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Intensity of a crossover signal relative to Lamb dips observed in Stark spectroscopy of methane

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ABSTRACT

We have observed twelve triplets of two Lamb dips and a crossover signal in Stark spectroscopy of the ν_3 band of methane using a comb-referenced sub-Doppler resolution spectrometer. The crossover signal consists of two crossover resonances in the Λ- and V-type three-level systems, which overlap each other in frequency under the first-order Stark shifts (Tyurikov et al., 1997). The intensity of the crossover signal relative to the Lamb dips agrees well with a simple calculation based on steady-state solutions of rate equations in weak saturation using the observed intensity ratio of the Lamb dips in the Λ-type three-level system.

1. Introduction

Saturated absorption spectroscopy has been applied extensively to enhance Doppler-limited spectral resolutions of thermal gaseous atoms and molecules by observing Lamb dips with sub-Doppler line widths from the early days of laser spectroscopy [1–7]. When two transitions share a common upper or lower level (Λ- or V-type three-level system) and are separated in frequency by less than the Doppler width, a crossover resonance is also observed in the middle of the Lamb dips of the two transitions [8,9]. The small frequency separation is usually caused by the hyperfine interaction and Stark and Zeeman effects in visible, infrared, and submillimeter spectroscopy [4,5,8,10]. The crossover resonance is useful for making assignments of the Lamb dips. In addition, even when the weaker Lamb dip is not observed, the transition frequency can be determined if the crossover resonance is observed together with the larger Lamb dip.

The intensity of crossover resonance was discussed for atoms and molecules [11–15]. For atoms, the crossover resonance is sometimes much larger than associated Lamb dips, for instance, the D lines of the alkali atoms [11,16,17]. Nakayama attributed the intensity distribution to the branching ratio of the spontaneous emissions in the three-level system [11]. For molecules, however, crossover resonance has not been observed to be the largest in the triplet, as far as we know [8,10,12,18].

We carried out Stark spectroscopy of the ν_3 band of methane using a comb-referenced sub-Doppler resolution spectrometer [19]. Twenty transitions between the E levels were observed across the vibrational band because they are subject to the first-order Stark shift and thereby sensitive to the Stark field [20,21]. Here E is one of the irreducible representations of the tetrahedral point group, T_d. In contrast, the levels of the other irreducible representations exhibit second-order Stark shifts. In the course of the study, we found that the crossover signal was the largest in the triplet for the Q- and R-branch transitions [19].

This paper presents an analysis of the relative intensity in the triplet. The crossover signal indeed contains two crossover resonances in the Λ- and V-type three-level systems, which overlap each other in frequency. This fact was observed using a 3.39 μm He-Ne laser [18]. The observed intensity distribution agrees with a simple calculation based on steady-state solutions of rate equations in weak saturation using the observed intensity of the Lamb dips.

2. Relative intensity of saturated absorption signals

2.1. Observed triplets in Stark spectra of methane

We recorded sub-Doppler-resolution Stark spectrum of the ν_3 band of methane and observed a number of triplets of the

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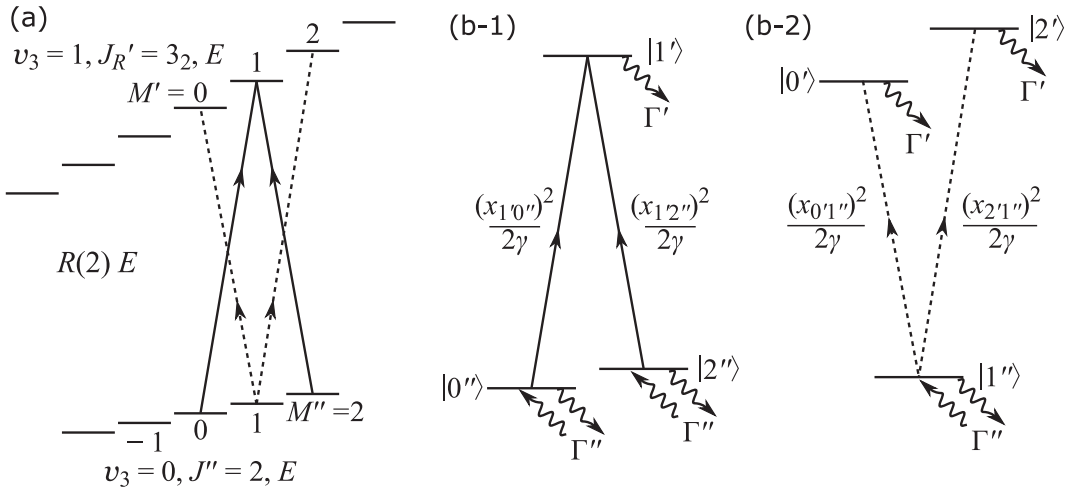

Fig. 1. (a) Energy level diagram of the $|\ v_3'' = 0, J'' = 2, E, M''\rangle$ and $|\ v_3' = 1, J_R' = 3_2, E, M'\rangle$ levels in the presence of the Stark field. Solid and dashed arrows indicate the Lamb dips of the Λ- and V-type three-level systems associated with the $M' = 1$ and $M'' = 1$ levels. Note that the magnitude of the energy separation is not scaled. (b) Rate equation models for the Λ(b-1)- and V(b-2)-type three-level systems. Here, x_{ij} is the Rabi angular frequency of the $|\ i\rangle \leftarrow |\ j\rangle$ transition, γ is a phase relaxation rate, and Γ' and Γ'' are population relaxation rates of the vibrational excited and ground states.

crossover signal and the associated Lamb dips [19]. Fig. 1(a) illustrates an energy level scheme of the $R(2)E$ transition in the presence of an electric field. Here, $|\ v_3'' = 0, J'' = 2, E, M''\rangle$ and $|\ v_3' = 1, J_R' = 3_2, E, M'\rangle$ are the lower (double-prime) and the upper (single-prime) levels of the transition; v_3 is a vibrational quantum number of the triply-degenerate v_3 stretching mode, J is the quantum number of the total angular momentum $\boldsymbol{J}$ defined as $\boldsymbol{J} = \boldsymbol{R} + \boldsymbol{l}$, where $\boldsymbol{R}$ and $\boldsymbol{l}$ are the rotational and vibrational angular momenta, and M is a projection of $\boldsymbol{J}$ onto the field direction. The rotational angular momentum quantum number R is attached to J as a subscript to identify the Coriolis-sublevels in the $v_3 = 1$ state. These E levels are subject to the first-order Stark effect, and the Stark shifts are thereby proportional to the value of M' or M''. The energy separation between the neighboring M levels in the $v_3 = 1$ state is at least an order of magnitude larger than that in the vibrational ground state because the vibration-induced permanent dipole moment in the $v_3 = 1$ state is larger by three orders of magnitude than the rotation-induced permanent dipole moment in the vibrational ground state [19].

The selection rule of the electric dipole transition is $E \leftrightarrow E$. Solid and dashed arrows in Fig. 1(a) indicate Λ- and V-type three-level systems of the $R(2)E$ transition, where the $M' = 1$ and $M'' = 1$ levels are shared in the Λ- and V-type three-level systems under the selection rule of $\Delta M = M' - M'' = \pm 1$. When the Stark field is strong enough to separate the neighboring M'' levels beyond the spectral resolution, a crossover signal and two Lamb dips are observed as a triplet for the Λ-type three-level system. Note that the crossover signal consists of two crossover resonances of the Λ- and V-type three-level systems, which overlap each other in frequency due to the first-order Stark effect. In contrast, two Lamb dips of the V-type three-level system are observed in frequency distant from the triplet because the Lamb dips are separated owing to the large Stark shift in the $v_3 = 1$ state. Hereafter, we focus on the triplets in the P-, Q-, and R-branch transitions of $J'' = 2$ to 7 associated with the $M' = \pm 1$ and $M'' = \pm 1$ levels.

2.2. Rate equation and absorption intensity

Fig. 1(b-1) illustrates a model based on rate equations for the Λ-type three-level system. The three levels are labeled M' or M''. The populations of the vibrational ground state, $N_{0''}$ and $N_{2''}$, decay at a rate of Γ'', and that of the vibrational excited state, $N_{1'}$, decays at a rate of Γ'. The optical field pumps the population of the vibrational ground state to the $v_3 = 1$ state at a rate of $(x_{i'j''})^2/2\gamma$ when $x_{i'j''} \ll \gamma$, where $x_{i'j''}$ is the Rabi angular frequency of the transition from the $j'' = M''$ level to the $i' = M'$ level and γ is the phase relaxation rate [22]. In the absence of the optical field, there is no population of the $M' = 1$ level in the $v_3 = 1$ state because the energy difference between the $v_3 = 1$ and vibrational ground states is much larger than the thermal energy at room temperature. The populations of the $M'' = 0$ and 2 levels in the vibrational ground state are both assumed to be N^{eq} in thermal equilibrium because the Stark shift between the two levels is the order of 1 MHz in frequency, and the corresponding energy difference is much smaller than the thermal energy at room temperature. The rate equations are then given as

$$\dot{N}_{0''} = \Gamma''(N^{\mathrm{eq}} - N_{0''}) - \frac{(x_{1'0''})^2}{2\gamma} N_{0''}, \tag{1}$$

$$\dot{N}_{2''} = \Gamma''(N^{\mathrm{eq}} - N_{2''}) - \frac{(x_{1'2''})^2}{2\gamma} N_{2''}, \tag{2}$$

and

$$\dot{N}_{1'} = -\Gamma' N_{1'} + \frac{(x_{1'0''})^2}{2\gamma} N_{0''} + \frac{(x_{1'2''})^2}{2\gamma} N_{2''}. \tag{3}$$

Here the Rabi angular frequency is given by

$$x_{1'M''} = \langle J''1; M''\Delta M | J''1; J'1\rangle \frac{\mu \mathcal{E}_{\mathrm{opt}}}{\hbar} \tag{4}$$

with $M'' = 0''$ or $2''$, where $\langle j_1 j_2; m_1 m_2 | j_1 j_2; JM\rangle$ is the Clebsch-Gordan coefficient with $\Delta M = \pm 1$, μ represents the magnitude of transition dipole moment of the v_3 band, $\mathcal{E}_{\mathrm{opt}}$ is the optical electric field amplitude, and $\hbar$ is the Dirac constant. Here we consider neither stimulated emissions nor vibrational relaxation processes between the $|\ 1'\rangle \leftrightarrow |\ 0''\rangle$ or $|\ 1'\rangle \leftrightarrow |\ 2''\rangle$ levels because saturation is weak, and rotational quenching processes are much faster than the vibrational one [23]. The detuning between the optical and transition frequencies is not explicitly considered because only the peak intensities of the triplet are of interest. Namely, the optical frequency is $\nu = \nu_{1'0''}$ and $\nu_{1'2''}$ for the Lamb dips, and $\nu = (\nu_{1'0''} + \nu_{1'2''})/2$ for the crossover resonance. Here $\nu_{1'M''} = (E_{1'} - E_{M''})/h$, where $E_{1'}$ and $E_{M''}$ are the