

FREQUENCY SHIFT OF OPTICAL TRANSITION IN THE FIELD OF A LIGHT WAVE

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We have observed that the resonant optical transition frequency in potassium vapor shifts by about  $10^9$  cps under the influence of a powerful ruby-laser pulse.

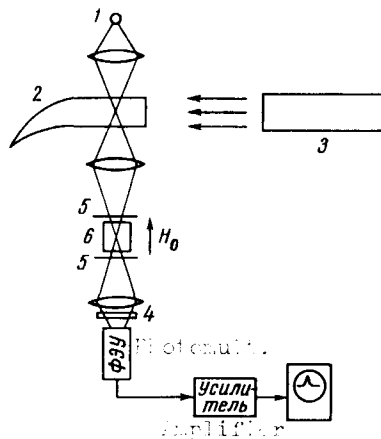
The shift  $\Delta W_i$  of the energy level  $i$  under the influence of the electric field  $E = 2E_0 \cos \omega t$  of a light wave is given by the formula [1]

$$\Delta W_i = \sum_k \left\{ \frac{|\langle i | \vec{d} \cdot \vec{E}_0 | k \rangle|^2}{E_i - E_k + \hbar\omega} + \frac{|\langle i | \vec{d} \cdot \vec{E}_0 | k \rangle|^2}{E_i - E_k - \hbar\omega} \right\} \quad (1)$$

where  $E_i$  and  $E_k$  are the energies of states  $i$  and  $k$  of the atom, and  $\langle i | \vec{d} \cdot \vec{E}_0 | k \rangle$  is the matrix element of the dipole transition between states  $i$  and  $k$ . Unlike the formula that describes the level shift in a constant electric field (Stark effect), the shift in the field of a light wave can increase resonantly when the light quantum energy  $\hbar\omega$  approaches the transition energy  $E_k - E_i$  of the atoms <sup>1)</sup>.

We investigated in the experiment the frequency shift of the optical resonant transition  $4S_{1/2} - 4P_{1/2,3/2}$  of potassium (principal doublet). It can be shown that the expected frequency shift of this transition is connected principally with virtual transition induced by the laser pulse from the ground level ( $4S_{1/2} - 4P_{1/2,3/2}$ ) and the excited level ( $4P_{3/2} - 6S_{1/2}$ ). The first pair of transitions is still sufficiently far from the resonances (the transition wavelengths are 7665 and 7699 Å, that of the laser is 6943 Å). The  $4P_{3/2} - 6S_{1/2}$  transition is much closer to resonance ( $\lambda = 6939$  Å). In spite of this, all these transitions make comparable contributions to the sought frequency shift of the investigated transition, owing to the difference in the oscillator strengths [3]. It is important that the ground and excited levels are shifted here by the ruby-laser light in opposite directions.

The experiment was organized as follows. Light from potassium lamp 1 was passed through vessel 2 with potassium vapor saturated at 100°C (see the figure). At the selected temperature, the vapor absorbed about 80% of the lamp's resonant radiation. It was expected that the transmission of light by vessel 2 would increase during the action of the pulse from laser 3, provided the resultant transition frequency shift is commensurate with the line width of the lamp radiation (it was assumed that this line was broader than the absorption line of the vapor). The transmission of the resonant light was recorded with a photomultiplier whose output was fed to a pulsed oscilloscope (4 - glass filters).



The scattered laser light in the registration channel was reliably cut out with FS-7 filters. Nonetheless, preliminary experiments have shown that the laser pulse is accompanied by scattered radiation with spectral components lying in the region of the registered potassium line. The mechanism of occurrence of this radiation has not yet been finally explained. To combat it, we used a special method of filtering the resonant line with the aid of the Faraday effect [4]. After passing through vessel 2, the light beam of the potassium lamp was made to pass through an auxiliary cuvette 6 filled with potassium vapor and placed between crossed polaroids 5. A local magnetic field of approximately 2 kOe was applied to cuvette 6. The magnetic field produced, besides splitting of the absorption line, also strong radiation of the plane of polarization of the light, but only in the nearest vicinity of optical resonance, so that by suitable choice of the magnetic field intensity the system was made to transmit almost all the resonant line, and to absorb the extraneous light. The entire apparatus behaves like a high-transmission optical filter with a bandwidth on the order of  $0.1 \text{ cm}^{-1}$ .

Under the conditions described, a distinct signal was recorded, evidencing a decrease in the absorption of the resonant light by the potassium atoms in vessel 2 during the time of action of the laser pulse (20 nsec); the laser operated in the monopulse mode by using bleaching filters KS-19. The amplitude of the signal corresponded to bleaching of the potassium vapor by sever dozen per cent. To verify that the change in the light absorption was not connected with some experimental errors we checked that (i) the signal vanished when the potassium light was turned off, (ii) the signal vanished when the potassium vapor was frozen out in vessel 2 (with the illumination on the photomultiplier maintained at the previous level), and (iii) the signal vanished when the operating mode of lamp 1 was forced so as to broaden the emission line (the broadening was confirmed by the observations).

The minimum laser radiation power density at which the bleaching signal was produced was  $\sim 10 \text{ MW/cm}^2$ , corresponding to an electric field intensity (in the light) of  $10^5 \text{ V/cm}$ . The half-width of the spectral emission line is estimated at  $\sim 3 \times 10^9 \text{ cps}$ , so that the observed shift was of the same order.

It must be noted that realization of a similar level shift in a constant electric field would call for field intensities of the order of  $10^6 \text{ V/cm}$ , which is at the limit of the experimental capabilities. Yet the production of such a field in a light wave (without producing breakdown in low-density vapor) is now perfectly feasible. The use of lasers therefore affords new possibilities of investigating the Stark effect in systems with transition frequencies that are either closer to or far from the laser emission frequency.

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- [2] A. Kastler, *J. Opt. Soc. Amer.* 53, 903 (1963).
- [3] D. S. Villars, *ibid.* 42, 552 (1952).
- [4] E. B. Aleksandrov, *Optika i spektroskopiya* 19, 455 (1965).

1) Kastler's group [2] has recently observed in the radio-frequency region a small re-

lative shift (on the order of 1 cps) of the magnetic nuclear sublevels of the ground state of mercury under the influence of 2537 Å resonant radiation.

#### ROTATIONAL STRUCTURE OF ULTRAVIOLET GENERATION OF MOLECULAR NITROGEN

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A laser generating in the ultraviolet and using molecular nitrogen was first described in [1]. So far, however, no data on the emission spectrum were published. Yet such data are essential both for an explanation of the physical mechanisms that lead to inversion and emission, and for practical applications. In this communication we present results of an investigation of the emission spectrum.

An ordinary laser was used with external mirrors and with windows at the Brewster angle. The discharge was excited with voltage pulses up to 40 kV in a tube of 3 mm i.d. and a discharge length ~ 90 cm. The current pulse duration was ~1.5 μsec. Mirrors with multilayer dielectric coatings were used, having a transmission ~ 40% in the  $\lambda \approx 3370$  Å region, as well as sputtered-aluminum mirrors.

Generation was observed at two bands (0-0 and 0-1) of the second positive nitrogen system  $C^3\Pi_u \rightarrow B^3\Pi_g$ . The generation power in the 0-0 band (3371 Å edge) is many times larger than in the 0-1 band (3577 Å edge). A considerable super-radiance effect is observed in the 0-0 band <sup>1)</sup>. Radiation with a single mirror has practically the same spectrum as the generation radiation, and differs only slightly in power. In investigation of the radiation from the tube without mirrors, a sharp increase was observed in several lines, compared with the normal spontaneous emission spectrum. This increase is apparently also connected with the super-radiance effect.

The results presented below were obtained at nitrogen pressures close to optimal: ~ 2 torr for the 0-0 band and ~1 torr for the 0-1 band. Spectrally pure nitrogen was used in the experiments, but impurities apparently play a minor role, for in practice similar generation could be observed when the discharge tube was filled with air.

The generation spectrum was investigated with a DFS-13 spectrograph with a 600 lines/mm grating. The spectrum of the 0-0 band was photographed in third order with dispersion ~1.3 Å/mm, and the 0-1 band in second order with dispersion ~ 2.0 Å/mm. In addition to the generation spectrum, to facilitate interpretation of the lines, we photographed the spectrum of the spontaneous emission. The comparison was against the iron and titanium lines. To eliminate random shifts, the generation spectrum was measured with a large number of plates. The estimated wavelength measurement accuracy is  $\Delta\lambda \approx 0.02$  Å for the 0-0 band and  $\Delta\lambda \approx 0.04$  Å for the 0-1 band.

The measurement results are listed in Tables 1 and 2.