Optical Sum-Frequency-Generation Interference in Atomic Sodium Vapor*

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Optical sum-frequency generation, due to either noncollinear excitation, or magnetic-field mixing, or to their combined effect, is observed in atomic sodium vapor. Novel interference effects are observed and explained.

Three-wave mixing[1] in unpolarized atomic vapors has recently been demonstrated for the first time: in sodium by the generation of $E2$ sum-frequency radiation (SFG) from noncollinear laser-beam excitation[2]; and in thallium by the generation of difference-frequency radiation (DFG) from collinear laser-beam excitation in the presence of a magnetic field.[3] We have repeated the sodium SFG experiment of Bethune, Smith, and Shen.[2] We find that if, in addition, we apply a magnetic field, we obtain enhanced SFG, and we report the observation of novel interference effects.

Our experimental procedure and apparatus are similar to those of Bethume, Smith, and Shen, except that the optical excitation fields are provided by two nitrogen-laser–pumped dye lasers producing 6-nsec-long pulses of frequencies ω_1 and ω_2 tunable near the 16 956-cm^{-1} $3^2S_{1/2}$-$3^2P_{1/2}$ and 17 594-cm^{-1} $3^2P_{1/2}$-4^2D Na absorption lines, respectively. The laser linewidths are 0.7 and 0.4 cm^{-1}, respectively. The laser frequencies are adjusted so that their sum ω_3 overlaps the Doppler-broadened two-photon $3^2S_{1/2}$-4^2D adsorption spectrum. The two noncollinear laser pulses are focused into and cross at the center of a cell containing argon buffer gas and a zone of Na vapor of length $Z \cong 25$ cm. The spot size d and confocal parameter s of each laser beam is about 0.5 mm and 20 cm, respectively, at the crossing point. Two orthogonal Helmholtz coils supply the requisite transverse magnetic field. The crossing angle θ ($\ll 1$) between $\vec{k}_1$ and $\vec{k}_2$ is always adjusted so that the phase-matching condition $\vec{k}_3 = \vec{k}_1 + \vec{k}_2$ is satisfied[2]; here $\vec{k}_{1,2}$ and $\vec{k}_3$ denote, respectively, the wave vectors in the Na vapor of the incident laser beams and the SFG wave of frequency ω_3. The detected energy of the SFG wave is averaged over several hundred shots by a PDP/8E computer. We always work at Na densities and incident laser intensities low enough so that the SFG intensity (for fixed geometry) varies as the product of the two laser intensities.[2]

In the absence of a magnetic field, our experimental results are in excellent agreement with those of Ref. 2. For example, when the laser electric-field polarizations are perpendicular to each other and one lies along $\vec{k}_1 \times \vec{k}_2$, the SFG signal is polarized along $\vec{k}_1 \times \vec{k}_2$, and its intensity varies as the square of the atomic Na density n (Fig. 1). When the laser polarizations are parallel and lie along $\vec{k}_1 \times \vec{k}_2$, no SFG is observed.

The application of a transverse magnetic field $\vec{H}$ (i.e., a field transverse to $\vec{k}_3$) produces dramatic effects. For example, parallel laser polarizations along $\vec{k}_1 \times \vec{k}_2$ now generate SFG. For $\vec{H}$ along

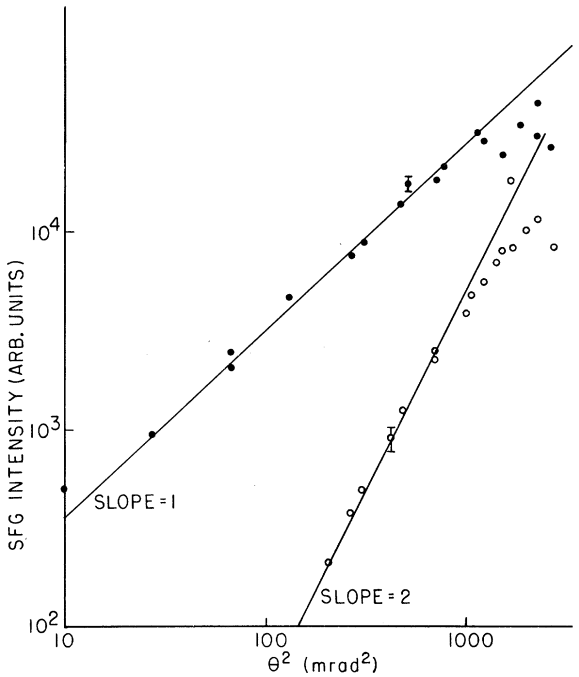

FIG. 1. SFG intensity vs θ^2. Conditions: $\tilde{\nu}_1 = \omega_1/2\pi c = 16\,915$ cm^{-1}; laser energies W' in cell $\cong 1$ μJ each. Note that $\theta^2 \propto n$, and that $\theta^2 = 800$ mrad^2 corresponds to $n \cong 1 \times 10^{16}$ atoms/cm^3. Open circles: $H = 0$, $\vec{E}_1$ along $\hat{y}$, $\vec{E}_2$ perpendicular to $\hat{y}$. Solid circles: $\vec{E}_{1,2}$ along $\hat{y}$, $\vec{H} = H\hat{x}$, $H = 37$ G. We believe that the deviation of the data from slopes 1 and 2, respectively, at higher values of n is caused by dimer absorption of the incident lasers. The vertical scales are not the same for the two sets of data.

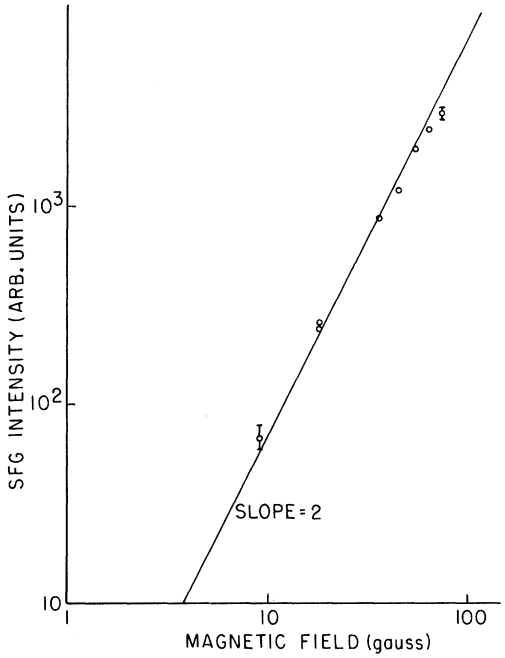

FIG. 2. SFG intensity vs H. Conditions: $n \cong 4\times 10^{14}$ cm^{-3}, $\theta \cong 3$ mrad, $E_{1,2}$ along $\hat{y}$, $\bar{\nu}_1 = \cong 16\,900$ cm^{-1}; $W' \cong 1\,\mu$J. 1 unit on the vertical scale corresponds to an SFG pulse energy of $\cong 10^{-17}$ J.

$\vec{k}_3 \times (\vec{k}_1 \times \vec{k}_2)$, sizable SFG signals linear in n (Fig. 1) and quadratic in H (Fig. 2) occur.

For orthogonal laser polarizations the effect of an applied magnetic field is more complicated. Let the magnetic field and the beam from one of the lasers be polarized along $\vec{k}_1 \times \vec{k}_2$. The SFG intensity $I(H)$ now depends on the *sign* of H; and there is a nonzero value of H for which the SFG intensity is a relative minimum (Fig. 3). If, instead, $\vec{H}$ is applied along $\vec{k}_3 \times (\vec{k}_1 \times \vec{k}_2)$, the SFG intensity $I(H)$ becomes a monotonically increasing function of $|\vec{H}|$. In either case, the SFG intensity varies linearly with n at low densities and quadratically with n at high densities.

Although the behavior of the field-dependent intensity $I(H)$ for the case in which the laser polarizations are orthogonal and $\vec{H}_1$ is along $\vec{k}_1 \times \vec{k}_2$ is complicated, it has a hidden simplicity which is uncovered by plotting (Fig. 4) separately $I(+H) - I(-H)$ and $I(H) + I(-H) - 2I(H=0)$—the former is found to vary simply as H, the latter as H^2. As we now show, this behavior results from the fact that the SFG is due to an interference of two terms, one of which is due to the noncollinearity of the laser beams, and the other to the applied magnetic field.

We first treat the case of monochromatic excitation (well-defined sum frequency ω_3). We denote the electric fields of the noncollinear beams by $2\,\mathrm{Re}\vec{E}_{1,2}\exp[i(\vec{k}_{1,2}\cdot\vec{x} - \omega_{1,2}t)]$. The ground states $3^2S_{1/2}$ and excited states 4^2D, labeled $|a\rangle$ and $|b\rangle$, respectively, are connected by an $E2$ matrix element $\overleftrightarrow{Q}_{ab}$. We assume that ω_3 is nearly equal to the energy separation Ω_{ba} between states $|b\rangle$ and $|a\rangle$. In the absence of a magnetic field,

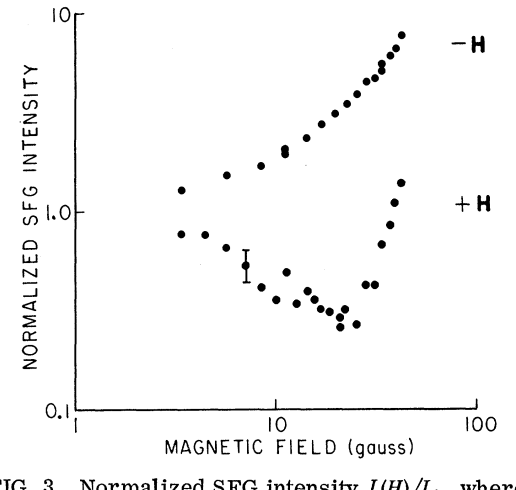

FIG. 3. Normalized SFG intensity $I(H)/I_0$, where $I_0 = I(H=0)$. Conditions: $\vec{E}_1$ along $\hat{y}$; $\vec{E}_2$ perpendicular to $\hat{y}$; $H = +H\hat{y}$ (lower data), $H = -H\hat{y}$ (upper data); $\bar{\nu}_1 \cong 16\,855$ cm^{-1}, $n \cong 1.5\times 10^{16}$ cm^{-1}, $W' \cong 1\,\mu$J.

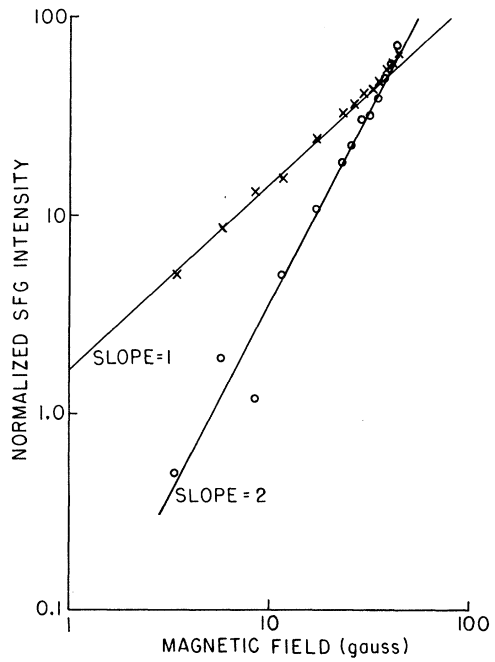

FIG. 4. Difference and sum of plots of Fig. 3. For crosses, the abscissa is $[I(+H) + I(-H) - 2I_0]/I_0$; for circles, the abscissa is $[I(+H) - I(-H)]/I_0$.

the two incident beams mix in the interaction or overlap volume of the vapor for form a macroscopic $E2$ moment density $2\,\mathrm{Re}\vec{\vec{Q}}_0 \exp[i(\vec{k}_3\cdot\vec{x}-\omega_3 t)]$,

$$\vec{\vec{Q}}_0(\omega_3)=\vec{\vec{\chi}}_Q^{(2)}:\vec{E}_1\vec{E}_2=n\pi\hbar^{-1}\textstyle\sum_{a,b} g(\omega-\Omega_{ba})\vec{\vec{Q}}_{ab}[\vec{\vec{\alpha}}_{ba}:\vec{E}_1\vec{E}_2]. \tag{1}$$

Here $2\pi\hbar$ is Planck's constant, $\vec{\vec{\chi}}_Q^{(2)}$ is the susceptibility tensor,[2] α_{ba} is the b-a element of the two-photon polarizability tensor,[4] and $g(\nu)$ is the complex two-photon line shape which for the $\vec{\vec{\chi}}^{(2)}$ of Ref. 2 is given by $\pi^{-1}(\nu+i\Gamma)^{-1}$, with the linewidth Γ as defined therein. We assume that the three-wave mixing is phase-matched, with θ ($\ll 1$) the angle between $\vec{k}_1$ and $\vec{k}_2$. Choosing coordinate axes $\hat{x}$, $\hat{y}$, and $\hat{z}$, with $\hat{z}$ along $\vec{k}_3$ and $\hat{y}$ along $\vec{k}_2\times\vec{k}_1$, and noting that in our experiment $|\omega_1-\omega_2|\ll\omega_1$, we find that the electric-field envelope $\vec{E}_0$ of the SFG wave leaving the interaction volume (proportional to the transverse component of $z\cdot\vec{\vec{Q}}_0$[5]) is given by

$$\hat{x}\cdot\vec{E}_0\cong n\alpha\theta k_3^2 l(\hat{x}\cdot\vec{E}_1)(x\cdot\vec{E}_2)(\omega_1/\omega_2-1),\quad \hat{y}\cdot\vec{E}_0\cong n\alpha\theta k_3^2 l\hat{z}\cdot(\vec{E}_2\times\vec{E}_1) \tag{2}$$

to lowest order in $\vec{E}_1$ and $\vec{E}_2$.[2] Here $n\alpha=|\vec{\vec{\chi}}_Q^{(2)}|$; and l is the length of the interaction volume. We refer to $\vec{E}_0$ as the "noncollinear" contribution to the SFG electric field. The interaction length l defined above is the smaller of Z, s, and the laser overlap length d/θ. We note that $\theta\propto\sqrt{n}$, since the dispersion is due to Na vapor alone.[2] At the densities at which we work, θ is large enough so that $l=d/\theta$ and the factor θl in Eq. (2) is actually independent of θ.

The application of an external magnetic field $\vec{H}$ mixes the various $|a\rangle$ states and $|b\rangle$ states, respectively, among themselves via the magnetic dipole interaction $-\vec{M}\cdot\vec{H}$, producing an additional $E2$ moment density $\vec{\vec{Q}}_H$. A perturbation expansion to lowest order in $\vec{E}_1$, $\vec{E}_2$, and $\vec{H}$ yields[6]

$$\begin{aligned}\vec{\vec{Q}}_H(\omega_3)=n\pi\hbar^{-1}\{&\sum_{a,b,b'}[g(\omega_3-\Omega_{b'a})-g(\omega_3-\Omega_{ba})]\vec{\vec{Q}}_{ab}[\vec{\vec{\alpha}}_{b'a}:\vec{E}_1\vec{E}_2][\vec{M}_{bb'}\cdot\vec{H}]\Omega_{bb'}^{-1}\\&-\sum_{a,a',b}[g(\omega_3-\Omega_{ba'})-g(\omega_3-\Omega_{ba})]\vec{\vec{Q}}_{ab}[\vec{\vec{\alpha}}_{ba'}:\vec{E}_1\vec{E}_2][\vec{M}_{a'a}\cdot\vec{H}]\Omega_{a'a}^{-1}\},\end{aligned} \tag{3}$$

where the various ground-state (excited-state) levels are denoted by a,a' (b,b').[7] We note that although $|\vec{\vec{Q}}_H(\omega_3)|\ll|\vec{\vec{Q}}_0(\omega_3)|$, the radiated field $\propto|\hat{z}\times[\hat{z}\cdot\vec{\vec{Q}}_H(\omega_3)]|$ may easily exceed $|\hat{z}\times[\hat{z}\cdot\vec{\vec{Q}}_0(\omega_3)]|$ since $\hat{z}\cdot\vec{\vec{Q}}_0$ is mainly along $\hat{z}$, whereas $\hat{z}\cdot\vec{\vec{Q}}_H$ is mainly perpendicular to $\hat{z}$. Utilizing the spherical symmetry of the unexcited atom, we obtain the form of $\vec{\vec{Q}}_H$ by defining the various rank combinations $T^{(J)}$ of $\vec{E}_1$ and $\vec{E}_2$: $T^{(0)}=\vec{E}_1\cdot\vec{E}_2$, $\vec{T}^{(1)}=\vec{E}_1\times\vec{E}_2$, and $\vec{\vec{T}}^{(2)}=\frac{1}{2}(\vec{E}_1\vec{E}_2+\vec{E}_2\vec{E}_1)-\frac{1}{3}\vec{\vec{I}}(\vec{E}_1\cdot\vec{E}_2)$, where $\vec{\vec{I}}$ is the unit dyadic. [A similar argument leads to the form of Eq. (2).] After combining the $T^{(J)}$'s with $\vec{H}$ to form symmetric second-rank tensors, we find that the electric-field envelope $\vec{E}'$ ($\propto$ transverse component of $\hat{z}\cdot\vec{\vec{Q}}_H$[5]) of the magnetically induced component of the SFG wave is of the form[6]

$$\vec{E}'(\omega_3)=nk_3^2 l\{\mu_1\hat{z}\cdot(\vec{E}_1\times\vec{E}_2)\vec{H}+\mu_2[\hat{z}\cdot(\vec{E}_2\times\vec{H})\vec{E}_1+\hat{z}\cdot(\vec{E}_1\times\vec{H})\vec{E}_2]\}. \tag{4}$$

It may be shown[6] that μ_1, $\mu_2(\omega_3)\cong\alpha\mu_B/\hbar\Gamma$, where μ_B is the Bohr magneton.[8] Comparison of Eqs. (2) and (4) shows that $|E'/E_0|\cong\mu_B H/\theta\hbar\Gamma$. Clearly, $\vec{E}_0$ and $\vec{E}'$ will interfere either constructively or destructively, depending on the signs of μ_1/α and μ_2/α, and on the direction of $\vec{H}$. Since $\theta\propto n^{1/2}$, the SFG power will come principally from the magnetic effect at low n (or high H), and from the noncollinear effect at high n (or low H). In the intermediate regime ($\mu_B H\cong\theta\hbar\Gamma$), the interference between $\vec{E}_0$ and $\vec{E}_1$ will be apparent, the energy of the SFG wave changing significantly under reversal of $\vec{H}$.

We now generalize the above description in which only a single frequency component is present in the lasers' sum-frequency spectrum. Letting $E_1\to E_1 f_1(\omega_1)$ and $E_2\to E_2 f_2(\omega_2)$, where $f_{1,2}(\omega)$ is the normalized Fourier transform of laser electric fields 1 and 2, respectively, we define $f(\omega_3)=\int f_1(\omega_1)f_2(\omega_3-\omega_1)d\omega_1$. The SFG mean intensity is then given by

$$I\propto\int|f(\omega_3)|^2|\vec{E}_0(\omega_3)+\vec{E}'(\omega_3)|^2 d\omega_3. \tag{5}$$

From Eqs. (2), (4), and (5), it follows that I may in general be written as a sum of three terms, the first independent of H and the other two proportional to H and H^2, respectively.

We now compare the above theoretical predictions with experiment. In all cases, $\vec{E}_1$ lies along $\hat{y}$. The experimental results divide themselves naturally into four sets:

(a) $\vec{H}$ and $\vec{E}_2$ along $\hat{y}$. According to Eqs. (2) and (4), both $\vec{E}_0$ and $\vec{E}'$ vanish; and, indeed, no SFG

is observed in this case. This result distinguishes $E2$ from $M1$ three-wave mixing, in which generation occurs when $\vec{H}$, $\vec{E}_1$, and $\vec{E}_2$ are parallel.[3]

(b) $\vec{H}+H\hat{x}$, $\vec{E}_2$ along $\hat{y}$. Only the H-dependent term contributes; hence, $I \propto n^2 l^2 H^2 |E_1|^2 |E_2|^2 \propto nH^2 |E_1|^2 \times |E_2|^2$, as observed (Figs. 1 and 2).

(c) $\vec{H}=\pm H\hat{y}$, $\vec{E}_2$ perpendicular to $\hat{y}$. From Eqs. (2), (4), and (5) we obtain

$$I \propto n^2 l^2 |E_1|^2 |E_2|^2 [\theta^2 \int |\mu_0(\omega)|^2 |f(\omega)|^2 d\omega \mp 2H\theta \int \mu_0(\mu_1 - \mu_2) |f(\omega)|^2 d\omega + H^2 \int |\mu_1 - \mu_2|^2 |f(\omega)|^2 d\omega],$$

which explains the shape of Fig. 3 and the simplicity of the combinations plotted in Fig. 4. The minimum of Fig. 3 ($\theta = 35$ mrad) occurs at $H = 25$ G which, for $\Gamma \cong 3\times 10^{10}$ sec^{-1},[2] is in reasonable agreement with the expected value $\cong \theta\hbar\Gamma/\mu_B \cong 100$ G.

(d) $\vec{H}=\pm H\hat{x}$, $\vec{E}_2$ perpendicular to $\hat{y}$. Since $\vec{E}_0$ is perpendicular to $\vec{E}'$, the intensities of the noncollinear and magnetically induced signals add, and $I(H) - I(H=0) \propto H^2$. This prediction was verified in our experiment.

To summarize, we have observed the effect of a magnetic field on SFG in atomic Na vapor. Depending on experimental conditions, the unperturbed SFG can be either enhanced or degraded. All observations are in agreement with theory. Similar effects should be observable in DFG.

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[7]In the limit $\Omega_{bb'} \to 0$, the ratio $[g(\omega - \Omega_{ba}) - g(\omega - \Omega_{b'a})]\Omega_{bb'}^{-1} \to dg/d\omega$ is well defined.

[8]Equivalently, the effect of the magnetic field in producing three-wave mixing is of order $(\vec{M}\cdot\vec{H})/\hbar\Gamma$, where Γ is the Doppler width if depolarizing collisions may be neglected. This result is derived in Ref. 6. We note that in Ref. 3 it was stated that for DFG on the $T1$ $6^2P_{1/2}$-$6^2P_{3/2}$ transition, the effect of $\vec{H}$ is of order $(\vec{M}\cdot\vec{H})/\hbar\Delta\omega$, where $\Delta\omega/2\pi$ is the $6^2P_{3/2}$-state hyperfine splitting (520 MHz). This statement is misleading; DFG will occur via magnetic mixing of *Zeeman* levels even in the absence of hyperfine splitting. The effect of $\vec{H}$ is always of the order of $(\vec{M}\cdot\vec{H})/\hbar\Gamma$. The estimate of Ref. 3 is valid, however, since for the transition in question, $\Delta\omega \cong$ the Doppler width Γ.

Thermal-Force Terms and Self-Generated Magnetic Fields in Laser-Produced Plasmas

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The several magnetic-source terms affecting laser-produced plasmas are presented and their importance discussed. The thermal force is shown to greatly modify the magnetic-field distribution in numerical simulations. Experimental implications are also discussed.

In 1971, self-generated magnetic fields were proposed as an explanation of some physical phenomena in laser-produced plasmas.[1] Since that time, their presence has been confirmed experimentally,[2,3] and a number of theoretical reasons have been given why they may be expected to have an important effect on the plasma dynamics.[4-7] We present here analysis of these effects, and draw attention to the features of the magnetic-field source terms which may assist further experimental studies.

In these studies, we use a computational model which has been described in detail elsewhere.[8] Using a simplified magnetic-source term, it has