\{ \left(\omega_{1p}^{pol} + \Omega_{k_o - p} - \omega_{k_o} \right)^2 - 4 \left(M'_{11}(p - k_o)\alpha_{k_o} \right)^2 \right\}^{1/2} . \tag{25}$$

Note that an "imaginary" gap appears in the case of Stokes scattering.

For the case of anti-Stokes scattering, the diagonalized Hamiltonian, in general, is :

$$\tilde{H}_{AS}^{phn} = \sum_{\mu p} \omega_\mu^{AS}(p) D_{\mu p}^+ D_{\mu p} , \tag{26}$$

where $\mu = 0, 1, 2, 3$ are branch numbers of phonoritons, $D_{\mu p}$ are the annihilation operator of anti-Stokes scattered phonoriton, we have :

$$D_{\mu p} = u_\mu(p) c_{p - k_o} + v_{1\mu}(p) B_{1p} + v_{2\mu}(p) B_{2p} + v_{3\mu}(p) B_{3p} , \tag{27}$$

here $u_\mu(p)$ and $v_{i\mu}(p)$ are the weights of the phonon and ith branch polariton ($i = 1, 2, 3$) in the μth branch phonoriton ($\mu = 0, 1, 2, 3$)

$$u_\mu(p) = \left\{ 1 + \frac{\left[M'_{11}(p - k_o)\alpha_{k_o} \right]^2}{\left(\omega_\mu^{AS}(p) + \omega_{k_o} - \omega_{1p}^{pol} \right)^2} + \frac{\left[M'_{21}(p - k_o)\alpha_{k_o} \right]^2}{\left(\omega_\mu^{AS}(p) + \omega_{k_o} - \omega_{2p}^{pol} \right)^2} + + \frac{\left[M'_{31}(p - k_o)\alpha_{k_o} \right]^2}{\left(\omega_\mu^{AS}(p) + \omega_{k_o} - \omega_{3p}^{pol} \right)^2} \right\}^{-1/2} ,$$

$$v_{i\mu}(p) = \frac{M'_{i1}(p - k_o)\alpha_{k_o}}{\omega_\mu^{AS}(p) + \omega_{k_o} - \omega_{1p}^{pol}} u_\mu(p) . \tag{28}$$

The values $\omega_{\mu p}^{AS}$ of the anti-Stokes scattered phonoritons are determined from the equation :

$$\frac{\Omega_{p - k_o}}{\omega_\mu^{AS}(p)} = 1 + \frac{\left[M'_{11}(p - k_o)\alpha_{k_o} \right]^2}{\omega_\mu^{AS}(p) \left(\omega_{1p}^{pol} - \omega_{k_o} - \omega_\mu^{AS}(p) \right)} + \frac{\left[M'_{21}(p - k_o)\alpha_{k_o} \right]^2}{\omega_\mu^{AS}(p) \left(\omega_{2p}^{pol} - \omega_{k_o} - \omega_\mu^{AS}(p) \right)} + + \frac{\left[M'_{31}(p - k_o)\alpha_{k_o} \right]^2}{\omega_\mu^{AS}(p) \left(\omega_{3p}^{pol} - \omega_{k_o} - \omega_\mu^{AS}(p) \right)} . \tag{29}$$

The equation (29) can be solved numerically, that will be done later in this paper for GaAs. By analogy with similar oscillator equation in atomic physics, we can say that the new gap Δ_i appear. These new gaps Δ_i are proportional to $(VN_o)^{1/2} M'_{i1}(p - k_o)$ so they not only depend on the properties of the material but are also proportional to the square root of the intensity of the pump light.

If we consider $\omega_{k_o} < \omega_T - \Omega_p$, only the lower branch ($i = 1$) of the polariton will be reconstructed and we return to the results of two-level model [20-21]

$$\begin{aligned} D_{op} &= u(p)c_{p-k_o} + v(p)B_{1p} \quad , \\ D_{1p} &= -v(p)c_{p-k_o} + u(p)B_{1p} \quad , \\ D_{2p} &= B_{2p} \quad , \\ D_{3p} &= B_{3p} \quad , \end{aligned} \tag{30}$$

where

$$\begin{aligned} u(p) &= \left\{ 1 + \frac{\left[M_{11}'(p-k_o)\alpha_{k_o}\right]^2}{\omega_1^{AS}(p) + \omega_{k_o} - \omega_{1p}^{pol}} \right\}^{-1/2} \quad , \\ v(p) &= \frac{M_{11}'(p-k_o)\alpha_{k_o}}{\omega_1^{AS}(p) + \omega_{k_o} - \omega_{1p}^{pol}}\, u(p) \quad , \end{aligned} \tag{31}$$

and we have :

$$\begin{aligned} \omega_{1,2}^{S}(p) &= \frac{1}{2}\left(\omega_{1p}^{pol} + \Omega_{p-k_o} - \omega_{k_o}\right) \pm \frac{1}{2}\left\{ \left(\omega_{1p}^{pol} - \Omega_{p-k_o} - \omega_{k_o}\right)^2 \right. \\ &\left. + 4\left(M_{11}'(p-k_o)\alpha_{k_o}\right)^2 \right\}^{1/2} \quad . \end{aligned} \tag{32}$$

Note that here a new gap Δ_1 appears :

$$\Delta_1 = \left\{ \left(\omega_{1p}^{pol} + \Omega_{p-k_o} - \omega_{k_o}\right)^2 \pm \left[\sqrt{VN_o}M_{11}'(p-k_o)\right]^2 \right\}^{1/2} \quad , \tag{33}$$

and it is proportional to the square root of the occupation number of the incident polariton.

The physical origin of the new gaps was explained clearly in the previous work [21]. The presence of strong pump polariton ω_{k_o} in the semiconductor makes any polariton with wave vector k interact with a phonon with wave vector $k \pm k_o$ and frequency $\omega_{ik}^{pol} \pm \omega_{k_o}$. In the system of semiconductor plus existing strong waves, the nonlinear phonon-polariton interaction lead to a linear coupling between phonon and polariton, it plays the role of new periodic "potential" or diffraction "grating" with spatial periodicity $2\pi/k_o$ for polaritons and phonons that satisfy the scattering condictions and produces the spectrum reconstruction.

4. Group velocity of phonoriton

Resonant Brillouin Scattering experiments in the most direct evidence of the existence of exciton-polaritons. Another dramatic direct proof of the existence of these coupled-modes is provided by time-of-flight measurements. In that experiments, the group velocity

of exciton-polaritons was measured by time-resolved spectroscopy [11-22,32-33]. It gives us an idea of using time-of-flight measurements for providing another direct test of the existence of phonoritons. For this purpose we calculate the group velocity of phonoritons.

The group velocity of polariton can be easily derived from the equation (8) by taking its derivative both sides, we have :

$$v_{ip}^{pol} = \psi_i^{Pt}(p)^2 v_p^o + \psi_i^H(p)^2 v_p^H + \psi_i^L(p)^2 v_p^L \quad , \tag{34}$$

where v_{ip}^{pol} is the group velocity of ith branch polariton and v_p^0, v_p^H, v_p^L are the group velocities of the photon, heavy exciton, light exciton at wave vector p, respectively:

$$\begin{aligned} v_{ip}^{pol} &= \frac{\partial \omega_{ip}^{pol}}{\partial p} \quad , \quad v_p^0 = \frac{1}{\sqrt{\epsilon_b}} \quad , \\ v_p^H &= \frac{p}{M_H}\theta(\omega_{Hp} - \omega_T) \quad , \quad v_p^L = \frac{p}{M_L}\theta(\omega_{Lp} - \omega_T) \quad , \end{aligned} \tag{35}$$

$\theta(x)$ is the step function :

$$\theta(x) = \begin{cases} 1 & \text{if } x \geq 0 \\ 0 & \text{if } x < 0 \end{cases}$$

Using the formulae (25) and (29) we can derive the following expressions for the group velocity of ith branch of phonoriton $v_{ip} = \frac{\partial \omega_i(p)}{\partial p}$,

$$\begin{aligned} v_{1p}^{S} &= x^2(p)v_{1p}^{Pol} + y^2(p)v_p^{Ph} \quad , \\ v_{2p}^{S} &= y^2(p)v_{1p}^{Pol} + x^2(p)v_p^{Ph} \quad , \end{aligned} \tag{36}$$

for the case of Stokes scattering, and :

$$v_\mu^{AS}(p) = u_\mu^2(p)v_{p-k_o}^{Ph} + v_{1\mu}^2(p)v_{1p}^{pol} + v_{2\mu}^2(p)v_{2p}^{Pol} + v_{3\mu}^2(p)v_{3p}^{pol} \quad , \quad \mu = 0,1,2,3 \quad . \tag{37}$$

for the case anti-Stokes scattering.

Here $v_p^{pol} = \frac{\partial \Omega_p}{\partial p}$ is the group velocity of phonon.

The expression (37) can be rewritten as :

$$\begin{aligned} v_\mu^{AS}(p) &= u_\mu^2(p)v_{p-k_o}^{Ph} + \sum_i \left[v_{1\mu}^2(p)\psi_i^{Pt}(p)^2 v_p^o \right. \\ &\left. + v_{i\mu}^2(p)\psi_i^H(p)^2 v_p^H + v_{i\mu}^2(p)\psi_i^L(p)^2 v_p^L \right] \quad . \end{aligned} \tag{38}$$

If we choose $\omega_{k_o} < \omega_T - \Omega_p$, then we can use results of the two-level model, we have :

$$\begin{aligned} v_o^{AS}(p) &= u^2(p)v_p^{Ph} + v^2(p)v_{1p}^{pol} \\ v_1^{AS}(p) &= v^2(p)v_p^{Ph} + u^2(p)v_{1p}^{pol} \end{aligned} \tag{39}$$

Now we concentrate on the case of anti-Stokes scattering with appearance of a new gap.

The behaviour of phonoriton group velocities may be seen directly from the slopeof their dispersion curve. For the simple two-level model, and in the absence of damping, if a new gap appeared, there will be a minimum of phonoriton group velocities at the energy $\omega_{k_o} + \Omega_p$. Even if damping is included, generally the phonoriton group velocities will decrease several order of magnitude as the phonoriton energy is changes via $\omega_{k_o} + \Omega_p$. The velocity of energy propagation may be experimentally determined by means of time resolved spectroscopy. The observation of the strong variation of the group velocity at energy $\omega_{k_o} + \Omega_p$ will provide another direct proof of the existence of phonoriton.

Acknowledgments

The authors would like to thank Professor J.L.Birman for his interest in this work and sending the galley proofs of his papers. Two of us (N.A.V. and N.T.Q.H.) would like to take this opportunity to express his gratitude to Prof. Abdus Salam, the International Atomic Energy Agency and UNESCO for hospitality at the ICTP, Trieste. Interest of Prof. Yu Lu in this work is much appreciated.

References

1. A.L. Ivanov and L.V. Keldysh, Zh. Eksp. Teor. Fiz. **84**, 404 (1982).
2. D.S. Chemla, W.H. Knox, D.A.B. Miller, S. Schmitt-Rink, J.B. Stark and R. Zimmerman, J. Lumin. **44**, 233 (1989).
3. T. Higashimura, T. Tida and T. Komutsu, Phys. Stat. Solidi (b) **150**, 431 (1988).
4. B.I. Greene, J.F. Mueller, J. Orenstein, D.H. Rapkine, S. Schmitt-Rink and M. Thakar, Phys. Rev. Lett. **61**, 325 (1988).
5. T. Ishihara, J. Phys. Soc. Jap. **57**, 2573 (1988).
6. A.L. Ivanov and V.V. Panashenko, Pis'ma Zh. Eksp. Teor. Fiz. **49**, 34 (1989) [JETP Lett. **49**, 39 (1989)].
7. M. Combescot and R. Combescot, Phys. Rev. B **40**, 3788 (1989).
8. M. Combescot, Phys. Rev. B **41**, 3517 (1990).
9. A.L. Ivanov, L.V. Keldysh and V.V. Panashenko, Zh. Eksp. Teor. Fiz. **99**, 641 (1991).
10. W. Brenig, R. Zeyher and J.L. Birman, Phys. Rev. B **6**, 4617 (1972).
11. E.S. Koteles, in *Excitons*, edited by E.I. Rashba and M.D. Sturge (North- Holland Publishing Company-Amsterdam. New York. Oxford, 1982).
12. R.G. Ulbrich and C. Weisbuch, in *Light Scattering in Solids III*, Vol.51 of *Topics of Applied Physics*, edited by M. Cardona and G. Gutherodt (Springer-Verlag, Berlin, 1982).
13. G. Winterling and E.S. Koteles, Solid State Commun. **23**, 95 (1977).
14. B. Sermage and G. Fishman, Phys. Rev. Lett. **43**, 1043 (1979); Phys.Rev. B **23**, 5107 (1981).
15. A.L. Ivanov and L.V. Keldysh, Zh. Eksp. Teor. Fiz. **84**, 404 (1982); Dokl.Acad.Nauk. SSSR, **264-265**, 1363 (1982).
16. A.L. Ivanov, Zh. Eksp. Teor. Fiz. **90**, 158 (1986).
17. L.V. Keldysh and S.G. Tikhodeev, Zh. Eksp. Teor. Fiz. **90**, 1582 (1986); **91**, 78 (1986).
18. N.A. Gippius, L.V. Keldysh and S.G. Tikhodeev, Zh. Eksp. Teor. Fiz. **90**, 2213 (1986).
19. A.L. Ivanov and G.S. Vyskovskii, Phys. Status Solidi (b) **150**, 443 (1988).
20. B.S. Wang and J.L. Birman, Solid State Commun. **75**, 867 (1990).
21. B.S. Wang and J.L. Birman, Phys. Rev. B **42**, 9609 (1990).
22. A. Baldereschi and N.O. Lipari, Phys. Rev. B **3**, 439 (1971).
23. O. Kane, Phys. Rev. B **11**, 3850 (1975).
24. G. Fishman, Ann. Phys. **5**, 5 (1980).
25. G. Fishman, Solid State Commun. **27**, 1097 (1978); J. Luminescence **18-19**, 289 (1979).
26. Hoang Ngoc Cam, Nguyen Van Hieu and Nguyen Ai Viet, Ann. Phys. **164**, 172 (1985).
27. Ch.Khamsasy, Nguyen Van Hieu and Nguyen Ai Viet, Phys. Stat. Sol.(b) **144**, 645 (1987).
28. Nguyen Van Hieu and Nguyen Ai Viet, Dokl. Acad. Nauk SSSR, **305** 608 (1989) ; **306**, 1110 (1989).

29. Nguyen Que Huong, Nguyen Toan Thang and Nguyen Ai Viet, Phys. Stat. Sol.(b) **161**, 637 (1990); Proc. NCSR Vietnam **3**, 31 (1991).
30. Nguyen Ai Viet, Sov. J. Part. Nucl. **20**, 466 (1989).
31. Nguyen Que Huong and Nguyen Ai Viet, Czech. J. Phys. B **41**, 277 (1991).
32. Y.Masumoto, Y.Y.Unuma, Y.Tanaka and S.Shionoya, J. Phys. Soc. Japan **47**, 1844 (1979).
33. R.G.Ulbrich and G.W.Fehrenbach, Phys. Rev. Lett. **43**, 963 (1979).