Selection of slow molecules by the methods of coherent transient processes

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The effect of slow particles was recorded for the first time in the kinetics of the coherent response of a molecular gas under transit conditions. The decrease in the decay rate with long time delays demonstrates that slow particles can be used for high-resolution, coherent, time-resolved spectroscopy. © *1995 American Institute of Physics.*

Slow particles are of great interest for the spectroscopy of gases. This is explained, on the one hand, by the fact that the translational motion of the particles determines the width and the shape of the spectral lines of rarefied gases. On the other hand, because their de Broglie wavelengths are long, slow particles should manifest purely quantum properties and the study of such objects could yield new information about the nature of particle interactions[1] and be useful for the development of a new field—neutral-particle interferometry.[2–4]

To decrease the intensity of the thermal motion, it is generally necessary to use a complicated technique of monokinetic beams of atoms or molecules or to confine cooled particles in traps. The particle-beam technique is extensively used in the high-resolution spectroscopy of atoms and molecules. In the case of cold-particle traps, important results have been obtained only for atoms.[5,6] Efficient methods for cooling molecules are not yet available. At the same time, molecular transitions have the small radiative linewidths which are of interest for high-resolution spectroscopy.

In the present work we have realized experimentally the selection of slow particles according to the transverse (relative to the direction of propagation of the exciting radiation) velocities of the translational motion by means of coherent transient processes — the photon echo and its modifications. The idea of particle selection according to radial velocities[7] consists of the following. The first pulse of the exciting radiation with a nearly Gaussian transverse intensity profile excites in the medium a nearly Gaussian transverse polarization distribution of the coherent emitters. During the time delay T before the next exciting pulse appears, this distribution "spreads out" as a result of the thermal motion of the particles.[8] The next exciting pulse with a transverse intensity profile similar to that of the first pulse excites only the particles which have remained in the region of the laser beam. As the delay time between the excitation pulses increases, particles with increasingly lower radial velocities interact with the next pulse and, correspondingly, participate in the generation of a coherent response. As a result, the coherent response generated after a long delay time $2T$ is produced by an ensemble of particles whose radial velocities are $v_r < a/T$, where a is the transverse size of the excitation-radiation field. The depletion of "fast" particles from the ensemble of the transverse distribution can be estimated

by calculating the average radial velocity u_r of the particles from the remaining "truncated" distribution of the translational radial velocities $f(v_r) = 2v_r/u^2 \exp(-v_r^2/u^2)$:

$$u_r = \int_0^{a/T} dv_r v_r f(v_r), \tag{1}$$

which leads to the expression

$$u_r = u\left[\frac{\sqrt{\pi}}{2}\Phi\left(\frac{a}{Tu}\right) - \frac{a}{Tu}\exp\left(-\frac{a^2}{T^2u^2}\right)\right], \tag{2}$$

where $\Phi(x)$ is the error function. This dependence gives a very sharp decrease of the average radial velocity and, correspondingly, the average kinetic energy of the selected particle ensemble as the time delay between the excited pulses increases. Under the conditions of our experiment the effective temperature of the ensemble can reach fractions of a degree.

The experiments were performed in a SF_6 gas on the transition P(33) of the A_2^1 vibrational mode $0 \rightarrow 1$ ν_3 by the stimulated photon echo method. The radiation in the 10P(18) line of a cw CO_2 laser, whose frequency was stabilized at the center of the experimental transition, was used to produce the required train of exciting pulses with the aid of an electro-optic shutter. An additional CO_2 laser with a controllable detuning of the frequency from the center of the transition in SF_6 served for heterodyning of the coherent response. This increased the sensitivity of registration. The geometric configuration of the exciting radiation beam and the low pressures of the experimental gas provided homogeneous transit conditions over the entire length of the experimental cell containing the gas. The transverse dimensions of the laser beam were checked at the entrance and exit of the cell by measuring the radial distribution of the power of the exciting pulses with the aid of a small diaphragm. The pressure of the SF_6 gas was varied from 0.2 to 0.5 mTorr. The collisional relaxation rate Γ_{coll} was equal to $(6-15)\times 10^3$ s^{-1}, which is much shorter than the decay due to the transit effect, which on the initial section of the kinetic curve can be estimated as $\Gamma_{tr} = u/a$, where $u = \sqrt{2k_BT^o/m}$ is the average velocity of the particles at the temperature T^o, k_B is Boltzmann's constant, and m is the mass of the gas particles. Under our experimental conditions with the gas at room temperature $\Gamma_{tr} \approx 0.7\times 10^5$ s^{-1}. Therefore, the condition $\Gamma_{coll} < \Gamma_{tr}$ was satisfied for the initial section of the kinetic curve and the transit effect predominated in the decay of the coherent response.

The stimulated photon echo (SPE) method was chosen because this coherent transient process with short delays T_{12} between the first and second exciting pulses is less sensitive to collisions in which the longitudinal velocity v_z changes.[9] At the same time, such collisions can play a large role in the decay of the photo echo in low-pressure gases with long delays between the exciting pulses.[10,11]

The kinetics of the decay of the coherent responses of a stimulated photon echo was studied for time delays T_{23} longer than the transit time for a room-temperature gas. As follows from Fig. 1, the echo power decays nonexponentially at a rate that decreases for long delays, exceeding the transit time at room temperature. The relaxation rate on the initial section of the curve in the figure is determined by collisions, since for short delays

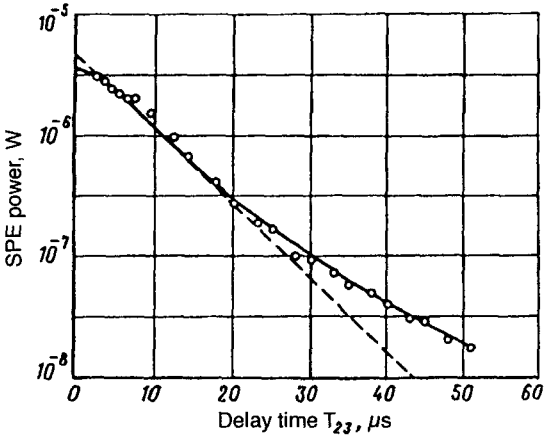

FIG. 1. Kinetics of the decay of a stimulated photon echo as a function of the delay time T_{23} between the second and third exciting pulses. Dots — Experiment; solid curve — estimate obtained from Eqs. (3) and (4); dashed line — tangent at the point $T_{23} = 10$ μs.

T_{23} the transit effects are still not manifested. The dashed line shows the relaxation rate near $T_{23} = 10$ μs. This section of the curve reflects the combined effect of collisional damping and the transit time of particles at room temperature. For long delay times T_{23} the coherent responses are produced mainly by slow particles with radial velocities $v_r \leqslant a/T_{23}$. The solid line is a theoretical estimate which was obtained taking into account the radial translational motion of the particles and also the spatial distribution of the exciting pulses and the heterodyning radiation, by analogy with Ref. 12. For the power recorded by the detector (taking into account the low power P_{SPE} of the coherent response and the constancy of the heterodyne power P_h) we obtain a dependence on the time delay T_{23} of the form $P_{det}(T_{23}) \propto 2\sqrt{P_{SPE}P_h}$, which leads to the formula

$$P_{SPE} \propto \frac{\exp(-2\Gamma_{coll}T_{23})}{[1+(T_{23}/\tau_{tr})^2]^2}, \tag{3}$$

where, as compared to Ref. 9, there is only a small correction to the transit time τ_{tr} in accordance with the equation

$$\tau_{tr} = (a/\sqrt{2}u)\sqrt{\frac{3a^{-2}+a_h^{-2}}{a_2^{-2}+a_h^{-2}}}, \tag{4}$$

where a_h is the width of the transverse distribution of the heterodyning-radiation field. To construct an approximate theoretical curve, we employed the values of a and a_h obtained by measuring the radial distributions of the exciting and heterodyning radiation beams.

The solid curve in Fig. 1 represents an estimate. To compare it with the experimental data, a more accurate account must be taken of the selection of slow particles — this selection is very sensitive to the specific spatial form of the exciting-radiation pulses. The complicated functional dependence of the amplitude of the coherent response on the spatial and temporal forms of the excitation pulses must also be taken into account.

The experimentally recorded drop in the rate of decrease of the number of particles generating a coherent response makes it possible to use slow particles with long delays (greater than 50 μs in our case). In turn, this guarantees improved resolution of time-resolved coherent spectroscopy[13,14] using slow particles; this improvement can reach 1 kHz under our experimental conditions. In addition, there is a real possibility of studying collisions of slow particles in a rarefied gas. Such studies are of interest, since there is the possibility of observing the quantum effects of interactions.

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