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Laser induced fluorescence measurements of microwave stimulated OH molecules from H₂O photodissociation

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Laser induced fluorescence of microwave stimulated OH molecules from H₂O photodissociation is investigated as a first step in a series of laboratory experiments to understand features of astronomical OH masers. The inversion between the $\Pi_{3/2}$, $J=7/2$ Λ -doublet states of OH generated via photodissociation of cold H₂O at 157 nm is shown to be 1.8:1. Within a microwave, Fabry–Perot cavity tuned to the resonance of one of the main hyperfine transitions in this Λ doublet it is possible to stimulate all inverted OH molecules. The linewidth of the two main microwave transitions is measured as a function of the microwave power and interaction time. The results show that Doppler broadening is negligible at short interaction times of the molecules with the microwave field. © 1996 American Institute of Physics. [S0021-9606(96)02231-3]

I. INTRODUCTION

Astronomical OH masers are very interesting objects, because they are found in the vicinity of massive star formation regions (“interstellar masers”) or in the outer shell of late stars (“stellar OH masers”). A proper understanding of the pump mechanism of these masers may allow to extract important information about the physics and the environment in these objects. The present experiments are an attempt to investigate only features of the interstellar OH masers under the assumption that they are pumped by photodissociation of H₂O. Interstellar masers (H₂O, OH) are found at a distance of around 10¹⁵ m from the central object close to the border of the HII region.^{1,2}

The pump scheme of these OH maser is still under debate despite a large amount of observational data.³ A star in a star forming region may have a temperature of about 25 000 K. Thirty percent of the black body radiation is then in the range of the first absorption band of water (135–190 nm). A first step photodissociation of water yields a population inversion between $^2\Pi_{3/2}$ Λ -doublet states of OH and the maser activity starts. To close the pump cycle, it is probable that in a second step photodissociation of OH to O+H or reactive collisions of OH with H₂ destroy OH and thus depopulate the lower maser level. The reactive collisions also regenerate water.⁴

The first measurements of the inversion between the OH Λ -doublet states in $^2\Pi_{3/2}$ from the photodissociation of H₂O in the VUV have been reported by Andresen *et al.*⁵ Using a heated gas cell, an effusive beam and a pulsed nozzle beam they showed that the inversion depends on the temperature of the water molecules. The highest inversion resulted from dissociation of cold H₂O molecules. Here, a pulsed nozzle beam of pure water was expanded into a vacuum chamber and a F₂-excimer laser (157 nm) dissociated the H₂O molecules.

Laser induced fluorescence (LIF) was used to probe the population distribution of the generated OH molecules. The degree of population inversion was derived from the ratio of the intensities of *P* or *R* lines probing the lower $\Pi_{3/2}$ Λ -doublet states and *Q* lines probing the upper states. ter Meulen *et al.*⁶ have measured population inversion between the OH- Λ -doublet hyperfine states as well as accurate transition frequencies in an OH beam maser experiment. The inversion was created either in the OH production or by applying electrostatic state selection.⁷

In this experiment, a molecular beam maser setup is used to investigate OH-maser action based on the photodissociation of cold H₂O between microwave Fabry–Perot cavity plates. This maser should work like an interstellar OH-maser driven by H₂O photolysis, so that it may be possible to obtain information about the maser conditions, oscillation behavior and polarization effects. First LIF measurements of microwave stimulated OH molecules from the H₂O photodissociation are presented.

II. EXPERIMENTAL METHOD

The experimental setup is shown in Fig. 1. A heated pulsed nozzle with a 0.8 mm hole is used to form a pulsed molecular beam of pure water with a pulse risetime of 60 μ s and a full width at half-maximum (FWHM) of 100 μ s. The H₂O backing pressure is 1 bar which is obtained by heating water in an oven up to 100 °C. The jet expansion into the vacuum chamber (10⁻⁶ mbar) yields rotationally cold H₂O in the electronic ground state X^1A_1 . Utilizing a 5 mm skimmer the central part of the beam is directed between the plates of a Fabry–Perot microwave cavity tuned to one of the $\Pi_{3/2}$, $J=7/2$, Λ -doublet hyperfine transitions (Fig. 2), 13.441 417 3 GHz for $F'=4 \rightarrow F=4$ and 13.434 637 4 GHz

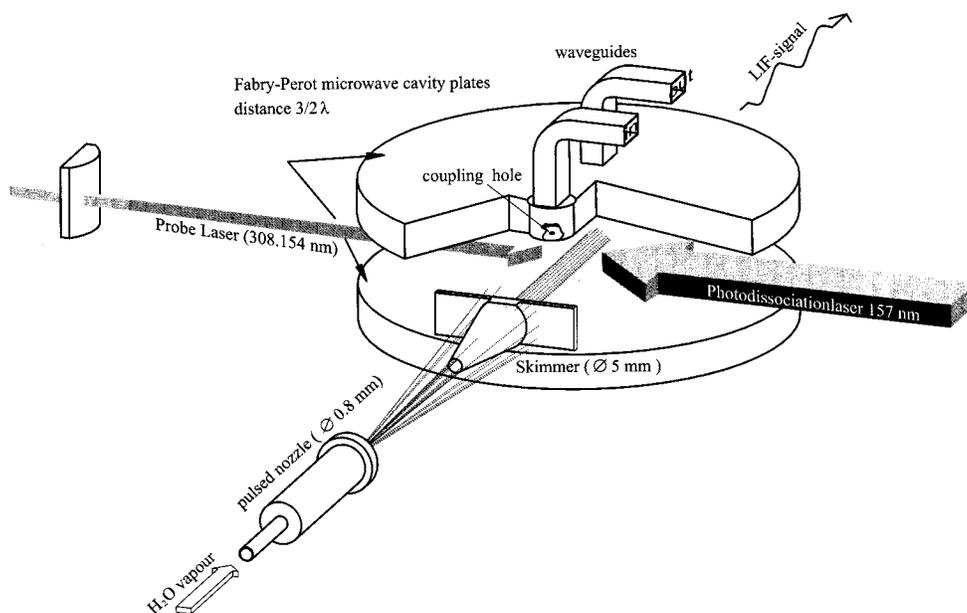


FIG. 1. Overview of the experimental setup in the vacuum chamber. The reaction volume is in the centre of the Fabry–Perot plates. In this experiment the outcoupling waveguide was not used.

for $F' = 3 \rightarrow F = 3$.⁷ The distance between nozzle and skimmer is 40 mm and the distance between skimmer and the centre of the cavity is 86 mm.

The Fabry–Perot cavity is part of a microwave system. A frequency stabilized klystron (frequency range 12–18 GHz) with a bandwidth < 1 Hz delivers the microwave power. The continuous output of the klystron is amplified

with a microwave amplifier (HP 83 006 A) up to a maximum power of 400 mW. A subsequent attenuator allows to vary the input power to the Fabry–Perot cavity. The diameter of the Fabry–Perot plates is 155 mm. The plate with the coupling holes is flat, the other one has a radius of curvature of 446 mm.

The distance between the cavity plates can be tuned from outside the vacuum chamber. This allows to change the resonance frequency without any change of the incoupling microwave power. The distance between the plates is ≈ 33 mm which corresponds to $3/2 \lambda$ for the $\Pi_{3/2}$, $J = 7/2$ Λ -doublet transition of OH, and the quality factor is about 6000.

The density of the H_2O -molecular beam is estimated by filling the vacuum chamber with H_2O up to a pressure at which the same LIF signal is obtained as in the molecular beam. It was checked that at these densities no significant absorption of laser radiation took place. Inside the cavity, the density of the molecular beam turns out to be about $4 \times 10^{12} \text{ cm}^{-3}$. The H_2O molecules are photodissociated in the center (see Fig. 1) of the cavity by VUV photons from an F_2 -excimer laser (Lambda Physik, LPF 205, 157 nm). The laser delivers maximum pulse energies of 40 mJ inside the vacuum chamber at a repetition rate of 10 Hz. To estimate the efficiency for H_2O photodissociation, we consider the excitation rate which is given by

$$\gamma = \sigma(\bar{\nu}) \int I(\nu) d\nu = \sigma(\bar{\nu}) \frac{N}{\tau F}$$

with $N = 3 \times 10^{16}$ photons/pulse, $\tau = 10$ ns excimer pulse length, $\sigma(\bar{\nu}) = 2 \times 10^{-18} \text{ cm}^2$ cross section for the H_2O absorption at $\lambda = 157 \text{ nm}$ and $F = 0.6 \times 1.3 \text{ cm}^2$ the area of the excimer profile.

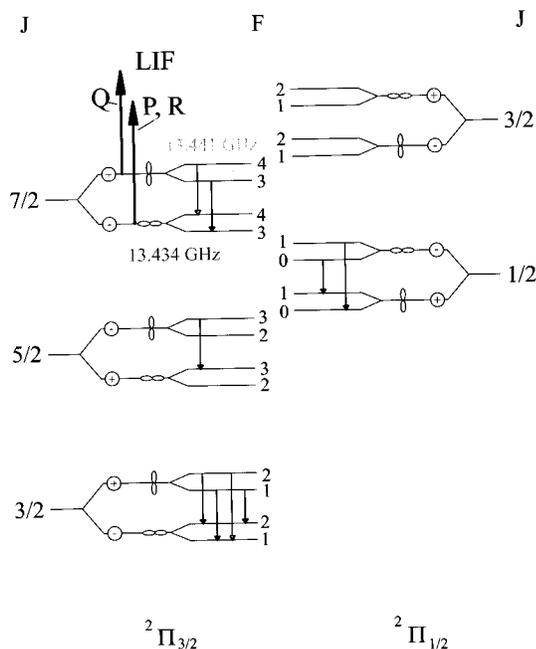


FIG. 2. The schematic energy level diagram shows the electronic ground state of OH. The arrows indicate observed interstellar OH-maser emissions.

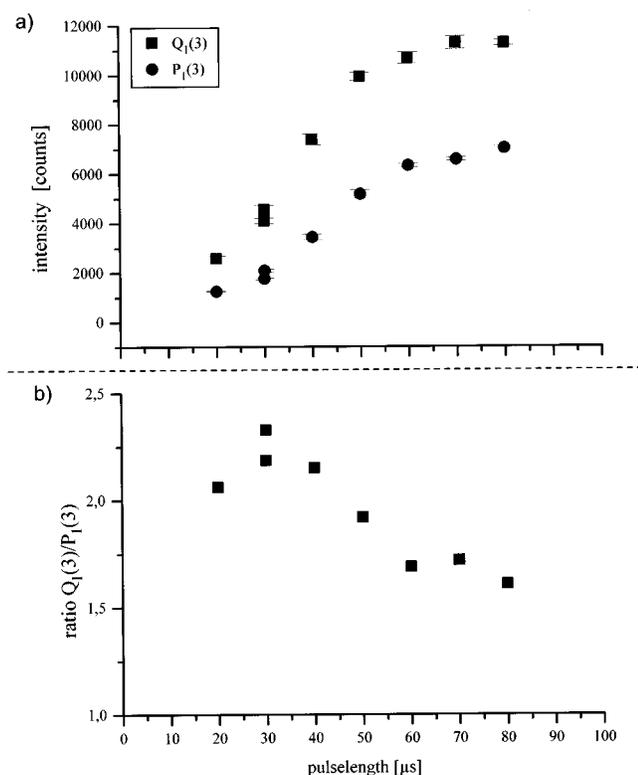


FIG. 3. (a) The LIF intensities of the $Q_1(3)$ and the $P_1(3)$ transitions as a function of time in the pulse. (b) The resulting ratios of $Q_1(3)/P_1(3)$ as a function of time in the pulse. The same measurements done for a constant background pressure of H_2O result in a ratio of $Q_1(3)/P_1(3)=1.25\pm 0.1$. Dividing the pulsed nozzle ratio by the background pressure ratio a maximum inversion of 1.8 is achieved.

The fraction of dissociated H_2O molecules is

$$\eta = \gamma\tau$$

and equals to an amount of 8%. With this fraction of dissociated H_2O molecules, the resulting OH density in the cavity is $3\times 10^{11} \text{ cm}^{-3}$. The distribution between the Λ -doublet levels and the population of the selected $\Pi_{3/2}$, $J=7/2$, Λ -doublet hyperfine levels yields an estimate of $\approx 1\times 10^{10}$ OH molecules/ cm^3 in the $F=3$ or $F=4$ hyperfine state.

The population distribution of the OH- Λ -doublet states is probed by LIF using a frequency doubled excimer pumped dye laser via the OH (${}^2\Pi-{}^2\Sigma$) absorption band around 308 nm. One of the important parameters in the experiments on the microwave stimulation of OH is the microwave line width, which is affected by the time that OH is exposed to the microwave field (“interaction time”). In the present case the interaction time of the produced OH molecules in the microwave field is determined by the delay between the dissociation and probe laser. The LIF signal is imaged on a CCD-camera (Wright P075) by means of a Maksutov–Newton-telescope and averaged over an area where the signal is strongest (20 000 pixels). Further averaging is done over 400 laser shots, to obtain a reasonable signal to noise ratio. The measured statistical error is then less than 5%.

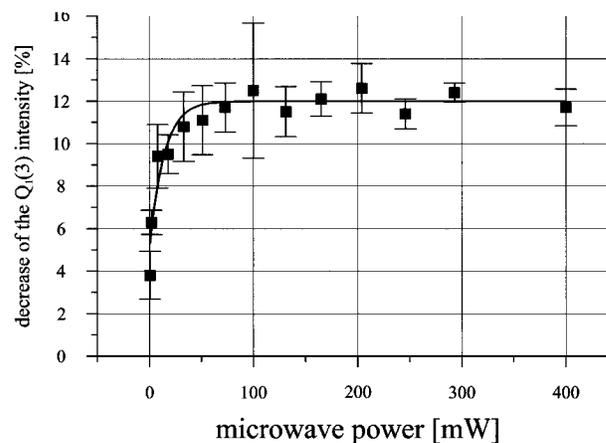


FIG. 4. Saturation behavior of the ${}^2\Pi_{3/2}$, $J=7/2$ Λ -doublet $F'=4\rightarrow 4$ transition in OH as a function of the microwave power.

The relative microwave induced population change between the Λ -doublet states is determined from the change in the population of the upper and lower Λ -doublet state with and without microwave stimulation via the $Q_1(3)$ and $P_1(3)$ transition, respectively. The dependence of the LIF signal on microwave power, frequency, and interaction time of the OH molecules with the microwave field is investigated.

III. RESULTS

In the following, we present experimental results about the inversion between the OH- Λ -doublet states as a function of the time in the pulsed water beam, the saturation of the microwave transition, the linewidth of the microwave transition as a function of microwave power, and finally the influence of the interaction time of the OH molecules with the microwave field. The photolysis within the H_2O pulse at different times affects the inversion between the Λ -doublet states. Using the same experimental setup shown in Fig. 1, the population of the $P_1(3)$ and $Q_1(3)$ Λ -doublet state is measured as a function of the time in the H_2O -molecular pulse without a microwave field. Figure 3(a) shows the measured intensities of the $Q_1(3)$ and $P_1(3)$ LIF signal as a function of time in the pulse. The ratio of $Q_1(3)/P_1(3)$ is plotted in Fig. 3(b). To avoid systematic errors, the following procedure is used. The ratio $Q_1(3)/P_1(3)$ is measured twice, (1) for the cold water in the molecular beam at short delays between dissociation and probe laser, and then (2) for water at 300 K at long delays (10 μs).

In the second case, the populations of the two Λ -doublet states are equal because water at 300 K does not give inversion and even if this were the case, equilibrium would be achieved because of the collisional redistribution at the long delay of 10 μs . Dividing the two ratios $Q_1(3)/P_1(3)$ for cold water by the ratio $Q_1(3)/P_1(3)$ for 300 K water yields a direct measure for the inversion.⁵ In the present case, we obtain for 300 K water a ratio $R=1.25\pm 0.1$. For the cold water, in the beam the inversion depends upon the part of pulse that is photolyzed. Based on the data in Fig. 3, the division by this ratio of 1.25 yields in a maximum inversion

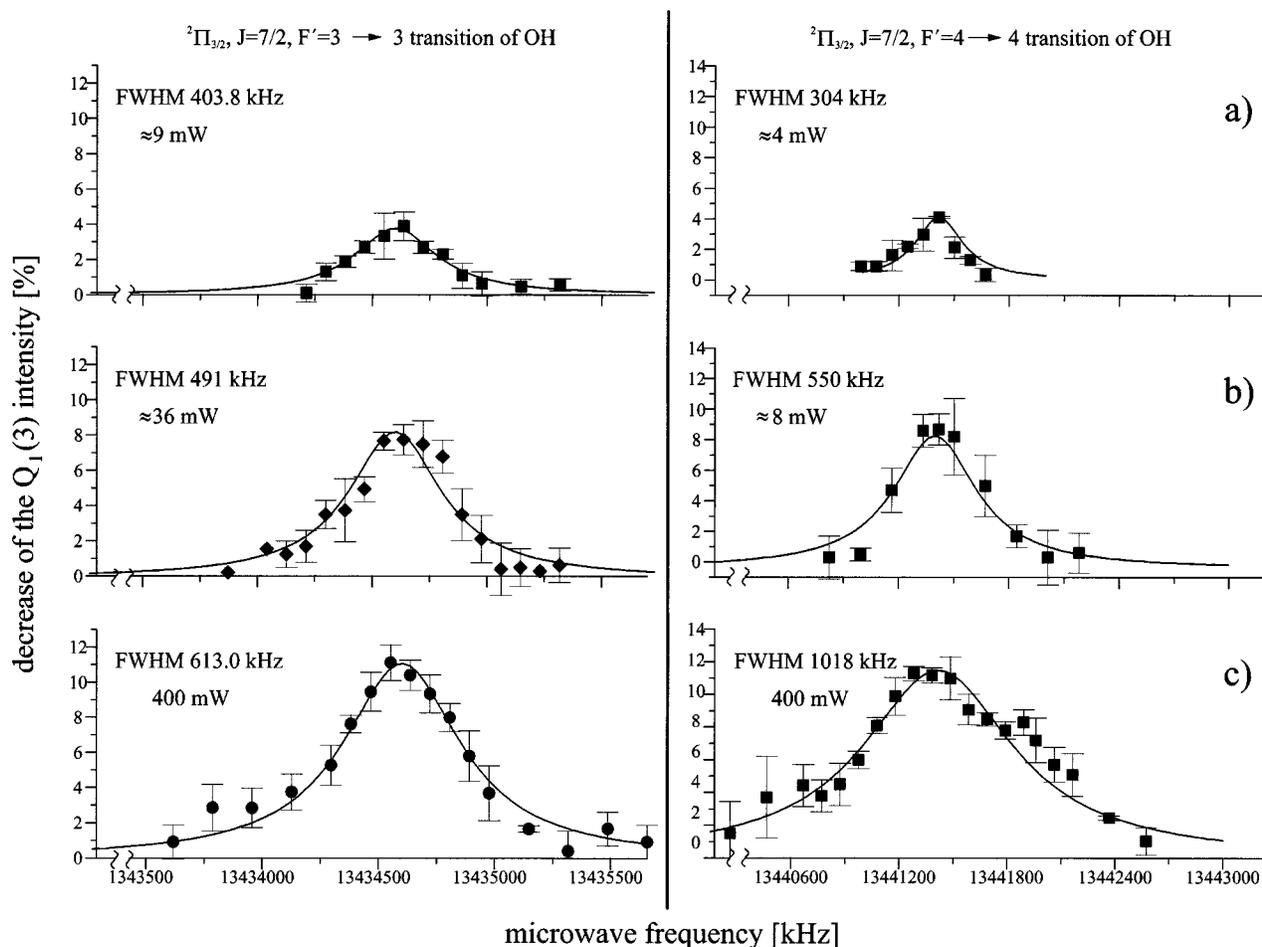


FIG. 5. Decrease of the $Q_1(3)$ intensity as a function of the microwave frequency and microwave power with a fixed interaction time of $1.5 \mu\text{s}$. The interaction time defines a minimum linewidth of 590 kHz.

of 1.8 after $30 \mu\text{s}$ and later in the pulse the inversion decreases down to 1.5. The pulsed nozzle opens very fast and results in a cold molecular beam and a high inversion only in the beginning (Fig. 3). At the end of the pulse, the expansion is less good and the inversion decreases.

Figure 4 shows the effect of microwave radiation on the population in the Λ -doublet state probed by the $Q_1(3)$ transition. Plotted is the decrease in the LIF signal. It should be kept in mind that the inversion implies that the state probed by the $Q_1(3)$ line is more populated than the state probed by the $P_1(3)$ line. The microwave field couples these two states and depletes the population in the state probed by $Q_1(3)$.

The decrease of the population in the upper Λ -doublet state probed by the $Q_1(3)$ line is shown in Fig. 4, as a function of the input power of the microwave field at the $F'=4 \rightarrow F=4$ hyperfine transition frequency. The interaction time that is the delay between dissociation and probe laser is fixed at $1.5 \mu\text{s}$. This transition is obviously saturated at a microwave power of 100 mW. A maximum intensity decrease of $12\% \pm 1\%$ is reached in the saturation case. At high power, the microwave field obviously stimulates all molecules down and reaches population equilibrium between the upper and lower $F=4$ state.

Via the $Q_1(3)$ line the probe laser excites simultaneously the states $F=3$ and $F=4$, whereas the microwave field affects only the population in the $F=4$ state. For the measured inversion of 1.8, the theoretically predicted decrease is 12.5%. The deviation from 12% to 12.5% is well within the error limits of the experiment. This value is based on the assumption of statistical population between the hyperfine states of OH after photolysis, resulting in a 9:7 statistical population of the $F=4$ and $F=3$ hyperfine states, respectively. For the $F=3 \rightarrow 3$ transition, a maximum decrease for the $Q_1(3)$ line of 9.7% is expected. Below 100 mW, of microwave power the population change is smaller.

The microwave induced decrease of the population in the state probed by the $Q_1(3)$ line is measured also as a function of microwave frequency. The microwave frequency is scanned over the full linewidth of the selected hyperfine transition at a fixed microwave power. Each data point encloses eight measurements of the population of the state probed by $Q_1(3)$ four with and four without microwave influence at the selected microwave frequency and microwave power. This is shown in Fig. 5. For a maximum power of 400 mW, the population decreases by $12\% \pm 1\%$ in case of both hyperfine transitions whereas at the lowest powers the

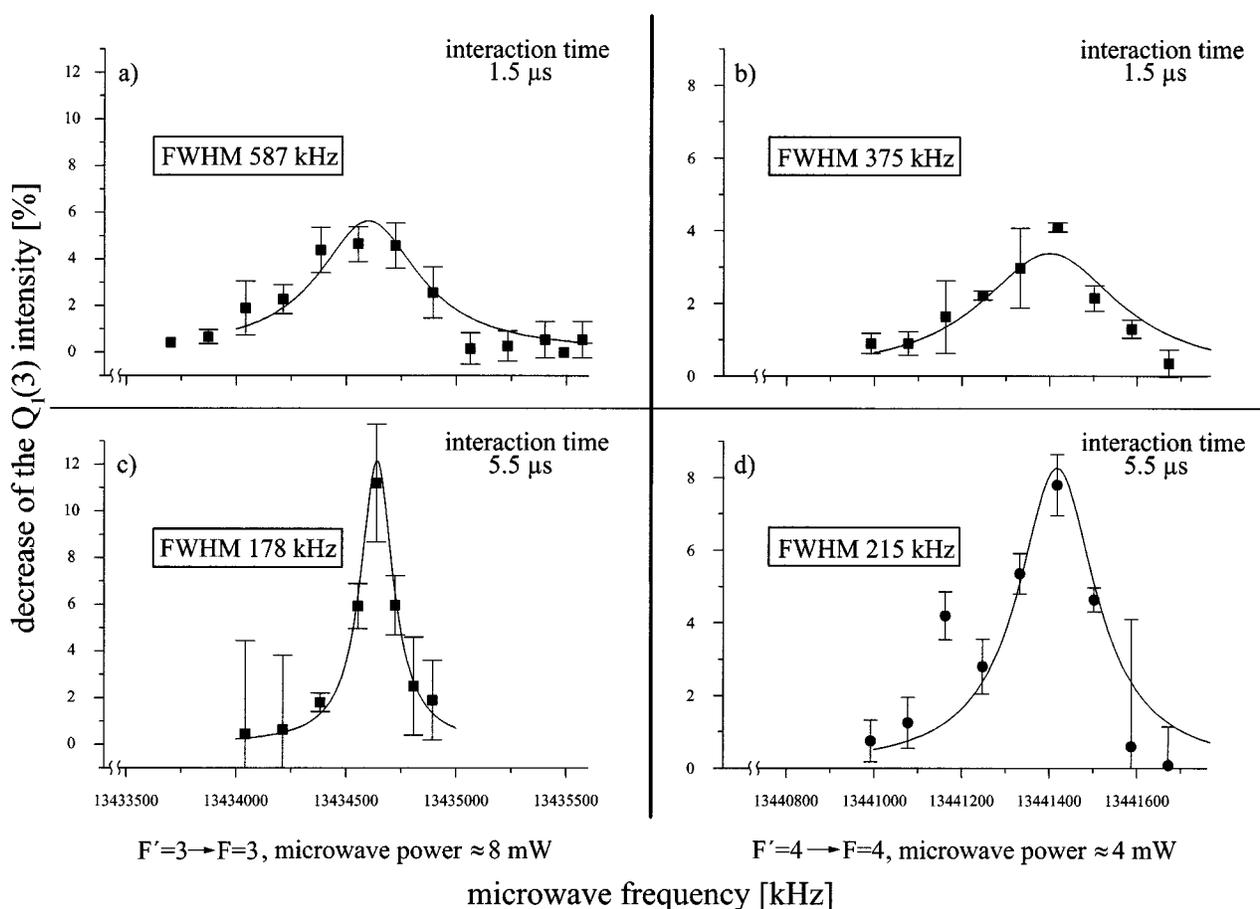


FIG. 6. Decrease of the $Q_1(3)$ intensity as a function of microwave frequency and the interaction time of the OH molecules with the microwave field.

decrease is only 4%. The interaction time is $1.5 \mu\text{s}$. A simple Lorentz shape fit is made to determine the FWHM of the hyperfine transitions at different powers of the stimulating microwave field. The experimental accuracy, however, is not very high.

The main contributions to the linewidth are the inhomogeneous Doppler and the homogeneous time of flight broadening. The OH molecules from the photodissociation of H_2O at 157 nm move with a velocity of 1600 m/s with an isotropic velocity distribution in first approximation. This results in a maximum Doppler shift of $\pm 70 \text{ kHz}$. The interaction time of the OH molecules with the microwave field is determined by the delay ($1.5 \mu\text{s}$) between the dissociation laser and the probe laser for the LIF detection. The linewidth can be calculated by Fourier analysis of the electric and magnetic field experienced by the molecules. For different transverse electric (TE) and magnetic (TM) fields, one gets different linewidths, as has been shown by Heuvel⁸ for cylindrical microwave cavities. As an example, he found for the TM_{010} mode $\Delta\nu \approx 0.89/\tau$ and for the TE_{011} mode $\Delta\nu \approx 1.20/\tau$. We did not perform the Fourier analysis for the present cavity, because the mode structure is quite complex. Instead, we assume that $\Delta\nu \approx 1/\tau$, which will give a reasonable estimate of the interaction time broadening. For $\tau = 1.5 \mu\text{s}$, a broadening of 670 kHz is obtained.

Despite saturation occurs at high microwave power, this will not cause any measurable line broadening. Saturation affects the molecular linewidth which is, however, much smaller than the present linewidth. Microwave line broadening due to saturation is typically in the kHz range. It is concluded that the time of flight broadening is the most important factor determining the linewidth of the hyperfine transitions. This is demonstrated in Fig. 6. The linewidth is measured at $\tau = 1.5 \mu\text{s}$ and $\tau = 5.5 \mu\text{s}$ with 4 and 8 mW microwave power for the $F=4 \rightarrow 4$ and $3 \rightarrow 3$ transitions, respectively. The longer interaction time results in a linewidth of $\approx 200 \text{ kHz}$ for both hyperfine transitions and in an increase of the stimulated OH molecules. The theoretical convolution of Doppler and time of flight broadening results in a FWHM of 200 kHz in this case and is in an excellent agreement with the experimental data.

Preparations are made to stimulate the OH emission by applying a pulsed microwave field. In this case, the interaction time will be determined by the length of the microwave pulse. Instead of probing the depletion of the inversion by LIF, the stimulated emission will be observed by superheterodyne detection.

Spontaneous maser oscillation of OH molecules from the photodissociation is not detectable with the present setup, because of the low density of OH-molecules per hyperfine

quantum state ($\approx 2 \times 10^{10} \text{ cm}^{-3}$) between the Fabry–Perot plates. The calculated threshold⁹ for spontaneous oscillation of the OH maser is reached at a density of OH molecules in the upper hyperfine state of $7 \times 10^{12} \text{ cm}^{-3}$. To investigate the oscillation behavior, a higher OH density between the Fabry–Perot cavity is necessary. This might be achieved by moving the nozzle directly in front of the Fabry–Perot plates without a skimmer and heating the H₂O reservoir to higher temperatures.

IV. CONCLUSION

As a first step to simulate interstellar OH masers pumped by VUV photodissociation of VUV in the laboratory, OH radicals are produced inside an open Fabry–Perot microwave cavity. A maximum population inversion of 1.8 obtained between the $J=7/2$ Λ -doublet states. The influences of Doppler and time of flight broadening on the stimulated emission are qualitatively and quantitatively understood.

In spite of the inhomogeneous Doppler broadening, it is possible to stimulate all inverted OH molecules down, by making the homogenous time of flight broadening larger

than the Doppler width. At long interaction times and high enough microwave power, the population inversion can be equilibrated completely.

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