

LETTER TO THE EDITOR

The Stimulated Raman Gain Spectrum of the $Q_{1 \leftarrow 0}(0)$ Transition of Solid Parahydrogen

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We have recently reported in a series of papers that infrared spectral lines of solid hydrogen may be observed sharper than those in Doppler-limited gaseous spectroscopy by one to two orders of magnitude (1-4). Such high resolution has revealed a variety of intricate spectral structures due to intermolecular and crystal field interactions. Even the simplest vibration-rotation transition $Q_{1 \leftarrow 0}(0)$ ($v = 1 \leftarrow 0$, $J = 0 \leftarrow 0$) showed a structure because it is caused by neighboring $J = 1$ molecules which make simultaneous transitions (2, 4, 5). The Raman spectrum of the $Q_{1 \leftarrow 0}(0)$ transition, however, is expected to show a sharp single line due to the exciton momentum selection rule $\Delta k = 0$. We report here our observation of such a transition under high resolution using the stimulated Raman gain effect.

A crystal of nearly pure parahydrogen (99.90% para, 0.06% ortho, 0.04% residual HD) with the length of 11.5 cm and the diameter of 1.7 cm was prepared at helium cryogenic temperature in a manner similar to that reported earlier (3). The concentration of the orthohydrogen was determined by the infrared absorption intensity of the $Q_{1 \leftarrow 0}(1)$ ($v = 1 \leftarrow 0$, $J = 1 \leftarrow 1$) transition (6). Well collimated beams (diameter ~ 1.5 mm) of 476.6 nm (20 981.09 cm^{-1}) Ar ion laser radiation and ~ 594.1 nm dye laser radiation were passed through the crystal to cause the stimulated Raman effect. The Ar ion laser power was ~ 300 mW. The two beams were separated after the crystal by a prism and a filter and the dye laser radiation was monitored by a PIN diode. The frequency of the Ar laser radiation was tone-burst modulated at 10 kHz with radiofrequency sidebands of 6 to 80 MHz, and the variation of the Raman gain was synchronously detected with a phase sensitive detector.

The observed signals are shown in Fig. 1. The trace in Fig. 1a shows the good signal-to-noise ratio observed with the rf frequency of ~ 80 MHz and the trace in Fig. 1b shows the sharp linewidth of $\Delta\nu \leq 7$ MHz (HWHM) observed with the rf frequency of 8 MHz. As expected, the spectral line is single without any structure apart from that caused by the rf sidebands. The transition wavenumber was measured using an iodine standard line and a Fabry-Perot spectrum analyzer. The measured wavenumber of 4149.75 cm^{-1} was in agreement with 4149.81 cm^{-1} reported earlier by Bhatnagar, Allin, and Welsh (7). The observed Raman gain was of the same order of magnitude as the theoretically estimated value of 5.0×10^{-3} (8). The clear singlet structure with no hint of asymmetry or satellites suggests that our crystal is fairly free from inhomogeneous broadening and shifts due to the complication of polycrystalline structure and stress. We have searched 2.5 cm^{-1} above and below the observed line and found no other spectral features. Because of the absence of other features we believe that our crystal is purely hexagonal close-packed (hcp). A crystal with some face-centered cubic structure would probably give an additional $Q_{1 \leftarrow 0}(0)$ line at a transition frequency different from the hcp transition frequency (7).

The sharpness of the observed line is not due to the localization of excitons as in most of our infrared spectral lines, but is due to the exciton momentum selection rule $\Delta k = 0$. The spectral linewidth is possibly caused by the scattering of Bloch waves by impurity $J = 1$ molecules. Abram *et al.* measured the decay of vibrational coherence by time resolved Raman gain spectroscopy (9). While they do not give the value of the decay explicitly, we read from their figure that the decay time at 0.22% ortho concentration is ~ 8 MHz. Our observed width of 7 MHz is, however, contaminated by the instrumental width due to the frequency instabilities of the Ar ion and the dye lasers, which are estimated to be on the order of a few megahertz. Reduction of the instrumental width will be attempted. Nevertheless, the present observation already amply demonstrates the purity of the quantum states of the hydrogen molecules in the solid and the $\Delta k = 0$ selection rule.

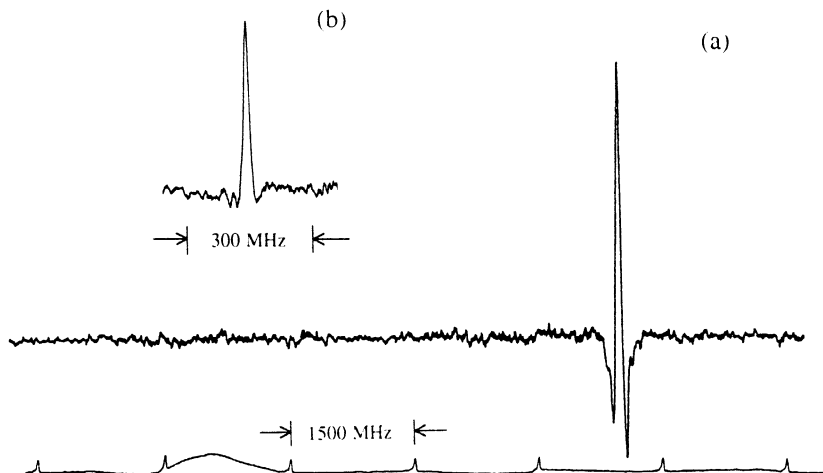


FIG. 1. Examples of the observed spectral line. (a) 80-MHz radiofrequency sidebands. The fine structure is due to overmodulated rf sidebands. The spacing of the spectrum analyzer markers is 1500 MHz. The feature on the left in the I_2 spectrum is at $16\,831.5127\text{ cm}^{-1}$. The detection time constant is 1 sec. (b) A high-resolution spectrum observed by using 8-MHz radiofrequency sidebands. The linewidth at half height at half maximum is ~ 7 MHz. The detection time constant is 1 sec.

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