Observation of radiation pressure exerted on molecules

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The first observation of pressure exerted by laser light on molecules is reported. A beam of Na_2 molecules interacting with the field of a train of short counterpropagating pulses was deflected through an angle of 7×10^{-4} rad.

Radiation pressure on atoms has attracted research interest for a long time now.[1–9] In contrast, there has been essentially no discussion of a radiation pressure exerted on molecules. A primary difficulty in efforts to observe a radiation pressure exerted on molecules is in arranging a cyclic interaction of the molecules with light, because of the large number of vibrational levels of the ground state, to which transitions can occur during spontaneous emission: The first event of spontaneous photon emission will prevent the molecule from interacting further with the field. Apparently the most realistic possibility is to observe a stimulated radiation pressure based on an alternation of events of absorption and stimulated emission in the field of two counterpropagating waves. Spontaneous emission would not be required for the existence of a radiation pressure in this case. We should point out that one of the simplest ways to arrange a stimulated radiation pressure in the field of a bichromatic standing wave, which was recently proposed in Refs. 10 and 11 and implemented experimentally in Ref. 12 for atoms, is in principle also suitable for molecules. A necessary condition here is that the interaction of the molecule with the laser field be brief in comparison with the spontaneous-emission time $1/\gamma$.

The unwanted spontaneous emission can be reduced by reducing the time the molecule spends in an excited state. As an example, we consider an idealized case: A molecule is interacting with a short π pulse propagating in the positive direction along the Z axis. Immediately thereafter, it interacts with a π pulse propagating in the opposite direction. As a result, after the absorption and stimulated emission of a photon, the momentum of the molecule changes by an amount $2\hbar k$, where $\hbar k$ is the momentum of the photon. The molecule spends essentially all its time in the ground state, and the effect of spontaneous emission is small. A sequential interaction with pairs of such pulses, with a repetition period T, will give rise to an average radiation-pressure force of $2\hbar k/T$ exerted on the molecule. We have previously studied[13] a more realistic model which incorporates the deviation of the pulse area from π, but which ignores relaxation processes during the application of the pulses. It was shown in that earlier paper that even in the case $\gamma T \gg 1$, and with a significant deviation of the pulse area from π, a significant fraction (up to 10%) of the molecules interacting with the light can be given a momentum on the order of $100\hbar k$.

The deflection of a beam of Na_2 molecules by the force of stimulated radiation pressure in the field of two counterpropagating pulse trains was studied on an exper-

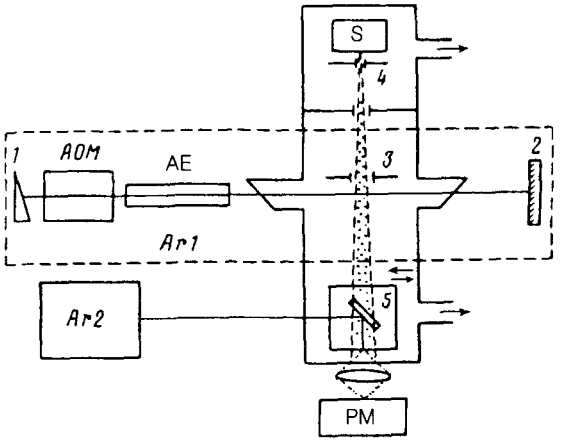

FIG. 1. Block diagram of the experimental apparatus. Ar1—Deflecting laser; Ar2—probe laser; AOM—acoustooptic modulator; S—source of the molecular beam; AE—active element; 1—Littrow prism; 2—output mirror; 3,4—iris diaphrams; 5—system for scanning the laser beam.

imental apparatus with the block diagram in Fig. 1. To achieve a pulsed effect on the molecules, we used active mode locking of an argon-ion laser ($\lambda = 488$ nm) by means of an acoustooptic modulator. The mode-locking regime was monitored with the help of a signal representing intermode beats. At a cavity length of 2 m, the pulse repetition frequency under mode-locking conditions was 75 MHz, with a duration $\leqslant 200$ ps. The distance from the molecular beam to the output mirror was 25 cm. With a time interval ~ 13 ns between successive pulses, the pulses thus acted on the molecule at intervals of 1.7 ns. The light from the laser excited the $X^1\Sigma(v''=3,\ J''=43) \rightarrow B^1\Pi(v'=6,\ J'=43)$ transition of the Na_2 molecule. The maximum average intracavity radiant power under mode-locking conditions was ~ 20 W.

The spatial distribution of the beam molecules was determined from fluorescence excited by a second argon laser, used as a probe. The probe beam was directed onto the beam of molecules, in a direction perpendicular to the latter beam and perpendicular to the axis of the cavity of the deflecting laser, at a distance $L = 35$ cm from the region in which the beam interacted with the deflecting laser. The probe beam was moved parallel to itself in the plane perpendicular to the axis of the molecular beam. The necessary spatial resolution was achieved by focusing the laser beam in such a way that the spot radius did not exceed 80 μm in the region in which the probe beam intersected the molecular beam. The fluorescence signal was detected with a photomultiplier.

The beam of Na_2 molecules was produced in a two-section vacuum chamber (with a residual pressure $< 5 \times 10^{-6}$ torr). The first section of the chamber held the beam source, at a temperature of 670 K. In the second section, the drift section, there were Brewster-angle windows for the beam from the deflecting laser. There were also windows for the entrance of the beam from the probe laser and for detecting the fluorescence of the beam. The divergence of the beam did not exceed 5×10^{-3} rad. The

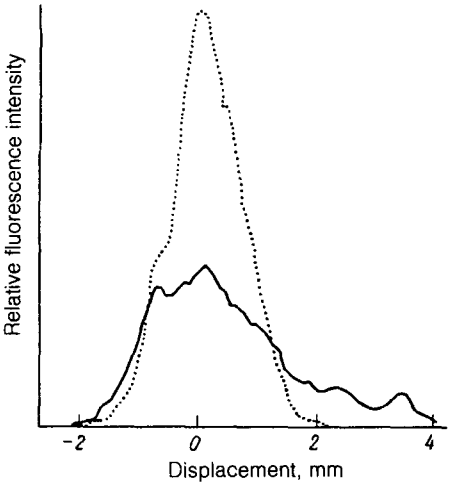

FIG. 2. Distribution function of the molecules in the observation plane. Dotted curve—Without the deflecting field; solid curve—with the deflecting field (the scale for the solid curve is larger by a factor of 25).

diameter of the molecular beam was 0.5 mm in the region in which it intersected the laser beam. During the experiment, the vacuum chamber was inside the cavity of the deflecting laser. The wave vector of the deflecting field was kept perpendicular to the axis of the molecular beam within 2×10^{-3} rad. The density of molecules in the region in which the molecular beam interacted with the field of the deflecting laser was 8×10^{7} cm^{-3} . The density of molecules in the $X^1\Sigma(v''=3,\ J''=43)$ was 1.2×10^{5} cm^{-3}.

The stimulated radiation pressure leads to a change in the distribution function of the molecules in the beam. This change is conveniently described in terms of the displacement of the center of gravity of the beam in the observation plane and in terms of the standard deviation from the center of gravity, which characterizes the spreading of the molecular beam.

Figure 2 shows the initial distribution function of molecules in the observation plane (with the deflecting laser turned off). Also shown here is a distribution function of the molecules after an interaction with the field of the deflecting laser, at an average intercavity power of 17 W. Contributing to the spreading of the beam, along with the nonzero width of the velocity distribution, is a momentum diffusion of the molecules accompanying the stimulated radiation pressure.

Figure 3 shows a plot of the displacement of the center of mass and of the spreading of the beam versus the power of the deflecting laser. As the power is raised, the displacement of the center of mass increases, as does the spreading of the molecular beam. These results agree with the theory for a low laser-beam power.[13] The maximum deflection of the center of mass of the beam was 7×10^{-4} rad. It follows that the average change in the momentum of the molecules which remained in the $X^1\Sigma(v''=3,\ J''=-43)$ state after the interaction with the field of the deflecting laser was $\sim20\hbar k$. Because of the spontaneous relaxation during the pulse, in connection with the time delay between the pulses and the deviation of the shape of the pulses from π, optical pumping significantly reduces the number of molecules at the working

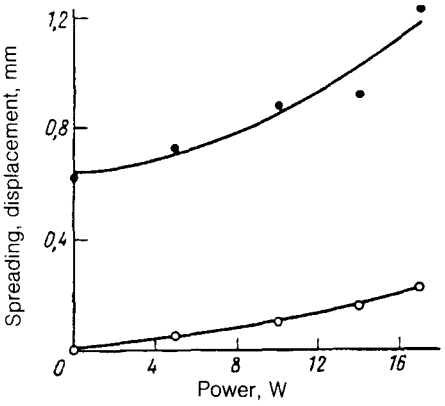

FIG. 3. Displacement of the center of mass (the lower curve) and the spreading of the beam versus the power of the deflecting laser.

level. At the highest power attainable in our experiments, about 3% of the molecules remained in the working level after the interaction with the deflecting light.

When the mode locking of the deflecting laser was turned off, essentially all the molecules deviated from resonance with the laser field because of the optical-pumping effect. As a result, there was absolutely no fluorescence due to the probe laser.

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[1] G. A. Askar'yan, Zh. Eksp. Teor. Fiz. **42**, 1567 (1962) [Sov. Phys. JETP **15**, 1088 (1962)].
[2] A. Ashkin, Phys. Rev. Lett. **25**, 1321 (1970).
[3] A. P. Kazantsev, Zh. Eksp. Teor. Fiz. **66**, 1599 (1974) [Sov. Phys. JETP **39**, 784 (1974)].
[4] T. W. Hänsch and A. L. Schawlow, Opt. Commun. **13**, 68 (1975).
[5] V. S. Letokhov *et al.*, Zh. Eksp. Teor. Fiz. **72**, 1328 (1977) [Sov. Phys. JETP **45**, 698 (1977)].
[6] P. D. Lett *et al.*, Phys. Rev. Lett. **61**, 169 (1988).
[7] C. Monroe *et al.*, Phys. Rev. Lett. **65**, 1571 (1990).
[8] M. Kasevich *et al.*, Phys. Rev. Lett. **66**, 2297 (1991).
[9] M. Zhu *et al.*, Phys. Rev. Lett. **67**, 46 (1991).
[10] A. P. Kazantsev and I. V. Krasnov, JETP Lett. **46**, 332 (1987).
[11] V. S. Voĭtsekhovich *et al.*, Zh. Tekh. Fiz. **58**, 1174 (1988) [Sov. Phys. Tech. Phys. **33**, 690 (1988)].
[12] V. S. Voĭtsekhovich *et al.*, JETP Lett. **49**, 161 (1989).
[13] V. S. Voĭtsekhovich *et al.*, Ukr. Fiz. Zh. **38**, 211 (1993).

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