Simultaneous determination of the Lande g factor and relaxation rate of the ground state for diatomic molecules using the Hanle effect

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If the absorption rate Γ_p on optical excitation of a diatomic molecule $(\alpha'', v'', J'') \rightarrow (\alpha', v', J')$ from the vibrational rotational level v'', J'' of the ground electronic state α'' is comparable with the rate of its thermalization γ, the possibility appears of observing the Hanle effect[1] of the lower level in the resonance series of laser-induced fluorescence (LIF) from the upper level (α', v', J').[2] The phenomenon bears a nonlinear character and is due to disruption of the polarization moments (PM) φ_g^x of the lower level by the external magnetic field B. The moments φ_g^x are connected by the light field with the PM of the upper level, and through them the Hanle effect is manifested in the LIF.[3,4] The effect is conveniently recorded using measurements of the degree of linear polarization P on imposition of a field along the direction of observation, orthogonal to a linearly polarized exciting laser beam and its vector E. When the width of the Hanle contour of the lower level, dependent on the ratio γ/g, where g is the Lande factor, is much less than that of the upper level, the effect of the ground state in its general features is manifested as an increase in P, and only in large fields sets in because of depolarization due to the Hanle effect of the excited level (see Fig. 1). Earlier[2,5] the problem was solved as follows: From the experimentally determined ratio γ/g using the known value of g to determine the relaxation rate γ for $(x^1\Sigma_g^+, v'', J'')$ of Na_2 and K_2. However, the values of the Lande g factors of diamagnetic ground electronic states have been measured only for alkali dimers by Ramsay's molecular beam method[6] and for (O_g^+) Te_2 by beat resonances[7]; their calculation is also difficult. In the present work a method of simultaneous determination of γ and g from experiment using the Hanle effect is proposed.

For the excited state extrapolation to conditions where there is no collisional relaxation permits from the Hanle signal determining $g\tau_{sp}$, where τ_{sp} is the spontaneous decay time.[1] If for the ground state one emerges into the collisionless region of concentrations, then with an exciting beam narrow in comparison with the diameter of the cell relaxation remains due to exchange of molecules aligned in the state (α'', v'', J'') under the action of the beam with the isotropic molecules of the remaining volume as a result of their motion, which sets in transverse to the laser beam. At first glance the rate of such a process is determined only by the diameter of the beam and can easily be calculated, as assumed by Drullinger and Zare.[8] But for a real, for example, Gaussian profile of a laser beam the pattern is much more complex, especially on effective devastation, when $\Gamma_p/\gamma \gg 1$.[9,10] Thus, the kinetics of relaxation of the ground state after switching off of pumping has a nonexponential charac-

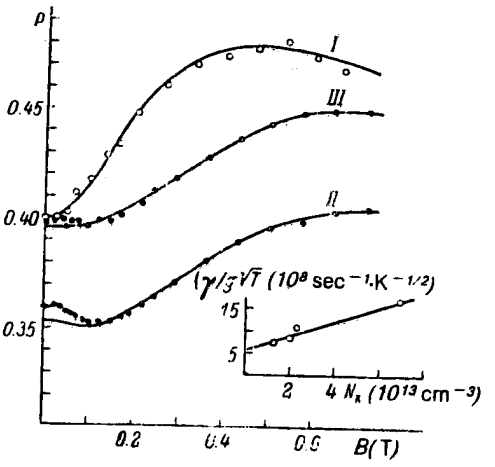

FIG. 1. Degree P of linear polarization of fluorescence of K_2 vs magnetic field B. I—for rectangular cross sections of laser beam of diameter $d_0 = 3$ mm at $T = 437$ K. $N_K = 2.3 \times 10^{13}$ cm^{-3}; II—for Gaussian profile of a beam at $T = 463$ K, $N_K = 7.1 \times 10^{13}$ cm^{-3}; III—the same on attenuation of pumping by a factor of 1.8. Below right—ratio $\gamma/g\sqrt{T}$ vs concentration of potassium atoms N_K.

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ter and depends on Γ_p/γ. Only for a beam of rectangular profile can one succeed in obtaining an analytical expression,[9] which is sufficiently well approximated by an exponent with index $\gamma_0 = 1.708\, v_{mp}/d_0$, where $v_{mp} = \sqrt{2kT/m}$ is the most probable velocity of molecules with mass m and d_0 is the beam diameter. In this case the relaxation rates on flight and in collisions is additive, i.e., in the thermal cell $\gamma = \gamma_0 + v_{rel}\sigma N$, where N is the particle concentration, $v_{rel} = \sqrt{8kT/\pi\mu}$ is the average relative velocity of the colliding partners, μ is the reduced mass, σ is the effective cross section of collisional relaxation; therefore, if in the experiment one creates a beam giving in cross section a homogeneous illuminated circle, the linear dependence of the ratio $\gamma/g\sqrt{T}$ on the concentration N is written in the form

$$\frac{\gamma}{g\sqrt{T}} = \frac{1.708}{gd_0}\sqrt{\frac{2\kappa}{m}} + \frac{\sigma N}{g}\sqrt{\frac{8kT}{\pi\mu}} \qquad (1)$$

and permits from the ordinate at $N = 0$ finding the Lande g factor and then changing to the relaxation rate γ and cross section σ.

In the experiment an LG-38 He–Ne laser of 60-mW power at the 632.8-nm wavelength was used in the TEM_{00q} lasing regime with a Gaussian beam width of 3 mm. Using a long-focal-length diverging lens the beam is broadened by 3 times, and its central part is isolated by a diaphragm of diameter $d_0 = 2.5$–4.0 mm. The monitoring of the power distribution in the beam cross section, performed by scanning with a lightguide with a small entrance aperture, showed homogeneity within the limits 20–30%. In potassium vapors at thermal equilibrium the transition ($\alpha'' = X\,^1\Sigma_g^+$, $v'' = 1$, and $J'' = 72$)$\rightarrow$($\alpha' = B\,^1\Pi_u$, $v' = 8$, $J' = 72$) was excited in the K_2 molecule. The degree of polarization on the Q_{16} line of the resonance series of LIF isolated by a DFS-12 monochromator as a function the field B created by an electromagnet ($B \leqslant 0.8$ T with the gap 40 mm, a rotating mirror in the gap guaranteed observation of LIF along B) by the method of photon counting with accumulation. Typical data of the experiment are given in Fig. 1 (curve I) for $d_0 = 3$ mm. The solid curve is a calculation using the system of kinetic equations for the PM in the limit $J'' \rightarrow \infty$.[3,4] Minimization of the deviations was performed on variation of the two nonlinear parameters γ/g and Γ_p. In Fig. 1 the dependence of the value $\gamma/g\sqrt{T}$ on the concentration of potassium atoms N_K determined from the temperature of the vapors,[11] which play the role of a thermal reservoir for a small impurity ($\sim 10^{-3}$) of K_2 dimers is given in Fig. 1. Extrapolation to $N_K = 0$ permitted determining the value of the Lande g factor using Eq. (1). The value averaged over the experiments for some diameters of the beam d_0 is $g = (1.16 \pm 0.15) \times 10^{-5}$ which agrees within the limits of error both with the result 1.18×10^{-5} obtained in Ref. 6 by the beam method for the entire thermally populated $X\,^1\Sigma_g$ state of K_2 and also with the preliminary result $(0.99 \pm 0.05) \times 10^{-5}$ obtained in Ref. 12 by the method of nonlinear resonance of beats for ($X\,^1\Sigma_g^+$, $v'' = 1$, $J'' = 72$) K_2.

Further, using the value of g found one can in Eq. (1) change to the relaxation rate γ. For points in Fig. 1 γ varies from $0.18 \times 10^-$ up to 0.42×10^6 sec^{-1}. The cross section σ we did not determine here due to possible systematic errors in determining the absolute values of the concentrations of potassium atoms in a narrow furnace-thermostat; the order of magnitude of σ corresponds to that determined in Refs. 2 and 13.

The method proposed here permits determining the magnetic moment with higher error than, for example, nonlinear resonance of beats,[7,12] however, it has an advantage in simplicity of realization. On the other hand, the coincidence of results obtained by various methods,[6,12] indicates in favor of the adequacy of the model used of describing the Hanle signal. Matters stand somewhat differently for a beam of Gaussian profile at large values of the nonlinearity parameter $\Gamma_p/\gamma \gtrsim 3$. The form of the Hanle signal is complicated—an additional peak at $B = 0$ appears, detected experimentally in Refs. 2 and 5, to explain which it is necessary to adopt PM of rank $\varkappa = 6$.[3] Here this structure was measured more carefully (see Fig. 1, curve II). The peak had an amplitude near $\Delta P \simeq 0.01$ and almost disappeared on attenuation of excitation by 1.8 times (see Fig. 1, curve III) and also on a concentration increase. Here in the case given in Fig. 1 (curve II) we do not succeed in attaining qualitative coincidence of the calculated curve (solid line) with experiment. It is not excluded that manifestation of the narrow peak more expressed in comparison with calculation is connected with the nonexponential character of relaxation—with prolongation in the kinetics of thermalization due to flight across a beam of Gaussian profile,[10] which is capable of increasing the contribution of a narrower structure.

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