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# RESONANT BRILLOUIN SCATTERING IN SPATIALLY DISPERSIVE MEDIUM+

E.L. Albuquerque and C.E.T. Gonçalves da Silva*

*Departamento de Física, Universidade Federal do Rio Grande do Norte, 59 000 - Natal, RN, Brazil*
**Instituto de Física, UNICAMP, 13 100 - Campinas, SP, Brazil*

Résumé - Nous présentons quelques aspects qualitatifs, qui découlent de la symétrie, de la diffusion Brillouin à l'interface vide-milieu cubique dispersif. Nous démontrons l'absence d'effet du polariton d'exciton sur le pic dû à l'onde de Rayleigh.

Abstract - We discuss qualitative, symmetry dependent, features of Brillouin scattering by an interface vacuum - cubic dispersive medium. The peak due to Rayleigh wave is shown to be unaffected by exciton polariton formation.

Spatial dispersion (SD) effects due to wavevector dependence of the dielectric function have been extensively studied since the phenomenological model of Pekar /1/ and Hopfield and Thomas /2/. Recently, the great development of tunable lasers stimulated a very thorough investigation of these effects and led to the elucidation of several important features of SD materials. In particular, resonant Brillouin scattering (RBS) is a very useful tool to clarify their fundamental properties, mainly in reasonably pure, direct-gap semiconductors at sufficiently low temperatures /3, 4, 5, 6/.

The purpose of this communication is to present some theoretical considerations about RBS via exciton polaritons in a SD medium. Due to space limitations, we discuss here only qualitative features of the spectra. Complete details, including numerically evaluated Brillouin spectra, are given in a forthcoming paper by the authors.

Let an electromagnetic wave, of frequency $\omega_I$ and wavevector $\vec{k}_I$ be incident, at an angle $\theta_I$, from the vacuum into a semi-infinite medium which shows SD effect. The SD medium is assumed to occupy the half-space $z < 0$ with a flat surface in the $z = 0$ plane. The SD medium can be described by a dieletric function $\varepsilon(\omega, \vec{k})$ given by

$$\varepsilon(\omega, \vec{k}) = \varepsilon_\infty + \chi(\omega, \vec{k}) \qquad (1)$$

where $\varepsilon_\infty$ is the background dielectric constant, and

$$\chi(\omega, \vec{k}) = s(\omega_0^2 - \omega_I^2 - i\omega_I \Gamma + D\,k^2)^{-1} \qquad (2)$$

is the exciton susceptibility. In (2), $s$ is the oscillator streght, $\omega_0$ is the frequency of the uncoupled mode, $\Gamma$ is the damping coefficient and $D = \hbar.\omega_0/m^*$, where $m^*$ is the exciton effective mass.

In the SD medium, the exciton polariton interacts with a crystal excitation of wavevector $\vec{Q}$ and frequency $\omega$ to produce a scattered wave of frequency $\omega_S = \omega_I - \omega$. This interaction is mediated by a deformation potential, where an i-polarized (i can be x, y, or z) acoustic mode

$$\mu^i = \mu^i\,(Q^z)\,\exp\,(i\,\vec{Q}.\vec{r} + i\,\omega\,t) \qquad (3)$$

changes the band gap of the SD medium by an amount

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$$\delta E_g = -c(Q^z)\ \mu^{ij}\ (Q^z)^* \tag{4}$$

where $\mu^{ij}$ means $\partial\mu_i/\partial_r j$ . This effect can be considered in the equation of motion of the exciton field $\vec{p}$ by augmenting it, as a perturbation, with the term

$$\eta^{ijkl}\ p^j\ \mu^{kl}\ (Q^z)^* \tag{5}$$

Here, $\eta^{ijkl}$ is a fourth-rank tensor which measures the coupling between $\vec{p}$ and $\vec{\mu}$; i,j are labels related with the polarization of the incident and scattered light respectively and k, l are labels related with the polarization of the acoustic phonons. In (5), the convention that repeated superscript are summed over is adopted. If we consider a SD medium with cubic symmetry, the only non-vanishing components of $\eta$ are /7/ $\eta^{xxxx} = \eta^{11}$, $\eta^{xxyy} = \eta^{12}$, $\eta^{yzyz} = \eta^{44}$ and their symmetric counterparts deduced by permutation of the indices x, y and z.

For an incident light wave, s-polarized, we have two transverse exciton polariton branches excited, while for an incident light wave, p-polarized, the longitudinal mode is also excited. Therefore, for scattering by bulk acoustic phonons, this means that we have, for appropriate incidence frequency $\omega_I$, (a) four stokes (or anti-stokes) resonant lines in s → s scattering, (b) six resonant lines in either s → p or p → s scattering, and (c) nine resonant lines in p → p scattering.

Table I shows all these results, where we have considered the electric field of the light $\vec{E} = (0, E^y, 0)$ for s-polarization and $\vec{E} = (E^x, 0, E^z)$ for p-polarization.

| Non-vanishing tensor | Type of scattering | Phonon polarization | Rayleigh wave |
|---|---|---|---|
| $\eta^{yyxx}$ | s → s | xz - plane | No |
| $\eta^{yyzz}$ | s → s | xz - plane | Yes |
| $\eta^{xyxy}$ | s → p | y - direction | No |
| $\eta^{zyzy}$ | s → p | y - direction | No |
| $\eta^{xxxx}$ | p → p | xz - plane | No |
| $\eta^{xxzz}$ | p → p | xz - plane | Yes |
| $\eta^{zzzz}$ | p → p | xz - plane | Yes |
| $\eta^{zzxx}$ | p → p | xz - plane | No |
| $\eta^{zxzx}$ | p → p | xz - plane | Yes |
| $\eta^{xzxz}$ | p → p | xz - plane | Yes |
| $\eta^{yxyx}$ | p → s | y - direction | No |
| $\eta^{yzyz}$ | p → s | y - direction | No |

TABLE 1

The last column of Table 1 gives information about the possibility or not to have the propagation of surface waves of the Rayleigh type. One important question about this mode is whether it gives information about the exciton polariton in the Brillouin spectrum. To answer this question let us consider the projection on the x-y plane of the scattered wave vector $\vec{k}_s$ (see Figure 1).

The Rayleigh wave is characterized by a 2D wave vector $\vec{q}$, with frequency $\omega = v_R|\vec{q}|$. The kinematic equation for stokes scattering is

$$\vec{k}_{I//} = \vec{k}_{s//} + \vec{q} \tag{6}$$

Therefore, from (6) we have that

$$q^2 = \left(\frac{\omega_s}{c}\right)^2 \sin^2\theta_s + \left(\frac{\omega_I}{c}\right)^2 \sin^2\theta_I - 2\,\frac{\omega_s\omega_I}{c^2}\,\sin\theta_s\,\sin\theta_I\,\cos\phi \tag{7}$$

As $q = \omega/v_R$ and $\omega = \omega_I - \omega_s$ we have, together with (7), three simultaneous equations for q, $\omega$, and $\omega_s$. The solution of these equations determines the frequency $\omega_s$ at which the sharp scattering peak due to Rayleigh wave is seen, and hence $\omega_s$ is determined solely by x-y plane values $\vec{k}_{I//}$ and $\vec{k}_{s//}$. However, as $\vec{k}_{I//}$ and $\vec{k}_{s//}$ are the same for external light and all exciton-polariton branches, there is no splitting of Rayleigh peak due to exciton-polariton formation! This is a very important information since the Rayleigh peak could mask some resonant peaks.

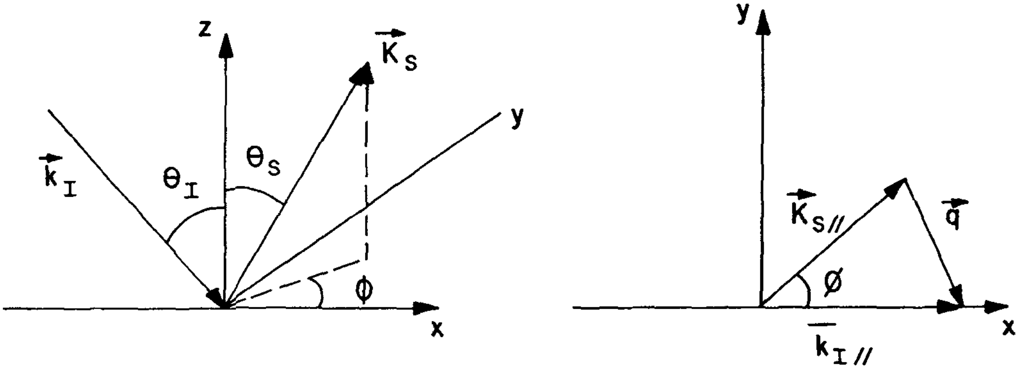


Figure 1: To illustrate the projections of the incident ($\vec{k}_I$) and scattered ($\vec{k}_s$) wavevetor and the Rayleigh wavevector $\vec{q}$.

So far we have discussed RBS considering that the interface between the SD medium and vacuum is flat. However, as it was pointed out by Loudon /8/ there is another mechanism that can also scatter light: the so-called surface-ripple mechanism. The general form of the cross section in this case is /8, 9/

$$\frac{d^2\sigma}{d\Omega\, d\omega_s} \;\alpha\; < |u^z\,(o)|^2 >_{Q^x,\ \omega} \tag{8}$$

where the constant of proportionality includes factors depending on $\theta_I$, $\theta_s$, Ø and the optical properties of the two media. The power spectrum <...> contains a $\delta$-function peak from the Rayleigh wave and broadened structure from the bulk acoustic modes. However, neither the Rayleigh peak nor the broad structure can be split by the exciton-polaritons branches, although the constant of proportionality must depend on properties of excitonic medium.

In summary, we have discussed the qualitative, symmetry dependent, properties of Brillouin scattering of light by a cubic spatially dispersive medium with a single exciton branch coupled to the eletromagnetic modes. Quantitative results, as well as the natural extensions of this work to include realistic exciton dispersion /10/ will be presented elsewhere. Of great interest also is the study of layered semiconductor structures, e.g., super-lattices /11/, which we are undertaking.

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