PHOTON-CORRELATION ENHANCEMENT OF SHG AT 10.6 μm

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Second-harmonic-generation (SHG) experiments have been performed at 10.6 μm using a phase-matched tellurium crystal and both amplitude-stabilized and chaotic incident radiation. A factor of two enhancement in the rate of SHG has been observed for the chaotic source over the single-mode stable laser source, verifying theoretical predictions.

The connection between the rate of a multiple-photon (or nonlinear) process and the statistics of the radiation field inducing the process has been the subject of theoretical discussions by many authors [1-8]. It seems clear from this work that for the particular case of a two-photon process, induced by a chaotic source under usual conditions, the rate is expected to be twice as large as that induced by a single-mode amplitude-stabilized laser source of the same intensity. Such an enhancement, by a factor close to two (1.88 ± 0.05), has been observed by Shiga and Imamura [9] for the double-quantum photoelectric emission process [10-14] in Cs_3Sb, using an Nd-doped glass laser (1.06 μm). In this communication, we report an observed enhancement of precisely a factor of two (2.0 ± 0.1) in the rate of second-harmonic-generation (SHG) at 10.6 μm, using the CO_2 laser.

The experimental arrangement is shown in fig. 1 and is similar to that used by Shiga and Imamura [9]. The flowing-gas Brewster-window laser, which has a diffraction grating output coupling, is 200 cm long, and is operated on a single mode and a single line. It is passively Q-switched by means of an SF_6 cell inserted in the laser cavity. The emitted pulses have a peak power of approximately 200 W, a half-width of about 0.4 μsec, and a repetition rate of ≈ 10 kHz. The irradiance profile of the beam was determined to be gaussian, to very good approximation, by sweeping with a 100 μm slit. The laser was operated substantially above threshold. Thus, the radiation pulses arose from an amplitude-stabilized source operating on a single line, and on a single longitudinal and transverse mode (TEM_{00}). Excess noise, arising from photon correlations, was therefore absent from the radiation emitted by the laser [15]. The plane of polarization lies perpendicular to the plane of the paper, as shown in fig. 1.

The radiation at the output of the laser is focused by a germanium lens, through insertable calcium fluoride attenuators, onto a barium fluoride flat which is divided into two parts: half of the front surface of the flat has been roughened with 100 mesh grit to form a diffuser, the other half has been optically polished. The entire rear surface of the flat is polished. The flat is mounted on a translation stage (indicated by arrows in fig. 1) which places either the roughened or polished surface of the flat at the focus of the germanium lens. The radiation is then collected by a barium fluoride lens and a flat aluminum mirror, focused to a point, and then allowed to diverge before impinging on a potassium chloride flat, the entire rear surface of which has been roughened as described above. Thus, when the roughened portion of the barium fluoride flat is inserted in the beam, the radiation field at each point on this second diffuser is a superposition of the many coherent secondary sources generated by the first diffuser, all having random and independent phases. This diffuser is therefore assumed to be a radiation source

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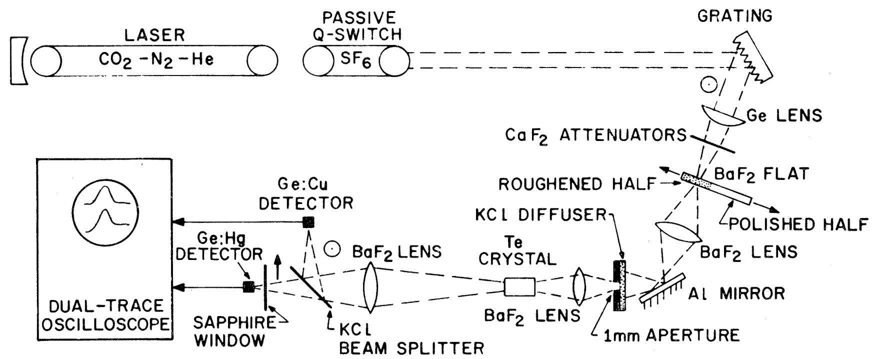

Fig. 1. Experimental arrangement.

whose electric field obeys gaussian statistics in coordinate space, and is hereafter referred to as a chaotic source [9]. If, however, the polished portion of the barium fluoride flat is inserted in the beam, the radiation field at the second diffuser maintains the amplitude-stabilized nature of the coherent laser output. Thus, changing the position of the barium fluoride flat has the effect of converting the radiation source from amplitude-stable to chaotic.

The radiation from this second diffuser is collected by a barium fluoride lens and is allowed to impinge on a 9.8 mm long single-crystal tellurium sample at the appropriate phase-match angle for optimum SHG (14° from the c axis) [16]. A 1 mm diameter circular aperture, which is smaller than the beam size at the second diffuser, is used to mask the diffuser surface in order to insure that the irradiance distribution of the imaged spot on the tellurium crystal remains essentially the same for both the coherent and the chaotic experiments. The irradiance distributions were, in fact, experimentally measured and the size of the imaged spot was shown to change by less than 7%. The radiation exciting from the tellurium crystal consists of an unconverted fundamental component at 10.6 μm and an SHG component at 5.3 μm, both of which are collected by another barium fluoride lens. The 10.6 μm radiation is sampled by an uncoated potassium chloride beamsplitter and is detected by a Ge:Cu detector operated at 4°K. The 5.3 μm radiation is detected by a Ge:Hg detector, also operated at 4°K, with a sapphire window which prevents the 10.6 μm radiation from entering it. The polarizations of both the fundamental and the SHG were experimentally verified to lie in the planes indicated in fig. 1. The outputs from both detectors were displayed on a Tektronix type 454 dual-trace oscilloscope.

A run was performed by first placing the polished portion of the movable barium fluoride flat in the beam. This provided an amplitude-stable source at the SHG crystal. The peak radiation powers at 10.6 μm (P_{ω}) and at 5.3 μm ($P_{2\omega}$) were then measured from the two oscilloscope traces and plotted as the triangular data points presented in fig. 2. Different values of P_{ω} were obtained by placing various thicknesses of calcium fluoride attenuators in the beam before the barium fluoride flat (see fig. 1). The incident radiation power density was sufficiently low so that free-carrier reduction of SHG efficiency did not occur [17]. Thus, the fundamental radiation transmitted through the tellurium crystal, and registered on the oscilloscope was a true indication of the incident radiation power. The line drawn through the triangles has a slope of 2 on a log-log plot, as it should for the two-photon SHG process ($P_{2\omega} \propto P_{\omega}^2$). Then, the roughened portion of the barium fluoride flat was placed in the beam and an identical experiment was performed; the data points are indicated as circles in fig. 2. It may be seen that once again the slope of the curve is identically 2. In this case, corresponding to the chaotic source, the curve lies above the previous case (for the amplitude-stabilized source) by a factor of 2.0 ± 0.1. Therefore, the expected enhancement for the SHG rate, obtained by using a chaotic source rather than an amplitude-stabilized source, has been demonstrated at 10.6 μm.

Nevertheless, it would not be practical to convert a coherent source into a chaotic source as we have done in this experiment, because of the loss of incident radiation power which occurs in placing the diffusers and other optics in the beam. It should, however, be useful for laser

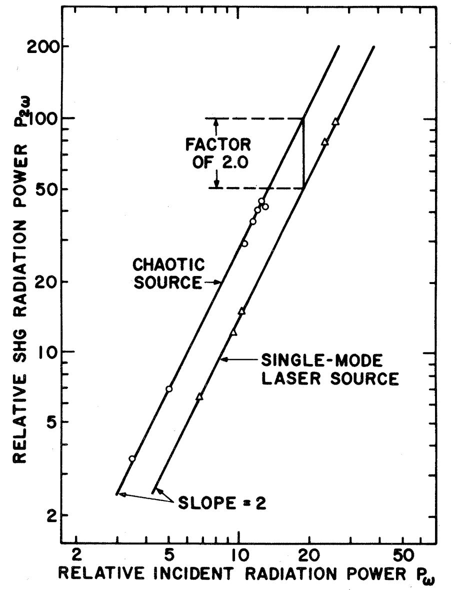

Fig. 2. SHG radiation power at 5.3 μm versus incident radiation power at 10.6 μm (relative units).

radiation containing many modes, where the radiation statistics approach those of the chaotic source, thus yielding the observed enhancement [18-20]. Finally, we note that the expected enhancement becomes larger for higher-order processes [1]; for chaotic sources possessing precise first-order coherence, the m-photon rate is enhanced by a factor of $m!$ over the single-mode ideal laser source. Thus, third-harmonic-generation (THG) should be enhanced by a factor of $3! = 6$. For a general radiation field of arbitrary statistical properties, the enhancement has been shown [1] to be given by a factor which in general differs from unity [2,21,22].

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