NUMERICAL EVALUATION OF THE SATURATION EFFECT IN THE PURELY ROTATIONAL SPECTRUM OF H_2O

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UDC 535

Numerical evaluations of the roles played by the various nonlinear effects are important in connection with the transport of intense monochromatic radiation in the atmosphere and in connection with the use of lasers in spectroscopic devices. Below we carry out a numerical evaluation for one such effect – the spectroscopic saturation effect [1, 2] – for the case of the purely rotational spectrum of H_2O vapor.

The saturation effect [1] consists of an additional broadening of a spectral line and reduction of the line intensity due to an equalization of the populations of the energy levels involved in the transition. We adopt the following arbitrary criterion for determining the degree of saturation of a spectral line:

$$\frac{S(\varepsilon^2)}{S(0)} = \frac{1}{2}, \tag{1}$$

where $S(\varepsilon^2)$ is the intensity of the line distorted by the saturation effect; and $S(0)$ is the intensity ordinarily used for spectroscopic purposes [3].

To find $S(\varepsilon^2)$ we use the usual definition of the line intensity in terms of the absorption coefficient $K(\omega, \varepsilon^2)$, which depends on the square of the field intensity ε as a parameter in the case of an intense field [1]:

$$S(\varepsilon^2) = \int_{-\infty}^{\infty} K(\omega, \varepsilon^2)\, d(\omega - \omega_0) \tag{2}$$

In the notation of [1], we can write $K(\omega, \varepsilon^2)$ as

$$K(\omega, \varepsilon_z^2) = \frac{4\pi\omega_0 N}{\hbar c}\left(1 - e^{-\frac{\hbar\omega_0}{\kappa T}}\right)\frac{e^{-\frac{E_b}{\kappa T}}}{Z}\sum_\lambda |(\mu_z)_{ab}^\lambda|^2 \frac{\gamma}{(\omega - \omega_0)^2 + \gamma^2 + |(\mu_z)_{ab}^\lambda|^2 \frac{\varepsilon^2}{c^2\hbar^2}} \tag{3}$$

[the frequency ω and the half-width γ in Eq. (3) are given in reciprocal centimeters]. From Eqs. (2) and (3) we find

$$S(\varepsilon^2) = \frac{4\pi^2\omega_0 N}{\hbar c Z}\left(1 - e^{-\frac{\hbar\omega_0}{\kappa T}}\right) e^{-\frac{E_b}{\kappa T}} \sum_\lambda |(\mu_z)_{ab}^\lambda|^2 \frac{\gamma}{\sqrt{\gamma^2 + |(\mu_z)_{ab}^\lambda|^2 \frac{\varepsilon^2}{c^2\hbar^2}}}. \tag{4}$$

TABLE 1

$J_\tau \rightarrow J'_{\tau'}$	γ cm^{-1}	L	ε [esu/cm]
$7_{-7}-6_{-1}$	0,068	0,17860	28,2395
$8_{-7}-7_{-3}$	0,080	0,02365	97,7990
$6_{-3}-5_5$	0,052	0,00022	567,9268
$2_{-2}-2_1$	0,095	0,8333	9,5683
$4_{-2}-4_0$	0,095	3,6541	6,0742
$12_{-4}-12_{-2}$	0.079	9,5872	5,1945
2_1-3_{-1}	0,095	0,3000	19,3008
$5_{-5}-6_{-1}$	0,080	0,1239	37,0108
$7_{-7}-8_1$	0,057	0,0006	417,5127

We can write the matrix element for the z component of the dipole moment in the basis of the wave functions of a rigid asymmetric top $\Psi_{J\tau m}$ (we assume that the matrix element in terms of the vibrational wave functions is equal to the constant dipole moment $\mu_0 = 1.884 \cdot 10^{-18}$ esu [4]):

$$(\mu_z)_{ab}^\lambda \rightarrow \mu_0 \langle J'\tau'm' \,|\, \Phi(z_y) \,|\, J\tau m \rangle.$$

Here $\Phi(z_y)$ is the direction cosine. Using the standard

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procedure for evaluating the matrix elements for direction cosines [5], we find the following expression for the sum over λ ($\sum_{mm'}$ in our notation):

$$\sum_m (1J0m \mid J'm)^2 \frac{3}{2J'+1} L\mu_0^2 \frac{\gamma}{\sqrt{\gamma^2 + (1J0m \mid J'm)^2 \frac{1}{2J'+1} L \frac{\varepsilon^2\mu_0^2}{c^2\hbar^2}}}.$$

Here L is the line strength tabulated, e.g., in [6]. Using $S(0) \sim \mu_0^2 L$ and Eq. (1), we find the following final expression for ε^2:

$$6\gamma \sum_m \frac{(1J0m \mid J'm)^2}{2J'+1} \frac{1}{\sqrt{\gamma^2 + \frac{(1J0m \mid J'm)^2}{2J'+1} L \cdot \mu_0^2 \frac{\varepsilon^2}{c^2\hbar^2}}} - 1 = 0. \qquad (5)$$

From Eq. (5) we can find ε in electrostatic units per centimeter; to express the results in volts per centimeter we must multiply the result by 300. Finally, with the solution of Eq. (5) we can find the intensity I (in watts per square centimeter) associated with this electric field from [7] I (W/cm^2) = (ε^2(V/cm)/361). A program was written for an M-20 computer to solve Eq. (5).

Some of the calculated results are shown in Table 1; the half-width γ and line strength L were taken from [8]. From this table we see that saturation is achieved most easily in the spectral regions in which the intense lines (L > 1) are grouped. Analysis of calculations for 60 lines from all three spectral branches shows that the field leading to saturation can be estimated from the simple equation $\varepsilon^2 \simeq (C/L)$, where C is a "constant," i.e., increases quite slowly with increasing half-width. At $\gamma = 0.07$ cm^{-1}, e.g., we have C $\simeq$ 150 for the P branch, C $\simeq$ 180 for the Q branch, and C $\simeq$ 185 for the R branch.

The authors thank T. K. Pasichenko for assistance in the calculations.

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