OPTICAL PUMPING AND DISSOCIATION OF RUBIDIUM MOLECULES BY A LASER PULSE

N.N. Kostin, V.A. Khodovoi, V.V. Khromov, and N.A. Chigir'
S.I. Vavilov State Optical Institute
Submitted 25 October 1971
ZhETF Pis. Red. 14, No. 11, 589 - 592 (5 December 1971)

The phenomenon of optical pumping (sometimes called optical orientation) of atoms is well known and consists in the general case of producing a non-equilibrium distribution of the atoms over the sublevels of the ground state [1 - 3]. We report here an attempt to observe an analogous phenomenon in vapors of diatomic molecules (see also [4, 5]).

1. In the first experiment we observed the kinetics of saturation of absorption of the radiation of a ruby laser operating in the spike generation regime with an off-duty ratio of about 10 and a total pulse duration 700 μsec, saturated by vapor of molecular rubidium Rb_2 (transitions $^1\Sigma_g - {}^1\Pi_u$ and $^1\Sigma_g - {}^1\Sigma_u$). A pulse of laser radiation passing through the Rb_2 vapor, with a leading front duration not exceeding 20 μsec, was registered on the screen of a long-persistence oscilloscope (S1-29). At an incident laser pulse intensity I_0 on the order of 10 W/cm^2 and a Rb_2 vapor pressure of about 0.1 mm Hg, we observed a stretching of the leading front of the envelope of the pulse passing through the vapor, up to 100 - 200 μsec (Fig. 1). There was no such stretching at lower ($I_0 \ll 10$ W/cm^2) and higher intensities ($I_0 \gg 10$ W/cm^2).

2. In the second experiment we photographed the Rb_2 absorption spectrum both during the time of the laser pulse (with approximately the same parameters) and after its termination. The trial radiation source employed was an ISK-25 lamp with flash duration 20 μsec. In this experiment, the optical density at the ruby-laser emission frequency was ∿6. The following was established:

a) Under the influence of the laser radiation, a decrease of absorption takes place uniformly over the entire $^1\Sigma_g - {}^1\Pi_u$ band. The time of action of the laser radiation necessary for complete bleaching of the vapor in the entire absorption band is ∿100 μsec and does not depend on the intensity I_0 if the latter exceeds a definite level (10^3 W/cm^2)[1].

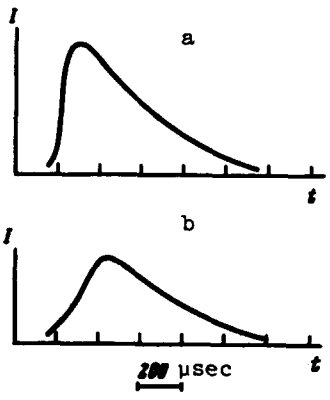

Fig. 1. a - Incident radiation pulse, b - pulse of radiation transmitted through Rb_2 vapor.

b) After the termination of the laser pulse, the absorption is uniformly reestablished in both red bands with a time constant ∿2 × 10^{-2} sec. Figure 2 shows the dependence of the absorption coefficient of Rb_2 on the time after the termination of the laser pulse, measured at the laser-emission wavelength.

We propose that the most probable mechanism of the observed phenomena is the dissociation of the Rb_2 molecules into Rb atoms under the influence of the laser radiation, followed by a slow (∿10^{-2} sec) restoration of the Rb_2 molecules. From an analysis of the absorption and fluorescence spectra of Rb_2 under the influence of ruby-laser radiation it follows that the absorption of the laser radiation gives rise to transitions from the vibrational levels v" = 5 - 10 of the ground $^1\Sigma_g$

[1] Complete bleaching of the vapor was observed also in the $^1\Sigma_g - {}^1\Sigma_u$ band.

state to the vibrational levels v' = 2 - 7 of the excited $^1\Pi_u$ state [6]. Direct transitions from the levels v" = 0 - 4, corresponding to the maximum of the absorption band, do not take place, since the ruby-laser emission frequency is lower than the frequency of the 4 - 0 transition in the $^1\Sigma_g$ - $^1\Pi_u$ band. The stretching of the leading front of the envelope of the pulse passing through the vapor, observed under these conditions, is evidence of depletion of a number of levels of the ground state (formation of a non-equilibrium population) and subsequent relaxation to these levels of the molecules from all other vibrational states. The uniform saturation of the absorption noted in the second experiment, in the band 6600 - 7400 Å within a time ∿100 μsec, indicates that the thermalization of the distribution of the populations over the rotational-vibrational sublevels of the ground state occurs under our conditions within a time ∿100 μsec.

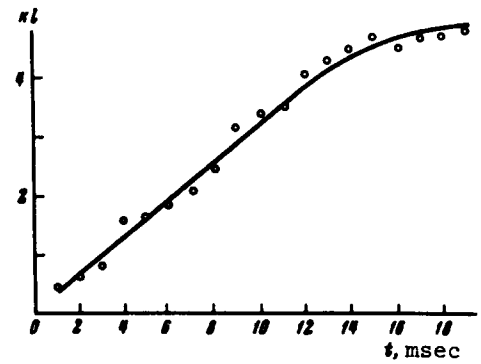

Fig. 2

3. To determine the factor governing the time of total bleaching of the Rb_2 vapor, we attempted to investigate the kinetics of the saturation of the absorption of Rb_2 under the influence of a giant ruby-laser pulse. To this end a cell with rubidium vapor was illuminated simultaneously by radiation of a giant pulse of one laser and the radiation of another ruby laser operating in the spike-generation regime. The radiation intensity of the spike laser was attenuated to a level corresponding to its linear absorption. The width of the radiation spectrum of both lasers and their frequency mismatch did not exceed 0.03 cm^{-1}. That such a degree of agreement between the frequencies of the two lasers is sufficient was confirmed by the decreased absorption of the beam of the spiked laser during the time of action of the giant pulse. Under these conditions, no residual bleaching of the molecular rubidium was observed for the time intervals larger than 3×10^{-7} sec after the action of the giant pulse. It follows therefore that the population of the rotational-vibrational level depleted by the radiation of the giant pulse occurs within a time shorter than 3×10^{-7} sec at the employed molecular vapor pressure (the pressure of the atomic vapor under these conditions was ∿10 mm Hg). The presence of such a fast relaxation in the ground state suggests that the time of complete bleaching of the Rb_2 vapor (∿10^{-4} sec) is determined by the time of dissociation of the Rb_2 molecules from the excited $^1\Pi_u$ state.

Molecular vapors were used by us for simultaneous passive Q switching and decreasing the frequency instability of the ruby and neodymium laser radiations, to values less than 10^8 Hz [7, 8]. It was already noted in [8] that generation of a series of weak neodymium-laser pulses is connected with the presence of residual bleaching of the Cs_2 vapor.

We believe that further investigation of the observed phenomena will clarify their mechanism and determine the constants of the molecular dissociation and recombination, and also of the rate of molecular relaxation in the ground and excited states. By using the radio-optical spectroscopy methods for the observation of non-equilibrium distributions of the molecules over the sublevels of the ground and excited electronic states (vibrational, rotational, Zeeman, and Stark), optically-pumped molecular vapor can be used to measure the frequencies of molecular transitions, and also the constants of the Zeeman and Stark splitting.

The authors are deeply grateful to Prof. A.M. Bonch-Bruevich for support and interest in the work.

[1] A. Kastler, J. de Physique 11, 255 (1950).
[2] G.V. Skrotskii and T.G. Izyumov, Usp. Fiz. Nauk 73, 423 (1961) [Sov. Phys. -Usp. 4, 177 (1961)].
[3] A. Kastler and C. Cohen-Tannoudji, Progr. Opt. 5, 3 (1966).
[4] R.E. Drullinger and R.N. Zare, J. Chem. Phys. 51, 5531 (1969).
[5] J.H. Simpson, D.S. Bayley, and E.C. Eberlin, Bull. Amer. Phys. Soc., Ser. 11, 16, No. 1, 107 (1971).
[6] N.N. Kostin, M.P. Sokolova, V.A. Khodovoi, and V.V. Khromov, Zh. Eksp. Teor. Fiz. 62, No. 2 (1972) [Sov. Phys.-JETP 35, No. 2 (1972)].
[7] A.M. Bonch-Bruevich, N.N. Kostin, V.A. Khodovoi, and V.V. Khromov, ZhETF Pis. Red. 12, 354 (1970) [JETP Lett. 12, 242 (1970)].
[8] N.N. Kostin, V.A. Khodovoi, and N.A. Chigir', Opt. Spektrosk. (in press).

EXPERIMENTAL OBSERVATION OF ELECTRONIC SHOCK WAVES IN A COLLISIONLESS PLASMA

A.A. Ivanov, L.L. Kozorovitskii, V.D. Rusanov, R.Z. Sagdeev, and D.N. Sobolenko
Submitted 28 October 1971
ZhETF Pis. Red. 14, No. 11, 593 - 596 (5 December 1971)

The existence of collisionless thermal waves in a plasma was first demonstrated experimentally in [1]. The occurrence of ion-acoustic noise on the front of a thermal wave was noted in [2, 3], i.e., the feasibility of a stationary discontinuity was demonstrated [4].

In the present paper we considered experimental proofs of the existence of a stationary heat discontinuity in a collisionless plasma (electronic shock wave) and the connection of its parameters with the theoretical concepts [4, 5].

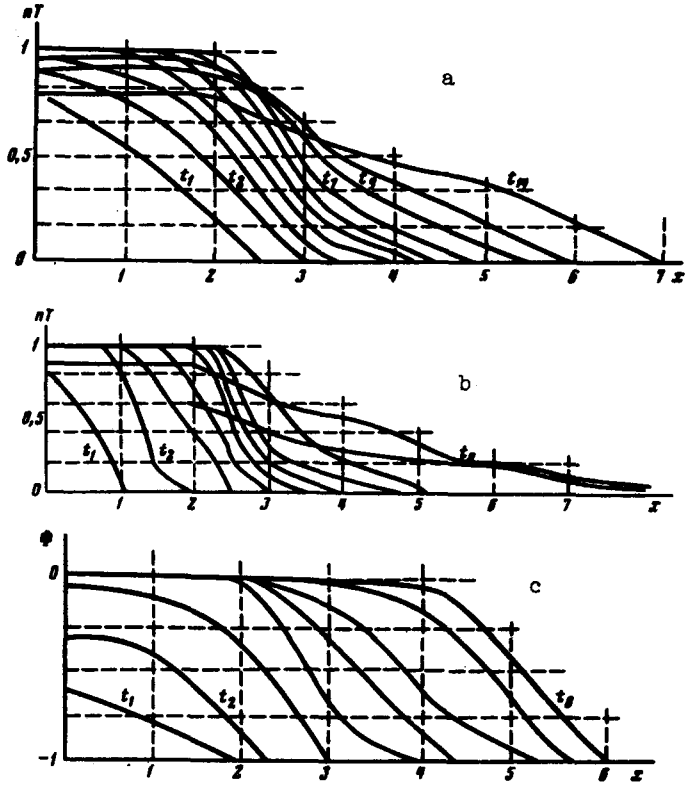

Fig. 1. a - Instantaneous profiles of $n_h T_h(x)$ for a discharge in hydrogen: $\Delta t = 10^{-8}$ sec, nT is in relative units, $\Delta x = 10$ cm; b - instantaneous profiles of $n_h T_h(x)$ for a discharge in argon: $\Delta t = 10^{-8}$ sec, $n_h T_h$ is in relative units, $\Delta x = 5$ cm; c - instantaneous profiles of Φ for a discharge in argon: $\Delta t = 10^{-8}$ sec, Φ is in relative units, $\Delta x = 5$ cm.