

Analytical solutions for infrared-laser-driven diffusion in polyatomic gas mixtures

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We use an effective bimodal velocity distribution in a five-level model to analytically examine the problem of laser-driven gas transport in mixtures of an absorbing gas and a buffer gas. Analytic expressions are given for the steady-state active-gas density distribution in a closed tube for a uniform excitation profile. The model also predicts that laser-induced transport may occur without a buffer gas if collisions favoring simultaneous vibrational deactivation and velocity change occur. Profiles are given for $\text{CH}_3\text{F}:\text{He}$ mixtures when a CO_2 laser is used to excite the $Q_{12,2}$, ν_3 band absorption.

INTRODUCTION

In 1979, Gel'mukhanov and Shalagin reported on the effect of light-induced diffusion (LID) in gases.¹ In a later paper that year the use of the method for estimating transport coefficients was discussed.² In 1980 a more rigorous theory for the LID effect for an ideal two-level system was presented.³ This work was based on the solution of the coupled set of density-matrix and kinetic equations. Although this work clearly explains the effect, it could not be readily applied to polyatomic systems where several degrees of freedom and many quantum states are involved.

The experimental efforts in LID were first performed by Ansignin and Atutov.⁴ These experiments were the first deliberate measurements, while LID effects may have been observed and unexplained by others previously.⁵ The work in Ref. 4 was performed under flowing conditions and utilized sodium vapor as the absorber and helium as the buffer gas. Although these were not closed-tube experiments, a two-level model calculation was given for the profile which might exist in such an experiment. The calculations indicated that at modest dye-laser intensities all the sodium could be confined to a length less than 1 mm. This experimental result led to questions concerning the use of this method as a gas separation scheme for polyatomics which absorb in the infrared.

The first such experiment was performed on SF_6 in H_2 and He mixtures.⁶ These experiments were performed using the CO_2 laser 9P(20) excitation of the SF_6 and measured by spectrophotometer and mass-spectroscopy methods at the end of the cell. Subsequent to this work another experiment was performed in order to more specifically address the questions of gas separation. These experiments were performed on $^{12}\text{CH}_3\text{F}:\text{He}$ mixtures.⁷

The most recent publication dealing with the LID effect in polyatomics is the first to discuss the significantly more complicated theoretical questions which arise in complicated molecules absorbing infrared radiation.⁸ This paper presents a reasonably complete set of coupled rate equations but fails to compare results of mass-spectroscopy experiments to the theory. The equations given in Ref. 8 can only be treated numerically and serve little purpose for identifying the importance of the various rates in the LID effect. Without any analytic results it is difficult to understand the LID effect in polyatomics.

In this paper, we have modeled the interaction of a complicated polyatomic with the radiation field. In addition to an analytic solution for the gas profile in a closed cell, we predict a new effect in LID. The model predicts that LID may occur even if the excited- and ground-state transport cross sections are equal, if collisions favoring simultaneous vibrational deactivation and velocity direction change occur.

KINETIC MODEL

The essence of the LID phenomenon is the following. Let the frequency ν of a plane monochromatic wave slightly differ from the frequency ν_0 of the transition of the molecule. Then because of the Doppler effect the velocity distribution in the ground and excited states becomes asymmetrical. Depending on the ratio of homogeneous to inhomogeneous widths, the asymmetry can vary in its shape. The most important effect of this asymmetry is that the average velocity in each state is no longer equal to zero. Hence, in each of the states involved there is directed movement which can be characterized by flows.

At this point it is interesting to note that the *size*

of an atom is different depending on the state of excitation. This is a hard-sphere picture describing the state dependence of the transport coefficients. When the absorbing atoms or molecules are mixed with another gas, the presence of the second gas produces friction with the flows of the absorbing species. However, the friction forces for excited and nonexcited atoms are different due to the difference in transport coefficients. Thus a net force emerges with which the second buffer gas acts on the absorbing atoms and causes them to have directed motion.

The laser-induced diffusion effect in a closed system can be said to reach equilibrium when the flow due to Fickian diffusion exactly balances the laser-induced flow. Since the system is closed, the total pressure along the cell will be constant while the partial pressures of the active gas and the buffer gas will be functions of position. Calling the flux due to the LID effect J_A , and the density of the active gas ρ_A , we have the equilibrium condition

$$\vec{J}_A = D_{AB} \vec{\nabla} \rho_A .$$

D_{AB} is the mutual diffusion coefficient of the gas mixture. This equation is the starting point for our theory for polyatomic LID. The rate-equation model developed here will be used to determine J_A .

We derive the general form for J_A based on a model of a mass moving through a dissipative medium (buffer gas) driven by a constant force (thermal translational bath). This is based on viewing dissipation as any process that changes the direction of an active-gas molecule. The equation of motion for such a system is given by

$$m\ddot{x} + \beta\dot{x} = F .$$

The solution of this equation yields a limiting velocity

$$v_0 = F/\beta .$$

β is a function of buffer-gas density, mass, and collision cross section with the active molecule in the particular quantum state of interest. We are, therefore, interested in the limiting velocity for active molecules in the ground vibrational state and those in the excited vibrational state connected by the pump laser.

The macroscopic flow J_A will be a result of velocity distributions summed over the relevant quantum states of the polyatomic system. The derivation of J_A will be given in terms of the molecular velocity group selected by the laser, V , and the ratio $\beta_g/\beta_e = \phi$. β_e and β_g are the friction constants for the excited vibrational state and the ground state, respectively. The derivation of an estimate for ϕ is given in the Appendix.

In order to avoid the problems associated with a velocity distribution function, we have developed a model which utilizes a bimodal velocity distribution function in conjunction with a three-level rate-equation approximation for the optical-pumping process. The use of a single speed and only two directions is reasonable since cross sections for elastic processes are nearly velocity independent. With this approximation the effect of only having two velocities can be corrected for by use of an effective interacting population. In addition, since velocity averaged relaxation rates obtained from other experiments are utilized for the vibrational and rotational processes, these parameters are insensitive to the form of the velocity distribution function.

The level scheme which results is shown with the associated rates in Fig. 1(a). The + and - subscripts refer to molecular velocities copropagating and counterpropagating with the laser field. The levels and rates used in the model are defined as follows: $|0\rangle$, rotational state of the ground vibrational level connected by the pump transition; $|1\rangle$, rotational state of the excited vibrational level connected by the pump transition; $|2\rangle$, level representing the remainder of the population of the rotational manifold of the excited vibrational state; W_p , velocity equilibration rate; W_r , rotational equilibration rate; η , rate of vibrational relaxation from all remaining

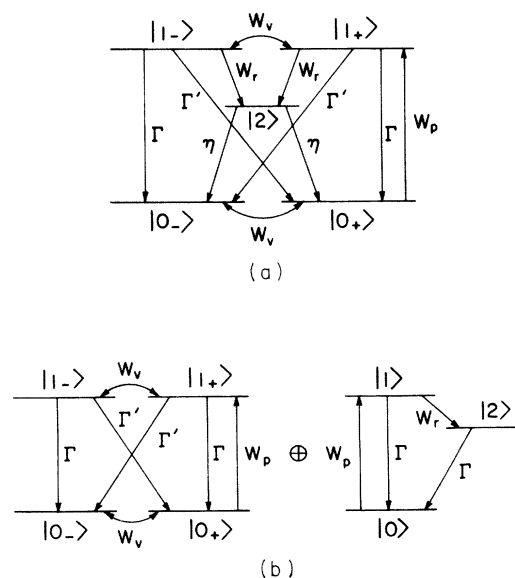


FIG. 1. (a) Five-level model with bimodal velocity distribution. (b) Two-model approximation to the five-level scheme utilized to generate the rate equations.

levels in the excited vibrational state; Γ , rate of vibrational relaxation without velocity change from the upper rotational state connecting the pump transition; Γ' , rate of simultaneous vibrational relaxation and velocity exchange from the upper rotational state connecting the pump transition; W_p , rate of pumping.

The rates defined above are all functions of buffer-gas density with the exception of W_p (for the Doppler-broadened limit only). The rate of rotational equilibration is assumed to be composed of a self-equilibration term, given by $\pi \Delta v$ where Δv is the self-broadened linewidth, and a term proportional to the buffer-gas density. Rates η , Γ , and Γ' can have two types of pressure dependences in the usual case since at low pressures wall collisions can play an important role in vibrationally deactivating some molecular species.⁸ In the LID problem, however, we exclude the $1/p$ diffusion dominated relaxation path for the states $|1_+\rangle$ and $|1_-\rangle$ since once a molecule changes direction toward the wall it is lost from the flow equation. However, η must include both relaxation by wall collision as well as by binary collisions. The general form for η in terms of the active-gas density ρ_A and the buffer-gas density ρ_B is given by

$$\eta = \frac{A}{(\rho_A + \rho_B)d^2} + B\rho_A + C\rho_B.$$

A is a constant proportional to the mutual diffusion coefficient of the mixture, d is the cell diameter, and B and C are homogeneous and heterogeneous deactivation rate constants, respectively.

The rate W_v which represents the loss of a velocity asymmetry is estimated using kinetic theory solutions for the problem of a nonequilibrium gas mixture.⁹ The result is clearly dependent on density as well as the mass ratios of the active and buffer gas since momentum transfer is the relevant variable. The rate utilized is based on the derivation in Ref. 9 and is given by

$$W_v = \frac{\frac{16}{3}(2\pi kT)^{1/2} \frac{\sigma}{\pi} \left(\frac{m_a}{m_b} \right)^{1/2}}{m_a^{1/2} \left[\frac{m_a}{m_b} + 1 \right]^{3/2}},$$

where σ is the ground-state hard-sphere collision cross section, T is the average temperature, and m_a and m_b are the masses of the active and buffer gases, respectively.

Finally, we conclude the discussion of the relevant rates with the form of W_p . In order to analytically

treat the problem at hand we use an expression for W_p which includes a mixed-broadening linewidth to include Doppler, collisional, and Rabi effects. The average linewidth is given by

$$(\Delta v_T)^2 \simeq (\Delta v_h)^2 + (\Delta v_D)^2 + \Omega^2.$$

Δv_h , Δv_D , and Ω are the homogeneous, Doppler-broadened, and power-broadened linewidths, respectively. The result is a Doppler linewidth function which is modified by the substitution of Δv_T . Calling this function $g(v)$, we have the following expression for $W_p(v)$:

$$W_p(v) = \frac{Ic^2 g(v)}{8\pi n^2 h v^3 t_{sp}}.$$

I is the intensity of the pump laser, n is the index of refraction, h is Planck's constant, t_{sp} is the spontaneous lifetime of the pump transition, and c is the speed of light.

The rate equations for the scheme shown in Fig. 1(a) result in a set of five linearly independent equations. This set was difficult to solve and resulted in an overly complicated expression involving terms having large powers of the rates which did not contribute significantly to the result. In order to better understand the effects of the various rates, we have developed an equivalent scheme based on solving two separate level models and combining the results. Since $W_v > W_r$, the molecules which were involved in the $|1_+\rangle \rightarrow |2\rangle$ and $|1_-\rangle \rightarrow |2\rangle$ processes no longer possess velocity information. This is equivalent to the assumption that once a molecule having a well defined velocity ($|1_+\rangle$ or $|1_-\rangle$) suffers a rotational state-changing collision, it statistically no longer carries any velocity information. In the rate equations this is reflected by level $|2\rangle$ having equal numbers of copropagating and counterpropagating molecules.

Based on these assumptions, the scheme shown in Fig. 1(a) may be decomposed into the combination of schemes shown in Fig. 1(b). The left-hand side of Fig. 1(b) describes the velocity asymmetry induced in the molecules participating in the cycle and the right-hand side is used to account for the storage effect of the lumped rotational manifold of the excited vibrational state. Thus the two models can be solved simultaneously with a conservation of particles equation relating them.

RATE EQUATIONS

The rate equations associated with the models shown in Fig. 1(b) are given as follows:

$$\begin{aligned}\dot{N}_0^+ &= W_p(N_1^+ - N_0^+) + \Gamma N_1^+ + \Gamma' N_1^- \\ &\quad - W_v(N_0^+ - N_0^-), \\ \dot{N}_0^- &= \Gamma N_1^- + \Gamma' N_1^+ + W_v(N_0^+ - N_0^-), \\ \dot{N}_1^+ &= W_p(N_0^+ - N_1^+) - \Gamma N_1^+ - \Gamma' N_1^+ \\ &\quad - W_v(N_1^+ - N_1^-), \\ \dot{N}_1^- &= -\Gamma' N_1^- - \Gamma N_1^- + W_v(N_1^+ - N_1^-).\end{aligned}$$

The number densities of the states have been denoted by N_0^+ , N_0^- , N_1^+ , and N_1^- , respectively. In addition, the assumption that the velocity equilibration rate W_v is independent of vibrational state has been made.

The rate equations describing the effect of rotational equilibration and vibrational bottlenecking are given as follows:

$$\begin{aligned}\dot{N}_0 &= W_p(N_1 - N_0) + \Gamma(N_1 - fN_0) + \Gamma(N_2 - gN_0), \\ \dot{N}_1 &= W_p(N_0 - N_1) - \Gamma(N_1 - fN_0) - W_r(N_1 - hN_2), \\ \dot{N}_2 &= W_r(N_1 - hN_2) - \Gamma(N_2 - gN_0),\end{aligned}$$

The quantities f , g , and h are relative populations for the levels at equilibrium and are defined by

$$\frac{N_1}{N_0} = f, \quad \frac{N_2}{N_0} = g, \quad \frac{N_1}{N_2} = h.$$

These sets of rate equations must, in addition, satisfy

$$\begin{aligned}\lambda &= [4W_p W_v^2 + (W_p \Gamma' + 4W_v^2 + 3W_p W_v)(\Gamma' + \Gamma)^2 + W_p W_v(\Gamma' - \Gamma)]^{-1}, \\ \xi &= \left[1 - \frac{W_p W_r + W_r W_v(f + g) + W_p W_v g + \Gamma^2 g}{W_p \Gamma(2 + g) + (1 + f + g)\Gamma^2 + W_p W_r(1 + 2h) + W_r \Gamma(1 + h)(1 + f + g)} \right].\end{aligned}$$

The active-gas flux can now be calculated using the steady-state population densities derived. The bimodal expression for J_A is then given by

$$J_A = F \left[\frac{1}{\beta_e} (N_1^+ - N_1^-) + \frac{1}{\beta_g} (N_0^+ - N_0^-) \right].$$

Using the approximation $\beta_e \sim \beta_g = \beta$, we have that

$$J_A = \frac{F}{\beta} [\phi(N_1^+ - N_1^-) + (N_0^+ - N_0^-)].$$

The quantity F/β represents the "terminal" velocity of an active-gas molecule in the medium of the buffer gas. This picture is only approximate and applies only in conditions where many collisions are required to alter the velocity direction of an active molecule. The expression F/β can then be replaced by the velocity of molecules capable of interacting with the radiation field. In terms of the pump detuning $\delta\nu = \nu_0 - \nu_l$; where ν_0 is the molecular line

center and ν_l is the laser frequency, we have that

$$\begin{aligned}N_1^+ + N_1^- + N_0^+ + N_0^- &= N', \\ N' + N_2 &= N, \\ N_1^+ + N_1^- &= N_1, \\ N_0^+ + N_0^- &= N_0.\end{aligned}$$

Since level 2 is assumed to always possess a symmetric velocity distribution, the contribution to the instantaneous local flow will be due to N_0^+ , N_0^- , N_1^+ , and N_1^- . The steady-state expressions for each level population as calculated using both sets of rate equations are given as follows:

$$\begin{aligned}N_0^+ &= N\xi\lambda W_v [W_p W_v \\ &\quad + (\Gamma + \Gamma')(W_p + \Gamma + \Gamma' + 2W_v)], \\ N_0^- &= N\xi\lambda [W_p W_v^2 + (W_p W_v + W_p \Gamma' + 2W_v^2)(\Gamma' + \Gamma) \\ &\quad + W_v(\Gamma + \Gamma')^2 + W_p W_v(\Gamma - \Gamma')], \\ N_1^+ &= N\xi\lambda W_v W_p (\Gamma + \Gamma' + W_v), \\ N_1^- &= N\xi\lambda W_v^2 W_p.\end{aligned}$$

In the steady-state solutions above, the quantities ξ and λ are rate dependent and given by

center and ν_l is the laser frequency, we have that

$$F/\beta = \frac{c \delta\nu}{\nu_0}.$$

The direction of F/β is specified by the sign of $\delta\nu$. The direction of J_A is, however, specified by the sign of both $\delta\nu$ and $(\phi - 1)$.

Using the expressions derived we can now give an analytic result for the macroscopic active-gas flux:

$$\begin{aligned}J_A &= \frac{c \delta\nu W_p}{\nu_0} \\ &\quad \times [W_v(\Gamma' + \Gamma)(\phi - 1) - \Gamma'(\Gamma' - \Gamma)] N\xi\lambda.\end{aligned}$$

Pumping-induced flow vanishes if $\phi = 1$ and $\Gamma' = \Gamma$. However, it should be pointed out that if $\Gamma \neq \Gamma'$ then we predict a flow even when the transport properties in the ground state and excited vibrational state are identical.

ACTIVE-GAS DENSITY PROFILE

The steady-state distribution of active gas in a closed tube in the absence of convection will be derived with some simplifying assumptions. The assumptions involved are listed below.

- (1) The rates involved in the model are constant throughout the length of the cell.
- (2) The only spatial variation present is due to the active gas.
- (3) There is no coupling between the pump intensity profile and the LID steady-state profile of the active gas.

Assumptions (1) and (2) are justifiable under conditions where $\rho_B \gg \rho_A$. Assumption (3) is justifiable when the cell length is shorter than the reciprocal of the absorption coefficient or when the pump intensity is always maintained above the saturation intensity.

The steady-state distribution occurs when the diffusive flow balances the LID flow. This equality was given in Eq. (1). It is important to note that the value of the total density N , given in the expression for J_A , is not the total active-gas density but only the fraction participating in the optical-pumping cycle. We must therefore relate this quantity to the total active-gas density ρ_A . Calling the fractional occupation of the $|0\rangle$ state f_{JK} and the fraction of molecules having the correct velocity to interact with the pump laser $\epsilon(\nu_0, \nu_l, I, I_s)$, we have that

$$N = \rho_A f_{JK} \epsilon(\nu_0, \nu_l, I, I_s),$$

where I is the average laser intensity, ν_0 is the molecular line-center frequency, and ν_l is the laser frequency. Calling $G(\nu)$ the normalized Gaussian line-shape function for Doppler broadening, we have that

$$\epsilon \sim \int_{\nu_l - \Delta\nu_0/2}^{\nu_0 + \Delta\nu_0/2} G(\nu) d\nu - \left| \int_{\nu_0}^{\nu_l + \Delta\nu_0/2} G(\nu) d\nu \right|.$$

The hole burnt is approximated as rectangular and given by

$$\Delta\nu_0 = \Delta\nu_h (1 + I/I_s)^{1/2}.$$

The balance equation can be simplified in one dimension and gives a simple differential equation for $\rho_A(x)$:

$$\begin{aligned} \frac{d\rho_A(x)}{dx} &= \frac{-1}{D_{AB}} \frac{c \delta\nu}{\nu_0 f_{JK} \epsilon} \xi \lambda \\ &\times [W_v(\Gamma' + \Gamma)(\phi - 1) - \Gamma'(\Gamma' - \Gamma)] \\ &\times W_p \rho_A(x). \end{aligned}$$

Letting

$$\Pi = \frac{c \delta\nu \xi \lambda W_p}{D_{AB} \nu_0 f_{JK} \epsilon} [W_v(\Gamma' + \Gamma)(\phi - 1) - \Gamma'(\Gamma' - \Gamma)]$$

we can immediately write down the solution

$$\rho_A(x) = \rho_A^0 \exp(-\Pi x).$$

ρ_A^0 is a normalization constant which can be related to the initial uniform density ρ_0 by

$$\rho_A^0 = \frac{\Pi L \rho_0}{1 - \exp(-\Pi L)},$$

where L is the length of the cell.

CALCULATIONS FOR CH₃F:He MIXTURES

In this section we give results for the LID effect in CH₃F:He mixtures. The CH₃F:He system was chosen for several reasons. CH₃F is a well studied molecule from both the kinetics point of view as well as spectroscopically. The main reason for this is its exceptional characteristics as an optically pumped submillimeter laser gas.¹⁰ In addition, the CH₃F:He system is the only system for which an excited-state elastic collision cross section has been determined.

The CH₃F molecule is a symmetric top with a strong coincidence with the 9P(20) line of the ¹²CO₂ laser. We present in Table I the relevant data we utilized to investigate our analytical LID results.

The rate-equation solutions were computed for CH₃F:He mixtures containing 0.1 torr of CH₃F. Curves are shown in Fig. 2 for two pressures of He. The calculations are performed for a CO₂ laser pump locked at the 9P(20) line center and producing a uniform intensity of 50 W/cm². The results are plotted relative to the initial uniform density of CH₃F. The results indicate a confinement region on the order of 20 cm for the 5-torr He mixture. These

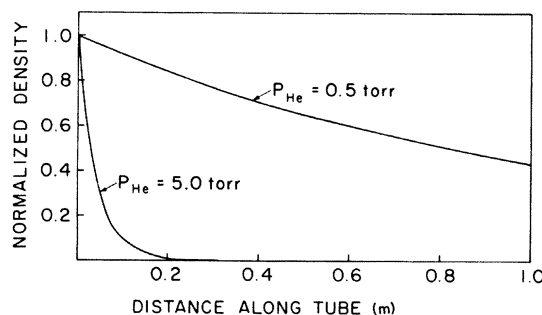


FIG. 2. Spatial variation of CH₃F density for laser-induced CO₂ diffusion in He. CH₃F pressure is 0.10 torr and CO₂-laser intensity is 50 W/cm².

TABLE I. Relevant data used in investigation of our analytical results.

		Reference
$\nu_0 - \nu_1$	44 MHz	11
$\sigma_e(\text{CH}_3\text{F}:\text{He})$	40 Å ²	12
ϕ	0.9963	calculated with 12
I/I_S	5	calculated
D_{AB}	387 cm ² /sec	calculated from 12
W_r	2×10^5 cm ⁻¹ torr ⁻¹ (He)	13
W_v	1.3×10^5 sec ⁻¹ torr ⁻¹ (He)	9
Γ	$600P_{\text{CH}_3\text{F}} + 1.6 \times 10^3$ sec ⁻¹ torr ⁻¹ (He)	14

results cannot be compared to the experiments mentioned in the introduction as a profile has never been measured.

APPENDIX

We give a simple method for determining the ratio of ground-state to excited-vibrational-state elastic collision cross sections using spectroscopic data and virial coefficients. Since we have an experimentally determined value for $\sigma_e(\text{CH}_3\text{F}:\text{He})$, we can use this as a given constant. We are interested in determining $\phi = \sigma_g / \sigma_e$.

We may define a collision cross section in terms of collision radii for the active gas R_A and the buffer gas R_B by

$$\sigma_{g,e} = \pi(R_A^{g,e} + R_B)^2.$$

The ratio of cross sections ϕ may be written as

$$\phi = \frac{\pi(R_A^e - \delta R + R_B)^2}{\sigma_e},$$

where $R_A^e = R_A^g + \delta R$.

The ratio of the average molecular radius of the active molecule in the ground and electronic state may be estimated spectroscopically. Recalling that the rotational constants of polyatomics are functions of vibrational quantum number and the type of mode excited, we may use this to estimate δR .

The rotational constant B for a given mode is inversely proportional to the molecular moment of inertia. The moment of inertia is then related by the square of the average point-mass distribution. Thus the inverse ratio of rotational constants can serve to estimate R_A^e / R_A^g .

The result can be used to determine δR :

$$\delta R \approx -R_A^e \left[\left(\frac{B^e}{B^g} \right)^{1/2} - 1 \right].$$

Using this expression, we may express ϕ in terms of R_B and σ_e as follows:

$$\phi = \left\{ \left(\frac{B^e}{B^g} \right)^{1/2} + \left(\frac{\pi}{\sigma_e} \right)^{1/2} \left[1 - \left(\frac{B^e}{B^g} \right)^{1/2} R_B \right] \right\}^2.$$

The remaining parameter is the value of R_B . This can be determined for simple systems such as He from the van der Waals constant b for pure buffer-gas mixtures. The expression relating these two quantities is¹⁵

$$b = \frac{4\pi}{3} (R_B)^3.$$

The values for B^g and B^e for CH_3F $v=1$, ν_3 mode excitation were extracted from high-resolution spectroscopy studies.¹⁶ The value of $R_B = 2.5$ Å for He was deduced from pressure-volume data for pure He mixtures.¹⁷ The excited ν_3 mode elastic collision cross section σ_e was taken from the ultrahigh-resolution pure rotational collisional narrowing experiments in Ref. 12. These values result in $\phi = 0.9963$.

CONCLUSIONS

We have developed an analytically solvable, rate-equation model for the process of laser-induced diffusion in polyatomic gases. The model utilizes an effective bidirectional velocity distribution with a kinetic model. The profile of active gas is derived for closed tubes. The model also predicts an interesting new effect in LID. The results indicate that LID may occur even when the collision cross sections in the two states connected by optical pumping are the same if collisions allowing simultaneous velocity direction change and vibrational relaxation are present.

Calculations have been performed for uniform intensity pumping of $\text{CH}_3\text{F}:\text{He}$ mixtures by a line-

center 9P(20) CO₂ laser. These calculations are performed using a novel spectroscopic method for determining the excited- and ground-state collision cross sections. The results indicate that confinements on the order of centimeters can take place under strong excitation. These values are reasonable as compared to calculations performed on idealized two-level systems such as sodium, considering the

level dilution factor present when a polyatomic molecule possessing a rotation-vibration structure is utilized.

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