lengths attainable in storage rings would further improve both the linewidth and the average power output.

Because the gain falls at short wavelengths, a higher electron current will be required to support laser operation in the visible and the ultraviolet. Based on the small-signal gain equations,[4,10,11] sufficient current has been stored in existing electron storage rings to sustain laser operation at wavelengths as short as 1200 Å.[9]

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Optical Second-Harmonic Generation in Gases: "Rotation" of Quadrupole Moment in Magnetic Field

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We report optical second-harmonic generation in gases in a transverse dc magnetic field. An electric quadrupole moment of sodium or calcium vapor is induced by a two-photon transition. The effect of the magnetic field is to transform the wave function within both the ground and the excited magnetic sublevels, and to make a new component of the quadrupole moment which can generate a collinear second harmonic. Transverse magnetization is not important in the present case.

Optical second-harmonic generation (SHG) in centrosymmetric media is possible only under certain restricted conditions. The experiments so far reported can be grouped by the conditions employed: (1) The second harmonics from the electric quadrupole moment induced by noncollinear fundamental fields were observed in an anisotropic crystal[1,2]; (2) those from the electric quadrupole and the magnetic dipole interaction were observed on the boundary surfaces of isotropic materials[3,4]; and (3) SHG by a dipole interaction at a noncentrosymmetric surface layer was deduced from an experiment on an adsorbed surface layer.[5] Bethune, Smith, and Shen[6] recently demonstrated sum-frequency generation with a resonant noncollinear excitation in sodium vapor. SHG was not possible in the $3S$-$4D$ transition. Hänsch and Toschek,[7] on the other hand, discussed a collinear three-wave mixing in the background medium with transverse magnetization.

In this Letter we describe an experiment in

which a second harmonic was easily generated from sodium and calcium vapor in a transverse dc magnetic field. It has been shown that, by a resonant two-photon transition, gaseous atoms are brought into a coherent excited state.[8] Let us suppose that this two-photon transition is made from S to D levels by left circularly polarized light. This excitation is then accompanied by a component of electric quadrupole moment. (Magnetic dipole transition is forbidden.) If we assume that the quantization axis z along the direction of the wave, this is expressed by $\vec{Q}(+2)$ since it is associated with the transition $\Delta m = +2$, where m is the magnetic quantum number of the S and D levels. This component of the quadrupole moment cannot emit radiation in the direction of $\pm z$.[9] If, however, we apply a dc magnetic field transverse to this direction, the wave function receives a rotation transformation within both the ground and the excited magnetic sublevels around the magnetic field. This brings about, as a function of time, a new component $\vec{Q}(+1)$ which is the moment associated with the transition $\Delta m = +1$. This component in turn can emit the second harmonic in the direction parallel to the z axis. Note that transverse magnetization is not important in the above SHG process.

The second-harmonic effective dipole moment $\vec{P}^{Q}(2\omega)$ is,[10] to first order in the magnetic field,

$$P_j^{Q}(2\omega) = \sum_{kl} \chi_{zjklx} \circ 2k_z E_k^{(\omega)} E_l^{(\omega)} H_x^{(0)}, \qquad (1)$$

where the magnetic field direction is taken as x axis, or more generally

$$\vec{P}^{Q}(2\omega) = \vec{\chi} E^{(\omega)} \{ 2\vec{k} \cdot (\vec{E}^{(\omega)} \times \vec{H}^{(0)}) \},$$

where ω and $\vec{k} = (0, 0, k_z)$ are the fundamental frequency and wave vector, and $\vec{E}^{(\omega)}$ and $\vec{H}^{(0)}$ are the fundamental electric and the dc magnetic field, respectively. The susceptibility tensor components satisfy the relations $2\chi_{zxxyx} = 2\chi_{zxyxx} = \chi_{zyyyx} = -\chi$. Details of the susceptibility tensor form and its magnitude will be discussed in a later section of the paper for convenience.

We have carried out an experiment to demonstrate the above prediction choosing mainly the two-phonon resonance of the $3^2S_{1/2} - 3^2D_{3/2,5/2}$ transition of sodium. A large quadrupole moment is known for this transition.[11] A flash-lamp–pumped dye laser with two intracavity Fabry-Perot interferometers was used to excite the two-photon transition. The output pulse at 685 Å (spectral width, 0.5 Å; pulse length, 500 nsec) was focused on the sodium vapor in a heat-pipe oven through a 50-cm focal length lens. A dc magnetic field was applied perpendicular to the light beam axis by a Helmholtz coil up to the maximum value of 70 G. On increasing the magnetic field, a signal at the wavelength 3428 Å, which is exactly the second harmonic wavelength, was observed in the direction parallel to the fundamental. We found that the second harmonic (SH) signal intensity increased quadratically with the magnetic field as shown in Fig. 1. The fundamental intensity here was about 3 kW, and that of the SH was about 2 mW at the maximum value of the magnetic field. The tuning of the laser within 1.5 cm^{-1} of the two-photon resonance gave four orders of magnitude enhancement. Phase match-

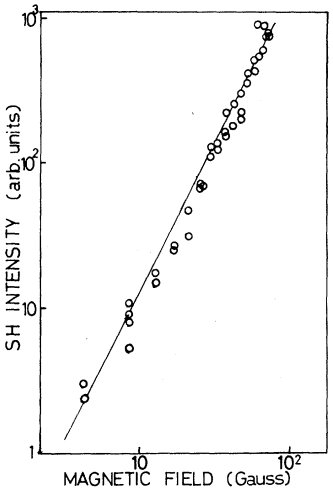

FIG. 1. SH intensity vs magnetic field. Laser intensity, 3 kW (total); sodium number density, 1.2×10^{15} cm^{-3}. The straight line has a slope of 2. The maximum SH power corresponds to 2 mW.

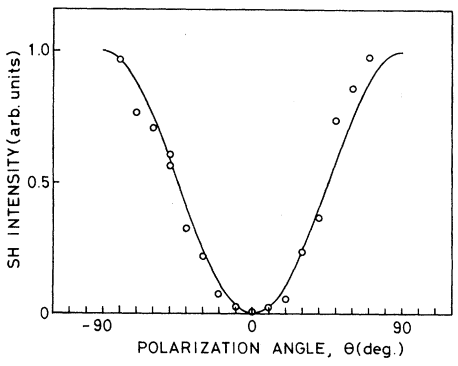

FIG. 2. SH intensity vs angle between laser polarization and magnetic field. Sodium number density, 8.5×10^{14} cm^{-3}; magnetic field, 60 G. The theoretical curve $\sin^2\theta$ is given.

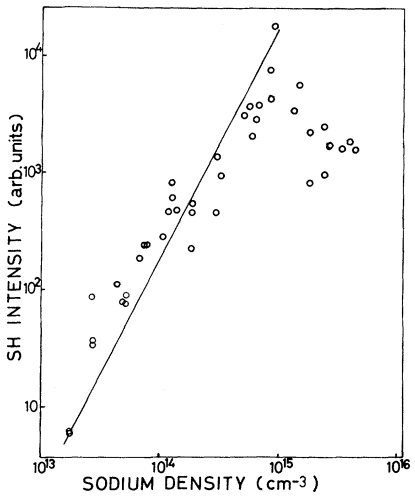

FIG. 3. SH intensity vs sodium number density. Laser intensity, 3 kW (total); magnetic field, 60 G. The straight line has a slope of 2.

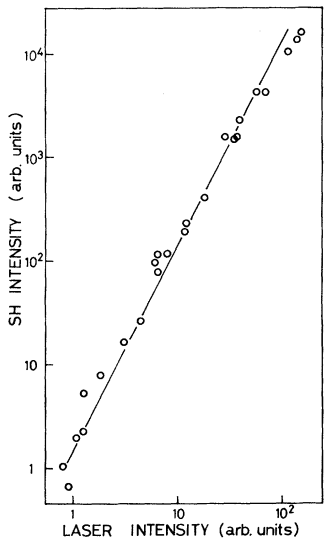

FIG. 4. SH intensity vs laser intensity. Sodium density, 1×10^{15} cm^{-3}; magnetic field, 70 G.

ing is satisfied since the coherence length is 26 cm at the sodium density $N = 10^{15}$ cm^{-3}.

The polarization dependence of the SH was checked next. When a circularly polarized fundamental was used, the SH was also circularly polarized. When the fundamental is linearly polarized at an angle θ from the x axis, SH was also linearly polarized parallel to the fundamental, and its intensity varied as $\sin^2\theta$ as shown in Fig. 2. This is in remarkable agreement with the theoretical prediction for the quadrupole interaction. Furthermore, we examined a dependence of the SH intensity on the atomic number density in order to confirm that the SH is coherent emission from the coherent two-photon excited state. A result is shown in Fig. 3. The SH intensity varies as the square of the density between 10^{13} and 10^{14} cm^{-3} as expected.[12]

A similar experiment was done on the $4s^2\,{}^1S_0$-$4s3d\,{}^1D_2$ transition of calcium.[8] A ruby-laser-pumped, mode-locked dye laser[8] was used in this case. A second harmonic was observed at 4576 Å by a two-photon resonant excitation at 9153 Å. An important point here is that the ground level has no multiplicity, and the excited level has no hyperfine splittings, so that the transformation of the quadrupole moment is possible only by transformation of the wave functions in the magnetic sublevels of $4s3d\,{}^1D_2$.

In order to understand a more detailed mechanism of SHG, we examined the fundamental power dependence and the transient character of SH. We know that a transverse magnetization or a coherent superposition state among the magnetic sublevels of the electronic ground and/or excited levels can be generated as a result of a strong selective excitation of the sublevels by the optical pumping under the transverse magnetic field. If this magnetization were responsible for SHG, the SH intensity should not vary simply as the square of the fundamental intensity, and furthermore, the SH should follow the slowly precessing magnetization (about 50 nsec/rad at 10 G). First, the dependence of the SH intensity on the fundamental was checked, and the result is shown in Fig. 4. SH increases fairly well as the square of the fundamental. Second, an 80-nsec-long rectangular pulse, or two 3-nsec pulses with 10-nsec separation, were used as a fundamental. The SH outputs were confirmed to have no delayed growth in either case.

It is now realized that the principal part of our experiment can be explained by a steady-state analysis for optical transition. Let us denote the ground, the intermediate, and the final levels of the two-photon transition by a, b, and c, and the angular momentum and the magnetic sublevels of level α ($\alpha = a$, b, and c) by J_α and M_α, respectively. When left circularly polarized light $\vec{E} = E[\hat{x}\cos(\omega t - kz) + \hat{y}\sin(\omega t - kz)]$ is incident on the atomic system, matrix elements of the density operator are given by a perturbation approximation in the magnetic field strength H. The off-diagonal elements between levels $|c, M+1\rangle$ and

$|a, M\rangle$, where M takes on proper allowed values for the transition, are given by

$$\rho_{cM+1\,aM}=\frac{\gamma_c H(J_{-1})_{cM+1\,cM+2}\rho_{cM+2\,aM}-\rho_{cM+1\,aM-1}(J_{-1})_{aM-1\,aM}\gamma_a H}{2\hbar(\Omega_{ca}-2\omega-i\Gamma_{ca})}+(\text{other terms}), \tag{2}$$

where Ω_{ca} and Γ_{ca} are the resonance frequencies and the Doppler linewidth of the transition c-a, respectively. In Eq. (2) $\rho_{cM+2\,aM}$ and $\rho_{cM+1\,aM-1}$ are the steady-state two-photon coherences induced by the optical interaction alone. These are transformed to $\rho_{cM+1\,aM}$ by the magnetic interaction; the angular momentum operator $J_{-1}=J_x-iJ_y$, gyromagnetic ratios γ_c and γ_a of the levels c and a, respectively, and the magnetic field H appear as a rotation operator around the x axis. The time allowed to rotate is limited by the frequency denominator in Eq. (2). The other terms in Eq. (2) include those of the two-photon transition starting from the transverse magnetization which is proportional to $\rho_{aM-1\,aM}$ or $\rho_{cM+1\,cM+2}$. These terms are of higher order in the laser intensity,[7] except when $\rho_{aM-1\,aM}$ or $\rho_{cM+1\,cM+2}$ is prepared by any other means. (The preparation by Boltzmann distribution in the transverse magnetic field has negligible effect.) These terms actually have negligible contribution as was confirmed by the experiment. The component $\bar{Q}(+1)$ of the quadrupole moment is obtained from the relation $\bar{Q}=N\,\mathrm{Tr}\{\bar{q}\rho\}$, where $\bar{q}=e\bar{r}\bar{r}$ is the quadrupole moment operator. The expression for the nonlinear current $\bar{j}^{(2\omega)}=\partial^2(Q_{zx}, Q_{zy}, Q_{zz})/\partial z\,\partial t$ gives the susceptibility which conforms with Eq. (1),

$$\chi_{zxxyx}=\frac{N}{8\hbar^3}\sum_{(J_c J_b M)}(q_{-1})_{aM\,cM+1}\{(p_{+1})_{cM+1\,bM}(p_{+1})_{bM\,aM-1}(J_{-1})_{aM-1\,aM}\gamma_a$$
$$-\gamma_c(J_{-1})_{cM+1\,cM+2}(p_{+1})_{cM+2\,bM+1}(p_{+1})_{bM+1\,aM}\}$$
$$\times[(\Omega_{ca}-2\omega-i\Gamma_{ca})^2(\Omega_{ba}-\omega)(2J_a+1)]^{-1}, \tag{3}$$

where $q_{-1}=-e[zx+xz-i(zy+yz)]/2$ is a component of $\bar{q}$, and $p_{+1}=-e(x+iy)$ is a component of the electric dipole moment $\bar{p}$. In the calculation we may neglect hyperfine splittings within the Doppler width $\Gamma_{ca}(=0.05\ \mathrm{cm}^{-1})$. Neglect of the ground-state hyperfine splitting of sodium (0.06 cm^{-1}) also may not affect the essential part of the result.

We can calculate an order of magnitude of $|\chi_{zxxyx}|$ in the case of sodium using the known quadrupole matrix element.[11] For $\Omega_{ca}-2\omega=0$, $\Omega_{ba}-\omega=2400\ \mathrm{cm}^{-1}$, $H=70$ G, $\gamma_c(D_{5/2})=1.68$ MHz/G, $\gamma_c(D_{3/2})=1.2$ MHz/G, and $\gamma_a=2.80$ MHz/G, we find $\chi^{(2\omega)}\equiv 2k|\chi_{zxxyx}|H/N=9.6\times10^{-26}$ esu. If we compare $\chi^{(2\omega)}$ with $\chi^{(3\omega)}$ of the third harmonic generation which is also two-photon resonant to the $3S$-$3D$ transition, we find that these two are of comparable order when $E=20$ esu. The former is also compared to $\chi^{(\omega_1+\omega_2)}\sin\varphi\simeq10^{-25}$ esu of the sum-frequency generation,[6] where $\varphi=24$ mrad is one half of the angle between the noncollinear input beams. (Note that $\chi^{(2\omega)}$ is 240 times unfavored by the large single-photon off-resonance.)

In the present SHG, the rotation of the wave functions in the magnetic sublevels in the presence of the coherent optical quadrupole moment is most essential. This rotation may be regarded as a "rotation" of the second-harmonic quadrupole moment when the gyromagnetic ratios γ_a and γ_c are equal. The "rotation angle" is then $\gamma H\Gamma_{ca}^{-1}$. This leads to H^2 dependence of the SHG. An increase of the magnetic field is desired for the SH efficiency. In a recent experiment we applied about ten times stronger magnetic field, and obtained an enhancement of a factor of several times ten over the maximum value in Fig. 1. A saturation effect is, however, expected for the further increase of the magnetic field, i.e., when $\gamma H\Gamma_{ca}^{-1}\sim1$. Another effect expected is a deviation of the polarization angle of SH from that of the fundamental.

In conclusion, we have shown that the second-harmonic generation in gases is easily observable when an electric quadrupole moment is induced by a two-photon resonance, and the transverse dc magnetic field transforms the quadrupole moment. The effect should become more efficient by an increased magnetic field, or by a longer optical coherence time.

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Note added.—After the manuscript was completed, we noticed that A. Flusberg, T. Mossberg, and S. R. Hartmann [Phys. Rev. Lett. 38, 59 (1977)] reported a difference-frequency generation (DFG) due to a magnetic dipole transition. They explained the effect by a mixing of hyperfine split levels by an external magnetic field. Our

treatment of SHG, which should be applicable also to the DFG case, is that the initially degenerate magnetic sublevels are mixed by the external magnetic field. The mixing of hyperfine split levels is of secondary importance. The example of calcium is important in this respect.

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Evidence of Parametric Decay and Brillouin Backscatter Excited by a CO_2 Laser

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Experimental measurements of the scattered infrared emission in the vicinity of 10 μm are presented. The shape and power dependence of the backscattered spectrum indicates the presence of the Brillouin backscatter instability. The sidebands on the side-scattered radiation are found to be strongest for emission perpendicular to the electric field vector of the incident radiation. This is interpreted as being the result of the parametric decay instability.

The interaction of high-power laser radiation with plasmas is of current interest because of possible use of such lasers in thermonuclear fusion. The absorption or reflection mechanisms, that are important aspects of laser-target interactions, have generated a considerable number of theoretical papers. Experimentally, the existence of nonlinear effects in plasmas has be surmised from plasma emission at $\frac{3}{2}$ and 2 times the 1.06-μm frequency of glass lasers and at second and third harmonics for CO_2.[1-6] Although studied in considerable detail in the interaction of microwaves with plasma,[7,8] this is the first experiment that shows convincing evidence of the parametric decay instability in laser produced plasma. Elegant evidence of stimulated backscatter has been obtained by Ripin *et al.*,[9] as determined by the rapid rise and directional properties of the reflected radiation. We also present new measurements on the existence and effect of the Brillouin backscatter instability.

The CO_2 laser used in these experiments consisted of a mode-locked oscillator, optical gate, and four stages of amplification.[10] It produced 5 J in a 1.5-ns pulse. The laser beam was focused by an off-axis parabolic mirror, equivalent to $f/2$ optics, to fluxes up to 10^{13} W/cm^2. The scattered spectra was obtained with a 1-m grating spectrograph and a HgCdTe detector with a rise time of 1 ns. A schematic of the experimental setup is shown in Fig. 1. The target consisted of a ribbon of 150-μm-thick polyethylene. In order to ensure that the spectral structure observed was not inherent to the laser beam, the wavelength spectra of the incident laser beam and that of the scattered signal were taken simultaneously on several runs. It should be pointed out that the spectra presented were taken on a shot-to-shot basis and that the signal was time-integrated during the CO_2 laser pulse (the rise time of the detector is about equal to the laser pulse length).

Results of the backscatter experiments are shown in Fig. 2. Although a similar spectrum