Investigation of the shape of nonlinear resonances at low pressures

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Direct spectroscopic observation of elastic scattering of colliding excited particles into small angles $\sim 10^{-2}$ rad is first reported. The effect of these collisions on the shape of narrow resonances in methane is investigated.

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1. Qualitative characteristics of shock broadening and displacement of narrow resonances in a low-pressure gas have been identified.[1,2] Collisional broadening and displacement due to nonlinear effects depend on a gas density for which a binary collision model is known to be correct (working gas pressure $\sim 10^{-3}$ tor). The identified phenomena were attributed[1,2] *to the effect of small-angle elastic scattering of* particles without a phase reduction. If the Doppler frequency shift $\Delta\omega_D = ku\theta$ in the course of scattering through a characteristic angle θ ($\theta \sim 10^{-2}$ rad) is greater than homogeneous line width 2Γ, particles fail to interact with a field after colliding (k is

the wave number, u is the mean thermal velocity). Line broadening in this pressure region is determined by the total scattering cross section. The homogeneous linewidth increases with increasing gas pressure and at a certain time 2Γ may be greater than $\Delta\omega_D$. Thus, upon colliding a particle continues to interact with the field and the collisions without phase reduction fail to broaden the resonance. In this case broadening is determined by inelastic processes and phase-reducing collisions. Therefore, in the presence of collisions that preserve that phase of the dipole moment broadening at low ($ku\theta \gg \Gamma$) and high ($ku\theta \ll \Gamma$) pressures may vary considerably. Pre-1972 theoretical investigation of the collisional effects on the shape of resonances—based on the use of phenomenological constants that characterize the collisional frequency—have not even qualitatively explained the results of Refs. 1 and 2. The first exact theoretical investigations[3,4] based on the use of kinetic equations for the density matrix—in which the incoming and outgoing terms are expressed in terms of scattering amplitudes—agree qualitatively with the results of Refs. 1 and 2. More recently, nonlinear dependence of resonance shock broadening in a low-pressure gas was also observed in NH_3,[5] I_2,[6] CO_2,[7,8] and Xe.[9] Calculations of the dependence of resonance linewidth on gas density in CO_2[10] are in a satisfactory quantitative agreement with the experiment.[8] We should note that at $ku\theta \ll \Gamma$ the resonance shape is complex. It consists of a narrow part and a base whose width is directly dependent on the characteristics of differential scattering cross sections.[3,4] In this work we first report on an investigation of resonance shape in the region of low pressures and the direct spectroscopic observation of elastic small-angle scattering of excited particles.1)

2. Figure 1 shows the experimental setup. The frequency of reference laser 1 was shifted from the methane line center by 8 MHz. Test laser 2 with an internal methane cell was "attached" by means of a system of frequency self-tuning (FST) to the frequency of laser 1 with a shift f which was varied in the range 1–15 MHz by coupling a sawtooth voltage to a FST frequency discriminator. This allowed us to scan laser 2 frequency with respect to the center of the methane absorption line. The measuring

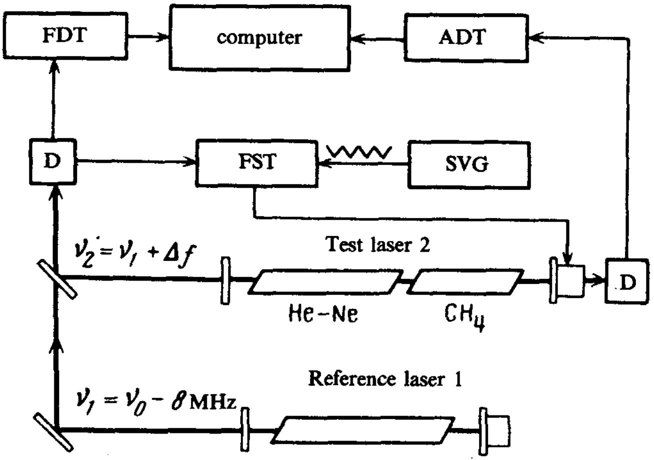

FIG. 1. Experimental setup: D—photodetector; FST—electronic system of frequency self-tuning; SVG—sawtooth voltage generator; FDT—frequency digital transducer; ADT—amplitude frequency transducer.

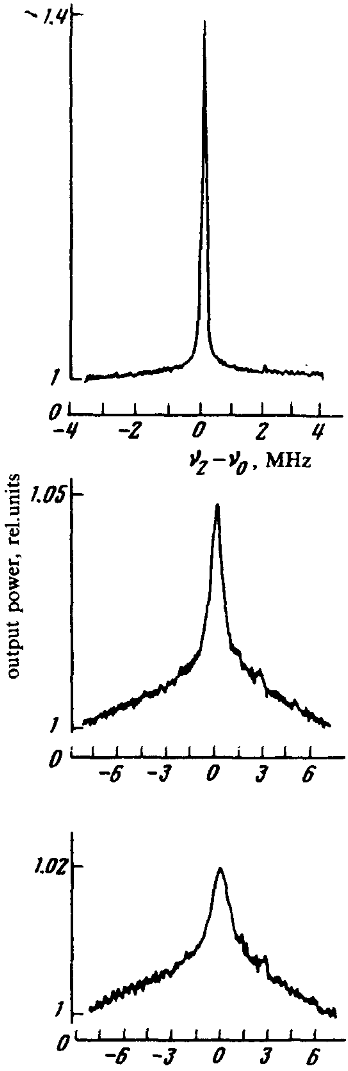

FIG. 2. Shape of resonance in methane at different helium pressures: *a*—$P_{\mathrm{CH4}} = 1$ mtor, $P_{\mathrm{He}} = 0$; *b*—$P_{\mathrm{CH4}} = 1$ mtor, $P_{\mathrm{He}} = 20$ mtor; *c*—$P_{\mathrm{CH4}} = 1$ m tor, $P_{\mathrm{He}} = 43$ mtor.

system consisted of a 512-channel spectral analyzer based on the M-400 computer. A laser difference frequency ($\nu_2 - \nu_1$) signal was coupled to one channel and signal proportional to the output power of laser 2 to the other. In contast to traditional methods of investigating the resonance shapes—which are based on the recording of the variable component of output power as the laser frequency is modulated—we recorded the power resonance directly. This permitted us to increase the sensitivity of recording resonance tails. However, effects of drift and power fluctuations increased sharply. The latter were eliminated in the course of signal processing by a computer. The laser power level was corrected after each passage of frequency through the resonance. The experimental setup was designed to investigate the resonance shape for a contrast ratio of ~0.001.[2)] Increased sensitivity is associated with a reduction in the power output drift.

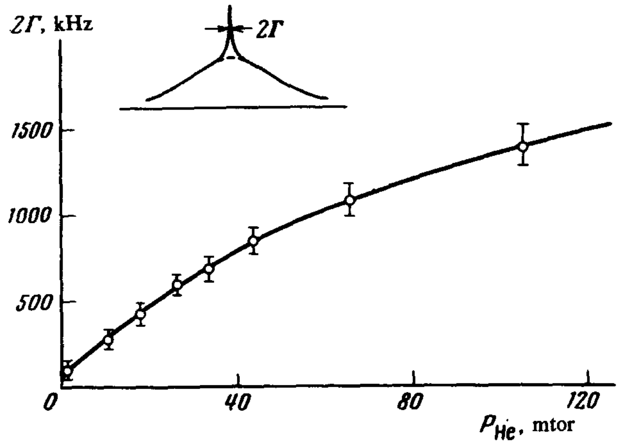

FIG. 3. Dependence of resonance width 2Γ on helium pressure. Methane pressure ~10^{-3} tor.

3. Figure 2 shows the resonance shape at different helium pressures in the methane cell. At ~10^{-3}-tor methane pressures the resonance shape is Lorentzian with a ~70-kHz width. Addition of helium broadens the resonance line and a ~2–3-MHz base appears. Processing of results showed that the width of the narrow part of resonance nonlinearly depends on the pressure (Fig. 3). This is highly significant for theory confirmation.[3,4] Resonance broadening in the 1–10-mtor pressure range is ~20 MHz/tor and is determined by the total elastic scattering cross section.

We observed a small displacement of the base with respect to resonance center which may be due to two different shock displacements. The base shape at low pressures (~10 mtor), when the collisional frequency is low, is directly related to characteristics of the differential scattering cross section. Data in Fig. 2b yield a value of the characteristic angle θ that may be ~1°. The relative amplitude of the base increases with increasing pressure. At $P_{He} = 0.1$ tor, base amplitude is comparable to the amplitude of the sharp portion of the resonance.

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[1)] Results of this work were reported by the authors at the VI International Conference on Atomic Physics (Riga, August 1978).

[2)] The computer-based measuring system was designed by Borisov and Gusev[11] to whom the authors express deep gratitude.

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