Effect of Idler Attenuation on the Spontaneous Parametric Scattering of Intense Light in $LiNbO_3$ and $NH_4H_2PO_4$†

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The forward-scattered intensity in the spontaneous parametric process is found to decrease significantly as the idler frequency goes sufficiently far into the 5.7-μ and 1.4-μ infrared absorption bands of $LiNbO_3$ and $NH_4H_2PO_4$, respectively. These results indicate that, unlike the previously studied cases in GaP and $LiNbO_3$, the corresponding optical nonlinear susceptibility has essentially no resonance increase and its ionic contribution remains small near these absorption bands. A formula for the forward-scattered intensity is obtained which agrees very well with the observed effects of idler attenuation on the spontaneous parametric scattering in both $LiNbO_3$ and $NH_4H_2PO_4$.

THERE has recently been considerable interest in Raman scattering by polaritons[1–4] and the spontaneous parametric scattering of intense light[5] in nonlinear optical crystals. These two processes can, however, become closely related. When the frequency of the idler wave in the parametric process approaches any lattice resonance (*Reststrahl*) mode that is both infrared and Raman active, the idler wave is of mixed photon and phonon character and becomes the polariton. In this case, the two processes become indistinguishable and the intensity of the scattered light is determined by, among other things, the nonlinear optical susceptibility $X^{(2)}$ that could have both electronic and ionic contributions. Far above the lattice resonance, the electronic contribution dominates over the ionic contribution. In previously studied cases, notably in gallium phosphide[1] and near the 634-cm^{-1} *Reststrahl* band of lithium niobate,[2] $|X^{(2)}|^2$ was generally found to have a strong resonance because of the ionic contribution near the lattice resonance where the intensity attenuation constant α_i of the polariton, or the idler wave, is also resonantly large. One of the implications of this is that the gain in the stimulated parametric process, which is proportional to $|X^{(2)}|^2/\alpha_i$, will remain unaltered as the idler frequency is swept through its absorption region.[1,4] As will be shown, this also means that the scattered intensity in the spontaneous parametric process would remain essentially unaffected as the idler frequency is swept through its absorption region.

On the other hand, if the lattice resonance is only weakly Raman active, even though the idler wave is still of mixed photon and phonon character and becomes the polariton near this lattice resonance, the ionic contribution to the nonlinear susceptibility $X^{(2)}$ will remain small as the idler frequency approaches this resonance region. The process can be considered as parametric scattering by polaritons. In this case, there will be no resonance increase in $|X^{(2)}|^2$ to compensate for any increase in α_i as the idler frequency is swept through this particular absorption region.

We have measured the integrated intensity of the forward-scattered light in the spontaneous parametric process as the idler frequency approaches the 5.7-μ and 1.4-μ absorption bands of $LiNbO_3$ and $NH_4H_2PO_4$ (ADP), respectively. These absorption bands are of particular importance since they limit the transparency windows of these crystals extending from the visible to the near infrared. The measured signal intensity was found to decrease significantly as the idler absorption increased and became proportional to α_i^{-1}, when α_i was sufficiently large. These results are consistent with the results on $LiNbO_3$ reported recently by Klyshko[6] and with our earlier speculations[7] on ADP. The decrease in the spontaneous parametric intensity here shows that there is no resonance in the nonlinear suceptibility $|X^{(2)}|^2$ near these absorption bands, which is presumably due to the fact that the ionic contribution to $X^{(2)}$ is negligible in these particular cases. The absence of any resonance increase in $|X^{(2)}|^2$ to compensate for the increase in idler attenuation in these frequency ranges is also of considerable importance since it would limit the tuning range over which optical parametric oscillation can be achieved using these crystals.

The primary light sources used to study the absorption regions of $LiNbO_3$ and ADP were, respectively, the 5145 and 4880 Å Ar^+ laser lines. The incident beam was polarized as an extraordinary wave, while the signal and idler were ordinary waves. In the case of

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[1] W. L. Faust and C. H. Henry, Phys. Rev. Letters **17**, 1265 (1966).

[2] S. K. Kurtz and J. A. Giordmaine, Phys. Rev. Letters **22**, 192 (1969).

[3] H. E. Puthoff, R. H. Pantell, B. G. Huth, and M. A. Chacon, J. Appl. Phys. **39**, 2144 (1968).

[4] C. H. Henry and C. G. Garrett, Phys. Rev. **171**, 1058 (1968).

[5] See the extensive references in, for example, D. Magde and H. Mahr, Phys. Rev. **171**, 393 (1968); R. L. Beyer and S. E. Harris, *ibid.* (to be published); J. P. Budin, B. Godard, and J. Ducing, IEEE J. Quant. Electron **QE-4**, 831 (1968); D. Weinberg, Appl. Phys. Letters (to be published); D. N. Klyshko, Zh. Eksperim. i Teor. Fiz. **55**, 1006 (1968) [English transl.: Soviet Phys.—JETP **28**, 522 (1969)].

[6] D. Klyshko, in Fourth USSR Nonlinear Optics Symposium, Kiev, 1968 (unpublished).

[7] T. G. Giallorenzi and C. L. Tang, Appl. Phys. Letters **12**, 376 (1968).

$LiNbO_3$, phase matching in the desired wavelength range for the collinear parametric process perpendicular to the optical axis of the crystal was achieved through temperature tuning the crystal from approximately 150 to over 500°C. In the case of ADP at room temperature, phase matching and tuning was achieved through crystal rotation. A standard phase-sensitive detection system, together with polarizing prisms, cutoff filters, and a $\frac{3}{4}$-m Jarrell-Ash monochromator was used to select and photoelectrically measure the parametrically scattered signal intensity in the forward direction. Other details of the experimental setup, with the exception of the oven used, are described in Ref. 7.

Figure 1 gives the signal wavelength and corresponding intensity obtained at different temperatures for a 1-cm-long $LiNbO_3$. The crystal experimental tuning curve is included in the figure for reference. The solid curve corresponds to the calculated intensity. The collinear integrated signal intensity for a given detector solid angle and for 90° phase matching is given by the following formula[8,9]:

$$P_s=\frac{(\Delta\epsilon_1)^2\hbar\omega_s^4\omega_i L P_p \Delta\Omega_{\rm det}}{2\pi C^4 n_s n_i n_p |n_s-n_i|}, \qquad (1)$$

which was derived for the case of no absorption at either the signal or the idler wavelengths. In the absence of idler absorption, which starts at about 4.2 μ for the idler, the agreement between the experimental and theoretical results is very good. However, beyond 5900 Å, the measured signal intensity decreased drastically. In this region, the estimated idler absorption length $2/\alpha_i$ becomes shorter than the crystal length L. To account for the effect of idler absorption on the signal intensity in the spontaneous parametric process, the result of Eq. (1) must be further multiplied by a reduction factor[10]

$$R=(2/\pi)\tan^{-1}(2\sigma/\alpha_i), \qquad (2)$$

where σ is the width of the distribution of the mismatch of the wave vectors for the pump, idler, and signal waves, or $|\mathbf{k}_p-\mathbf{k}_i-\mathbf{k}_s|$, in the absence of any absorption. The value of σ for the case of 90° pumping, as in the case of $LiNbO_3$, is primarily determined by the crystal length and is equal to π/L; therefore, the reduction factor needed here is

$$R(90°\text{ pumping})=(2/\pi)\tan^{-1}(2\pi/\alpha_i L), \qquad (2')$$

which in the limit of large $\alpha_i L$ reduces to $4/\alpha_i L$. In the limit of no absorption, or $\alpha_i=0$, R always reduces to 1. The dashed curve in Fig. 1 corresponds to the solid curve multiplied by the reduction factor R given in Eq. (2'), assuming no dispersion in the nonlinear susceptibility. The very good agreement, between the experimental points and the dashed curve, shows that there is no resonance increase in the nonlinear susceptibility $|X^{(2)}|^2$ or its ionic contribution is negligible in the frequency range we studied, where the idler absorption is already quite large. This, of course, does not mean that as the idler frequency goes further into these absorption bands or at other *Reststrahl* bands of the crystal, the nonlinear susceptibility will not show any increase.

In terms of the notation of Ref. 1, the nonlinear susceptibility near a *Reststrahl* band is of the form

$$X^{(2)}(\omega_s=\omega_p-\omega_i)\sim d_E[1+C\omega_0^2/(\omega_0^2-\omega_i^2-i\omega_i\Gamma)], \qquad (3)$$

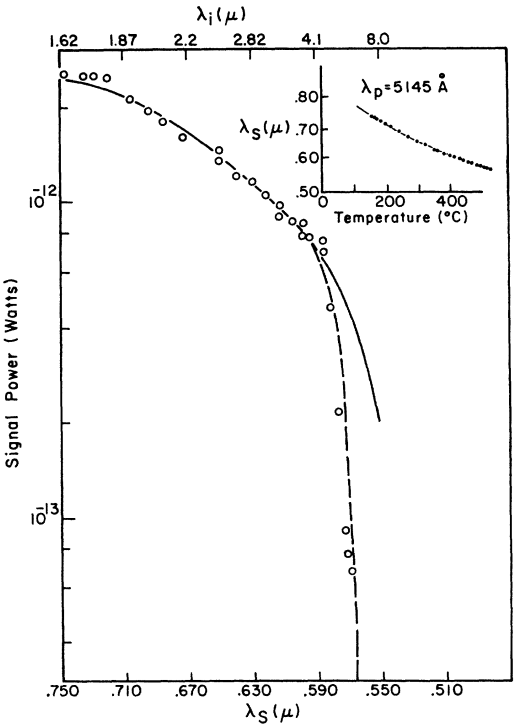

FIG. 1. Forward-scattered intensity in the spontaneous parametric scattering of light in $LiNbO_3$. The solid curve corresponds to the calculated intensity neglecting the effect of absorption. The dashed curve includes the effect of idler absorption. The experimental tuning curve is included for reference. The incident power was 50 mW in the crystal; the last three points were obtained with $P_p=200$ mW and rescaled.

[8] T. G. Giallorenzi and C. L. Tang, Phys. Rev. **166**, 225 (1968); the following misprints in this reference should be noted: In the denominator of Eq. (36), πC^4 should read $2\pi C^4$ and $n_e^2(\omega_p\frac{1}{2}\pi)$ should read $n_e(\omega_p\frac{1}{2}\pi)$.

[9] Equation (36) of Ref. 8, where $\Delta\epsilon_1=4\pi\sum_{ijk}X_{ijk}^{(2)}e_i 0_j 0_k$. Here e_i, 0_j and 0_k are the projections of the unit polarization vectors for the extraordinary pump, and ordinary signal and idler waves on $\hat{i},\hat{j},\hat{k}$; ω_s and ω_i are the signal and idler frequencies; $\Delta\Omega_{\rm det}$ is the solid angle subtended by the detector; P_p is the pump power in the crystal.

[10] The effect of idler absorption on signal intensity can be accounted for, approximately, by averaging the signal power for a distribution of attenuated idler waves. This is done by replacing in Eq. (24) of Ref. 8

$$\int\delta(\mathbf{k}_p-\mathbf{k}_i-\mathbf{k}_s)d\mathbf{k}_i \quad \text{by} \quad G(\alpha_i)/G(0) \quad \text{where}$$

$$G(\alpha_i)=\frac{1}{\sigma}\int_0^\sigma\frac{2}{\pi}\frac{\alpha_i}{4(\Delta k)^2+\alpha_i^2}d\Delta k=\frac{1}{\pi\sigma}\tan^{-1}\left(\frac{2\sigma}{\alpha_i}\right).$$

To determine the normalizing constant $G(0)$, we know that for $\alpha_i=0$, $G(0)=\frac{1}{2}\sigma$; therefore, the normalized reduction factor $R\equiv G(\alpha_i)/G(0)=(2/\pi)\tan^{-1}(2\sigma/\alpha_i)$. A detailed theory of this will be published elsewhere.

where the first term represents the electronic contribution and the second term represents the ionic contribution. ω_0 is the resonance angular frequency of the associated transverse optical phonon and Γ is the corresponding lifetime. The absorption band[11] near 5.7 μ in $LiNbO_3$ is a two-phonon combination band of the fundamental longitudinal mode with E symmetry at 878 cm^{-1}; the next lower observed transverse mode is at 670 cm^{-1}. Based on this information and the estimated values of α_i,[12] we estimate the values of ω_0 and Γ to be used in Eq. (3) in conjunction with the data points near 5700 Å, as shown in Fig. 1 to be 2.5×10^{14} and 7.5×10^{12} sec^{-1}, respectively. Since at 5700 Å we still have not seen the ionic contribution, $|C|$ must be much less than 0.5 for this particular ir-absorption band of $LiNbO_3$; for comparison, $|C|$ for the 634-cm^{-1} mode of $LiNbO_3$ has been estimated by Giordmaine[13] to be about 6.7. A value of $|C|$ much smaller than 0.5 for the ir-absorption band we studied is, however, not unexpected because the 5.7 μ two-phonon combination band is not observed[10] in the Raman spectra of $LiNbO_3$. The corresponding Raman cross section, which gives an indication of the size of $|C|$, is expected to be orders of magnitude smaller than the single-phonon 880-cm^{-1} mode Raman cross section, which has been found[11] to be at least an order of magnitude smaller than that for the 634-cm^{-1} mode.

Figure 2 gives similar tuning curve and intensity data for a 5-cm-long ADP crystal. These data go much further into the idler absorption band than the results of Ref. 7 due to increased available incident intensity. Again, the solid curve corresponds to the calculated intensity[7,8] neglecting any absorption at the signal and idler wavelengths. With idler absorption, we have to multiply the solid curve by the reduction factor R, Eq. (2). In the case of ADP where the pump beam is not at 90° to the optical axis of the crystal, the value of σ is primarily determined by the pump-beam divergence, not by the crystal length as in the 90° case. σ is then determined from the tuning curve and the estimated divergence $\Delta\theta_p$ of the pump beam, or $\sigma=(n_s/C)(d\omega_s/d\theta_p)\Delta\theta_p$. Two values of pump-beam divergences were used in the experiment, and the results are shown in Fig. 2 along with the corresponding calculated results, assuming no dispersion in the nonlinear susceptibility. Again, the very good agreement shows that the decrease in the signal intensity can be accounted for

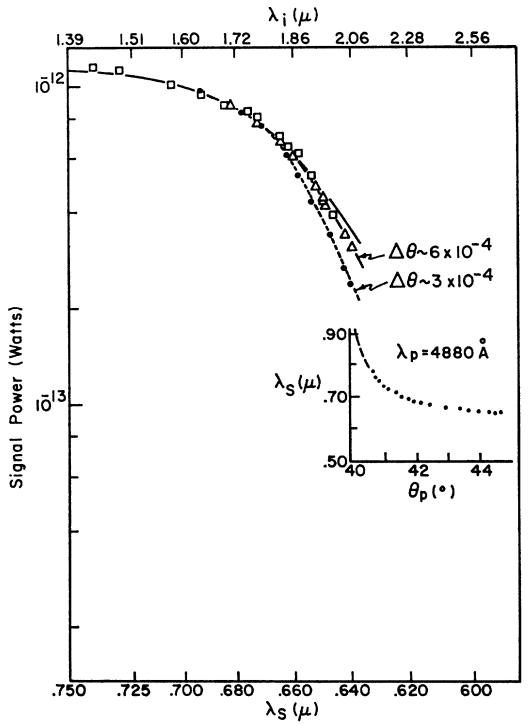

FIG. 2. Forward-scattered intensity in the spontaneous parametric scattering in ADP. The experimental tuning curve is also included for reference. The solid curve corresponds to the calculated results neglecting absorption, while the dashed curves include the effects of idler absorption. The boxes and triangles refer to two different runs with pump beam divergence $\Delta\theta_p\cong6\times10^{-4}$ rad, while the dots refer to a single run with $\Delta\theta_p\cong3\times10^{-4}$ rad. The incident power was 400 mW in the crystal.

when both the idler absorption and the pump-beam divergence are properly taken into account and there is no increase in the nonlinear susceptibility in the frequency range we studied. This is, again, presumably due to the lack of ionic contribution to the nonlinear susceptibility. However, because of the lack of suitable data on the ir or Raman spectra of ADP, it is not possible to place any upper limit on the value of $|C|$ defined in Eq. (3) on the basis of our results. The present experimental results, together with the results of Refs. 1 and 2, clearly indicate that the effect of resonant idler absorption on the optical nonlinear susceptibility, and consequently on the signal intensity in the spontaneous parametric process, and on the gain in the stimulated parametric process will depend upon the strength of the Raman scattering in the regions of interest.

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[11] A. S. Barker and R. Loudon, Phys. Rev. **158**, 433 (1967).

[12] Reference 11 gives the room-temperature values of α_i. For other temperatures, we multiplied the room-temperature values by $1+2/[\exp(\hbar\omega_i/kT)-1]$ which approximately accounts for the two-phonon combination band temperature dependence [A. S. Barker (private communication)].

[13] Reference 2 gives $d_E'/d_Q'=0.3$; using this, we calculate $|C|=(d_Q'/d_E')\cdot(\omega_p^2/\omega_0^2)=6.7$, using the notations of Refs. 1 and 4.