Interaction of atoms in a light field

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The effect of a resonant light field on the interaction between two identical atoms is considered in the limiting cases of large and small distances between the atoms. The interaction potential for atoms at distances greatly exceeding the atomic distance, but much smaller than the resonant wavelength, is considered in the first part of the study in the adiabatic approximation. It is shown that for some atomic states the interaction potential is spherically symmetric and has the shape of a potential well whose parameters depend strongly on the field strength. For a strong field $E \sim 10^6$ V/cm the well depth is ~ 100 K, the radius is ~ 10 Å, and the number of bound states is $\sim 10^2$. For a weak field ~ 1 V/cm the well size becomes comparable with the wavelength and the depth is $\sim 10^{-4}$ K. The lifetime of a quasimolecule in such states may considerably exceed the lifetime of the atom in the excited state. This may be of interest in the spectroscopy of narrow atomic and molecular resonances. The interaction force between two atoms in the wave zone is considered in the second part of the paper. It is shown that it can be regarded as the result of pressure exerted on one atom by light scattered by the other atom. Depending on the field intensity, the atoms may either attract or repulse each other. In a strong field the force obeys the Coulomb law with an effective charge which depends on the field strength and atom velocities. In a gas of finite density the interaction becomes comparable with the light-pressure force. This may be important in studies of the kinetics of atoms in a resonant field.

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1. INTRODUCTION

In this study we consider the interaction of identical atoms located in a resonant light field. We consider the two characteristic limiting cases of small and large distances between the particles relative to the resonant wavelength. It is known that the unexcited atoms interact in the near zone according to the Van der Waals law. The resonant field "switches on" the dipole-dipole interaction. If the field is sufficiently strong the effective interaction potential can differ considerably from the dipole one. These potentials are found in the first part of this study (Sec. 2).

The potential curves for some states have the form of wells in which there can be a fairly large number of bound states. The well parameters depend on the field strength and can vary in a wide range. Thus, the two atoms in the light field can form a molecule whose size is large compared with the size of the atoms and is determined by the applied field. The lifetime of this molecule is determined by the spontaneous emission of the atoms. This effect is not taken into account in this study, however.

Another important approximation is the assumption that the atoms move slowly (the adiabatic approximation). In this sense we are considering the case opposite to that in the theory of atomic collisions in a light field.[1-3] In this theory transitions between the resonance levels during the collision are taken into account and the particle trajectory is assumed to be unperturbed (straight-line). For thermal atoms the kinetic energy is usually considerably greater than the interaction potential at the Weisskopf radius. This condition is violated for low energy atoms. In particular, for bound states the kinetic energy is less than the potential energy. In this case it is natural to use the adiabatic approximation. Therefore, to find the interaction potential we shall drop the atom kinetic energy operator. The correctness of this approximation is discussed below.

We note that the distortion of the oscillator potential of the molecule in the direction of the resonant-field polarization was studied in Ref. 4.

In the second part of this study (Sec. 3) we consider the interaction of the atoms in the wave zone. In this case retardation effects are important and the interaction is nonpotential. It can be viewed as the result of pressure of an atom exerted by the light scattered by the other atom. This pressure can be either positive or negative, that is, the atoms can attract or repulse each other. At large distances the atoms interact in accord with the Coulomb law with an effective charge that depends on the external field. The features of the atomic interaction in a weak and strong external fields are studied in detail.

2. INTERACTIONS OF THE ATOMS IN THE NEAR ZONE

Let us consider the interaction of the atoms at distances $a \ll r \ll 2\pi/k$, where a is the size of the atom and $2\pi/k$ is the light wavelength. Neglecting the spatial dependence, we can write for the field

$$\mathbf{E} \exp[-i(\omega_0+\Delta)t] + \text{c.c.} \qquad (1)$$

where ω_0 is the transition frequency and Δ is a small frequency difference. The field amplitude $\mathbf{E}$ can be assumed real with no loss of generality.

The Hamiltonian of the system of two atoms in the

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adiabatic approximation is

$$H=(\mathbf{d}_1^{+}+\mathbf{d}_2^{+})Ee^{-i\Delta t}+(\mathbf{d}_1\mathbf{d}_2^{+}-3(\mathbf{d}_1\mathbf{n})(\mathbf{d}_2^{+}\mathbf{n}))/r^3+\text{H.c.} \quad (2)$$

where $\mathbf{n}=\mathbf{r}/r$ and $\mathbf{d}_1$ and $\mathbf{d}_2$ are the dipole-moment operators. The first term takes into account the resonant interaction of the atoms with the field and the second corresponds to the dipole-dipole interaction. The problem is to find the eigenvalues $\varepsilon(\mathbf{r})$ of the Hamiltonian as functions of the distance between the atoms. The terms $\varepsilon(\mathbf{r})$ are potentials that determine the relative motion of the atoms.

We consider next the $0-1$ transition in more detail. We introduce the following state amplitudes: ψ_0—both atoms are in an S state, ψ_{i0}—the first atom is in a P_i state and the second is in an S state, ψ_{0i}—the first atom is in an S state and the second is in a P_i state, and ψ_{ij}—the first atom is in a P_i state and the second is in a P_j state. The original equations then have the following form (repeated indices are summed over):

$$\begin{aligned}
(\varepsilon-\hbar\Delta)\psi_0&=dE_i(\psi_{i0}+\psi_{0i}),\\
\varepsilon\psi_{i0}&=d\psi_{ij}E_j+dE_i\psi_0+\varepsilon_0(\delta_{ij}-3n_in_j)\psi_{0j},\\
\varepsilon\psi_{0i}&=dE_j\psi_{ji}+dE_i\psi_0+\varepsilon_0(\delta_{ij}-3n_in_j)\psi_{j0},\\
(\varepsilon+\hbar\Delta)\psi_{ij}&=dE_i\psi_{0j}+dE_j\psi_{i0}.
\end{aligned} \quad (3)$$

where $\varepsilon_0=d^2/r^3$ and d is the dipole-moment matrix element. Since the Hamiltonian (2) is invariant to particle exchange, the system (3) breaks up into two independent systems for the vectors $\psi_{\pm i}=(\psi_{i0}\pm\psi_{0i})/\sqrt{2}$:

$$\varepsilon\psi_+=\frac{2d^2\mathbf{E}(\mathbf{E}\psi_+)}{\varepsilon-\hbar\Delta}+\frac{d^2[\mathbf{E}^2\psi_++\mathbf{E}(\mathbf{E}\psi_+)]}{\varepsilon+\hbar\Delta}+\varepsilon_0(\psi_+-3\mathbf{n}(\mathbf{n}\psi_+)), \quad (4)$$

$$\varepsilon\psi_-=\frac{d^2[\mathbf{E}^2\psi_--\mathbf{E}(\mathbf{E}\psi_-)]}{\varepsilon+\hbar\Delta}-\varepsilon_0(\psi_--3\mathbf{n}(\mathbf{n}\psi_-)). \quad (5)$$

For $\psi_-=0$ and $\psi_+\neq 0$ the dipole moments of the atoms are parallel and for $\psi_+=0$ and $\psi_-\neq 0$ they are antiparallel.

There are two singled-out directions of $\mathbf{E}$ and $\mathbf{r}$ in the problem. The Hamiltonian (2) is invariant to reflection of the vectors $\mathbf{d}_1$ and $\mathbf{d}_2$ in the place of the vectors $\mathbf{E}$ and $\mathbf{r}$. Therefore, each of equations (4) and (5) splits into equations containing only the state vectors which are normal ($\psi_{\pm n}$) or tangential ($\psi_{\pm t}$) relative to the $(\mathbf{E},\mathbf{r})$ plane.

2.1. The antisymmetric case

In the antisymmetric case, when only the vector ψ_{-n} is nonzero, equation (5) gives

$$\begin{aligned}
&(\varepsilon+\varepsilon_0)(\varepsilon+\hbar\Delta)=(\mathbf{dE})^2,\\
&\varepsilon=-{}^1/_2\{(\varepsilon_0+\hbar\Delta)\pm[(\varepsilon_0-\hbar\Delta)^2+4(\mathbf{dE})^2]^{1/2}\}.
\end{aligned} \quad (6)$$

The graph of $\varepsilon(r)$ is given in Fig. 1. The term $\varepsilon(r)$ are spherically symmetric. This is a consequence of the adiabatic approximation, in which $\hat{H}$ is invariant under reflection of the dipoles in the $(\mathbf{E},\mathbf{r})$ plane. Therefore, for slowly varying $\mathbf{r}$ the vector ψ_{-n} is always perpendicular to the radius vector $\mathbf{r}$. For small r the lower branch behaves as $\varepsilon(r)\approx-\varepsilon_0(r)$, which corresponds to the attraction of two antiparallel dipoles perpendicular to the radius vector. Here the upper branch is finite: $\varepsilon(r)\approx-\hbar\Delta$. In this case both atoms are in an excited state, so that the dipole interaction is switched off.

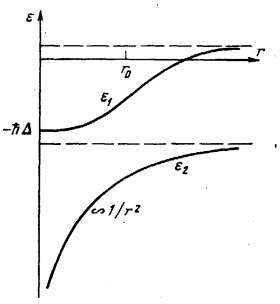

FIG. 1. The functions $\varepsilon(r)$ for the antisymmetric case. $\varepsilon_1(r)\to\frac{1}{2}([(\hbar\Delta)^2+(2dE)^2]^{1/2}-\hbar\Delta)$ at large r.

The upper potential has the form of a well, whose size r_0 is approximately given by the condition $\varepsilon_0(r)\approx[(\hbar\Delta)^2+4(\mathbf{d}\cdot\mathbf{E})^2]^{1/2}$. At this point the dipole potential is comparable to the Stark splitting of the levels. For $r\gg r_0$ the position of the terms is given by the Stark splitting.

There is an essential difference between the two attractive potentials shown in Fig. 1. In the potential $-\varepsilon_0(r)$ the particles fall toward the center. At small r a particle acquires a large velocity, so that the adiabatic approximation can break down. In addition, the integrals of the motion can be violated because of the exchange interaction at small distances.[1)] As a result, the atoms can cross over to the decay potential and the lifetime in the bound state will be very small.

When moving to the upper potential well with finite angular momentum the atoms are always far from each other and have small velocities. Taking dE for the well depth, we estimate the number of bound states in the well at $N=(dEM)^{1/2}r_0\hbar^{-1}$, where M is the atomic mass. In a strong field $E=10^6$ V/cm we have for Na atoms, for example, about 100 K for the well depth, about 10 Å for the well radius, and $N\sim100$. In this case a sizable number of the atoms [of order $(100\text{ K}/T)^{3/2}\sim0.1$, where T is the temperature of the atoms] can be in bound states. In a weak field $E=1$ V/cm the well depth is about 10^{-4} K, the well radius is comparable to the wavelength, and $N\sim10$. The parameters of the potential well as a function of the applied field vary in a wide range.[2)]

The lifetime of the atoms in a bound state is limited by transitions, due to spontaneous emission, to decay states. However, the dipole radiation of atoms having antiparallel dipole moments and located at a distance much smaller than the wavelength is decreased considerably.[6] The lifetime in an antisymmetric bound state can therefore greatly exceed that of the free atom in an excited state.

We now consider the equation for ψ_{-t}. Setting the determinant of (5) equal to zero, we find a fourth-order equation for $\varepsilon(r)$. We restrict ourselves to small distances $r\ll r_0$. Then the roots of (5) are easy to find, either by dropping the first term in the square brackets on the right-hand side or be setting the left-hand side of the equation equal to zero. In the first case we have unbounded solutions $\varepsilon(r)\approx-\varepsilon_0(r)$ and $\varepsilon(r)\approx2\varepsilon_0(r)$, corresponding to the potentials for dipole moments per-

pendicular and parallel to the radius vector. In the second case the solutions at small distances are bounded. Setting $\varepsilon(r) = -\hbar\Delta + \delta\varepsilon(r)$, where $\delta\varepsilon(r)$ is small, we get from (5) the following two solutions:

$$\delta\varepsilon(r)=0, \quad \delta\varepsilon(r)=\frac{(dE)^2}{2\varepsilon_0(r)}(3\cos^2\theta-1), \tag{7}$$

where θ is the angle between the vectors **E** and **r**. The second root changes sign when θ is changed, so there is no well in this case.

2.2. The symmetric case

In the case of parallel dipole moments we have from (4) for ψ_{+n}

$$(\varepsilon-\varepsilon_0)(\varepsilon+\hbar\Delta)=(dE)^2,$$
$$\varepsilon(r)=\tfrac{1}{2}(\varepsilon_0-\hbar\Delta)\pm\tfrac{1}{2}[(\varepsilon_0+\hbar\Delta)^2+4(dE)^2]^{1/2}. \tag{8}$$

The difference from the case described by formula (6) is due to the change of the sign in front of $\varepsilon_0(r)$. Because of this the interaction potentials are repulsive, as seen from Fig. 2.

We shall investigate Eq. (4) for ψ_{+t} likewise only in the limit $r \ll r_0$. Dropping the first and second terms on the right-hand side of (4), we find the roots corresponding to the purely dipole interaction $\varepsilon(r) \approx \varepsilon_0(r)$ and $\varepsilon(r) \approx -2\varepsilon_0(r)$. Setting

$$\varepsilon(r)=-\hbar\Delta+\delta\varepsilon(r), \quad \delta\varepsilon(r)\ll|\hbar\Delta|,$$

and keeping only the second and third terms on the right-hand side of (4), we have

$$\delta\varepsilon(r)=\frac{(dE)^2}{\varepsilon_0(r)}\left(-\frac{3}{4}\sin^2\theta\pm\left[1+\left(\frac{3}{4}\sin^2\theta\right)^2\right]^{1/2}\right). \tag{9}$$

For the plus sign, $\delta\varepsilon$ is positive and increases with increasing r. Thus, the interaction potential of parallel dipoles also has a well of finite depth (not spherically symmetric), whose center is at the origin. Setting

$$\varepsilon(r)=\hbar\Delta+\delta\varepsilon(r), \quad \delta\varepsilon(r)\ll|\hbar\Delta|$$

and keeping only the first and third terms on the right-hand side of (4), we find

$$\delta\varepsilon(r)=0, \quad \delta\varepsilon(r)=\frac{(dE)^2}{\varepsilon_0(r)}(1-3\sin^2\theta). \tag{10}$$

When the angle between **E** and **r** is changed the second root changes sign, so there is no well in this case.

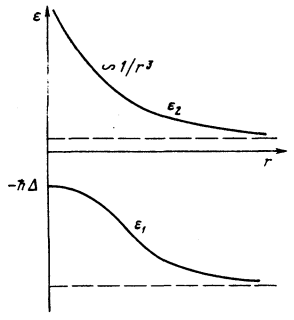

FIG. 2. The functions $\varepsilon(r)$ for the symmetric case. $\varepsilon_1(r) \to \frac{1}{2}(\hbar\Delta + [(\hbar\Delta)^2 + (2dE)^2]^{1/2})$ at large r.

This investigation thus shows that the presence of a resonant field together with the unbounded potential wells of the dipole interaction causes wells of finite depth to appear. The parameters of these wells are determined by the field strength and their centers are at the origin. This latter fact simplifies greatly the finding of the terms for $r \ll r_0$. Apparently, there are no other potential wells with minima at a finite distance from the origin.

3. INTERACTION OF THE ATOMS AT LARGE DISTANCES

It is well known[7] that in the absence of an external field the interaction of atoms in the wave zone $kr \gg 1$ is described by the potential

$$U(r)=-23c\hbar\alpha_1\alpha_2/4\pi r^7, \tag{11}$$

where α_1 and α_2 are the polarizabilities of the atoms at zero frequency. Virtual photons are then exchanged between the atoms. In an external resonant field the atoms exchange real photons and the interaction force grows considerably and varies with distance as r^{-2}. This force arises as a result of the pressure exerted by the light scattered by the atoms. We note that light-pressure forces for a single atom were studied in Refs. 8 and 9.

The general expression for the force **F** acting on the dipole moment $\mathbf{p}(t)e^{-i\omega_0 t} + \text{c.c.}$ of an atom in a field $\mathbf{E}(\mathbf{r}, t)e^{-i\omega_0 t} + \text{c.c.}$, averaged over the small period of oscillations $2\pi/\omega_0$, has the form

$$\mathbf{F}=p_i(t)\nabla E_i^*(\mathbf{r}, t)+\text{c.c.} \tag{12}$$

Let us find the force $\mathbf{F}_{ab}$ with which an atom a acts on an atom b. We shall denote the coordinates and velocities of the atoms by $\mathbf{r}_a$, $\mathbf{v}_a$ and $\mathbf{r}_b$, $\mathbf{v}_b$. The field at the point $\mathbf{r}=\mathbf{r}_b$ is written as

$$\mathbf{E}_0(\mathbf{r}_b, t)+\mathbf{E}_{1a}(\mathbf{r}_b, t),$$

where $\mathbf{E}_0(\mathbf{r}, t)=\mathbf{E}_0 e^{-i\Delta t+i\mathbf{k}\cdot\mathbf{r}}$ is a strong external field and $\mathbf{E}_{1a}(\mathbf{r}, t)$ is the weak field emitted by atom a. Accordingly, we write the dipole moment of the atom b as $\mathbf{p}_b(t) = \mathbf{p}_{0b}(t) + \mathbf{p}_{1b}(t)$, where $\mathbf{p}_0$ and $\mathbf{p}_1$ are the dipole-moment components induced by the fields $\mathbf{E}_0$ and $\mathbf{E}_{1a}$.

The polarizations of the fields $\mathbf{E}_0$ and $\mathbf{E}_{1a}$ are different, which greatly complicates the calculation. However, in the cases of a weak or a strong external field, which will be studied in detail below, the following simplification can be made. In a weak external field the polarizability of the atom, which determines the interaction, is isotropic. This isotropy disappears in a strong field, but the resonance frequencies of the polarizability along and across the field differ considerably. In the resonance approximation the contributions from the transverse and longitudinal components of the polarizability can be separated. Below we consider only the contribution from the longitudinal part. This allows us to assume that the atoms have two levels and the vectors **p** and **E** can be viewed as scalar quantities.

To find $p(t)$ it is necessary to use the equation for the atom-density matrix (q is the difference between the

populations of the upper and lower levels)

$$\left(\frac{d}{dt}+\frac{\gamma}{2}\right)p=i\frac{d^2E}{\hbar}q, \quad \frac{dq}{dt}+\gamma(1+q)=\frac{2i}{\hbar}(pE^*-\text{c.c.}). \tag{13}$$

The width of the upper level is denoted by γ and the lower level is assumed to be the ground state.

For the weak field we have the expression

$$E_{1a}(\mathbf{r}_b,t)=\frac{k^2e^{ik_0r}}{r}p_{0a}\left(t-\frac{r}{c}\right)\sin\theta, \quad k_0=\frac{\omega_0}{c},$$

where the retardation time is $\tau=|\mathbf{r}|/c$, $\mathbf{r}=\mathbf{r}_b(t)-\mathbf{r}_a(t-\tau)$, and θ is the angle between the vectors $\mathbf{E}_0$ and $\mathbf{r}$. To find ∇E_1 at large distances it is sufficient to differentiate only e^{ik_0r}. Then to second order in the weak field we have the following expression for the force acting on atom b:

$$\mathbf{F}_b=\mathbf{F}_{0b}+\mathbf{F}_{ab}^{(1)}+\mathbf{F}_{ab}^{(2)}, \quad \mathbf{F}_{ab}^{(1,2)}=2k\mathbf{n}\,\mathrm{Im}\,(p_{0,1b}E_{1a}(\mathbf{r}_b,t)), \tag{14}$$

where $\mathbf{F}_{0b}=\hbar\mathbf{k}\gamma W$ is the light-pressure force on atom b in the field of a traveling plane wave and

$$W=\frac{|dE_0|^2}{\hbar^2(\Delta^2+\gamma^2/4)+2|dE_0|^2}$$

is the population probability of the upper level. The functions referring to different atoms will henceforth have a subscript a or b and differ from the function without the subscript by the replacement of Δ by $\Delta_{a,b}=\Delta-\mathbf{k}\cdot\mathbf{v}_{a,b}$.

In a homogeneous light field $\mathbf{F}_{0b}$ does not depend on the atomic coordinates, while $\mathbf{F}_{ab}$ depends on the distance $\mathbf{r}$ between the atoms. The force $\mathbf{F}_{ab}$ is central, but not potential. It is directed along $\mathbf{r}$ or opposite to $\mathbf{r}$, depending on the sign of the work performed on atom b by the weak field. If the weak field is absorbed by atom b, then atom a repulses atom b. On the other hand, if the weak signal induces emission by atom b (this is possible in an external field), then atom a attracts atom b. If the atoms interact at large distances, then $\mathbf{F}_{ab}\neq\mathbf{F}_{ba}$, that is, the action is not equal to the reaction. This appears most clearly in the case where the external field acts only on one of the atoms. In the following, however, we shall assume that both atoms are in a homogeneous field.

We note that at large distances the interaction potential of the two excited atoms in the absence of an external field has the form $\text{const}\cdot r^{-1}\cos(\mathbf{k}\cdot\mathbf{r})$ (Ref. 10). In an external field the nature of the interaction is changed considerably, namely, together with terms containing factors like e^{ikr}, in the expression for the force $\mathbf{F}_{ab}$ there are terms which vary slowly with r. In averaging over a volume containing many wavelengths, the oscillatory terms disappear. Therefore, below we shall calculate only the slowly varying part of the force $\mathbf{F}_{ab}$, which gives the main contribution to the interaction of the atoms at $kr\gg1$.

Using the stationary solution of Eqs. (13) for p_0, we find the following expression for the first-order interaction force:

$$\mathbf{F}_{ab}^{(1)}=\tfrac{3}{2}n\mathbf{r}r^{-1}\hbar\gamma W_aW_bV^{-2}[(\gamma_1^2+\Delta_a\Delta_b)\sin\varphi+\gamma_1(\Delta_a-\Delta_b)\cos\varphi], \tag{15}$$
$$\varphi=kr-\mathbf{kr}, \quad V=|dE_0|/\hbar.$$

If the atoms are located in a line coincident with the direction of the light propagation so that $\mathbf{k}\cdot\mathbf{r}=kr$, the interaction force varies as $1/r$. The absence of oscillations is due to the fact that in forward scattering the atom a radiates a field coherent with the incident field.

In the case $\Delta\gg kv$, γ, V we have

$$\mathbf{F}_{ab}^{(1)}=\tfrac{3}{2}n\mathbf{r}r^{-1}\hbar\gamma(V/\Delta)^2\sin\varphi$$

and the sign of the force does not change within a single Fresnel zone.

The order of magntidue of the ratio $F_{ab}^{(1)}/F_{0b}$ is $(kr)^{-1}\ll1$. The collective action of the atoms can lead to a considerable change of the force acting on a given atom. In a light beam of large cross section the force $\mathbf{F}_{ab}^{(1)}$ becomes rapidly oscillating in the far Fresnel zones. In this case it is necessary to take into account the second-order force, in which there are no oscillations. In the presence of an external field $E_0(t)$ the response of an atom $p_1(t)$ to a weak field $E_1(t)$ can be written as

$$p_1(t)=\int_0^\infty d\tau(\alpha(\tau)E_1(t-\tau)+\beta(\tau)E_1^*(t-\tau)e^{-2i\Delta(t-\tau)}).$$

When the external field is small, β is also small and α is the isotropic polarizability of the atom. In a strong external field, in accord with the above discussion, α and β are the longitudinal components of the polarizability.

In substituting $p_1(t)$ into (14) the second term can be dropped, since it leads to the appearance of an oscillatory factor $e^{\pm2ikr}$. The formula for the force now is of the form

$$\mathbf{F}_{ab}^{(2)}=\frac{2k^3\mathbf{n}\sin^2\theta}{r^2}\mathrm{Im}\int_0^\infty d\tau\alpha_b(\tau)\mathcal{F}_a(\tau)\exp\left\{-ik\dot{r}(t)\tau+\frac{ik\ddot{r}(t)\tau^2}{2}\right\},$$
$$\dot{r}(t)=\mathbf{nv}, \quad \ddot{r}=(\mathbf{v}^2-(\mathbf{nv})^2)/r, \quad \mathbf{v}=\mathbf{v}_b-\mathbf{v}_a. \tag{16}$$

Here $\mathcal{F}_a(\tau)=\langle p_{0a}^*(\tau)p_{0a}(0)\rangle$ is the correlator for the dipole moment of atom a. The angle brackets denote averaging over the fluctuations arising because of spontaneous emission.

The Fourier transform of the correlator $\mathcal{F}(\tau)$ determines the spectrum of the resonance fluorescence of the atom and has been calculated in a number of studies.[11-13] The emission spectrum of the atom contains coherent (monochromatic) and incoherent (nonmonochromatic) radiation components. Formula (16) takes into account the effect on atom b by both radiation components of atom a. The relative contributions from these components change considerably as a function of the external field. Expansion of $r(t-\tau)$ in a series in the retardation time τ was carried out in (16) up to terms of second order. The first term $k\dot{r}(t)\tau$ describes the Doppler shift of the frequency and the second term $k\ddot{r}(t)\tau^2/2$ arises because of the difference of the spherical wave from a plane wave. This can be important only for distances which are not too large $r<r_1$ [see (21)]. The function $\mathcal{F}(\tau)$ is found from the system of kinetic equations (13) with special initial conditions and with a special right-hand side. The polarizability $\alpha(\tau)$ is found from the solution of the linearized system of equations (13), which, as is well known, is exactly

solvable in a monochromatic field.

It is convenient to carry out the subsequent calculations in the frequency representation. Dropping the factor $\exp(ik\ddot{r}\tau^2/2)$ in (16) for the time being (its role is discussed in Sec. 3.1), the general expression for the interaction force is

$$\mathbf{F}_{ab}^{(2)}=-\frac{9n\gamma^2\hbar\sin^2\theta}{8kr^2}\operatorname{Im}\int_{-\infty}^{+\infty}\frac{d\omega}{2\pi}\frac{A_b(\omega)B_a(\omega+\nu)}{D_b(-\omega)D_a(\omega+\nu)}, \tag{17}$$

where

$$\begin{aligned}D(\omega)&=(\omega-i\gamma)(\omega-\mu_+)(\omega-\mu_-)-4V^2(\omega-i\gamma/2),\\ A(\omega)&=(1-2W)[(\omega+i\gamma)(\omega+\mu_+)+2V^2\omega/\mu_-],\\ B(\omega)&=-\frac{iW}{(\omega-i0)}(D(\omega)+2V^2(\omega-i\gamma)),\\ \nu&=(kn-k)v,\quad \mu_\pm=\pm\Delta+i\gamma/2.\end{aligned} \tag{18}$$

Here $A(\omega)/D(-\omega)$ is the polarizability and the quantity $B(\omega)/D(\omega)$ characterizes the emission spectrum of the atom. In (17) we have used the relation $\gamma=4d^2k^3/3\hbar$.

Let us now consider several typical limiting cases.

3.1. Weak field

In a weak field ($W\ll 1$) we have $A(\omega)/D(-\omega)=-(\omega+\mu_+)^{-1}$. Only the coherent component is important in the emission spectrum, so that $B(\omega)/D(\omega)=-iW(\omega-i0)^{-1}$. With the two cases of large and small distances in mind, we use the general formula (16):

$$\mathbf{F}_{ab}^{(2)}=\frac{2\varkappa\mathbf{n}}{r^2}\operatorname{Re}\int_0^\infty d\tau\exp\left\{-\frac{\gamma}{2}\tau+i(\Delta_b-\nu)\tau+\frac{ik\ddot{r}(t)\tau^2}{2}\right\}, \tag{19}$$

where

$$\varkappa=\frac{9}{16}\frac{h\gamma^2\sin^2\theta W_a}{k}.$$

Expression (19) has a simple form for $|\Delta_b-\nu|\gg\gamma$, when the integral can be written as an expansion in inverse powers of the effective frequency difference:

$$\mathbf{F}^{(2)}=\frac{n\varkappa}{r^2}\left[\frac{\gamma}{(\Delta_b-\nu)^2}-\frac{2k\ddot{r}(t)}{(\Delta_b-\nu)^3}\right]. \tag{20}$$

The ratio of the second term to the first can be written as r_1/r, where

$$r_1=\frac{k(v\sin\vartheta)^2}{\gamma|\Delta_b-\nu|}; \tag{21}$$

ϑ is the angle between the vectors $\mathbf{k}$ and $\mathbf{n}$. Under ordinary conditions the parameter $kv/\gamma\sim 10^2$. Therefore, for frequency differences which are not too large, $kr_1\gg 1$. The maximum value of r_1 can be estimated from (21), setting $|\Delta_b-\nu|\sim\gamma$, so that $r_{1\max}\sim k^{-1}(kv/\gamma)^2\sim 0.1$ cm.

At distances $k^{-1}<r<r_1$ the second term in (20) becomes the most important. In this case the sign of the force coincides with the sign of $\nu-\Delta_b$, since $\ddot{r}(t)$ is always positive. The attraction between the atoms at $(\Delta_b-\nu)>0$ can be explained as follows.

From (19) we see that a field with a frequency difference $\Delta_b-\nu+k\ddot{r}(t)\tau/2$ which increases linearly with time acts on the atom. In the adiabatic approximation the population of the upper level is inversely proportional to the square of the frequency difference. The decrease of the population of the upper level can be viewed as the result of the stimulated emission of photons of the weak field. Therefore, the work done by the radiation field of atom a on atom b is negative, which corresponds to an attractive force. We note that the effect of changing the sign of the work done by the field on the atom was noticed in a study of the absorption spectrum of an atom accelerated by an external field.[14]

Therefore, at $r<r_1$ the atoms can either attract or repulse each other depending on the sign of the frequency difference and the force varies with distance as r^{-3}. At large distances $r>r_1$ the difference of the spherical wave from a plane wave can be neglected ($\ddot{r}(t)\approx 0$) and the force can be written in the form of the Coulomb law with an effective charge $g(\mathbf{n})$:

$$\mathbf{F}_{ab}^{(2)}=\frac{\mathbf{n}g^2(\mathbf{n})}{r^2},\quad g^2(\mathbf{n})=\frac{\varkappa\gamma}{(\Delta_b-\nu)^2+(\gamma^2/4)}. \tag{22}$$

The square of the "induced" charge is proportional to the product of the probabilities for the excitation of atom a and the absorption of the scattered field by atom b. Estimate of the maximum value of g for $W_a\sim 1$ and $|\Delta_b-\nu|\sim\gamma$ gives

$$g\sim(ka)e\sim 10^{-3}e. \tag{23}$$

The effective charge turns out to be smaller than the electron charge e by a factor equal to the multipole expansion parameter ka, where a is the Bohr radius.

The force $F_{ab}^{(2)}$ is considerably greater than the interaction force ∇U of the atoms without a field and smaller than the light pressure F_{0b} by a factor equal to the small parameter of the problem $1/kr$:

$$\frac{|\nabla U|}{F_{ab}^{(2)}}\sim\frac{c\hbar(ka)^4}{e^2(kr)^6}\ll 1,\quad \frac{F_{ab}^{(2)}}{F_{0b}}\sim\frac{1}{(kr)^2}\ll 1. \tag{24}$$

In a gas of finite density the interaction of the atoms increases because of the slow fall off of the Coulomb force. The total force

$$\mathbf{f}_b=\sum_{a\neq b}\mathbf{F}_{ab}^{(2)}$$

acting on atom b diverges linearly at large r. The maximum distance is limited by the photon mean free path. From this we find that the total force can be comparable to the light-pressure force $f_b\sim F_{0b}$. This is a rough estimate, since it does not include the vector nature of the force. This shows, however, that in large volumes of the gas comparable to the weak field absorption length, the total force acting on a particle can differ markedly from F_{0b}.

3.2. Strong field

The calculation of the force according to expression (17) reduces to finding the residues at the poles, which are determined by the zeros of the function

$$D(\omega)=(\omega-\omega_1)(\omega-\omega_2)(\omega-\omega_3).$$

The ω_i are the eigenfrequencies of the two-level atomic system in an external field. In a strong field they are equal to

$$\omega_{1,3}=\pm\Omega+i\gamma_1, \quad \omega_2=i\gamma(1-2V^2/\Omega^2),$$
$$\Omega=(\Delta^2+4V^2)^{1/2}, \quad \gamma_1={}^1/_2\gamma(1+2V^2/\Omega^2) \tag{25}$$

and the Stark shift of the levels is considerably larger than the natural width, $\Omega \gg \gamma$. The polarizability of the atom at the eigenfrequencies ω_i is significantly different. At the unshifted frequency ω_2 it is less than the polarizability at the sideband frequencies by a factor of Ω/γ. Therefore, closing the integration contour in the lower half-plane, it is sufficient to take into account only the contributions from the poles at $\omega=\omega_1$ and $\omega=\omega_3$.

As a result we obtain the following expression[3]

$$\mathbf{F}_{ab}^{(2)}=\frac{9n\hbar\gamma^2\sin^4\theta(1-2W_b)}{32kr^2\Omega_b\Delta_b}\operatorname{Re}\left\{-(\Omega_b-\Delta_b)^2\frac{B_a(-\omega_{1b}+\nu)}{D_a(-\omega_{1b}+\nu)}+(\Omega_b+\Delta_b)^2\frac{B_a(-\omega_{3b}+\nu)}{D_a(-\omega_{3b}+\nu)}\right\}. \tag{26}$$

The interaction strength is determined by the sum of two terms, each of which is proportional to the product of some effective oscillator strength at the frequency ω_1 or ω_3 and the intensity of the emission of atom a at the same frequencies. The terms in (26) have different signs, so the force F_{ab} can be of either sign.

The emission spectrum $\operatorname{Re}(B(\omega)/D(\omega))$ consists of two shifted components at the frequencies $\pm\Omega$ and two unshifted ones. One of the components at zero frequency corresponds to coherent emission. Because of the motion of the atoms their eigenfrequencies differ. The force $F_{ab}^{(2)}$ depends considerably on the relation between ω_{ia} and ω_{jb} and reaches a maximum when one of the components of the emission spectrum of atom a coincides with an eigenfrequency of atom b. This happens when the condition

$$\operatorname{Re}(\omega_{ia}+\omega_{jb})=\nu \tag{27}$$

is satisfied. The interaction force is thus a resonant function of the velocities of the atoms. For an arbitrary relation between $\Delta_{a,b}$, ν, and V the force F_{ab} has many resonances corresponding to condition (27). We shall restrict our consideration to the simple case when $kv \ll \Delta$, V and the eigenfrequencies of atoms a and b are close. Formula (26) has the form

$$\mathbf{F}_{ab}^{(2)}=\frac{9n\hbar\gamma^2\sin^4\theta(1-2W)}{32kr^2\Delta\Omega}\left\{\frac{(\Omega-\Delta)^2}{\mathbf{k}_+\mathbf{v}+2i\gamma_1}-\frac{(\Omega+\Delta)^2}{\mathbf{k}_-\mathbf{v}+2i\gamma_1}\right\}. \tag{28}$$

The force (28) has resonances when the velocity $\mathbf{v}$ is perpendicular to one of the vectors $\mathbf{k}_\pm=\mathbf{k}(1\pm\Delta/\Omega)+\mathbf{n}k$. For an arbitrary orientation of $\mathbf{n}$ and $\mathbf{k}$ the force $F_{ab}^{(2)}$ can be attractive or repulsive, depending on the direction of the velocity.

4. CONCLUSION

Out of the broad range of problems related to the interaction of atoms in an external field, we have considered two in the present study. In the first part we studied the adiabatic potentials of atoms in the near zone. It was shown that in a light field the interaction potential has the form of a well whose parameters depend on the field strength. The atoms can therefore form a quasimolecule of dimensions larger than those of the atoms. The bound states can apparently be experimentally observed as fine structure in studying the Stark shift of the levels in gases.

The second part of the study was devoted to finding the interaction forces of the atoms at large distances. It was shown that the interaction coincides with the Coulomb law and the effective charge depends on the field and velocity of the atoms. In weak fields the atoms repulse each other and in strong fields both repulsion and attraction are possible.

In a gas of finite density the force of the interaction of an atom with the other particles can be comparable to the light-pressure force. This can be important in studying the kinetics of atoms in a resonant field. From this viewpoint the experiment of Ref. 15 is of interest, in which an anomalous distribution of the gas under the influence of light pressure was observed.

In conclusion we note that particles can exchange not only photons, but also phonons. This can also lead to the appearance of long-range forces. It is possible that forces of this type play a role in the interaction of protein molecules.[16]

[1)] In strong fields the size of the bound state can be comparable to the range of the exchange potential.[5]

[2)] The bound states for a purely dipole interaction induced by a nonresonant field were discussed in Ref. 2.

[3)] In finding the contribution from the longitudinal component in a strong external field it is necessary to take the projection of the weak field vector on the direction of the strong field vector, i. e., multiply E_{1a} again by $\sin\theta$.

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Experimental investigation of the feasibility of application of the wavefront reversal phenomenon in stimulated Mandel'shtam-Brillouin scattering

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The possibilities of applying stimulated Mandel'shtam-Brillouin scattering (SMBS) to laser-mediated thermonuclear fusion problems and, in particular, to the decoupling of amplifier stages, shaping of pulse profiles, etc., are considered. The stimulated Mandel'shtam-Brillouin scattering is obtained experimentally under nonstationary conditions by exciting the system with radiation from a photodissociation iodine laser. A stationary scattering regime is also attained with a pump pulse of 5–10 μsec duration. Under these conditions, the operation of an amplifier with a SMBS mirror is studied experimentally in the case of weak input signals ($I_{in} \sim 10^{-3}$ W/cm^2) and a gain per pass of $\sim 10^6$ is attained. Particular attention is paid to the quantitative determination of the characteristics of the degree of pump-beam reproducibility and of the compensation of the optical inhomogeneities of the laser medium, and also to the clarification of the conditions under which they are observed. The range of pump intensities in which complete reproduction of the angular spectrum is observed is found experimentally. The dependence of the compensation accuracy on the degree of reproduction of the angular spectrum is obtained.

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INTRODUCTION

In recent years there has been an ever broadening interest in the phenomenon of wave front reversal in nonlinear processes. High directivity of the scattered radiation was apparently first observed in Refs. 1–3 for stimulated Mandel'shtam Brillouin scattering (SMBS) and in Refs. 4 and 5 for stimulated Raman scattering (SRS). However, only publication of Ref. 6 has made it clear that the field of the scattered radiation is, under certain circumstances, the complex conjugate of the pump field, and that this effect can be used for the compensation of phase distortions of laser emission. In particular, this phenomenon was used in Ref. 7 for compensation of optical inhomogeneities in a ruby amplifier. The publication of these works stimulated theoretical and experimental investigations of the phenomenon of wave front reversal and consideration of the feasibility of its application (see, for example, Refs. 8–20).

The present work is devoted to the study of the possibility of application of this phenomenon to the excitation of SMBS by the emission of photodissociation lasers (PL), which have been developed[21-24] along with others for the solution of laser-mediated thermonuclear fusion (LTF) problems. The experimental investigations were conducted principally in a stationary scattering regime, in which the pulse length of the laser radiation was significantly greater than the lifetime of the acoustical phonons. In this regime, the phenomenon of wave front reversal should be accomplished in the purest form. Special attention was paid to the experimental investigation of the quantitative characteristics of the degree of reproducibility and of the compensation, and the limiting conditions under which they are observed were determined. The results are of interest not only in LTF but also for problems of optical communication[14] and the acceleration of macro- and micro-bodies.[25-29]

1. SOME POSSIBILITIES OF THE APPLICATION OF SMBS TO THE PROBLEM OF LTF

As is known, elements of interstage decoupling and a system of decoupling of the output stages from the target are necessary in LTF systems.

For these purposes, it is expedient to consider, in addition to Kerr and Faraday shutters and phototropic shutters, also shutters that operate on stimulated scattering (SMBS or SRS). Figure 1 shows one of the possible schemes of decoupling amplifiers from a target with the use of SMBS. The radiation of the amplifier by means of a beam-splitting mirror ($R \approx 0.5$) is fed to two SMBS cells. The reflected Stokes radiation is focused on the target. Under conditions in which the spontaneous radia-

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