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MEASUREMENT OF ELASTIC SCATTERING CROSS SECTIONS IN A GAS BY LASER SPECTROSCOPY METHODS

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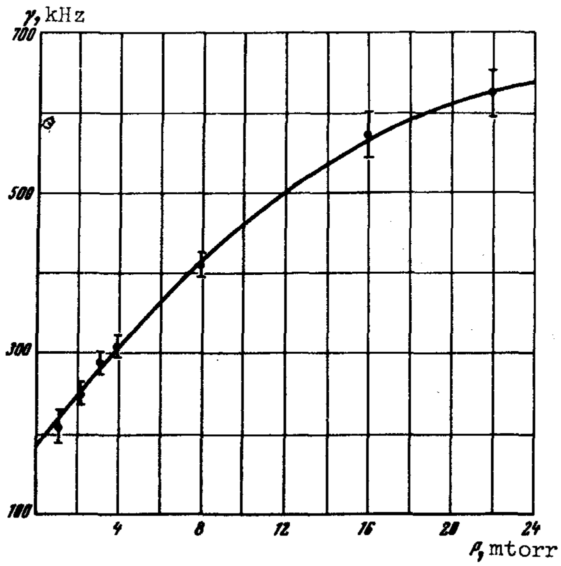
1. The main information concerning the interaction potential of particles colliding in a gas has been obtained from studies of the elastic scattering cross sections of atomic and molecular beams. We report here for the first time the measurement of the scattering cross sections by methods of high-resolution laser spectroscopy. We call attention to new features of the behavior of the Lamb dip in molecular gases of low pressure, the nonlinear dependence of its width and shift on the pressure, and the difference between its impact broadening and the Doppler-contour broadening. These features, which are connected with the nature of the scattering of two-level systems, make it possible to separate the contribution of the angular scattering of atoms, and the role of phase-shifting and quenching collisions in the line broadening.

2. In the case of low-pressure gases it is not always possible to identify the impact line width of the spontaneous emission with the Lamb-dip width. The emission line width for collisions is determined by the scattering amplitudes at both levels and was obtained in [1, 2]. To find the width of the Lamb dip it is necessary to solve the kinetic equation for the density matrix in the standing-wave field. The gas-kinetic approach developed in [3] makes it possible to express the departure and arrival terms in this equation through the exact amplitudes for scattering by the levels m and n. In a qualitative study, however, we can use the fact that the width of the Lamb dip in a gas is essentially equal to the time of coherent interaction between the atom and the field. The influence of the collisions on the shape of the Lamb dip will depend essentially on the specific nature of the scattering of the two-level system. Two qualitatively different cases are then encountered.

1) The collisions are accompanied by total loss of the coherence between the levels m and n. In this case the scattering amplitudes for the two levels differ appreciably. The line width of the radiating atom is [1, 2]

$$\gamma = Nv(\sigma_m + \sigma_n) + \gamma_0 , \qquad (1)$$

where σ_m and σ_n are the total elastic cross sections for levels m and n, v is the average velocity of the atoms, N is the density of the scattering centers, and γ_0 is determined by the inelastic scattering at the levels m and n, and also by the line width in the absence of collisions. The width of the Lamb dip is given in this case by (1).



Pressure dependence of the width of the Lamb dip in methane at $\lambda = 3.39\ \mu$.

2) The coherence between levels is retained in the scattering (there is no randomization of the oscillator phase). The amplitude of the elastic scattering at levels m and n are equal in this case. Case (2) differs in principle from (1), since the time of coherent interaction of the atom with the field depends significantly on the ratio of the characteristic angle $\bar{\theta}$, at which the scattering takes place, to the quantity γ/kv (kv is the Doppler width). If $\bar{\theta} \gg \gamma/kv$, then the atom leaves the interaction after the collision, and the width of the Lamb dip is

$$\gamma = 2Nv\sigma + \gamma_0 \ . \qquad (2)$$

For $\tilde{\theta} \sim \gamma/kv$, the atoms scattered through angles $\theta < \gamma/kv$ do not leave the region of coherent interaction with the field, i.e., they cannot be distinguished from the unscattered ones (there is no phase randomization!). Excluding these atoms, we obtain

$$\gamma = 2Nv \int_{\gamma/kv}^{\pi} d\theta \sin\theta \, 2\pi\sigma(\theta) + \gamma_0 \ , \qquad (3)$$

where $\sigma(\theta)$ is the differential elastic-scattering cross section. If we use (3) to measure σ, then we have a situation analogous to that in experiments on scattering of atomic beams. The role of the resolving power of the instrument is played in our case by the quantity γ/kv.

3. We investigated the impact broadening of the Lamb dip in methane at $\lambda = 3.39\ \mu$ and pressures $2 \times 10^{-2} - 2 \times 10^{-4}$ Torr (He-Ne laser with internal CH_4 absorbing cell). The figure shows the dependence of the width of the Lamb dip $\gamma(P)$ on the pressure P, obtained from the generation-power peak with allowance for its broadening by the field and for the nonlinear pulling of the generation frequency. At low pressures, the $\gamma(P)$ plot is linear with a slope 30 ± 2 MHz/Torr. This value agrees with the results of [4, 5]. At higher pressures the slope of the curve decreases and tends to 10 ± 5 MHz/Torr.

4. The observed $\gamma(P)$ dependence corresponds to the analysis present in Sec. 2. According to [6], 90% of the CH_4 molecules are scattered through angles $\theta \gtrsim 10^{-2}$, so that γ depends linearly on the pressure in the region $\gamma \sim 10^4 - 10^5$ Hz. The decrease of the slope of the curve at large P indicates that some of the atoms begin to be scattered through angles $\theta \sim \gamma/kv$ without loss of coherence. The difference between the slopes of the curve at small and large pressures indicates that elastic scattering without phase shift plays an important role. This indicates that the scattering amplitudes are approximately equal for both levels. Additional proof of the small role of the phase-shifting collisions is the difference in the broadening of the Lamb shift and of the Doppler contour. According to our measurements and the data of [7], the broadening of the Doppler contour is 7.8 ± 0.8 MHz/Torr and 7.4 ± 0.4 MHz/Torr, respectively. This broadening may be due to inelastic scattering, and also to a

certain difference between the scattering amplitudes[1]. The small line shift Δ [4] confirms to some extent the conclusion that the phase randomization plays a minor role. The sum of the total scattering cross section for both levels is $(10 \pm 0.3) \times 10^{-14}$ cm^2. The determination of the cross section for scattering at each level with such an accuracy calls for an explanation of the mechanism responsible for the broadening of the dispersion part of the Doppler contour. Since this mechanism cannot be regarded as exactly established, the measurement accuracy of the scattering cross section at each level, in accordance with the considerations advanced above, will be determined by the ratio of the impact broadening of the Lamb dip and of the Doppler contour. The total scattering cross section and the cross section for elastic scattering at each level are equal to $\sigma = (5 \pm 1.2) \times 10^{-14}$ cm^2. The inflection in the $\gamma(P)$ curve occurs at $\gamma \simeq 500$ kHz, corresponding to a characteristic scattering angle $\overline{\theta} \simeq 0.5 \times 10^{-2}$ rad. If it is assumed that the interaction is determined by the c/r^6 law, then it can be shown that the total cross section is connected by the transport ratio $\sigma_{tr} \simeq \sigma(\overline{\theta})^{1/3}$. The calculation yields $\sigma_{tr} \simeq 8 \times 10^{-15}$ cm^2. This estimate agrees with the assumed value of the gas-kinetic cross section of methane [8].

5. In accord with the foregoing analysis, we should expect singularities in the behavior of the shift of the Lamb dip at low pressures; these singularities are very important for the development of optical standards with high reproducibility of the frequency. At low pressures, when $kv\overline{\theta} \gg \gamma$, the atom goes out of the interaction region after the collision, regardless of whether phase randomization does or does not take place, and consequently makes no contribution to the shift Δ_L of the Lamb dip. Only the small number of atoms scattered in the angle region γ/kv produce a shift Δ_L. Recognizing that $\gamma \sim Nv\sigma$, we can expect $\Delta_L \sim (Nv\sigma/kv\overline{\theta})\Delta$. Since Δ depends linearly on the pressure, Δ_L depends quadratically on the density. For methane we have $d\Delta/dP \sim 100$ kHz/Torr at $\gamma \sim 10^{-4}$ Hz ($kv\overline{\theta} \sim 10^6$ Hz) and $\Delta_L \simeq 1$ Hz, ensuring a reproducibility of the frequency as a function of the pressure better than 10^{-13}. With increasing pressure, the $\Delta_L(P)$ dependence tends to become linear, and $\Delta_L \lesssim \Delta$ when $\gamma \gg kv\overline{\theta}$. Thus, the characteristics of the differential cross section of elastic scattering, and particularly $\overline{\theta}$, can be measured by using the difference between the shifts of the Lamb dip and of the line center.

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[1]It follows from a comparison of the results of [3] and [9] that collisions with phase randomization lead to equal broadening of the Doppler contour and the Lamb dip. Elastic scattering without phase randomization leads to a narrowing of the Doppler contour, which is determined by the transport cross section σ_{tr}.

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MULTIPHOTON DISSOCIATION, PREDISSOCIATION, AND AUTOIONIZATION OF THE HYDROGEN MOLECULE

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We report here observations of dissociation and ionization of the hydrogen molecule by the second harmonic of a neodynium laser. We show that an important role can be played in these processes by multiphoton transitions into highly-excited H_2^* Rydberg states. Owing to the strong interaction of the nuclear motion with the electron motion, the excitation of these states is accompanied by a fast nonradiative transition into the continuous spectrum of the electronic wave function (auto-ionization) or the nuclear wave function (predissociation) of the molecule.

Our experiment enables us to compare the probabilities of direct multiphoton ionization of H_2 and auto-ionization of optically excited Rydberg states. The latter channel is more probable. Multiphoton ionization of H_2 can proceed via predissociation in oscillations during the oscillations, as well as by direct transition of the excited terms into the continuous spectrum of the nuclear wave function, with formation of hydrogen atoms in the ground state H (n = 1) and the excited state H* (n = 2).

Multiphoton dissociation of molecules in a field of optical frequency is usually associated, in the main, not with electronic transitions but with excitation of nuclear oscillations. The vibrational transitions can be noticeable

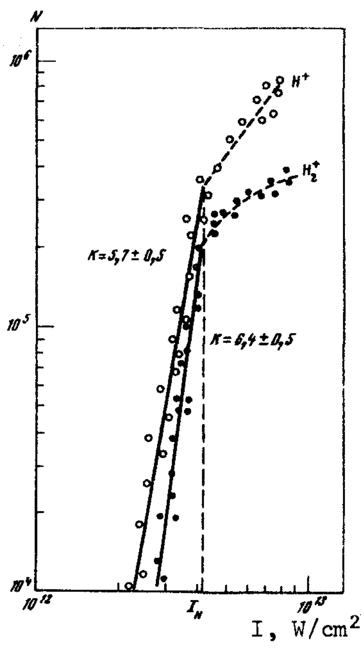

Fig. 1. Number of ions produced as a result of dissociation (N_{H^+}) and ionization ($N_{H_2^+}$) of H_2 molecules vs. the intensity I. The breaks on the curves at $I_H = 4 \times 10^{12}$ W/cm² corresponds to saturation of the ion signals, i.e., to dissociation (with subsequent ionization of the atoms) or ionization of all $n_0V = 4 \times 10^5$ molecules in the focal region ($n_0 = 4 \times 10^{12}$ is the density of H_2 in the focal region, $V = (1 \pm 0.4) \times 10^{-7}$ cm³ is the effective volume of interaction [5] for six-photon processes.

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