Observations of second-harmonic generation in isotropic vapors tested at low power

Victor Mizrahi and D. P. Shelton
Department of Physics, University of Toronto, Toronto, Ontario, Canada M5S 1A7
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Observations of second-harmonic generation in Na vapor with a pulsed laser have been reported. We have checked these observations with a sensitive cw technique at low power. No second-harmonic generation is observed, giving support to proposed mechanisms requiring high-power pulses.

The observation of second-harmonic generation (SHG) in atomic Na vapor with a single laser beam and no applied external fields has been reported by Miyazaki, Sato, and Kashiwagi,[1] despite the expected absence of SHG on the basis of symmetry. They argue that this is due to static-field generation resulting from the interaction of the high-power laser pulse with the atomic medium. Others have also reported similar effects,[2,3] although there has been some discussion as to the cause.[4] By making use of a cw laser system built for dc electric-field-induced second-harmonic generation (ESHG) experiments in gases,[5,6] we have been able to test for this effect at low power.

About 500 mw (300 watt/cm^2) from a cw dye laser tuned to 589.3 nm (midway between the sodium D lines) is weakly focused into a Na heat pipe cell. A double prism spectrometer tuned to the second-harmonic frequency eliminates the fundamental, and ultraviolet photons are detected with a photomultiplier tube operating in the photon counting regime. Substantial ultraviolet is observed due to a variety of mechanisms, such as investigated by previous workers.[7] However, this is not second-harmonic generation.

A sensitive test for SHG in the Na vapor is provided by preceding the Na cell with an ESHG cell containing gaseous fluoroform (CHF_3) as the active medium. Periodic phase matching[8] ensures a strong ESHG signal. Any second harmonic generated in the Na cell will have a fixed phase relationship to that from the ESHG cell. The total observed signal will therefore be given by

$$S_{\rm obs} \propto S_{\rm ESHG} + 2\cos\phi\sqrt{S_{\rm ESHG}S_{\rm SHG}} + S_{\rm SHG} + S_{\rm uv} , \quad (1)$$

where $S_{\rm ESHG}$ is the ESHG signal due to the CHF_3 alone, $S_{\rm SHG}$ is the SHG signal (if any) from the Na alone, $S_{\rm uv}$ is the background uv signal from the Na, and ϕ is the relative phase shift between the second-harmonic electric field from the CHF_3 and the Na. The second term results from interference between the ESHG and the SHG wave. By modulating the dc field applied to the CHF_3, and using synchronous photon counting techniques, only the first two terms in (1) are detected. As the amplitude of the ESHG wave is proportional to the applied static field, the second term changes sign upon reversal of the static-field polarity. Comparing measurements made with positive (S_+) and negative (S_-) polarity we therefore find

$$\frac{S_+}{S_-} = \frac{1+2\cos\phi(S_{\rm SHG}/S_{\rm ESHG})^{1/2}}{1-2\cos\phi(S_{\rm SHG}/S_{\rm ESHG})^{1/2}} . \quad (2)$$

For $S_{\rm SHG} \ll S_{\rm ESHG}$ this simplifies to

$$\frac{S_+}{S_-} - 1 = 4\cos\phi\left[\frac{S_{\rm SHG}}{S_{\rm ESHG}}\right]^{1/2} . \quad (3)$$

In general, the phase factor ϕ is unknown, but by repeating the measurement with the Na cell moved one half coherence length (defined as the distance in the medium over which the second-harmonic and the fundamental generating wave get out of phase by $\pi/2$ radians) of the air (about 5 mm) closer to the ESHG cell, the phase factor changes from $|\cos\phi|$ to $|\sin\phi|$. The larger of $|\cos\phi|$ and $|\sin\phi|$ is greater than or equal to $1/\sqrt{2}$ for any ϕ. Therefore, taking the result with the largest deviation guarantees that

$$\frac{S_{\rm SHG}}{S_{\rm ESHG}} \le \frac{1}{8}\left|\frac{S_+}{S_-} - 1\right|^2 . \quad (4)$$

A series of measurements were made over a range of Na pressures from 50 mTorr up to 630 mTorr, without buffer gas. No significant SHG from the Na vapor was observed, and using (4) we have determined that

$$\frac{S_{\rm SHG}}{S_{\rm ESHG}} < 2\times 10^{-4} . \quad (5)$$

Since ESHG count rates of about 1700 counts/sec were observed, the upper bound on the SHG from the Na is 0.3 count/sec. (It is estimated that for this system about one second-harmonic photon in 60 is eventually detected and counted.) At the higher sodium densities an $S_{\rm uv}$ background signal comparable to $S_{\rm ESHG}$ was observed.

As a demonstration of the sensitivity of the system, the Na cell was replaced with a simple quartz $\lambda/2$ plate with optic axis perpendicular to the direction of the laser beam propagation. The quartz acts as a second-harmonic generator, and with the plate rotated about its optic axis for maximum signal (which coincides with zero polarization rotation of the beam), $S_{\rm SHG}$ (quartz) $\simeq$400 counts/sec was easily observed. Note that the quartz is not phase matched, so that only one coherence length ($\simeq 10$ μm)

plays an active role.

The failure to detect significant SHG from the Na vapor, despite the proven sensitivity of the system, gives support to those models of SHG which rely upon the existence of high-power laser pulses, such as the static-field generation theory of Miyazaki *et al.* We would suggest that the technique of using an ESHG cell in series with the vapor cell would also be useful for those observing the effect at high powers, as this provides an unambiguous test for true second-harmonic generation.

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[1]K. Miyazaki, T. Sato, and H. Kashiwagi, Phys. Rev. A **23**, 1358 (1981).

[2]T. Mossberg, A. Flusberg, and S. R. Hartmann, Opt. Commun. **25**, 121 (1978).

[3]R. W. Boyd (private communication).

[4]D. S. Bethune, Phys. Rev. A **23**, 3139 (1981).

[5]Victor Mizrahi and D. P. Shelton, Phys. Rev. A **31**, 3145 (1985).

[6]Victor Mizrahi and D. P. Shelton, Phys. Rev. A **32**, 3454 (1985).

[7]J. K. Chen, C. Y. R. Wu, C. C. Kim, and D. L. Judge, Appl. Phys. B **33**, 155 (1984), and references therein.

[8]D. P. Shelton and A. D. Buckingham, Phys. Rev. A **26**, 2787 (1982).