Hidden structure of the quasistatic wing of the atomic line of rubidium

T. A. Vartanyan, Yu. N. Maksimov, S. G. Przhibel'skiĭ, and V. V. Khromov
State Optical Institute

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Laser saturation spectroscopy methods are used to study the quasistatic wing of the atomic line of Rb formed by collisions between Rb and Xe atoms. The nonlinear excitation spectrum of atomic fluorescense shows a hidden structure whose nature is explained within the framework of the semiclassical theory in terms of the inhomogeneity of the ensemble of colliding atoms.

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In recent years considerable interest has arisen in nonlinear optical phenomena in colliding atomic systems.[1,2] This is due to both the nontrivial nature of the phenomena itself [1] and to purely practical needs–the development of excimer and gasdynamic recombination lasers, study of interatomic interaction potentials, kinetics of gaseous chemical reactions, etc. In this report we demonstrate the effectiveness of saturation spectroscopy methods[3] for studying atomic collisions.

The object of study was the Rb + Xe system with its well-known system of terms (Fig. 1).[4] Excitation of the short-wave wing of the D_2-line of Rb (transition $X\,^2\Sigma_{1/2}$–$B\,^2\Sigma_{1/2}$) was carried out by means of a pulsed tunable dye laser ($\lambda = 750$–780 nm, $\tau = 20$ nsec, $\Delta\lambda = 5$–$10\ \text{cm}^{-1}$). The integral intensity of Rb line luminescence ($\lambda = 780$ nm) was measured by a FEU-84 photomultiplier at right angles with the pumping beam direction through a MDR-2 monochromator. The Rb and Xe vapor pressure was 10 m Torr and 30 Torr, respectively.

The linear excitation spectrum is shown in Fig. 2(a). Spectral feature near $\lambda_s = 760$ nm (blue satellite) and the sharp cut-off of the spectrum on the shortwave

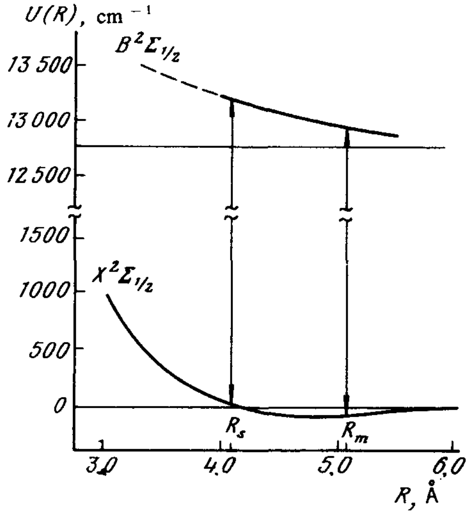

FIG. 1. System of the Rb + Xe terms.[4]

side is explained [4] by the parallelism of the terms $X\,^2\Sigma_{1/2}$–$B\,^2\Sigma_{1/2}$ in the vicinity of $R \approx R_s = 4.1$ Å (Fig. 1). Saturation of atomic luminescence intensity was observed at increased excitation power over the entire wavelength region in question. Figure 2(b) shows the shape of the excitation spectrum taken at laser intensity $I = 3\times10^8$ W/cm^2. For convenience, both spectra are normalized to the signal level at a single point ($\lambda = 765$ nm). Clearly, the greatest spectral deformation occurs at $\lambda_m = 775$ nm and $\lambda_s = 760$ nm.

Highly detailed data on the Rb + Xe terms makes the problem of interpretation of the revealed features of nonlinear spectrum easier. Actually, the wavelength $\lambda_m = 775$ nm is nearly equal to the transition wavelength between the ground and excited states at the point $R_m = 5$ Å (Fig. 1). This implies that the formation of a dip in this region may be explained by depopulation of the lower states which is accompanied by photo-dissociation of the van-der-Waals molecules. Thus, the depth of the dip characterizes the number of such molecules, and measurement of the intensity I_0 at which they form provides a method of determining in principle their production rate. We should also note that the number of van-der-Waals molecules is small and their presence is sufficiently detectable only in the nonlinear excitation spectrum (Fig. 2).

Deformation of the spectrum in the region $\lambda \approx \lambda_s$ is associated with the properties of collisions. Insofar as the effectiveness of excitation of an atomic system in the quasistatic region is determined by the length of its "being in resonance" with the radiation field,[5] it is qualitatively clear that the satellite deformation depends on the saturation of electronic-translational transitions which correspond to the slowest relative motion of atoms near each other.

Most interesting, we believe, is the "kinetics" of deformation of the excitation spectrum in the vicinity of a satellite as a function of laser power [Fig. 2(c)] whose value increased 3-fold for each of the successive spectra 1–5. Clearly, the altogether lightly deformed satellite has an even less deformed narrow core. In order to study this

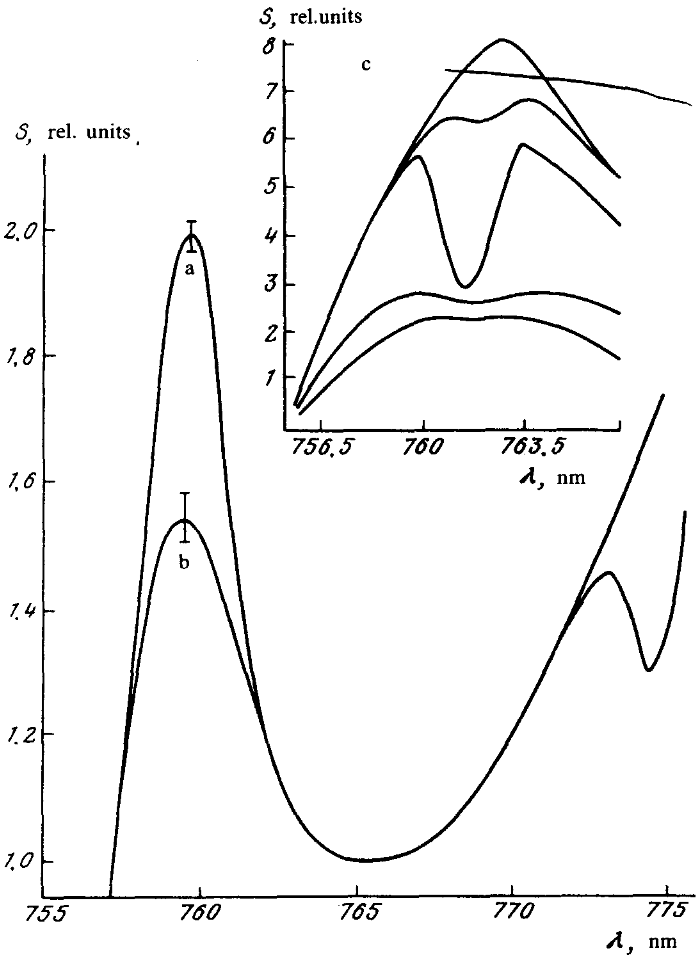

FIG. 2. Excitation spectra of atomic luminescence (a—at excitation intensity $I = 10^5$ W/cm²; b—$I = 3\times10^8$ W/cm²).

effect of inhomogeneous saturation more fully, we measured the dependence of the intensity of atomic fluorescence on the excitation power at $\lambda = \lambda_s$. Both experiments indicate the presence of a small group in the inhomogeneous set of colliding atoms whose probability of excitation by radiation with $\lambda = \lambda_s$ is considerably higher than that of the remaining atoms. As is known,[5] the transition probability is highest for those atoms for which the classical precession point is near the point R_s where the terms are parallel. We associate saturation of precisely this portion of the set with the occurrence of a plateau in the nonlinear function (Fig. 3) and a narrow dip in the satellite contour (Fig. 2). Subsequent behavior of the nonlinear function is explained by the slow saturation of the remaining portion of the atomic set whose collision energies and impact parameters provide means for a rapid non-stop passage through the neighborhood of point R_s.

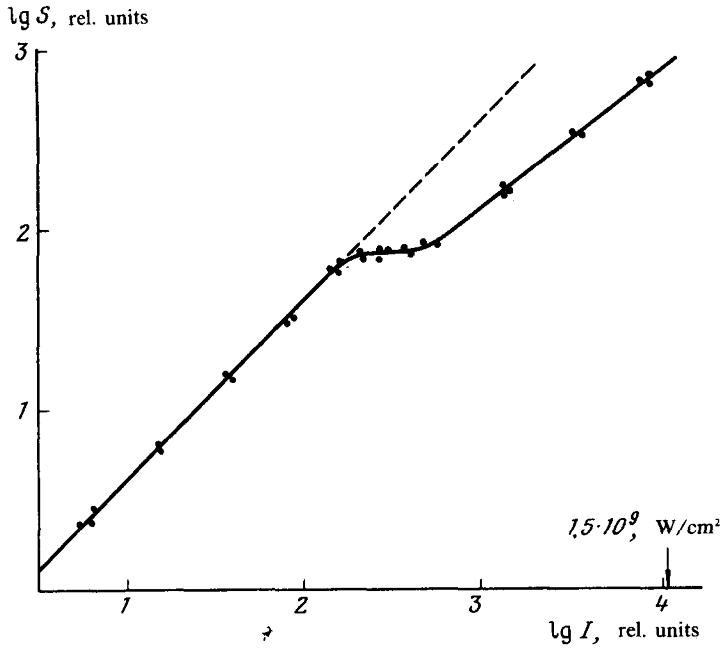

FIG. 3. Dependence of atomic luminescence intensity on excitation power ($\lambda_{exc} = 760$ nm).

Theoretical analysis of the satellite saturation pattern was carried out within the framework of quasistatic theory of atomic line wings. The transition probability linear with intensity has the following from near R_s[6]:

$$w = \left(\frac{dE}{\hbar(kg^2)^{1/5}}\right)^2 \left| \int_{-\infty}^{\infty} dy \ \exp i \ \{y^5/5 - 2y^3/3 \ (R_s - R_p)(g/k^2)^{-1/5} - (\omega - \omega_s) \ y(kg^2)^{-1/5} + (R_s - R_p)^2 y(g/k^2)^{-2/5}\} \right|^2 \quad (1)$$

where E is the laser field intensity, d is the dipole matrix element of electron transition, $2gk = U_2''(R_s) - U_1''(R_s)$ is rate of divergence of terms, $2mg = -U_{1,2}'(R_s)$ is interatomic interaction force at point R_s, and R_p is the precession point at the lower term, and m is the reduced mass. Equation (1) is a function of two parameters $X = (R_s - R_p)(g/k^2)^{-1/5}$ and $C = (\omega - \omega_s)(kg^2)^{-1/5}$. This condition is determinant in the explanation of inhomogeneities of the satellite spectral structure.

The principal part of the satellite is formed by collisions with thermal energy T. In this case, $R_s - R_p \approx T/mg$ and, since in the subject system $T/mg \gg (g/k^2)^{1/5}$, the parameter $X \gg 1$ for the principal portion of the set. In this region of the parameter X the asymptotic expression for transition probability

$$w(c, X) = 2^{2/3} \pi (kg^2)^{-2/5} (dE/\hbar)^2 X^{-2/3} \cos^2\left(\frac{8}{15} X^{5/2}\right) \Phi^2 \{C(4X)^{-1/3}\} \quad (2)$$

shows that the condition $C \approx X^{1/3}$ (here Φ is the Airy function) defines the satellite width $\Delta_s \approx X^{1/3}(kg^2)^{1/5}$, and the condition $w(C \approx X^{1/3}) \approx 1$ the minimum amplitude of a field which saturates the satellite $E_s \approx \hbar\Delta_s/d$.

The region of maximum values of w in Eq. (1) lies in the vicinity of $X = 0.96$ and $C = 1.08$. The "spectral" width of the maximum region $\Delta C \approx 1$ determines the dip width in a satellite $\Delta_h \approx (kg^2)^{1/5}$, and an expression for the maximum transition probability $w_m \approx \pi(dE/\hbar\Delta_h)^2$ permits us to evaluate ($w_m \approx 1$) the minimum field amplitude which forms the dip, $E_h \approx \hbar\Delta_h/d$. The relative depth of the dip is associated with the "coordinate" width of the maximum region $\Delta X = 1$.

At the following reasonable values of the system parameters $k = 10^{30}$ sec^{-1} cm^{-2}, $g = 10^{15}$ cm sec^{-2}, and $d = 3 \times 10^{-18}$ CGSE the quantities determined above correspond to $\Delta_s = 120$ cm^{-1}, $\Delta_h = 30$ cm^{-1}, and $E_h = 300$ CGSE ($I_h \approx 10^7$ W/cm^2), and, regardless of the crudeness of estimate, agree satisfactorily with the values of the corresponding measured quantities.

Further, past the plateau, satellite saturation (see Fig. 2) may also be described in terms of the linear quasi-classical theory. Determination of the dependence of w in Eq. (1) on the velocity v with which the atoms pass through the neighborhood of point R_s, yields $w \sim v^{-4/3}$ which appears to be more clearly defined than the Landau-Zener dependence $w \sim v^{-1}$ at small values of v. This condition explains qualitatively the more prominent manifestation of nonlinearity in the function $S(I)$ than under the Landau-Zener conditions.

In conclusion we should note that identification of the hidden structure in the quasistatic wing of the atomic line which is dependent on atomic collisions constitutes a direct proof of the inhomogeneity of its broadening. Unlike the Doppler broadening, this broadening is associated with inhomogeneity of a set of relative energies and impact parameters of colliding atoms. We should also point out that the occurrence of narrow dips in the nonlinear excitation (absorption) spectra is possible only in the situations when the slopes of terms in the vicinity of a point of classical transition are similar. Spectral characteristics of such a hidden structure in the linear spectrum are determined—as was shown above—by the fine parameters of interatomic interactions and may be used to determine the latter.

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