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PHASE FLUCTUATIONS AND LINEWIDTHS IN A SOLID STATE ANTI-STOKES RAMAN OSCILLATOR

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Introduction

Assuming the fundamental processes to be polariton scatterings, in some papers [1,2] we have been dealing with special problems of the stimulated Raman effect in crystals and have presented appropriate theoretical results. Especially in [2] on the basis of an easy model [1,3] we have investigated the stationary behaviour of an anti-Stokes Raman oscillator. So we could calculate in detail the threshold value, the frequency shifts, and the steady state photon (polariton) numbers of the several modes in dependence on detuning, damping, and coupling. Furthermore, from these calculations predictions with regard to useful ratios κ_1/κ_{-1} for experiments are possible (κ_{-1}, κ_1 - damping parameters of Stokes and anti-Stokes modes). As it was mentioned in [2] such an anti-Stokes Raman oscillator is very suitable for realizing lasers with more than one tunable frequency.

In this paper we will complete our results with some investigations on the behaviour of the linewidths of the Stokes mode and anti-Stokes mode. Thereby we will be interested in the detailed dependence of the linewidths on detuning, damping, and pumping.

It is known [3,4] that in such a laser-like oscillator the phase fluctuations of the incident laser flux as well as the spontaneous noise of all modes involved in the scattering processes contribute to the linewidths considered here. On the other hand, in real situations the influence of the phase fluctuations of the incident laser pump wave will be dominant in comparison with that of the spontaneous one[3,5]. In this sense we will neglect all the spontaneous noise contributions to the linewidths.

It sould be emphasized that the results found in this way are also valid for all other laser-like oscillators that can be described by a nonlinear quasi-resonant interaction (of the lowest order) of four light waves.

Theory

Let us begin with some theoretical aspects that are important for the following. The behaviour of an anti-Stokes Raman oscillator can be described mathematically by a set of four nonlinear coupled differential equations of first order /1/ for the polariton creation (respectively, annihilation) operators $B^+, B^+_{-1}, B^+_0, B^+_1$ referring to infrared, Stokes, laser, and anti-Stokes modes in the given order. These equations are supposed to be quantum mechanical Langevin equations (therefore they contain dissipation and fluctuation terms in a quantum mechanically consistent manner). Furthermore, the pump term in the equation for the laser mode is assumed to be $\hat{F}^*_0 \exp\{i(\omega_0 t + \psi_p(t))\}$ with the appropriate correlation function

$$\langle F^+_0(t_1) F_0(t_2) \rangle = |\hat{F}_0|^2 \exp\{-\tfrac{1}{2}\Delta_P|t_1 - t_2| + i\omega_0(t_1 - t_2)\} \qquad (1)$$

(Δ_P is the linewidth of the incident pump wave).

For solving this set of equations above threshold Haken's ansatz is shown to be useful /3,5/. Such an ansatz means with respect to the operator B^+_{-1}

$$B^+_{-1}(t) = (r_{-1} + a_{-1}(t)) \exp\{i(\Omega_{-1} t + \phi_{-1} + \psi_{-1}(t))\} \qquad (2)$$

(for the other modes the situation is quite analogous). In Eq. 2 r_{-1} is the squareroot of the steady state photon number N_{-1} (calculated in /2/): $r_{-1} = (N_{-1})^{1/2}$. Ω_{-1} (also calculated in /2/) and ϕ_{-1} are frequency and phase of the Stokes mode under stationary conditions. $a_{-1}(t)$ and $\psi_{-1}(t)$ mean the amplitude and phase deviations from their steady state values. From /1/ the stationary phases are easily seen to be

$$\phi_0 - \phi_{-1} - \phi = \delta_1 - \arctan \frac{2\Delta\omega_{-1}}{\kappa_{-1}}, \qquad (3)$$

$$\phi_1 - \phi_0 - \phi = \pi + \delta_2 + \arctan \frac{2\Delta\omega_1}{\kappa_1}, \qquad (4)$$

$$2|\hat{F}_0| \cos(\phi_P - \phi_0) = \kappa_1 \frac{r_1^2}{r_0} + \kappa_{-1} \frac{r_{-1}^2}{r_0} + \kappa_0 r_0 \qquad (5)$$

(with the notation of the indices as mentioned above; $T_1 = -i|T_1|\exp(-i\delta_1)$ - coupling constant between laser, Stokes, and infrared modes, $T_2 = -i|T_2|\exp(-i\delta_2)$ - coupling constant between anti-Stokes, laser, and infrared modes; $\Delta\omega_{-1} = \omega_{-1} - \Omega_{-1}$, $\Delta\omega_1 = \omega_1 - \Omega_1$ where ω_{-1}, ω_1 are the unperturbed frequencies).

By quantum mechanical quasi-linearization (we are only interested in a small deviation from the steady state behaviour) the ansatz (2) finally leaves us with a set of coupled linear differential equations for the amplitude and phase deviations (that are assumed to be small) only. After calculating the solution that can be obtained by standard methods we get the mean squares $\langle\psi^2_{-1}(t)\rangle$, $\langle\psi^2_1(t)\rangle$ in an easy manner. From these mean squares the linewidths considered here are defined by those contributions that are proportional to time t /3,5/. These contributions (to the above mean squares) divided by t directly lead to the linewidths of Stokes and anti-Stokes modes.

Results

In /2/ (for the most important case $|T_1|^2 = |T_2|^2$ we will restrict ourselves to) the region $0.4 \leq \kappa_{-1}/\kappa_1 \leq 0.9$ was shown to be useful with respect to experiments. Therefore we will only present results for this region (in accordance

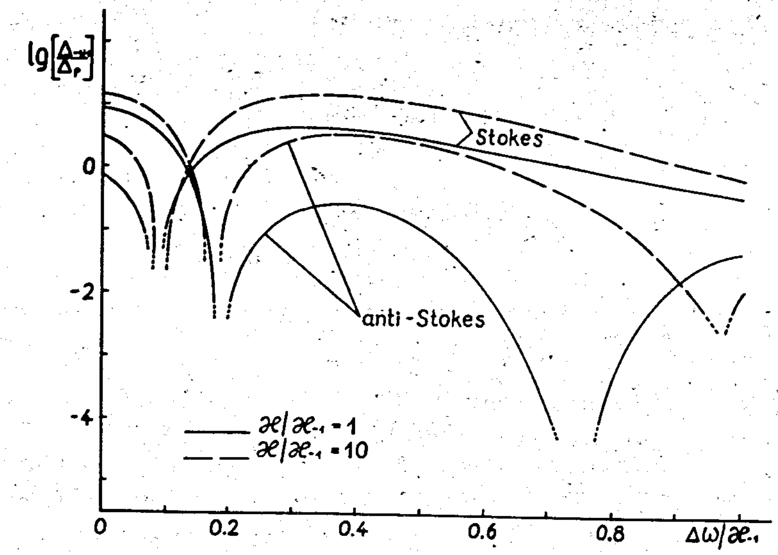

Fig. 1. The Stokes and anti-Stokes linewidths (relative to the linewidth of the pump wave) are shown for the two infrared damping parameters $\kappa = \kappa_{-1}$ and $\kappa = 10\kappa_{-1}$ ($P = 1.1$, $\rho = 1.0$, $\beta^{-1} = 0.7$) as functions of $\Delta\omega/\kappa_{-1}$

with the notation introduced in /2/ we will again use the abbreviation $\beta^{-1} = \kappa_{-1}/\kappa_{1}$).

For a first orientation with regard to the general properties of the linewidths it is sufficient to look at Fig. 1. There the behaviour of Stokes and anti-Stokes linewidths (relative to the linewidths Δ_P of the incident pump wave) is shown for a middle value $\beta^{-1} = 0.7$ and the two assumed ''boundary'' dampings of the infrared mode, $\kappa = \kappa_{-1}$ and $\kappa = 10\kappa_{-1}$, as a function of $\Delta\omega/\kappa_{-1}$ ($\omega_1 - \omega_0 - \omega = \Delta\omega$ is the detuning). The other parameters that enter in the theory are assumed to be: $P = 1.1$ (the pump parameter P is defined by $P = (4|\hat{F}_0|^2/\kappa_0^2)/N_0$; above threshold it must be greater than 1) and $\rho = \kappa_0/\kappa_{-1} = 1$.

First of all, a relative deep and narrow minimum in the region of small detuning $\Delta\omega/\kappa_{-1}$ is seen to be characteristic of the linewidth of the Stokes mode. In this connection we note that all minima appearing in our curves

are indicated by short dotted lines because the density of $\Delta\omega/\kappa_{-1}$ points chosen in the numerical calculations did not allow an exact localization of the minima. On the other hand, a more exact calculation does not pay for it is clear that in the case of such deep minima (for example, the relative decrease of the Stokes linewidth may be of the order $10^5 - 10^6$) the spontaneous noise (of the phases) of all modes will contribute to the linewidths essentially. This means, in practice, the minima will be expected to be not so deep as it is shown in Fig. 1, for example, and their values will be limited by the spontaneous noise. For our investigations it seems only remarkable that the Stokes linewidth (for specially chosen β^{-1}) can be very small in the region $0 < \Delta\omega/\kappa_{-1} \leq 2.0$.

Secondly, with respect to the linewidth of the anti-Stokes mode two minima were found. From Fig. 1 the

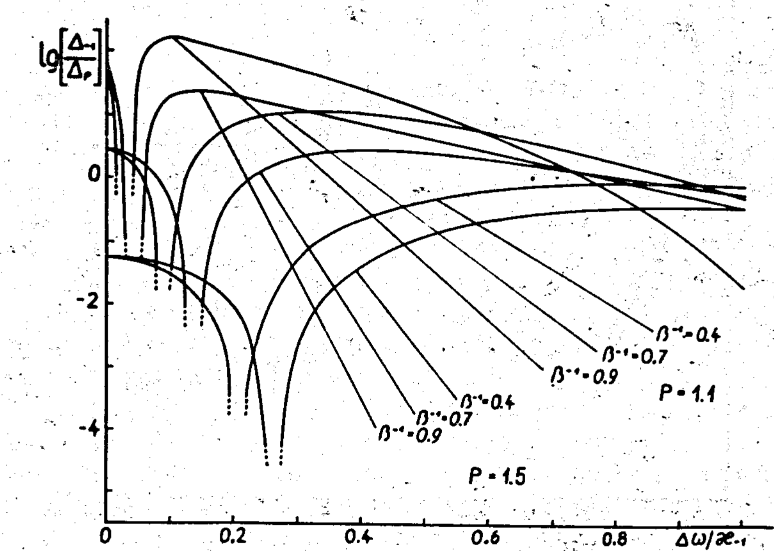

Fig. 2. The Stoke linewidth (relative to the linewidth of the pump wave) is shown for the two pump parameters $P = 1.1$ and $P = 1.5$ and for various ratios $\beta^{-1} = \kappa_{-1}/\kappa_{1}$ ($\rho = 1.0$, $\kappa = 6\kappa_{-1}$) as a function of $\Delta\omega/\kappa_{-1}$

second minimum at the end of the $\Delta\omega/\kappa_{-1}$ axis is expected to be not so a narrow one. Further, it is seen that the anti-Stokes linewidth (in dependence on $\Delta\omega/\kappa_{-1}$) can be greater as well as smaller than the linewidth of the Stokes mode. In a first approximation one could say that the anti-Stokes linewidth will be greater than the Stokes linewidth only in the region of small $\Delta\omega/\kappa_{-1}$ values with the mean position of the minima (this relates to the first minimum of the anti-Stokes mode) of both modes as the upper limit.

Last not least the results represented in Fig. 1 show an increase of the linewidths with the infrared damping parameter κ/κ_{-1}.

The linewidths of the Stokes and anti-Stokes modes estimated in /1/ are noted to be not correct.

In Figs. 2-5 the dependence of the linewidths on other physical quantities entering in the theory is explained. The

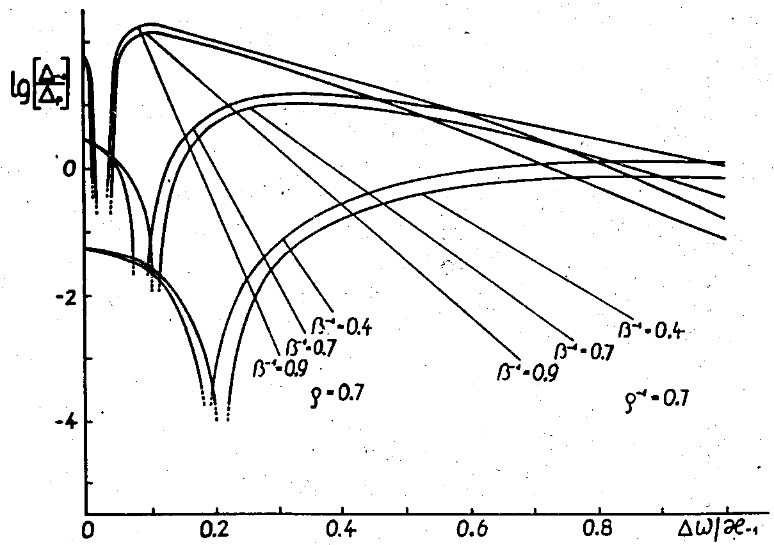

Fig. 3. The Stokes linewidth (relative to the linewidth of the pump wave) is shown for the two values $_{-1}\kappa_0/\kappa_{-1}=0.7$ and $\kappa_{-1}/\kappa_0=0.7$ and for various ratios $\beta^{-1}=\kappa_{-1}/\kappa_1$ (P=1.1, $\kappa=6\kappa_{-1}$) as a function of $\Delta\omega/\kappa_{-1}$.

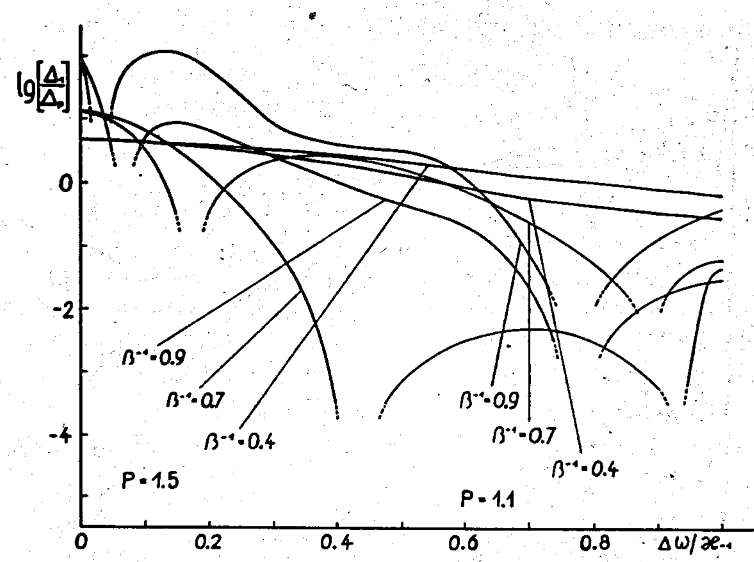

Fig. 4. The anti-Stokes linewidth (relative to the linewidth of the pump wave) is shown for the two pump parameters P=1.1 and P=1.5 and for various ratios $\beta^{-1}=\kappa_{-1}/\kappa_1$ (ρ=1.0, $\kappa=6\kappa_{-1}$) as a function of $\Delta\omega/\kappa_{-1}$.

results refer to a middle infrared damping parameter $\kappa=6\kappa_{-1}$.

So in Fig. 2 for two pump parameters P the dependence of the Stokes linewidth on the parameter β^{-1} is shown (ρ is assumed to be 1). From the plotted curves the position of the minimum is found to shift up to larger $\Delta\omega/\kappa_{-1}$ values if the parameter β^{-1} decreases. Simultaneously the relative decrease of the linewidth becomes smaller. It does not exist in the case of very small β^{-1} ($\beta^{-1}\lesssim 0.1$). Now let us look at curves with equal β^{-1} but different P. Roughly speaking, above the mean position of the minima (with respect to different P values) the Stokes linewidth decreases with increasing P. Below this point it increases with P. It is to be noted that in the limit $\Delta\omega/\kappa_{-1}\to 0$ the linewidth does not depend on the pump parameter.

By investigating the dependence of the Stokes linewidth on the damping parameter ρ (appropriate curves are plot-

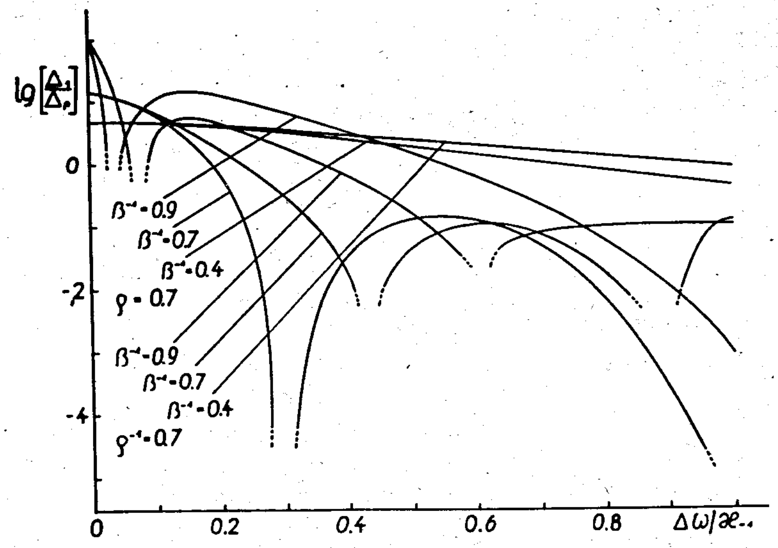

Fig. 5. The anti-Stokes linewidth (relative to the linewidth of the pump wave) is shown for the two values $\kappa_0/\kappa_{-1} = 0.7$ and $\kappa_{-1}/\kappa_0 = 0.7$ and for various ratios $\beta^{-1} = \kappa_{-1}/\kappa_1$ ($P = 1.5$, $\kappa = 6\kappa_{-1}$) as a function of $\Delta\omega/\kappa_{-1}$.

ted in Fig. 3 for $P = 1.1$) the behaviour was found to be completely similar to that illustrated in Fig. 2 (with ρ instead of P).

With regard to the anti-Stokes mode the situation is similar but more complicated because of the existence of two minima. This is shown in Figs. 4-5. Contrary to the results presented in Figs. 2-3 for $\beta^{-1} \lesssim 0.4$ minima do not appear and in the limit $\Delta\omega/\kappa_{-1} \to 0$ the linewidth is always found to be decreasing with β^{-1} (below $\beta^{-1} \approx 0.2$ the Stokes linewidth slowly increases with decreasing β^{-1}).

As it was mentioned above all results presented in Figs. 1-5 refer to the region $0.9 \gtrsim \beta^{-1} \gtrsim 0.4$. Generally the anti-Stokes oscillator was found to be stable for any value $\beta^{-1} < 1$ ($\kappa_{-1} < \kappa_1$). In the other case ($\kappa_{-1} \gtrsim \kappa_1$) the oscillator is stable only for $\Delta\omega/\kappa_{-1} \gtrsim 0.05$. This fact

could be connected with the necessity to include higher order interactions into the calculations (cf. [2]).

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