

# Experimental investigation of the polarizability of the $H_2$ molecule in the excited vibrational state

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We observed and measured by an interference method the change produced in the dielectric constant of hydrogen by excitation of the vibrational state in SRS. The polarizability of the first-excited vibrational state of the  $H_2$  molecule was determined.

The polarizability of molecules in the excited state  $\kappa^{(e)}$  determines the Stark shift of this state in an optical wave, and also the change of the dielectric constant of a substance, due to its excitation;<sup>[1]</sup> this last phenomenon can lead to self-action of the light.<sup>[2-4]</sup> Experimental data on the values of  $\kappa^{(e)}$  are therefore undoubtedly of interest, but no such measurements have been reported in the literature.

It was proposed in<sup>[1]</sup> to determine the polarizability of the excited states, which is produced in matter by radiation, from interference measurements of the nonlinear dielectric constant. In the present paper we demonstrate the feasibility of such measurements even for vi-

brational states whose polarizability difference is in the general case smaller than in the case of the electronic states.

The measurement object was chosen to be the  $H_2$  molecule, since the values of  $\kappa^{(e)}$  have been calculated for it<sup>[5, 6]</sup> for the first excited vibrational state.

The experimental setup is shown in Fig. 1. The pump was the second harmonic of a single-mode neodymium laser ( $\lambda_p = 0.53 \mu$ ). The bulk of the pump was focused by lens  $L_1$  (focal length  $f = 300$  cm) into a Mach-Zender type interferometer ( $P_2$ ,  $P_6$ ,  $M_1$ , and  $M_3$ ). One arm of the interferometer included chamber  $C_2$  (of length  $L_2 = 4$  cm) filled with hydrogen at a pressure  $P_2 = 60$  atm.

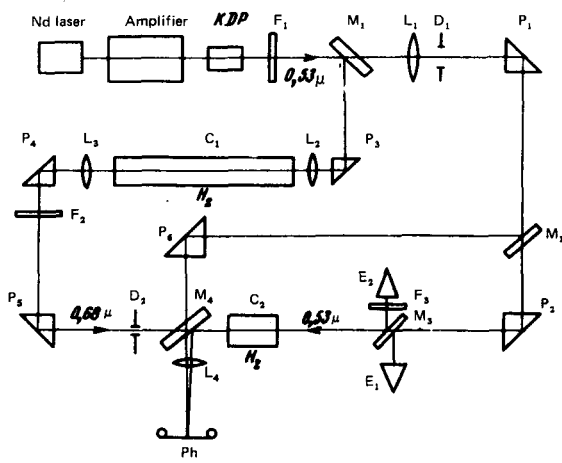


FIG. 1. Block diagram of experimental setup:  $C_1, C_2$  - chambers with compressed  $H_2$ ;  $M_1-M_4$  - mirrors,  $P_1-P_5$  - prisms;  $L_1-L_4$  - lenses,  $E_1, E_2$  - calorimeters,  $Ph$  - photographic film,  $F_1-F_3$  - light filters.

The vibrations of the  $H_2$  molecules were excited in the chamber  $C_2$  during the course of amplification of the Stokes SRS component ( $\lambda_s = 0.68 \mu$ ), obtained in chamber  $C_1$  ( $L_1 = 75$  cm,  $P_1 = 60$  atm). The filter  $F_2$  passed only the Stokes SRS component, which was then directed to chamber  $C_2$  in a direction opposite to that of the pump beam.

Excitation of the vibrations by opposing pump and Stokes-radiation beams prevents production of the anti-Stokes SRS component (including the axial component<sup>(7)</sup>), which disrupts the excited state.

The interferometer was adjusted in such a way that fringes of finite width were localized inside the chamber  $C_2$  and were projected by lens  $L_4$  on the photographic film. Simultaneously with the interference pattern, we registered the pump energy fed to chamber  $C_2$  (with calorimeter  $E_1$ ), and the Stokes component emerging from this chamber (with calorimeter  $E_2$ ).

Typical interference patterns are shown in Fig. 2a. The direction of the bending of the fringes corresponds to a positive increment, due to SRS, to the refractive index of the gas. In the absence of Stokes radiation in chamber  $C_2$ , there was no observed bending of the fringes (Fig. 2b).

From the bending of the interference fringes one can obtain the difference between the polarizabilities of the  $H_2$  molecule in the excited and in the ground vibrational states. To this end, we calculate the increment of the optical path in chamber  $C_2$ , due to the excitation of  $H_2$ . At the pump frequency, this increment, at the end of a pulse of duration  $t_{pul}$ , amounts to<sup>(1)</sup>

$$\Delta l = \frac{2\pi N}{n(\omega_p)} \int_0^{L_2} \chi_{nl} dz = \frac{\pi N(\kappa^{(e)} - \kappa^{(g)})}{n(\omega_p)} \int_0^{L_2} (\eta_0 - \eta) dz. \quad (1)$$

where  $n(\omega_p)$  is the linear part of the refractive index for the pump,  $N$  is the density of the number of molecules,  $\chi_{nl}$  is the real part of the nonlinear susceptibility of one molecule (see<sup>(11)</sup>), and  $\eta_0$  and  $\eta$  are the population dif-

ferences of the ground and excited vibrational states at the start and at the end of the pump pulse.

The following conditions were satisfied in the experiment: 1) The amplitudes of the pump field  $\mathcal{E}_p$  and of the Stokes field  $\mathcal{E}_s$  ( $E_{p,s} = \mathcal{E}_{p,s} \exp[i\omega_{p,s}t] + c. c.$ ) were such that the populations of the transition did not saturate during the pulse time. 2) The spatial length of the pulses was much larger than  $L_0$ . 3) The spatial distribution of the pump intensities was close to Gaussian

$$\left( |\mathcal{E}_{p,s}|^2 \sim \exp\left[-\frac{x^2}{a_{p,s}^2} - \frac{y^2}{b_{p,s}^2}\right] \right)$$

and their dimensions remained practically unchanged after passing through the chamber  $C_2$ .

We can therefore use the following relations to calculate  $\Delta l$ :

$$\eta_0 - \eta = 4\pi^{-4} T \eta_0 |r|^2 \int_0^{t_{pul}} |\mathcal{E}_p|^2 |\mathcal{E}_s|^2 dt. \quad (2)$$

$$\frac{d|\mathcal{E}_s|^2}{dz} = -\frac{8\pi^2 N |r|^2 T \eta_0}{\hbar^3 n_0(\omega_s) \lambda_s} |\mathcal{E}_p|^2 |\mathcal{E}_s|^2. \quad (3)$$

where  $T^{-1}$  is the half-width of the transition line and  $r$  is the composite matrix element for the Raman process.

Substituting (2) and (3) in (1) and expressing  $|\mathcal{E}_s|^2$  at the center of the beam as it leaves the chamber in terms of the total energy  $W_S$  of the Stokes energy and the parameters  $a_s$  and  $b_s$ , we obtain the difference of the polarizabilities:

$$\kappa^{(e)} - \kappa^{(g)} = \frac{\Delta l}{\lambda_p} \frac{\lambda_p}{\lambda_s} \frac{n(\omega_p)}{n(\omega_s)} \frac{\hbar \pi c a_s b_s}{W_S - W_{S,in}}. \quad (4)$$

Here  $W_{S,in}$  is the Stokes-component energy entering chamber  $C_2$ .

For a pump energy  $W_p = 0.15$  J and for  $W_{S,in} = 5 \times 10^{-3}$  J at  $a_s b_s \approx 1.5 \times 10^{-3}$  m<sup>2</sup> we obtained an energy  $W_S = 15 \times 10^{-3}$  J and observed a relative deviation  $\Delta l/\eta_p \approx 0.75$  at the center of the beam.

Taking into account the error in the determination of the energy, of the beam energy, and of the value of  $\Delta l/\eta_p$ , we find that the polarizability difference between the ground ( $v=0$ ) and excited ( $v=1$ ) vibrational states at the pump frequency is  $(1.2-1.8) \times 10^{-41}$  A-sec-m<sup>2</sup>/V.

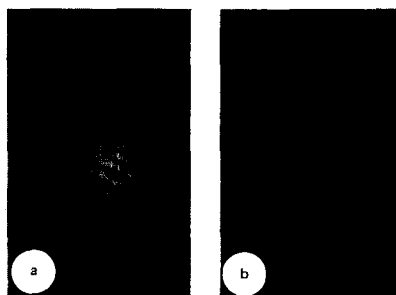


FIG. 2. Photographs of the interference fringes in the Mach-Zender interferometer: a - in the presence of Stokes radiation in chamber  $C_2$ , b - in the absence of Stokes radiation.

The calculated values are  $1.55 \times 10^{-41[6]}$  and  $1.8 \times 10^{-41}$  A-sec-m<sup>2</sup>/V. [5]

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<sup>1</sup>Since the lifetime  $\tau \approx 5 \times 10^{-6}$  of the excited state<sup>[8]</sup> is much larger than the pulse duration  $t_{pul} = 2 \times 10^{-8}$  sec, the registered interference pattern is that established at the end of the pump pulse.

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