Manifestation of states of quasibound motion of atoms in the nonlinear excitation spectrum of resonant fluorescence

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The shape of the excitation spectra of atomic fluorescence in the system Rb + Xe was investigated experimentally. The observed change in the shape of the line wing with increasing excitation intensity is attributed to saturation of the states of the RbXe* quasimolecule. The investigated dependence of the intensity of the fluorescence on the Xe concentration has made it possible to conclude that the main channel that ensures the emission is the rotational predissociation of the excited RbXe* molecules produced in the radiation field. The recorded nonlinear dependence of the intensity of the emission of the atomic line on the excitation power is attributed to field quenching of the quasibound states of the RbXe* molecule.

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1. An investigation of the optical processes in a system of colliding atoms (quasimolecules) is of interest for problems of diagnostics of interatomic interactions, as well as for the study of the possiblity of directed action of radiation on atomic processes. In either case, special attention should be paid to processes in which the participating light quantum has a frequency in the region of the static wings of the colliding atoms. The light interacts effectively in this case only with deformed atoms whose transition frequency is substantially shifted during the time of the collision. Thus, the radiation, on the one hand, probes the atoms in the states of the strong interaction, and on the other, it becomes involved in a process in which an essential role is played by the character of the relative particle motion.

The use of lasers for the investigation of such processes is quite promising, since it facilitates the problem of their registration and uncovers a possibility of using nonlinear spectroscopy methods for their study.[1] Observation of nonlinear optical processes in the far wings of atomic lines requires, according to estimates, large radiation powers which in general are hardly attainable in experiment[1,2] because of the short time that the system stays at resonance with the exciting field.

At present there are three known experimental studies of nonlinear inelastic light-scattering processes in a system of colliding atoms.[2-4] In these studies, a nonlinear dependence of the emission S_a of atomic vapor (Rb, Cs, and Ca) at the transition frequency of the isolated atom (resonance fluorescence) was observed at relatively low intensity of the exciting radiation, $I \gtrsim 10^8$ W/cm^2. To explain the character of this dependence,[4,5] the hypothesis was advanced that the observed nonlinearity is due to saturation of the populations of the quasibound molecular states lying above the dissociation energy of the upper binding term. Although the quasibound motion of the molecules (rotational dissociation) is well known[6] and there is no doubt that saturation of such states is possible in principle, the question of the existence of a contribution of the proposed process to the formation of the nonlinearity observed in Refs. 2–4 remains open.

The purpose of the present investigation is twofold. First, an experimental check on the validity of the assumption made in Refs. 4 and 5 that the dominant contribution to the recorded nonlinearity is made by deactivation of rotational predissociation states. Second, to demonstrate the effectiveness of the saturation spectroscopy method for the observation of such close-collision motions whose registration by methods of linear spectroscopy is difficult.

2. The object of the investigation was a system of colliding Rb and Xe atoms. The choice of this system was dictated by a number of factors. First, by the presence of detailed information on this system[7] (its level scheme is shown in Fig. 1). Second, by the proximity to the simple model systems considered in Refs. 4 and 5 (without the binding term in the electronic ground state and in the presence of such a term in the excited state). Finally, by the possibility of its excitation by a real source of tunable radiation, which in the experiment was a organic dye laser (pulse length 20 nsec, lasing wavelength $\lambda_{exc} = 820$–890 nm, spectrum width 5–10 cm^{-1}). The emission of the Rb+Xe mixture was registered at a right angle to the direction of the exciting beam through an MDR-2 monochromator. The pressure of the Rb and Xe vapor in the cell as 1–3 mTorr and 10–30 Torr, respectively.

3. Before we report the experimental results, we consider (Fig. 2) the excitation of the system by radiation whose frequency ω is less than the frequency ω_0 of the atomic transition of the Rb atom. In accordance with the quasiclassical premises (the Franck-Condon principle) the excitation takes place near the resonance $\hbar\omega = U_2(R_\omega) - U_1(R_\omega)$, which is realized when the Rb and Xe atoms collide at a relative distance R_ω between them. In the case when the relative energy of the collision is $\mathscr{E} < \hbar(\omega_0 - \omega)$, the absorption of the quantum leads to formation of an excited molecule (RbXe)*. In the opposite case ($\mathscr{E} > \hbar(\omega_0 - \omega)$) it causes the atoms to move apart, Rb being in the excited $5P$ state and Xe in the ground state. The first process leads to the appearance of the registered spontaneous radiation in the region of the molecular band, and the second leads to atomic emission at the frequency ω_0. An essential feature of the investigated system is the monotonic dependence of R_ω on ω: Smaller values of ω correspond to smaller R_ω. This is the cause of a circumstance of

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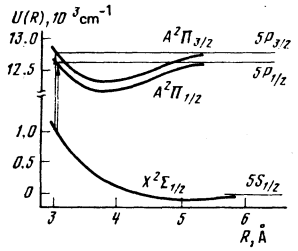

FIG. 1. Working terms of the system Rb+Xe. Within the limits of the accuracy of Gallagher's data[7] the spin-orbit splitting does not depend on R. The lengths of the arrows determine the frequencies ω_c.

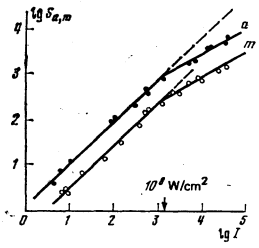

FIG. 3. Dependence of the emission intensity $S_{a,m}$ on the excitation intensity I (λ_{exc}=820 nm): a—of an atomic doublet, m—of the molecular band (λ_{rec}=840 nm).

importance for the further investigation, namely that laser radiation with $\omega<\omega_c$ (see Fig. 2) excites only states in which the atoms move apart, and cannot excite molecular states.

4. We measured in the experiment the intensities of the molecular (S_m) and atomic (S_a) emissions as functions of the laser excitation intensity I. The measurement results are shown in Fig. 3. We discuss first the character of the molecular emission.

The plot of $S_m(I)$ deviates substantially from a straight line at $I>I_m\approx 10^8$ W/cm². The nonlinearity in this process, just as the one observed in Ref. 4, can be attributed to optical quenching (deactivation) of the excited molecule (RbXe)* by the powerful radiation field in the vicinity of the point R_ω. We note that the characteristic power I_m is much less than that necessary for strong saturating excitation of the atoms per collision.[4] The difference between the probabilities of the excitation and deexcitation of the molecule by laser radiation is connected with the difference between the time that the system is at resonance with the field in the ground diverging state of motion (10^{-13} sec) and the bound excited state (10^{-8} sec).

The plot $S_a(I)$ of the atomic emission (Fig. 3) is similar to $S_m(I)$ of the molecular emission. The characteristic value $I_a\approx 10^8$ W/cm² for the atomic emission is close to I_m for the molecular emission and, while somewhat larger than the latter, is still surprisingly low if it is recognized that in excitation of the atomic emission in both states—the lower and now also the upper—the particles move apart (separation states), and the time

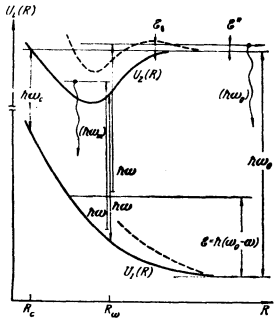

FIG. 2. Excitation scheme of molecular and atomic emission. The dashed curves show the effective (radial) potentials for $l\gg 1$. Transitions with $\omega<\omega_c$ occur in the region of $R<R_c$; the ordinates represent the quantity $U_i(R)=U_{1,2}(R)+\hbar^2 l(l+1)/2\mu R^2$.

that the quasimolecule is at resonance with the field is too short for the appearance of a noticeable optical quenching of the fluorescence.

5. The nonlinearity observed in $S_a(I)$ can be attributed to the fact[4,5] that the Rb atoms optically excited into separation states can become temporarily bound with Xe atoms. The quasibound character of these atomic states in the presence of attraction between the atoms is ensured by the rotational barrier of the relative-motion potential. The excited quasibound Rb atoms contribute to the atomic emission only after tunneling decay of the quasimolecule (RbXe)*. The de-excitation of the latter in induced transitions (in the vicinity of R_ω) decreases the intensity of the atomic emission.

We note that laser radiation can be used to excite the colliding atoms Rb and Xe both in states of quasibound motion with energy $0<\mathscr{E}<\mathscr{E}_b$ (see Fig. 2) and into states of rapid separation with $\mathscr{E}>\mathscr{E}_b$. Contributions to the atomic emission is made by both processes, but the recorded nonlinearity can be attributed only to the contribution of the former, since the appreciable laser de-excitation at relatively low intensity is possible only for a quasibound pair (during the time of its tunneling decay).

From the explanation proposed for the nonlinearity it follows that the latter cannot be observed even at $I>I_a$ if the frequency of the exciting radiation is $\omega<\omega_c$. Indeed, light with $\omega<\omega_c$ cannot excite quasibound-motion states and excites only the rapid separation states with $\mathscr{E}>\mathscr{E}_b$, inasmuch as at any instant $\hbar l$ of the colliding atoms the minimum separation energy $\mathscr{E}_{min}(l)=U_2(R_\omega)+\hbar^2 l(l+1)/2\mu R_\omega^2$ is always larger than the height of the barrier: $\mathscr{E}_b(l)=U_2(R_b)+\hbar^2 l(l+1)/2\mu R_b^2$ (R_b is the coordinate of the position of the maximum centrifugal potential—the barrier). This follows from the already noted singularity of the system $U_2(R_\omega)>0$, $R_\omega<R_b$ and from

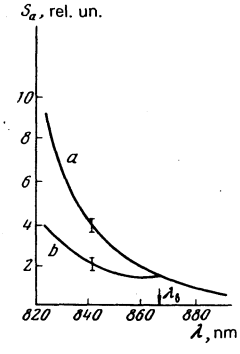

FIG. 4. Long-wave excitation spectra of atomic emission at $I=10^6$ (a) and $I=10^9$ (b) W/cm².

the obvious $U_2(R_b) < 0$.

For an experimental check on the described interpretation of the nonlinearity we investigated the excitation spectrum of the atomic fluorescence at low ($I \ll I_a$) and high ($I > I_a$) excitation powers. In the experiment, Rb + Xe pairs were excited by focused laser radiation whose power was decreased by neutral light filters. The obtained spectra are shown in Fig. 4. It is seen that in excitation with $\lambda_{exc} > 867$ nm both spectra agree within the limits of the measurement accuracy.[1)] The difference between the linear and saturated spectra manifests itself clearly in the region $\lambda_{exc} < \lambda_c$.

Within the limits of the accuracy of the available data the experimental value of λ_{exc} agrees well with the cutoff wavelengths $\lambda_c(\Pi_{1/2})$ and $\lambda_c(\Pi_{3/2})$ calculated from the known terms (see Fig. 1). The observed large value $\lambda_c(\Pi_{1/2}) = 865$ nm is very close to λ_{exc}, and the smaller one $\lambda_c(\Pi_{3/2}) = 855$ nm is located in the deformed region of the spectrum and therefore does not mainfest itself as a singularity.

7. The results presented within the framework of the developed concepts offer unequivocal evidence in favor of the proposed explanation of the nonlinearity mechanism. However, for the concretely investigated system Rb + Xe the same results can be interpreted also in a different manner. In fact, a contribution to the atomic emission in the case of short-wave $\lambda_{exc} < \lambda_c$ excitation can be made also by dissociation of the (RbXe)* molecules when they collide with the xenon atoms:

$$Rb^*Xe + Xe \rightarrow Rb^* + 2Xe \rightarrow Rb + \hbar\omega_0 + 2Xe.$$

If this mechanism is decisive in the formation of the atomic emission, then the nonlinearity of $S_a(I)$ and the deformation of the short-wave part of the spectrum are attributed to the aforementioned de-excitation of the molecular states of (RbXe)*.

In principle, the two considered mechanisms (both tunnel and impact) can jointly cause the observed nonlinear phenomena. However, the contribution of each of them to the intensity of the atomic emission depends differently on the xenon concentration. The efficiency of the tunnel mechanism is a linear function of the concentration, while that of the impact mechanism is a quadratic function in the case of excitation in the region $\lambda_{exc} < \lambda_c$ and a linear one in the region $\lambda_{exc} > \lambda_c$. This concentration difference between the mechanism makes it possible to establish the role of each of them in the observed nonlinearities.

The dependence of the linear excitation spectrum of the atomic emission on the pressure of the xenon was investigated with a specially produced installation that includes a high-power incandescent lamp and two MDR-2 monochromators for the monochromatization of the exciting and recording radiations. The obtained spectra are shown in Fig. 5 (the resolution is 6 nm). It is seen that the spectrum deformation due to the nonlinear dependence of the intensity of the emission on the Xe concentration is significant only at high (>30 Torr) pressures,[2] and the spectra obtained at 10 and 30 Torr

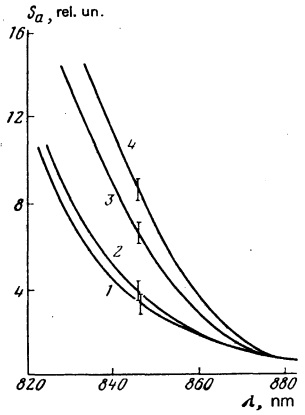

FIG. 5. Linear excitation spectra of atomic emission at different Xe concentrations. Curves 1, 2, 3, and 4 correspond to 10, 30, 100, and 300 Torr, respectively.

agree within the limits of the measurement accuracy. This is evidence that the contribution of the collisional dissociation to the observed nonlinearities is negligible at low pressures (10–30 Torr).

From the presented experimental results we can draw the following conclusions. A large fraction of the Rb and Xe atoms excited by the collision are in a state of quasibound motion. The dissociation of the excited quasimolecule (RbXe)* proceeds via tunneling of the atom through the centrifugal barrier. The nonlinearity of $S_a(I)$ at high excitation powers (and low pressures) is due to the field-induced de-excitation of the predissociated molecules. The characteristic value of I_a makes it possible to estimate[5] the average lifetime of the predissociating molecule: $\tau_{tun} = 10^{-9} - 10^{-10}$ sec. The conclusions are obviously valid in general for all systems with an attracting upper term. This gives grounds for assuming that the optical excitation of the quasibound motions in a system of colliding atoms and their recording by methods of nonlinear spectroscopy will find use in various branches of laser physics and chemistry, in view of the timeliness of research on excimer and quasimolecule system.

[1)] The quantity that normalized the spectra was the intensity of the atomic emission excited at $\lambda_{exc} = 890$ nm. The linear $S_a(I_{\lambda=890})$ dependence was verified specially in the entire range of measurement of I.

[2)] The spectra were normalized to the intensity of the emission excited at $\lambda_{exc} = 879$ nm $> \lambda_c$. We recall that emission with $\lambda_{exc} > \lambda_c$ does not produce excited molecules, although molecular emission does take place. Therefore the intensity of the atomic emission in this region depends linearly on the Xe concentration also under conditions when the impact mechanism predominates.

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Quantum theory of stimulated processes in a free-electron laser in a strong field

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General quantum-mechanical equations are derived for an electron in a spatially periodic magnetic field and in the field of an electromagnetic wave. The limits of applicability of the calculations in the lowest order of perturbation theory are obtained. It is shown that for gain calculations these limits are much wider than in the general case owing to the substantial cancellation of the higher-order corrections that contribute to the gain. The saturation parameter μ is determined. The asymptotic dependence of μ on the gain is obtained at $\mu > 1$. The spectral properties of the gain, namely the width and the shift of the resonant maximum, which depend on the field intensities, are investigated. An analytic expression is obtained, at $\mu > 1$, for the maximum (in the spectrum) gain, which decreases in proportion to $E_0^{-3/2}$ with increasing intensity E_0 of the amplified wave.

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1. INTRODUCTION

Amplification[1] and generation[2] of radiation produced when a beam of relativistic electrons is scattered by a spatially-periodic potential of a time-independent magnetic field were recently observed experimentally, for the first time ever, at Stanford University. The theory of the process was developed in a large number of papers[3-15] both on the basis of a classical description[5-11] and a quantum one.[3,4,12-15] At the same time, and independently, a theoretical and experimental investigation was made of spontaneous radiation in systems of this type–undulator (Wiggler) radiation.[16-18]

One of the principal results of Refs. 3–15 is the derivation of formulas for the gain of a weak trial wave (in the "weak signal" approximation). As shown by us earlier[15] these calculations are equivalent to a direct quantum-electrodynamic calculation in the lowest order of perturbation theory (in first order in the magnetic field and first order in the field of the electromagnetic wave). Saturation effects were considered in a number of papers by way of qualitative estimates[6,9,12] and also on the basis of a numerical solution of simplified equations in a classical model.[10,11] No analytic solutions have been apparently published to date. The present paper is devoted to a theoretical description of the amplification of an intense external wave when electrons are scattered by a spatially periodic strong magnetic field. We shall use one of the simplest variants of the quantum-mechanical description of the electron motion in the classical fields, proposed in our earlier papers[15] and based on the interpretation of these phenomena in terms of stimulated bremsstrahlung and absorption. On the basis of the analysis of the exact equations we find the conditions for applicability of the calculations in the lowest order of perturbation theory. In the general case these conditions are much more stringent than in the calculation of the gain. We shall obtain the solutions for the equations in different ranges of variation of the field intensities, including the asymptotic solution for a strong field and a description of the saturation effect.

To estimate various parameters we shall frequently use below the data of Ref. 2. We shall therefore assume that the following relations hold,

$$\varepsilon \gg m \gg \omega \gg q_0,$$

where ε and m are respectively the energy and mass of the electron, $q_0 = 2\pi/\lambda_0$, λ_0 is the period of the magnetic field, and ω is the frequency of the amplified wave; we use a system of units in which $\hbar = c = 1$. Just as in all the preceding papers[1-14] (with the exception of Ref. 12), we use the approximation of the given field of the electromagnetic wave, assuming its amplitude to be constant; this is justified by the *a posteriori* smallness of the gain per pass.

2. FORMULATION OF PROBLEM. PRINCIPAL EQUATIONS

We consider the initial problem, assuming that the interaction of the electron with the magnetic field is turned on at a certain instant of time (the instant when it enters the region where the magnetic field exists), and lasts for a limited time $t = L/v \approx L$, where v is the electron velocity and L is the length of the magnet. A direct check shows that the equations obtained in this

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