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RESONANT SECOND HARMONIC GENERATION IN POTASSIUM VAPOR

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ABSTRACT

Picosecond pulses are used to study resonant second harmonic generation in potassium vapor. Although the process is both microscopically and macroscopically forbidden, it can readily be observed. The results can be quantitatively understood by a multiphoton-ionization-initiated, dc-field-induced, coherent transient model.

Resonant second harmonic generation (SHG) in atomic vapor, first reported in 1978,[1] is a nonlinear optical process not yet fully understood. Both microscopically and macroscopically, it is forbidden by symmetry, and yet experimentally it is readily observable. Over the past years, many mechanisms have been proposed to explain the observations, but none seems to be highly successful.[2] Among them, the dc-field-induced mechanism initiated by laser ionization appears most plausible,[1,3,4] although the observed dependence of SHG on pump power and absence of correlation between SHG and degree of ionization were not explained. All previous investigations employed nanosecond pulse excitations. They were not able to follow the time development of the process which can help identify the responsible mechanism. For example, the creation of a dc field by laser ionization generally has a response time of ~ 100 ps. Only with picosecond or sub-picosecond pulse excitation can it be time-resolved. We report here a study of resonant SHG in potassium vapor using tunable picosecond pump pulses.[5,6] We show that laser-ionization-initiated dc-field-induced SHG explains the observations quantitatively.

The experiment was performed with amplified 2-ps dye laser pulses at 10 Hz and ~ 200 μJ/pulse propagating and generating SH in a potassium/argon mixture in a heat pipe.[5] The potassium vapor pressure was kept at 0.6 torr and the argon buffer pressure was variable between 5 and 100 torr. We discuss here only resonant SHG from the 4s-9d transition of K; results from the 4s-11s and 4s-10d transitions showed similar characteristics. With a pump intensity of 10^{10} W/cm^2 over a length of 10 cm in a K (0.6 torr)/Ar (20 torr) mixture, a SH signal of 100 photons/pulse highly collimated along the pump beam was observed. As a function of pump intensity, the SH output exhibited an I^8 dependence at low intensities and started to saturate at ~ 10^{10} W/cm^2. These results are quite different from those obtained with nanosecond pump pulses.[2] In the latter case, SHG efficiency as high as 10^{-5} with an input intensity of 10^9 W/cm^2 and an I^2 dependence before saturation have been reported. However, as we shall see later, both cases can be understood as resulting from dc-field-induced SHG.

How can resonant SHG in atomic vapor ever be observed with picosecond pump pulses? Estimates of electric quadrupole and various other mechanisms yield orders-of-magnitude weaker signals than what has been observed.[7] The dc-field-induced SHG could give a roughly correct signal if the dc field were developed instantaneously

following ionization. However, the dc field only builds up when electrons diffuse out of the ionization region, and takes ~ 100 ps to reach the maximum value, as shown by a representative example[6] in Fig. 1. Clearly, during the picosecond pulse, the dc field generated is negligibly small, and the dc-field-induced SHG should be insignificant. This seems to contradict what was observed.

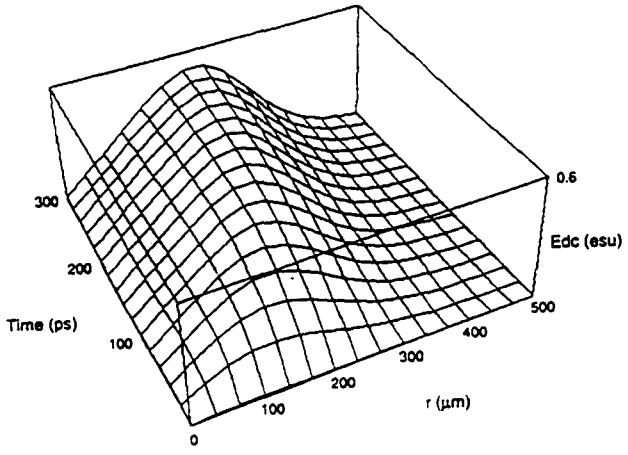

Fig. 1. Numerical simulation of E_{dc} initiated by a picosecond or femtosecond laser pulse. Initial electron density is 3×10^{11} cm^{-3} with a cylindrical radius of 170 μm.

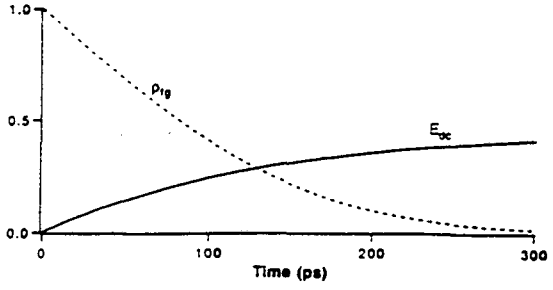

Fig. 2. Time development of E_{dc} and ρ_{fg} for 0.6 torr of K in 10 torr of Ar at $I(\omega)$ = 10 GW/cm^2 and r = 200 μm.

The dilemma can be solved if we realize that the resonant SHG actually consists of two steps, a two-photon coherent excitation (e.g., from 4s to 9d) followed by coherent radiation at the SH frequency. The coherent excitation decays with a dephasing time, which is of the order of 100 ps in our K/Ar mixture.[8] In the absence of a dc field, this coherent excitation can only yield a quadrupole radiation. The dc field, however, can break the symmetry and make the forbidden radiation electric-dipole allowed. Therefore as long as the coherent excitation and the dc field buildup overlap in time, significant SHG can be expected. This is illustrated in Fig. 2. From the above model, one would find decreasing SHG with increasing Ar buffer pressure since the dephasing time of the coherent excitation is inversely proportional to the Ar pressure.[8] Figure 3 describes such a behavior and shows quantitative agreement between theory and experiment.

To test the model further, we adopted a time-delayed pump/probe scheme.[5] The picosecond pump and probe pulses, orthogonally polarized, were overlapped at a small angle of 0.2° in the vapor mixture. The pump was used to ionize the vapor and generate the dc field $E_{dc}(t)$ while the probe pulse created the coherent excitation $\rho_{fg}(t)$. The intensity of the probe pulse was so adjusted that it was not sufficient to produce detectable SHG by itself. Only with the help of the pump-created $E_{dc}(t)$ could the probe pulse yield significant SHG (along the probe beam), which must depend on the overlap of $E_{dc}(t)$ and $\rho_{fg}(t)$. We can calculate the SHG versus the time delay between the pump and the probe and compare the result with experiment. As shown in Fig. 4 for two representative cases, the agreement is satisfactory. The discrepancy in the 10-100 ps region is most likely due to our neglect of the non-diffusive electron motion in the calculation of $E_{dc}(t)$. Note that SHG is appreciable even when the probe pulse is ahead of the pump pulse. This is because the probe-generated $\rho_{fg}(t)$ has a decay profile that

extends out to overlap with the pump-generated $E_{dc}(t)$. Thus, Fig. 4 constitutes a very strong evidence supporting our model.

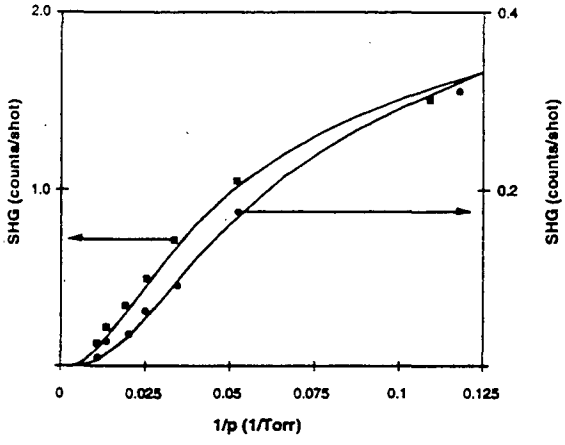

Fig. 3. SHG versus Ar pressure in 0.6 torr of K with $I(\omega) = 14$ GW/cm^2 (squares) and 8 GW/cm^2 (circles). The lines are theoretical.

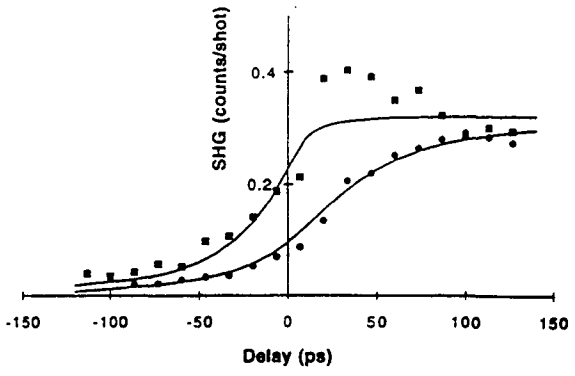

Fig. 4. SHG versus time delay between pump and probe at $I(\omega) = 12$ GW/cm^2 (squares) and 8 GW/cm^2 (circles). The lines are theoretical.

We now discuss how well the model predicts other characteristic features of resonant SHG by picosecond pulses. On the pump intensity dependence, we know that at low pump intensities, the dc-field-induced SH output should be proportional to $|E_{dc}|^2 I^2$. In our experiment, E_{dc} results from three-photon ionization of potassium and hence $E_{dc} \propto I^3$. This then leads to an SHG signal proportional to I^8, in agreement with the experimental observation. At higher pump intensities, SHG deviates from I^8 as E_{dc} begins to saturate. Calculation shows that the quasi-steady-state value of E_{dc} rapidly saturates and becomes independent of ionization (or I) for ion or electron density above $\sim 10^{11}$/cm^3. Similarly, the dependence of SHG on potassium density N_K can be explained. The SH output is proportional to $N_K^2 |E_{dc}|^2$. At low densities of K, $E_{dc} \propto N_K$ and hence SHG $\propto N_K^4$, but as N_K increases, both E_{dc} and SHG appear to saturate, as was indeed observed experimentally.

Saturation of E_{dc} with ionization is actually the reason behind the anomalous results of resonant SHG in atomic vapor by nanosecond laser pulses.[2] In this case, E_{dc} quickly rises up (in time) to a constant value independent of the pump laser intensity as long as the intensity is sufficiently strong. Accordingly, the dc-field-induced SH signal appears nearly proportional to I^2. It also explain why SHG has no correlation with the level of ionization. Marmet, et. al.[4] have calculated E_{dc} resulting from three-photon ionization in a diffusive atomic beam and found that on average E_{dc} also saturates.

We can also calculate the spatial profile of the SHG output. Following Bethune,[3] we find that the nonlinear polarization for the dc-field-induced SHG should have the form

$$\vec{P}(2\omega) = \chi_b E_o^2 E_{dc} [\vec{x}/r + (\alpha + 1)\vec{y}/r] \exp(-2r^2/R^2)$$

and a SH output proportional to

$$|P(2\omega)|^2 = |\chi_b|^2 E_o^4 E_{dc}^2 [x^2/r^2 + |\alpha + 1|^2 y^2/r^2] \exp(-4r^2R^2)$$

where $\alpha \equiv \chi_a/\chi_b$ and $|\alpha + 1|^2$ is the ratio of SH outputs polarized parallel and perpendicular to the pump polarization. For $\alpha = 3$, the spatial profile of SHG appears to have two lobes along the input polarization and the SH polarization is mainly parallel to the input polarization. These predictions apply to the 4s-9d resonant SHG, and are found to agree very well with experiment. For other transitions, α has different values and the spatial distributions of SH intensity and polarization should change accordingly.

From what we have discussed, it is clear that we now have a fairly good understanding of resonant SHG in atomic vapor. The process is dc-field-induced with the dc field created by multiphoton ionization. With picosecond pump pulses, the SHG is not instantaneous; it comes from two-photon coherent excitation that is allowed to radiate by the ionization-induced dc field. The above picture would predict similar results if femtosecond pump pulses are used. This is indeed what was observed in a recent experiment.[9] So far, we have not discussed SHG at 4s-np resonances which are forbidden in two-photon absorption but allowed in SH emission. In this case, the dc-field-induced SHG again can be operative as the dc field makes the forbidden two-photon transition allowed. This is presumably the case with nanosecond pump pulses and has been reported.[2] With picosecond pulses, however, the dc field created by laser ionization only becomes significant long after the pump pulse is over, and therefore would not help the two-photon 4s-np excitation. Consequently, the 4s-np resonant SHG is expected to be very weak. Confirmation of this prediction is currently in progress.

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