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Atomic collisions in a strong electromagnetic field

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It is shown that the long-range part of the interatomic interaction is significantly altered in a strong electromagnetic field. Instead of the Van der Waals law the asymptotic potential is described by a dipole-dipole R^{-3} law. The impact line broadening and line shift in a strong nonresonant field are calculated. The possibility of formation of bound states of two atoms in a strong light field is discussed.

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1. The effect of a strong electromagnetic field on the characteristics of inelastic atomic and molecular collisions has been investigated in a number of recent papers.^[1-3] The main effect studied in these papers is that the energy of the strong field photon cancels the resonance defect of the colliding systems. As a result the inelastic transition probabilities are significantly enhanced.

In the present paper we wish to point out that a strong light field changes also the interaction of colliding atoms at large distances. This affects the characteristics of the elastic scattering and, consequently, the collision width and shift of spectral lines, which can be verified most simply by the following example. Let two different atoms in the s -state collide in a nonresonant, linearly polarized electromagnetic field \mathcal{E} of frequency ω . Dipole moments $\alpha_A(\omega)\mathcal{E}_0 \cos\omega t$ and $\alpha_B(\omega)\mathcal{E}_0 \cos\omega t$ are then induced in the atoms, where $\alpha_{A,B}$ are the atomic polarizabilities (it is assumed that the atoms are separated from each other by a distance R , much smaller than the wavelength of light). The long-range part of the atomic interaction, averaged over a period of the field, is obviously

$$U = \frac{\alpha_A \alpha_B \mathcal{E}_0^2}{2R^3} (1 - 3 \cos^2 \theta), \quad (1)$$

where θ is the angle between the vectors \mathcal{E}_0 and \mathbf{R} . Without a field the atomic interaction is caused by Van der Waals forces, i. e., $U_0 = c_6 R^{-6}$. The interaction (1), thus, becomes dominant if $R > (c_6 / \alpha_A \alpha_B \mathcal{E}_0^2)^{1/2}$.

We assume that atom A is in the ground state, and atom B is excited. For an estimate one can take $\alpha_A \sim 10$ a.u., $\alpha_B \sim 10^2$ a.u., and the constant c_6 can be estimated by the Slater-Kirkwood equation^[4]: $c_6 \sim 10^2$ a.u.

In fields $\mathcal{E}_0 \sim 10^{-2}$ a.u. the interaction (1) exceeds the Van der Waals one at $R > 10$ a.u.

2. The modified interaction is quite important at significantly lower fields if the field frequency is close to the resonance frequency of one of the atoms (atom A , for example). We treat this problem in detail. Let the field be in resonance with the transition between the ground and first excited states of atom A . This atom can be then described by a density matrix for a two-level system, including spontaneous transitions into the ground state. The relation between the duration of atomic flight through the light beam T and the excited-state lifetime $1/\gamma$ is an important factor in this case. In real situations the intensity distribution of the field over the cross section of the beam has a characteristic dimension, therefore the duration of field involvement is of the order of the time of flight through the beam. For thermal velocities in a beam of diameter $\sim 0.1-1$ cm the condition $\gamma T \gg 1$ can always be assumed to be satisfied. This implies that the two-level system is described only by the induced solution of the equations of motion for the density matrix. As is well-known, this solution is (see, for example,^[5])

$$\rho_{12} = -\frac{V}{2} (\rho_{11} - \rho_{22}) \frac{\varepsilon - i\gamma_{12}}{\varepsilon^2 + \gamma_{12}^2} e^{-i\omega t}, \quad (2)$$

$$\rho_{11} - \rho_{22} = \frac{\varepsilon^2 + \gamma_{12}^2}{\varepsilon^2 + \gamma_{12}^2 + \gamma_{12} V^2 / \gamma}, \quad \rho_{11} + \rho_{12} = 1.$$

Here $v = \mathcal{E}_0 d_A^0$, d_A^0 is the transition dipole moment, γ_{12} is the luminescence line width of atom A , and ε is the field frequency, measured from the line center of the atomic transition ω_0 .

Generally speaking, γ and γ_{12} in Eq. (2) can differ appreciably from each other, since γ contains, besides

radiative damping, contributions of inelastic collisions, while γ_{12} contains contributions of elastic collisions as well. In what follows we consider the range of pressures in which the contributions to γ and γ_{12} by collisions of A atoms with each other can be neglected. These collisions between identical atoms are described by a dipole-dipole mechanism even in the absence of a light field. In this case the smallness criterion of the collision width as compared to the radiative one is of the well-known form $N_A \lambda^3 \ll 1$, where N_A is the density of gas A , and $2\pi\lambda$ is the transition wavelength. For the visible region this condition restricts the density of gas A to 10^{15} cm^{-3} . For higher densities γ_{12} depends on V , and Eq. (2) with a constant γ_{12} is no longer valid (in this connection see Lisitsa and Yakovlenko^[3]).

The average dipole moment of atom A in the presence of a field is expressed in terms of the nondiagonal element of the density matrix:

$$d_A(t) = 2d_A^0 \text{Re } \rho_{12} = \frac{d_A^0 V (-\varepsilon \cos \omega t + \gamma_{12} \sin \omega t)}{\varepsilon^2 + \gamma_{12}^2 + \gamma_{12} V^2 / \gamma}. \quad (3)$$

As far as atom B is concerned the field is nonresonant, therefore its dipole moment is determined, as usual, by the polarizability $d_B = \alpha_B \mathcal{E}_0 \cos \omega t$. We thus obtain for the time-averaged atomic interaction the dipole-dipole law:

$$U_3 = c_3(V) R^{-3} (1 - 3 \cos^2 \theta), \quad c_3(V) = -\frac{\alpha_B}{2} \frac{\varepsilon V^2}{\varepsilon^2 + \gamma_{12}^2 + \gamma_{12} V^2 / \gamma}. \quad (4)$$

It follows from (3) and (4) that the quantity $|d_A|$, and consequently also the interaction energy, reaches a maximum value at frequencies

$$\varepsilon_{\text{opt}} = \pm (\gamma_{12}^2 + \gamma_{12} V^2 / \gamma)^{1/2}.$$

The constant c_3 in the extremum points at $V^2 \gg \gamma \gamma_{12}$ is linear in the field, unlike (1), and is of the order of $c_3(V) \sim \alpha_B d_A^0 \mathcal{E}_0$. At exact resonance ($\varepsilon = 0$), $c_3(V)$ vanishes as a consequence of the adiabatic atomic interaction with the field in the range of anomalous dispersion.

In the presence of an electromagnetic field the Van der Waals part of the interaction is also changed. As is well-known, it is obtained as a result of including the change of the wave function of first order in the dipole-dipole interaction. The distinctive feature of this case consists of the fact that the resonant field singles out only two levels of atom A . The derivation in the Appendix leads to the physically clear result:

$$c_6(V) = \sum_{m \neq n} | \langle d_A^0 d_B^0 - 3R^{-2} (d_A^0 \mathbf{R})(d_B^0 \mathbf{R}) \rangle_{1, n, 2m} |^2 \left(\frac{\rho_{11}}{\omega_{mn} + \omega} - \frac{\rho_{22}}{\omega_{mn} - \omega} \right), \quad (5)$$

i. e., U_6 consists of two parts corresponding to two levels of atom B with their populations and Van der Waals constants. Unlike the dipole-dipole constant $c_3(V)$, the quantity $c_6(V)$ does not depend so critically on the field and on the frequency deviation from resonance. In order of magnitude the quantity c_6 is approximately equal to the Van der Waals constant without a field. Another difference is that c_6 is independent of the angle θ between the vectors \mathcal{E}_0 and \mathbf{R} .

3. We calculate now the collision line broadening and line shift of atom B due to the adiabatic loss of phase synchronism caused by the interactions (4) and (5). In the linear trajectory approximation $\mathbf{R} = \mathbf{r} + \mathbf{v}t$ the total phase shift η is determined by the equation

$$\eta(\mathbf{r}, \mathbf{v}) = -\frac{3\pi}{8} \frac{c_6}{v r^3} - \frac{2c_3}{v r^2} \sin^2 \psi \cos 2\varphi, \quad (6)$$

where ψ is the angle between the relative velocity of the atoms \mathbf{v} and the direction of the field \mathcal{E}_0 , φ is the azimuthal angle in the plane perpendicular to \mathbf{v} , and \mathbf{r} is the impact parameter.

The cross section σ'' determining the line shift $\Delta\omega = N_A v \sigma''$, where N_A is the density of atoms A , equals (see, for example, [6])

$$\sigma'' = \frac{1}{2} \int \sin \eta r dr d\varphi \sin \psi d\psi = -2\pi \int_0^\infty r dr \int_0^\pi dx J_0 \left[\frac{2c_3}{r^2 v} (1-x^2) \right] \sin \left(\frac{3\pi c_6}{8v r^3} \right) \quad (7)$$

(the integration with respect to ψ in (7) corresponds to averaging over the relative-velocity directions of the atoms). As seen from (6) and (7), there exists a characteristic value of the constant c_3 (and, consequently, of the field V and its frequency ε), at which the contributions of both terms to the phase η are equal. We introduce a parameter λ defined by the equation

$$\lambda = 2(8/3\pi)^{1/2} c_3 c_6^{-1/2} v^{-3/2} \quad (8)$$

and express the asymptotic value of σ'' in its terms. In the weak-field region ($|\lambda| \ll 1$) we obtain

$$\frac{\sigma''(V)}{\sigma''(0)} = 1 - \frac{\sin(2\pi/5)}{3\pi} \Gamma^2(\nu/5) \lambda^2. \quad (9)$$

In the opposite limiting case

$$\frac{\sigma''(V)}{\sigma''(0)} = \frac{\Gamma^2(\nu/5) \cos(\pi/5)}{2\sqrt{2} \Gamma(\nu/5) \cos(2\pi/5)} |\lambda|^{-1}. \quad (10)$$

The Van der Waals line shift is negative in the absence of a field (see Eqs. 5 and 6). The dipole-dipole interaction in a strong electromagnetic field gives a shift of opposite sign. Therefore, the total collision line shift decreases in absolute value with increasing field and tends to zero as $1/V$ in the strong-field limit when the field frequency is tuned in such a manner that it is always near the critical values¹⁾ $\varepsilon = \varepsilon_{\text{max}}$. The characteristic value of the field corresponding to $\lambda \sim 1$ is approximately 10^{-4} a. u. For a flux density 10^6 W/cm^2 , atom velocity $\sim 10^4 \text{ cm/sec}$, and pressure 10^{-2} Torr the line shift due to the dipole-dipole interaction is of the order of 10^2 Hz .

The following results are obtained for the cross section σ' of the collision line broadening:

$$\begin{aligned} \sigma'(V)/\sigma'(0) &= 1 + \frac{\sin(2\pi/5)}{3\pi} \Gamma^2(\nu/5) \lambda^2, \quad |\lambda| \ll 1, \\ \sigma'(V)/\sigma'(0) &= \frac{4\pi}{3} \Gamma^2(\nu/5) \sin \frac{\pi}{5} |\lambda|, \quad |\lambda| \gg 1. \end{aligned} \quad (11)$$

For the same pressure, flux density, and atom velocity values as given above the field-dependent part of the

collision width is of the same order of magnitude as the shift, and is approximately 300 Hz.

4. The discussion above refers to the overwhelming majority of atoms whose kinetic energy is of the order of the thermal energy. A small fraction of atoms with kinetic energy of relative motion $\lesssim |U_3|$ can form a bound molecular complex AB^* . We have in mind the range of binding energies E in which bound states are mostly formed by the dipole-dipole part of the full interaction. Naturally, bound states are possible even without a field, owing to the Van der Waals forces. The corresponding levels are deeper in energy, i. e., they occur at higher values of $|E|$ ($E < 0$).

The E value separating these two regions can be found by calculating the density of bound levels $\nu(E)$ in the quasiclassical approximation (for this it is necessary that the total number of levels be large):

$$\nu(E) = \frac{(2\mu)^{3/2}}{2(2\pi)^2} \int d\mathbf{R} (E - c_3 R^{-3} (1 - 3 \cos^2 \theta) + c_6 R^{-6} - AR^{-12})^{1/2}, \quad (12)$$

where μ is the reduced mass of atoms A and B . The integration is performed over the region of \mathbf{R} in which the radicand is positive. To remove the divergence of the integral as $R \rightarrow 0$, we have included in (12) a term describing repulsion at small distances in the traditional form AR^{-12} ($A \sim 10^6$ a. u.). As follows from (4), the sign of c_3 depends on the sign of the frequency deviation from resonance ϵ . Therefore, dipole-dipole attraction occurs in the angular region $\theta_0 < \theta < \pi - \theta_0$ for $\epsilon > 0$, and in the regions $0 < \theta < \theta_0$, $\pi - \theta_0 < \theta < \pi$ for $\epsilon < 0$, where $\theta_0 = \arccos(1/\sqrt{3})$. An analysis of (12) shows that at low energies $|E| \ll c_3^2/4c_6 \ll c_3^4/3A^{-1/3}$ the last two terms under the square root sign can be neglected (in this case the region $R \sim |c_3/E|^{1/3}$ provides the main contribution to the integral).

Thus, at $|E| \ll c_3^2/4c_6$, the bound states are of dipole-dipole origin, and the density of levels (for any sign of ϵ) is given by:

$$\nu(E) = \frac{\sqrt{6}}{27} \mu^{3/2} |c_3| |E|^{-3/2}. \quad (13)$$

A factor $\frac{1}{2}$ was introduced in (12) and (13) to take into account the twofold level degeneracy with respect to the sign of the momentum z -projection. Requiring that the number of levels described by Eq. (13) be large, we arrive at the condition $\mu^{3/2} c_3^2 |E|^{-3/2} \gg 1$. This inequality implies that at the extremal frequencies the flux density should be much higher than 10^8 W/cm² for $\mu \sim 20$ proton masses. It is significant that the position and number of the bound-state energy levels depend on the light field intensity, i. e., unusual nonlinear optical effects are generated.

The formation of bound AB^* complexes should be manifested in the structure of the absorption spectrum of a weak signal that is resonant with the transitions of atom B . Satellites of molecular complexes should appear against the background of the absorption lines contour, this being the analog of vibrational-rotational structure of molecules. Unlike ordinary molecules, the AB^* system considered here is characterized by a low

binding energy and by a large equilibrium internuclear distance. As a consequence the coupling between the nuclear and electronic degrees of freedom is weak. Therefore, the transition dipole moment in the absorption coefficient $K(\omega)$ can be assumed to be independent of the molecular quantum numbers:

$$K(\omega) = -\frac{\pi\omega}{4} |d_{ab}|^2 \sum_{\mu,\nu} |\langle \mu | \nu \rangle|^2 (N_{a\mu} - N_{b\nu}) \delta(\omega - \omega_{a\mu, b\nu}). \quad (14)$$

Here a and b number the upper and lower electronic terms, μ and ν are the quantum numbers with respect to the nuclear degrees of freedom for the terms a and b , respectively, $\langle \mu | \nu \rangle$ is the overlap integral of the nuclear wave functions, and $N_{a\mu}$ and $N_{b\nu}$ are the number of atoms in states ($a\mu$) and ($b\nu$). The summation in (14) extends over both the discrete and continuous spectrum. The presence of the δ -function in (14) presupposes that the intervals of molecular structure are large in comparison with the width of the electronic term (an estimate is given below).

It is easily realized that three groups of terms can be distinguished in the contour $K(\omega)$: 1) transitions between continuum states of both electronic terms; 2) between discrete states; 3) between discrete and continuum states. The basic features of the first type of transitions were considered above. Qualitative conclusions may be drawn concerning the two other groups: group 2) contributes a structure of δ -shape peaks to the spectrum, and group 3) provides step-like singularities.

We estimate now the term molecular-structure parameters that determine the scales of the indicated singularities of the spectrum. The level density (13) corresponds in the sense of its E dependence to a one-dimensional potential well. Numbering all levels downward in succession, i. e., without account of the quantum number of the z -projection of the angular momentum, it follows from (13) that

$$E_n = 3^3 (n+q)^2 / 8\mu^3 c_3^2, \quad 0 < q < 1, \quad n=0, 1, 2, \dots \quad (15)$$

This equation gives the relative arrangement of molecular structure terms in the quasiclassical approximation. The characteristic frequency determining the intervals between molecular satellites of the electronic transition is of the order of $10^2 \mu^{-3} (\alpha_B d_A^0 \mathcal{E}_0)^{-2}$ and amounts (at the μ and α_B values used above) to $\sim 10^8$ Hz for a flux density $\sim 10^9$ W/cm². Thus, the strong-field-dependent dipole-dipole interaction provides a structure located much closer to the line center than the usual Van der Waals interaction ($\sim 10^{11}$ Hz).

It should be pointed out, however, that experimental detection of this structure is, obviously, rather difficult due to the small number of AB^* pairs. Their density N_{AB} equals $N_0 N_A N_B / N_T$, where N_A and N_B are the densities of the atoms A and B , $N_T = (\mu k T / 2\pi \hbar^2)^{3/2}$, T is the temperature, and N_0 is the total number of states with negative energy: $N_0 = \int \nu(E) dE$. For $T \sim 300$ K, $N_A \sim 10^{14}$ cm⁻³, and $N_0 \sim 10^2$ the relative number of pairs N_{AB}/N_B is only 10^{-10} .

APPENDIX

We start from the Schrödinger equation for two interacting atoms A and B in a linearly polarized field $\mathcal{E} = \mathcal{E}_0 \cos \omega t$. The Hamiltonian of this system is

$$H = H_A^0 + H_B^0 - (d_A^0 + d_B^0) \mathcal{E} + U. \quad (\text{A.1})$$

Here $H_{A,B}^0$ are the Hamiltonians of atoms A and B , and U is their interaction energy, which in the following is assumed to be

$$U = \frac{d_A^0 d_B^0}{R^3} - 3 \frac{(d_A^0 R)(d_B^0 R)}{R^5}. \quad (\text{A.2})$$

The total wave function is represented in the form of a series in the wave functions of the individual atoms, including the interaction of atoms with the field:

$$\Psi = \sum_{\alpha, n} C_{n\alpha} \psi_\alpha \varphi_n. \quad (\text{A.3})$$

The Greek indices (α) denote states of atom A , and the Latin (n) states of atom B .

Since atom A is at resonance with the field, it follows that by separating the two resonating levels we find the explicit form of the wave functions $\psi_\alpha (\alpha = \pm)$:

$$\begin{aligned} \psi_+ &= a_1 \psi_1 e^{-iE_1 t} + a_2 \psi_2 e^{-iE_2 t}, \\ \psi_- &= i a_2^* \psi_1 e^{-iE_1 t} - i a_1^* \psi_2 e^{-iE_2 t}, \end{aligned} \quad (\text{A.4})$$

where $\psi_{1,2}$ are the eigenfunctions of the zeroth Hamiltonian H_A^0 , and the coefficients $a_{1,2}$ take into account the interaction with the field (see, for example, [7]). The wave functions φ_n of atom B are found by taking into account the first order correction in the field \mathcal{E} and are given, for example, in [7]. To take into account spontaneous transitions in atom A we assume that a correction describing spontaneous radiation is included in the field \mathcal{E} , and that the coefficients $a_{1,2}$ contain, respectively, random phases. By definition the density matrix ρ_{ij} is an average of the products of coefficients in expansion (A.4) over the random phases, i. e., $\rho_{ij} = \langle a_i^* a_j \rangle$. In this case the normalization $|a_1|^2 + |a_2|^2 = 1$ is retained in the approximation given.

Substituting expansion (A.3) into the Schrödinger equation, we obtain an equation for the coefficients $C_{n\alpha}$. We give, for example, the equation for C_{n+} (the equation for C_{n-} is similar)

$$\begin{aligned} i \left(\frac{\partial}{\partial t} + \frac{P^2}{2\mu} \right) C_{n+} &= - \left\{ a_1^* a_2 \sum_m \frac{V_{nm} U_{1m, 2n} \omega_{mn}}{\omega_{nm}^2 - \omega^2} \cos \omega t + \text{r.c.} \right\} C_{n+} \\ &+ \sum_m C_{m+} e^{i\omega_{mn} t} (a_1^* a_2 U_{1n, 2m} + a_2^* a_1 U_{2n, 1m}) \\ &- i \sum_m C_{m+} (a_1^* e^{i\omega_0 t} U_{1n, 2m} - a_2^* e^{i\omega_0 t}) e^{-i\omega_{nm} t}. \end{aligned} \quad (\text{A.5})$$

Here $V_{nm} = d_{nm}^0 \mathcal{E}_0$. There are two types of terms in this equation: the first term in the curly brackets does not contain the high frequency $\sim \omega_0$, and therefore gives the dipole-dipole interaction (4) when averaged over the high-frequency vibrations and the random phases, since $\langle \langle a_1^* a_2 \cos \omega t \rangle \rangle = \langle \rho_{12} \cos \omega t \rangle_\omega$. The two remaining terms in (A.5) contain fast oscillations and lead to the Van der Waals interaction. To obtain the explicit form of this interaction we proceed as follows. We represent the solution of Eq. (A.5) as a sum of a slowly varying part \bar{C}_{n+} and a quickly oscillating correction δC_{n+} of frequency ω . Corrections of the type

$$\langle \delta C_{m+} a_{1,2}^* a_{2,1} e^{i\omega_{nm} t} \rangle, \quad \langle \delta C_{m-} a_{1,2}^* e^{i(\omega_{nm} \mp \omega_0) t} \rangle, \quad (\text{A.6})$$

to the second and third term appear then in the equation for \bar{C}_{n+} . In calculating the oscillating parts all terms not explicitly containing rapidly oscillating exponents should be neglected. As a result we find

$$\begin{aligned} \delta C_{n+} &\rightarrow - \sum_m \bar{C}_{m+} e^{i\omega_{nm} t} \left(\frac{a_1^* a_2 U_{1n, 2m}}{\omega_{nm} - \omega} + \frac{a_2^* a_1 U_{2n, 1m}}{\omega_{nm} + \omega} \right), \\ \delta C_{n-} &\rightarrow i \sum_m \bar{C}_{m+} e^{i\omega_{nm} t} \left(\frac{a_1^* U_{1n, 2m} e^{-i\omega_0 t}}{\omega_{nm} - \omega_0} - \frac{a_2^* U_{2n, 1m} e^{i\omega_0 t}}{\omega_{nm} + \omega_0} \right) \end{aligned} \quad (\text{A.7})$$

The symbol \rightarrow denotes that only terms giving dominant contributions to the average (A.6) were included in calculating $\delta C_{n\pm}$. Substituting (A.7) and (A.6) in the equation for $\bar{C}_{n\pm}$ and averaging with respect to fast oscillations and random phases with account of the normalization $|a_1|^2 + |a_2|^2 = 1$, we arrive at the Van der Waals interaction (5).

¹We point out that the dynamic Stark shift, not considered here, equals $-\alpha_B \mathcal{E}_0^2/4$ and is independent of pressure.

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