

Laser-induced translational absorption in noble gases

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A theory of the absorption coefficient for inverse Raman scattering (IRS) on translation degrees of freedom in compressed monatomic gases is developed. The advantages ensuing from laser induction of the translational absorption spectrum compared with induction by an external electrostatic field are demonstrated.

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We propose in the present paper a new procedure for observing translational absorption spectra induced by binary collisions in pure noble gases. The difficulty of investigating such system by spectroscopy is due to the fact that in the absence of external fields the collisions of identical atoms (in contrast to collisions of unlike atoms) are not accompanied by the appearance of an induced dipole moment, i.e., they are not active in absorption. In principle, the activity of pair collisions can be induced^[1] by turning on an external electrostatic field \mathbf{E} . In this case the dipole $\vec{\mu}$ localized on a given pair

$$\vec{\mu} = \hat{\alpha} \mathbf{E} = 2\alpha_0 \mathbf{E} + \hat{\Delta}(\mathbf{R}) \mathbf{E} \quad (1)$$

($\hat{\alpha}$ is the polarizability tensor of the pair of interacting atoms, and α_0 is the polarizability of the free atom), contains a term $\hat{\Delta} \mathbf{E}$ that depends on the interatomic distance R . The depth of modulation of the dipole $\vec{\mu}$ by the translational motion, however, is quite small, so that an attempt^[2] to observe this effect ended in failure.

There are grounds for assuming that a transition from a static to a rapidly alternating (“laser”) field will increase the cross section of absorption in the anti-Stokes region relative to the laser frequency ω_1 . In other words, it is proposed to use the inverse Raman scattering (IRS) phenomenon^[3] to observe translational absorption in monatomic gases. The onset of IRS is connected with interaction of a weak external field of frequency ω_2 ($\omega_2 > \omega_1$) with the electric dipole moment induced the atoms or molecules by the laser field.

We assume that the molecules remain in the ground electronic state ϵ_0 after the transition. The wave functions of the initial and final states of the “molecules + field” system can then be written in the form

$$|I\rangle = |\epsilon_0\rangle |i\rangle |n_1\rangle |n_2\rangle; \quad |F\rangle = |\epsilon_0\rangle |f\rangle |n_1 + 1\rangle |n_2 - 1\rangle,$$

where $n_{1,2}$ are the numbers of the corresponding photons, while i and f are the quan-

tum numbers describing the motion of the nuclei in the electronic ground state. The electric-dipole approximation in second-order perturbation theory leads to the following expression for the transition rate

$$P_{IF} = \frac{8\pi^3}{V^2} n_2 (n_1 + 1) \omega_2 \omega_1 |\langle i | (\mathbf{e}_1 \cdot \hat{\mathbf{a}} \mathbf{e}_2) | f \rangle|^2 \delta(\omega_{fi} - \omega)$$

($\mathbf{e}_{1,2}$ are the polarization unit vectors, V is the volume, and $\omega = \omega_2 - \omega_1$), from which we have left out factors that are of no importance for the subsequent analysis and are connected with the possible degeneracy of the state I and F . Assuming that the intensities of the two light fluxes satisfy the inequalities $I_1 \gg I_2$ and $I_1 \gg \hbar\omega_1 c/V$, and there are no other channels for a change in the photon numbers we obtain the absorption coefficient in the Lambert-Beer law for the flux I_2 :

$$K_2(\omega) = \frac{8\pi^3}{\hbar c^2} I_1 \omega_2 2\text{th}(\hbar\omega/2kT) W(\omega), \quad (2)$$

which contains an even spectral function $W(\omega)$ given by

$$W(\omega) = \frac{1}{V} \sum_{if} \frac{\rho_{ii} + \rho_{ff}}{2} |\langle i | (\mathbf{e}_1 \cdot \hat{\mathbf{a}} \mathbf{e}_2) | f \rangle|^2 \delta(\omega_{fi} - \omega), \quad (3)$$

where ρ is the molecular density matrix. If we identify the absorption frequency for the static regime ($\omega_1 = 0$) with the detuning ω in the dynamic regime and assume that the mean squares of the intensities of the fields in both cases are equal, then (2) leads to an appreciable increase of the absorption coefficient on going to the dynamic regime. The frequency factor ω_2/ω increases the absorption cross section on account of the frequency of the oscillations of the induced dipole moment with which the light wave ω_2 interacts. Under ordinary conditions, the frequencies of the translational collisions in compressed inert gases are of the order of several dozen cm^{-1} .^[4] Thus, the use of a strong visible-band laser field as a source provides a gain up to ~ 400 times.

To estimate the condition for the observability of the translational IRS in pure monatomic gases, let us obtain, in the binary-collision approximation, the zeroth moment of the spectral function (3):

$$M_0 = \int_{-\infty}^{\infty} W(\omega) d\omega = n^2 \text{Sp} \{ \rho (\mathbf{e}_1 \cdot \hat{\mathbf{A}} \mathbf{e}_2)^2 \},$$

where n is the number of atoms per unit volume. Introducing the isotropic $\bar{\Delta} = 1/3 \text{Sp} \hat{\Delta}$ and anisotropic $\gamma = \Delta_{\parallel} - \Delta_{\perp}$ components of the induced-polarizability tensor $\hat{\Delta}$, we can calculate M_0 by using the \mathbf{R} coordinate representation. After averaging over the orientations of the vector \mathbf{R} we obtain

$$M_0 = 4\pi n^2 \int_0^{\infty} dR R^2 g(R) \left\{ \bar{\Delta}^2(R) (\mathbf{e}_1 \cdot \mathbf{e}_2)^2 + \frac{\gamma^2(R)}{45} [3 + (\mathbf{e}_1 \cdot \mathbf{e}_2)^2] \right\},$$

where $g(R)$ is the radial-distribution function. If we neglect the electron exchange, then $\hat{\Delta} \ll \gamma = 6\alpha_0^2 R^{-3}$.^[5] For the model of hard-spheres of diameter σ we have at $\mathbf{e}_1 \parallel \mathbf{e}_2$:

$$M_o = \frac{64\pi n^2 \alpha_c^4}{15\sigma^3}$$

The function (3) can be approximated by a dispersion curve. It is then easy to derive the formula for the maximum value of the absorption coefficient (2):

$$K_2^{max} \cong \frac{256\pi^3 \alpha_o^4 n^2}{15c^2 \sigma^3 kT} \omega_2 I_1. \quad (4)$$

From (4) we can now determine the experimental conditions under which the optical density can have an arbitrary specified value, say 0.1, which is larger by three orders of magnitude than that realized in^[2]. For example, for a flux $I_1 = 30 \text{ MW/cm}^2$ at $\gamma = 0.69 \mu\text{m}$ (ruby laser) we find that in Xe we have $\alpha_o = 4.05 \text{ \AA}^3$ and $\sigma = 3.85 \text{ \AA}$, at room temperature and at a density 200 amagat, the indicated optical density can be obtained over a path of 60 cm.

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