Collision-assisted second-harmonic generation from Rydberg atoms

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Second-harmonic generation in rubidium vapor has been studied in the absence of external symmetry-breaking dc fields. It is shown that binary collisions between atoms in the presence of the fundamental laser beam leads to the generation of a coherent second-harmonic beam in accordance with a recent theoretical proposal [A. Elçi and D. Depatie, Phys. Rev. Lett. **60**, 688 (1988)].

Second-harmonic generation (SHG) in atomic vapors has been of great interest in recent years[1-7] and the given explanations for the occurrence of this effect in centrosymmetric atomic systems are based on the contributions of high-order multipole transitions,[1] spatial nonuniformities in the laser beam wave front,[2] ionization of atoms in intense fields,[2,3] or collision-assisted mechanisms.[4,5]

In a series of experiments, Dinev and co-workers[4,6] provided evidence that atomic collisions make an important contribution toward the SHG. This is a reasonable result since the large size of Rydberg atoms and their small binding energy make them very sensitive to collisions with other atoms. Furthermore, the corresponding energy separation between Rydberg levels is so small that these states can easily be mixed through collisions which may act effectively as a parity symmetry-breaking mechanism. The l-mixing model originally introduced in Ref. 4 is based on this reasoning and it has been the most used model to describe SHG in vapor although it fails to explain several important points such as the generation of a well-collimated second-harmonic (SH) beam, as well as the SH intensity behavior as a function of the atomic density and laser beam intensity.

In this article we present experimental results showing that the SHG originating from np levels ($n=15-30$) in rubidium has its main characteristics described by the recent theoretical model proposed by Elçi and Depatie.[7]

We used rubidium atomic vapor in a heated cell as the nonlinear medium. A Nd:YAG (where YAG represents yttrium aluminum garnet) laser-pumped dye laser (pulse duration of 5 ns, 5 Hz repetition rate, peak power of ≈ 30 kW) operating as a narrow bandwidth laser (≈ 0.1 cm^{-1}) in the wavelength range of 590–610 nm was used. The intensity and polarization of the laser beam could be controlled using filters, glan-prisms, and $\lambda/2$ plates.

The laser beam was linearly polarized and focused with a 20-cm foal lens into the 25-cm-long cell containing rubidium vapor and argon buffer gas. The vapor density was varied between 4×10^{14} and 5×10^{6} atoms/cm^3. The uv beam in the range of 298–303 nm was analyzed using a 1.4-m double monochromator (resolution of 0.05 nm). Appropriate filters were used to cut off the fundamental beam. The SH signal was detected with a 1P28 photomultiplier tube, a boxcar averager, and a chart recorder.

Two-photon resonant SHG from excited s, p, and d states was studied as a function of the pump-laser intensity $I(\omega)$, Rb atomic density N, and the buffer gas pressure.

The well-collimated SH beam was detected through a monochromator placed about 1 m from the heated cell. Figure 1 shows the several observed transitions between nl states ($n=15,\ldots,30$) and the ground $5s$ state of rubidium. This result was obtained scanning the laser frequency in the region between 590 and 605 nm. The heated cell temperature was 593 K, corresponding to a rubidium vapor density of 3.3×10^{16} atoms/cm^3 and a buffer gas pressure of 7 Torr was used. As shown in Fig. 1 the intensity of the SH signal decreases for increasing values of n. The relative intensities of the several lines were studied as a function of the temperature and the gas buffer pressure. For example, we observed that for a fixed cell temperature of 600 K the SHG efficiency measured was 3×10^{-6} for the $16d\rightarrow5s$ transition, 6×10^{-7} for the $18s\rightarrow5s$ transition, and 1×10^{-6} for the $17p\rightarrow5s$ transition. These values were independent of the Ar pressures in the region between 0.1 and 7 Torr. However, when the Ar pressure was increased up to 200 Torr the SH signal decreased one order of magnitude probably due to phase mismatch. For equal pressures of Rb and Ar (~7 Torr) used in most of the present experiments no parametric emission from the neighboring lower states was observed except for the $17s\rightarrow5s$ line.

The intensity dependence of the second-harmonic signal as a function of the dye laser intensity is shown in Fig. 2. For these measurements the laser frequency was adjusted to resonate with a two-photon transition and the monochromator was tuned to the SH frequency. The intensity of the fundamental beam was varied using calibrated neutral density filters. As illustrated in Fig. 2, several second-harmonic lines present the same law dependence with respect to the laser intensity. At low intensities, we found a dependence greater than I^2 which changes to a quadratic law in the region 100–400 MW/cm^2 and then to a linear behavior for intensities larger than 400 MW/cm^2. The solid curve was obtained using the "polariton states" formalism introduced in Ref.

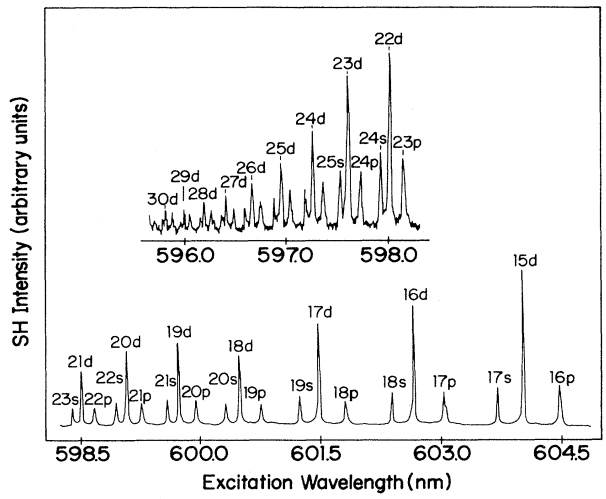

FIG. 1. Two-photon excitation spectrum of atomic rubidium. (Rb density: 3.3×10^{16} atoms/cm^3; buffer gas pressure: 7 Torr).

7. The assumptions made and the fitting parameters will be discussed below.

The SHG dependence with the atomic density was also studied for several transitions and the results are presented in Fig. 3 for two representative transitions. The Rb atomic density N was varied changing the cell temperature. A quadratic dependence with N for the SHG originating from s and d states was obtained while for the transition $np\rightarrow5s$ a quartic dependence was observed as shown in Fig. 3. The two different behaviors with the atomic density is evidence that different processes are contributing to SHG from s, d, and p states.

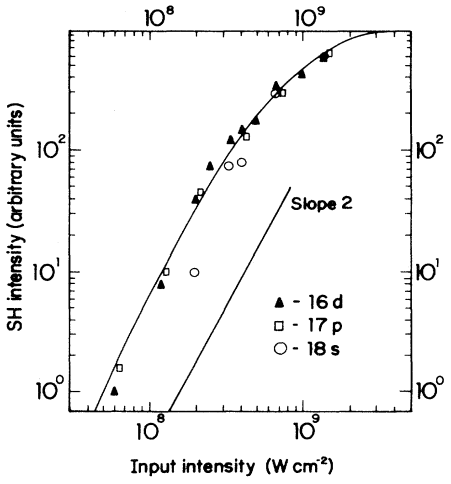

FIG. 2. Normalized SH intensity from 16d, 17p, and 18s states as a function of pump intensity. The predictions of Elçi and Depatie (Ref. 7) are illustrated by the solid line obtained from Eq. (1).

In order to understand the present experiments we first analyze the experimental results using the l-mixing model of Dinev *et al.*[4] According to this approach during collisions between the Rydberg atom and a rare-gas perturber the s or d states are mixed with p states and thus the collisionally induced dipole moment can radiate at the second-harmonic frequency. The induced second-harmonic field is thus proportional to the product between Rb and rare-atom densities. Consequently, the second-harmonic intensity $I(2\omega)$ should present a quadratic dependence with the Rb density and with the laser intensity $I(\omega)$. These conclusions agree with the observed behavior of $I(2\omega)$ as a function of the Rb density for the s and d transitions (Fig. 3) but fail to explain its behavior with $I(\omega)$ as shown in Fig. 2. Another important fact is that for low Ar pressure (0.1–7 Torr) the conversion efficiency does not change. We also found that for low temperature (495 K) no Ar buffer gas was necessary to observe a small SH signal from a sealed cell con-

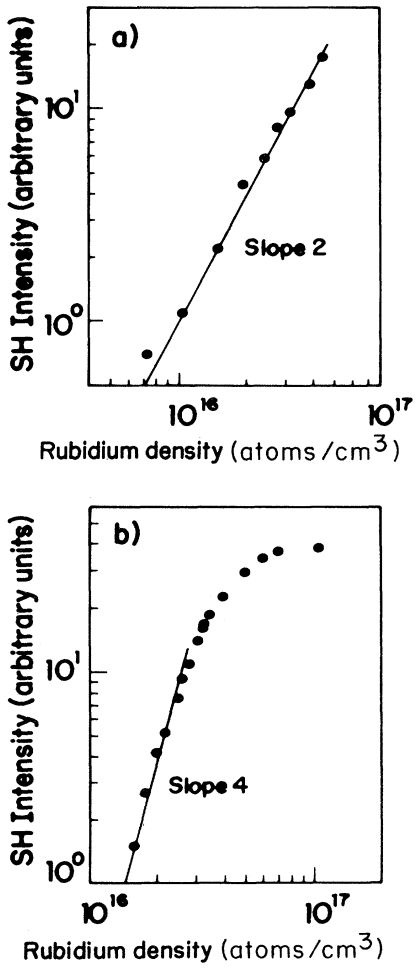

FIG. 3. Log-log dependence of SH intensity on Rb atomic density for 7 Torr of argon: (a) 16d ^2D-5s ^2S transition; (b) 17p ^2P-5s ^2S transition.

taining Rb in a 10^{-6}-Torr pressure. These results indicate that collisions between Rydberg atoms and a buffer gas are not necessary to generate the second-harmonic signal. Furthermore, the l-mixing model, as originally proposed in Ref. 4, cannot explain the quartic behavior with N obtained for the $np \rightarrow 5s$ transitions. Notice also that another fundamental problem with this model is to explain how the emitted second-harmonic radiation from the several atoms add coherently resulting in a well-collimated SH beam.

An alternative way to analyze the experiment is according to the formalism recently introduced by Elçi and Depatie.[7] In order to get a better insight concerning the physical processes involved in the SHG we first notice that very large cross sections for Rb Rydberg states colliding with ground-state Rb atoms in excess of 10^{-12} cm^2 have been reported.[8] On the other hand, collisions between Rydberg Rb with Ar atoms have cross sections of 10^{-14} cm^2.[9]

To explain the SHG using the formalism of Ref. 7, one has to think in terms of the interaction of the laser field with a pair-collision complex. Accordingly, the observed transitions can be described as corresponding to dipole-allowed transitions involving the mixed parity components of the pair-collision-complex states. The collision-induced second-harmonic polarization is proportional to the density of the pair-collision complexes. Although for the $s,d \rightarrow 5s$ transitions one obtains $I(\omega) \propto N^2$, for $np \rightarrow 5s$ transitions the SH intensity is proportional to the quartic power of the atomic Rb density indicating that collision complexes of Rb-Rb are involved. Thus in the case of the $np \rightarrow 5s$ transitions, the binary atomic collisions allow the coupling between the laser field to its second harmonic via the generation of "polariton states" which are expressed as a linear combination of states formed by the product between photon states and scattering states involving a pair of atoms. The phase information between the scattering events is thus preserved as shown in Ref. 7.

Strong evidence in favor of the "polariton states" model can be obtained studying the second-harmonic intensity behavior as a function of the laser beam intensity. Following Ref. 7 it can be shown that the SH intensity as a function of the incident laser intensity is given by

$$I(2\omega) \propto \left[-\frac{\zeta}{1+\zeta^2} + \frac{3}{\zeta} - \frac{3}{\zeta^2}\tan^{-1}\zeta \right]^2 , \qquad (1)$$

with $\zeta = (4\pi\mu/h\Delta)\sqrt{I/c\epsilon_0}$, where μ represents the dipole moment corresponding to the $5s \rightarrow 5p$ transition, Δ is the laser frequency detuning, I is the laser pump intensity, h is the Planck's constant, c is the speed of light, and ϵ_0 is the vacuum permitivity. From Eq. (1) we find that for a small laser intensity $I(2\omega)$ scales as I^3. However, for increasing values of I the SH signal increases at first as I^2 and then as I. For very large values of the incident laser beam a behavior with I^{-1} is predicted.

In order to apply the theoretical approach of Elçi and Depatie[7] to the present experimental conditions we consider that the laser beam of frequency ω couples the ground ($5s$) and the first excited ($5p$) states of the Rb atoms via a strong single-photon nonresonant electric dipole transition. The laser frequency detuning (Δ) with respect to the $5p$ level will be different for each selected two-photon resonant Rydberg state and for the whole set of levels studied it varies between 3957.09 and 3721.36 cm^{-1}.

The intensity parameter ζ can be estimated for the present experiments using $\mu = 7.8$ D, the pump laser intensity employed, and the appropriate Δ value for each wavelength used. For example, for the conditions corresponding, to Fig. 2, we used $\Delta = 3767.5$ cm^{-1} which corresponds to a laser wavelength of 603 nm such that $2\hbar\omega$ is in resonance with the energy separation between $17p$ and $5s$ states. The intensity parameter obtained is $\zeta = 7.6 \times 10^{-6}\sqrt{I}$. This value should be compared with the one obtained by a least-squares fitting of the experimental results ($7.6 \times 10^{-6}\sqrt{\beta I}$) with $\beta = 25$. The physical origin of the β factor is attributed to the self-focusing effect which contributes to reduce the confocal beam parameters effectively increasing the laser power density in the focal region.

A very good adjustment between Eq. (1) and the experimental values for the line $17p$ can be obtained as shown in Fig. 2, where the results for $16d$ and $18s$ states were also plotted. The behavior of the various recorded second-harmonic transitions as a function of $I(\omega)$ is quite similar to the one displayed in the Fig. 2 supporting the main assumptions of the polariton states model. An analogous behavior of the intensity dependence of the SHG signal was also reported in Ref. 5 although no explanation was given.

Notice that although the intensity dependence of the $s,d \rightarrow 5s$ SH transitions agree with the prediction of Ref. 7 (see Fig. 2), the quadratic dependence with the Rb density indicates that other mechanisms are involved. Accordingly further measurements were performed to investigate the possibility of other mechanisms.

The first experiment was performed to check the influence of the possible multiphoton ionization processes on the SHG. As in Ref. 5, we found that the simultaneous presence of the pump dye laser and an intense 1064-nm laser beam does not increase the efficiency of the SHG indicating that the nonlinear process is not enhanced due to the charge separation following multiphoton ionization of the Rb vapor as suggested in Ref. 2. Moreover, for a Rb density $\sim 5 \times 10^{16}$ atoms/cm^3 and an infrared laser peak power of 100 kW, the SH process is completely suppressed probably because the Rb plasma produced by the infrared beam reduces the phase matching for the SHG.

In another experiment we verified that no backward uv signal was produced. Moreover, the SH temporal behavior was studied with a resolution of 2 ns. We observed that duration of the SH pulses was reduced by a factor ~ 1.4 with respect to the laser pulses and no time delay was observed. The results indicate that the uv signal corresponding to $s,d \rightarrow 5s$ transitions is not due to amplified spontaneous emission or superfluorescence. Certainly about this awaits further analysis.

In conclusion, we have presented an experimental

study of SHG in Rb vapor for *ns*, *np*, and *nd* states, $n=15-30$. For the $np \rightarrow 5s$ transitions the very good agreement with the predictions of the "polariton states model" provides strong evidence that random binary collisions play a fundamental role in the coherent second-harmonic generation in atomic vapors. The partial agreement for the $s,d \rightarrow 5s$ transitions indicates that other mechanisms have to be invoked to explain those emissions.

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