# Heterodyne generation of polaritons

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(Received 22 August 1972; revised manuscript received 21 May 1973)

A quantum-mechanical analysis is given which takes into account the damping of polaritons excited in a crystal by the two-beam method. The Heisenberg equations of motion are linearized by an approximation, well adapted to this method, in which the two beams are symmetrically considered. We get the polariton $P$ function which allows us to study the coherence and the time evolution of the polariton beam.

## I. INTRODUCTION

In crystals having optical modes exhibiting both Raman and infrared activities, it is possible to study polaritons by the Raman effect. As in the very center part of the Brillouin zone, the polariton frequency varies, these crystals are a potential material for tunable sources of infrared radiation.[1-5] If one can achieve stimulated Raman scattering, coherent infrared radiations are obtained; this is a possible way to generate far-infrared electromagnetic waves. There are two ways to obtain stimulated Raman scattering. In the first one a single pumping-light beam is used; as it propagates inside the crystal, it generates spontaneous polaritons and Stokes photons. As the number of these particles increases, stimulated emission becomes more and more important. A great number of coherent polaritons are emitted only for a sufficient pumping power. This pumping power is lowered in the experiment of Ref. 5 by putting the crystal inside an optical resonator in order to increase the electromagnetic energy density at the Stokes-beam frequency. In the second way, that we shall call the two-beam method (TBM), two coherent light beams, with a frequency difference equal to that of the expected polaritons, are simultaneously sent inside the crystal. Because of the nonlinearity associated with Raman scattering, a beat is driven between the two light beams: stimulated polaritons are generated with rather low powers of the two pumping-light beams. This heterodyne method to obtain a stimulated effect has been suggested by Kastler[6] and Kroll[7] for Brillouin scattering. Papoular[8] has generalized their proposition to Raman scattering by optical modes of crystals. We have experimentally shown[9] that optical phonons can be generated by the TBM with a very high intensity. Independently Coffinet and de Martini[2] and the present authors[1] have excited polariton modes by the TBM and have characterized infrared radiations outside the crystal.

Because of the recent progress observed in the tunable-dye-laser field, the TBM has interesting technical applications; it can also provide information on the solid-state properties of crystals.

In this paper we are considering a crystal having only one infrared-active optical mode which is not supposed to be undamped; we study the time behavior of the number of polaritons excited by the TBM and the coherency of the corresponding beam. Glauber's formalism[10] for coherent states is used. The damping of polaritons is introduced by considering the lattice-vibration anharmonicity.[11-13] We do not use the approximation usually made to study parametric interactions, i.e., undepleted higher-frequency pumping beam, because it does not apply to the TBM, in which the two light beams must be symmetrically considered.

## II. HAMILTONIAN OF THE SYSTEM

We shall first derive the Hamiltonian for the polariton-damping process. We consider this damping as a consequence of the anharmonicity of the mechanical vibrations; in the expression for the potential energy we drop all the terms of order higher than three. The Hamiltonian describing the interaction between polaritons and lattice vibrations can then be written as[11,13]

$$H_I = \hbar (b^{\dagger}_{-\vec{k}_3 r_3} + b_{\vec{k}_3 r_3}) \sum_{\vec{k}_i \vec{k}_j r_i r_j}^{N} \sum V \begin{pmatrix} r_i & r_j & r_3 \\ \vec{k}_i & \vec{k}_j & \vec{k}_3 \end{pmatrix} \Delta(\vec{k}_i + \vec{k}_j + \vec{k}_3)(b^{\dagger}_{-\vec{k}_i r_i} + b_{\vec{k}_i r_i})(b^{\dagger}_{-\vec{k}_j r_j} + b_{\vec{k}_j r_j}).$$

The sums are taken over the $N$ wave vectors in the Brillouin zone and over all branches, $r_i$, $r_j$ of the frequency spectrum: $b^{\dagger}_{-\vec{k}_\lambda r_\lambda}$, $b_{\vec{k}_\lambda r_\lambda}$ $(\lambda = i,j)$ are phonon creation and annihilation operators, $b^{\dagger}_{-\vec{k}_3 r_3}$ and

$b_{\vec{k}_3 r_3}$ are the creation and annihilation operators for the polarization field[14] and $V(\ )$ is a coupling coefficient which satisfies[13]

$$V\begin{pmatrix} r_i & r_j & r_3 \\ \vec{k}_i & \vec{k}_j & \vec{k}_3 \end{pmatrix} = V^*\begin{pmatrix} r_i & r_j & r_3 \\ -\vec{k}_i & -\vec{k}_j & -\vec{k}_3 \end{pmatrix} .$$

The expression $b^\dagger_{-\vec{k}_3 r_3} + b_{\vec{k}_3 r_3}$ can be written as a linear combination of the operators describing polaritons.[14]

In crystals having one infrared-active mode, polaritons corresponding to the upper branch of the dispersion curve cannot be excited by the Raman effect,[15] hence we shall only consider in $H_I$ the terms describing the interaction of a polariton from the lower branch with two lattice phonons. The effect of the other terms is supposed to be negligible as long as the coupling described by $V(\ )$ remains weak; this will be assumed in the following.

$H_I$ can then be written

$$H_I = \hbar\mu(\omega_3) \sum_{\vec{k}_i \vec{k}_j}^{N} \sum_{r_i r_j} i\Delta(\vec{k}_3 - \vec{k}_i - \vec{k}_j) \left[ V\begin{pmatrix} r_i & r_j & r_3 \\ -\vec{k}_i & -\vec{k}_j & \vec{k}_3 \end{pmatrix} a_{\vec{k}_3 1} b^\dagger_{\vec{k}_i r_i} b^\dagger_{\vec{k}_j r_j} - V^*\begin{pmatrix} r_i & r_j & r_3 \\ -\vec{k}_i & -\vec{k}_j & \vec{k}_3 \end{pmatrix} a^\dagger_{\vec{k}_3 1} b_{\vec{k}_i r_i} b_{\vec{k}_j r_j} \right] ,$$

$$\mu(\omega_3) = \left( 4\pi\beta \frac{\omega_3/\omega_0}{(1 - \omega_3^2/\omega_0^2)^2 + 4\pi\beta} \right)^{1/2} ,$$

where $\omega_3$ and $4\pi\beta$ are respectively the polariton frequency and the mode oscillator strength; and $a^\dagger_{\vec{k}_3 1}$ and $a_{\vec{k}_3 1}$ are the creation and annihilation operators for polaritons from the lower branch of the dispersion curve. We shall write $H_I$ in a more simple form,

$$H_I = \hbar\mu(\omega_3) \sum_{i,j} (K_{ij} a_3 b_i^\dagger b_j^\dagger + K_{ij}^* a_3^\dagger b_i b_j) ,$$

$$K_{ij} = i\Delta(\vec{k}_3 - \vec{k}_i - \vec{k}_j) V\begin{pmatrix} r_i & r_j & r_3 \\ -\vec{k}_i & -\vec{k}_j & \vec{k}_3 \end{pmatrix} .$$

$\mu(\omega_3)$ is a quantity ranging from zero to unity; it expresses the fact that the damping of polaritons exists only through their phononlike part and so is growing as the frequency $\omega_3$ gets closer to the resonance frequency $\omega_0$.

Let us now derive the complete Hamiltonian for the system studied. For a Raman process, the Hamiltonian $H$ is usually written as[16]

$$H = \hbar\omega_1 a_1^\dagger a_1 + \hbar\omega_2 a_2^\dagger a_2 + \hbar\omega_3 a_3^\dagger a_3 - \hbar K(a_1^\dagger a_2 a_3 + a_1 a_2^\dagger a_3^\dagger) ;$$

$a_1$, $a_2$, $a_1^\dagger$, $a_2^\dagger$ are annihilation and creation operators of photons 1 (frequency $\omega_1$) and photons 2 (frequency $\omega_2$). Photons 1 and photons 2, respectively, belong to the higher-frequency beam (HF) and to the lower-frequency beam (LF); they are also called pumping and Stokes photons. $K$ is a coupling coefficient.

We then get the final form of the complete Hamiltonian

$$H_t = H + \sum_l \hbar\omega_l b_l^\dagger b_l + H_I .$$

The term $\sum_l \hbar\omega_l b_l^\dagger b_l$ represents all the phonon modes existing in the crystal, and acts as a reservoir of harmonic oscillators describing the loss mechanism to which the polaritons are coupled by means of $H_I$.[11]

With regard to the loss mechanism and following Refs. 11 and 17–19, we make the assumptions that the reservoir of oscillators has a very large number of closely spaced energy levels, and that it is only slightly affected by the interaction with the polariton field.

## III. STUDY OF TBM IN CASE OF POLARITON DAMPING

To study the properties and the time behavior of the polariton beam, we use the Heisenberg equations of motion.[20] Setting

$$a_{1,2}(t) = A_{1,2}(t) e^{-i\omega_{1,2} t} ,$$

we obtain

$$i\frac{da_3}{dt} = \omega_3 a_3 - KA_1 A_2^\dagger e^{-i(\omega_1 - \omega_2)t} + \sum_{i,j} \mu(\omega_3) K_{ij}^* b_i b_j , \tag{1}$$

$$i\frac{d}{dt}(b_i b_j) = (\omega_i + \omega_j) b_i b_j + [b_i b_j, H_I] . \tag{2}$$

Following Senitzki[18] and Opie[17] the commutator of the last equation is replaced by

$$a_3 \mu(\omega_3) \left\langle \left[ b_i(0) b_j(0), \sum_{pq} K_{pq} b_p^\dagger(0) b_q^\dagger(0) \right] \right\rangle .$$

The lattice phonons being in thermal equilibrium, we have, setting

$$\bar{n}_i = \langle b_i^\dagger(0) b_i(0) \rangle = (e^{\hbar\omega_i/kT} - 1)^{-1} ,$$

$$i\frac{d}{dt}(b_i b_j) = (\omega_i + \omega_j) b_i b_j + 2\mu(\omega_3) K_{ij} a_3 (1 + \bar{n}_i + \bar{n}_j) . \tag{3}$$

Equation (1) being nonlinear, we are making reasonable assumptions to linearize it with respect to $A_1$ and $A_2^\dagger$. First, the two pumping-light beams being powerful and coherent, the correspondence principle allows the replacement of the operators $a_1(t)$ and $a_2^\dagger(t)$ by the complex quantities $\alpha_1(t)$ and $\alpha_2^*(t)$. Before entering the crystal the two light beams have about equal intensities. Afterwards,

as the interaction occurs inside the crystal, the LF beam is amplified while the HF beam is attenuated; if we neglect the energy given to the polariton field, we can assume, as a first approximation, that $\alpha_1(t)\alpha_2^*(t)$ remains constant and equal to $\alpha_1(0)\alpha_2^*(0)$.

Equation (1) then becomes

$$i\frac{da_3}{dt} = \omega_3 a_3 - K\alpha_1(0)\alpha_2^*(0)e^{-i(\omega_1-\omega_2)t} + \sum_{i,j}\mu(\omega_3)K_{ij}^* b_i b_j \ . \quad (4)$$

Equations (3) and (4) are solved by taking the Laplace transforms[21] $\tilde{a}_\lambda(s)$ of the various operators $a_\lambda(t)$ and $\tilde{B}_{ij}(s)$ of the operator $b_i(t)b_j(t)$:

$$i[s\tilde{a}_3(s) - a_3(0)] = \omega_3\tilde{a}_3(s) + \sum_{ij}\mu(\omega_3)K_{ij}^*\tilde{B}_{ij}(s) - \frac{K\alpha_1\alpha_2^*}{s+i(\omega_1-\omega_2)} \ , \quad (5)$$

$$i[s\tilde{B}_{ij}(s) - b_i(0)b_j(0)] = (\omega_i+\omega_j)\tilde{B}_{ij}(s) + 2\mu(\omega_3)K_{ij}\tilde{a}_3(s)[1+\bar{n}_i+\bar{n}_j] \ . \quad (6)$$

$a_3(0)$, $b_i(0)$, $b_j(0)$ are the operators $a_3(t)$, $b_i(t)$, and $b_j(t)$ taken at time $t=0$.

To excite, with the TBM, $\omega_3$ frequency polaritons, $\omega_1$ and $\omega_2$ have to be chosen in such a way that $\omega_1-\omega_2=\omega_3$; if we suppose that this relation is fulfilled then we have

$$\tilde{a}_3(s) = \left(a_3(0) - i\sum_{ij}\mu(\omega_3)\frac{K_{ij}^* b_i(0)b_j(0)}{s+i(\omega_i+\omega_j)} + i\frac{K\alpha_1\alpha_2^*}{s+i\omega_3}\right) \Bigg/ \left(s+i\omega_3+\sum_{ij}2\mu^2\frac{|K_{ij}|^2(1+\bar{n}_i+\bar{n}_i)}{s+i(\omega_i+\omega_j)}\right). \quad (7)$$

We have[21]

$$a_3(t) = \frac{1}{2\pi i}\int_{\epsilon-i\infty}^{\epsilon+i\infty} ds\, e^{st}\tilde{a}_3(s) \ ,$$

where $\epsilon$ is a small positive number.

It is shown in the Appendix that

$$a_3(t) = u(t)a_3(0) + \sum_{ij}v_{ij}(t)b_i(0)b_j(0) + w(t)\alpha_1\alpha_2^* \ , \quad (8)$$

$$u(t) = e^{-i\omega_3 t}e^{-(\gamma/2)t - i\Delta\omega t} \quad (9)$$

$$v_{ij}(t) = -K_{ij}^*[e^{-i(\omega_i+\omega_j)t} - \exp(-\tfrac{1}{2}\gamma t - i\Delta\omega t - i\omega_3 t)]/[\omega_3 - (\omega_i+\omega_j) - i(\tfrac{1}{2}\gamma) + \Delta\omega] \quad (10)$$

$$w(t) = K\frac{1-e^{-(\gamma/2)t-i\Delta\omega t}}{\Delta\omega - \frac{1}{2}i\gamma}e^{-i\omega_3 t} \ . \quad (11)$$

$\frac{1}{2}\gamma$ and $\Delta\omega$ are defined in the Appendix.

From (8) one obtains the normal characteristic function $\chi_N(\eta_3)$ for the polariton beam

$$\chi_N(\eta_3) = \mathrm{Tr}(\rho e^{\eta_3 a_3^\dagger(t)}e^{-\eta_3^* a_3(t)}),$$

where $\rho$ is the density operator at time $t=0$:

$$\chi_N(\eta_3) = \exp(\eta_3 w^*\alpha_1^*\alpha_2 - \eta_3^* w\alpha_1\alpha_2^*]\,\mathrm{Tr}\{\rho\exp[\eta_3 u^* a_3^\dagger(0)]\exp[-\eta_3^* u a_3(0)] \times\exp\left(\eta_3\sum_{ij}v_{ij}^* b_i^\dagger(0)b_j^\dagger(0)\right)\exp\left(-\eta_3^*\sum_{ij}v_{ij}b_i(0)b_j(0)\right) \ . \quad (12)$$

From (12), we obtain:

$$\chi_N(\eta_3) = \exp[\eta_3 w^*\alpha_1^*\alpha_2 - \eta_3^* w\alpha_1\alpha_2^*]\chi_N'(\eta_3) \ ,$$

where $\chi_N'(\eta_3)$ is the normal characteristic function that would be obtained for the polariton field in the absence of the LF and HF beams. $\chi_N'(\eta_3)$ only describes the interaction between the polaritons which are in the crystal and the loss mechanism. According to the very general hypothesis made in Sec. II about the phonon reservoir, it can be shown

that this interaction describes a thermalization process.[17-19,22] If one assumes that the coupling between the polaritons and the loss mechanism has been existing since $t=-\infty$, then in the absence of any other coupling the polaritons are in thermal equilibrium at time $t$. $\chi'_N(\eta_3)$ is therefore the normal characteristic function associated with a polariton field in thermal equilibrium. Thus, we have[20,23]

$$\chi'_N(\eta_3)=e^{-|\eta_3|^2\bar{n}_3},$$

with

$$\bar{n}_3=(e^{\hbar\omega_3/kT}-1)^{-1};$$

hence

$$\chi_N(\eta_3)=\exp[-|\eta_3|^2\bar{n}_3+\eta_3 w^*\alpha_1^*\alpha_2 - \eta_3^* w\alpha_1\alpha_2^*]. \quad (13)$$

The expression of the $P(\alpha_3,t)$ function of polaritons is easily deduced,[23]

$$P(\alpha_3,t)=(1/\pi\bar{n}_3)\exp\{-[|\alpha_3-\bar{\alpha}_3(t)|^2]/\bar{n}_3\}, \quad (14)$$

where $\bar{\alpha}_3(t)=w(t)\alpha_1\alpha_2^*$. It is well known[10] that the $P$ function corresponding to the superposition of two fields is the convolution of the $P$ functions of each field.

Equation (14) shows that the polariton field obtained at time $t$ is the superposition of two fields that we shall call field I and field II. Field I is a Gaussian one, whose variance $\bar{n}_3$ does not depend upon time; it corresponds to polaritons existing initially inside the crystal and which are not amplified by the TBM. Field II is a coherent one, with a $P$ function given by $P_{II}=\delta(\alpha_3-\bar{\alpha}_3(t))$, it describes the signal, i.e., the polaritons created and amplified by the TBM. Therefore there is a coherent signal since the very beginning of the interaction; here is a noticeable advantage of the TBM over a method using only one beam. This result is obtained because the vacuum fluctuations of the LF beam are the only noise source for field II[24,25]; as soon as $a_2$ is replaced by $\alpha_2$ the quantum noise of this beam is neglected: it is then natural for the signal to have only a coherent component.

Neglecting $\Delta\omega$, we obtain from (14) the mean number $\langle n_3(t)\rangle$ of created polaritons at time $t$:

$$\langle n_3(t)\rangle=\bar{n}_3+(4K^2n_1n_2/\gamma^2;(1-e^{-\gamma t/2})^2, \quad (15)$$

where $n_1=n_1(0)=|\alpha_1|^2$ and $n_2=n_2(0)=|\alpha_2|^2$.

The expression (15) shows that the effect of damping is a growth limitation of the intensity of the coherent part of the signal. This coherent component exists for any value of $\gamma$; the maximum number of polaritons created inside the crystal is equal to $4K^2n_1n_2/\gamma^2$; the larger is the damping the smaller is this number which is also proportional to the product of the intensities of the incident-light beams.

We would like to emphasize the fact that the previous result is independent of the model chosen to describe the damping of polaritons; this is due to the fact that in the TBM the two beams are coherent and powerful enough to allow the replacement of the product $a_1(t)a_2^\dagger(t)$ by the complex quantity $\alpha_1(0)\alpha_2^*(0)$. Within this approximation the characteristic function, Eq. (12), will always be the product of two independent quantities, one of them describing the beat driven in the crystal by the LF and HF beams. The $P$ function, which is the Fourier transform of (12), will always be the convolution of two $P$ functions, one of them expressing the existence of a coherent field in the Glauber sense.

## IV. CONCLUSION

We have shown that, even in the case of damping, the polaritons created by the TBM are immediately coherent; this property of the polariton beam does not depend upon the model chosen to describe the losses. The unavoidable effect of damping due to lattice anharmonicity limits the growth of the signal.

## APPENDIX

One has to evaluate the function

$$a_3(t)=\frac{1}{2\pi i}\int_{\epsilon-i\infty}^{\epsilon+i\infty} ds\, e^{st}\tilde{a}_3(s).$$

Setting $s=\epsilon+iy$, we obtain

$$a_3(t)=\frac{1}{2\pi i}\int_{-\infty}^{+\infty} dy\, e^{iyt}e^{\epsilon t}\left[\left(a_3(0)-\sum_{ij}\frac{\mu(\omega_3)K_{ij}^*b_i(0)b_j(0)}{y+\omega_i+\omega_j-i\epsilon}+\frac{K\alpha_1\alpha_2^*}{y+\omega_3-i\epsilon}\right)\Big/(y+\omega_3-i\epsilon+I)\right], \quad (16)$$

with

$$I=-2\sum_{ij}\mu^2(\omega_3)\frac{|K_{ij}|^2(1+n_i+n_j)}{y+\omega_i+\omega_j-i\epsilon}. \quad (17)$$

Let us suppose that the energy levels of the phonon reservoir are very closely spaced, the summation in (17) can be replaced by an integral:

$$\sum_{ij}\to\int_0^\infty\rho(\omega_i)\,d\omega_i\int_0^\infty\rho(\omega_j)\,d\omega_j,$$

where $\rho(\omega)$ is the density of energy states of the reservoir:

$$I = -2\mu^2 \int_{-\infty}^{+\infty} \rho(\omega_i)\, d\omega_i \int_{-\infty}^{+\infty} \rho(\omega_j) \times \frac{|K(\omega_i, \omega_j)|^2 (1 + \bar{n}(\omega_i) + \bar{n}(\omega_j))}{y + \omega_i + \omega_j - i\epsilon}\, d\omega_j \,. \tag{18}$$

Here we have explicitly taken into account the frequency dependence of $K_{ij}$, $\bar{n}_i$, $\bar{n}_j$. The limits of integration have been taken from $-\infty$ to $+\infty$ since the denominator is very weak if $y + \omega_i + \omega_j = 0$. Taking the new variables $\omega' = \omega_i + \omega_j$ and $\omega'' = \omega_i - \omega_j$, (18) becomes

$$I = -\mu^2 \int_{-\infty}^{+\infty} [d\omega'/(y + \omega' - i\epsilon)] A(\omega') \tag{19}$$

with

$$A(\omega') = \int_{-\infty}^{+\infty} \rho[\tfrac{1}{2}(\omega' - \omega'')]\rho[\tfrac{1}{2}(\omega' + \omega'')] \times |K(\omega', \omega'')|^2 \{1 + \bar{n}[\tfrac{1}{2}(\omega' - \omega'')] + \bar{n}[\tfrac{1}{2}(\omega' + \omega'')]\}\, d\omega''$$

It is well known that[26]

$$\lim_{\epsilon \to 0} (x - i\epsilon)^{-1} = \mathcal{P}\left(\frac{1}{x}\right) + i\pi\delta(x) \,,$$

$\mathcal{P}$ standing for principal value, then

$$I = -\mu^2 \mathcal{P} \{\int_{-\infty}^{+\infty} d\omega' [A(\omega')/(y + \omega')]\} - i\pi\mu^2 A(-y) \,. \tag{20}$$

As long as the coupling remains weak between the polariton field and the reservoir, the function to be integrated in (16) has only noticeable values when $y$ is near $-\omega_3$, which is the pole of (16) when $K_{ij} = 0$. So $y$ can be taken equal to $-\omega_3$ in (20):

$$I \simeq \Delta\omega - \tfrac{1}{2} i\gamma \tag{21}$$

with

$$\gamma = 2\pi\mu^2 A(\omega_3) \,,$$

$$\Delta\omega = -\mu^2 \mathcal{P} \{\int_{-\infty}^{+\infty} d\omega' [A(\omega')/(\omega' - \omega_3)]\} \,.$$

Going back to (16) and using (21), it is easy to obtain the functions $u(t)$, $v_{ij}(t)$, and $w(t)$.


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