Spontaneous-Field-Induced Optical Second-Harmonic Generation in Atomic Vapors

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It is shown that optical second-harmonic generation is possible in dispersive atomic vapors since a static electric field is spontaneously created by the fundamental laser beam. When multiphoton ionization of atoms occurs simultaneously in the interaction, the spontaneous field and second-harmonic generation are significantly enhanced. Our experimental results for atomic sodium vapor irradiated by picosecond Nd-doped yttrium-aluminum-garnet laser pulses agree well with the theoretical predictions.

Optical second-harmonic generation (SHG) or three-wave mixing in atomic vapors and gases has recently been studied extensively.[1,2] Under the electric dipole ($E1$) approximation, SHG in centrosymmetric media is strictly forbidden by parity conservation or symmetry. However, the application of an external electric field to the medium makes is possible to observe SHG due to $E1$ interaction. This process is known as electric-field-induced SHG[3-5] and was demonstrated in atomic gases by several authors.[1] On the other hand, if higher-order electric-quadrupole or magnetic-dipole moments are taken into account in atomic nonlinear susceptibilities,[6] SHG is also possible under certain restricted conditions.[2]

In this paper, we present theory and experiment to show that a laser pulse with a spatial intensity gradient irradiating a dispersive atomic vapor induces a static electric field and generates second harmonic (SH) in the atomic medium due to the $E1$ interaction. In this case, the presence of any external field applied to the medium or any resonance condition is not necessary. It is also shown that multiphoton ionization of atoms, which is inevitably brought about by the irradiation with an intense laser pulse,[7,8] increases the spontaneous electric field and resulting SH radiation.

Let us consider an isotropic and nonabsorptive atomic vapor with dielectric constant $\epsilon(\omega)$. The atomic vapor is irradiated by a short laser pulse with some spatial and temporal field distributions. The laser frequency ω is assumed to be far from any resonance of the atomic transitions. It is our first purpose to derive static fields generated in the medium during the laser pulse. The static fields may be introduced into the ordinary quantum-mechanical perturbation theory to describe nonlinear optical processes.[4,5] The starting point of theory is a momentum conservation equation per unit volume of the system concerned. This equation gives a radiation force $\vec{f}$ working on a unit volume of the medium. Obviously, one has to consider two time scales for the quantities of interest[6]: One is a fast time scale determined by the laser frequency, and the other is a slow time scale, that on which the temporal history of a laser pulse changes. To find the static fields induced on the slow time scale, we take an average of the quantities over a cycle of field oscillation. The cycle-averaged form of the equation[9] may be written as

$$\langle\vec{f}\rangle = -\nabla\langle\overleftrightarrow{\sigma}\rangle + \partial\langle\vec{g}\rangle/\partial t, \tag{1}$$

where the brackets $\langle\ \rangle$ mean "cycle averaged," $\overleftrightarrow{\sigma}$ is the Maxwell stress tensor associated with the high-frequency electromagnetic fields $\vec{E}$ and $\vec{B}$, which denotes radiation pressure per unit volume, and $\vec{g} = (\vec{E}\times\vec{B})/4\pi c$ is the field momentum density. The stress tensor in a dielectric medium[9] of which $\epsilon - 1$ is proportional to the atomic or electric-dipole number density N_0 is given by

$$\overleftrightarrow{\sigma} = (8\pi)^{-1}(E^2+B^2)\overleftrightarrow{I} + (4\pi)^{-1}(\epsilon\vec{E}\vec{E} + \vec{B}\vec{B}), \tag{2}$$

where $\overleftrightarrow{I}$ is the unit tensor. The static electric field $\langle\vec{E}_s\rangle$ induced in the medium may be derived from Eqs. (1) and (2) by the relation $\langle\vec{f}\rangle = -eN_0\langle\vec{E}_s\rangle$. When some anisotropy in $\langle\vec{E}_s\rangle$ is produced, we have a static magnetic field $\langle\vec{B}_s\rangle$ from $\nabla\times\langle\vec{E}_s\rangle$.

We assume that the incident laser beam with lowest-order (TEM_{00}) Gaussian distribution of field is linearly polarized along the x direction and weakly focused at the center ($z = 0$) of the atomic vapor with a confocal beam parameter b. In the propagating region $|z| \lesssim b$ of the focused beam, the incident field can be approximated by a plane wave with only radial field distribution. In this region, since $B^2 = \epsilon E^2$ and $\langle g\rangle = I/c^2$, Eqs. (1) and (2) reduce to

$$\langle\vec{f}\rangle = \nabla(I/2nc)[(1-\epsilon)\overleftrightarrow{I} + 2\epsilon\vec{k}\vec{k}] + \vec{k}(1/c^2)\partial I/\partial t, \tag{3}$$

where $I=nc\langle E^2\rangle/4\pi$ is the laser intensity in the medium with refractive index $n=\sqrt{\epsilon}$, and $\vec{k}$ is a unit vector along the z direction.[10] In the derivation of $\langle\vec{E}_s\rangle$, we take into account multiphoton ionization of atoms, which readily occurs in atomic vapor irradiated by an intense laser pulse and exerts a large effect on the dielectric constant of the medium.[7] For an atomic vapor consisting of one-electron atoms such as alkali-metal atoms, the dielectric constant including the ionization effect[7,11] is expressed as

$$\epsilon(\omega)=1-4\pi e^2N_e/m\omega^2 + (4\pi e^2N/m)\textstyle\sum_i[f_i/(\omega_i^2-\omega^2)], \quad (4)$$

ignoring the contribution of ionic electrons to ϵ, where N_e and N are the electron and ground-state-atom densities, respectively, and f_i is the oscillator strength for the transition i with excitation frequency ω_i.[12] Since the initial atomic number density is $N_0=N+N_e$ and $N=N_0\exp(-F)$ in multiphoton ionization,[7] we have the form for a component of $\langle\vec{E}_s\rangle$ in cylindrical coordinates,

$$\langle E_s\rangle_r=\frac{2\pi e}{mc}\left[\sum_i\frac{f_i}{\omega_i^2-\omega^2}(1-KF)\exp(-F) - \frac{1}{\omega^2}\{1-(1-KF)\exp(-F)\}\right]\frac{\partial I}{\partial r}, \quad (5)$$

with a similar nonzero component $\langle E_s\rangle_z$ and $\langle E_s\rangle_\theta=0$, where $F=\int_{-\infty}^{t}W\,dt$ is concerned with the degree of ionization at time t which is characterized by the multiphoton ionization probability W at ω for a specific kind of atoms,[7] and K is the minimum number of incident laser photons necessary to ionize a single atom. From $\nabla\times\langle\vec{E}_s\rangle$, we have $\langle\vec{B}_s\rangle$ of the form

$$\langle B_s\rangle_\theta=\frac{4\pi e}{mc}\sum_i\frac{f_i\omega_i^2}{(\omega_i^2-\omega^2)^2}(1-KF) \times\exp(-F)\frac{\partial I}{\partial r}, \quad (6)$$

and $\langle B_s\rangle_r=\langle B_s\rangle_z=0$. By setting $F=0$ in Eqs. (5) and (6), one obtains the forms of $\langle\vec{E}_s\rangle$ and $\langle\vec{B}_s\rangle$ in a pure atomic vapor without charged species, and for $F\to\infty$, those in a fully ionized plasma. Note that the spontaneous fields generated do not depend on the particle densities, and that $\langle\vec{B}_s\rangle$ originates only from the atomic dispersion.

Thus we can expect to observe SHG induced by the spontaneous fields in an atomic vapor. In the off-resonant case of interest, it should be the SHG induced by $\langle\vec{E}_s\rangle$, what is called electric-field–induced SHG, that we can observe most easily. The effective second-harmonic (SH) dipole moment $\vec{P}_{2\omega}$ in this process[4,5] is associated with $\langle\vec{E}_s\rangle$ and the incident high-frequency field $\vec{E}$ according to

$$\vec{P}_{2\omega}=\chi^{(3)}\langle\vec{E}_s\rangle\vec{E}\vec{E}, \quad (7)$$

where $\chi^{(3)}$ is the effective third-order nonlinear susceptibility. Equations (5) and (7) show that only the radial component of the dipole moment can emit SH radiation to be observed in the z direction. Therefore, the SH radiation predicted is polarized in the radial direction, and it vanishes at the beam center ($r=0$) because of its radial polarization and of the form of $\partial I/\partial r\propto rI$ in Eq. (5) for a Gaussian beam. From Eq. (7), the local and temporal SH intensity $I_{2\omega}$ generated along the z direction is then given by

$$I_{2\omega}\propto\chi^2N^2\langle E_s\rangle_r^2I^2, \quad (8)$$

where χ is the effective nonlinear susceptibility per atom, and we have set $\chi^{(3)}=\chi N$ for an atomic vapor.[4]

To demonstrate the SHG process predicted above, we have made an experiment using atomic Na vapor and a mode-locked Nd-doped yttrium-aluminum-garnet laser system ($\lambda=1.064\ \mu$m;

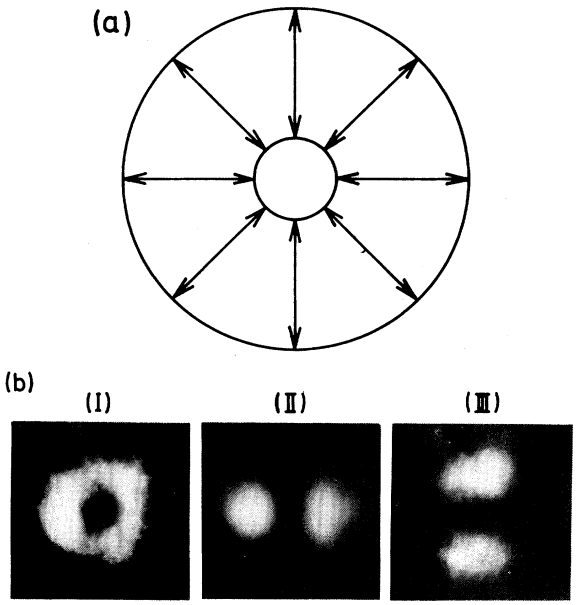

FIG. 1. (a) Scheme of the SH polarization. (b) Photographs of the SH beam pattern observed (I) with no polarizer and behind (II) horizontal and (III) vertical polarizers. Each photograph was taken by about 100 superimposed shots of laser (with horizontal polarization) at $(2-3)\times10^{12}$ W/cm^2.

pulse duration, 28 psec; output energy $\lesssim 50$ mJ) which has been described in detail elsewhere.[7] The measured spatial distribution of the laser beam was in good agreement with a Gaussian profile. The laser beam was focused by a lens with focal length 46 cm into a heat-pipe–type oven containing Na vapor with pressure range from 0.1 to 1 Torr. Above the laser intensity of about 5×10^{11} W/cm^2 where Na atoms would be partially ionized through a multiphoton process,[7] the SH radiation at $\lambda = 532$ nm could be observed. The SH light was monitored photographically and by a detection system composed of a 30-cm spectrometer and an optical multichannel analyzer (OMA). The SH signal of the OMA was integrated during the laser pulse to give the relative SH energy.

First, we observed photographically the polarization and spatial pattern of the SH beam which show the most distinctive properties to confirm the theoretical predictions. The results are shown in Fig. 1. For an incident linearly polarized beam, the SH light was found to be linearly polarized in the radial direction. At the same time, the SH radiation was found to vanish at the center ($r = 0$) of the beam: A ringlike spatial pattern of the SH was observed.[13] These facts observed are in perfect agreement with the theoretical predictions for the SHG expected.

Next, we measured a dependence of the SH signal $S_{2\omega}$ on initial Na density N_0. Since $\langle E_s\rangle_r$ in Eq. (8) does not depend on N_0, the SH signal should be proportional to N^2, i.e., to N_0^2, at a fixed fundamental intensity when phase matching is satisfied. The coherence length in the present case is only 6 cm at the initial Na density 5×10^{15} cm^{-3}. However, one can expect in the Na vapor partially ionized during the laser pulse that the coherence length becomes much longer,[7] and then the phase matching is satisfied over the effective interaction region and duration for the SHG. Therefore, the measurement was performed at a high input intensity, and the result is shown in Fig. 2. The SH signal observed increases quadratically with N_0 over the Na density region, con-

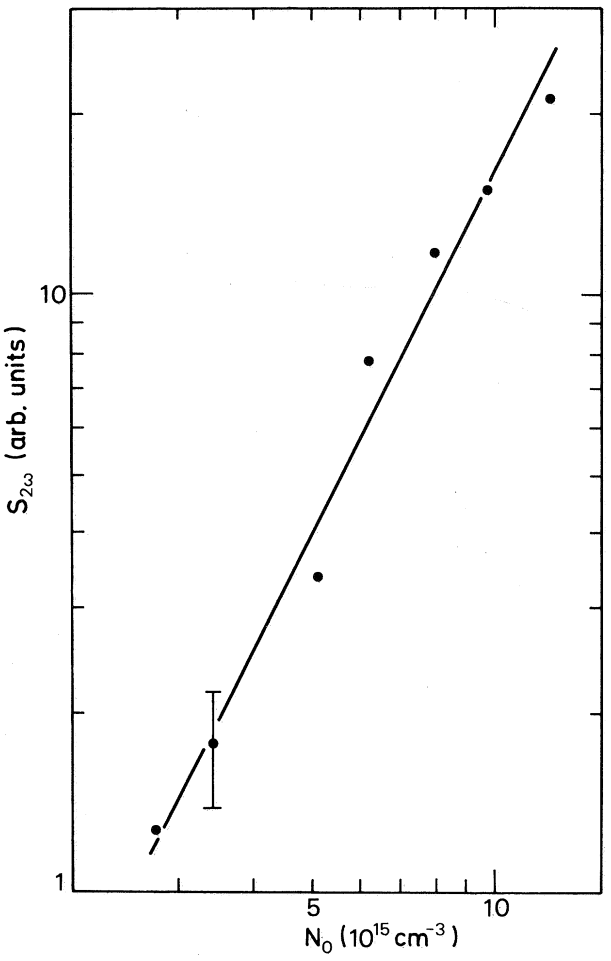

FIG. 2. SH signal vs initial Na density. Laser intensity $I_0 = 2.8\times10^{12}$ W/cm^2. The solid line has a slope of 2.

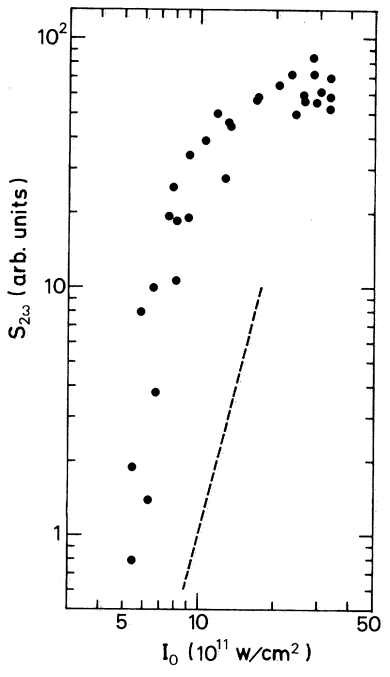

FIG. 3. SH signal vs laser intensity. Initial Na density $N_0 = 5.3\times10^{15}$ cm^{-3}. The dashed line with a slope of 4 is indicated for comparison.

sistent with the above prediction.

We also measured a dependence of the SH signal on the fundamental peak intensity I_0, and the result is shown in Fig. 3. The dependence observed has a slope steeper than 4 (a dashed line indicated in Fig. 3) at relatively low input intensities. This can be explained qualitatively as follows. If there is no ionization of atoms in the Na vapor, $\langle E_s \rangle_r \propto I$ with $F = 0$ in Eq. (5). Then Eq. (8) predicts a fourth-power dependence of the SH intensity or energy on I. On the other hand, when the fundamental pulse causes multiphoton ionization of atoms, $\langle E_s \rangle_r^2 N^2/I^2$ is an increasing function of F, i.e., I, in the region of small value of F. Thus Eqs. (5) and (8) suggest a dependence of $I_{2\omega}$ on I steeper than a slope of 4.[14]

In the present experiment, the maximum value of $\langle E_s \rangle_r$ estimated at $I_0 = 3.0 \times 10^{12}$ W/cm^2 is about 50 V/cm. In this paper, we have neglected secondary processes such as macroscopic current and charge separation between electron and ion distributions in the ionized vapor, because of the short interaction time. However, if a long laser pulse ($\gtrsim 1$ nsec) is used in the experiment, the secondary processes may enhance the spontaneous electric field and the SH radiation. Also, it may be possible to observe another type of SHG[2,6] due to presence of the static magnetic field $\langle B_s \rangle_0$.

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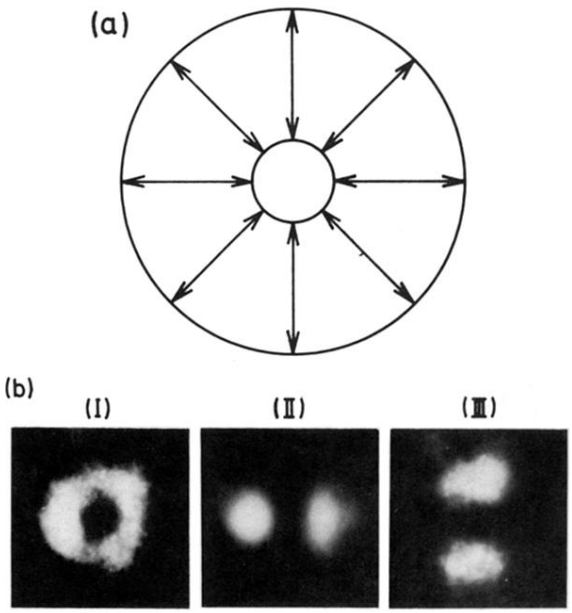
[11]See, e. g. J. B. Gerardo, J. T. Verdeyen, and M. A. Gusinow, J. Appl. Phys. 36, 3526 (1965).

[12]The dispersion terms in Eq. (4) are several orders of magnitude less than unity under the conditions of interest. This assumed in the subsequent part of this paper.

[13]No apparant dependence of the SH signal on angle between the fundamental and SH polarizations was observed.

[14]The saturation of $S_{2\omega}$ in Fig. 3 can be explained by the rapid decrease of Na atoms during the intense laser pulse. See Ref. 7.



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