Investigation of light-induced drift of ammonia molecules

A. E. Bakarev, A. K. Folin, and P. L. Chapovskiĭ

Institute of Automation and Electronics, Siberian Division, USSR Academy of Sciences

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With ammonia molecules as the example, a theory that contains no free parameters is developed for the light-induced drift effect. An experimental investigation of the effect is carried out. The experimental and theoretical data agree within the limits of the measurement errors. It is established from a comparison of the experimental and theoretical data that the relative change of the diffusion coefficient of ammonia molecules excited by the vibrational mode is determined by the change of the dipole-dipole part of the interaction potential and amounts to $\Delta D/D = (2.5 \pm 0.4)\cdot 10^{-2}$.

INTRODUCTION

Considerable progress has by now been made in investigations of light-induced drift (LID)[1] of molecules.[1)] The LID effect was observed in a number of molecules, such as CH_3F (Ref. 3), NH_3 (Ref. 4), CH_3Br (Ref. 5), and CH_3OH (Ref. 6). A theoretical model providing a correct quantitative description of the effect was developed and compared with experiment.[7,8] The contribution of a number of concomitant effects in experiments on LID of molecules have been analyzed.[8,9] The use of the LID effect to separate isotopic[3,10] and nuclear spin modification of molecules has been demonstrated.

The still employed approach to the investigation of LID of molecules is not free, however, of a number of shortcomings. Thus, the parameter $\Delta\nu/\nu$ that enters in the theory of LID of molecules, equal to the relative change of the transport frequency of an excited molecule, remained free in this approach.[2)] To find this parameter it was necessary to compare the theory with LID experimental results. The frequency dependence of the LID effect could be measured only by using the effect itself, and its calculation called for precise spectroscopic data and was possible only for thoroughly studied objects.

We show in the present paper that the LID of molecules can be investigated using an approach free of the indicated shortcomings. We have developed a theory of the LID of ammonia molecules, investigated the effect experimentally, and compared the theory with the measurement results.

1. THEORY OF LID OF MOLECULES

We derive relations that describe the stationary distribution of the absorbing molecules in a cell with closed end faces. We consider the LID produced by a monochromatic traveling wave that is at resonance with the vibrational-rotational transition of the molecule. We assume that the buffer-particle concentration ρ_b in the cell is much higher than the concentration ρ of the absorbing molecules.

The velocity and rotational-level distributions of the molecules in excited $\rho_m(J,\mathbf{v})$ and $\rho_n(J,\mathbf{v})$ vibrational states are described by the kinetic equations[13]

$$\begin{aligned} \mathbf{v}\nabla\rho_m(J,\mathbf{v}) &= S_m(J,\mathbf{v})+\rho P_J(\mathbf{v}), \\ \mathbf{v}\nabla\rho_n(J,\mathbf{v}) &= S_n(J,\mathbf{v})-\rho P_{J+\sigma}(\mathbf{v}). \end{aligned} \tag{1}$$

Here $S_\tau(J,\mathbf{v})$ with $\tau = m, n$ are the collision integrals of the absorbing and buffer molecules; $\rho P_J(\mathbf{v})$ is the rate of excitation of the molecules by radiation into the state J, $\mathbf{v}$; the subscripts $\sigma = 0$, -1, and $+1$ correspond to the Q, P, and R absorption lines of the rotational modes. The spontaneous decay of the vibrational level is neglected in (1).

To calculate the spatial distribution of the concentration of the absorbing molecules, we multiply Eq. (1) by $\mathbf{v}$, integrate with respect to $\mathbf{v}$, sum over J, and add the resultant equations. We get ultimately

$$\int \mathbf{v}(\mathbf{v}\nabla\rho(\mathbf{v}))\,d\mathbf{v} = \sum_{J,\tau}\int S_\tau(J,\mathbf{v})\,\mathbf{v}\,d\mathbf{v}, \quad \rho(\mathbf{v}) = \sum_{J,\tau}\rho_\tau(J,\mathbf{v}). \tag{2}$$

The LID effect produces in molecular systems a weak disequilibrium in the total velocity distribution of the absorbing molecules. We can therefore assume approximately that $\rho(\mathbf{v}) \approx \rho W(\mathbf{v})$, where $W(\mathbf{v})$ is the Maxwell distribution in velocity.

For the moments of the collision integrals in (2) we use, following Ref. 14, the representation

$$\int S_\tau(J,\mathbf{v})\,\mathbf{v}\,d\mathbf{v} = -\nu_\tau \mathbf{j}_\tau, \quad \mathbf{j}_\tau = \sum_J \int \rho_\tau(J,\mathbf{v})\,\mathbf{v}\,d\mathbf{v}, \tag{3}$$

where ν_τ is the transport frequency of the collisions of the absorbing particles in the state τ with the buffer ones. We assume the frequencies ν to be independent of the characteristics of the laser emission. Taking Eqs. (3) and the approximation $\rho(\mathbf{v}) = \rho W(\mathbf{v})$ into account, we get from (2)

$$\tfrac{1}{2}v_0^2\nabla\rho = -\nu_m \mathbf{j}_m - \nu_n \mathbf{j}_n, \quad v_0 = (2k_B T/M)^{1/2}. \tag{4}$$

In the experiments described below we use long and thin cells ($L/R \sim 10^3$). This makes it possible to change over in (4) to characteristics averaged over the cross section and to neglect the small inhomogeneity of the concentration of the absorbing particles in the transverse direction:

$$\tfrac{1}{2}v_0^2 d\rho/dz = -\nu_m j_m - \nu_n j_n, \tag{5}$$

where j_m and j_n are the fluxes of the excited and unexcited particles, averaged over the cell cross sections. Under stationary conditions the total flux of the absorbing particles is zero in any cross section of the cell, therefore $j_m = -j_n$ and we get from (5)

$$\tfrac{1}{2}v_0^2 d\rho/dz = -(\nu_m - \nu_n) j_m. \tag{6}$$

Since a small spatial inhomogeneity influences little the flux j_m, it can be obtained from the locally homogeneous equation (1). Repeating the transformations described above, we get

$$j_m=\rho\,\overline{\int P(\mathbf{v})v_z\,d\mathbf{v}/\mathrm{v}_m},\quad P(\mathbf{v})=\sum_J P_J(\mathbf{v}),\tag{7}$$

where the superior bar denotes averaging over the cell cross section. Combining next Eqs. (6) and (7), we get

$$\frac{d\rho}{dz}=-\frac{\Delta\mathrm{v}}{\mathrm{v}}\frac{2}{v_0{}^2}\rho\,\overline{\int P(\mathbf{v})v_z\,d\mathbf{v}},\quad \frac{\Delta\mathrm{v}}{\mathrm{v}}\equiv\frac{\mathrm{v}_m-\mathrm{v}_n}{\mathrm{v}_m}.\tag{8}$$

To continue, we must specify the form of the function $\rho P(\mathbf{v})$. An approximation widely used in nonlinear spectroscopy[13] is

$$\rho P_J(\mathbf{v})=\frac{2\Gamma|G_J|^2}{\Gamma^2+(\Omega_J-\mathbf{kv})^2}[\rho_n(J+\sigma,\mathbf{v})-\rho_m(J,\mathbf{v})],\tag{9}$$

$$G_J=\frac{Ed_J}{2\hbar},\quad \Omega_J=\omega-\omega_J,\quad k=\frac{\omega}{c},$$

where ω_J, Γ, and d_J are the frequency, homogeneous half-width, and dipole moment of the transition $J+\sigma\rightarrow m,J;\omega$ and E are the frequency and amplitude of the electromagnetic wave. The relations we need can be obtained, following Ref. 15, using for the form of $\rho P(\mathbf{v})$ an assumption more general than (9), viz.,

$$\rho P(\mathbf{v})\sim Sf(\omega-\mathbf{kv})[\rho_n(J+\sigma,\ \mathbf{v})-\rho_m(J,\ \mathbf{v})],\tag{10}$$

where f is a certain arbitrary function of $\omega-\mathbf{k}\cdot\mathbf{v}$.

We assume also that the laser radiation is absorbed without saturation. It will be shown below that this is precisely the situation realized under our experimental conditions. Then

$$\rho_n(J+\sigma,\ \mathbf{v})-\rho_m(J,\ \mathbf{v})\approx\rho_n(J+\sigma,\ \mathbf{v})\approx\rho W_B(J)W(\mathbf{v}),\tag{11}$$

where W_B is the Boltzmann distribution of the particles over the rotational levels. Using the equality

$$\int P_J(\mathbf{v})v_z\,d\mathbf{v}=-\frac{kv_0{}^2}{2}\frac{d}{d\omega}\int P_J(\mathbf{v})\,d\mathbf{v},\tag{12}$$

which is valid under the assumptions (10) and (11), and recognizing that the linear absorption coefficient is $\alpha=\hbar\omega\rho P/S, P\equiv\int P(\mathbf{v})d\mathbf{v}$, we get

$$\frac{d\rho}{dz}=\frac{\Delta\mathrm{v}}{\mathrm{v}}\overline{\rho P}\frac{k}{\alpha/\rho}\frac{d(\alpha/\rho)}{d\omega}.\tag{13}$$

The factor

$$\frac{1}{\alpha/\rho}\frac{d(\alpha/\rho)}{d\omega}$$

was taken out in (13) from under the averaging sign, since it is independent of the coordinates in the absence of saturation.

It is convenient to express the quantity $\overline{\rho P}$ in (13) in terms of the radiation power concentration W_g absorbed by the gas per unit cell length

$$W_g=\overline{\rho P}\hbar\omega.\tag{14}$$

Using (13) and (14), we easily obtain an equation that relates the drop $\Delta\rho$ in concentration of the absorbing molecules per unit cell length with the radiation power concentration ΔS_g absorbed by the gas:

$$\Delta\rho=\frac{\Delta\mathrm{v}}{\mathrm{v}}\frac{\Delta S_g}{\hbar\omega}\frac{k}{\alpha/\rho}\frac{d(\alpha/\rho)}{d\omega}.\tag{15}$$

The signs in this equations are chosen such that positive $\Delta\rho$ correspond to the increase ΔS_g of the concentration along the radiation propagation direction is always positive.

Equations such as (15), which relate the drop of the concentration of the absorbing particles to the absorbed radiation power density, were first obtained in LID theory in Ref. 7. Calculation of $\Delta\rho$ by the method developed in Ref. 7 called for high-precision spectroscopic data, such as the positions of the absorption lines, their homogeneous widths, and others. In calculations using (15), however, it is possible to dispense with these data by using direct measurements of the functions $\alpha^{-1}d\alpha/d\omega$ provided that ρ has a negligible dependence on the radiation frequency (that the LID effect is small), when

$$\frac{1}{\alpha/\rho}\frac{d(\alpha/\rho)}{d\omega}=\frac{1}{\alpha}\frac{d\alpha}{d\omega}.$$

2. CALCULATION OF THE FACTOR $\Delta\mathrm{v}/\mathrm{v}$

The representation (3) with transport collision frequencies that are independent of the radiation characteristics makes it possible to connect the factor $\Delta v/v$ with the change of the diffusion of the excited molecule:

$$\Delta\mathrm{v}/\mathrm{v}=-\Delta D/D,\quad \Delta D/D\equiv(D_m-D_n)/D_m.\tag{16}$$

As shown in Ref. 16, for ammonia molecules in a polar buffer gas, the change of the diffusion coefficient is due to the change of the dipole-dipole part of the intermolecular potential. We calculate $\Delta D/D$ by the numerical method proposed in Ref. 16. The diffusion coefficient of the polar molecules is given by[17]

$$D=\frac{3}{8(\rho+\rho_b)}\left(\frac{k_BT}{\pi M}\right)^{1/2}(\sigma_0\langle\Omega^{(1,1)*}\rangle)^{-1},\tag{17}$$

where σ_0 is the Stockmayer-potential parameter and $\Omega^{(1,1)*}$ is the reduced Chapman-Cowling integral. The angle brackets denote averaging over the mutual orientations of the colliding molecules.

In our case, the molecule excitation changes only the dipole-dipole part of the intermolecular potential; we therefore obtain from (17)

$$\frac{\Delta D}{D}=-\frac{d\langle\Omega^{(1,1)*}\rangle}{d\delta}\frac{\delta}{\langle\Omega^{(1,1)*}\rangle}\frac{\Delta\mu_1}{\mu_1},\tag{18}$$

where $\delta=\mu_1\mu_2/2\varepsilon_{12}\sigma_{12}{}^3$; ε, and μ are the parameters of the Stockmayer potential; is the dipole moment of the molecule; the indices 1 and 2 pertain respectively to the absorbing and buffer particles.

To calculate $\Delta D/D$ from Eq. (18) we must know the change $\Delta\mu_1/\mu_1$ of the dipole moment. This quantity can be calculated for each rotational transition in the vibrational band v_2 from the data of Ref 18. To compare the value of $\Delta\mu_1/\mu_1$ obtained in this manner with experiment, however, it is necessary to average over the rotational levels, since the molecules' LID is actually determined by diffusion coefficients averaged over J. The lack of detailed information on the rotational-relaxation processes leads in the upshot to a noticeable error in the calculation of the relative change of the dipole moment: $\Delta\mu_1/\mu_1 = -0.16 \pm 0.02$. The calculation of $\Delta D/D$ can be carried out further by numerically differentiating the function $\Omega^{(1,1)*}$ tabulated in Ref 17. The parameters σ_0 and ε needed for the calculations are also given in Ref. 17. We ultimately get

$$\Delta D/D \approx 2.5\cdot 10^{-2}. \tag{19}$$

3. ALLOWANCE FOR RADIATION ABSORPTION BY THE CELL WALLS

According to (15), the concentration drop of the absorbing molecules in proportional to the concentration ΔS_g of radiation power absorbed by the gas. This quantity is difficult to measure because of the absorption by the cell walls. It will be shown in the present section that the radiation loss to the walls can be accounted for if the radiation is absorbed by the gas and by the walls in a linear regime, with absorption coefficients that are independent of the radiation intensity.

The radiation power densities W_w and W_g absorbed per unit cell length by the walls and by the gas are connected by the relation

$$W_w = \gamma W_g/\alpha, \tag{20}$$

where γ and α are the coefficients of radiation absorption by the cell walls and by the gas. Combining eqs. (13), (14), and (20), we obtain

$$\frac{1}{\rho}\frac{d\rho}{dz} = \frac{\Delta\nu}{\nu}\frac{\alpha}{\gamma\rho}\frac{W_w}{\hbar\omega}\frac{k}{\alpha}\frac{d\alpha}{d\omega}. \tag{21}$$

Equation (21) can be easily integrated (α is by assumption independent of the radiation intensity):

$$\ln\frac{\rho_2}{\rho_1} = \frac{\Delta\nu}{\nu}\frac{\alpha}{\gamma\rho}\frac{\Delta S_w}{\hbar\omega}\frac{k}{\alpha}\frac{d\alpha}{d\omega}, \tag{22}$$

where ρ_2 and ρ_1 are the concentrations of the absorbing molecules at the ends of the cell, and ΔS_0 is the radiation power density absorbed by the walls, referred to the cell cross section. Combining next Eqs. (15) and (22) we obtain

$$\Delta S_g \ln\frac{\rho_2}{\rho_1} = \frac{\alpha}{\gamma\rho}\Delta\rho\Delta S_w. \tag{23}$$

To determine experimentally the power density S_g absorbed by the gas it is necessary to measure the radiation power at the exit from a cell without gas (S_1) and with gas in the cell (S_2). Of course, S_2 should be measured when the stationary state of the light-induced separation has already been reached. Using (23), we obtain for the power density absorbed by the gas

$$\Delta S_g = [S_1 \exp(\gamma L) - S_2][1 + \gamma\rho \ln(\rho_2/\rho_1)(\alpha\Delta\rho)^{-1}]^{-1}. \tag{24}$$

Without allowance for the radiation absorption by the cell walls ($\gamma = 0$) we have $\Delta S_g = S_1 - S_2$. Allowance for absorption by the walls increases ΔS_g in our case by 10–15%.

4. AMMONIA LID MEASUREMENT PROCEDURE

The LID of ammonia molecules was investigated by separating the isotopic modification of NH_3. As in Ref. 4, the absorbing molecules were ${}^{15}NH_3$ (absorption line $aR(6, 0)$ of ν_2 oscillation), and the buffer molecules were ${}^{14}NH_3$. We used the the natural isotope mixture ${}^{15}NH_3:{}^{14}NH_3 = 1:270$.

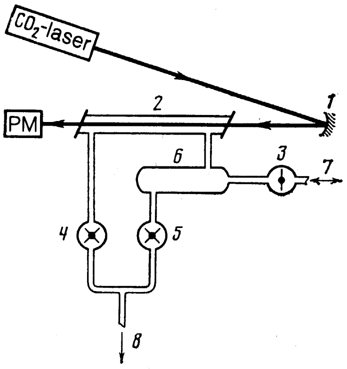

FIG. 1. Diagram of experimental facility for the study of the LID effect: 1—spherical mirror ($R = 1$ m), 2—cell with gas, 3—valve, 4 and 5—proportioning valves, 6—ballast volume, 7—vacuum duct for inlet and evacuation of gas mixture, 8—vacuum duct to mass.

Ammonia is prone to strong physical adsorption. Under these conditions, reliable information on the magnitude of the LID can be obtained from the stationary values of he isotopic enrichment, which we characterize by an enrichment coefficient $\beta = K_0/K - 1$, where $K_0 = (\rho/\rho_b)_0$ is the gas composition established by the LID effect, and $K = \rho/\rho_b$ is the initial composition. Since $\rho \ll \rho_b$ in the experiments, the enrichment is simply connected with the decrease of the concentration of the absorbing molecules:

$$\Delta\rho = \beta\rho. \tag{25}$$

A diagram of the setup for the LID investigation is shown in Fig. 1. The ammonia molecule drift is effected in a copper cell with inside diameter 3 mm and length 1.5 m. The system had two valves with the aid of which it was possible to extract and analyze the compositions of gas batches from the cell and from the ballast volume. The isotope composition and enrichment were measured with an MI-1201 mass spectrometer. Owing to the strong adsorption of the ammonia the measurement of the mixture composition called for repeated "scrubbing" of the analytic part of the mass spectrometer by samples of the investigated mixture.

The gas composition was determined by measuring the ${}^{15}NH_3$ and ${}^{14}NH_3$ ion currents. Serious difficulties were raised by the proximity of the peaks of ammonia (${}^{15}NH_3$) and water, traces of which were present in the vacuum system. It became possible to measure the ${}^{15}NH_3$ peak amplitude reliably at a mass spectrometer resolution ~ 3000 (in which case the ammonia and water peak are well resolved).

To investigate the ammonia LID we used a low-pressure CO_2 laser operating on the R 10 line of the CO_2 vibrational band 02^00–00^01. The rotational bands were selected by a diffraction grating in the laser cavity. The laser frequency was stabilized by an electronic system setting the CO_2 center at the maximum of the lasing power. The output radiation power was ~ 10 W. The linearly polarized CO_2 laser radiation was focused by a spherical mirror ($f = 50$ cm) on the entrance end of the cell. The confocal parameter (kw_0^2) past the mirror was 20 cm. The TEM_{00} mode of the CO_2 laser is effectively converted in the linearly polarized EH_{11} waveguide mode. The field distribution in the cell was measured in Ref. 8. We calculate the absorption coefficient for the EH_{11} mode from the equation[19]

$$\gamma \approx \left(\frac{2{,}41}{k}\right)^2 \frac{1}{R^3} \mathrm{Re}\left[\frac{n^2+1}{2(n^2-1)^{1/2}}\right], \tag{26}$$

where R is the radius of the tube and n is the complex refractive index $n = 14.2 - i64.5$ for copper.[20] The absorption coefficient of a copper tube with $R = 1.5$ mm is $\gamma = 1.22\cdot 10^{-3}$ cm^{-1}, and the transmission of a tube 1.5 m long is 0.83. The measured cell transmission was noticeably lower, about 0.7. This is due to losses in matching the TEM_{00} mode of the CO_2 laser to the EH_{11} mode of the waveguide. Clearly, the radiation loss to matching hardly distorts the results of the calculation of ΔS_g from Eq. (24).

5. MEASUREMENT OF THE SPECTROSCOPIC CHARACTERISTICS

The approach used in the present paper to the LID problem obviates the need for detailed data on the absorbed transition of the molecule. To calculate the LID from (15) it is necessary only to measure the logarithmic derivative of the absorption coefficient normalized to the density of the absorbing particles.

The measurement setup is shown in Fig. 2. The radiation source was a waveguide CO_2 laser (its description can be found in Ref. 21) using ~1 W of radiation and frequency tuning range ~300 MHz. The laser beam passed through a collimator (light-beam diameter 6 mm) and was split into two. One beam passed through the measurement cell (1.6 cm diameter, length 40 cm) with the investigated gas and was incident on photoreceiver 3. The second beam served as the reference and was recorded by photoreceiver 4. The signals from the two photoreceivers were processed by electronic circuitry in analog form, such that the output signal was proportional to the absorption coefficient of the gas in the cell. This signal was fed to Y input of an X-Y plotter. The X input was an electric signal proportional to the voltage across a piezoceramic element used to vary the length of the cavity of the CO_2 laser. The nonlinearity of the piezoceramic was taken into account in the measurement of the frequency dependence of the absorption coefficient.

A typical plot of the absorption-coefficient frequency dependence is shown in Fig. 3. Plots of this form, measured at different pressures, were used to calculate $\alpha^{-1}d\alpha/d\omega$ at the frequency of the center of the $9R$ 10 line of the CO_2 laser (in view of the large cross section of the measuring cell and the low radiation power, the frequency dependence of ρ as a result of the LID effect can be neglected, and it can be assumed that $(\alpha/\rho)^{-1}d(\alpha/\rho)/d\omega = \alpha^{-1}d\alpha/d\omega)$. These measurements were made both with gas having a natural isotope composition and with enriched gas.

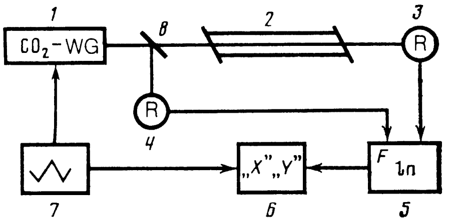

FIG. 2. Diagram of facility for the measurement of the absorption coefficient: 1—waveguide CO_2 laser, 2—cell with gas, 3 and 4—photoreceivers, 5—differential logarithmic amplifier, 6—x-y plotter, 7—laser-frequency control block, 8—beam-splitting plate.

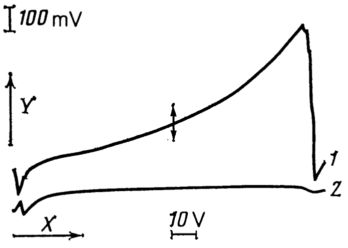

FIG. 3. Typical plot of absorption spectrum in the tuning range of the waveguide CO_2 laser—curve 1 (the arrow shows the position of the center of the CO_2 laser $9R$ 10 line). Curve 2 was obtained in the absence of absorbing gas.

In another approach to the analysis of LID of molecules (see, e.g., Refs. 3, 10, and 16), the laser-frequency function $\alpha^{-1}d\alpha/d\omega$ is not measured but calculated. It was difficult to use this approach for ammonia for lack of unambiguous data on the absorption-line parameters. Thus, for example, the published data on the deviation of the ammonia absorption-line frequency from the CO_2 laser 9R10 line range from $\Omega = -125$ MHz (Ref. 22) to $\Omega = -306$ MHz (Refs. 23 and 24).

6. ANALYSIS OF LID MEASUREMENT RESULTS

The theoretical LID model and the calculation of the radiation power absorbed by the gas, described in the present paper, are valid in the absence of field saturation. The gas pressure region in which this condition is met was found by experiment. Figure 4 shows a plot of α/α_0 vs the gas pressure. Here α is the gas absorption coefficient in the separation cell (Fig. 1) in a strong fields $S \sim 10^2$ W/cm^2, and α_0 is the unsaturated absorption coefficient obtained by measurements in a weak field $S \sim 1$ W/cm^2 (Fig. 2). It can be seen from the data of Fig. 4 that at pressures exceeding 7 Torr the field saturation in the separation cell can be neglected at our radiation intensities.

The model developed above for the LID of ammonia molecules contains no free parameters. The validity of the model can therefore be verified by comparing with experiment both the magnitude of the effect and its dependence on the parameters of the problem. Figure 5 shows the measured

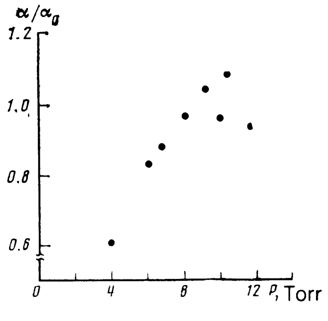

FIG. 4. Ratio of absorption coefficient in the gas in a strong (α) and weak (α_0) field at various gas pressures in the cell.

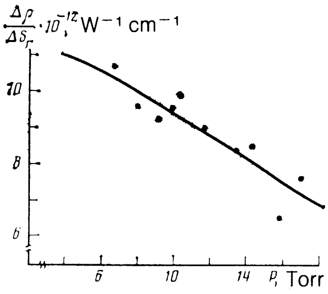

FIG. 5. Results of measurement (●) and calculation (solid line) of the normalized concentration drop $\Delta\rho/\Delta S_g$ at various gas pressures in the cell.

and calculated normalized concentration drop $\Delta\rho/\Delta S_g$ at various gas pressures in the cell. The measurement of $\Delta\rho/\Delta S_g$ consisted of measurement of β, S_1, and S_2 and calculation of $\Delta\rho$ and ΔS_g from Eqs. (25) and (24). The calculation of $\Delta\varphi/\Delta S_g$ consisted of calculation of $\Delta v/v$ and measurement of $\alpha^{-1}d\alpha/d\omega$ at various pressures.

The data of Fig 5 show that, within the limits of the $\Delta\rho/\Delta S_g$ measurement errors, the ammonia-molecules LID model developed in the present paper agrees with experiment. The most important are, in our opinion, three circumstances: the validity of relation (16), the possibility of calculating the factor $\Delta D/D$ only with allowance for the dipole-dipole part of the intermolecular potential, and the possibility of determining the first moment of the function $P(\mathbf{v})$ by measuring the frequency dependence of the absorption coefficient.

The change of the ammonia-molecule diffusion coefficient, determined from the experimental data using Eqs. (15) and (16), is $\Delta D/D = (2.49 \pm 0.11)\cdot 10^{-2}$ (with a fiducial probability 0.9). In our opinion, the possible systematic error of this quantity does not exceed 10% and is determined mainly by the radiation-power-density measurement error.

In conclusion, the authors thank F. Kh. Gel'mukhanov, A. I. Parkhomenko, A. M. Shalagin, and D. A. Shapiro for support and for helpful discussions.

[1)] A bibliography of experimental work on LID of atoms can be found, for example, in Ref. 2.

[2)] Progress was made in a recent theoretical paper[12] devoted to the calculation of the $\Delta v/v$ factors of CH_3F, NH_3, and SF_6.

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