Radiative Collisions

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Elementary excitation transfer events involving simultaneous quantum emission and absorption by colliding atoms are considered. The probabilities of these events can be high and can strongly influence the kinetics of atomic level populations in a strong electromagnetic field with a frequency close to resonance. The conditions required for the relevant experiments are discussed briefly.

We shall use the expression "radiative collision of atoms"[1,2] to name the collisional transfer of excitation from one atom to another, through the respective emission and absorption of a light quantum having a frequency close to the energy difference between the transitions occurring in the colliding atoms. A reaction wherein atom X goes from state 2 to state 1, atom Y goes from state 1 to state 2 (Fig. 1), and a quantum of frequency ω is emitted, will be described by[1)]

$$X(2) + Y(1) \rightleftarrows X(1) + Y(2) + \hbar\omega. \qquad \text{(I)}$$

We are here considering frequencies ω that are close to the frequency $\omega_0 \equiv (E_{21}^X + E_{12}^Y)/\hbar$ and that are emitted predominantly when the separation of the atoms considerably exceeds their diameters. At close distances the atomic levels are considerably distorted. The energy difference of the transitions then differs appreciably from $\hbar\omega_0$; consequently only a negligibly small probability remains that a quantum of frequency $\omega \approx \omega_0$ will be emitted at close distances.[2)] This situation will be discussed in greater detail in Sec. 3.

Reaction (I) will be studied by the methods of perturbation theory. The unperturbed Hamiltonian will be the sum of the Hamiltonians of the interacting atoms; the perturbation will be the interaction of the atoms with each other and with an electromagnetic field. Assuming that the characteristic "transit time" is much shorter than the half-lives of the levels [determined by all processes except (I)], the resonance case could be limited to a two-level scheme. It is clear, however, that in the first order of perturbation theory the transition matrix element (along with the probability of the considered collision) will vanish, because reaction (I) then represents simultaneous transitions to new states of three pair-wise interacting objects. The appropriate matrix element will be different from zero only in the second-order perturbation case. The relations required for the study of reaction (I) will now be presented in Sec. 1.

1. In the Hamiltonian

$$\hat{H} = \hat{H}_0 + \hat{V}(t) \qquad (1.1)$$

of a quantum object let $\hat{V}(t)$ represent a small perturbation of the Hamiltonian $\hat{H}_0$. Using the equations for the coefficients $a_\gamma(t)$ in the expansion of the system's wave function in terms of the eigenfunctions of $\hat{H}_0$, it can be shown[2] that a resonance transition, i.e., a transition between states γ_1 and γ_2, for which the quantity

$$\hbar\Delta\omega \equiv \mathscr{E}_{\gamma_1} - \mathscr{E}_{\gamma_2}$$

is sufficiently small, is described by the differential equations

$$\begin{aligned} i\hbar\dot{a}_{\gamma_1} &= \mathscr{V}_{\gamma_1\gamma_1}a_{\gamma_1} + \mathscr{V}_{\gamma_1\gamma_2}e^{i\Delta\omega t}a_{\gamma_2}, \\ i\hbar\dot{a}_{\gamma_2} &= \mathscr{V}_{\gamma_2\gamma_2}a_{\gamma_2} + \mathscr{V}_{\gamma_2\gamma_1}e^{-i\Delta\omega t}a_{\gamma_1}, \end{aligned} \qquad (1.2)$$

where

$$\mathscr{V}_{\gamma_a\gamma_b} = V_{\gamma_a\gamma_b} + \sum_{\gamma\neq\gamma_1,\gamma_2} \frac{V_{\gamma_a\gamma}V_{\gamma\gamma_b}}{\mathscr{E}_{\gamma_b} - \mathscr{E}_\gamma} \quad (a, b = 1, 2). \qquad (1.3)$$

For the applicability of (1.2) it is sufficient to fulfill the small-perturbation condition

$$|V_{\gamma_a\gamma}| \ll |\mathscr{E}_{\gamma_a} - \mathscr{E}_\gamma| \quad (a = 1,2;\ \gamma \neq \gamma_1, \gamma_2) \qquad (1.4)$$

and the adiabatic perturbation condition

$$\tau_V \gg \hbar/|\mathscr{E}_{\gamma_a} - \mathscr{E}_\gamma|, \quad (a = 1,2;\ \gamma \neq \gamma_1, \gamma_2), \qquad (1.5)$$

where τ_V is the characteristic variation time of $\hat{V}(t)$.

2. A radiative collision is conveniently characterized by its cross section, i.e., the cross section for the excitation transfer (I) in the field of a plane monochromatic wave of frequency ω. Assuming in advance that the wavelength $2\pi c/\omega$ of the field greatly exceeds the characteristic separations of the atoms involved in (I), we use

$$\mathbf{E}(t) = \mathbf{E}_0 \cos(\omega t) \qquad (2.1)$$

to describe the electric field in the collision region. This is considered to be a classical field; in the present problem a quantum field leads only to a complication of the notation.

Assuming also classical motion of the nuclei, we represent the radiative collision cross section by

$$\sigma(v, \omega, E_0) = 2\pi \int_0^\infty w(\rho, v, \omega, E_0)\,\rho\,d\rho, \qquad (2.2)$$

where $w(\rho, u, \omega, E_0)$ is the probability of transition (I)

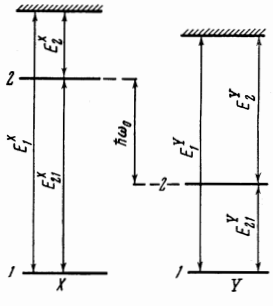

FIG. 1. Atomic levels.

[1)] State 2 is not necessarily the first excited state of the atom.
[2)] We disregard accidental equality of $\hbar\omega_0$ and the difference between the energy levels at close distances.

in an atomic collision with the impact parameter ρ and relative velocity v within the field defined by (2.1).

Assuming a dipole-dipole interaction,[3] we write the Hamiltonian of the system in the form

$$\hat{H} = \hat{H}_0 + \hat{V}(t), \quad \hat{H}_0 = \hat{H}^X + \hat{H}^Y, \tag{2.3}$$

$$\hat{V}(t) = R^{-3}(t)\gamma_{ij}d_i^X d_j^Y + (\mathbf{d}^X + \mathbf{d}^Y)\mathbf{E}(t). \tag{2.4}$$

Here $\hat{H}^X$ and $\hat{H}^Y$ are the Hamiltonians of noninteracting atoms X and Y; d_i^X and d_j^Y are the vector operators of the dipole moments; $\mathbf{R}$ is the vector separation of the atomic nuclei; $\gamma_{ij} = \delta_{ij} - 3e_ie_j$, where δ_{ij} is the Kronecker symbol and $e_i = R_i/R$ is a unit vector along the axis connecting the nuclei. Here and henceforth we shall understand that summation is performed over repeated "spatial" indices [over i and j in (2.4)], in accordance with the usual convention of tensor algebra.

To determine the probability of the transition (I) we use a system of equations analogous to (1.2):

$$\begin{gathered} i\dot{a}_1 = V_1a_1 + V e^{i\Delta\omega t}a_2, \quad i\dot{a}_2 = V_2a_2 + V e^{-i\Delta\omega t}a_1, \\ a_1(-\infty) = 1, \quad a_2(-\infty) = 0, \end{gathered} \tag{2.5}$$

where

$$V_1 = \frac{C_1}{R^6} + B_1E_0^2, \quad V_2 = \frac{C_2}{R^6} + B_2E_0^2, \quad V = \frac{C_3}{R^3}E_0 \tag{2.6}$$

are the matrix elements of the perturbation (2.4), taken in the second order of perturbation theory. The coefficients C and B represent the following expressions[2]:

$$C = \frac{1}{\hbar}\sum_{\xi,\eta}\frac{|\gamma_{ij}\langle\xi'|d_i^X|\xi\rangle\langle\eta'|d_j^Y|\eta\rangle|^2}{\mathscr{E}^X_{\xi'\xi} + \mathscr{E}^Y_{\eta'\eta}},$$

with $C_1 = C(\xi' = 2, \eta' = 1)$, $C_2 = C(\xi' = 1, \eta' = 2)$;

$$B = \frac{1}{4\hbar}[|\langle\xi'|\hat{\alpha}_{EE}^X(\omega)|\xi'\rangle|^2 + |\langle\eta'|\hat{\alpha}_{EE}^Y(\omega)|\eta'\rangle|^2],$$

with $B_1 = B(\xi' = 2, \eta' = 1)$, $B_2 = B(\xi' = 1, \eta' = 2)$;

$$C_3 = \frac{1}{4\hbar}\gamma_{ij}[\langle 1|d_j^Y|2\rangle(\langle 2|\hat{\alpha}_{iE}^X(\omega_{12}^Y)|1\rangle + \langle 2|\hat{\alpha}_{iE}^X(\omega)|1\rangle)$$
$$+ \langle 2|d_i^X|1\rangle(\langle 1|\hat{\alpha}_{jE}^Y(\omega_{21}^X)|2\rangle + \langle 1|\hat{\alpha}_{jE}^Y(\omega)|2\rangle)].$$

Here $\hat{\alpha}_{ij}^Z(\omega)$, the tensor polarizability operator for an atom having charge Z in a field of frequency ω, is given by the matrix

$$\langle\zeta_1|\hat{\alpha}_{ij}^Z(\omega)|\zeta_2\rangle = \sum_\zeta\left(\frac{\langle\zeta_1|d_i^Z|\zeta\rangle\langle\zeta|d_j^Z|\zeta_2\rangle}{E^Z_{\zeta,\zeta} + \hbar\omega} + \frac{\langle\zeta_1|d_j^Z|\zeta\rangle\langle\zeta|d_i^Z|\zeta_2\rangle}{E^Z_{\zeta,\zeta} - \hbar\omega}\right)$$

(the index E of the polarizability operators designates the component along the vector $\mathbf{E}_0$). We shall, for the time being, ignore angular dependences in the coefficients C and B and also the variation in the velocity of atomic approach,[4] assuming $R = \sqrt{\rho^2 + v^2t^2}$. In this paper the cross section and other characteristics of the radiative collision will be calculated only for the frequency

$$\omega' = \omega_0 + (B_2 - B_1)E_0^2, \tag{2.7}$$

which will here be called the "center" of the line (see Section 3).

[3]The applicability of the dipole-dipole approximation will be discussed in Sec. 3.

[4]The applicability of these assumptions will also be discussed in Sec. 3.

The system (2.5) was solved asymptotically in[3], where the formula derived for the transition probability $w = |a_2(\infty)|^2$ is

$$w = \left|\int_{-\infty}^{\infty} V(t)\cos\left\{\int_0^t[(\Delta\omega + V_2 - V_1)^2 + 4V^2]^{1/4}dt'\right\}dt\right|^2. \tag{2.8}$$

The limitations on the applicability of this formula are not entirely clear, although it includes the Born approximation as a special case, agrees with the exact solution at resonance ($\Delta\omega = 0$, $V_1 - V_2 = 0$) and for a rectangular potential [$V_1 - V_2 = 0$, $V(t) = \text{const}$ for $|t| < T$, $V(t) = 0$ for $|t| > T$], and for crossing the levels gives values close to the Landau-Zener formula.

For $\omega = \omega'$ Eq. (2.9) yields

$$w = \left|\int_{-\infty}^{\infty}\frac{C_3E_0}{R^3(t)}\cos\left\{\int_0^t\left(\left[\frac{C_{12}}{R^6(t)}\right]^2 + \left[\frac{2C_3E_0}{R^3(t')}\right]^2\right)^{1/4}dt'\right\}dt\right|^2, \tag{2.9}$$
$$C_{12} = |C_1 - C_2|.$$

We shall now consider the two limiting cases of large and small fields E_0. The second term of the radical in (2.9) exceeds the first term when the nuclear separation exceeds a certain value:

$$R > a, \quad a \equiv (2C_{12}/E_0C_3)^{1/3}. \tag{2.10}$$

Consequently, with sufficiently large E_0 the first term is significant only for small values of R, which do not contribute appreciably to the cross section. In this case the probability of (I) is

$$w = \sin^2\left(\int_{-\infty}^{\infty}\frac{C_3E_0}{R^3(t)}dt\right) = \sin^2\left(\frac{2C_3E_0}{v\rho^2}\right), \tag{2.11}$$

so that for the radiative collision cross section we obtain

$$\sigma_1 = \pi^2C_3E_0/v. \tag{2.12}$$

The "strong field" condition required for the validity of these formulas is obtained from the relation

$$\sqrt{\sigma_1/\pi} \gg a, \tag{2.13}$$

which gives[5]

$$E_0 \gg (v^3C_{12}^2)^{1/3}/2C_3. \tag{2.14}$$

In the case of relatively weak fields the second term of the radical in (2.9) may be neglected. The probability of transition (I) is then

$$w_2 = E_0^2C_3^2\left|\int_{-\infty}^{\infty}\cos\left(C_{12}\int_0^t\frac{dt'}{R^6(t')}\right)\frac{dt}{R^3(t)}\right|^2$$
$$= E_0^2\left(\frac{C_3}{v\rho^2}\right)^2\left|\int_{-\infty}^{\infty}\frac{\cos(f(x)C_{12}/v\rho^5)\,dx}{(1+x^2)^{3/2}}\right|^2 \tag{2.15}$$

where

$$f(x) \equiv \frac{x}{4(1+x^2)^3} + \frac{3x}{8(1+x^2)} + \frac{3}{8}\operatorname{arctg}x. \tag{2.16}$$

From (2.15) we obtain the cross section for (I) in weak fields:

$$\sigma_2 = 3\pi C_3^2C_{12}^{-2/3}v^{-4/3}E_0^2. \tag{2.17}$$

Finally, with arbitrary E_0 the cross section for (I) is given by the general expression

$$\sigma = 2\pi C_3^2C_{12}^{-2/3}v^{-4/3}E_0^2\chi\left(\frac{2E_0C_3}{C_{12}^{2/3}v^{4/3}}\right), \tag{2.18}$$

[5]We note that the field is here assumed to be considerably weaker than the atomic field.

where

$$\chi(\alpha)=\int_0^\infty \frac{dy}{y^3}\left|\int_{-\infty}^{\infty}\frac{dx}{(1+x^2)^{3/2}}\cos\left\{\frac{1}{y^3}\int_0^x[(1+z^2)^{-6}+\alpha^2y^6(1+z^2)^{-3}]^{1/2}dz\right\}\right|^2 \tag{2.19}$$

When the argument of the function is much larger than unity (2.18) becomes (2.12); for small arguments it becomes (2.17).

3. We shall now discuss the applicability of the relations derived in Section 2. The most important simplifying assumptions were: a) dipole-dipole interaction, b) rectilinear and uniform motion of the colliding atoms, and c) negligible angular dependence of the parameters C_{12} and C_3.

Rectilinear trajectories are provided for by the left-hand member of the relation

$$\hbar/M\rho_0 \ll v \ll v_0 = 2.2\cdot 10^8 \text{ cm/sec}, \tag{3.1}$$

where M is the reduced mass of the colliding atoms and ρ_0 is the characteristic linear dimension of the region that contributes most to the probability of the transition being analyzed.

The assumption of dipole-dipole interaction is justified when the contribution to transition (I) with radiation of frequency ω' comes from separations that greatly exceed the atomic diameter.[6] Regarding ρ_0 as the interatomic separation at which the level shifts are of the order of the inverse "transit time," we obtain[7]

$$\rho_0 \sim (C_{12}/v)^{1/5} \gg a_0 \tag{3.2}$$

or

$$v \ll C_{12}/a_0^5 \sim v_0, \tag{3.3}$$

which agrees with the right-hand member of (3.1).

The neglect of the angular dependence of C_{12} and C_3 means, in fact, that the results of Sec. 2 are true except for a factor of the order of unity. In the formulas of Sec. 2 the roles of C_{12} and C_3 can be performed by their angular averages.

In the present work the squares of the parameters were averaged. As previously, it is convenient to calculate for the limiting cases of strong and weak fields. In the strong-field case we shall assume that the atomic dipole moments are quantized along the z axis, in the direction of the vector $\mathbf{E}_0$; their average values are projected upon the line connecting the nuclei:

$$\mathbf{dE}_0 = d_z E_0, \quad \gamma_{ij} d_i^X d_j^Y = d_z^X d_z^Y (1-3\cos^2\theta),$$

where θ is the angle between the field direction and the axis connecting the nuclei. In the weak-field case we take the direction of R as the quantization axis z′:

$$\mathbf{dE}_0 = d_{z'} E_0 \cos\theta', \quad \gamma_{ij} d_i^X d_j^Y = -2 d_{z'}^X d_{z'}^Y.$$

The averaging of the squares of the parameters is now reduced to the averages $\overline{(1-3\cos^2\theta)^2} = 4/5$, $\overline{\cos^2\theta}^2 = 1/3$.

For collisions in weak fields the atomic levels can be considered degenerate (to the same degree of accuracy as the entire analysis) with respect to the dipole moment projection on the z′ axis, because the field does not separate the levels strongly and in the region where splitting due to interatomic interaction is important it does not determine the order of magnitude of the cross section. Here it is reasonable to introduce a radiative collision cross section that is averaged over the initial states of atoms X and Y and is summed over their final states.[2]

In sufficiently strong fields

$$E_0 \gg v^{3/5} C_{12}^{1/10} |B_1 - B_2|^{1/2} \tag{3.4}$$

we find that as a result of Stark splitting only one transition will, generally speaking, contribute to a line; the other transitions will not be resonant.[8]

We note that selection rules exist for radiative collisions. The probability of (I) vanishes when the following quantity equals zero:

$$C_3 \sim \langle 1|d_i^Y|2\rangle(\langle 2|\hat{a}_{j_q}^X(\omega_{12}^Y)|1\rangle + \langle 2|\hat{a}_{j_q}^X(\omega)|1\rangle) + \langle 2|d_i^X|1\rangle(\langle 1|\hat{a}_{j_q}^Y(\omega_{21}^X)|2\rangle + \langle 1|\hat{a}_{j_q}^Y(\omega)|2\rangle). \tag{3.5}$$

When we limit ourselves to the case where LS coupling can be assumed in the atoms we have the selection rules

$$\begin{aligned} &\Delta S^X, \quad \Delta S^Y = 0; \\ &\Delta L^Y = 0;\ 2 \text{ with } \Delta L^X = 1, \\ &\Delta L^X = 0;\ 2 \text{ with } \Delta L^Y = 1. \end{aligned} \tag{3.6}$$

In the strong field of (3.4), we add to (3.6) a set of selection rules with respect to the magnetic quantum numbers M^X and M^Y.

4. When the cross section for the radiative collision is known we can evaluate the influence of the reaction (I) on the kinetics of atomic level populations and on the field intensity. In the case of strong electric fields, which at the present time are provided by means of lasers, (I) can have large influence on the population kinetics. For example, if in (2.12) we assume $u \sim 10^5$ cm/sec and $E_0 \sim 10^7$ V/cm (which corresponds to the intensity $I \sim 10^{10}$ W/cm) and let $C_3 \sim 10$ at. units, we obtain the radiative collision cross section $\sigma \sim 10^{-14}$ cm^2, which considerably exceeds the usual gas-kinetic cross sections. At the same time the cross section for direct excitation transfer (disregarding the special Landau-Zener or Rosen-Zener cases) is adiabatically small when the energy defects are of the order of a few eV.[9]

To illustrate the possible effect of radiative collisions on a resonant field we shall now evaluate light amplification and absorption at the "center" of a line having the frequency (2.7). Let us consider the weak-field case. The populations (N_ξ^X, N_η^Y) of the atomic levels are assumed to be given, and the light is propagated along the z axis. Then for a change of light-wave intensity in a layer of thickness dz we have

$$dI = \hbar\omega\,\langle\bar{\sigma}_2 v\rangle\, g\left(\frac{N_2^X N_1^Y}{g_2^X g_1^Y} - \frac{N_1^X N_2^Y}{g_1^X g_2^Y}\right) dz. \tag{4.1}$$

Here $g \equiv g_2^X g_1^X g_2^Y g_1^Y$ is the product of the statistical weights of the atomic levels in the initial and final states of reaction (I); $\bar{\sigma}_2$ is the radiative collision cross

[6] In the present work we shall assume that at these distances the exchange interaction between the given atoms is unimportant.

[7] In connection with the condition (3.4) we note that for weak fields ρ_0 cannot be equated to the square root of the cross section.

[8] This pertains to the monochromatic wave (2.4). Averaging over the projections of the moments can be performed in this case if the line width of the applied field greatly exceeds the Stark splitting.

[9] In the argument of the exponential factor in the transition probability a negative quantity of the order of Massey's parameter appears. For $\hbar\omega_0 \sim 1$ eV, $v \sim 10^5$ cm/sec, and $\rho_0 \sim 10^{-7}$ cm we have $\omega_0\rho_0/v \sim 10^3$.

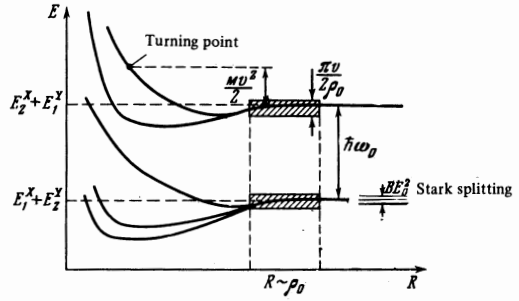

FIG. 2. Levels of a quasimolecule.

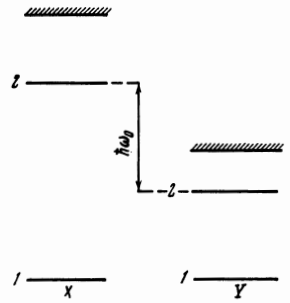

FIG. 3

section averaged over the initial and final states; the angular brackets here denote averaging over the relative velocities of the colliding atoms.

When the atomic populations satisfy the condition

$$N_2^X N_1^Y / g_2^X g_1^Y > N_1^X N_2^Y / g_1^X g_2^Y, \tag{4.2}$$

light amplification ($|E_{21}^X| > |E_{21}^Y|$) occurs; when the inverse condition is satisfied light is absorbed. The relation (4.2) can, in principle, be satisfied easily in a recombination regime; for example, state 2 of atom X could be selected as a metastable state and state 2 of atom Y as a state that is well depopulated by electronic impacts. In weak fields the light amplification dI is proportional to the square of the field amplitude; we can use the concept of "linear" gain defined by

$$\varkappa = \frac{d}{dz}\ln I(z). \tag{4.3}$$

For a plane wave we have

$$I = \frac{c}{4\pi}\overline{E_0^2(t)} = \frac{c}{8\pi}E_0^2, \tag{4.4}$$

and in accordance with (4.1) we obtain

$$\varkappa = \frac{24\pi^2}{c}\langle v^{-3/5}\rangle \overline{\hbar\omega C_3^2 C_{12}^{-2/5}}\, g\left(\frac{N_2^X N_1^Y}{g_2^X g_1^Y} - \frac{N_1^X N_2^Y}{g_1^X g_2^Y}\right), \tag{4.5}$$

where c is the velocity of light. Taking $\overline{\hbar\omega C_3^2 C_{12}^{-2/5}}$ ~ 100 at. units and v ≈ 10^5 cm/sec, we have

$$24\pi^2 c^{-1}\langle v^{-3/5}\rangle \overline{C_3^2 C_{12}^{-2/5}}\, g\hbar\omega \approx 3\cdot 10^{-38}\ \text{cm}^5.$$

Thus, with the quite high but actually achievable concentrations $N_2^X/g_2^X \approx 10^{15}$ cm^{-3} and $N_1^Y/g_2^Y \approx 3\times 10^{19}$ cm^{-3}, it becomes possible to reach a gain $\kappa \sim 10^{-3}$ cm^{-1} that is sufficient for laser action on the basis of radiative collisions betweens atoms X and Y. The gain associated with radiative collisions was calculated in[1] using a somewhat different approach to the solution of the problem but obtaining close results.

In strong fields the growth of radiation intensity is proportional to its square root; therefore the "linear" gain introduced formally by (4.3):

$$\varkappa = \frac{2\pi^{3/2}C_3}{v}\sqrt{\frac{c}{I}}(N_{k_2}^X N_{n_1}^Y - N_{k_1}^X N_{n_2}^Y) \tag{4.6}$$

is diminished as I increases.

In the present paper the different characteristics of radiative collisions have been considered only for the "center" of a line. This term is somewhat arbitrary, because the spectral line of a radiative collision can exhibit one or more peaks at frequencies different from ω′. For example, this effect can be associated with an exchange interaction that shifts levels when the interatomic separations are much smaller ($R \ll \rho_0$) while it enhances the probability of quantum emission at such distances. Moreover, when motion occurs in repulsive states the "turning points" (Fig. 2) can make appreciable contributions to transitions at other frequencies. However, the proper allowance for these effects and the analysis of an entire line contour for radiative collisions probably requires numerical calculations for a specific pair of atoms.

The inverse problem would undoubtedly be of interest, namely to investigate the character of an interatomic interaction on the basis of the radiative collision line shape (for emission or absorption). We now discuss briefly the scheme of experiments for observing radiative collisions and analyzing them in considerable detail. At the present time we can count on parametrically tuned lasers emitting pulses at about a megawatt in the 8000—14000 Å range. A mixture of dense alkali metal vapors would be suitable for the experiments. Pulsed irradiation of an alkali vapor by means of a laser tuned to the resonance frequency $\omega_{21}^Y = |E_2^Y - E_1^Y|/\hbar$ of the appropriate atom can produce a sufficiently large population N_2^Y of its first excited state. A second laser tuned to the frequency $\omega \approx \omega_0$ associated with process (I) can appreciably populate state 2 of atom X from state 2 of atom Y.

In selecting specific pairs of atoms X and Y for such experiments the following considerations must be taken into account: a) Process (I) must be allowed [see (3.5) and (3.6)]; b) the frequencies ω_{12}^Y and ω_0 should not be close to other characteristic frequencies of the atoms; c) for observation of the concentration N_2^X it is desirable that between the X(2) state and the ground state of X there be an intermediate state to which an allowed spontaneous optical transition goes from the X(2) state; d) it is desirable that the optical transition Y(1) → Y(2) be strong and that C_3 be a sufficiently large constant (these constants are large for alkali metals).

The foregoing considerations indicate that a mixture of cesium and potassium vapors, for example, is suitable, with the $6p^2P_{1/2}$ state of Cs for Y(2) and the $5s^2S_{1/2}$ state of potassium for X(2).

In conclusion, we wish to draw attention to an additional experimental possibility. If state 2 of atom X "lies in the continuum" of atom Y (see Fig. 3), then process (I) can greatly facilitate the photoionization of level 2 of atom Y by a quantum of frequency $\omega \approx \omega_0$. In this case X(2) is like a self-ionized state of the XY quasi-molecule. Under suitable conditions (concentrations and selected states of X and Y etc.) this process will lower the critical value of the light field that is required for the breakdown of an X-Y mixture at the focus

of a laser pulse with the frequency $\omega \approx \omega_0$. This effect has apparently been observed experimentally (see[4] and especially[5]).

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