SPECTRAL BROADENING OF PICOSECOND 1.06 μ PULSE IN KBr

W. YU and R.R. ALFANO *
City College of New York, Physics Department, New York, New York 10031, USA

and

C.L. SAM and R.J. SEYMOUR
Philips Laboratories, 345 Scarborough Road, Briarcliff Manor, N.Y. 10510, USA

Received 11 April 1975

Spectral broadening of picosecond 1.06 μ pulses in KBr crystal is measured. The electronic Kerr effects and avalanche ionization are employed to interpret the broadening and its upper limit at the Stokes side in KBr and NaCl.

It is well known that high power ($\sim 10^8$ W) second harmonic pulses of mode-locked Nd: glass laser at 0.53 μ experience spectral superbroadening in the order of ± 4000 cm^{-1} in many materials [1]. This broadening has been studied in some detail. Several authors have suggested various mechanisms to account for it [1–3]. The broadening of the fundamental beam at 1.06 μ, particularly on the Stokes side, is not well documented. The intrinsically higher power ($10^9 - 10^{10}$ W) in the 1.06 μ pulses leads one to speculate that the Stokes spectral broadening might be broad enough to provide a high power broad-band coherent source in the near and medium infrared region. The purpose of this note is to report an experimental study of the spectral broadening of 1.06 μ pulses in KBr. A KBr crystal was chosen because of its excellent optical quality, a suitable transmission range in the infrared (0.2 – 33 μ) [4], and availability in large crystals.

Experimentally, a mode-locked Nd: glass laser pulse train was directed into a 10-cm long KBr crystal. The pulse width was measured by two photon fluorescence method to be 9 psec. Using an inverted telescope the spot size of the incident beam was reduced to 2 mm diameter with peak power density of about 10^{11} W/cm^2 with a shot-to-shot power fluctuation of approximately ±30%. The frequency broadened signal in the forward direction was collected in a 0.2 sr solid angle and focused onto the slit of a $\frac{1}{2}$ -meter Jarrell–Ash spectrometer, which has a linear dispersion 32 Å/mm at the exit slit. A photovoltaic Ge detector with 1 mm^2 active area was placed at the exit slit and the output scanned over different wavelengths was recorded on an oscilloscope. Due to the long time constant of the detector (0.2 μsec) the signal was integrated over the entire pulsetrain. The spectral range of the detection system is 0.5 to 2 μ. The signal intensity corrected with respect to the efficiencies of the filter, spectrometer, and detector, and normalized with respect to the intensity at 1.2 μ is displayed in fig. 1 as a function of wavelength. The fluctuations in the signal level were about ±30%. On the Stokes side the maximum intensity occurs at 1.2 μ. When the signal drops to 10^{-1} the span of the spectral broadening is $\Delta\nu_s = 3200$ cm^{-1} on the Stokes side, and $\Delta\nu_{as} = 4900$ cm^{-1} on the anti-Stokes side. No measurement was made in the rejection band of filters (0.8 – 1.15 μ). Beyond 1.6 μ the signal level falls off rapidly. At 1.8 μ the signal is 10^{-2} and at 2 μ no detectable signal can be observed.

The shape of the broadened spectrum in fig. 1 is similar to 0.53 μ broadening in, for example, NaCl [1]. It therefore appears that very little energy is con-

* Supported in part by a grant from Philips Laboratories and an NSF grant.

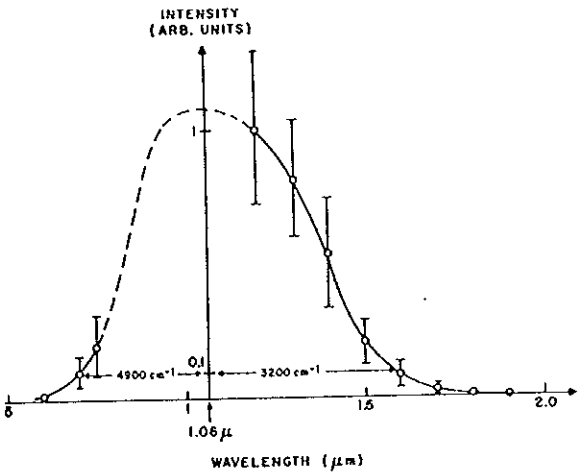

Fig. 1. The relative emission intensity vs. emission wavelength. Exciting wavelength = 1.06 μ; material: KBr.

tained in wavelengths beyond 2 μ. To verify this point the entire signal output was directed into a liquid N_2 cooled PbS detector. Two sets of data were taken, one with a filter which passes 1.2 to 3 μ. After corrections for the spectral responses of the filters, the ratio of the two measurements provides an approximate upper limit of the average power between 2 and 3 μ, which was found to be $I_{av}(2\text{–}3\ \mu) < 10^{-3}$.

The power conversion into the broadened frequencies was estimated by comparing the broadened signal with the stimulated Raman scattering (SRS) signal from nitrobenzene under identical experimental arrangements by replacing the KBr crystal with a 10-cm long nitrobenzene cell. The SRS signal has the same spectral width as the laser line width, $\delta\nu = 80\ \mathrm{cm}^{-1}$. This was larger than the 32 cm^{-1} spectral width of the Ge detector. The measured ratio I(broadened)/I_{SRS} at 1.30 μ is about 0.1. Taking a 0.01% conversion efficiency of SRS in nitrobenzene [5], the power conversion is about 10^{-5} at 1.3 μ, 10^{-6} at 1.6 μ, and 10^{-8} at 2–3 μ. These numbers translate to 1 kW per cm^{-1} at $\Delta\nu = -2000\ \mathrm{cm}^{-1}$, 0.1 kW per cm^{-1} at $\Delta\nu = 3200\ \mathrm{cm}^{-1}$, and 1 W per cm^{-1} between $\Delta\nu = 4400\ \mathrm{cm}^{-1}$ and $\Delta\nu = 6000\ \mathrm{cm}^{-1}$.

Recently, several mechanisms such as self-phase modulation due to the electronic nonlinear refractive index [1], electron avalanche breakdown [2], and collinear non-phasematched four-wave mixing due to vibrational resonance absorption [3] have been proposed to explain superbroadening of picosecond pulses in matter. In the present experiment KBr is transparent from $5 \times 10^4\ \mathrm{cm}^{-1}$ to $300\ \mathrm{cm}^{-1}$. Thus, the absence of a resonance absorption band in KBr on either the anti-Stokes or the Stokes side of the incident frequency seems to rule out the non-phase-matched vibrational resonance parametric four-photon processes [3] as a probable contributor to the broadening. Spectral broadening due to phase modulation is proportional to the negative time rate of change of the nonlinear refractive index as in the equation

$$\Delta\nu = -\frac{\omega_L}{c} l_{eff} \frac{\partial\, \Delta n}{\partial t}\,, \tag{1}$$

where ω_L is the laser frequency, l_{eff} is the effective length within the crystal over which the broadening takes place, and Δn is the sum of nonlinear refractive index changes derived from electronic and plasma contributions, i.e., $\Delta n = \Delta n_{el} + \Delta n_{pl}$. From the time development of the electron plasma formation, Bloembergen [2] indicates the plasma contribution to the broadening is negligible during the formation of the Stokes emission due to Δn_{el}. Furthermore, since Re Δn_{pl} is negative, it will only contribute to anti-Stokes broadening. It thus appears that the main contribution to Stokes broadening in KBr arises from self-phase modulation (SPM) due to the electronic nonlinear refractive index change $\Delta n_{el} = n_2 E_L^2$ which introduces a frequency sweep

$$\Delta\nu_{SPM} = -\frac{\omega_L}{c} l_{eff} \frac{\partial\, \Delta n_{el}}{\partial t}$$

$$\lesssim -\frac{\omega_L}{c} l_{eff}\, \delta\nu n_2 E_L^2\,, \tag{2}$$

where E_L is the maximum field attained in the material, n_2 the electronic nonlinear coefficient, $\delta\nu$ the spectral width of the laser.

The broadening of 0.53 μ pulses in NaCl has been measured [1] to reach $-3900\ \mathrm{cm}^{-1}$ on the Stokes side and $7300\ \mathrm{cm}^{-1}$ on the anti-Stokes side. The similarities between various physical properties of KBr and NaCl, and the nearly identical shapes of the broadened spectra, strongly suggest that the broadening mechanisms are the same in both cases. (NaCl is transparent between $5 \times 10^4\ \mathrm{cm}^{-1}$ to $550\ \mathrm{cm}^{-1}$.) It is therefore interesting to compare our result for KBr with those of ref. [1] for NaCl. From (1) one

expects the Stokes broadening to be several times wider in the KBr case, due to the nearly 10 times larger input power; however, it turned out to be narrower. To account for the discrepancy, we note that self-focusing was observed in the NaCl case. In the present experiment with KBr filament structures appeared on the tail end of the anti-Stokes spectrum, while an average of one in five shots resulted in an observable damage spot in the crystal. These evidences indicate that self-focusing also occurred in KBr. Under these conditions, Yablonovich and Bloembergen [6] have postulated that the growth of the electric field inside the filament is limited to a maximum value by the onset of avalanche breakdown. This maximum field E_L is on the order of the breakdown field strength E_{br}. When the electric field approaches this value, the electron density grows exponentially so that the real part of Δn_{pl} can be approximated by [2,6,7]

$$\mathrm{Re}\,\Delta n_{pl} = -\frac{2\pi e^2\tau^2}{n_0 m(1+\omega_L^2\tau^2)} N_0 \exp[\alpha(E)t_p] , \quad (3)$$

where the notations of ref. [6] have been adopted. $\alpha(E)$ is the ionization rate at field E and t_p is the laser pulse duration. Due to the exponential growth of $\mathrm{Re}\,\Delta n_{pl}$, its magnitude quickly exceeds those of Δn_{el} so that E_L is attained when

$$\left(\frac{\partial\,\Delta n_{el}}{\partial E}\right)_{E_L} = -\left(\frac{\partial\,\mathrm{Re}\,\Delta n_{pl}}{\partial E}\right)_{E_L} \quad (4)$$

substituting (3) into (4) it can be derived that

$$\frac{n_2 E_L^2}{\mathrm{Re}\,[\Delta n_{pl}(E_L)]} = -\tfrac{1}{2}\,\alpha(E_L)t_p\left(\frac{\partial \ln \alpha}{\partial \ln E}\right)_{E=E_L} . \quad (5)$$

From eq. (2) of ref. [7], we find that $\alpha(E_L)t_p$ has to be about 18 for the free electron density to grow to breakdown proportions. The filament size is stabilized at this time and free electron absorption becomes significant. Consequently, the effective length l_{eff} in (2) is about an absorption length [2], i.e.,

$$l_{eff} \sim \tfrac{1}{2}\,\frac{c\tau}{\mathrm{Re}\,\Delta n_{pl}} .$$

Using these results eq. (2) can be rewritten as

$$\Delta\nu \simeq \tfrac{9}{2}\,\omega_L\tau\delta\nu_p\left(\frac{\partial \ln \alpha}{\partial \ln E}\right)_{E_L} . \quad (6)$$

Using the approximation $E_L \sim E^{br}$ alkali halides [6] and the relation $\alpha(E_L) \approx 18 t_p^{-1}$ [7] the value of $(\partial \ln \alpha/\partial \ln E)_{E_L}$ for NaCl can be evaluated from a plot of ionization rate against breakdown field in, for example, fig. 1 of ref. [6]. Furthermore, Fradin et al. [8] and Smith et al. [9] have measured the breakdown fields of various alkali halides for pulses of different durations up to few tens picosecond. From their data, interpolation to picosecond region shows approximately that

$$\left(\frac{\partial \ln \alpha}{\partial \ln E^{br}}\right)^{4\,ps}_{NaCl} \sim \left(\frac{\partial \ln \alpha}{\partial \ln E^{br}}\right)^{9\,ps}_{KBr} . \quad (7)$$

Substituting (7) into (6), we find

$$\Delta\nu_{NaCl} \sim 2\Delta\nu_{KBr} \quad (8)$$

which is consistent with the observed Stokes broadening measurements. In addition, the following calculation provides a numerical check for the validity of eq. (6). Bloembergen [2] has estimated the free election collision time τ to be 3×10^{-15} sec. From fig. 1 of ref. [6], the value of $(\partial \ln \alpha/\partial \ln E_{br})$ deduced at $\alpha = 4.5 \times 10^{12}$ sec^{-1} (corresponding to $t_p = 4$ psec) is 1.7. Using these numbers in (6), the calculated values of the broadening in NaCl and KBr are $\Delta\nu_{NaCl} \sim 6600$ cm^{-1}, and $\Delta\nu_{KBr} \sim 3300$ cm^{-1}, in reasonably good agreement with the measured values 3900 cm^{-1} in NaCl [1], and 3200 cm^{-1} in KBr from this experiment.

An important consequence of this result is that when laser pulses of the same frequency and duration produce self focusing in alkali halide crystals, the spectral broadening due to SPM, (i.e., the Stokes broadening) will be independent of incident power density. This statement will also hold for materials other than KBr which possess a similar dependence of ionization rate on breakdown field as NaCl. In our experiment, the limiting electric field and therefore, the maximum Stokes broadening of 1.06 μ pulses in KBr has been achieved. Further increases in input power density would not generate a broader spectrum by SPM. Nevertheless, the high instantaneous power of this broadband radiation in the 1–2 μ region could be used as a source for additional nonlinear interactions; for example, three wave mixing of the Stokes radiation with the fundamental laser beam would generate infrared radiation beyond 3 μ.

In conclusion, the Stokes broadened spectrum of mode-locked 1.06 μ pulses in KBr has been measured.

Under self focusing conditions, a broadband near infrared radiation of high power extending out to 1.6 μ has been observed. The energy contained in radiation beyond 2 μ has been found to be small. The experimental results have been interpreted using the avalanche ionization theory postulated by Yablonovich and Bloembergen.

We would like to thank Dr. S.K. Kurtz for critically reading the manuscript and Dr. R. Bhargava for helpful suggestions.

References

[1] R.R. Alfano and S.L. Shapiro, Phys. Rev. Lett. 24 (1970) 548, and 591.
[2] N. Bloembergen, Opt. Commun. 8 (1973) 285.
[3] A. Penzkofer, A. Lauberau and W. Kaiser, Phys. Rev. Lett. 31 (1973) 863;
A. Penzkofer, Opt. Commun. 11 (1974) 265.
[4] H.A. Szemanski, Theory and Practice of Infrared Spectroscopy (Plenum Press, N.Y. 1964) p. 27.
[5] M.J. Colles, Opt. Commun. 1 (1969) 169.
[6] E. Yablonovich and N. Bloembergen, Phys. Rev. Lett. 29 (1972) 907.
[7] D.W. Fradin, N. Bloembergen and J.P. Letellier, Appl. Phys. Lett. 22 (1973) 635.
[8] D.W. Fradin, E. Yablonovich and M. Bass, in Laser Induced Damage in Optical Materials: 1972, A.J Glass and A.H. Guenther, ed. (NBS Special Publ. No. 372).
[9] W.L. Smith, J.H. Bechtel and N. Bloembergen, to be published.