

# Optical excitation of colliding atoms to a state of quasibound motion

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A theoretical analysis is given of the excitation of colliding atoms by strong optical radiation whose frequency is appreciably different from the resonance transition frequency of the isolated atom. The process is described within the framework of the adiabatic approximation of the theory of inelastic atomic collisions. The excitation probability is obtained for the atom as a function of the intensity of the exciting radiation. It is shown that the nonlinear character of this dependence is due to the deexcitation of the excited quasimolecule produced in the radiation field. The results are used to explain the experimentally established dependence of the intensity of atomic fluorescence of alkali metal vapors on the excitation intensity. The specific features of the proposed nonlinearity mechanism are reflected at moderate levels of excitation in the nature of this function which differs from the Landau-Zener-type dependence usually discussed in the literature.

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1. The recent interest in optical phenomena in systems of colliding atoms (quasimolecules) has arisen for at least two reasons. Firstly, optical detection of collisions can be used to investigate the interatomic interaction potential<sup>[1]</sup> and, secondly, radiation is an effective means of influencing the collision process and, in particular, the relative motion of the atoms.<sup>[2,3]</sup>

Until quite recently, only those quasimolecular processes that were linear in the intensity were investigated. The advent of high-density sources of optical radiation has meant that nonlinear optical phenomena in quasimolecules have become amenable to investigation. Recent theoretical<sup>[4–6]</sup> and experimental<sup>[2–7]</sup> work has been concerned with nonlinear phenomena at field intensities such that the probability of absorption or emission by the quasimolecule of a quantum of radiation during the collision time was close to unity. Constructive theoretical results have been obtained<sup>[8]</sup> in the approximation of a given relative motion of colliding atoms, independent of the radiation field. It will be shown below that the inclusion of the influence of the field on the motion of the atoms is essential for the correct description of many nonlinear processes, even when the given-motion approximation is justified for the description of the corresponding linear process. This shows that studies of nonlinear phenomena in quasimolecules may become useful in the diagnostics of atomic collisions.

2. The research reported here was stimulated by the experiments of Bonch-Bruevich *et al.*<sup>[2,3]</sup> and was concerned with the interpretation of the dependence of the emission of rubidium and cesium vapor at the atomic transition frequency  $\omega_0$  on the intensity of the exciting radiation whose frequency  $\omega$  was not in resonance with  $\omega_0$  and lay in the static wing of the atomic line. It was shown that the observed inelastic scattering of light was due to binary atomic collisions. Under the conditions of the above experiments, the resonance defect  $\hbar(\omega_0 - \omega)$  was appreciably greater than the thermal energy of the colliding atoms and, therefore, there was

a substantial reduction in the velocity of their relative motion during this process.

The atomic fluorescence intensity  $S$  was found to increase monotonically with increasing intensity  $I$  of the exciting radiation, and became a nonlinear function of the latter for  $I > I_s \approx 4 \times 10^9 \text{ W/cm}^2$ . The nature of this nonlinearity, which was observed for the first time, was explained by the saturation of translational electronic transitions in the quasimolecule in the neighborhood of the “rotation-crossing” point.

The proposed explanation produced a value of  $I_s$  that was in agreement with experiment, but the overall form of the function remained unexplained. The stationary theory of transitions in the neighborhood of rotation-crossing points<sup>[8]</sup> shows that the transition probability decreases exponentially with increasing  $I$  for  $I > I_s$ . The inhomogeneous character of the excitation associated with different directions of the axis of the quasimolecule relative to the electric field vector in the incident radiation<sup>[9]</sup> does not, in this case, explain the transformation of the decreasing dependence into an increasing one, and predicts only a change from an exponential variation to the power law function  $S \sim I^{-1/2}$  for  $I \gg I_s$ .

In view of the particular features of the experiment (the atomic fluorescence was excited by a series of short, picosecond pulses<sup>[2]</sup>), it was suggested that the nonstationary character of the excitation process might have had a correcting influence on the above function. However, although subsequent investigations, performed with longer exciting pulses (approximately  $10^{-8} \text{ sec}$ ), resulted in the smaller value  $I_s \approx 5 \times 10^7 \text{ W/cm}^2$ , the form of the function  $S(I)$  was exactly the same, so that the previously proposed explanations turned out to be inconsistent.

In this paper, we shall give a different explanation of the origin of the above nonlinearity, which will be free from the defects of the explanation given previously. The main cause of the nonlinearity can be found in the

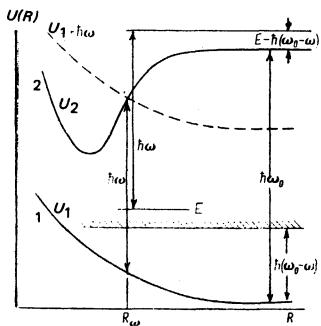


FIG. 1. Optical nonresonant excitation of atomic fluorescence in a system of colliding atoms.

existence of quasistationary states of the excited quasimolecule formed in the radiation field.

In what follows, we shall examine the optical excitation of quasibound motions of atoms, and will consider the importance of these processes in the explanation of the above function.

3. Consider free-free transitions<sup>1)</sup> within the framework of the Born-Oppenheimer approximation (Fig. 1) for a system of colliding atoms in the presence of a field  $F \cos \omega t$ , between the continuum of states 1 (unexcited atoms) and the continuum of excited states 2, subject to the conditions  $\omega < \omega_0$  and  $\omega_0 - \omega \ll \omega_0$ . The latter condition enables us to confine our attention to almost-resonant processes and to use the rotating-field approximation in the description of the transition dynamics. Within the framework of this approximation, a simple redefinition of the position of the potential for the lower state (Fig. 1) enables us to reduce the problem of transitions in an alternating field to a problem in the stationary theory of inelastic atomic collisions, with term-coupling constant depending on the field amplitude  $F$ . The resonance points  $R_\omega$ , which are the solutions of the equation

$$U_1(R_\omega) + \hbar\omega = U_2(R_\omega), \quad (1)$$

have the same role in this process as the term-crossing points in the theory of nonadiabatic collisions.<sup>[10]</sup>

The emission at the atomic transition frequency  $\omega_0$  can contain contributions due to unexcited atoms colliding with relative energy  $E > \hbar(\omega_0 - \omega)$  after the quasimolecule consisting of the atoms, and excited in the neighborhood of  $R_\omega$ , dissociates into an excited and an unexcited atom. The dynamics of these atoms is determined by the shape of the potential  $U_{12}(R)$  for radial motion:

$$U_{12}(R) = U_2(R) + \hbar^2 l(l+1)/2\mu R^2. \quad (2)$$

where  $\hbar l$  is the orbital angular momentum of the resulting atoms and  $\mu$  is their reduced mass.

The potential  $U_{12}(R)$  corresponding to the excited bound state has a centrifugal barrier which "traps" the atom in the quasimolecule with energy  $0 < E < E_B$  (Fig. 2) in the region of the resonance point  $R_\omega$ , and thus ensures that the field exerts a prolonged influence on the quasimolecule.

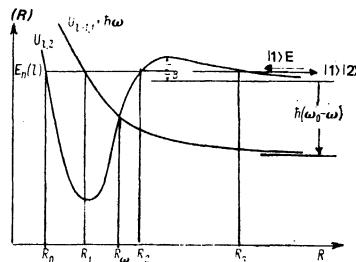


FIG. 2. Formation of long-lived quasimolecules. The atoms collide in the unexcited states  $|1\rangle$  and depart in states  $|1\rangle$  and  $|2\rangle$ .

4. The problem of optical transitions between states 1 and 2 will now be formulated in the language of the stationary theory of multichannel scattering.<sup>[11]</sup> The dynamics of both relative motion and electronic excitation of the atoms in the radiation field is described by the following set of coupled equations within the framework of the resonance approximation:

$$\begin{cases} (E - H_1 - \hbar\omega) \psi_1 = 2V\psi_2, \\ (E - H_2) \psi_2 = 2V\psi_1, \end{cases} \quad (3)$$

where  $\psi_{1,2}(R)$  are the wave functions for the relative motion of the atoms occupying electronic states 1 and 2, respectively. The Hamiltonian  $H_{1,2}$  has the form

$$H_{1,2} = -\frac{\hbar^2}{2\mu} \Delta_R + U_{1,2}(R), \quad (4)$$

where  $V$  is the operator corresponding to the interaction between the quasimolecule and the field and, in this particular scheme, has the form

$$4V = -D(R)F = -D(R)F \cos \theta, \quad (5)$$

where  $D(R)$  is the dipole moment associated with the 1-2 electronic transition.

Henceforth, we shall confine our attention to the case of large detuning from resonance:  $\hbar(\omega_0 - \omega) \gg D(R)F$  and will suppose that the main contribution to the excitation of the quasimolecule is provided by the neighborhood of the point  $R_\omega$ . Since the 1-2 transition is assumed to be optically allowed, the dependence of  $D$  on the separation between the nuclei can be neglected in the zero-order approximation, and we shall henceforth consider that  $D(R) = D(R_\omega) = D$ .

Expanding the wave functions  $\psi_{1,2}(R)$  in terms of the spherical harmonics  $Y_{lm}$ :

$$\psi_{1,2}(R) = \sum_{l,m} i^l R^{-1} \chi_{1,2}^{l,m}(R) Y_{lm}(\theta, \varphi), \quad (6)$$

and substituting this in (3), we obtain the following infinite set of equations for the amplitudes  $X_{1,2}^{l,m}$ :

$$\begin{aligned} (E - H_1 - \hbar\omega) \chi_1^{l,m} &= V_{lm} (\chi_2^{l-1,m} + \chi_2^{l+1,m}), \\ (E - H_2) \chi_2^{l,m} &= V_{lm} (\chi_1^{l-1,m} + \chi_1^{l+1,m}) \end{aligned} \quad (7)$$

where

$$H_{l,z} = -\frac{\hbar^2}{2\mu} \frac{d^2}{dR^2} + U_{l_0,2}(R) \quad (8)$$

and

$$2V_{l,m} = -DF[(l^2 - m^2)/(2l-1)(2l+1)]^{1/2}. \quad (9)$$

We shall now assume that the main contribution to the excitation of fluorescence is provided by atomic collisions other than head-on collisions, so that  $l \gg 1$ . In the classical limit, the angular momentum component is  $m = l \sin \theta$ , where  $\theta$  is the angle between D and F, because the angular momentum vector is perpendicular to the line joining the nuclei, i.e. to D/D. Thus, in the classical limit,  $V_{l,m} = V$  [see Eq. (5)].

Next, we consider the situation where, owing to the presence of the centrifugal barrier, the system has quasistationary levels in the state corresponding to the upper term. The position  $E_n$  and width  $\Gamma_n$  of these levels depend on  $l$ . It is well known<sup>[11]</sup> that the probability of inelastic scattering is a function of the energy  $E$  of the colliding particles and exhibits a resonance in the neighborhood of  $E \approx E_n$ . In our case, the position and width of these resonances are functions of field intensity.

5. The scattering problem will now be formulated as follows: Before the field is introduced, the atoms are in the eigenstate  $\Psi_E$  corresponding to the lower term with angular momentum  $l_0 - 1$  and energy  $E$  (the normalization is chosen so that the current density in each of the two traveling waves producing the standing wave  $\Psi_E$  is equal to unity). When the field is turned on, reflected waves appear in all channels while the incident current is present only in the state corresponding to the lower term and, as before, is equal to unity.

When  $E$  is close to the resonance energy  $E_0(l_0)$ , and the resonances corresponding to different  $l$  are well resolved (do not overlap), the most probable transitions are those from the initial state to the state with  $l = l_0$ . Transitions to other states with  $l \neq l_0$  are of lower probability, especially for large departures from resonance.<sup>2)</sup> The set of equations given by (7) can therefore be simplified by neglecting the "nonresonant" terms in them. The result is

$$\begin{aligned} (E - H_{l_0-1,1} - \hbar\omega) \chi_1^{l_0-1,m} &= V \chi_2^{l_0,m}, \\ (E - H_{l_0+1,1} - \hbar\omega) \chi_1^{l_0+1,m} &= V \chi_2^{l_0,m}, \\ (E - H_{l_0,2}) \chi_2^{l_0,m} &= V(\chi_1^{l_0+1,m} + \chi_1^{l_0-1,m}). \end{aligned} \quad (10)$$

In accordance with the above formulation of the problem, we write the formal solution of (10) for  $\chi = \chi_2^{l_0,m}$  in the form

$$\chi(R) = V^2 G_0 (\hat{G}_+ + \hat{G}_-) \chi + V G_0 \Psi_E, \quad (11)$$

where  $\hat{G}_0$  and  $\hat{G}_{\pm}$  are the operators  $(E - H_{l_0,2} + is)^{-1}$  and  $(E - H_{l_0 \pm 1,1} - \hbar\omega + is)^{-1}$  (with  $s \rightarrow 0$ ), whose spectral distribution has the asymptotic behavior of a diverging wave:

$$G_0(R, R') = \sum_{\delta} \frac{\varphi_{\delta}(R)\varphi_{\delta}(R')}{E - \delta + is}, \quad s \rightarrow 0, \quad (12)$$

where  $\varphi_{\delta}(R)$  are the eigenfunctions (standing waves) for the term corresponding to the potential  $U_{l_0,2}(R)$ , normalized to the  $\delta$ -function of the energy in the continuous part of the spectrum. The symbol  $\sum_{\delta}$  represents summation over the discrete, and integration over the continuous, spectra.

Substituting

$$\chi(R) = \sum_{\delta} c_{\delta} \varphi_{\delta}(R) \quad (13)$$

in (11) and using the spectral representation of Green's function (12), we obtain the following integral equation for the coefficient  $c_{\delta}$ :

$$c_{\delta} = (E - \delta + is)^{-1} \left\{ V^2 \sum_{\delta'} c_{\delta'} (\varphi_{\delta}, [\hat{G}_+ + \hat{G}_-] \varphi_{\delta'}) + V(\varphi_{\delta}, \Psi_E) \right\}. \quad (14)$$

This equation can be simplified by recalling that the wave functions corresponding to the continuous spectrum with  $\delta \approx E_0$  can be approximately factorized in the inner region  $[R_0, R_2]$ <sup>[12]</sup>

$$\varphi_{\delta}(R) = g(\delta) \Phi(R), \quad (15)$$

where  $\Phi(R)$  is the wave function for the bound state with energy  $E_0$ , normalized to unity, which appears if the barrier is assumed to be impenetrable and

$$g^2(\delta) = \frac{1}{2\pi} \frac{\Gamma_0}{(\delta - E_0)^2 + \Gamma_0^2/4} \quad (16)$$

where  $\hbar = 1$  (we shall use this system of units henceforth).

Using the factorized wave functions (15) to evaluate the integrals in (14), which is justified since the neighborhood of  $R_0$ , which lies within the interval  $[R_0, R_2]$ , provides the dominant contribution, we obtain

$$c_{\delta} = g(\delta) (E - \delta + is)^{-1} \left\{ W \sum_{\delta'} c_{\delta'} g(\delta') + V(\Phi, \Psi_E) \right\}, \quad (17)$$

where

$$W = V^2 (\Phi, [\hat{G}_+ + \hat{G}_-] \Phi). \quad (18)$$

Equation (17) can be solved by multiplying both sides by  $g(\delta)$ , then summing over  $\delta$  and, finally, evaluating the sum

$$\sum_{\delta} c_{\delta} g(\delta)$$

and substituting it into the right-hand side of (17). The resulting expression for  $c_{\delta}$  enables us to determine  $X(R)$  from the expansion given by (13):

$$\chi(R) = V(1 - QW)^{-1} \int dR' G_0(R, R') \Psi_E(R'), \quad (19)$$

where

$$Q = \sum_s \frac{g^2(\mathcal{E})}{E - \mathcal{E} + is} = \frac{1}{E - E_0 + i\Gamma_0/2}. \quad (20)$$

The asymptotic expression for  $\chi(R \rightarrow \infty)$  determines the density of the current of excited atoms which, for the above normalization, is dimensionless and equal to the transition probability  $w$ . The asymptotic form of (19) can readily be determined with the aid of Green's function in the form

$$G_0(R, R') = -\left(\frac{2\pi}{v_2}\right)^{1/2} \exp\left\{i\left(k_2 R - \frac{\pi l_0}{2} + \delta_E\right)\right\} \varphi_E(R') \quad (21)$$

and the expression given by (15) which determines  $\varphi_E(R)$  in the region of the main contribution to the integral (19). In these expressions,  $v_2 = k_2/\mu = [(E - U_{l_02}(\infty))/2\mu]^{1/2}$  is the velocity of the outgoing atoms and  $\delta_E$  is the elastic scattering phase for the potential  $U_{l_02}(R)$ . Evaluation of the current with the aid of (19), (21), and (15) yields

$$w = \frac{2\pi V^2 g^2(E)}{|1-QW|^2} |\langle \Phi, \Psi_E \rangle|^2. \quad (22)$$

6. Let us now examine the expression given by (22) in the quasiclassical approximation. In the region in which we have a linear dependence on the field intensity, the transition probability is given by

$$w_0 = \frac{1}{2\pi} \frac{\Omega_0 \Gamma_0}{(E - E_0)^2 + \Gamma_0^2/4} p_-, \quad (23)$$

where  $\Omega_0$  is the vibrational quantum (frequency of oscillations in a well with energy  $E_0$ ) and

$$p_- = \frac{8\pi V^2}{v_u \Delta U_u} \cos^2\left(S_- + \frac{\pi}{4}\right) \quad (24)$$

is the probability of the inelastic process under the conditions of term crossing,<sup>[10]</sup> which is linear in intensity,  $v_u = [(E - U_{l_02}(R_\omega))/2\mu]^{1/2}$  is the velocity at the term-crossing point (resonance point),  $\Delta U' \equiv U'_{l_02}(R_\omega) - U'_{l_0-1,1}(R_\omega) \approx U'_2(R_\omega) - U'_1(R_\omega)$  is the rate at which the terms diverge at  $R_\omega$ , and  $S_-(E)$  is the difference between the classical action evaluated for the potential  $U_{l_02}(R)$  in the region  $[R_0, R_\omega]$  and the action for the potential  $U_{l_0-1,1}(R)$  in the interval  $[R_1, R_\omega]$  (see Fig. 2).

When  $E = E_0$ , the probability  $w_0$  reaches its maximum value  $2\Omega_0 p_- / \pi \Gamma_0$ , which, in the case of narrow resonance  $\Omega_0 \gg \Gamma_0$ , is much greater than the linear Landau-Zener transition probability  $p_-$  evaluated without taking the barrier into account. When  $w_0 > 1$ , the linear approximation is not valid even if  $p_- \ll 1$ , and it is essential to take into account the transition saturation effect.

To determine the form of  $w$  in the nonlinear region, we use the method of stationary phase and the quasiclassical representation of Green's function in the form

$$G_+(R, R') = -\frac{2i}{v_1} \exp\left\{i\left(\int_{R_1}^{R_2} k_+(R) dR - \frac{\pi}{4}\right)\right\} \cos\left\{\int_{R_1}^{R_2} k_+(R) dR - \frac{\pi}{4}\right\}, \quad (25)$$

where  $k_+(R) = [2\mu(E - U_{l_0+1,1}(R) - \hbar\omega)]^{1/2}$ ,  $v_1 = k_+(\infty)/\mu$ ,

and  $R^\pm$  are the turning points of the potentials  $U_{l_0+1,1}(R)$ . We shall assume that  $R$  and  $R'$  are greater than  $R^\pm$ , and will use  $R_>$  to represent, as usual, the greater and smaller of the values  $R$  and  $R'$ . We shall write the result in the form

$$W = \Delta - i \frac{\Omega_0}{4\pi} p_-, \quad (26)$$

where  $\Delta$  is a real quantity proportional to  $V^2$ ,  $p_+ = p_- + p_*$ , and  $p_*$  is analogous to  $p_-$  but is associated with the potential  $U_{l_0+1,1}(R)$ .

Substituting (26) in (22), and using (20) and (23), we obtain the following expression:

$$w = \frac{1}{2\pi} \frac{\Omega_0 \Gamma_0 p_-}{(E - E_0 - \Delta)^2 + (\Gamma_0/2 + \Omega_0 p_*/4\pi)^2}, \quad (27)$$

from which it is clear that  $\Delta$  determines the intensity-dependent shift of the quasistationary level and  $\Omega_0 p_-$  determines its field width. As in the linear case, the transition probability is a resonant function of the energy of the colliding atoms, since  $\Delta(E)$  and  $p(E)$  are slowly-varying functions on the scale of the resonance width.

We must now examine the limits of validity of (27), which was obtained by solving the truncated set of equations (10) in which the nonresonant amplitudes  $\chi_2^{l_0+2,m}$  were neglected. In the first order in the field, the equation for the larger of these,  $\chi_2^{l_0-2,m}$  is

$$(E - H_{l_0-2,2}) \chi_2^{l_0-2,m} = V \chi_1^{l_0-1,m}, \quad (28)$$

which is analogous to the equation for the resonant amplitude  $\chi_2^{l_0,m}$  in the approximation linear in the field. The probability of the  $\chi_1^{l_0-1,m} \rightarrow \chi_2^{l_0-2,m}$  transition is, therefore, given by (23), in which the energy difference in the denominator is close to the difference between the energies of the neighboring levels,  $E_n(l_0)$  and  $E_n(l_0 - 2)$ , i.e., the energy of the rotational quantum  $\epsilon_{rot}(l_0)$ . Therefore, the nonresonant amplitudes can be neglected, provided the field-broadened resonances can be well resolved:

$$\epsilon_{rot}(l_0) \approx \frac{\hbar l_0}{\mu R_u^2} \gg \frac{\Gamma_0}{2} + \frac{\Omega_0}{4\pi} p_-, \quad (29)$$

and this restricts the allowed power levels. Of course, the field should not, under these conditions, produce an appreciable change in the energy structure of the system ( $p \ll 1$ ).

It is important to note that, if the term-crossing point (Fig. 2) can lie above the dissociation energy of state 2 for large  $l$ , and, at the same time, remain in the region of the quasibound motion, then it can also be a turning point at the same time. The quantity  $p_-$  in (23) is then given not by (24) but by the expression given in<sup>[10]</sup>.

Next, consider the expression given by (27). It is noticeable that  $w$  is a nonmonotonic function of the radiation intensity  $I \sim V^2$ : at low intensities,  $w \sim I$ , whereas, at high intensities, when the field width exceeds the

detuning  $E - E_0$ , the probability varies as  $\sim I^{-1}$ . This somewhat unexpected result can easily be explained in the language of quasistationary states. In particular,  $w$  is the probability of finding the excited atom well away from the region of quasibound motion [ $R_0, R_2$ ], and its values determine the competition between field deexcitation ("downward" transition) and tunneling dissociation of the quasimolecule. During the time  $\tau(E)$  necessary for the tunneling dissociation,<sup>[12]</sup> the radiation frequently ( $\approx \tau(E)\Omega$  times) stimulates the deexcitation of the quasimolecule as the oscillating atom traverses the neighborhood of the point  $R_\omega$ . As the intensity increases, the probability of deexcitation begins to predominate over the probability of tunneling dissociation because the former increases with increasing  $I$ . The probability of detecting a free excited atom will, therefore, fall, beginning with a certain  $I(E)$ .

7. We now use (27) to interpret the function  $S(I)$ . Since the incident current is noncoherent both in  $l$  and  $E$ , we shall calculate the current  $J_l$  of excited atoms flying apart with angular momentum  $l$ :

$$J_l = \int \{w_{l-1}(E)j_{l-1}(E) + w_{l+1}(E)j_{l+1}(E)\} dE = \sum_n 2\pi\Gamma_{n,l} j_l(E_{n,l}) \frac{p_{n,l}}{q_{n,l} + p_{n,l}}; \quad (30)$$

where  $j_{l\pm 1}(E)$  is the energy (thermal) distribution in the current of atoms colliding with  $l \pm 1$ ,  $w_{l\pm 1}(E)$  is the probability of transition from these states to the quasidiscrete levels  $E_{n,l}$  in the potential  $U_{12}(R)$  over which the sum is evaluated, and  $q_{n,l} = 2\pi\Gamma_{n,l}/\Omega_{n,l}$  is the barrier factor (probability of tunneling dissociation) for these levels. We have neglected the variation of  $\Delta(E)$ ,  $p_4(E)$ , and  $j_{l\pm 1}(E)$  on the scale of the resonance width,<sup>[3]</sup> and have substituted  $j_{l+1} \approx j_{l-1} \approx j_l$  at the end. To determine the resultant current, we must sum (29) over  $l$  and average over  $\theta$ . The resultant current obtained in this way is the sum of nondecreasing functions of the field intensity and, therefore, corresponds to a nondecreasing function  $S(I)$ . This result is in qualitative agreement with experiment.

For a quantitative comparison with experimental data, we note that the intensity characterizing the transition from the linear to the nonlinear region is determined by the minimum value of  $q_{n,l}$  in the expression for the resultant current. Estimates performed within the framework of the quasiclassical approximation for the Morse potential with the parameters given in<sup>[3]</sup> show that states with  $q_{n,l} < 10^{-4}$  are easily realized for  $l \gtrsim 100$ , which corresponds to  $\Gamma_{n,l}^{-1} > 10^{-8}$  sec. However, the maximum value of the dissociation time of the excited quasimolecule with which the atomic fluorescence can be associated must be less than the radiative decay time  $\gamma^{-1} \approx 10^{-8}$  sec since, unless the tunneling process succeeds in taking place in the time  $\gamma^{-1}$ , the frequency of the photon emitted by the quasimolecule will be substantially different from  $\omega_0$ . Hence, the characteristic value of the field  $F_s$  can be estimated from (in cgs units)

$$\pi D^2 F_s^2 / 2\hbar v_\omega \Delta U_\omega' \approx \gamma/\Omega.$$

For the parameter values adopted in<sup>[2]</sup>,  $\Delta U_\omega' = 0.5 \times 10^{-4}$  dyn/cm,  $D = 3 \times 10^{-18}$  cgs,  $v_\omega = 2 \times 10^4$  cm/sec,  $\gamma = 10^8$

$\text{sec}^{-1}$ , and  $\Omega = 10^{12} \text{ sec}^{-1}$ , we have  $I_s \approx 10^7 \text{ W/cm}^2$  which is in reasonable agreement with the experimental result under prolonged excitation. It is important to note that the value of the intensity for which the nonlinearity of the above process sets in ( $p \approx q \ll 1$ ) is much smaller than that corresponding to the Landau-Zener nonlinearity ( $p \approx 1^{[5]}$ ). Complete comparison between the experimental and theoretical functions  $S(I)$  requires, as already noted, averaging over  $\theta$  and summation over the contributions to the emission due to all resonances and the over-barrier states ignored in (29). The former can easily be carried out in a general form but does not lead to an important change in the final result: the function  $S(I)$  remains a saturating function for  $I \rightarrow \infty$ . The summation is a relatively difficult and independent problem. Nevertheless, it is clear that the presence of the broad resonances  $q_{n,l} \gg p_{n,l}$  ( $I > I_s$ ) and over-barrier states provides nonsaturating contributions to the function  $S(I)$ . This, and the fact that  $I_s$  is close to the measured value, may be regarded as an indication that the above model correctly explains the origin of the nonlinearity of  $S(I)$ .

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<sup>1)</sup>We follow the transition scheme discussed in [2].

<sup>2)</sup>The conditions for the validity of this approximation will be specified more precisely later.

<sup>3)</sup>The function  $S(E)$  varies slowly with  $E$  on the scale of the resonance width because the quasiclassical phase changes by  $\pi$  between resonances.

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