Time-dependent Brillouin scattering of an intense polariton wave

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It is shown that the amplitude of the anti-Stokes line in a Brillouin-scattering spectrum can oscillate with time following the onset of an intense coherent polariton wave. The initial evolution of the correlated noise of scattered polaritons is investigated under conditions such that the intensity of the transmitted polariton wave greatly exceeds the stimulated-scattering threshold.

The analogy between the behavior of stimulated emission and scattering close to threshold, on the one hand, and second-order phase transitions, on the other, has been pointed out and discussed numerous times in the literature.[1–3] The similarity lies in the onset of a macroscopically coherent state superposed on intense fluctuation noise that increases as the threshold is approached. The interaction of the fluctuations with one another and with the coherent mode therefore plays a decisive role in both cases. A fundamental difference between the two is that whereas in phase transitions the fluctuations are in thermodynamic equilibrium, in stimulated emission they are excited by an external source. The statistical properties of the fluctuation ensemble in this second case are therefore, generally speaking, not universal but are determined by the properties of the excitation source and of its interaction with the medium. In particular, they can be described by some additional correlations, such as the correlation of the scattered polaritons and phonons in the specific problem discussed below. In addition, many stimulated processes (particularly scattering) are observed as a rule under essentially nonstationary conditions (pulsed excitation). Establishment of a stationary fluctuation pattern near threshold, on the other hand, is a long-time process. The question of the time-dependent onset of coherence is here much more significant than in the typical formulation of the second-order phase-transition problem. Furthermore, the time available for passage through the regions below and near threshold, is quite possibly too short for the noise level to reach values that can influence substantially the onset of a coherent mode. The evolution of the fluctuations can also be strongly influenced by the size of the samples, whose linear dimensions can be comparable with or even smaller than the photon mean free path.

Our present aim is to investigate certain phenomena of this kind, using as a very simple example the Brillouin scattering of polaritons in semiconductors. The stationary behavior of this system was investigated in Refs. 4 and 5, where the variation of the dispersion of the scattered polaritons under the influence of the exciting radiation and their spectral distribution was studied. We use here the same approach as in Refs. 4 and 5, based on introducing two types of Green's function—retarded and correlational.[6,7] As noted above, a characteristic feature of this system is the onset of the correlation of the scattered polaritons and the emitted phonons, and also of the scattered polaritons with one another. This additional coherence is manifested, in particular, in the intensity oscillations of anti-Stokes scattering following abrupt application of the pump wave. From the formal standpoint, this coherence is accounted for in natural fashion by the appearance of the so-called anomalous Green's functions, similar to the functions introduced by Belyaev[8,9] and by Gor'kov[9,10] in superfluidity and superconductivity theories. This also alters substantially the character of the equations that describe the spectral distribution of the scattered particles. The usual kinetic equation for an incoherent and strongly nonequilibrium many-particle system is replaced by an equation similar to the equation, well know in quantum radiophysics, for the density matrix of a two-level system in a resonant external field, with the off-diagonal matrix elements replaced by the anomalous Green's functions.

Especially noteworthy is also the physical meaning of the self-energy of these equations. In the so-called triangular representation, the off-diagonal elements (retarded and advanced) of the self-energy matrix are related, as in the equilibrium case,[9] to the polarizability of the medium and determine the propagation of the field in it. On the contrary, the diagonal element of the self-energy matrix plays the role of a correlator of the flucutuating field sources. In thermodynamic equilibrium, this term is uniquely related to the off-diagonal elements by the fluctuation-dissipation theorem, but under the conditions far from equilibrium considered here, it becomes a fully autonomous nontrival physical characteristic of the problem.

More specifically, we consider here the evolution of the Stokes and anti-Stokes components in the spectrum of scattered polaritons following an abrupt application to the crystal of an electric field of frequency ε_0 close to that of the polariton resonance. We use the equations obtained[5] for normal and anomalous Greens's functions by a diagram technique for nonequilibrium processes.[6,7] We confine ourselves here to the τ-approximation, i.e., assume that the normal polarization operators are independent of the external-field amplitude, and only terms linear in the field amplitude are included in the anomalous polariton-phonon polarization

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operator that takes into account the correlations between the scattered polaritons and phonons. The normal polarization operators, whose imaginary parts specify the widths of the corresponding levels, are integrals over a large range of frequencies and momenta, and vary little in fields that are not too strong, so long as the spectrum restructuring and the change of the occupation numbers are concentrated in regions of frequencies and momenta small compared with ε_0 and $\mathbf{p}_0$ (where $\mathbf{p}_0$ is the quaasimomentum of the polariton wave). An exception, as shown in Ref. 5, is the behavior of the system near the stimulated-scattering threshold. In our problem involving the onset of the field, however, even this case is not dangerous (meaning that the τ-approximation can be used), since the accumulation of weakly damped phonoritons (mixed polariton-phonon modes), which alters the polarization operators, is localized in a narrow spectral range (compared with the combined width of the polariton and phonon levels), and proceeds quite slowly.

We use a mixed momentum-time representation of the Green's functions. In the diagram technique for nonequilibrium processes, the Green's functions are 2×2 matrices. In the "triangular" representation, which we shall use for the most part, the off-diagonal components are retarded and advanced Green's functions, while the nonzero diagonal component is the statistical function. Our procedure is the following. We first solve the equations for the retarded and advanced functions. To obtain a unique solution these equations must be supplemented by boundary conditions. Since the retarded (advanced) functions are proportional to the mean values of the commutators of the corresponding fields, at equal times the normal and anomalous functions are equal to $+i$ and zero, respectively. To find the intensities of the Stokes and anti-Stokes components, we next calculate the statistical functions (proportional to the mean values of the anticommutators), using[6] equations of the type $F = FG^r \Omega G^a$ (Ω is the statistical component of the polarization operator). We shall also find it convenient to combine[4] the normal and anomalous Green's functions into 4×4 matrices whose retarded, advanced, and statistical components are 2×2 matrices similar to (4) (see below). These 4×4 matrices and their temporal 2×2 components will be designated by appropriate lower-case letters.

The equations for the advanced Green's functions take the form

$$\begin{bmatrix} i\dfrac{\partial}{\partial t}-\varepsilon_{pol}(\mathbf{p}) & -\Phi(\mathbf{p},t)\exp(-i\varepsilon_0 t) \\ -\Phi(\mathbf{p},t)\exp(i\varepsilon_0 t) & \pm\left(i\dfrac{\partial}{\partial t}-\varepsilon_{ph}(\mathbf{p})\right)\end{bmatrix}$$

$$\times g^a(t,t',\mathbf{p})=\delta(t-t')I, \tag{1}$$

$$g^a(t,t',\mathbf{p})\begin{bmatrix} -i\dfrac{\partial}{\partial t'}-\varepsilon_{pol}(\mathbf{p}) & -\Phi(\mathbf{p},t')\exp(-i\varepsilon_0 t') \\ -\Phi(\mathbf{p},t')\exp(i\varepsilon_0 t') & \pm\left(-i\dfrac{\partial}{\partial t'}-\varepsilon_{ph}(\mathbf{p})\right)\end{bmatrix}$$

$$=\delta(t-t')I. \tag{2}$$

Here

$$I=\begin{bmatrix}1&0\\0&1\end{bmatrix},\quad \varepsilon_{pol}(\mathbf{p})=\varepsilon+i\gamma_{pol},\quad \varepsilon_{ph}(\mathbf{p})=\pm u|\mathbf{p}-\mathbf{p}_0|+i\gamma_{ph},$$

is the polariton dispersion law, u the speed of sound in the semiconductor, $\gamma_{pol(ph)}$ the reciprocal polariton (phonon) lifetime,

$$\Phi(\mathbf{p},t)=\Phi_{\mathbf{p}}\theta(t)=D(|\mathbf{p}-\mathbf{p}_0|n_0/2\hbar\rho u)^{1/2}\theta(t), \tag{3}$$

D the deformation-potential constant for the polariton, ρ the semiconductor density, n_0 the spatial density of the coherent-mode polaritons,

$$\theta(t)=\begin{cases}0, & t<0\\ 1, & t>0\end{cases},$$

$$g^a(t,t',\mathbf{p})=\begin{bmatrix} G^a_{pol}(t,t',\mathbf{p}) & G^a_{\Diamond}(t,t',\mathbf{p},\mathbf{p}_0-\mathbf{p}) \\ G_\times{}^a(t,t',\mathbf{p}_0-\mathbf{p},\mathbf{p}) & G^r_{ph}(t',t,\mathbf{p}_0-\mathbf{p})\end{bmatrix}, \tag{4}$$

$G^a_{\rm pol}$ the advanced Green's function of the scattered polariton, $G^r_{\rm ph}$ the retarded one of the photon, and G^a_Δ and $G^a_\times$ the advanced phonon-polariton anomalous Green's functions. Here and elsewhere the superscripts and subscripts refer to the anti-Stokes and Stokes components, respectively. It is assumed that the electromagnetic field is applied at the instant $t=0$. The functions (4) satisfy the boundary conditions

$$g^a(t,t+0,\mathbf{p})=\begin{bmatrix} i & 0\\ 0 & \pm i\end{bmatrix}. \tag{5}$$

The Stokes and anti-Stokes components in (1), (2), and (5) differ in sign because the positive- and negative-frequency parts of the customarily employed Green's function are chosen for these respective components.

The solutions of Eqs. (1) and (2), satisfying conditions (5), take the following forms: For $t, t'<0$

$$g^a(t,t',\mathbf{p})$$
$$=i\begin{bmatrix}\exp\{i\varepsilon_{pol}(t'-t)\} & 0\\ 0 & \pm\exp\{i\varepsilon_{ph}(t'-t)\}\end{bmatrix}\theta(t'-t). \tag{6}$$

For $t<0<t'$

$$G^a_{pol}(t,t',\mathbf{p})=[A_1\exp(i\varepsilon_1 t')+A_2\exp(i\varepsilon_2 t')]\exp(-i\varepsilon_{pol}t),$$
$$G^a_\Diamond(t,t',\mathbf{p},\mathbf{p}_0-\mathbf{p})$$
$$=B\exp(-i\varepsilon_{pol}t)[\exp(i(\varepsilon_1-\varepsilon_0)t')-\exp(i(\varepsilon_2-\varepsilon_0)t')], \tag{7}$$
$$G_\times{}^a(t,t',\mathbf{p}_0-\mathbf{p},\mathbf{p})=B\exp(-i\varepsilon_{ph}t)[\exp(i\varepsilon_1 t')-\exp(i\varepsilon_2 t')],$$
$$G_{ph}{}^r(t',t,\mathbf{p}_0-\mathbf{p})$$
$$=\pm\exp(-i\varepsilon_{ph}t)[A_2\exp(i(\varepsilon_1-\varepsilon_0)t')+A_1\exp(i(\varepsilon_2-\varepsilon_0)t')].$$

Finally, for $t, t'>0$

$$G^a_{pol}(t,t',\mathbf{p})$$
$$=\theta(t'-t)[A_1\exp(i\varepsilon_1(t'-t))+A_2\exp(i\varepsilon_2(t'-t))],$$
$$G^a_\Diamond(t,t',\mathbf{p},\mathbf{p}_0-\mathbf{p})=\theta(t'-t)B\exp(-i\varepsilon_0 t')[\exp(i\varepsilon_1(t'-t))-\exp(i\varepsilon_2(t'-t))], \tag{8}$$
$$G_\times{}^a(t,t',\mathbf{p}_0-\mathbf{p},\mathbf{p})=\theta(t'-t)B\exp(i\varepsilon_0 t)[\exp(i\varepsilon_1(t'-t))-\exp(i\varepsilon_2(t'-t))],$$

$$G_{ph}^r(t', t, \mathbf{p}_0-\mathbf{p}) = \pm\theta(t'-t)[A_2 \exp(i(\varepsilon_1-\varepsilon_0)(t'-t)) + A_1 \exp(i(\varepsilon_2-\varepsilon_0)(t'-t))].$$

Here

$$A_{1,2} = i(\varepsilon_{2,1}-\varepsilon_{pol})/(\varepsilon_{2,1}-\varepsilon_{1,2}), \quad B = \pm i\Phi_p/(\varepsilon_1-\varepsilon_2), \qquad (9)$$

and the functions $\varepsilon_{1,2}(\mathbf{p})$ are the split "phonoriton" terms[4,5]:

$$\varepsilon_{1(2)}(\mathbf{p}) = \varepsilon_0 + {}^1/_2\{\varepsilon_p - \varepsilon_0 \pm u|\mathbf{p}-\mathbf{p}_0| + i\Gamma + (-)[(\varepsilon_p - \varepsilon_0 \mp u|\mathbf{p}-\mathbf{p}_0| + i\gamma)^2 \pm 4\Phi_p^2]^{1/2}\},$$
$$\Gamma = \gamma_{pol} + \gamma_{ph}, \quad \gamma = \gamma_{pol} - \gamma_{ph}. \qquad (10)$$

Since the scattering probability increases with momentum transfer [see (3)], the splitting of the phonoriton terms is greatest for backscattering into modes located near the intersection of the unperturbed polariton spectra and the absorbed and emitted phonons; the characteristic frequencies $\varepsilon_\pm$ and momenta $\mathbf{p}_\pm$ of the latter are determined from the conditions

$$\varepsilon_\pm = \varepsilon_{\mathbf{p}_\pm}, \quad \varepsilon_\pm \mp u|\mathbf{p}_\pm - \mathbf{p}_0| = \varepsilon_0, \quad \mathbf{p}_\pm \| \mathbf{p}_0. \qquad (11)$$

At $\mathbf{p} \sim \mathbf{p}_\pm$ the occupation numbers of the anti-Stokes and Stokes components of the scattered polaritons are maximal. To find them, we calculate first the statistical normal and anomalous Green's functions, using the equation[5]

$$f(t, t', \mathbf{p}) = \begin{bmatrix} F_{pol}(t, t', \mathbf{p}) & F_\diamond(t, t', \mathbf{p}, \mathbf{p}_0-\mathbf{p}) \\ F_\times(t, t', \mathbf{p}_0-\mathbf{p}, \mathbf{p}) & F_{ph}(t', t, \mathbf{p}_0-\mathbf{p}) \end{bmatrix}$$
$$= \iint_{-\infty}^{\max(t,t')} dt_1\, dt_2\, g^r(t, t_1, \mathbf{p})\, \omega(t_1, t_2, \mathbf{p})\, g^a(t_2, t', \mathbf{p}), \qquad (12)$$

where the statistical components of the polarization operators take in the τ-approximation the form

$$\omega(t_1, t_2, \mathbf{p}) = \delta(t_1-t_2)\begin{bmatrix} -2i\gamma_{pol}(1+2N_{0,pol}) & 0 \\ 0 & -2i\gamma_{ph}(1+2N_{0,ph}) \end{bmatrix}, \qquad (13)$$

where

$$N_{0,ph} = [\exp(u|\mathbf{p}-\mathbf{p}_0|/k_BT)-1]^{-1},$$

$$N_{0,pol} = [\exp(\varepsilon_p/k_BT)-1]^{-1}$$

are the bare "thermal" occupation numbers of the scattered polaritons and phonons, while the retarded Green's functions are Hermitian adjoints of the advanced ones: $g^r(t,t',\mathbf{p}) = [g^a(t',t,\mathbf{p})]^+$.

The equal-time statistical functions are connected with the occupation numbers by the relations (α = pol, ph)[6,7]

$$N_\alpha(\mathbf{p}, t) = [iF_\alpha(t, t, \mathbf{p}) - 1]/2. \qquad (14)$$

Simple calculations yield[1)]

$$N_{pol}(\mathbf{p}, t) = N_{0,pol} + \left[N_{0,ph} \mp N_{0,pol} + \frac{1\mp1}{2}\right]\frac{\Phi_p^2}{\omega_p^2 + (\gamma_1-\gamma_2)^2}$$
$$\times\left[(1-e^{-2\gamma_1 t})\frac{\gamma_{ph}-\gamma_1}{\gamma_1} + (1-e^{-2\gamma_2 t})\frac{\gamma_{ph}-\gamma_2}{\gamma_2} + \frac{2(\omega_p^2+\gamma\Gamma)}{\omega_p^2+\Gamma^2} - \frac{2(\omega_p^4+\omega_p^2(\Gamma^2+\gamma^2)+\gamma^2\Gamma^2)^{1/2}}{\omega_p^2+\Gamma^2} e^{-\Gamma t}\sin(\omega_p t + \psi_{pol})\right], \qquad (15)$$

where

$$\omega_p = \mathrm{Re}\,(\varepsilon_1-\varepsilon_2), \quad \gamma_{1,2} = \mathrm{Im}\,\varepsilon_{1,2},$$
$$\mathrm{tg}\,\psi_{pol} = (\omega_p^2+\gamma\Gamma)/\omega_p(\Gamma-\gamma).$$

To obtain the corresponding equation for $N_{ph}(\mathbf{p}_0-\mathbf{p}, t)$, we must interchange the subscripts pol and ph in (15) (and, in particular, replace γ by $-\gamma$).

We consider first the anti-Stokes scattering [the superscript in (10) and (15)]. Equation (15) describes the establishment of the stationary occupation numbers of the scattered polaritons. This process is relaxational in weak fields, i.e., at $\Phi_{\mathbf{p}_+} < \Gamma$ and oscillatory in strong fields $\Phi_\mathbf{p} > \Gamma$. The oscillation frequency equals the splitting of the phonoriton terms; oscillations set in when the splitting exceeds the sum of the line widths.

If the electromagnetic field is turned on for a time τ, it can be easily shown that

$$N_\alpha(t>\tau) = N_{0,\alpha}[1-\exp(-2\gamma_\alpha(t-\tau))] + N_\alpha(\tau)\exp(-2\gamma_\alpha(t-\tau)), \qquad (16)$$

(α = pol, ph). If, for example, $\tau \sim \omega_{\mathbf{p}_+} \ll \Gamma^{-1}$, the anti-Stokes component modulation depth is large and small changes of the duration of the passing pulse τ or of the intensity of the transmittted wave can give rise to considerable changes, from τ to Γ^{-1}, of the duration of the back-reflected anti-Stokes pulse.

For Stokes scattering, the splitting of the phonoriton terms in the central part of the line near $\mathbf{p} \sim \mathbf{p}_-$ is always small compared with Γ. No line-center oscillations are therefore produced. In sufficiently strong fields, at $\Phi_{\mathbf{p}_-} > (\gamma_{ph}\gamma_{pol})^{1/2}$, the sign of the damping γ_2 is reversed[5] in a certain momentum region near $\mathbf{p}_-$, and stimulated scattering sets in. It follows from (15) that in this case the number of scattered polaritons and of emitted phonons increases exponentially with time. If $\Phi_{\mathbf{p}_-} \gg (\gamma_{ph}\gamma_{pol})^{1/2}$ and there is no thermal source of polaritons ($N_{0,pol} = 0$), we have

$$N_{pol}(\mathbf{p}_-, t) \approx {}^1/_4(1+N_{0,ph})\exp(2\Phi_{\mathbf{p}_-}t). \qquad (17)$$

Note that $\omega_\mathbf{p} > \Gamma$ at the end points of the Stokes line and oscillations of the occupation numbers are possible.

In strong fields $\Phi_{\mathbf{p}_-} \gg (\gamma_{ph}\gamma_{pol})^{1/2}$ phonoriton backscattering causes a rapid buildup of the transmitted-phonoriton-wave fluctuations, consisting of correlated pairs of polaritons with momenta $\mathbf{p}_0 + \mathbf{k}$ and $\mathbf{p}_0 - \mathbf{k}$. The approach developed here permits a description of the initial stage of the onset of the noise, for times $t < \Gamma^-$ when the intensity of the scattered polariton wave is still low compared with that of the transmitted wave, and the fixed pump approximation is applicable. In second order of perturbation theory in the polariton-phonon intersection, we obtain[2] at $N_{0,\mathrm{pol}} = 0$ the following expressions for the density of the "noise" polaritons near the transmitted wave

$$N_{pol}(\mathbf{p}_0+\mathbf{k}, t) = \langle a^+(\mathbf{p}_0+\mathbf{k}, t)\, a(\mathbf{p}_0+\mathbf{k}, t)\rangle$$

and for the equal-time noise correlator $\langle a(\mathbf{p}_0-\mathbf{k}, t)\, a(\mathbf{p}_0+\mathbf{k}, t)\rangle$ (where $a(\mathbf{p}, t)$ is the polariton annihilation operator in the Heisenberg representation and $|\mathbf{k}| \ll |\mathbf{p}_0|$):

$$N_{pol}(\mathbf{p}_0+\mathbf{k},t)\approx \mu^2(1+N_{0,ph})^2\int\frac{d^3p}{(2\pi)^3}\exp[2\Gamma(\mathbf{p},\mathbf{k})t]$$
$$\times\{[\varepsilon(\mathbf{p}_0+\mathbf{k})-\varepsilon_0-\Omega(\mathbf{p},\mathbf{k})]^2$$
$$+[\Gamma(\mathbf{p},\mathbf{k})+\gamma_{pol}]^2\}^{-1}|B(\mathbf{p})B(\mathbf{p}-\mathbf{k})|^2$$
$$\times[\gamma_2(\mathbf{p}-\mathbf{k})-\gamma_{pol}][\gamma_2(\mathbf{p})-\gamma_{ph}][\gamma_2(\mathbf{p}-\mathbf{k})\gamma_2(\mathbf{p})]^{-1}, \quad (18)$$
$$\langle a(\mathbf{p}_0-\mathbf{k},t)a(\mathbf{p}_0+\mathbf{k},t)\rangle\approx\mu^2(1+N_{0,ph})^2\exp(-2i\varepsilon_0 t)$$
$$\times\int\frac{d^3p}{(2\pi)^3}\exp[2\Gamma(\mathbf{p},\mathbf{k})t]\{[\varepsilon(\mathbf{p}_0+\mathbf{k})-\varepsilon_0-\Omega(\mathbf{p},\mathbf{k})]^2$$
$$+[\Gamma(\mathbf{p},\mathbf{k})+\gamma_{pol}]^2\}^{-1}B^*(\mathbf{p})B^*(\mathbf{p}-\mathbf{k})A_1(\mathbf{p})A_1(\mathbf{p}-\mathbf{k})$$
$$\times[\gamma_2(\mathbf{p})-\gamma_{ph}]$$
$$\times[\gamma_2(\mathbf{p}-\mathbf{k})-\gamma_{ph}][\gamma_2(\mathbf{p})\gamma_2(\mathbf{p}-\mathbf{k})]^{-1}, \quad (19)$$

where

$$\mu^2={}^1/_4D^2|\mathbf{p}_--\mathbf{p}_0|/\hbar\rho u,$$
$$\Omega(\mathbf{p},\mathbf{k})=\mathrm{Re}\,[\varepsilon_2(\mathbf{p})-\varepsilon_2^*(\mathbf{p}-\mathbf{k})],$$
$$\Gamma(\mathbf{p},\mathbf{k})=-\mathrm{Im}\,[\varepsilon_2(\mathbf{p})-\varepsilon_2^*(\mathbf{p}-\mathbf{k})]. \quad (20)$$

Using (9) and (10) we obtain

$$\langle a(\mathbf{p}_0-\mathbf{k},t)a(\mathbf{p}_0+\mathbf{k},t)\rangle\approx\exp(-2i\varepsilon_0 t)N_{pol}(\mathbf{p}_0+\mathbf{k},t) \quad (21)$$

and

$$N_{pol}(\mathbf{p}_0,t)\approx(1+N_{0,ph})^2\exp(4\Phi_{\mathbf{p}_-}t)\Gamma/\Phi_{\mathbf{p}_-}. \quad (22)$$

We emphasize that Eqs. (18)–(22) are valid in strong fields, when the transmitted-wave intensity exceeds substantially the stimulated-scattering threshold, and during the initial stage of the process, at times t satisfying the condition $\Phi_{\mathbf{p}_-}^{-1}\lesssim t\ll\Gamma^{-1}$.

We note in conclusion that the question of the effects observable outside the crystal calls for additional consideration of the coefficient of polariton passage through the crystal boundary, which is also changed by the change of the polariton spectrum.

APPENDIX

By calculating (12) with the aid of (13) and (6)–(8) we obtain, e.g., at $t>t'$

$$F_\alpha(t,t',\mathbf{p})=\tilde F_\alpha(t,t',\mathbf{p})+(1+2N_{0,\alpha})G_\alpha^r(t,t',\mathbf{p}),$$
$$F_\Diamond(t,t',\mathbf{p},\mathbf{p}_0-\mathbf{p})$$
$$=\tilde F_\Diamond(t,t',\mathbf{p})\pm(1+2N_{0,ph})G_\Diamond^r(t,t',\mathbf{p},\mathbf{p}_0-\mathbf{p}), \quad (A.1)$$
$$F_\times(t,t',\mathbf{p}_0-\mathbf{p},\mathbf{p})$$
$$=\tilde F_\times(t,t',\mathbf{p})-(1+2N_{0,pol})G_\times^r(t,t',\mathbf{p}_0-\mathbf{p},\mathbf{p}),$$

where $\alpha=$ pol, ph and $\tilde f=0$ if $0>t>t'$ and $t>0>t'$. If $t>t'>0$ we have

$$\tilde F_{pol}(t,t',\mathbf{p})=-i|B|^2(E_{11}+E_{22}-E_{12}-E_{21}), \quad (A.2)$$
$$\tilde F_\Diamond(t,t',\mathbf{p})$$
$$=\mp i\exp(-i\varepsilon_0 t')B^*(A_2E_{11}-A_1E_{22}+A_1E_{12}-A_2E_{21}), \quad (A.3)$$
$$\tilde F_\times(t,t',\mathbf{p})$$
$$=\pm i\exp(i\varepsilon_0 t)B(A_2^*E_{11}-A_1^*E_{22}-A_2^*E_{12}+A_1^*E_{21}), \quad (A.4)$$

where

$$E_{ij}=(1\mp1+2N_{0,ph}\mp2N_{0,pol})\exp(-i\varepsilon_i^*t+i\varepsilon_jt')$$
$$\times\{1-\exp[i(\varepsilon_i^*-\varepsilon_j)t']\}[i(\varepsilon_i^*-\varepsilon_j)-2\gamma_{ph}]/i(\varepsilon_i^*-\varepsilon_j), \quad (A.5)$$

$\varepsilon_i=\varepsilon_i(\mathbf{p})$ [see (10)], and $i=1,2$. To obtain the function $\tilde F_{ph}(t,t',\mathbf{p})$, we must interchange the subscripts pol and ph in (A.2) and (A.5)

The correlation properties of the noise near the transmitted wave are described by the corresponding components of the normal and anomalous Green's functions of the direct polaritons[5]

$$D(\mathbf{p}_0+\mathbf{k},t,t'),\quad D_\Diamond(\mathbf{p}_0+\mathbf{k},\mathbf{p}_0-\mathbf{k},t,t').$$

It is convenient to continue by transforming from the "triangular" to the "$\pm$" representation of the Green's functions, in which[6,7]

$$G^{+-}={}^1/_2(F-G^r+G^a),\quad G^{-+}={}^1/_2(F+G^r-G^a),$$
$$G^{++}=\theta(t-t')G^{-+}+\theta(t'-t)G^{+-},$$
$$G^{--}=\theta(t'-t)G^{-+}+\theta(t-t')G^{+-}. \quad (A.6)$$

In the "$\pm$" representation the functions D and $D_\Diamond$ are related to the polariton correlation functions by

$$D^{+-}=(\mathbf{p}_0+\mathbf{k},t,t')=-i\langle a^+(\mathbf{p}_0+\mathbf{k},t')a(\mathbf{p}_0+\mathbf{k},t)\rangle,$$
$$D_\Diamond{}^{+-}(\mathbf{p}_0+\mathbf{k},\mathbf{p}_0-\mathbf{k},t,t')=-i\langle a(\mathbf{p}_0-\mathbf{k},t')a(\mathbf{p}_0+\mathbf{k},t)\rangle. \quad (A.7)$$

In the τ approximation the backscattering of the phonoritons into modes close to the transmitted wave is disregarded, and in the absence of a thermal polariton source ($N_{0,\mathrm{pol}}=0$) we have[5]

$$D_0^{+-}=D_{0,\Diamond}=0,\quad D_0^{-+}(\mathbf{p},t>t')=-i\exp[i\varepsilon_{pol}^*(\mathbf{p})\cdot(t'-t)]. \quad (A.8)$$

In second order of perturbation theory in the polariton-phonon interaction, taking diagrams a and b of Fig. 1 into account, we obtain for the equal-time functions the following expressions

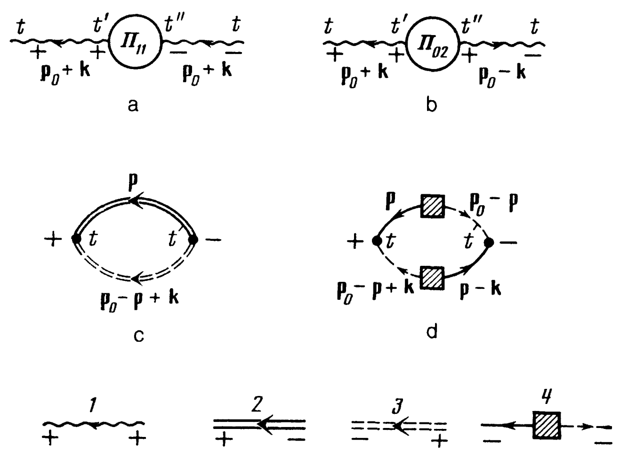

FIG. 1. Diagrams of second-order perturbation theory in the polariton-phonon interaction for the Green's function $D^{+-}(\mathbf{p}_0+\mathbf{k},t,t)$ (a), $D_\Diamond^{+-}(\mathbf{p}_0+\mathbf{k},\mathbf{p}_0-\mathbf{k},t,t)$ (b), and for the polarization operators $\Pi_{11}^{+-}(\mathbf{p}_0+\mathbf{k},t,t')$ (c), $\Pi_{02}^{+-}(\mathbf{p}_0+\mathbf{k},\mathbf{p}_0-\mathbf{k},t,t')$ (d). Notation: $1-D_0^{++}$, $2-G_{pol}^{+-}$, $3-G_{ph}^{-+}$, $4-G_\Diamond^{--}$

$$D^{+-}(\mathbf{p}_0+\mathbf{k},t,t)=\iint\limits_{-\infty}^{t} dt'\,dt''D_0^{-+}(\mathbf{p}_0+\mathbf{k},t,t')$$

$$\times\ \Pi_{11}^{+-}(\mathbf{p}_0+\mathbf{k},t',t'')D_0^{-+}(\mathbf{p}_0+\mathbf{k},t'',t), \qquad (A.9)$$

$$D_\diamond^{+-}(\mathbf{p}_0+\mathbf{k},\mathbf{p}_0-\mathbf{k},t,t)=\iint\limits_{-\infty}^{t} dt'\,dt''D_0^{-+}(\mathbf{p}_0+\mathbf{k},t,t')$$

$$\times\ \Pi_{02}^{++}(\mathbf{p}_0+\mathbf{k},\mathbf{p}_0-\mathbf{k},t',t'')D_0^{-+}(\mathbf{p}_0-\mathbf{k},t,t''), \qquad (A.10)$$

where the normal and anomalous polarization operators (diagrams c and d of Fig. 1) are equal to

$$\Pi_{11}{}^{+-}(\mathbf{p}_0+\mathbf{k},t,t')$$

$$=-2i\mu^2\int\frac{d^3p}{(2\pi)^3}G_{pol}(\mathbf{p},t,t')G_{ph}{}^{+-}(\mathbf{p}_0-\mathbf{p}+\mathbf{k},t,t'), \qquad (A.11)$$

$$\Pi_{02}^{+-}(\mathbf{p}_0+\mathbf{k},\mathbf{p}_0-\mathbf{k},t,t')=-2i\mu^2\int\frac{d^3p}{(2\pi)^3}$$

$$\times G_\diamond^{+-}(\mathbf{p},\mathbf{p}_0-\mathbf{p},t,t')G_\diamond^{-+}(\mathbf{p}-\mathbf{k},\mathbf{p}_0+\mathbf{k}-\mathbf{p},t',t), \qquad (A.12)$$

$$\Pi^{++}(t,t')=-\theta(t-t')\Pi^{-+}(t,t')-\theta(t'-t)\Pi^{+-}(t',t). \qquad (A.13)$$

In the derivation of (A.9) and (A.10) we used the fact that in the "$\pm$" representation the Green's functions are independent of the time index at the maximum time. Calculating (A.9) and (A.10) with the aid of (A.1)–(A.5) for Stokes scattering far above the stimulated-scattering threshold, we obtain Eqs. (18) and (19).

1) The final expressions for the statistical Green's functions (12) are given in the Appendix [Eqs. (A.1)–(A.5)].

2) The calculations are given in the Appendix [Eqs. (A.6)–(A.13)]

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