3) It is easy to show that this is due to the fact that $CO_2$ has no absorption lines in the $Q$ branches.

4) Electro-ionization CO and $CO_2$ lasers of high pressure (~10 atm) can be continuously tuned within a sufficiently wide range.[13] The driving laser can therefore be a CO laser of higher pressure than an amplifier tuned to $\nu \approx 1930.2\ \mathrm{cm}^{-1}$.

5) We use as measure of the duration of the output pulse its duration at a level $r = 1/3$ of $I_m$, since the energy of the output pulse is concentrated in the base of the pulse (see Fig. 1).

6) The maximum intensity of the input pulses that are effectively amplified as a result of the one-photon process, can be easily estimated from the known formulas for the gain of a short pulse.[17] Its value for a CO laser at $\tau_p \sim 10^{-9}$ sec is $10^5-10^6$ W/cm$^2$.


1. A. M. Prokhorov, Usp. Fiz. Nauk **85**, 599 (1965) [Science **149**, 821 (1965)].
2. P. P. Sorokin and N. Braslau, IBM, J. Res. Develop. **8**, 338 (1964).
3. G. L. Gurevich and Yu. G. Khronopulo, Izv. Vyssh. Uchebn. Zaved. Radiofiz. **8**, 493 (1965).
4. T. I. Il'inova, Vestn. Mosk. Univ. Fiz. Astron. No. 4, 79 (1966).
5. V. S. Letokhov, Pis'ma Zh. Eksp. Teor. Fiz. **7**, 221 (1968) [JETP Lett **7**, 169 (1968)].
6. G. M. Krochik and Yu. G. Khronopulo, Pis'ma Zh. Eksp. Teor. Fiz. **21**, 585 (1975) [JETP Lett. **21**, 274 (1975)].
7. H. P. Juen, Appl. Phys. Lett. **26**, 505 (1975).
8. R. L. Carman, Phys. Rev. A **12**, 1048 (1975).
9. K. J. McNeil and D. J. Walls, J. Phys. A Gen. **8**, 104 (1975).
10. E. B. Gordon and Yu. L. Moskvin, Zh. Eksp. Teor. Fiz. **70**, 1729 (1976) [Sov. Phys. JETP **43**, 901 (1976)].
11. V. R. Blok, G. M. Krochik, and Yu. G. Khronopulo, Zh. Eksp. Teor. Fiz. **76**, 46 (1979) [Sov. Phys. JETP **49**, 23 (1979)].
12. V. S. Butylkin, A. E. Kaplan, Yu. G. Khronopulo, and E. I. Yakubovich, Rezonansnye vzaimodeistviya sveta s veshchestvom (Resonant Interactions of Light with Matter), Nauka, 1977.
13. V. A. Danilychev, O. M. Kerimov, and I. B. Kovsh, Tr. Fiz. Inst. Akad. Nauk SSSR **85**, 49 (1976).
14. K. Boyer, Los Alamos Scientific Laboratory, Presentation at Japan--US Seminar on Laser Interaction with Matter, Kyoto, September 24–29, 1972.
15. N. V. Karlov and Yu. B. Konev, Spravochnik po lazeram (Laser Handbook), Vol. 1, Sovetskoe radio, 1978, p. 133.
16. S. S. Penner Quantitative Molecular Spectroscopy and Gas Emissivities, Addison-Wesley, 1959.
17. Ya. N. Khanin, Kvantovaya Radiofizika (Quantum Radiophysics), Vol. 2, 1976, p. 130.


Translated by J. G. Adashko

# Radiative collisions of atoms with excitation transfer at low resonance defects


D. S. Bakaev and Yu. A. Vdovin

*Moscow Engineering Physics Institute*



(Submitted 10 July 1979)
Zh. Eksp. Teor. Fiz. **78**, 497–506 (February 1979)



The effective cross section for the transfer of excitation from atom to atom in an external electromagnetic field with absorption and emission of a photon is calculated for small resonance defects. The radiative-collision line contour is determined in the strong- and weak-field approximations. It is shown that allowance for the angular dependence of the dipole-dipole interaction of the atoms affects significantly the line contour near resonance.



PACS numbers: 34.10. + x


Atomic collisions accompanied by emission or absorption of a photon (radiative collisions) are being intensively investigated both theoretically and experimental (see Yakovlenko's review[1]). We consider in this paper radiative collisions with excitation transfer

$$B^{\cdot}(2)+A(1)+\hbar\omega \rightleftharpoons B(1)+A^{\cdot}(2)$$

in an external electromagnetic field, when the photon energy $\hbar\omega$ is close to the energy difference $\hbar\omega_0$ between the initial and final states of the atomic systems (see Fig. 1):

$$E_2^{(A)}+E_1^{(B)}-E_1^{(A)}-E_2^{(B)}=E_{21}^{(A)}-E_{21}^{(B)}=\hbar\omega_0. \tag{1}$$

A resonant situation is produced here (the resonance defect is small) and one can expect relatively large effective cross sections if the electromagnetic field is strong enough. The recent advent of high-power tunable lasers has stimulated to a considerable degree the interest in these processes.

These collisions were first investigated theoretically for the resonant case by Gudzenko and Yakovlenko[2] and in later studies.[3–8] Ostroukhov, Smirnov, and Shlyapnikov[9] have considered the situation when the detuning from resonance is large enough and calculation of the cross section calls for knowledge of the quasimolecular terms of the atomic system. Excitation transfer in a

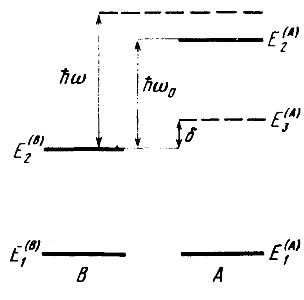


FIG. 1. Level scheme of atomic system.



0038-5646/80/020250-05$02.40

resonant field from strontium to calcium atoms was experimentally observed in Refs. 10 and 11, and from europium to strontium atoms in Ref. 12.

The operator of the interaction of the atoms with one another and with an external electromagnetic field of strength $\mathbf{E}$ and of frequency $\omega$ is given by

$$u(t)=[(\mathbf{d}_1\mathbf{d}_2)R^2-3(\mathbf{d}_1\mathbf{R})(\mathbf{d}_2\mathbf{R})]/R^5+(\mathbf{d}_1+\mathbf{d}_2)\mathbf{E}, \tag{2}$$

where $\mathbf{d}_1$ and $\mathbf{d}_2$ are the dipole-moment operators of the colliding atoms $B$ and $A$, respectively; $R^2=v^2t^2+\rho^2$; $v$ is the relative velocity of the atoms, and $\rho$ is the impact parameter. The atom motion can be regarded as along a straight line with constant relative velocity.

We denote by $a_1$ and $a_2$ the amplitudes of the states corresponding respectively to excited atom $B$ and unexcited $A$ on the one hand, and to unexcited atom $B$ and excited $A$, on the other. The equations for the amplitudes are of the form

$$\begin{aligned} i\dot{a}_1&=V_1a_1+Va_2e^{i(\omega-\omega_0)t},\\ i\dot{a}_2&=V_2a_2+Va_1e^{-i(\omega-\omega_0)t} \end{aligned} \tag{3}$$

with initial conditions $a_1(-\infty)=1$ and $a_2(-\infty)=0$, where $\omega-\omega_0$ is the resonance defect;

$$V_1=C_1/R^6+B_1E_0^2,\quad V_2=C_2/R^6+B_2E_0^2,$$
$$V=BE_0/R^3$$

are the matrix elements of the interaction operator, taken in second-order perturbation theory, and $E_0$ is the field amplitude. It is assumed that the excited state of the atom $B$ is connected with its ground state by an allowed dipole transition.

The general expressions for the coefficients $C$ and $B$ were obtained in Ref. 2. It must be noted, however, that the coefficient $B$, and hence the transition probabilities, becomes appreciable if the initially unexcited atom has an intermediate level connected with the ground state by a dipole transition, the frequency of transition to which is close to the transition frequency of the atom $B$. Denoting the energy of the intermediate state by $E_3^{(A)}$, we have (see Fig. 1)

$$E_3^{(A)}-E_1^{(A)}=E_{31}^{(A)}=E_{21}^{(B)}+\delta,$$

where $\delta\ll E_{21}^{(B)}$. This is precisely the situation realized in the experiment.[12] In this case the principal role is played by transitions through this intermediate level, and approximately we have

$$B=\frac{1}{4\hbar\delta}\left(\delta_{ij}-\frac{R_iR_j}{R^2}\right)(d_i^{(1)})_{12}\sum_t[(d_E^{(2)})_{2t}(d_j^{(2)})_{t1}+(d_j^{(2)})_{2t}(d_E^{(2)})_{t1}]. \tag{4}$$

Here $(d_i^{(1)})_{mn}((d_i^{(2)})_{mn})$ stands for the matrix elements of the $i$-th projection of the dipole moment of the atom $B$ ($A$) taken between the states $m$ and $n$. The summation is over all the states corresponding to the intermediate level with energy $E_3^{(A)}$. The index $E$ denotes the component directed along the field $E_0$.

The expression for $B$ is valid under the condition that $\delta$ is small compared with the atomic frequencies of the transitions, but is at the same time large compared with the resonance defect. For the system (3) to be valid it is also necessary that $\delta$ be large compared with the effective values of $V_1$, $V_2$ and $V$. We note that the resonant situation is realized also under the condition

$$E_{23}^{(A)}=E_{21}^{(B)}+\delta.$$

A consistent allowance for the possible level degeneracy in the system (3) is a rather complicated matter. Neglect of this degeneracy, however, replacement of the matrix elements $V_1$, $V_2$, $V$ by their values averaged over the angles, as was done for example in Refs. 2–7, obscures some singularities of the line contour of the radiative collisions near resonance. We shall therefore take into account the level degeneracy quasiclassically,[13] replacing the matrix element of the dipole-moment operator of the atom $B$ by the classical vector

$$(d_i^{(1)})_{12}\to d_{12}^{(1)}\mathbf{n}/(2j+1)^{1/2}, \tag{5}$$

where $d_{12}^{(1)}$ is the modulus of the reduced matrix element of dipole-moment operator of the atom $B$ for the transition under consideration, $j$ is the angular momentum of the excited state, and $\mathbf{n}$ is a unit vector in the direction of the dipole moment of the atom, with subsequent averaging of the final expressions over the directions of the vector $\mathbf{n}$. For the atom $A$, the selection rules for the transition between the ground and excited state in the case of $L$–$S$ coupling yield

$$\Delta l=0,2,\quad \Delta s=0.$$

For a $0\to 0$ transition, both atomic states are degenerate. However, if the initial or final state (or both) of the atom $A$ are degenerate, then this degeneracy is taken into account in analogy with (5), followed by averaging of the obtained expressions. This procedure is valid for transitions between states with large intrinsic angular momenta $j$, when the arising corrections are of the order of $1/j$. In the general case, the numerical factors obtained in this approach are correct only in order of magnitude. Nonetheless, the line-contour fine structure revealed in this manner seems to be general in character and is not connected with the indicated simplifications. In particular, such a structure was revealed also in our earlier paper,[8] where the degeneracy in the atom was lifted by a magnetic field.

Our task is to calculate the effective cross section of the excitation transfer as a function of the frequency of the electromagnetic field:

$$\sigma(\omega)=2\pi\int_0^\infty\langle|a_2(\infty)|^2\rangle\rho d\rho, \tag{6}$$

where the angle brackets denote averaging over the directions of the dipole-moment vector of the atom $B$ (and of the atom $A$ in the case of a transition between degenerate states), as well as over the orientations of the collision plane relative to the electromagnetic field polarization vector. The effective cross section $\sigma$ must also be averaged over the Maxwellian velocity distribution. We solve the system (3) in the weak- and strong-field approximations.[2]

## 1. STRONG-FIELD APPROXIMATION

This approximation means that over the effective distances the diagonal matrix elements $C_i/R^6 (i=1,2)$ are small compared with $V$ and can therefore be left out. As seen from the result (10), (11) (see below), we have in order of magnitude

$$R_{\mathrm{eff}}\sim\sigma^{1/2}\sim(|B_0|E_0/\hbar v)^{1/2}.$$

Consequently the inequality $C_i/R^6 \ll V(i=1,2)$ is satisfied under the condition

$$E_0 \gg (v^3 C_i^2)^{1/5}/|B_0|. \tag{7}$$

With this inequality taken into account, the system (3) takes the form

$$i\dot{b}_1 = Ve^{i\Delta\omega t}b_2, \quad i\dot{b}_2 = Ve^{-i\Delta\omega t}b_1, \tag{8}$$
$$b_1(-\infty)=1, \quad b_2(-\infty)=0,$$

where

$$b_k(t)=a_k(t)\exp(iB_kE_0^2t), \quad k=(1,2),$$
$$\Delta\omega=\omega-\omega_0+E_0^2(B_1-B_2), \tag{9}$$

$\Delta\omega$ is the resonance defect with account taken of the term shift due to the dynamic Stark effect.

We assume first that a transition between states with zero angular momentum takes place first in the atom $A$ ($0 \to 0$ transition). In this case the angular momentum of the intermediate level is equal to unity, and from among the three intermediate states only the state with $m=0$ contributes to $B$ [see (4)]. The expression for $V$ is then

$$V=\frac{BE_0}{R^3}=\left[(\mathbf{ne})-\frac{3(\mathbf{Rn})(\mathbf{Re})}{R^2}\right]\frac{B_0E_0}{R^3}, \tag{10}$$

where

$$B_0=d_{12}^{(1)}d_{23}^{(2)}d_{31}^{(2)}/6\hbar\delta(2j+1)^{1/2},$$

$d_{23}^{(2)}$ and $d_{31}^{(2)}$ are the reduced matrix elements of the dipole-moment of the atom $A$ for the transition between the considered states (see, e.g., Ref. 14); $\mathbf{e}$ is the unit vector of the electromagnetic-field polarization. The final expressions must be averaged both over the directions of the vector $\mathbf{n}$ and over the directions of $\mathbf{e}$ (averaging over the orientations of the colllsion plane relative to the direction of the field polarization). The system (8) with the interaction operator (10) is equivalent to the system considered in Ref. 13 with another interaction constant. At small resonance defects $\Delta\omega$, using the results of Ref. 13, we have

$$\sigma(\lambda)=\frac{\pi B_0E_0}{8v}\left\{1-\frac{8}{9}\lambda^2[\ln(\lambda^2)+5{,}93]\right\}, \tag{11}$$

where $\lambda=2(B_0E_0)^{1/2}|\Delta\omega|/\pi v^{3/2}$ is a dimensionless smallness parameter. Expression (1) is valid at $\lambda \ll 1$. At $\lambda=0$ we have exact resonance.

Assume now that the transition in atom $A$ is between degenerate states. Replacing the matrix elements of the dipole-moment operator of the atom $A$ in accord with (5), we have

$$V=\frac{BE_0}{R^3}=\left[(\mathbf{n}_1\mathbf{n}_2)-\frac{3(\mathbf{n}_1\mathbf{R})(\mathbf{n}_2\mathbf{R})}{R^2}\right](\mathbf{n}_2\mathbf{e})\frac{B_0'E_0}{R^3}, \tag{12}$$

where

$$B_0'=d_{12}^{(1)}d_{23}^{(2)}d_{31}^{(2)}/2(2j_2+1)\hbar\delta(2j_1+1)^{1/2};$$

$\mathbf{n}_1$ and $\mathbf{n}_2$ are unit vectors pertaining to atoms $B$ and $A$, respectively, and are defined in (5).

The system (8) with interaction operator (12) can also be investigated on the basis of the procedure developed in Ref. 13. It must be borne in mind that the operator $V$ contains now an additional factor $(\mathbf{n}_2\cdot\mathbf{e})$, and the expression for the effective cross section should be averaged also over the direction of the vector $\mathbf{e}$. Using the results of Ref. 13 and averaging additionally over

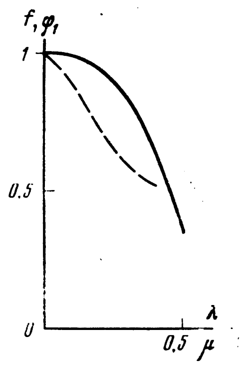


FIG. 2. Radiative-collision line shape in the strong-field approximation. Solid curve—the function $f(\lambda)$, dashed—the function $\varphi_1(\mu)$.

the directions of the vector $\mathbf{e}$, we obtain

$$\sigma(\lambda)=\frac{\pi^3B_0E_0}{16v}\left\{1-\frac{16}{27}\lambda'^2[\ln(\lambda'^2)+5.58]\right\}, \tag{13}$$

where $\lambda'=2(B_0'E_0)|\Delta\omega|/\pi v^{3/2}$ and the condition $\lambda' \ll 1$ must be satisfied.

Equation (13) has exactly the same structure as (11), and differs only in the numerical coefficients. The frequency dependence of the cross sections (11) and (13) determines the line shape of the radiative transition near resonance. It is shown for the transition $0 \to 0$ in Fig. 2, where $f(\lambda)$ denotes the function $\sigma(\lambda)/\sigma(0)$. It is seen that in this approximation the line shape is symmetrical about the resonance shifted by the Stark effect [see (9)]. A characteristic feature of the function $f(\lambda)$ is that there is no maximum at $\lambda=0$. This line shape is due to the form of the interaction operator $V$ and to its angular dependence. If we use in lieu of the operator $V$ its angle-averaged value, i.e., a scalar operator in the form $C/R^3$, where $C$ is a constant (the quadratic combination are naturally averaged out), then the effective cross section has a maximum at $\lambda=0$. If the intrinsic angular momenta of the considered levels are not large, then the use of the quasiclassical transformation (5) introduces errors in the numerical values of the constants in the expressions for the effective cross sections (11) and (13), but the common structure of these expressions is apparently preserved. This is indicated, in particular, by the agreement between expressions (11) and (13), as well as by the fact that in Ref. 8, where the quasiclassical transformation (5) was not used, an expression was obtained for the effective cross section of the radiative transition in a form similar to (11) and (13).

The effective cross sections (11) and (13) depend on the relative velocity $v$ of the atoms. If the excitation-transfer process is observed in a gas phase at thermal equilibrium, then the effective cross section (11) or (13), or more accurately the quantities $\sigma v$ that determine the level population, must be averaged over the Maxwellian distribution. Such an averaging, however, cannot be carried out consistently, since expressions (11) and (13) are valid only at $\lambda \ll 1$, and the parameter $\lambda$ itself depends on the relative velocity. If, however, the value of $\lambda$ corresponding to the maximum of the Maxwellian distribution function is small, then such an averaging can be carried out approximately, recognizing that the effective cross section must decrease rapidly with increasing resonance effect, i.e., at $\lambda > 1$. Therefore, assuming approximately that expressions (11) and (13) are valid up to $\lambda \sim 1$ and that $\sigma = 0$ at $\lambda > 1$, we obtain for the $0 \to 0$ transition

$$\langle\sigma v\rangle = 0.125\pi^3 B_0 E_0 \varphi_1(\mu), \tag{14}$$

where

$$\varphi_1(\mu) = 1 + \mu^2(1.28\ln^2(\mu) + 9.38\ln(\mu) + 4.98),$$
$$\mu = 2(B_0 E_0)^{1/2}|\Delta\omega|/\pi u^{3/2},$$

and for a transition between degenerate states we have

$$\langle\sigma v\rangle = 0.1\pi^3 B_0' E_0 \varphi_2(\mu), \tag{15}$$

where

$$\varphi_2(\mu) = 1 + \mu^2(1.88\ln^2(\mu) + 12.1\ln(\mu) + 8.52),$$
$$\mu = 2(B_0' E_0)^{1/2}|\Delta\omega|/\pi u^{3/2},$$

$u = (2kT/M)^{1/2}$, $M$ is the reduced mass of the atoms, and $T$ is the temperature of the system. Expressions (14) and (15) are valid at $\mu \ll 1$. Thus, radiative excitation transfer line shape averaged over the Maxwellian distribution, the function $\varphi_1(\mu)$, is shown in Fig. 2 dashed. As already noted earlier (see Ref. 2), the radiative-transition cross section is proportional in the strong-field approximation to the amplitude $E_0$ of the field, rather than to its strength.

## 2. WEAK-FIELD APPROXIMATION

In the weak-field case, when an inequality inverse to (7) is satisfied, the system (3) can be solved by perturbation theory, assuming $V$ to be small. We then obtain for the transition probability

$$|a_2(\infty)|^2 = \left|\int_{-\infty}^{\infty} dt V(t) \exp\left\{-i\left[\Delta\omega t + \int_0^t \varkappa(t')dt'\right]\right\}\right|^2, \tag{16}$$

where $\varkappa(t) = (C_1 - C_2)/R^6$. We confine ourselves in this case to the $0 \to 0$ transition. $V(t)$ is then defined by expression (10). Replacing in the general expression for the difference $C_1 - C_2$ (Ref. 2) the true matrix elements of the dipole-moment operator by their quasiclassical expression (5) and summing over intermediate states of the atom $A$ that differ in the value of the magnetic quantum number, we obtain

$$C_1 - C_2 = C_6[1 + 3(\mathbf{nR})^2/R^2], \tag{17}$$

where

$$C_6 = -|d_{12}^{(1)} d_{13}^{(2)}|^2/3(2j+1)\hbar\delta.$$

After substituting (16) in (6) and averaging over the directions of the vector $\mathbf{e}$, numerical integration yielded the effective cross section $\sigma$ as a function of the frequency $\omega$.

To determine the singularities of the behavior of the cross section near resonance, we expand expression (16), assuming $\Delta\omega$ to be small. We retain the quantities that can contribute to the transition probability terms proportional to $\Delta\omega^2$:

$$|a_2(\infty,\Delta\omega)|^2 - |a_2(\infty,\Delta\omega=0)|^2 = -\Delta\omega \int_{-\infty}^{\infty} dt_1 \int_{-\infty}^{\infty} dt_2 V_1 V_2 |t_2 - t_1|$$
$$\times \sin\left(\int_{t_1}^{t_2} \varkappa(t')dt'\right) + \int_{-\infty}^{\infty} dt_1 \int_{-\infty}^{\infty} dt_2 V_1 V_2 \sin(\Delta\omega t_1)\sin(\Delta\omega t_2)\cos\left(\int_{t_1}^{t_2} \varkappa(t)dt\right)$$
$$-\Delta\omega^2 \int_{-\infty}^{\infty} dt_2 V_2 \int_{-\infty}^{\infty} dt_1 V_1 t_1^2 \left[\cos\left(\int_{t_1}^{t_2} \varkappa(t)dt\right) - \cos\left(\int_{\infty\,\mathrm{sign}(t_1)}^{t_2} \varkappa(t)dt\right)\right]$$
$$+2\int_{-\infty}^{\infty} dt V \left[\cos\left(\int_{-\infty}^{t} \varkappa(t)dt\right)\theta_- + \cos\left(\int_{\infty}^{t} \varkappa(t)dt\right)\theta_+\right], \tag{18}$$

where

$$\theta_\pm = \int_0^{\infty} dt V(\pm t)[\cos(\Delta\omega t) - 1].$$

We substitute in expression (6) for the cross section the quantity $|a_2(\infty,\Delta\omega)|^2$ from (18) and average over the direction of the electric-field vector $\mathbf{e}$ relative to the collision plane. Using the smallness of the resonance defect $\Delta\omega$, we integrate with respect to the impact parameter $\rho$ and obtain an expression for the excitation-transfer cross section in the form

$$\sigma(\lambda) = \frac{\rho_0^4}{\rho_B^2}\left\{4{,}9 - \frac{2\pi}{15}\left\langle \lambda\,\mathrm{sign}(C_6)\,1.425 \int_{-1}^{1} d\xi_1 \int_{-1}^{1} d\xi_2 \left|\frac{\xi_2}{(1-\xi_2^2)^{1/2}}\right.\right.\right.$$
$$\left. - \frac{\xi_1}{(1-\xi_1^2)^{1/2}}\right| h|G(\xi_2,\xi_1)|^{-1/5} + \lambda^2\left[\ln|\lambda|\left(\int_{-1}^{1} d\xi[\varphi\ln(|G(\xi,1)G(\xi,-1)|)\right.\right.$$
$$\left.+10\chi] + 13.3\right) + \int_{-1}^{1} d\xi_1 \int_{-1}^{1} d\xi_2 h\left(\frac{\xi_1\xi_2}{(1-\xi_1^2)^{1/2}(1-\xi_2^2)^{1/2}}\ln(|G(\xi_2,\xi_1)|)\right.$$
$$\left. + \xi_2^2 \ln\left(\left|\frac{G(\xi_2,\mathrm{sign}(\xi_1))}{G(\xi_2,\xi_1)}\right|\right)\right) + \int_{-1}^{1} d\xi[0.1\varphi(\ln^2(|G(\xi,-1)|)$$
$$+\ln^2(|G(\xi,1)|)) + (\chi + 0.19\varphi)\ln(|G(\xi,-1)G(\xi,1)|) + 1.92\chi$$
$$\left.\left.\left. + 4\tau\ln(|G(\xi,1)/G(\xi,-1)|) + 0.51\right]\right\rangle\right\}, \tag{19}$$

where

$$h = 1 - 3(\mathbf{R}_1\mathbf{n})^2 - 3(\mathbf{R}_2\mathbf{n})^2 + 9(\mathbf{R}_1\mathbf{n})(\mathbf{R}_2\mathbf{n})(\mathbf{R}_1\mathbf{R}_2),$$
$$\varphi = 1 - 3(\mathbf{Rn})^2 - 3n_x^2 + 9(\mathbf{Rn})\xi n_x,$$
$$\chi = n_y^2 - n_x^2 - 3(\mathbf{Rn})[n_y(1-\xi^2)^{1/2} - n_x\xi],$$
$$\tau = n_x n_y - 1.5(\mathbf{Rn})[n_x(1-\xi^2)^{1/2} + n_y\xi],$$
$$R_x = \xi, \quad R_y = (1-\xi^2)^{1/2}.$$

The vectors $\mathbf{R}_1$ and $\mathbf{R}_2$ lie in the same plane $xy$. Averaging over the directions of the vector $\mathbf{n}$ is designated by the angle brackets. As before, $\lambda$ denotes the small parameter of the problem. In the weak-field approximation we have $\lambda = \Delta\omega\rho_B/v$, where $\rho_B = (C_6/v)^{1/5}$, $\rho_0 = (E_0B_0/v)^{1/2}$. The function $G(\xi_2, \xi_1)$ is of the form

$$G(\xi_2,\xi_1) = G(\xi_2) - G(\xi_1),$$
$$G(\xi) = 0.5(n_y^2 - n_x^2)\xi(1-\xi^2)^{5/2} + 0.25(1 + 0.5n_x^2 + 2.5n_y^2)$$
$$\times\{\xi(1-\xi^2)^{3/2} + 1.5\xi(1-\xi^2)^{1/2} + 1.5\,\mathrm{arctg}[\xi/(1-\xi^2)^{1/2}]\} - n_x n_y(1-\xi^2)^3.$$

The integrals in (19) were calculated with a computer. The final effective cross section $\sigma(\lambda)$ is given by

$$\sigma(\lambda) = 4.9E_0^2B_0^2C_6^{-2/5}v^{-8/5}\{1 - \mathrm{sign}(C_6)\lambda\cdot 0.11 - \lambda^2[0.35\ln(|\lambda|) + 0.62]\}, \tag{20}$$

where $|\lambda| \ll 1$. A characteristic feature of this cross section is that even near resonance it is no longer symmetric, and its maximum is shifted in a direction determined by the sign of $C_6$, namely towards the less

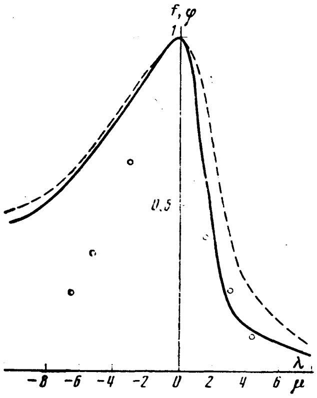


FIG. 3. Radiative-collision line shape in the weak-field approximation. Solid curve—the function $f(\lambda)$, dashed—the function $\varphi(\mu)$, points—experimental.

steep (power-law) decrease at large $|\lambda|$. The shift of the maximum of the cross section and the asymmetry of the line shape are due to van der Waals interaction of the atoms. We note that a similar contour-asymmetry effect near resonance takes place also in the usual theory of impact broadening of spectral lines.[15]

The line shape of the radiative transition is determined by the function $f(\lambda)=\sigma(\lambda)/\sigma(0)$. This function is shown graphically in Fig. 3 at $C_6>0$. At small $|\lambda|$ it is determined by (20), and at large $|\lambda|$ it is obtained by numerical integration. The line shape of the radiative collision, after averaging over the Maxwellian distribution, will be characterized by the function $\varphi(\mu)=\langle\sigma v\rangle/\langle\sigma(0)u\rangle$. At $|\mu|\ll 1$ this function takes the form

$$\begin{aligned}\varphi(\mu)=&1-\mathrm{sign}(C_6)0.17\mu\\&+\mu^2\{0.31\ln^2(|\mu|)\\&+1.34\ln(|\mu|)\\&+0.63+\mathrm{sign}(C_6)0.2\}.\end{aligned}\tag{21}$$

The function $\varphi(\mu)$ is shown in Fig. 3 by the dashed line.

In the experiments of Refs. 10 and 11 (see also Ref. 7) they investigated the excitation transfer from strontium to calcium atoms:

$$\mathrm{Sr}(5p^1p^0)+\mathrm{Ca}(4s^2\,{}^1s)+\hbar\omega(4977\,\text{Å})\to\mathrm{Sr}(5s^2\,{}^1s)+\mathrm{Ca}(4p^2\,{}^1s).$$

Thus, the transition $0\to 0$ is realized for the calcium atoms. For this transition, according to the estimates of Ref. 7, $C_6=4.2\cdot 10^7\ \mathrm{cm^{-1}}\cdot\text{Å}^6$ (our definition of the constants differs in sign from that used in Ref. 7). At an irradiation intensity on the order of $5\times 10^5\ \mathrm{W/cm^2}$, used in the experiment, and at a temperature 300 K, an inequality inverse to (7) is certainly satisfied, i.e., the weak field approximation is valid. The corresponding experimental data are marked in Fig. 3 as functions of the dimensionless parameter $\lambda$. In the calculation of the parameter $\lambda$ we used the constant $C_6$ indicated above. It is seen that the calculated curve accounts well for the rapidly decreasing (exponential) line wing. The observed shift of the maximum is determined by expression (21). Since the dynamic Stark effect is negligibly small at the electromagnetic field intensity used in the experiment (the shift is of the order of $10^{-11}\ \mathrm{cm^{-1}}$), the observed shift is due entirely to the interaction of the atoms. The experimental data show also a decrease faster than calculated in the slowly decreasing (power-law) line wing. This is apparently due to the need for taking into account, for this frequency region, a more realistic form of the interaction potential of the atoms, i.e., refinement of the employed long-range part of the potential, which plays the principal role in the region of the maximum of the cross section.

Experiment[12] revealed excitation transfer from europium atoms (states ${}^8P_{9/2,7/2,5/2}$) to strontium atoms (transition ${}^1S_0\to{}^1D_2$). The irradiation intensity was $10^6$ $\mathrm{W/cm^2}$, and the temperature was 1200 K, so that the weak-field approximation was again satisfied in this case. The experimentally observed line also took the form of an asymmetric curve. In accord with (21), the sign of shift of the maximum and the character of the asymmetry are determined by the sign of $C_6$ (by the sign of $\delta$).


[1]S. I. Yakovlenko, Kvantovaya Elektron. (Moscow) 5, 259 (1978) [Sov. J. Quantum Electron. 8, 442 (1978)].
[2]D. I. Gudzenko and S. I. Yakovlenko, Zh. Eksp. Teor. Fiz. 62, 1686 (1972) [Sov. Phys. JETP 35, 877 (1972)].
[3]V. L. Lisitsa and S. I. Yakovlenko, Zh. Eksp. Teor. Fiz. 66, 1550 (1974) [Sov. Phys. JETP 39, 759 (1974)].
[4]S. Geltman, J. Phys. B 9, L569 (1976).
[5]P. L. Knight, J. Phys. B 10, L195 (1977).
[6]A. Gallagher and T. Holstein, Phys. Rev. A16, 2413 (1977).
[7]S. E. Harris and J. C. White, IEEE J. Quantum Electron. 13, 972 (1977).
[8]D. S. Bakaev and Yu. A. Vdovin, Opt. Spektrosk. 44, 651 (1978).
[9]I. I. Ostroukhova, B. M. Smirnov, and G. V. Shlyapnikov, Zh. Eksp. Teor. Fiz. 73, 166 (1978) [Sov. Phys. JETP 46, 86 (1978)].
[10]D. B. Lidow, R. W. Falcone, J. E. Young, and S. E. Harris, Phys. Rev. Lett. 36, 462 (1977); 37, 1590(E) (1976).
[11]R. W. Falcone, W. R. Green, J. C. White, J. E. Young, and S. E. Harris, Phys. Rev. A15, 1333 (1977).
[12]Ph. Cahurac and P. E. Toschek, Phys. Rev. Lett. 40, 1087 (1978).
[13]Yu. A. Vdovin and V. M. Galitskiĭ, Zh. Eksp. Teor. Fiz. 69, 103 (1975) [Sov. Phys. JETP 42, 51 (1975)].
[14]L. D. Landau and E. M. Lifshitz, Kvantovaya mekhanika (Quantum Mechanics), Fizmatgiz, 1963, p. 121 [Pergamon].
[15]J. Szudy and W. Baylis, J. Quant. Spectrosc. Radiat. Transfer 17, 681 (1977).


Translated by J. G. Adashko