

# Degeneracy of energy levels in the cooperative Raman scattering of light

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We have observed a significant difference in the pulse shape of the rotational cooperative Raman scattering of light by ortho and para hydrogen. This difference is caused by the different degree of degeneracy of the energy levels.

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The cooperative processes in the emission and scattering of light have attracted the interest of many authors in recent years (see review<sup>1</sup>). Despite the fact that most experimental studies involved molecular and atomic systems with degenerate energy levels, the degeneracy effects have not been investigated explicitly.

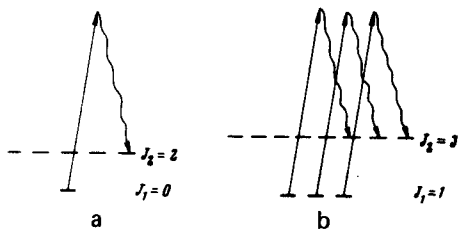


FIG. 1. Diagrams of transitions between magnetic sublevels for CRS in para and ortho hydrogen (a and b, respectively).

In our work we have investigated the rotational cooperative Raman scattering (CRS) by para and ortho hydrogen in order to determine the degeneracy effects. Since rotational scattering corresponds to the anisotropic part of the polarizability tensor,<sup>2</sup> the degeneracy of the energy levels of hydrogen molecules on the projection of the total momentum is of fundamental importance for the scattering process.<sup>1)</sup> The  $J_1 = 0 \rightarrow J_2 = 2$  transition (see Fig. 1) in para hydrogen is a model, purely two-level system, since the single transition  $M_1 = 0 \rightarrow M_2 = 2$  contributes to the CRS when circular polarization is excited. In contrast, for the scattering by the  $J_1 = 1 \rightarrow J_2 = 3$  transition in ortho hydrogen the three transitions  $M_1 = 1, 0, -1 \rightarrow M_2 = 3, 2, 1$  make a contribution.

A single-frequency, single-mode neodymium-glass laser ( $\lambda = 1.056 \mu\text{m}$ ) was used in the experiments. The rise time of the laser pulse reduced to 3–4 nsec by means of an external electro-optic shutter. The polarization of the laser radiation was converted from linear to circular, and then it was amplified and focused by a lens with  $F = 61 \text{ cm}$  into a 90-cm-long hydrogen cell. The energy of the laser pulse was 0.26 J. The spot area at the  $e^{-1}$  level at the neck of the focused beam was  $3 \times 10^{-4} \text{ cm}^2$ , and the average intensity in this spot was  $I \approx 10^{10} \text{ W/cm}^2$ . The cell windows were tilted at a  $5^\circ$  angle. The scattered radiation was isolated by a monochromator, and its pulse was recorded on a I2-7 oscilloscope. The pulse of the excitation radiation was recorded on another oscilloscope at the same time.

Normal hydrogen is a mixture of 25% para hydrogen and 75% ortho hydrogen. Scattering by the ortho hydrogen ( $\Delta\omega = 587 \text{ cm}^{-1}$ ) is observed at room temperature in this mixture. To obtain CRS by the para hydrogen ( $\Delta\omega = 354 \text{ cm}^{-1}$ ), we increased its content to more than 90%, using the method described in Ref. 4. In each case, scattering occurred only in the propagation direction of the laser pulse. The CRS spectrum consisted of the first Stokes component, and the polarization was contracircular. The scattered radiation was concentrated in one diffraction mode of the active volume; its power level for the conditions described below was less than 10% of the laser power.

Figure 2 shows oscillograms of the laser and scattered pulses. There is a considerable delay between the excitation front and the scattering pulse. The scattering pulse in para hydrogen has the transient fluctuations characteristic of CRS in a medium.<sup>3,6</sup> Figure 2a shows an oscillogram obtained at a pressure  $p = 1.1 \text{ atm}$  and room temperature. Under these conditions the measured delay time of the first CRS maximum from the middle of the leading edge of the excitation pulse amounts to  $t_0 = 10 \pm 3 \text{ nsec}$ , and the period of the fluctuations is  $\tau_0 = 4.5 \pm 1.5 \text{ nsec}$ . The CRS

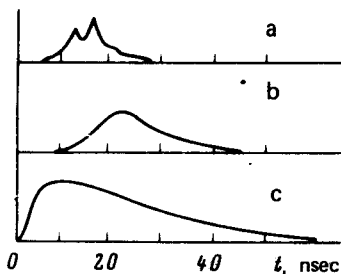


FIG. 2. Oscillograms of the CRS pulses in para hydrogen (a) and in ortho hydrogen (b), and an oscillogram of the excitation pulse (c).

pulses with a long fluctuation period also had a longer delay.

We shall evaluate the times  $t_0$  and  $\tau_0$  for para hydrogen by using the formulas from Ref. 6. For our conditions the wavelength of the Stokes scattering is  $\lambda_s = 1.097 \mu\text{m}$ , the length of the active volume is  $z = 10 \text{ cm}$ , the population difference of the initial and final states is  $n_0 \approx 10^{19} \text{ cm}^{-3}$ , and the polarizability anisotropy is  $\gamma_0 = 0.31 \times 10^{-24} \text{ cm}^3$ . We thus obtained  $t_0 = 10 \text{ nsec}$  and  $\tau_0 = 2 \text{ nsec}$ , in good agreement with the measurements. By using the results of Ref. 7 we found that the polarization dephasing time under these conditions is  $T_2 = 3 \text{ nsec}$ .

Figure 2b shows an oscillogram of the CRS in ortho hydrogen at  $p = 0.7 \text{ atm}$ . In contrast to the scattering in para hydrogen, the CRS pulse in ortho hydrogen has a smooth envelope with no fluctuations, whereas the parameters  $n_0 \approx 10^{19} \text{ cm}^{-3}$  and  $T_2 = 4 \text{ nsec}$  (Ref. 7) are close to those of para hydrogen.

As follows from the transition diagrams of Fig. 1, the large difference in the scattering in ortho and para hydrogen is attributable to the fact that in the case of para hydrogen the transition between the magnetic sublevels of the initial and final states contributes to the scattering, while in the case of ortho hydrogen there are three contributing transitions. In the case of ortho hydrogen the interaction of the different transitions with the excitation and scattered fields is determined by the different matrix elements of the scattering tensor. Therefore, the characteristic times at which the avalanche-like transition of molecules from the initial sublevel to the final sublevel occurs are different for each pair of sublevels. The delay times  $t_0(J_1 M_1, J_2 M_2)$  and the avalanche durations  $\tau_0(J_1 M_1, J_2 M_2)$  for the different  $J_1 M_1 \rightarrow J_2 M_2$  transitions, which are estimated in a manner similar to that in Ref. 6, are listed in Table I.

The delay times listed in the table agree with the measured value of  $t_0 = 19 \pm 5 \text{ nsec}$ . According to this table, the differences  $\delta\tau_0$  in the avalanche duration are

TABLE I.

Transition	$\tau_0(J_1 M_1; J_2 M_2)$ , nsec	$t_0(J_1 M_1; J_2 M_2)$ , nsec
$J_1 M_1 \rightarrow J_2 M_2$		
1 - 1 3 1	3.1	16.3
1 0 3 2	2.9	15.1
1 1 3 3	2.8	14.3

small, and the differences  $\delta t_0$  in the delay times are comparable to the avalanche durations:  $\delta t_0 \ll \tau_0$ . Since the avalanche transition of the molecules is accompanied by the emission of the scattering-field pulse, such a relationship between the transition times means that the scattering pulses corresponding to the avalanches between different pairs of sublevels follow each other with an interval that is slightly less than their duration. Because of this, the transient fluctuations of the scattering intensity, which are characteristic of the CRS in nondegenerate systems, can be smeared out in this case. Therefore, the observed differences in the shape of the CRS pulse in para and ortho hydrogen are caused, in our opinion, by the level degeneracy.

<sup>1)</sup> We note that purely vibrational CRS in hydrogen, which was reported in Ref. 3, is caused by the isotropic part of the polarizability.<sup>2,4</sup> Therefore, the degeneracy of the projection of the total momentum is unimportant in this case.

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