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Cooling and capture of atoms and molecules by a resonant light field

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A detailed classical analysis is presented of the method of cooling and capturing atoms and molecules by a resonant light field. The light-pressure forces acting on a particle in a three-dimensional standing light wave are determined. It is shown that these forces can be used for effective cooling and spatial capture of cold particles in the nodes or antinodes of a light field. The use of the method for the observation of narrow spectral lines in optical spectra or atoms or molecules, and for spectroscopic investigations for exceedingly small numbers of atoms, is considered.

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1. INTRODUCTION. FORMULATION OF PROBLEM

It is known that an atom or a molecule is acted upon in a light field by light-pressure forces[1] due to the recoil produced when the atom or molecule scatters a light photon. Thus, in a nonresonant light field the particles are acted upon by a striction force[2–4] due to the Compton scattering of the light by the atom or molecule. In a resonant light field, the particles are acted upon by forces of two types: the force of the spontaneous light pressure, and the force which we shall call henceforth the induced-light-pressure force. For example, in the case of a one-dimensional standing light wave, the force of the spontaneous light pressure is due recoil following absorption of a plane light wave by the particle and emission of a spherical light wave,[5,6] while the induced light-pressure force is produced by recoil due to induced absorption and emission of a plane light wave.[6,7]

An important and highly promising region of application of the light pressure is optical spectroscopy of ultrahigh resolution without Doppler broadening, based on registration of narrow spectral lines in atoms or molecules whose motion is either completely stopped or considerably limited by the light-pressure forces.[4] Indeed, the elimination of the particle motion in a low-pressure gas leads not only to a vanishing of the Doppler broadening of the spectral lines, but simultaneously to a vanishing of all the broadening sources due to the particle motion[8] (broadening due to the finite time-of-flight of the particles through the light beam, broadening due to the quadratic Doppler effect, etc.), and makes it possible in principle to obtain spectral lines with widths determined only by the properties of the quantum transitions of the atoms or molecules themselves. In addition, the use of light pressure to limit the spatial displacement of the particles in a low-pressure gas and for a prolonged containment of particles in fixed region of space makes it possible to carry out spectroscopic investigations of extremely small numbers of atoms or molecules, and permit in principle to realize ultrahigh-resolution spectroscopy of individual atoms or molecules (monatomic spectroscopy).

Earlier studies[4,9–11] considered new possibilities of spectroscopy without Doppler broadening of the atoms and molecules, based on the use of the light pressure to alter the velocity distribution of particles in a low-pressure gas. It was proposed[4] to use the nonresonant recoil force to capture particles in the field of a standing light wave. It turns out that in a standing light wave having a frequency outside the resonant transitions of the atom or molecule, particles having sufficiently small velocity projections on the light-wave propagation direction execute, under the influence of the nonresonant recoil force, finite vibrational motions near the nodes or antinodes of the field. As a result of these vibrational motions of the atom or molecule, a narrow resonance can exist at the center of the Doppler-broadened absorption line observed in the direction of the wave vector of the light wave; this resonance has a width determined by the natural line width and by the flight broadening.[4,11] In the case of three-dimensional standing light wave, this method makes possible the spatial capture of slow particles and permits registration, for any direction of the sounding light field, of a narrow resonance with a width

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determined only by the natural broadening. Subsequently[9] the idea was advanced that the force of the spontaneous light pressure can be used to slow down (cool) atoms to velocities corresponding to the natural width of the resonance transition, by isotropic irradiation of the low-frequency half of the Doppler-broadened line of the optical transition with a resonant light field and the ensuing narrowing the Doppler-broadened atomic lines.

A common basic shortcoming of the two proposed methods is the requirement that the light field have an appreciable intensity ($\gtrsim 10^3$ W/cm^2). In the first approach,[4] the strong-field requirement is due to the smallness of the nonresonant recoil force, while in the second[9] it is due to the need for saturating the entire Doppler contour of the optical transition. It was shown,[10] however, on the basis of a qualitative classical analysis of the motion of particles in the field of a standing light wave, that by using the light pressure acting on the atom or molecule in the resonant field of a standing light wave it is possible, at moderate intensities of the light field (~0.01–0.1 W/cm^2) to cool practically all the particles in a low-pressure gas, and simultaneously capture an appreciable fraction of the cold particles in the region of the light field.

Understandably, a rigorous analysis of the proposed method must be based on a consistent quantum-mechanical analysis of the motion of the particles in the resonant light field. We note that certain quantum-mechanical aspects of a similar problem have already been considered.[12] However, all the main regularities, both with respect to the motion of the mass center of the particle in the resonant light field and with respect to the singularities of the emission or absorption spectra of the cold captured particles, can be determined even from a classical description of the motion of the mass centers of the particles. Naturally, the internal motions of a particle interacting with a resonant light field should be considered quantum-mechanically in any approach.

The purpose of the present paper is to present a detailed analysis of the previously proposed[10] method of spectroscopy, without Doppler broadening, of atoms and molecules in a low-pressure gas, based on the cooling and capture of the particle by the resonant forces of the light pressure. We present below a classical analysis of the motion of the mass centers of the particles in one- and three-dimensional standing light waves under the influence of the resonant light-pressure forces, and carry out a classical analysis of the emission (absorption) spectra of cold captured particles. Principal attention will be paid to the case of a three-dimensional standing light wave, inasmuch as only three-dimensional spatial cooling and capture of the particles by the forces of the light pressure make it possible to use in full measure the advantages of the given method as a method of spectroscopy of ultrahigh resolution of the individual atoms and molecules.

2. FORCES ACTING ON A PARTICLE IN THE FIELD OF A STANDING LIGHT WAVE

It is known that an atom or molecule in a light field $\mathbf{E}=\mathbf{E}(\mathbf{r}, t)$ whose wavelength is much larger than the particle dimensions is acted upon by a force[13]

$$\mathbf{F}=\nabla(\langle\mathbf{d}(\mathbf{r}, t)\rangle\mathbf{E}(\mathbf{r}, t)), \tag{1}$$

where the differentiation operator ∇ acts only on $\mathbf{E}(\mathbf{r}, t)$, and the average dipole moment of the atom or molecule can be expressed in terms of the elements of the density matrix of the particle $\rho_{ij}=\rho_{ij}(\mathbf{r}, t)$:

$$\langle\mathbf{d}(\mathbf{r}, t)\rangle=\mathrm{Sp}[\rho(\mathbf{r}, t)\mathbf{d}(\mathbf{r}, t)]. \tag{2}$$

Let us find the forces acting on a particle in the field of a three-dimensional standing light wave made up of the three one-dimensional standing light waves:

$$\mathbf{E}(\mathbf{r}, t)=2E_0\cos\omega t(\mathbf{e}_x\cos kz+\mathbf{e}_y\cos kx+\mathbf{e}_z\cos ky). \tag{3}$$

The one-dimensional problem, obviously, is a particular case, and the corresponding results can be obtained from the results of the three-dimensional problem at $\mathbf{e}_y=\mathbf{e}_z=0$. We assume that the light wave is at resonance only with one transition of the particle with frequency ω_0 (the resonance approximation) and assume further that the lower level g of the resonant transition is the ground level, while the width of the upper level e is determined only by spontaneous decay at a rate to the ground level. Then the equations for the density-matrix elements are[14]

$$\begin{aligned} i\left(\frac{\partial}{\partial t}+\mathbf{v}\frac{\partial}{\partial\mathbf{r}}\right)\rho_{mm}&=V_{mn}\rho_{nm}-V_{nm}\rho_{mn}\pm i\gamma\rho_{ee},\\ i\left(\frac{\partial}{\partial t}+\mathbf{v}\frac{\partial}{\partial\mathbf{r}}\right)\rho_{mn}&=V_{mn}(\rho_{nn}-\rho_{mm})-\frac{i}{2}\gamma\rho_{mn}, \end{aligned} \tag{4}$$

where

$$m, n=e, g;\quad V_{nm}=V_{mn}^{*}=-2V_0e^{\pm i\Omega t}\sum_{\alpha}\cos kr_{\alpha};\quad V_0=dE_0/2\hbar.$$

The plus and minus signs pertain respectively to $m=e$, $n=g$ and $m=g$, $n=e$; d is the matrix element of the projection of the dipole moment of the particle on one of the axes of the coordinate system; the index α labels the projection of the radius vector of the particle and runs through the values $\alpha=x, y, z$; $\Omega=\omega-\omega_0$ is the detuning of the field frequency relative to the center of the line of the atomic or molecular transition.

In a classical treatment of the motion of the particles in the light field, it must be assumed that the de Broglie wavelength of the particle is much smaller than the wavelength of the light. This condition means simultaneously that the classical treatment is valid only for those particles whose momentum greatly exceeds the momentum of the absorbed or emitted photon, i.e.,

$$Mv\gg\hbar k. \tag{5}$$

For this reason, the particle velocity in (4) can be regarded as constant. At constant v, the stationary solutions of (4) can be obtained by successive approximations through an expansion of the density matrix elements in a spatial Fourier series, and the light-pressure force can be determined from relations (2) and (1).

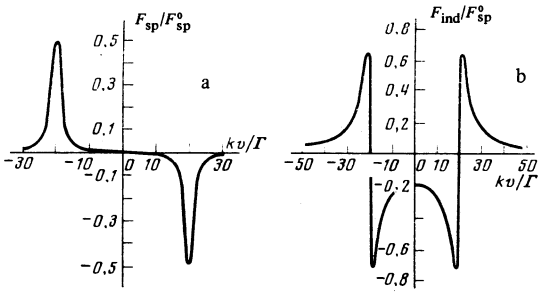

FIG. 1. The spontaneous light-pressure force normalized to $F^0_{sp}=2\hbar k\Gamma$ (a) and the force of the induced light pressure in a one-dimensional standing light wave (b) as functions of the particle velocity at $G=1$ and $\Omega/\Gamma=-20$. The forces were calculated at the spatial points corresponding to the maximum values of F_{sp} and F_{ind}.

Since it is of interest to know the average force acting on the particle, and since nonrelativistic particles ($v \ll c$) traverse during the period T of the field oscillations distances $l=vT$ which are much shorter than the light wavelength $\lambda=cT$, it follows that the resultant expression must also be averaged over the period of the field oscillations.

Finally, in the first nonvanishing approximation, the force acting on a particle in the field of a three-dimensional standing light wave consists of two parts:

$$\mathbf{F}=\mathbf{F}_{sp}+\mathbf{F}_{ind}, \tag{6}$$

where the force of the spontaneous light pressure is

$$\mathbf{F}_{sp}=\frac{2\hbar k\Gamma G}{1+G\Sigma}\Big(\sum_\alpha \mathbf{e}_\alpha \sin kr_\alpha\Big)\Big[\sum_\alpha (\mathscr{L}_-{}^\alpha-\mathscr{L}_+{}^\alpha)\sin kr_\alpha\Big], \tag{7}$$

and the force of the induced light pressure is

$$\mathbf{F}_{ind}=\frac{2\hbar k\Omega G}{1+G\Sigma}\Big(\sum_\alpha \mathbf{e}_\alpha \sin kr_\alpha\Big)\Big\{\sum_\alpha\Big[\Big(1-\frac{kv_\alpha}{\Omega}\Big)\mathscr{L}_-{}^\alpha + \Big(1+\frac{kv_\alpha}{\Omega}\Big)\mathscr{L}_+{}^\alpha\Big]\cos kr_\alpha\Big\}. \tag{8}$$

We have introduced above the notation

$$\mathscr{L}_\pm{}^\alpha=\frac{\Gamma^2}{(\Omega\pm kv_\alpha)^2+\Gamma^2},\quad G=\frac{2V_0^2}{\Gamma^2},\quad \Gamma=\frac{\gamma}{2},\quad \Sigma=\sum_\alpha(\mathscr{L}_-{}^\alpha+\mathscr{L}_+{}^\alpha).$$

For a one-dimensional standing light wave, expressions for the spontaneous and induced light pressure forces, which are particular cases of (7) and (8), have been derived earlier[10] (Fig. 1).

It must be emphasized that the two components into which the light-pressure force has been resolved represent the two processes of resonant interaction of the particle with the field. Absorption of plane light waves by the particle and emission of a spherical optical wave gives rise to the force of the spontaneous light pressure, while the induced absorption and emission of plane light waves gives rise to the forces of the induced light pressure.[1)] The difference between the forces (7) and (8) becomes particularly clear at $\Gamma=0$, i.e., in the case of a two-level system with infinitesimally narrow levels, when the force of the spontaneous light pressure vanishes identically and we are left only with the force due to the induced absorption and emission of the light waves.

We note finally two important circumstances. First, in a three-dimensional standing light wave (and also in a two-dimensional one) both the spontaneous light-pressure force and the induced light-pressure force contain new terms which are missing, in the same approximation, in the case of a one-dimensional standing light wave.[10] These terms have a spatial dependence of the form $\sin kr_\alpha \sin kr_\beta$, in the case of the spontaneous light-pressure force and of the form $\sin kr_\alpha \cos kr_\beta$ in the case of the induced light-pressure force ($\alpha\neq\beta$; $\alpha,\beta=x,y,z$), and determine the forces acting on the particle in a single one-dimensional standing light wave, when the nonvanishing of the average dipole moment of the particle is due to its interaction with another one-dimensional standing light wave. Second, in the higher approximations, which take into account the higher spatial harmonics in the density-matrix elements, the forces (7) and (8) will contain terms due to coherent scattering of the light both within the limits of a single standing light wave, and from one standing light wave into others. The contribution of such processes to the force of the light pressure is of course small for the saturation parameter values $G\lesssim 1$ of interest to us, and we confine ourselves henceforth to an investigation of the light-pressure forces in only the first nonvanishing approximation.

3. MOTION OF ATOMS AND MOLECULES IN THE FIELD OF A STANDING LIGHT WAVE

Generally speaking, the motion of a particle in the field of a standing light wave is due to the joint action of the forces of the spontaneous and induced light pressure. These forces, however, act in essentially different manners on the particles in a low-pressure gas. The force of the spontaneous light pressure results on the average in a directional acceleration of the particles: at $\Omega>0$ the particles are accelerated by the field, and at $\Omega<0$ they are slowed down; owing to the resonant character of the force, the acceleration or deceleration effect is maximal for those particles whose velocity projections on the directions of the wave vectors of the light waves satisfy the relations

$$v_\alpha=\pm\Omega/k. \tag{9}$$

The force of the induced light pressure causes either oscillatory motion of the particles or, if the particle velocity is high enough, spatially periodic modulation of the particle velocity. For this reason, it is of interest to consider the singularities of the motion of the particle under the influence of each of the light-pressure forces. Such an analysis can be carried out most completely in the case of a one-dimensional standing light wave.

From the equation for the motion of a particle under the influence of the spontaneous light-pressure force it is possible to obtain, by lowering the order of the dif-

ferential equation, an equation of motion that contains only the coordinate and velocity of the particle:

$$4G\frac{R_{rec}}{\hbar\Omega}\left(\frac{\Gamma}{\Omega}\right)^3[2\xi-2\xi_0+\sin 2\xi_0-\sin 2\xi]$$
$$=\frac{1}{5}(\eta^5-\eta_0^5)+\frac{2}{3}\left[(1+G)\frac{\Gamma^2}{\Omega^2}-1\right](\eta^3-\eta_0^3)$$
$$+\left(1+\frac{\Gamma^2}{\Omega^2}\right)\left[(1+2G)\frac{\Gamma^2}{\Omega^2}+1\right](\eta-\eta_0). \quad (10)$$

We have introduced here the dimensionless coordinate ($\xi=kz$) and dimensionless velocity ($\eta=k\dot{z}/\Omega$) of the particle along the wave vector of the field, which is directed for the sake of argument along the z axis; $R_{rec}=(\hbar\omega)^2/2Mc^2$ is the recoil energy of the particle, and allowance is made for the initial conditions $\xi(0)=\xi_0$, $\eta(0)=\eta_0$.

Equation (10) at $\Omega>0$ describes the cooling of the particle under the influence of the force of the spontaneous light pressure. An analysis of the equation shows that at $\Omega<0$ the resonant particles ($v_g=\pm\Omega/k$) move on the average with a constant deceleration, which is maximal at $|\Omega|\gg\Gamma$ and does not depend on the value of the detuning;

$$|\dot{v}_z|=\frac{2G}{1+G}\frac{R_{rec}}{\hbar}\frac{\Gamma}{k}, \quad (11)$$

and is very small at $|\Omega|\ll\Gamma$:

$$|\dot{v}_z|=\frac{8G}{1+2G}\frac{R_{rec}}{\hbar}\frac{\Gamma}{k}\frac{\Omega^2}{\Gamma^2}. \quad (12)$$

From the equation of particle motion under the influence of the induced light-pressure force

$$\frac{1}{\Omega^2}\frac{d^2\xi}{dt^2}=\frac{2R_{rec}}{\hbar\Omega}\frac{G[(1-\eta)\mathscr{L}_-^2+(1+\eta)\mathscr{L}_+^2]}{1+G[\mathscr{L}_-^2+\mathscr{L}_+^2]}\sin 2\xi, \quad (13)$$

we obtain in similar fashion an equation of motion that contains likewise only the coordinate and the velocity of the particle:

$$4G\frac{R_{rec}}{\hbar\Omega}\frac{\Gamma^2}{\Omega^2}(\cos 2\xi-\cos 2\xi_0)=\frac{1}{2}(\eta^4-\eta_0^4)$$
$$+\left[(3+2G)\frac{\Gamma^2}{\Omega^2}-1\right]\cdot(\eta^2-\eta_0^2)+4\frac{\Gamma^2}{\Omega^2}\left(1+\frac{\Gamma^2}{\Omega^2}\right)(1+G)\ln\frac{1+\Gamma^2/\Omega^2-\eta^2}{1+\Gamma^2/\Omega^2-\eta_0^2}. \quad (14)$$

Equation (14) describes both the finite vibrational motion—capture of the particles—and the infinite vibrational motion of the particles with a bounded variation of the velocity. A joint analysis of Eqs. (13) and (14) shows that the particles can be captured at the points $\xi=\pi/2+n\pi$ ($n=0,1,2,\ldots$), i.e., at the nodes of the light wave, if $\Omega>0$, and at the points $\xi=n\pi$, i.e., at the antinodes of the light wave, if $\Omega<0$. If the particle is initially at a node or antinode of the wave, then at a particle velocity $|\eta_0|<\eta_{capt}$ it will execute vibrations near the equilibrium point, with an amplitude not exceeding one quarter of the wavelength of the light. If $|\eta_0|>\eta_{capt}$, then the particle will execute infinite motion, and the particle velocity will vary periodically in space from η_{min} to η_0, where η_{min} can be obtained from (14) in which we put $\cos 2\xi-\cos 2\xi_0=\pm 2$ respectively for $\Omega=\mp|\Omega|$, and η_{capt} can also be obtained from (14) if, in addition, we put $\eta_0=0$.

Both the cooling and the capture of the particles by the resonant recoil forces are of interest only for strong transitions of the atoms or molecules, for which a typical relation is $R_{rec}\ll\hbar\Gamma$. Under these conditions we find from (14) that the limiting value of the projection of the velocity of the dragged particles is equal to

$$|v_z^{capt}|=\frac{|\Omega|}{k}\eta_{capt}=2\left(\frac{\hbar\Gamma}{M}\right)^{1/2}\left[\frac{G(|\Omega|/\Gamma)}{1+2G+(\Omega/\Gamma)^2}\right]^{1/2}, \quad (15)$$

and the maximum limiting velocity of the dragged particles

$$|v_z^{capt}|_{max}=\left(\frac{\hbar\Gamma}{M}\right)^{1/2}\left(\frac{2G}{\sqrt{1+2G}}\right)^{1/2} \quad (16)$$

is reached at the detuning equal to the field-broadened homogeneous line width:

$$|\Omega|=\Gamma(1+2G)^{1/2}. \quad (17)$$

We note that at $G\approx 1$ the condition (16) for the maximum velocity of the captured particles corresponds, apart from a factor on the order of unity, to equality of the kinetic energy of the particles to the half-width $\hbar\Gamma$ of the energy level. The depth of modulation of the velocity of the resonant particles ($|v_g|=|\Omega|/k\gtrsim\Gamma/k$) under the influence of the induced light-pressure force at $R_{rec}\ll\hbar\Gamma$ is determined by the following relation:

$$\left|\frac{\Delta v_z}{v_z}\right|=\frac{R_{rec}}{\hbar|\Omega|}\frac{G}{1+G}\left(\frac{\Gamma}{\Omega}\right)^2. \quad (18)$$

The motion of the particles in the field of a three-dimensional standing light wave is much more complicated, for in this case any projection of the light-pressure force depends on all the coordinates and on all the velocity projections of the particle. Nonetheless, the qualitative singularities of the motion of the particle remain the same as before. At $\Omega<0$ the spontaneous light-pressure force causes the resonant particles to move with a constant deceleration determined by relations (11) and (12) in the case when only one velocity projection of the particle is at resonance with the field, and by relations

$$|\dot{v}_\alpha|=\frac{2G}{1+3G}\frac{R_{rec}}{\hbar}\frac{\Gamma}{k}, \quad \text{if} \quad |\Omega|\gg\Gamma, \quad (19)$$

$$|\dot{v}_\alpha|=\frac{8G}{1+6G}\frac{R_{rec}}{\hbar}\frac{\Gamma}{k}\left(\frac{\Omega}{\Gamma}\right)^2, \quad \text{if} \quad |\Omega|\ll\Gamma, \quad (20)$$

in the case when all three particle velocity projections satisfy the condition (9). The limiting value of particle velocity projection, at which the particle is still captured at a node or antinode of a three-dimensional standing light wave also differs little from the corresponding one-dimensional expression (15), and at $R_{rec}\ll\hbar\Gamma$ we have

$$|v_\alpha^{capt}|=2\left(\frac{\hbar\Gamma}{M}\right)^{1/2}\left(\frac{3G|\Omega|/\Gamma}{1+6G+(\Omega/\Gamma)^2}\right)^{1/2}. \quad (21)$$

In this case, the maximum value of the limiting velocity projection of the captured particle

TABLE I.

Resonant transition	Mg^{24} $3^1S_0-3^1P_1$	Ca^{40} $4^1S_0-4^1P_1$	Sr^{88} $5^1S_0-5^1P_1$
Wavelength λ, Å	2852.13[20]	4226.73[20]	4607[21]
Dipole moment d, D	6.04	7.24	7.05
Natural line half-width Γ, MHz	39.4	17.4	12.8
Saturation intensity I_s, W/cm²	0.442	0.060	0.034
Doppler half-width $\Delta\omega_D$, MHz (at $T=300$ K)	1600	835	516
Recoil energy $R_{rec}/\hbar$, kHz	102	28	11
Gas cooling times (at $G=1$):			
τ'_{cool}, μsec	130	550	1200
τ''_{cool}, μsec	7.6	27.3	82.4
Frequency of the oscillations of the dragged atoms Δ, kHz (at $G=1$)	2000	698	376
Temperature of the cold captured atoms T, °K (at $\Omega=-\Gamma$)	$1.9\cdot10^{-3}$	$8.3\cdot10^{-4}$	$6.1\cdot10^{-4}$

$$|v_{\alpha\,capt}|_{max}=\left(\frac{\hbar\Gamma}{M}\right)^{1/2}\left(\frac{6G}{\sqrt{1+6G}}\right)^{1/2} \tag{22}$$

is also reached at a detuning equal to the field-broadened homogeneous line width:

$$|\Omega|=\Gamma(1+6G)^{1/2}. \tag{23}$$

Finally, the velocity modulation of the resonant particle is determined by relation (18) when only one projection of the velocity is at resonance with the field, and by the relation

$$\left|\frac{\Delta v_\alpha}{v_\alpha}\right|=\frac{R_{rec}}{\hbar|\Omega|}\frac{G}{1+3G}\left(\frac{\Gamma}{\Omega}\right)^2, \tag{24}$$

when all three velocity projections satisfy Eq. (9).

4. COOLING AND CAPTURE OF ATOMS AND MOLECULES

The foregoing analysis of particle motion shows that the resonant light-pressure forces can be used for an effective cooling and capture of atoms and molecules in the field of a standing light wave. Let us examine in detail the most important case of a three-dimensional standing light wave. It is clear from (11) and (19) and also from (18) and (24), that in the case of adiabatic change of frequency of the three-dimensional standing light wave from $\omega_{in}\approx\omega_0-\Delta\omega_D$ to $\omega_f\approx\omega_0-\Gamma$, practically all the particles can be slowed down by the spontaneous light-pressure force to velocities corresponding to the natural line width of the resonant transition. The maximum rate of cooling of the gas will be reached when the field frequency is scanned linearly in time, the minimum cooling time being

$$\tau'_{cool}=\frac{1+3G}{2G}\frac{1}{\Gamma}\frac{|\Omega_{in}-\Omega_f|}{R_{rec}/\hbar}\approx\frac{1+3G}{2G}\frac{\Delta\omega_D}{R_{rec}/\hbar}\frac{1}{\Gamma}. \tag{25}$$

For example, for optical transitions of atoms at typical values $\Delta\omega_D\sim10^3$ MHz, $R_{rec}/\hbar\sim10^{-2}$–10^{-1} MHz, and $\Gamma\sim1$–10 MHz, and at a field intensity corresponding to a saturation parameter $G\sim1$, the minimum cooling time is $\tau'_{cool}\sim10^{-2}$–10^{-4} sec. For strong optical transitions of the atoms (see Table I) the cooling time is even shorter, $\tau'_{cool}\sim10^{-4}$–10^{-3} sec, and is comparable or even shorter than the time between the collisions in the case of gas pressures $\lesssim10^{-3}$ Torr.

Cooling of particles by adiabatic scanning of the field frequency is meaningful and possible only up to velocities $|v|\sim\Gamma/k$, i.e., at field frequency deviations down to detunings $\sim\Gamma$. With further decrease of the detuning the spontaneous light-pressure force tends to zero and the cooling efficiency decreases sharply. If, however, the detuning is fixed, then the particles slowed down to $v\sim\Gamma/k$ will continue to be cooled by the force of the spontaneous light pressure.

The process of further cooling of the particles is determined by two factors: cooling under the influence of the average value of the spontaneous light-pressure force, which leads to a narrowing of the velocity distribution function, and fluctuations of the spontaneous light-pressure force, which lead to a spreading of the velocity distribution. The cold-particle velocity distribution function determined by the indicated factors can be obtained by a method that makes use of the solution of the Langevin stochastic equation[15]

$$\dot{v}=-gv+\zeta(t), \tag{26}$$

where

$$g=\frac{8R_{rec}}{\hbar}\frac{G|\Omega|/\Gamma}{[(\Omega/\Gamma)^2+1][(\Omega/\Gamma)^2+1+6G)]}, \tag{27}$$

and the first term in the right-hand side of (26) describes the average force acting on the slow particle ($|v|\ll\Gamma/k$), while the second term describes a random force independent of the particle velocity, with a mean value $\langle\zeta(t)\rangle=0$. The fluctuations of the spontaneous light-pressure force are due to two physically different causes: fluctuations of the number of photons absorbed by the particle, and the spontaneous emission of light with equal probability in all directions. In the case of a three-dimensional standing light wave, the contributions of both processes to the mean squared particle velocity are equal,[12] and the diffusion coefficient is

$$D=\frac{2}{3}n\left(\frac{\hbar k}{M}\right)^2, \tag{28}$$

where

$$n=\rho_{ee}2\Gamma=6G\frac{\Gamma^2}{\Omega^2+\Gamma^2(1+6G)}\Gamma \tag{29}$$

which is the average number of photons scattered by the cold particle per unit time, should be twice as large as the diffusion coefficient of the process of random walk of the particle in velocity space.[16] Taking (27) and

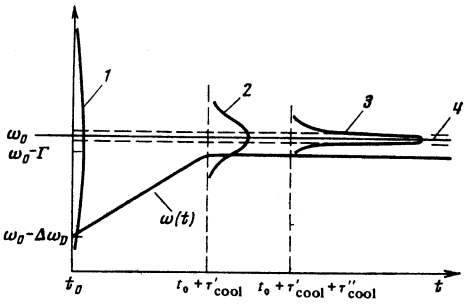

FIG. 2. Time variation of the frequency $\omega(t)$ of a three-dimensional standing light wave and the corresponding narrowing of the particle velocity distribution function. The widths of the distribution functions correspond to: 1—$2(2kT/M)^{1/2}$; 2—$2\Gamma/k$; 3—$2(2\hbar\Gamma/M)^{1/2}$; 4—capture region of width $2(2\hbar\Gamma/M)^{1/2}(3G/(1+3G))^{1/2}$.

(28) into account, the velocity distribution function of the cold particles is described by the Maxwellian distribution

$$W(v,t)=\left[\frac{\pi D}{g}(1-\exp(-2gt))\right]^{-1/2}\exp\left[-\frac{[v(t)-v(0)\exp(-gt)]^2}{(D/g)(1-\exp(-2gt))}\right] \tag{30}$$

which narrows down with a characteristic time $\tau=g^{-1}$ to the limiting half-width (of the mean squared velocity)

$$u=\left(\frac{D}{g}\right)^{1/2}=\left(\frac{\hbar\Gamma}{M}\right)^{1/2}\left(\frac{|\Omega|}{\Gamma}+\frac{\Gamma}{|\Omega|}\right)^{1/2}. \tag{31}$$

The minimal mean squared velocity of the cold particles

$$u_{min}=(2\hbar\Gamma/M)^{1/2} \tag{32}$$

is reached according to (31) at a detuning $|\Omega|=\Gamma$. The cooling time at the indicated detuning $\Omega=-\Gamma$,

$$\tau''_{cool}=\frac{1+3G}{4G}\frac{1}{R_{rec}/\hbar}\ln\frac{\hbar\Gamma}{4R_{rec}} \tag{33}$$

turns out to be much shorter than the adiabatic-cooling time τ'_{cool}. For the values of R_{rec} indicated above, at $G\sim 1$, we have $\tau''_{cool}\sim 10^{-5}$–$10^{-4}$ sec.

Thus, in the case of adiabatic variation of the frequency of the three-dimensional standing light wave, from a detuning $\Omega=-\Delta\omega_D$ to a detuning $\Omega=-\Gamma$, and at a subsequent fixed detuning $\Omega=-\Gamma$, practically all the particles in the low-gas pressure turned out to be slowed down by the spontaneous light-pressure force. Since, in addition, the limiting velocities of the dragged particles at $\Omega=-\Gamma$

$$|v_\alpha^{capt}|=\left(\frac{2\hbar\Gamma}{M}\frac{3G}{1+3G}\right)^{1/2} \tag{34}$$

practically coincide with the minimal mean squared velocity of the cold particles, it follows that an appreciable fraction of the cold particles is captured at antinodes of the light wave (Fig. 2).

We note that by using relation (32) in the form

$$u_{min}=(2kT/M)^{1/2}, \tag{35}$$

where k is Boltzmann's constant, we can introduce the concept of the temperature of the cold particles

$$T=\hbar\Gamma/k. \tag{36}$$

For the values of Γ given above, this temperature is $T\sim 10^{-4}$–10^{-5} K.

Attention should be called to an important conclusion of the stochastic analysis of radiative cooling of particles with minimum kinetic energy of the cold particles $E_{kin}^{min}=\hbar\Gamma$. From qualitative considerations one should expect cooling to the recoil energy R_{rec}. Actually this is not the case, because the particle cooling due to the resonant character of the particle interaction with a field at half-width Γ is bounded by the diffusion growth of particle energy as a result of the fluctuations in the emission directions and the number of the absorbed photons.

5. OPTICAL SPECTRA OF COLD CAPTURED PARTICLES

The slowing down of the particles and the limitation of their spatial motion by the light-pressure forces leads to a substantial change in the emission (absorption) spectra of a low-pressure gas. We note immediately two singularities in the registration of narrow lines in the spectra of cold captured particles. First, from the point of view of ultrahigh resolution spectroscopy, the observation of the optical spectra is productive only on transitions that have nothing in common with the resonant transition of the level. If this condition is not satisfied, the absorption (emission) line on the trial transition will be broadened because of the dynamic Stark effect[14] to a value determined by the widths $2\Gamma(1+G)^{1/2}$ of the resonant transition. Second, registration of nonzero absorption on such an unbound trial transition is possible only for those cold captured particles which were prepared beforehand, in special mixed states, prior to the start of the cooling process. In a mixed initial state the probability amplitude of the particle is different from zero, at least for one level of the resonant transition and for one level of the trial transition. In this case, obviously, the initial probability amplitude of finding the particle on one of the levels of the resonant transition 1–2 (for example, $a_1(t=0)\neq 0$) will be responsible for the cooling and capture of the particle by the strong resonant field, while the probability amplitude of finding the particle on one of the levels of the trial transition (for example, $a_3(t=0)\neq 0$) will be responsible for the absorption (emission) on the trial transition 3–4 in the weak field (Fig. 3).

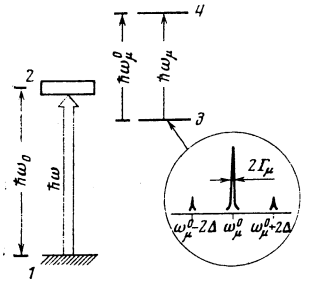

FIG. 3. Scheme of the particle energy levels used for cooling and capture on the resonant transition 1–2 and for observation of a narrow spectral line on the trial transitions 3–4.

From the classical point of view, the changes in the spectra of the cold captured particles are due to the frequency modulation of the spectrum as a result of periodic motion of the particles near the antinodes of the field, with a frequency (at $\Omega = -\Gamma$)

$$\Delta = \left(\frac{R_{rec}}{\hbar}\Gamma\frac{4G}{1+3G}\right)^{1/2}. \quad (37)$$

The most appreciable change in the spectra can be expected when the natural line width $2\Gamma_\mu$ of the trial transition is much less than the modulation frequency Δ. In this case the emission (absorption) line on the trial transition splits into a number of components, the distances between which are multiples of the oscillation frequency of the cold captured particles, and whose widths are determined by the natural width of the investigated transition. In the opposite case, $2\Gamma_\mu \gtrsim \Delta$, the components of the spectra are superimposed on one another and only line broadening takes place.

Assuming that for the trial transition we have $2\Gamma_\mu \ll \Delta$, let us consider the intensity of the spectral components, following the classical approach used by Shapiro[17] to estimate the Mössbauer effect. Assume that a trial transition of frequency ω_μ^0 interacts with a light wave

$$\mathbf{E}_\mu = \mathbf{E}_\mu^0 e^{-i\varphi}, \quad (38)$$

the propagation of which is determined by the wave vector $\mathbf{k}_\mu$. The oscillations of one particle along the wave vector of the trial field

$$r_\mu(t) = r_\mu^0 \sin\Delta t = (v_\mu/\Delta)\sin\Delta t \quad (39)$$

modulates the phase of the wave and this, in turn, leads to a change in the amplitude of the field

$$\mathbf{E}_\mu = \mathbf{E}_\mu^0 e^{-i\omega_\mu^0 t}\exp\left(-i\frac{v_\mu}{\lambda\!\!\!^{-}_\mu\Delta}\sin\Delta t\right) = \mathbf{E}_\mu^0 e^{-i\omega_\mu^0 t}\sum_{m=-\infty}^{+\infty} J_m\left(\frac{v_\mu}{\lambda\!\!\!^{-}_\mu\Delta}\right)e^{im\Delta t}. \quad (40)$$

Here $\lambda\!\!\!^{-}_\mu = \lambda_\mu/2\pi$ is the relative length of the light wave, v_μ is the projection of the particle velocity on the vector $\mathbf{k}_\mu$ in the antinode of the resonant light wave. The combined spectrum of the cold captured particles is determined, according to (40), by the expression

$$I_\mu = I_\mu^0 \sum_{m,n=-\infty}^{+\infty} \overline{J_m\left(\frac{v_\mu}{\lambda\!\!\!^{-}_\mu\Delta}\right)J_n\left(\frac{v_\mu}{\lambda\!\!\!^{-}_\mu\Delta}\right)}\, e^{i(m-n)\Delta t}, \quad (41)$$

where the bar over the Bessel functions denotes averaging over the projections of the particles velocities.

The most interesting from the point of view of ultrahigh resolution spectroscopy is the case when the averaged particle oscillation amplitude u/Δ is smaller than the length $\lambda\!\!\!^{-}_\mu$ of the trial light wave. Under this condition, the most intense is the undisplaced line of the spectrum, with frequency ω_μ^0. The intensity $I(\omega_\mu^0)$ of the undisplaced line can be easily obtained from (41) by using the asymptotic form of the Bessel functions. Accurate to the square of the ratio $u_{min}/\lambda\!\!\!^{-}_\mu\Delta \sim \omega_\mu^0/\omega_0$ we have

$$\frac{I(\omega_\mu^0)}{I_\mu^0} = 1 - \frac{1+3G}{4G}\left(\frac{\omega_\mu^0}{\omega_0}\right)^2. \quad (42)$$

The intensities of the components having frequencies $\omega_\mu^0 \pm k\Delta$, for odd k, are equal to zero in the considered approximations. Therefore the closest to the undisplaced lines are components with frequencies $\omega_\mu^0 \pm 2\Delta$, whose intensities, likewise accurate to the square of the ratio $u_{min}/\lambda\!\!\!^{-}_\mu\Delta$, are

$$\frac{I(\omega_\mu^0 \pm 2\Delta)}{I_\mu^0} = \frac{1+3G}{8G}\left(\frac{\omega_\mu^0}{\omega_0}\right)^2. \quad (43)$$

We note that even though the experimental observation of the absorption spectra of the cold captured particles is possible only in one selected direction, the trial light field itself, just as a resonant field, should form a three-dimensional standing light wave, for only in such a field configuration will the particles be unable to leave the capture region under the influence of the recoil of the trial field.

6. CONCLUSIONS. APPLICATIONS OF THE METHOD

The foregoing analysis shows that cooling and capture of particles by a resonant light field can become one of the methods of laser spectroscopy of ultrahigh resolution, making possible observation, in optical spectra of atoms and molecules, of narrow spectral lines whose positions are determined only by the frequencies of the trial transitions, and whose width is determined only by the natural broadening of the spectral lines. Naturally, the realization of the considered method is possible only for those particles for which the resonant and trial transitions have no common level and which can be brought with sufficient efficiency to superposition states.

In addition to the possibility of substantially increasing the resolving power of laser spectroscopy, this method is also of considerable interest for a radial increase in the sensitivity of detection of atoms and for spectroscopic investigations of exceedingly small numbers of atoms. It is known that modern methods of detection of low atom concentrations, for example, fluorescence methods, make it possible to detect $\sim 10^2$–10^3 at/cm^3.[18,19] In this case, however, the actual number of atoms employed, owing to their escape from the investigated volume, for example to the walls of the evacuated cell, within a time $\sim 10^{-3}$–10^{-5} sec, amounts to $\sim 10^5$–10^8 at/cm^{2}sec, and it is actually necessary to operate with a much larger number of atoms. The use of the method of cooling and capture of particles, owing to the abrupt increase of the time of containment of the particles in the investigated volume, should greatly decrease the number of atoms needed for their detection. For example, for a vacuum with a residual pressure $p \sim 10^{-10}$ Torr and at an average thermal velocity of their residual particles $u \sim 10^4$ cm/sec, for a typical total particle scattering cross section $\sigma \sim 10^{-14}$ cm^2 the time of containment of the cold captured atoms should be

$$\tau = (\sigma n u)^{-1} \sim 1 \text{ hour}.$$

It should be noted that this use of the method can be of particular interest for spectroscopic investigations of elements that do not exist in nature, for example, for

monatomic spectroscopy of artificially obtained atoms of transuranium elements.

We note finally that the most suitable objects for the first investigations by this method are apparently even isotopes of alkaline-earth metals, in which one can use as resonant transitions the strong allowed transitions of the $n^1S_0 - n^1P_1$ type. The main characteristics of the cooling and capture process for three such isotopes are given in the table.

The power level required to saturate the resonant transitions of the isotopes in question is of the order of 100 mW at wavelengths that are quite attainable with tunable cw dye lasers. The laser-radiation line widths needed for the first experiment (much smaller than the natural width, for example on the order of 1 MHz) are likewise perfectly attainable in the cw regime. All this makes the new approach to laser spectroscopy, demonstrated in the present paper, fully realistic.

[1)]This force is called "gradient force" in[6,7]. This designation should be regarded as less appropriate, since it indicates essentially only the formal method of obtaining the corresponding expression.

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Cooperative effect in Raman scattering of light

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A study is made of the influence of the interatomic correlation which arises in the process of emission of radiation on the spontaneous Raman scattering of light. It is shown that in the resonance scattering of a quasimonochromatic field, the shape of the Stokes component of the scattered pulse is identical with a superradiance pulse of the kind which appears in a medium of two-level atoms. If allowance is made for the anti-Stokes component, the scattering process becomes oscillatory.

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1. INTRODUCTION

Spontaneous emission from closely spaced atoms cannot be regarded as emission from isolated atoms because in the course of emission the atoms begin to interact with one another via the radiation field. The first quantum theory of the cooperative spontaneous emission from atoms was given by Dicke in[1]. He showed that the rate of cooperative emission depends on the distribution of the phases of atomic oscillators and can vary from γ_0 to $\gamma_0 N$, where γ_0 is the rate of the spontaneous decay of an excited state of an isolated atom and N is the number of atoms. Subsequently several authors[2–4] studied the temporal and spatial characteristics of cooperative spontaneous emission of radiation from atoms in macroscopic regions. It was shown in particular[2,3] that, in the quasiclassical approximation in the absence of an interatomic correlation at the initial moment, the radiation pulse has the shape of a hyperbolic secant on the time axis with a characteristic time $\tau_c \sim (\gamma_0 N/2)^{-1}$

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