Optical Difference-Frequency Generation in Atomic Thallium Vapor*

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Coherent radiation at the 1.28-μm electric-dipole-forbidden $6^2P_{3/2}$-$6^2P_{1/2}$ transition in thallium vapor has been produced by the simultaneous application of a magnetic field and two collinear, overlapping laser beams nearly resonant with the $6^2P_{1/2}$-$7^2S_{1/2}$ and $6^2P_{3/2}$-$7^2S_{1/2}$ transitions, respectively. The effect of the magnetic field is to mix hyperfine levels —atomic polarization effects are negligible. The optical difference-frequency power generated at low magnetic field varies as the square of the magnetic-field component normal to the laser-beam propagation direction.

Two types of optical-wave-mixing effects which have been studied extensively in the last 10 years are the following: Three-wave mixing in a crystal lacking inversion symmetry,[1] and four-wave mixing in an atomic vapor.[2] Both of these processes are allowed in the electric-dipole approximation. It is well known, however, that in the $E1$ approximation the mixing of three electromagnetic waves is forbidden by parity conservation in both atomic media and in crystals having inversion symmetry. If the higher-order $M1$ and $E2$ moments are taken into account, these restrictions do not exist. Indeed, optical three-wave mixing via both the $M1$ and $E2$ interactions have been reported. In a cooled (15 K) and magnetically polarized (40 kG) InSb crystal, difference-frequency generation (DFG) associated with a far-ir (90 cm^{-1}) $M1$ Raman spin-flip transition was observed.[3] Recently, uv sum-frequency generation (SFG) associated with the $5^2S_{1/2}$-$6^2D_{3/2}$ $E2$ transition has been observed in atomic sodium vapor excited by two noncollinearly propagating laser beams.[4]

It is generally recognized that, given two *collinearly* propagating laser beams, the presence of Zeeman-level polarization in a medium allows three-wave mixing to occur via either an $M1$ or $E2$ interaction.[5] However, for an unpolarized medium, an important effect which has not been considered before is the strong symmetry-breaking influence of a transverse magnetic field, i.e., an applied field transverse to the direction of propagation of the waves. Such a field may mix adjacent hyperfine energy levels in each of the two atomic states between which the $M1$ or $E2$ moment is induced. It may thus allow coherent $M1$ or $E2$ radiation to be emitted along the common direction of propagation. In this Letter we report optical DFG at the 7793-cm^{-1} frequency associated with the $6^2P_{1/2}$-$6^2P_{3/2}$ transition in atomic thallium. The thallium vapor is excited by two collinear laser beams in the presence of a weak static transverse magnetic field. We attribute the observed DFG to the mixing of adjacent hyperfine levels; atomic polarization effects are negligible.

Consider a medium made up of atoms, each having a ground state $|a\rangle$, and excited state $|b\rangle$ of the same parity, and other states $|n\rangle$ of opposite parity. We first assume that each state is nondegenerate. The atoms, all of which are initially in the ground state, are irradiated by two laser beams having the electric fields $2\,\mathrm{Re}\vec{E}_1 \times \exp[i(\vec{k}_1\cdot\vec{x} - \omega_1 t)]$ and $2\,\mathrm{Re}\vec{E}_2 \exp[i(\vec{k}_2\cdot\vec{x} - \omega_2 t)]$, respectively, where $\omega_1 - \omega_2 = \omega_{ba}$, the $b \to a$ transition frequency. The laser irradiation induces a macroscopic atomic excitation at the $b \to a$ transition frequency. If, for example, the $b \to a$ transition is $M1$-allowed, the macroscopic excitation may radiate coherent $M1$ radiation of frequency ω_{ba} and propagation vector $\vec{k}_1 - \vec{k}_2$. However, if the states $|a\rangle$ and $|b\rangle$ each contain several degenerate Zeeman levels $|m_a\rangle$ and $|m_b\rangle$, respectively, the net magnetization induced in the medium at ω_{ba} is the coherent sum of the magnetizations associated with the various $|m_b\rangle \to |m_a\rangle$ transitions. From symmetry considerations[5,6] it may then be shown that in the absence of laser saturation effects the magnetization $\vec{M}(\omega_{ba})$ induced at the frequency ω_{ba} is given by

$$\vec{M}(\omega_{ba}) = \alpha N \vec{E}_1 \times \vec{E}_2^*, \tag{1}$$

where $|\alpha| \cong \mathfrak{M}_{ba} \hbar^{-2} \gamma_{ba}^{-1} \sum_n p_{an} p_{nb} (\omega_1 - \omega_{na})^{-1}$, N is the atomic number density, p_{ij} ($\mathfrak{M}_{ij}$) is the $E1$ ($M1$) matrix element between states i and j ($= a, b, n$), $\hbar$ is Planck's constant divided by 2π, γ_{ba} is the linewidth of the $b \to a$ transition, and the sum is taken over n. For coherent emission of frequency $\omega_3 = \omega_{ba}$ and wave vector $\vec{k}_3$ to occur, phase matching requires that $\vec{k}_3 = \vec{k}_1 - \vec{k}_2$. For small enough N, dispersion of all three waves may be neglected; this condition was, unless otherwise stated, satisfied in each of our experiments. For negligible dispersion, phase matching occurs when beams 1 and 2 propagate in the same direction. However, only the component of $\vec{M}$ normal to $\vec{k}_3$ may radiate coherently; thus, Eq. (1) predicts that no DFG will occur. If we quantize along the direction of propagation z, we see that only $\Delta m = 0$, ± 2 transitions are induced by the Raman interaction with the two waves; however, it is well known that only $\Delta m = \pm 1$ transitions may radiate along z.[7] Thus the absence of DFG, as predicted by Eq. (1), may be viewed as an outcome of the conservation of the z component of angular momentum of an atom interacting with two electromagnetic quanta.[5]

Next consider the effect of applying a static transverse magnetic field $\vec{H}$ during the optical irradiation. The symmetry argument leading to Eq. (1) now fails because the Hamiltonian contains the term $-\vec{H}\cdot\vec{\mathfrak{M}}$. Symmetry considerations show that an additional magnetization $\vec{M}'(\omega_{ba})$ may be formed in the medium; to lowest order in $\vec{H}$, $\vec{E}_1$, and $\vec{E}_2$, it is given by

$$\vec{M}' = N\{\beta\vec{E}_1(\vec{E}_2{}^*\cdot\vec{H}) + \gamma\vec{E}_2{}^*(\vec{E}_1\cdot\vec{H}) + \delta\vec{H}(\vec{E}_1\cdot\vec{E}_2{}^*)\}. \tag{2}$$

The magnitudes of β, γ, and δ are of the order of $|\alpha\mathfrak{M}_{ij}(\hbar\omega_{ij})^{-1}|$, where $j = a$ or b and i is the nearest (hyperfine) level for which $\mathfrak{M}_{ij} \neq 0$. Since $\vec{M}'$ is normal to $\vec{k}_3$, DFG may occur, and according to Eq. (2), its intensity will vary as $|E_1|^2|E_2|^2H^2$. The effect of H may be quite large—for a magnetic field strong enough to cause level shifts comparable to the hyperfine splitting of either states $|a\rangle$ or $|b\rangle$ (a few hundred gauss in Tl), we expect M' to be comparable to $M_0 = \alpha NE_1E_2$. We speak of the **DFG** as being induced by the static magnetic field, since for the case considered it disappears in its absence. The DFG has the interesting property that its phase depends on the direction of $\vec{H}$; for according to Eq. (2), $\vec{M}'$ reverses phase if $\vec{H}$ is reversed in direction. Although we have described the DFG in terms of $M1$-allowed radiation, the even-parity $E2$ moment will contribute to the radiation as well if it is allowed by the selection rules for the $b\rightarrow a$ transition. Symmetry considerations show that for $E2$ as well, the intensity of the coherent emission is proportional to H^2. The extension to magnetically induced $M1$ or $E2$ SFG is straightforward.

Although the applied magnetic field also polarizes the ground-state Zeeman levels, the resulting difference-frequency magnetization should be of the order of r_1M_0, where $r_1 = \mu_B H/kT$. Here μ_B is the Bohr magneton, T is the temperature, and k is the Boltzmann constant. In the thallium experiment described below ($T \cong 1000°K$, $H < 100$ G), r_1 is no larger than 2×10^{-6}. On the other hand, the hyperfine-mixing effect results in a magnetization of the order of r_2M_0, where $r_2 = \mu_B H/\Delta W_b$ and ΔW_b is the b-state hyperfine splitting. [For thallium we may ignore the a-state ($6^2P_{1/2}$) mixing, since the $6^2P_{1/2}$ hyperfine splitting $\Delta W_a/h \cong 21$ GHz $\gg \Delta W_b/h \cong 500$ MHz.] Thus we obtain $r_2/r_1 \cong 4\times10^4$, and it is clear that the mixing effect dominates over the polarization. It is otherwise for the InSb experiment, in which the polarization caused by the magnetic field is responsible for the $M1$ **DFG**.[3]

The basic experimental setup we have used to observe **DFG** in atomic thallium vapor consists of two dye lasers, an oven-heated fused-silica thallium vapor cell, and a 3-nsec-risetime detection system consisting of a germanium avalanche photodiode and a wideband amplifier. A magnetic field of variable direction is produced in the cell by two orthogonal Helmholtz coils (for the transverse components) and one solenoid (for the longitudinal component). The dye lasers are pumped simultaneously by the same nitrogen laser; one (ω_1) is tuned near the 26 478-cm^{-1} $6^2P_{1/2}$-$7^2S_{1/2}$ transition frequency ω_0, and the other (ω_2) near the 18 685-cm^{-1} $6^2P_{3/2}$-$7^2S_{1/2}$ transition frequency $\omega_0-\omega_{ba}$. Each of the 5-nsec-long, 10-μJ laser pulses has a linewidth of less than 0.3 cm^{-1}. They are focused by a 30-cm-focal-length lens into a volume of diameter $d \cong 0.03$ cm and length $l \cong 3$ cm at the center of the thallium cell. When the frequency difference between the two lasers is adjusted to coincide with the 7793-cm^{-1} $6^2P_{3/2}$-$6^2P_{1/2}$ splitting, a collimated DFG 1280-nm pulse is emitted during the laser excitation along the common direction of propagation of the two beams. This optical pulse is detected efficiently by imaging the center of the thallium cell onto the surface of the Ge detector. The wavelength of the pulse is determined on a grating spectrometer; it must be distinguished from the 1300-nm superradiant burst[8] which occurs under certain conditions as a result of a $7^2P_{1/2}$-$7^2S_{1/2}$ atomic inversion.

The following are the observed DFG properties: (a) For fixed $\vec{H}$, the DFG power varies as N^2 (Fig. 1), as predicted by Eq. (2) when the phase mismatch φ in the wave-mixing region has a negligible effect. Using $\varphi \cong 2\pi c r_e fNl\omega_1(\omega_0{}^2-\omega_1{}^2)^{-1}$, where c is the speed of light, r_e is the classical

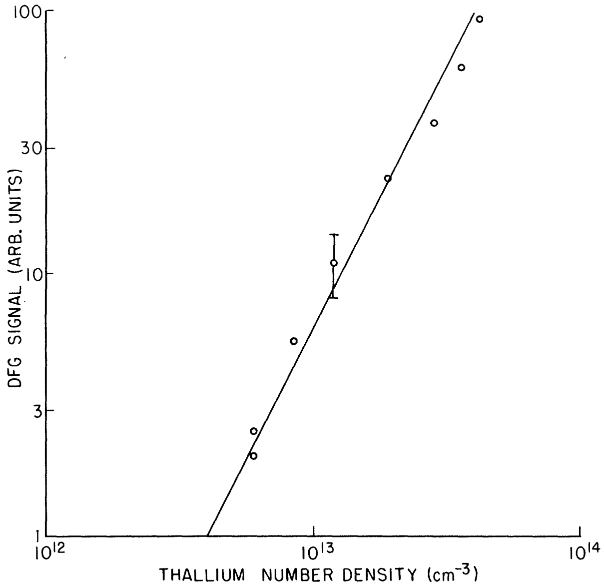

FIG. 1. DFG signal vs. Tl number density N. Conditions: $(\omega_1-\omega_0)/2\pi c=+5\ \mathrm{cm}^{-1}$, $H=90$ G. Laser linewidths and peak powers: $0.03\ \mathrm{cm}^{-1}$ and $\simeq 100$ W, respectively. Vertical scale: each unit $\cong 5\ \mu$W peak DFG power. The straight line drawn has a slope of 2 on the log-log plot. The error bar represents a typical standard deviation of the signal.

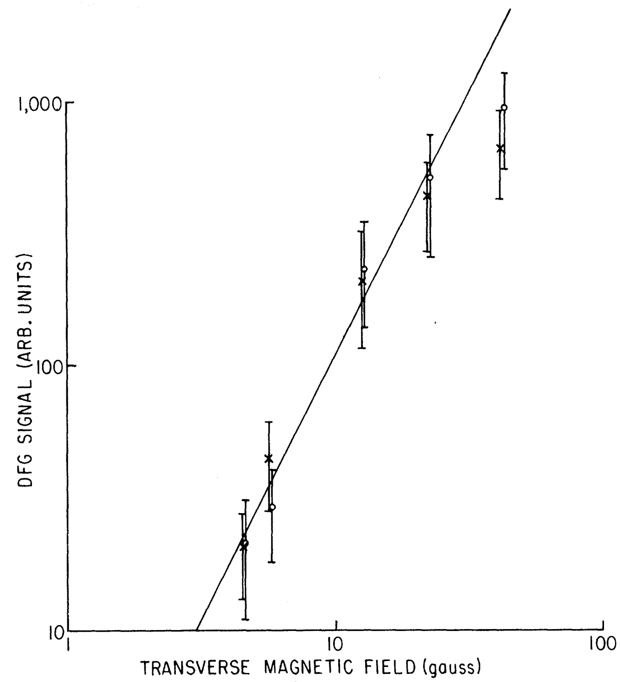

FIG. 2. DFG signal vs H. Conditions: $N=5\times10^{14}$ cm^{-3}, $(\omega_1-\omega_0)/2\pi c=+45\ \mathrm{cm}^{-1}$; the incident lasers have $\cong 1$ kW peak powers, $0.3\ \mathrm{cm}^{-1}$ linewidths, and are linearly polarized normal to each other; and $\vec{H}$ is directed along $\vec{E}_1$. Circles: $H>0$; crosses: $H<0$. The error bars are standard deviations of the data, each obtained from an average of about 15 oscilloscope traces. The straight line is drawn with a slope of 2 on the log-log plot.

electron radius, and $f\cong 0.13$ is the $6^2P_{1/2}$-$7^2S_{1/2}$ oscillator strength,[9] we find $\varphi\cong 0.5$ (and $\sin\varphi\cong\varphi$) for the largest N of Fig. 1.

(b) If $\vec{H}$ is aligned along either $\vec{E}_1$ or $\vec{E}_2$, then the polarization of the magnetic vector associated with the DFG is along $\vec{H}$ if $\vec{E}_1$ and $\vec{E}_2$ are parallel, but normal to $\vec{H}$ if $\vec{E}_1$ and $\vec{E}_2$ are perpendicular. This polarization dependence is predicted by Eq. (2).

(c) For H in a fixed direction transverse to the direction of propagation, the DFG power varies as H^2 (Fig. 2). For sufficiently low incident laser intensities, the DFG power is linear in the intensity of either input laser, in agreement with the $|E_1|^2|E_2|^2H^2$ dependence predicted by Eq. (2).[10] For higher laser intensities, the dependence is modified by the production of stimulated Raman scattering (see below).

(d) From Eq. (2), we expect a DFG peak power $P\cong c|2\pi dk_3 l\,\alpha N E_1 E_2 \mathfrak{M}_{b'b} H/\Delta W_b|^2$. Because of the resonance enhancement we may estimate α by including only the $7^2S_{1/2}$ level in the sum over intermediate states. Using $p_{an}\cong p_{nb}\cong 10^{-18}$ esu cm, $\mathfrak{M}_{ba}\cong\mathfrak{M}_{b'b}\cong\mu_B$, $\gamma_{ba}\cong 9\times10^8\ \mathrm{sec}^{-1}$ (the Doppler-broadened linewidth), we find $\alpha\cong 10^{-23}\ \mathrm{cm}^3/\mathrm{G}$ for the conditions of Fig. 1, and we expect P/N^2 $\cong 10^{-30}\ \mathrm{W\ cm}^6$. This agrees with the experimental value of $3\times10^{-31}\ \mathrm{W\ cm}^6$.

(e) The presence of a static electric field as large as 500 V/cm has no observable effect on the DFG.

(f) No DFG is observed in a longitudinal magnetic field as large as 300 G.

We note that the use of two lasers is not required for the excitation of the $6^2P_{1/2}$-$6^2P_{3/2}$ superposition. For sufficiently large N, a single laser of adequate power and of frequency near ω_0 excites the superposition through the production of forward stimulated Raman scattering of wavelength around 535 nm. We have indeed observed the DFG when only a 378-nm dye laser was used; this process may be viewed as parametric down-conversion. When the incident-laser-beam polarization is linear, the Stokes transition is unpolarized, and the dependence of the DFG intensity on the magnitude of a transverse $\vec{H}$ is qualitatively similar to that shown in Fig. 2. For an incident peak laser power of about 5 kW, $N\cong 10^{17}$ cm^{-3}, $H\cong 100$ G (transverse), and $(\omega_1-\omega_0)/2\pi c$

$\cong 70\ \mathrm{cm}^{-1}$, we obtain about 80 mW at 1280 nm. Under these conditions, the DFG efficiency is considerably reduced because phase matching no longer occurs within the divergence of the stimulated Stokes. ($\varphi \cong 70$ rad for collinear propagation.)

We note that the dependence of the polarization of the DFG on that of the incident laser depends on the type of multipole moment responsible for the DFG. For the $6^2P_{1/2}$-$6^2P_{1/2}$ Tl transition, both $M1$ and $E2$ moments have nonvanishing matrix elements. A theoretical calculation[11] indicates that the transition probability of $M1$ is 35 times larger than that of $E2$. Preliminary measurements of the DFG polarization and power dependence on the incident-laser-beam polarizations is in qualitative agreement with Eq. (2), with $\beta/\delta \cong 2$, $\gamma/\delta \cong -4$. From more careful measurements of the polarization dependence, it should be possible to ascertain the ratio between the $E2$ and $M1$ transition probabilities. This technique should be applicable to other atomic spectra as well.

In conclusion, we have observed DFG in an atomic vapor in the presence of a static magnetic field. Our technique does not require the use of noncollinear propagation to lift the symmetry which ordinarily suppresses three-wave mixing in an isotropic medium. The extension to magnetically induced SFG is straightforward.[12] Finally, we note that we have made preliminary observation of DFG on some $E1$-forbidden transitions in other atomic media: in particular, the 539-nm $7^2S_{1/2}$-$6^2S_{1/2}$ transition of Cs and the 497-nm $6^2S_{1/2}$-$5^2S_{1/2}$ transition of Rb. The physical effect appears to be different from that observed in Tl, however. In particular, the DFG occurs in the absence of a magnetic field, nor is it enhanced in its presence. Further study of these effects is under way.

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[1]A. Javan and A. Szöke, Phys. Rev. A 137, 536 (1965); M. D. Martin and E. L. Thomas, IEEE J. Quantum Electron. QE-2, 196 (1966); K. H. Yang, J. R. Morris, P. L. Richards, and Y. R. Shen, Appl. Phys. Lett. 23, 669 (1973); D. H. Auston, A. M. Glass, and P. LeFur, Appl. Phys. Lett. 23, 47 (1973).

[2]Shaul Yatsiv, William G. Wagner, Gerald S. Pincus, and Fred J. McClung, Phys. Rev. Lett. 15, 614 (1965); J. F. Young, G. C. Bjorklund, A. H. Kung, R. B. Miles, and S. E. Harris, Phys. Rev. Lett. 27, 1551 (1971); A. H. Kung, J. F. Young, G. C. Bjorklund, and S. E. Harris, Phys. Rev. Lett. 29, 985 (1972); P. P. Sorokin, J. J. Wynne, and J. R. Lankard, Appl. Phys. Lett. 22, 342 (1973); R. T. Hodgson, P. P. Sorokin, and J. J. Wynne, Phys. Rev. Lett. 32, 343 (1974).

[3]Van Tran Nguyen and T. J. Bridges, Phys. Rev. Lett. 29, 359 (1972); Terrence L. Brown and P. A. Wolff, Phys. Rev. Lett. 29, 362 (1972).

[4]D. S. Bethune, R. W. Smith, and Y. R. Shen, Phys. Rev. Lett. 37, 431 (1976).

[5]T. Hänsch and P. Toschak, Z. Phys. 236, 373 (1970).

[6]P. S. Pershan, Phys. Rev. 130, 919 (1963).

[7]E. U. Condon and G. H. Shortley, *The Theory of Atomic Spectra* (Cambridge Univ. Press, Cambridge, 1967).

[8]A. Flusberg, T. Mossberg, and S. R. Hartmann, Phys. Lett. 58A, 373 (1976).

[9]A. Gallagher and A. Lurio, Phys. Rev. 136, A87 (1964).

[10]The $|E_1|^2|E_2|^2H^2$ dependence of the DFG intensity distinguishes this effect from the much weaker Larmor precession of longitudinal Zeeman polarization in the presence of a transverse magnetic field. The DFG intensity due to the latter effect, should, as mentioned in Ref. 5, vary as a higher power of the laser intensities. In addition, the latter effect vanishes when $\vec{H}$, $\vec{E}_1$, and $\vec{E}_2$ are parallel, whereas Eq. (2) predicts a nonzero DFG intensity. Indeed DFG is observed when $\vec{H}$, $\vec{E}_1$, and $\vec{E}_2$ are parallel.

[11]B. Warner, Z. Astrophys. 69, 399 (1968).

[12]A. Flusberg, T. Mossberg, and S. R. Hartmann, to be published.