Electric-Field-Induced Second-Harmonic Generation with Reduced Absorption in Atomic Hydrogen

K. Hakuta,[a] L. Marmet, and B. P. Stoicheff
Ontario Laser and Lightwave Research Centre and Department of Physics, University of Toronto, Toronto, Ontario, Canada M5S 1A7
(Received 6 August 1990)

We show that dc-electric-field coupling of the $2s$ and $2p$ states in atomic hydrogen leads to resonantly enhanced second-order susceptibility with reduced absorption at the second-harmonic wavelength, and exact phase matching at the center of the Stark-split components.

PACS numbers: 42.65.Ky, 32.70.Jz, 32.80.−t

Recently, Harris, Field, and Imamoglu[1] proposed a method for obtaining high conversion efficiency in four-wave frequency mixing by making use of the Autler-Townes effect. The basis of their method is the application of a strong-coupling field between metastable and upper states to resonantly enhance the nonlinear susceptibility of the medium, and at the same time to induce transparency and a zero in the contribution of the resonance transition to the refractive index. In the present Letter, we demonstrate that similar characteristics may be realized with dc-electric-field-induced second-harmonic generation (SHG) in atomic hydrogen for the $n=2\rightarrow 1$ transition.

In the present example with a dc electric field, the process of SHG may be described in terms of the nonlinear susceptibility $\chi^{(3)}(-2\omega;\omega,\omega,\omega'=0)$, or with a $\chi^{(2)}(-2\omega;\omega,\omega)$ formulation, by incorporating the dc electric field in the atomic wave functions, that is, by using Stark-mixed eigenfunctions. We have used the latter method with $\chi^{(2)}$ expressed as[2]

$$\chi^{(2)}(-2\omega_a;\omega_a,\omega_a)=\frac{N}{2\hbar^2\epsilon_0}\sum_{i,j}\frac{\langle 1|\mu|i\rangle\langle i|\mu|j\rangle\langle j|\mu|1\rangle}{(\Omega_i-\omega_a)(\Omega_j-2\omega_a)}\,. \tag{1}$$

Here N denotes the density of hydrogen atoms. The states included in Eq. (1) are shown in the energy-level diagram[3] of Fig. 1. For the ground state, a field-free wave function $|1s\rangle$ was used since the Stark effect is negligible. The excited states $|2\rangle$ and $|3\rangle$ are the Stark-mixed states with $m=0$, which are expressed by linear combinations of field-free wave functions $|2s\rangle$ and $|2p\rangle$ as

$$\begin{aligned}|2\rangle&=\cos\theta|2s\rangle+\sin\theta|2p\rangle\,,\\ |3\rangle&=-\sin\theta|2s\rangle+\cos\theta|2p\rangle\,,\\ \tan 2\theta&=2\langle 2s|\mu|2p\rangle E_a/\Delta\Omega\,,\end{aligned} \tag{2}$$

where E_a denotes the value of the applied dc field, the dipole moment $\langle 2s|\mu|2p\rangle$ is $3ea_0$ or 7.6 D, and $\Delta\Omega$ is the energy splitting between the $2p$ and $2s$ states. With pump polarization perpendicular to the dc field, and by summing over the states $|2\rangle$ and $|3\rangle$, Eq. (1) becomes

$$\chi^{(2)}=\frac{NA}{4\hbar^2\epsilon_0\omega_a}\left[\frac{1}{\Omega_2-2\omega_a}+\left(\frac{-1}{\Omega_3-2\omega_a}\right)\right]\times\langle 2s|\mu|2p\rangle|\langle 1s|\mu|2p\rangle|^2\,. \tag{3}$$

Here

$$A^2=\sin^2 2\theta=\frac{4|\langle 2s|\mu|2p\rangle|^2E_a^2}{\Delta\Omega^2+4|\langle 2s|\mu|2p\rangle|^2E_a^2} \tag{4}$$

describes the Stark mixing of the $2s$ and $2p$ states by varying E_a. The same expression for $\chi^{(2)}$ is obtained for pump polarization parallel to the dc field. In Eq. (3), the complex quantities Ω_2 and Ω_3 denote the energies of Stark-split levels and include linewidths $\Gamma_2=\Gamma_{2p}\sin^2\theta$ and $\Gamma_3=\Gamma_{2p}\cos^2\theta$, where Γ_{2p} is the spontaneous decay rate of the $|2p\rangle$ state.

In the present three-wave mixing process, the mixed states $|2\rangle$ and $|3\rangle$ couple with state $|1\rangle$ through two-photon absorption and one-photon emission processes, and this coupling leads to a contribution to $\chi^{(2)}$ as a

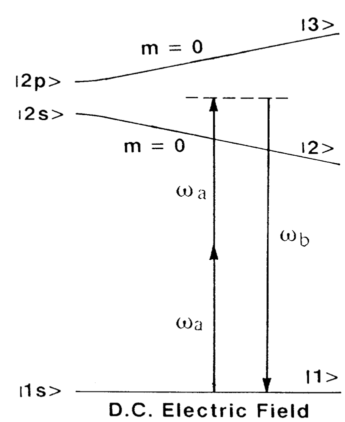

FIG. 1. Energy levels of atomic hydrogen involved in SHG induced by a dc field. States $|1\rangle$, $|2\rangle$, and $|3\rangle$ are the eigenstates under the dc field, with states $|2\rangle$ and $|3\rangle$ expressed by linear combinations of zero-field eigenstates $|2s\rangle$ and $|2p\rangle$.

cross term of eigenvector components. From Eq. (3), it is seen that $\chi^{(2)}$ is the sum of two terms, namely, the contributions from two paths via states $|2\rangle$ and $|3\rangle$. Since these cross terms have different signs for the paths via states $|2\rangle$ and $|3\rangle$, the minus sign in Eq. (3) results in the constructive interference in the nonlinear susceptibility predicted by Harris, Field, and Imamoglu.[1]

Since the second-harmonic radiation has polarization parallel to the dc field for both parallel and perpendicular pump schemes, the susceptibility $\chi^{(1)}$ which characterizes absorption and dispersion for the SH radiation has contributions from the Stark-mixed states $|2\rangle$ and $|3\rangle$ with $m=0$. It should be noted that this situation for $\chi^{(1)}$ is the same as that of Ref. 1 which involves electromagnetically induced transparency. The only difference is that in the present case the dc field (radiation with zero frequency) strongly mixes the metastable $|2s\rangle$ state with the state $|2p\rangle$ of the allowed transition. As discussed more fully recently,[4-6] the induced transparency has its physical origin in the interference due to spontaneous decay of two closely spaced states.[7] We have calculated $\chi^{(1)}$ for two closely spaced Stark-mixed states using the formulation of Ref. 7 and obtain

$$\chi^{(1)}=\frac{N}{\hbar\epsilon_0}\left\{\frac{\Delta\omega_{21}\Delta\omega_{31}(\Delta\omega_{31}\sin^2\theta+\Delta\omega_{21}\cos^2\theta)}{(\Delta\omega_{21}\Delta\omega_{31})^2+\Gamma_{2p}^2(\Delta\omega_{31}\sin^2\theta+\Delta\omega_{21}\cos^2\theta)^2}+i\frac{\Gamma_{2p}(\Delta\omega_{31}\sin^2\theta+\Delta\omega_{21}\cos^2\theta)^2}{(\Delta\omega_{21}\Delta\omega_{31})^2+\Gamma_{2p}^2(\Delta\omega_{31}\sin^2\theta+\Delta\omega_{21}\cos^2\theta)^2}\right\}|\langle 1s|\mu|2p\rangle|^2. \quad (5)$$

Here $\Delta\omega_{21}$ and $\Delta\omega_{31}$ denote the detuning of SH radiation for transitions between the ground state $|1\rangle$ and excited states $|2\rangle$ and $|3\rangle$, respectively. Since $\Delta\omega_{31}\sin^2\theta+\Delta\omega_{21}\times\cos^2\theta=0$ at the midpoint of the Stark-split levels, the medium becomes perfectly transparent to the SH radiation at $\omega_b=2\omega_a$. The dispersion also becomes zero at the midpoint. This distinct behavior between $\chi^{(2)}$ and $\chi^{(1)}$ is the origin of the resonantly enhanced second-harmonic generation with reduced absorption.

The behavior of $\chi^{(2)}$ and $\chi^{(1)}$ was numerically calculated by including the Doppler velocity distribution of H atoms. The position of the $|2s\rangle$ state was treated as that of the $2^2S_{1/2}$ state, and the position of $|2p\rangle$ was treated as the midpoint of the $2^2P_{3/2}$ and $2^2P_{1/2}$ states; that is, the $|2p\rangle$ state is located 0.165 cm^{-1} ($\Delta\Omega$) above the $|2s\rangle$ state. This approximation is good for fields higher than 4 kV/cm, where the Stark energy is sufficiently large compared to the spin-orbit interaction.

Figures 2(a)-2(c) show the tuning characteristics of $|\chi^{(2)}|^2$, Im$\chi^{(1)}$, and Re$\chi^{(1)}$ for various dc electric fields. It is clear that generation of SH radiation given by $|\chi^{(2)}|^2$ has a completely different behavior from the absorption of generated SH radiation, given by Im$\chi^{(1)}$. With increasing dc field, $|\chi^{(2)}|^2$ grows up to a field of 5 kV/cm, and the tuning curve splits into two components above fields of ~9 kV/cm. On the other hand, the peak of Im$\chi^{(1)}$ decreases with increasing field, and the absorption profile begins to split into two peaks above ~5 kV/cm. We draw special attention to the characteristics of these parameters at zero detuning, namely, at the center of the Stark-split components, for high dc fields. At 13 kV/cm, it is seen that $|\chi^{(2)}|^2$ still remains at ~50% of the peak value of the components, while Im$\chi^{(1)}$ decreases to ~3%, which is limited by residual absorption due to the Doppler tail. That is, the atomic medium becomes almost totally transparent at the center while maintaining an appreciable (resonantly enhanced) nonlinear susceptibility. Moreover, the resonance contri-

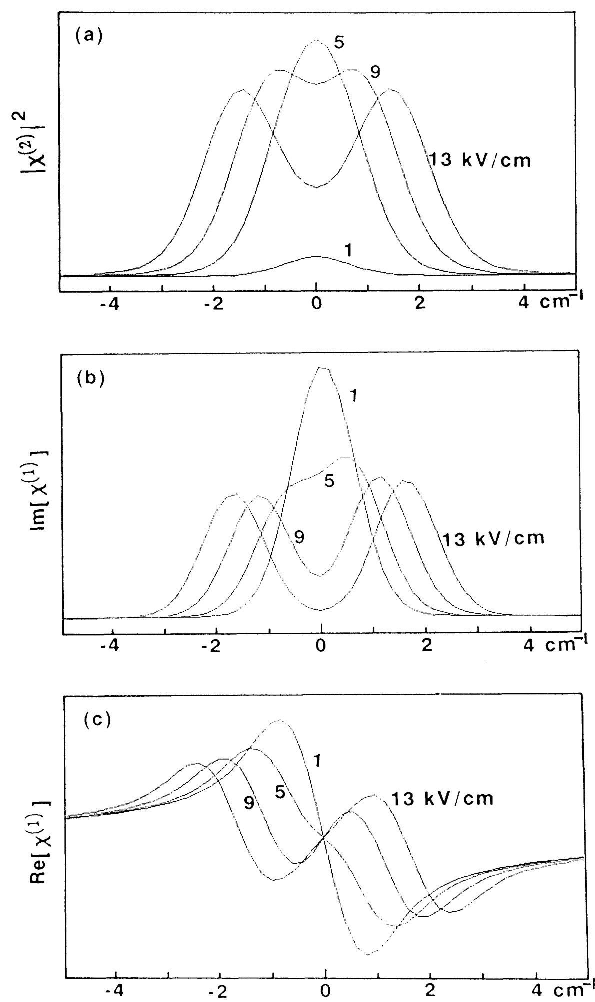

FIG. 2. Calculated characteristics of $|\chi^{(2)}|^2$ and $\chi^{(1)}$ for various dc fields as a function of detuning from the center of the Stark components.

bution to the refractive index, $\mathrm{Re}\chi^{(1)}$, takes a zero value at the center, thus satisfying the phase-matching condition. These results clearly demonstrate that the dc-field-induced SHG in atomic hydrogen may provide ideal conditions for high conversion efficiency at the center of the Stark-split components of the $n=2 \rightarrow 1$ transition.

We now discuss the results of recent experiments carried out to examine these theoretical predictions. The experimental arrangement is outlined here, with a detailed description to be given elsewhere.[8] A frequency-doubled dye laser was used to generate UV laser radiation with peak power of ~ 20 kW, pulse duration of 6 ns, and spectral width of 0.2 cm^{-1} (FWHM). Hydrogen atoms generated in a dc glow discharge of H_2 gas were diffused vertically through a nozzle of 0.5 mm diameter into a vacuum chamber maintained at 2×10^{-6} Torr. A dc electric field (from 0 to 13 kV/cm) was applied to the cw hydrogen beam, at the nozzle, by means of two horizontal-plate electrodes having a separation of ~ 2.0 mm. The UV laser beam was focused midway between the electrodes (~ 1.0 mm from the nozzle). Second-harmonic radiation was detected by a solar-blind photomultiplier (EMR 510G-08-17) after being dispersed relative to the incident beam by a monochromator (McPherson 225). Measurements of SH intensity were carried out under conditions of relatively low atomic density ($\sim 2\times10^{13}$ cm^{-3}) and short interaction length (1.5 mm) in order to obtain the behavior of $|\chi^{(2)}|^2$ directly, from the tuning characteristics of the SH intensity. The effects of absorption and phase mismatch were estimated to be, at most, 8% and 11%, respectively, for the whole tuning range (~ 10 cm^{-1}) of the measurements.

An important consideration in this series of experiments was how to monitor the absorption characteristics for the $n=2 \rightarrow 1$ transition. We have observed the absorption simultaneously with SH emission by monitoring the photoion current produced by three-photon ionization. Since the states $|2\rangle$ and $|3\rangle$ are mixed states of the $|2s\rangle$ and $|2p\rangle$ states which have different parity, it is possible to observe the absorption characteristics in two ways, by one-photon and two-photon processes. The one-photon absorption monitors the $|2p\rangle$ part of the mixed states, and the two-photon absorption monitors the $|2s\rangle$ part. As already mentioned, the two-photon absorption was readily obtained by measuring the ion current produced by two-photon-resonant three-photon ionization.

The tuning characteristics of the SH intensity and ion current at two dc fields are presented in Figs. 3(a) and 3(b) for pump polarization perpendicular to the dc field. For a field of 5.2 kV/cm, a smooth tuning curve is obtained for SH intensity while the tuning curve for ions clearly shows a shoulder on the high-frequency side. These features were explained by the calculated results, as shown by a comparison with Figs. 2(a) and 2(b), except that the shoulder of absorption appears on the opposite side from that of the calculated curve since the ion signal monitors the $|2s\rangle$ part of the mixed states. At 12.4-kV/cm field strength, both tuning curves split into two components: The SH signal midway between the components has an intensity of about half the peak value; on the other hand, the ion (or absorption) signal at the center drops to 6% of the peak value.[9] It is important to mention that the ion signal may have been overestimated, because the slightest elliptical polarization in the incident laser beam induces absorption to the $m=\pm 1$ states (located at the center of the Stark-split $m=0$ states) and produces a background absorption between the Stark-split components. For the present experiments, this background absorption has been estimated to be 3%. When these contributions to the ion signal are included, there is good agreement between the observed and calculated tuning curves.

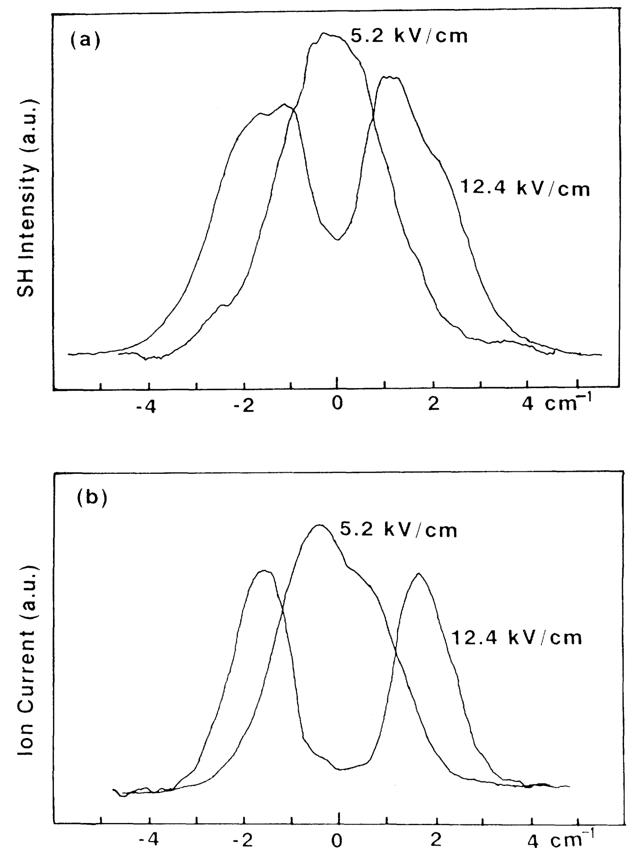

FIG. 3. Observed characteristics of SH intensity and ion current for two dc fields as a function of detuning from the center of the Stark components.

Figure 4 shows the dependence of the SH intensity on dc electric field as measured at the center of the two Stark components, for comparison with calculated values

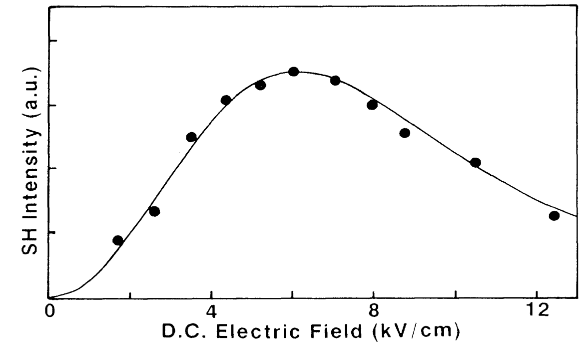

FIG. 4. SH intensity at the center as a function of dc field. The circles are the observed intensities, and the solid line denotes the theoretical dependence calculated from $|\chi^{(2)}|^2$.

of $|\chi^{(2)}|^2$. The agreement is excellent. It is noted that the SH intensity reaches saturation at $\sim$6 kV/cm, while Stark mixing is estimated from Eq. (4) to fully saturate at a low field of $\sim$3 kV/cm. The observed dependence of the SH intensity on applied field results as a natural consequence of the constructive interference in $\chi^{(2)}$ at the center. That is, the minus sign of Eq. (3) results in a cancellation of contributions from Lorentzian terms of states $|2\rangle$ and $|3\rangle$ for $\chi^{(2)}$ at the center. Moreover, $\chi^{(2)}$ at the center is expressed by a Doppler integral of the dispersion term which has a completely different dependence on the dc field from that of the Stark mixing.

We have shown theoretically and experimentally that Stark mixing between $|2s\rangle$ and $|2p\rangle$ states of atomic hydrogen can produce a resonantly enhanced nonlinear susceptibility for second-harmonic generation and at the same time produce reduced absorption for the generated radiation at the center of the Stark-split components. Also, the phase-matching condition can be satisfied exactly at the center. Although we have applied a dc electric field to mix two states instead of a high-frequency laser field, we believe this is the first demonstrated realization of the recent method proposed by Harris, Field, and Imamoglu[1] for achieving high conversion efficiency by nonlinear optical mixing. Further theoretical and experimental work is in progress in an attempt to scale up the present SHG method to high atomic density and long interaction length, in order to obtain intense coherent Lyman-α radiation.

We thank S. E. Harris for several helpful discussions. One of us (L.M.) is grateful to the University of Toronto for scholarships during the course of this study. This research was supported by the Natural Sciences and Engineering Research Council, the Premier's Council Technology Fund, and the University of Toronto.

[a]Visiting Scientist, University of Toronto, 1988–1989. Permanent address: Department of Applied Physics and Chemistry, and Institute for Laser Science, University of Electro-Communications, Chofu, Tokyo 182, Japan.

[1]S. E. Harris, J. E. Field, and A. Imamoglu, Phys. Rev. Lett. **64**, 1107 (1990).

[2]D. C. Hanna, M. A. Uratich, and D. Cotter, *Nonlinear Optics of Free Atoms and Molecules* (Springer-Verlag, Berlin, 1979), pp. 20–26.

[3]I. I. Sobelman, *Atomic Spectra and Radiative Transitions* (Springer-Verlag, Berlin, 1979), pp. 173–181.

[4]S. E. Harris, J. E. Field, A. Imamoglu, and J. J. Macklin, in *Proceedings of the International Conference on Quantum Electronics, 1990,* Technical Digest Series Vol. 8 (Optical Society of America, Washington, DC, 1990), pp. 220–222.

[5]A. Imamoglu, Phys. Rev. A **40**, 2835 (1989).

[6]A. Imamoglu and S. E. Harris, Opt. Lett. **14**, 1344 (1989).

[7]S. E. Harris, Phys. Rev. Lett. **62**, 1033 (1989).

[8]L. Marmet, K. Hakuta, and B. P. Stoicheff (unpublished).

[9]It should be noted that the present reduced ion signals cannot be attributed to the competition between harmonic generation and multiphoton-ionization processes described by J. J. Wynne [Phys. Rev. Lett. **27**, 751 (1984)] and by Blazewicz and co-workers [P. R. Blazewicz, M. G. Payne, W. R. Garrett, and J. C. Miller, Phys. Rev. A **34**, 5171 (1986); P. R. Blazewicz and J. C. Miller, Phys. Rev. A **38**, 2863 (1988), and references therein]. It can be readily understood from Eq. (8) of Ref. 6 that such competition may not be important for the present experiments at low density with small absorption.