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HYDROGEN LASER IN VACUUM ULTRAVIOLET AT ATMOSPHERIC PRESSURE

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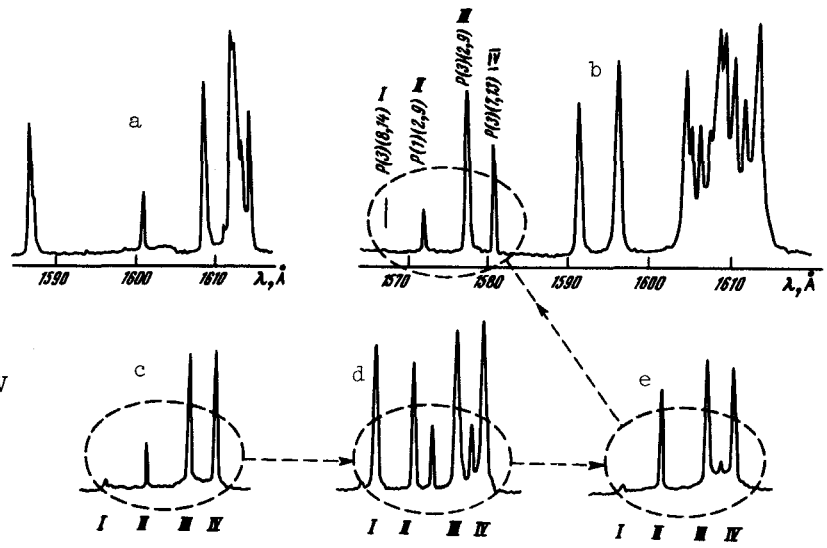
The first observation of superradiance in the vacuum ultraviolet on transitions of molecular hydrogen and deuterium at atmospheric pressure is reported. When the pressure is increased, intensity anomalies are observed in the superradiance spectrum for transitions terminating on vibrational levels near the dissociation limit of the H_2 molecule. Ten new laser transitions were observed.

Quantum electronics methods are being extended to the vacuum ultraviolet (VUV) band by developing pulsed VUV coherent-radiation lasers of high power [1, 2] and by finding new methods of multiplying the frequencies of high-power pulses in the visible band [3, 4]. One of the promising trends is to increase the power of H_2 lasers by increasing the pressure and changing over to four-level operation [5]. We report here significant progress made in this direction.

To obtain lasing at atmospheric pressure, we used a transverse discharge in a working volume $0.006 \times 1 \times 35$ cm^3 of a Blumlein flat transmission line measuring 35×60 cm with variable profile and wave resistance 5 Ω through an asymmetrically placed commutator with solid dielectric. The combination of high current density, $j = 10^5$ A/cm^2 with satisfactory spatial uniformity of the plasma improves greatly the conditions of laser excitation in a transverse discharge with a narrow channel. The electrodes were copper foil strips 50 μ thick with polished edges, glued between segments of polished glass at a distance 1 ± 0.05 cm. Visual indication of the short-wave radiation was through a uranyl-glass window with a control glass plate secured 15 cm away from the laser channel inside a vacuum cell.

In the entire investigated pressure interval, up to 1 atm, unidirectional radiation is observed, with divergence ∿2° in the plane of the working channel and ∿1° in the perpendicular plane, at a line voltage somewhat higher than the threshold value V_{thr}. The threshold voltage at atmospheric pressure, $V_{thr} = 28$ kV, is only double the value at $p = 0.1$ atm.

The spectra of the laser cell emission on the electronic transition $B^1\Sigma_u^+ \rightarrow X^1\Sigma_g^+$ of molecular hydrogen and deuterium at a gas pressure 1 atm were registered with the VMS-1 instrument with wavement-measurement accuracy 0.1 Å. Microphotograms of the emission spectra are shown in the figure. The observed radiation does not satisfy the line-intensity relations



Microphotograms of superradiance spectra of hydrogen and deuterium: a) D_2, 0.6 atm, V = 80 kV; b) H_2, 1 atm, 80 kV; c) H_2, 0.1 atm, 20 kV kV; d) H_2, 0.1 atm, 40 kV; e) H_2, 0.3 atm, 80 kV.

characteristic of spontaneous emission on the P and R branches, ortho- and paramodifications, and different vibrational transitions. Out of a total of several hundred molecular-hydrogen spontaneous-emission lines, the laser-cell emission contains only 23 lines.

The intensity ratios characteristic of the laser-cell emission lines are on the whole preserved when the gas pressure and line voltage are changed. Exceptions are the transitions P(3)(8-14) and P(3)(7-13) transitions (lines I and IV in the figure), which terminate at levels near the dissociation limit. At low gas pressure (Figs. c and d) the relatively weak line I becomes comparable with the most intense spectral lines, e.g., line III, when the voltage is increased. The intensity ratio of lines I and II changes, the former becoming more intense. An increase in the gas pressure is accompanied by a gradual overall weakening of the line intensity. Line I becomes particularly weak and disappears from the spectra at $p \gtrsim 0.3$ atm. The ratio of lines III and IV changes (Figs. d, e, b), the latter becoming less intense.

New laser emission lines were observed at the following wavelengths (in Å) for hydrogen: 1494.2 - P(3)(11-14), 1565 - R(1)(8-14), 1574 - P(2)(2-9), 1614.8 - P(4)(5-12), and 1616.5 - P(5)(5-12); the new lines for deuterium were 1589.0, 1592.3, 1602.1, 1606.3, and 1609.5 Å (the D_2 lines were not identified for lack of data on the spontaneous emission spectrum). Under optimal excitation conditions, the laser-pulse energy measured with a calibrated vacuum thermopile is $\sim$0.5 mJ.

The unidirectional character and small divergence of the radiation, as well as the general form of the observed emission spectrum, are unambiguous symptoms of superradiance in excitation in the traveling-wave regime.

When the gas pressure and the discharge-gap voltage are changed, the excitation rate of the vibrational-rotational levels remains practically constant within the limits of one electronic state, and the characteristic intensity ratio of the superradiance lines should remain constant. The observed anomalous increase of the intensity of the line P(3)(8-14) with increasing voltage is apparently due to the influence of dissociative deactivation. The collision lifetime of the level $v'' = 14, J'' = 3$ due to direct transitions to the continuous spectrum and on pre-dissociation levels in collisions is $\tau_{14-3}^{col} \approx [2^{\frac{1}{2}} N_0 \pi (r_0 + r_{v''})^2 v]^{-1} \times \exp(\Delta\varepsilon_{14-3}/kT) \approx 4.5 \times 10^{-11}$ sec-atm (r_0 and $r_{v''}$ are the effective radii of the molecule in the ground and excited vibrational states, and $\Delta\varepsilon_{14-3} \approx 100$ cm^{-1} is the energy needed to go over to the continuum). At $p = 0.1$ atm this time is shorter than the radiative lifetime of the upper working states ($\tau_{14-3}^{col} \approx 0.4$ nsec $< \tau_{rad} = 0.8$ nsec). Because of this, the time of existence of inversion for the P(3)(8-14) transition is much larger than for all other transitions, which are self-limited. An increase of the voltage on the Blumlein line, within certain limits, leads to an increase of the effective duration of the excitation pulse, and consequently to an increase of the relative emission energy in the P(3)(8-14) line. The observed decrease in the intensities of the P(3)(8-14) and P(3)(7-13) lines of hydrogen with increasing pressure is apparently due to a decrease in the gain, caused by the impact line broadening, which is apparently largest just for the levels with $v'' = 13$ and 14.

The experimentally observed relatively small excitation thresholds at $p = 1$ atm are evidence that there is no appreciable self-quenching of the hydrogen molecules[1]. The formation of a traveling superradiance wave at a length 30 cm indicates that the duration of the radiation pulse is much less than $L/c \approx 1$ nsec ($L = 35$ cm). At a pulse energy 0.5 mJ this corresponds to a peak power higher than 1 MW and a power density 200 MW/cm^2 at the cell exit.

The low effective duration of the excitation pulse, owing to the rapid decrease of the electric field and of the energy of the electrons in the discharge gap at the instant of breakdown, does not make it possible for the main advantage of the four-level scheme, the absence of a limitation on the lifetime of the gain and generation, to manifest itself. To excite a four-level collision high-pressure hydrogen laser it is therefore advisable to use strong-current electron beams, which are completely free of this shortcoming, and which ensure a high pumping level.

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MEASUREMENT OF THE CROSS SECTIONS OF SPIN EXCHANGE OF H ATOMS ($F = 1$, $m_F = 0$) ON PARAMAGNETIC O_2, NO, and NO_2 MOLECULES IN THE TEMPERATURE INTERVAL 310 - 390°K

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A hydrogen laser operating in the pulsed regime is used to measure the cross sections of spin exchange of H atoms in the hyperfine state ($F = 1$, $m_F = 0$) on the paramagnetic molecules O_2, NO, and NO_2. The cross sections are equal to 0.85×10^{-15} cm^2, 1.02×10^{-15} cm^2, and 0.88×10^{-15} cm^2, respectively.

When hydrogen atoms collide with paramagnetic molecules, the spin exchange process, which leads to a change of the hyperfine state of the hydrogen atoms, has a large cross section. On the other hand, it is known that in this case the chemical joining reaction with subsequent decay or collision stabilization of the complex has a high probability. The present paper is devoted to a measurement of the cross sections of spin exchange of hydrogen atoms with the paramagnetic molecules O_2, NO, and NO_2, to the dependence of the cross sections on the temperature, and to a comparison of the cross sections of spin exchange and of the chemical reaction of the hydrogen atoms with these molecules.

Calculations show [1 - 3] that the spin-exchange process is coherent, i.e., the phase information connected with the oscillating magnetic moment of the hydrogen-atom system is conserved during the course of the interaction. The ratio of the relaxation times T_1 of the population difference of the hyperfine levels and T_2 of the oscillating magnetic polarization is equal to 3/4 in the case of interaction of a hydrogen atom with a paramagnetic particle. The experimental values of the mean cross sections for the change of the hyperfine structure in spin exchange at 300°K are $\sigma(H\text{-}O_2) = (1.9 \pm 0.2)\times10^{-15}$ cm^2, $\sigma(H\text{-}NO) = (1.9 \pm 0.2)\times10^{-15}$ cm^2, and $\sigma(H\text{-}NO_2) \sim 2\times10^{-15}$ cm^2.

We measured the rate constants K_1^{se} and K_2^{se} of the spin exchange of a hydrogen atom in the state ($F = 1$, $m_F = 0$) colliding in the gas phase with the molecules O_2, NO, and NO_2 in the temperature interval 310 - 390°K.

The procedure [5, 6] consisted of measuring the cross sections for the relaxation of the hyperfine levels of the ground state of the hydrogen atoms in the collisions with the molecules of the investigated gas with the aid of a hydrogen maser operating in the pulsed regime. The inverted population of the levels ($F = 1$, $m_F = 0$) and ($F = 0$, $m_F = 0$) of the H atoms in the maser accumulating flask, placed in a microwave resonator, was produced by focusing the atom beam in an inhomogeneous magnetic field. The investigated gases were also admitted into the flask, and their concentration was in the interval $(0.5 - 3.5)\times10^{10}$ molecules/cm^3, while the H-atom concentration was about 10^8 atoms/cm^3. The oscillating magnetic polarization of the system of atoms was excited with a 90° pulse at the transition frequency, with duration that was short in comparison with T_1 and T_2. Since the inverted population did not exceed one-tenth of the threshold value for generation under the experimental conditions, the coherent-radiation signal of the system of atoms varied in time like $\exp(-t/T_2)$. The absolute value of the time was obtained from the decrease of the initial amplitude of this signal, from the time between the instant when the flow of focused atoms into the flask stopped and the start of the 90° pulse. The rate constants of the spin exchange were determined from the relation $T_{1,2} = (K_{1,2}^{se} n_B)^{-1}$, where n_B is the concentration of the investigated molecules (O_2, NO, or NO_2) in the accumulating flask.

The measurement results are listed in the table (in the case of NO_2, the obtained cross sections were extrapolated to the instant when the gas was admitted, to exclude the influence of its adsorption on the walls of the flask).

For all the partners, within the limits of the experimental accuracy, we obtained