

Light-induced gas drift under conditions of pulsed periodic excitation

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The drift of gases under the action of short light pulses is theoretically investigated. The phenomenon is due to the selective—with respect to the velocities—excitation of the particles and the fact that the particles in the ground state and those in the excited state elastically collide with the buffer-gas particles at different rates. The conditions under which the effect manifests itself most strongly are found, and it is shown that under these conditions the drift velocity constitutes an appreciable fraction of the most probable thermal velocity of the particles. The cases of dynamic and stochastic pulsed periodic excitation are analyzed. It is found that, in the case of the dynamic excitation, the drift can be reversed by changing the radiation intensity, an effect which has no analog in the continuous regime. The characteristics of the drift phenomenon are discussed in the cases in which it occurs in molecular and atomic media. It is shown that the highest drift velocities can be attained at moderate pulse repetition rates and radiation intensities.

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1. INTRODUCTION

Gel'mukhanov *et al.*^{1,2} have theoretically predicted and experimentally detected a new physical phenomenon: light-induced drift (LID) of gases. The essence of LID consists in the appearance of directed macroscopic motion of a gas during its interaction with a traveling electromagnetic wave that is at quasiresonance with some transition of the particles from the ground state. Here the presence in the system of an extraneous (buffer) gas is essential. The appearance of the drift is due to the selective—with respect to the velocities—optical excitation through the Doppler effect of the absorbing-gas particles and the fact that the particles in the excited state and those in the unexcited state elastically collide with the buffer-gas particles at different rates $\nu_{m,1}$ and $\nu_{n,1}$. Under optimal conditions the LID velocity constitutes an appreciable fraction of the characteristic thermal velocities. The direction of the drift coincides with, or is opposite to, the direction of propagation of the radiation, depending on the ratio of the rates ν_m and ν_n and the on sign of the detuning of the radiation frequency from the frequency of the corresponding transition.

The occurrence of the LID of gases under conditions of stationary excitation (SE), i.e., under the action of continuous radiation, are investigated in Refs. 1-6. At the same time, the study of this interesting phenomenon under conditions of pulsed periodic excitation (PPE) is important, since the use of pulsed light sources allows us to significantly broaden the possibilities of its experimental investigation and the region of its applications. The present paper is devoted to the theoretical analysis of the indicated problem.

In Sec. 2 we derive general expressions for the velocity distribution function and the mean drift velocity of the absorbing gas under conditions of excitation by short light pulses. The analysis shows that, in the case of pulsed periodic radiation with a large off-duty factor $\tau(\nu\tau)^{-1} \gg 1$ (τ is the pulse duration and ν is the pulse repetition rate), the LID velocity can be fairly high, al-

though on the face of it the smallness parameter α^{-1} should arise when we go over from SE to PPE. This is due to the fact that the anisotropy induced by the field of a short pulse in the velocity distributions for the particles at the energy levels exists during a time determined by the characteristic relaxation times ν_j^{-1} and $\nu_j^{-1} [\nu_j$ is the decay (quenching) rate of the excited state; $j = m, n]$. During this same time the velocity distribution function for all the absorbing particles is anisotropic, an anisotropy which develops after the passage of the pulse as a result of the difference in the rates ν_m and ν_n , and manifests itself as a LID of the gas. Thus, if the pulse repetition rate ν is close to the rates ν_m and ν_n , the LID velocity under PPE conditions can differ only insignificantly from the drift velocity in the SE regime.

Section 3 is devoted to the analysis of the LID phenomenon under conditions of coherent excitation by "rectangular" pulses with determinate field characteristics. The most interesting characteristic here is the possibility of reversing the drift by changing the field intensity I_i or the pulse duration τ . This effect is due to the following circumstances. In the case of coherent excitation by a "rectangular" pulse the populations of the energy levels of a particle oscillate with the Rabi frequency.⁷ This frequency is determined by both the pulse-field intensity I_i and the radiation-frequency detuning (with allowance for the Doppler shift) relative to the transition frequency. The latter circumstance gives rise to an energy-level-population-oscillation-phase difference between particles moving with different velocities. Accordingly, depending on I_i and τ , different velocity-defined groups of particles are preferentially excited, which leads to the dependence of the direction of drift on the indicated characteristics of the pulsed periodic radiation.

Section 4 is devoted to the investigation of the LID of gases under the action of short pulses whose characteristics undergo random fluctuations. The analogy between the effect occurring in the indicated regime and the effect occurring in the SE regime is followed.

In Sec. 5 we discuss the characteristics of the LID of gases in the PPE regime in the cases of molecular and atomic media. It is shown that significant drift velocities can be realized at moderate pulse repetition rates and moderate radiation intensities.

2. GENERAL EXPRESSIONS

Let us consider the interaction of a gas of two-level particles (one being the ground level) with a traveling electromagnetic wave:

$$E(x, t) = \frac{1}{2} [e(x, t)e^{-i(\omega t - kv)} + e^*(x, t)e^{i(\omega t - kv)}].$$

Below we shall label the ground state of the particles by the subscript n ; the excited state, by m . Under the assumption that the system contains a large quantity of buffer gas, this interaction is described, when the strong-collision model⁸ is used, by the following system of kinetic equations for the density matrix:

$$\begin{aligned} (\partial/\partial t + \nabla \nabla - i(\omega_0 + kv) + \Gamma) \rho_{nm}(v, r, t) &= iV^* e^{i\omega t} [\rho_n(v, r, t) - \rho_m(v, r, t)], \\ (\partial/\partial t + \nabla \nabla + v_m + v_n) \rho_m(v, r, t) &= v_m W(v) \rho_n(r, t) \\ &\quad - 2 \operatorname{Re} [iV e^{-i\omega t} \rho_{nm}(v, r, t)], \\ (\partial/\partial t + \nabla \nabla + v_n) \rho_n(v, r, t) &= v_n W(v) \rho_n(r, t) \\ &\quad + v_r \rho_m(v, r, t) + 2 \operatorname{Re} [iV e^{-i\omega t} \rho_{nm}(v, r, t)]. \end{aligned} \quad (2.1)$$

Here $V = -\varepsilon(r, t) d_{mn}/2\hbar$, d_{mn} is the matrix element of the dipole moment for the m - n transition, ω_0 and Γ are the frequency and the homogeneous halfwidth of this transition,

$$W(v) = (\pi^{1/2} v_0)^{-1} \exp[-(v/v_0)^2]$$

is the Maxwell velocity distribution, v_0 is the most probable thermal velocity, and

$$\rho_j(r, t) = \int \rho_j(v, r, t) dv, \quad j = m, n.$$

The symbols ν_0 , ν_m , and ν_n are explained above. The collisions of the absorbing particles with each other is neglected in view of the assumption that the relative concentration of these particles is low. The system of equations (2.1) allows us to investigate the LID effect for gases in the field of pulsed periodic radiation. We shall assume that the radiation is a train of identical pulses of duration τ , following each other at the rate of ν , and that the field amplitude is equal to zero in the intervals between the pulses. The medium is assumed to be optically thin. Here we consider the case in which the irradiated cell is located in a reservoir containing the parent gas mixture, and the absorbing and buffer gases can freely flow in and out of it through its open ends. On account of the smallness of the relative concentration of the absorbing particles, we neglect the deviation of the state of the buffer gas from the equilibrium state, and shall hereinafter be interested only in the state of the absorbing gas in the cell.

Let the following conditions be fulfilled along with the above-indicated ones:

$$\tau \ll \nu_q^{-1}, \nu_j^{-1}, \Gamma^{-1} \ll \nu^{-1}; \quad l/c \ll \nu_q^{-1}, \nu_j^{-1}. \quad (2.2)$$

Here l is the length of the cell and c is the velocity of light. The fulfillment of the conditions (2.2) implies the following: 1) the gas has time to return to the equilibrium state in the intervals between the pulses (the possibility of the free flow of the gas through the open cell ends, a possibility which precludes the appearance

of a density gradient, has been taken into account); 2) the velocity distributions for the particles at the energy levels after the passage through the cell of any radiation pulse are spatially homogeneous; 3) the relaxation of the velocity distributions for the particles in the m and n states occurs largely in the intervals between the pulses, the respective distribution functions remaining spatially homogeneous during the relaxation. It is clear from the foregoing that, under the conditions in question, the excitation of the medium by each pulse and the subsequent relaxation occur in entirely the same fashion. Therefore, the macroscopic gas velocity u averaged over a long period of time is equal to the velocity averaged over the period ν^{-1} . Accordingly, to compute the drift velocity u , it is sufficient to analyze the processes accompanying the passage through the medium of any one radiation pulse. Let us proceed to the analysis of this question.

Under the conditions in question the velocity distributions of the particles at the energy levels immediately after the passage through the cell of a radiation pulse coincide with the corresponding distributions for the particles at the inlet ($x = 0$) end of the cell. The latter can, if the rate at which the pulse-field-induced processes proceed is higher than the rates of the relaxation processes (the criteria are indicated in Secs. 3 and 4), be found from the following system of equations:

$$\begin{aligned} (\partial/\partial t - i(\omega_0 + kv)) \rho_{nm}(v, t) &= iV^*(t) e^{-i\omega t} [\rho_n(v, t) - \rho_m(v, t)], \\ (\partial/\partial t) \rho_m(v, t) &= -2 \operatorname{Re} [iV(t) e^{-i\omega t} \rho_{nm}(v, t)], \\ (\partial/\partial t) \rho_n(v, t) &= 2 \operatorname{Re} [iV(t) e^{-i\omega t} \rho_{nm}(v, t)] \end{aligned} \quad (2.3)$$

(here $V(t) = -\varepsilon(0, t) d_{mn}/2\hbar$) with the initial conditions

$$\rho_{nm}(v, 0) = 0, \quad \rho_m(v, 0) = 0, \quad \rho_n(v, 0) = W(v). \quad (2.4)$$

The system of equations (2.3) is obtained from (2.1) by discarding the relaxation terms and the terms containing space derivatives. The initial conditions (2.4) correspond to the equilibrium state of the gas. Evidently, the solutions to the equations (2.3) depend essentially on the form of the function $V(t)$, which is determined by the characteristics of the pulse field. In Secs. 3 and 4 we shall give the solutions corresponding to "rectangular" pulses with determinate and fluctuating field characteristics. Now let us arbitrarily denote the required solutions by $\rho_m(v, \tau)$ and $\rho_n(v, \tau)$, and proceed to the investigation of the relaxation processes.

The relaxation of the distributions $\rho_m(v, \tau)$ and $\rho_n(v, \tau)$ realized immediately after the passage of a field pulse through the cell is described by the following system of equations:

$$\begin{aligned} (\partial/\partial t + v_m + v_n) \rho_m(v, t) &= v_m W(v) \rho_n(t), \\ (\partial/\partial t + v_n) \rho_n(v, t) &= v_n W(v) \rho_n(t) + v_r \rho_m(v, t), \\ \rho_j(t) &= \int \rho_j(v, t) dv, \quad j = m, n \end{aligned} \quad (2.5)$$

with the initial conditions

$$\rho_m(v, 0) = \rho_m(v, \tau), \quad \rho_n(v, 0) = \rho_n(v, \tau). \quad (2.6)$$

The system (2.5) is obtained from (2.1) by neglecting the field terms [this also allows us to exclude the first of the equations (2.1) from the analysis] and the terms containing space derivatives. The solutions to the equations

tions (2.5) have the form

$$\begin{aligned}\rho_m(v, t) &= Y(v, \tau) \exp \{-(v_m + v_q)t\} + \rho_m(\tau) W(v) \exp(-v_q t), \\ \rho_n(v, t) &= \frac{v_q Y(v, \tau)}{v_n - v_m - v_q} \left[\exp \{-(v_m + v_q)t\} - \frac{v_n - v_m}{v_q} \right. \\ &\quad \left. \times \exp(-v_n t) \right] + W(v) [1 - \rho_m(\tau) \exp(-v_q t)], \\ Y(v, \tau) &= \rho_m(v, \tau) - \rho_n(\tau) W(v), \quad \rho_m(\tau) = \int_{-\infty}^{\infty} \rho_m(v, \tau) dv.\end{aligned}\quad (2.7)$$

The distribution functions $\rho_j(v, t)$ can each be split up into two parts: one isotropic (i.e., Maxwellian), and the other anisotropic (i.e., selective) and proportional to $\rho_n(v, \tau)$. The selective part of the distribution function $\rho_n(v, t)$ "dies out" over a period of time equal to $(\nu_m + \nu_q)^{-1}$. This is due to the fact that its relaxation is caused both by the "mixing" of the velocities as a result of the elastic collisions and by the transitions into the state n . Then the velocity-integrated population of the level m relaxes with the time constant ν_q^{-1} . The interpretation of the expression for $\rho_n(v, t)$ is complicated by the fact that for the level n , besides the relaxation processes that occur at the level itself, the arrival from the level n is important.

From (2.7) we find the velocity distribution function $\rho(v, t) = \rho_m(v, t) + \rho_n(v, t)$ for all the absorbing particles:

$$\rho(v, t) = W(v) + \frac{v_n - v_m}{v_n - v_m - v_q} Y(v, \tau) [\exp \{-(v_m + v_q)t\} - \exp(-v_n t)]. \quad (2.8)$$

The deviation of the distribution (2.8) from Maxwellian is due to the inequality of the rates ν_m and ν_n and the selective—with respect to the velocities—optical excitation of the particles. As noted above, the drift velocity averaged over a long period of time is equal to the drift velocity averaged over the time ν^{-1} :

$$u = v \int_0^{\infty} dt \int f(v, t) v dv. \quad (2.9)$$

Thus, from (2.8) and (2.9), allowing for the fact that $\nu \ll \nu_q, \nu_j$, we find

$$u = \frac{v_n - v_m}{v_n} \frac{v}{v_n + v_q} \int v \rho_m(v, \tau) dv. \quad (2.10)$$

From (2.10) we can (allowing for the fact that in real situations $\nu_m \sim \nu_q$) easily see that the drift velocity attains its maximum values when $|(\nu_n - \nu_m)/\nu_n| \sim 1$ and the values of the parameters ν_m and ν_q (allowed by the condition $\nu \ll \nu_q, \nu_j$) are as close as possible to ν . This is due to the fact that, as follows from (2.8), the nonequilibrium character of the velocity distribution function for the particles is most strongly pronounced in the indicated case, and the relaxation time of the function constitutes a significant fraction of the pulse repetition time.

Let us write out for comparison the expression giving the drift velocity under SE conditions³:

$$u_{SE} = \frac{v_n - v_m}{v_n} \int v \rho_m(v) dv. \quad (2.11)$$

Here $\rho_m(v)$ is the velocity distribution function for the particles in the state m in the SE regime. It is clear from a comparison of (2.10) and (2.11) that, when the conditions (2.2) are fulfilled, the smallness parameter in the PPE regime is the factor $\nu(\nu_m + \nu_q)^{-1}$, and not the reciprocal off-duty factor $\alpha^{-1} = \nu\tau$ for the pulsed period-

ic radiation. Further, within the framework of (2.2) $\nu(\nu_m + \nu_q)^{-1} \gg \nu\tau$, and the values of $\nu(\nu_m + \nu_q)^{-1} \sim 10^{-1}$ are admissible when the parameter α^{-1} is arbitrarily small. Thus, if

$$\left| \int v \rho_m(v, \tau) dv \right| \sim \left| \int v \rho_m(v) dv \right|,$$

then the situation can obtain in which $|u| \sim 10^{-1} |u_{SE}|$, which is a substantial quantity, since $|u_{SE}|$ under optimal conditions attains values of $(10^{-2} - 10^{-1})v_0$. The validity of this estimate for $|u|$ will be confirmed below.

Let us note that the obtained results have a general character, and do not depend on the specific characteristics of the pulses.

3. DYNAMIC EXCITATION

1. General relations

Let us consider the case in which the amplitude, the carrier frequency, and the initial phase of the electric field of the pulse are constant. Then the system (2.3) with the initial conditions (2.4) can be reduced to a third-order equation for the population difference

$$n(v, t) = \rho_m(v, t) - \rho_n(v, t)$$

between the states m and n :

$$\dot{n}(v, t) + \omega_R'^2 \tilde{n}(v, t) = 0, \quad \omega_R' = (4|V|^2 + \Omega'^2)^{1/2}, \quad (3.1)$$

$$\tilde{n}(v, t) = \frac{\partial n(v, t)}{\partial t}, \quad |V|^2 = \text{const}, \quad \Omega' = \Omega - x, \quad \Omega = \omega - \omega_n, \quad x = \mathbf{k} \mathbf{v}$$

with the initial conditions

$$n(v, 0) = -W(v), \quad \tilde{n}(v, 0) = 0, \quad \ddot{n}(v, 0) = 4|V|^2 W(v) \quad (3.2)$$

(Ref. 9). For $t \geq \tau$ the solution to Eq. (3.1) with the initial conditions (3.2) has the form

$$n(v, t) = n(v, \tau) = W(v) \left[\frac{4|V|^2}{\omega_R'^2} (1 - \cos \omega_R' \tau) \right]. \quad (3.3)$$

The expression (3.3) is the Rabi solution (see, for example, Ref. 7), which characterizes the dynamical variation of the population difference under the action of the field, and which takes account of the thermal motion of the particles. According to (3.3), the rate of radiation-induced transitions is determined by the quantity ω_R' . Thus, the requirement, formulated in Sec. 2, that the field-induced processes predominate over the relaxation processes has in the present case the form

$$\omega_R' \gg \nu_q, \nu_n, \Gamma \quad \text{for} \quad |x| \leq x_0 = kv_0.$$

Let us note that the solution (3.3) describes adequately the real situation only when the pulse field is switched on or off instantaneously.¹⁰

Using (3.3), we find the following expressions for $\rho_m(v, \tau)$ and $\rho_n(v, \tau)$:

$$\rho_m(v, \tau) = \frac{2|V|^2 W(v)}{\omega_R'^2} (1 - \cos \omega_R' \tau), \quad \rho_n(v, \tau) = W(v) - \rho_m(v, \tau). \quad (3.4)$$

Here we have allowed for the fact that $\rho_m(v, \tau) + \rho_n(v, \tau) = W(v)$, as follows from (2.3) and (2.4). The formulas (3.4) reflect the fact that the populations of the states m and n of each particle oscillate under the action of the pulse field with amplitude $2|V|^2/\omega_R'^2$ and frequency ω_R' that depend on the component v_x of the velocity \mathbf{v} of the particle along the wave vector \mathbf{k} of the radiation.

Connected with this are two factors that make the particle excitation selective with respect to the velocity components v_k . The oscillation amplitude of the population of the excited state is greatest and equal to $\frac{1}{2}$ in the case of particles for which $\Omega' = 0$, i.e., for which v_k is such that the corresponding Doppler shift $k v_k$ compensates for the detuning Ω . Thus, the selectivity factor connected with the dependence on v_k of the amplitude of the Rabi oscillations imposes a tendency toward preferential excitation of the "resonance" particles, and is responsible for the definite similarity that exists between the LID of gases in the dynamic PPE regime and the corresponding phenomenon occurring in the SE regime. The dependence of the frequency ω_R' on v_k produces a tendency toward preferential excitation of the particles for which v_k is such that $\omega_R' \tau = (1 + 2a)\pi$ (a are integers). This selectivity factor does not have an analog in the SE regime, and gives rise to a number of specific features that characterize the occurrence of the LID of gases under conditions of dynamic PPE. The role played by the two indicated factors in the appearance of the drift motion of the gas will be analyzed below.

Combining (3.4) and (2.10), we find for the drift velocity u and its component u along the wave vector the following expression:

$$u = \frac{k}{k} u = \frac{k}{k} \frac{2|V|^2 \beta v_0}{\pi^{1/2} x_0^2} \int_{-\infty}^{\infty} \frac{e^{-(x/x_0)^2} (1 - \cos \omega_R' \tau) x dx}{\omega_R'^2}, \quad (3.5)$$

$$\beta = \frac{v_k - v_m}{v_n} \frac{v}{v_m + v_q}.$$

It therefore follows that the drift velocity and the wave vector of the radiation are collinear, and that no drift occurs when $\Omega = 0$, which indicates that the particle excitation is symmetric about the center of the Maxwellian distribution.

2. The drift velocity for small and large τ

In the case of short pulse lengths (i.e., for $\omega_R' \tau \ll 1$ when $|x| \leq x_0$) we find from (3.5) that

$$u = |V|^2 \beta \Omega x_0 \tau^2 v_0 / 12. \quad (3.6)$$

In the present case the drift velocity is small (i.e., $|u| \ll v_0$) even for optimal β [let us recall that the conditions (2.2) admit of values of $|\beta| \sim 10^{-1}$]. This is due to the fact that, because of the condition $\omega_R' \tau \ll 1$ ($|x| \leq x_0$), only a small fraction of the particles undergo, as follows from (3.4), the transition into the excited state under the action of the pulse field. The drift is parallel or opposite to the direction of propagation of the radiation, depending on the sign of $(v_n - v_m)\Omega$, and has the direction as the drift that occurs under the same conditions [the same sign of the combination $(v_n - v_m)\Omega$] in the SE regime.³ Analysis of (3.4) shows that in the case of short τ the selectivity factor connected with the dependence on v_k of the Rabi-oscillation amplitude predominates, which leads to the indicated analogy with the SE regime.

In the other limiting case of long τ (the criteria are given below) that part of the integrand in the formula (3.5) which is proportional to the factor $\cos \omega_R' \tau$ is a rapidly oscillating function of the integration variable

x , and therefore makes a small contribution to the integral. By neglecting this small contribution, we can reduce the formula (3.5) to the form

$$u = \frac{\beta \pi^{1/2} v_0}{2} z'' \operatorname{Re} [z W(z)], \quad W(z) = e^{-z^2} \left[1 + \frac{2i}{\pi} \int_0^z e^{-t^2} dt \right]. \quad (3.7)$$

Here $W(z)$ is the probability function of the complex variable¹¹

$$z = z' + iz'', \quad z' = \Omega/x_0, \quad z'' = 2|V|/x_0.$$

Let us estimate the τ values necessary for the establishment of the regime (3.7) for different relations among the parameters x_0 , Ω , and $|V|$. In the case in which $|\Omega| \leq x_0$ and $|V| \ll x_0$ the dominant contribution to the integral in the formula (3.5) is made by the x domain where the factor $|V|^2/\omega_R'^2$, which represents a Lorentz contour with center at the point $x = \Omega$ and halfwidth $2|V|$, is significantly different from zero. In this case the variation scale of the τ -independent part of the integrand is equal to $|V|$. It is accordingly clear that the expression (3.5) assumes the form (3.7) if the parameter $\cos \omega_R' \tau$ undergoes in the indicated region oscillations whose scale is very small in comparison with $|V|$. The corresponding criterion, as is easy to verify, has the form

$$|V| \tau \gg 1. \quad (3.8)$$

In all the other cases the region that makes the dominant contribution to the integral and the variation scale of the τ -independent part of the integrand is determined by the Gaussian factor $\exp -(x/x_0)^2$. Arguments similar to those presented above lead, for $|\Omega| \leq x_0$ and $|V| \geq x_0$, to the criterion

$$|2|V| - (4|V|^2 + x_0^2)^{1/2}| \tau \gg 1, \quad (3.9)$$

and, for $|\Omega| \gg x_0$ and arbitrary values of $|V|$, to the criterion

$$|\Omega| x_0 \tau / \omega_n \gg 1, \quad \omega_n = (4|V|^2 + \Omega^2)^{1/2}. \quad (3.10)$$

Thus, in the case of pulse lengths τ that are long [in the sense of (3.8)–(3.10)], but satisfy the conditions (2.2), the drift velocity does not depend on τ , and is described by the formula (3.7). Analysis of the factor $z'' \operatorname{Re} [z W(z)]$ with the use of the tables of Ref. 11 shows that in magnitude this factor attains its maximum values, which are close to 0.1, when $|z'| \approx z'' \approx 1$. Thus, when $|\beta| \sim 10^{-1}$, and the indicated optimal conditions are satisfied, the drift velocity attains values $|u| \sim 10^{-2} v_0$, which confirms the estimate obtained in Sec. 2. It follows from (3.7) that the direction of the drift is determined, as in the case of small τ , by the sign of the combination $(v_n - v_m)\Omega$, and has the same direction as the drift that occurs in the SE regime.

It can be seen from (3.4) that, for large τ , the oscillation phases of the populations of the excited states of particles with arbitrarily close v_k values may differ significantly from each other. It is precisely this circumstance that is manifested in the occurrence of the very-small-scaled oscillations of the τ -dependent part of the integrand in the formula (3.5) as a function of x . The selectivity factor connected with the dependence of ω_R' on v_k accordingly imposes a tendency toward equiprobable excitation of the particles with positive and

negative v_k values, and does not guarantee the excitation asymmetry necessary for the appearance of LID [see the formula (2.10)]. Thus, in the case of large τ the drift motion owes its appearance only to the selectivity factor connected with the dependence of the Rabi-oscillation amplitude v_k . As a result, the LID occurring in the dynamic PPE regime in the case of large τ and the LID occurring in the SE regime bear a similarity to each other that extends to the formulas describing the drift velocities [see the formula (3.7) in the present paper and the formula (3.1) in Ref. 6].

Let us proceed to the analysis of those cases in which the dependence of the Rabi frequency on the particle-velocity component along the radiation wave vector can play an important role.

3. The drift velocity in the case of large detunings or strong fields

In the case of large detunings (i.e., for $|\Omega| \gg x_0$) or strong fields ($|V| \gg x_0$) the expression (3.4) for $p_m(v, \tau)$ in the region $|x| \gg x_0$ can be represented in the form

$$p_m(v, \tau) = \frac{2|V|^2 W(v)}{\omega_R^2} (1 + 4qy - sy^2) \{1 - \cos[\omega_R \tau (1 - 2qy + py^2)]\}, \quad (3.11)$$

$$s = \frac{\Omega x_0}{2\omega_R^2}, \quad q = \frac{x_0^2}{\omega_R^2} \left(1 - \frac{4\Omega^2}{\omega_R^2}\right), \quad p = \frac{2|V|^2 x_0^2}{\omega_R^4}, \quad y = \frac{x}{x_0}.$$

Let us note that the representation (3.11) is valid only when the conditions

$$|V|^2 |\Omega| x_0^2 \tau / \omega_R^3 \ll 1, \quad |V|^2 x_0^2 \tau / \omega_R^3 \ll 1, \quad (3.12)$$

which indicate the smallness of the "advance" of the population oscillation phase as a result of the presence of the terms of higher order in y than the quadratic term, are satisfied.

It follows from (3.11) that, under the conditions in question, the Rabi-oscillation amplitude depends weakly on the velocity component v_k . Therefore, it is *a priori* clear that the selectivity, connected with this dependence, of the particle excitation is insufficient for the appearance of high-velocity drift. The Rabi-oscillation phase for sufficiently large values of the parameter $\omega_R \tau$ depends essentially on v_k . This dependence, as will be seen below, guarantees a high excitation selectivity, and results in a strong manifestation of the LID effect.

Combining (3.11) and (2.10), we find that

$$v_m = \frac{2|V|^2 p v_0}{\pi^2 \omega_R^2} \int_{-\infty}^{\infty} e^{-v^2 y} dy (4qy - (1 + 4qy - sy^2) \cos[\omega_R \tau (1 - 2qy + py^2)]). \quad (3.13)$$

Let us consider in greater detail the case of large detunings in arbitrary fields. Under these conditions we can neglect in (3.11) the terms quadratic in y in comparison with the linear terms, and require in addition that

$$|V|^2 x_0^2 \tau / \omega_R^3 \ll 1. \quad (3.14)$$

In this approximation, it follows from (3.13) that

$$v_m = \frac{2|V|^2 \Omega x_0 p v_0}{\omega_R^2} \left[1 - \frac{\omega_R \tau}{2} \sin(\omega_R \tau) \exp\left\{-\left(\frac{\Omega x_0 \tau}{2\omega_R}\right)^2\right\} + D_2\left(\frac{\Omega x_0 \tau}{2\omega_R}\right) \cos(\omega_R \tau) \exp\left\{-\left(\frac{\Omega x_0 \tau}{2\omega_R}\right)^2\right\} \right]. \quad (3.15)$$

Here $D_2(a)$ is a parabolic cylinder function of the second order.¹² The absolute values of this function do not exceed 1.2, and are exponentially small at large values of the argument; $D_2(0) = -1$. The formula (3.15) assumes in the case of small τ ($\omega_R \tau \ll 1$) the form (3.6) and in the case large τ ($|\Omega| x_0 \tau / \omega_R \gg 1$) the form (3.7) with allowance for the fact that $|z'| \gg 1$. Thus, if the condition $|\Omega| x_0 \tau / \omega_R \gg 1$ can be realized within the framework of the condition (3.14) (and they are, generally speaking, not inconsistent), the expression (3.15) is valid for arbitrary values of τ , including the values in the region $|V|^2 x_0^2 \tau / \omega_R^3 \geq 1$. The first term in the square brackets in the formula (3.15) is connected with the dependence of the oscillation amplitude of the populations of the excited states of the particles on the velocity component v_k [see (3.11)]; the second, with the dependence on v_k of the frequency of these oscillations; and the third, with the "interference" of these dependences. The contributions to the magnitude of the drift velocity that correspond to the first and third terms are much smaller in magnitude than v_0 , in virtue of the smallness of the factor x_0 / ω_R . In absolute value, the second term attains its maximum values, which are approximately equal to $\omega_R^2 / (2e)^{1/2} |\Omega| x_0$, when $\tau \approx 2^{1/2} \omega_R / |\Omega| x_0$. Under these conditions, for $|V| \geq |\Omega|$ and the optimal $|\beta|$ values $\sim 10^{-1}$, the contribution corresponding to this term can, in absolute value, be of the order of $10^{-2} v_0$. Thus, the drift velocity can be comparable to the characteristic thermal velocities, and only insignificantly differ from the maximum drift velocities attainable in the SE regime.

The direction of the drift in the present case is determined not only by the sign of the combination $(v_n - v_m)\Omega$, as under SE conditions and in the cases considered above, but also by the factor $\omega_R \tau$. From the formula (3.15) it follows, in particular, that, for $\omega_R \tau > 1$ and $|\Omega| x_0 \tau / \omega_R \leq 1$, the drift can be reversed by changing the pulse-field intensity $I_1 = c|E|^2 / 8\pi \propto |V|^2$ or the pulse length τ . This characteristic is due to the following circumstances. From (3.11) it follows that, for $|\Omega| x_0 \tau / \omega_R \sim 1$, the difference between the phases of the oscillations of the excited-state populations for particles the components v_k of whose velocities differ by an amount of the order of v_0 can attain values close to π . At the same time, in the case of particles the v_k values for which differ by an amount significantly smaller than v_0 , this phase difference is insignificant. Therefore, the scale of the selectivity of the particle excitation with respect to the velocity components v_k is equal in this case to v_0 . From this and the fact that the Rabi oscillation amplitude depends weakly on v_k when $|\Omega| \gg x_0$ [see (3.11)], it is clear that there exist values of τ (depending on $|V|^2$ and Ω^2) at which the particles with positive or negative v_k values in the region $|v_k| \leq v_0$ are significantly more strongly excited. Evidently, to these two situations correspond the opposite directions of the mean excited-particle velocity

$$u_m \propto \int v p_m(v, \tau) dv,$$

and, consequently, the opposite directions of the drift.

For $|\Omega| x_0 \tau / \omega_R \ll 1$ or $|\Omega| x_0 \tau / \omega_R \gg 1$, the scale of the selectivity, connected with the dependence of ω_R' on v_k ,

of the particle excitation with respect to v_0 turns out to be much greater or much smaller than v_0 . In either case the corresponding selectivity factor plays a lesser role, which is manifested in the form of the realization of the regime (3.6) (when $\omega_R \tau \ll 1$) or the regime (3.7). The origin of the optimal condition $|\Omega| x_0 \tau / \omega_R \sim 1$ is accordingly clear.

The elucidated characteristics of the LID phenomenon are illustrated in Fig. 1, which was obtained through a computer analysis of the formula (3.5) under conditions close to the case $|\Omega| \gg x_0$. It can be seen from the graph that in the case under consideration the velocity u attains its maximum—in magnitude—value, which is equal to $-1.31 \times 10^{-2} v_0$, at $\tau = 2.5 x_0^{-1}$ ($\Omega x_0 \tau / \omega_R = 1.8$). For $\tau > 7.8 x_0^{-1}$ ($\Omega x_0 \tau / \omega_R > 5.6$) the drift velocity u does not depend on τ , and is equal to $-2.5 \times 10^{-3} v_0$.

Let us turn to the analysis of the case of strong fields (i.e., the case in which $|V| \gg x_0$) and arbitrary values of the detuning Ω . Under these conditions the terms quadratic in y in (3.11) may be of the same order of magnitude as, or even greater than, the linear terms. Let us, in view of this, take into account the dependence of the Rabi-oscillation frequency on v_0 , including the quadratic term. Then, neglecting the weak dependence on v_0 of the amplitude of these oscillations, we find from (3.13) the following expression:

$$u = \frac{2|V|^2 \beta v_0 Q \exp[-Q^2/(1+P^2)]}{\omega_R^2 (1+P^2)^{1/2}} \{ \sin(\omega_R \tau) \times (\cos \varphi - P \sin \varphi) + \cos(\omega_R \tau) (\sin \varphi + P \cos \varphi) \}; \quad (3.16)$$

$$Q = q \omega_R \tau, \quad P = p \omega_R \tau, \quad \varphi = \frac{1}{2} \operatorname{arctg} P - \frac{Q^2 P}{1+P^2}.$$

The factors depending on the quantities P and Q in (3.16) are, on account of the conditions $q \ll 1$ and $p \ll 1$, slower functions of the argument $\omega_R \tau$ than $\sin \omega_R \tau$ and $\cos \omega_R \tau$. Thus, the expression (3.16) describes an oscillatory type of rapid variation, slowly modulated in amplitude, of the drift velocity as the factor $\omega_R \tau$ is varied. It also demonstrates the possibility of reversing the drift at a fixed value of the factor $(\nu_n - \nu_m) \Omega$.

The maximum absolute values of u are attained in the region $|\Omega| \sim 1$, $P \leq 1$, $|V| \geq |\Omega|$ (for this to happen it is necessary that $|\Omega| \geq x_0$). Under these conditions, and for the optimal $|\beta|$ values $\sim 10^{-1}$, $|u|$ can be of the order of $10^{-2} v_0$. Notice that for $|\Omega| \gg x_0$ the formula (3.16) goes over into (3.15) if only the second term in the square brackets in the latter formula is taken into account. If $P \gg |Q|$ ($|\Omega| \ll x_0$), the drift velocity is low. This is due to the predominance under the present conditions of the quadratic "scatter" of the population-oscillation phases for particles with different v_0 values, which leads to the rapid reduction of the excitation-selectivity scale. Analysis shows that the formula (3.16) is applicable in the region where the u -variation amplitude, given by the formula, significantly exceeds the modulus of the τ -independent contribution to the drift velocity in (3.13).

The case $|\Omega| \sim |V| \sim x_0$, which has not been analyzed, lends itself at arbitrary τ to only a numerical treatment. Figure 2 shows a plot, obtained through a computer analysis of the formula (3.5), of u vs τ under conditions when $\Omega = 2|V| = x_0/2$ and $\beta = -10^{-1}$. The plot is markedly asymmetric about the $u = 0$ axis. This is due to the fact that the conditions under consideration are the optimal conditions for the manifestation of the excitation-selectivity factor stemming from the dependence of the Rabi-oscillation amplitude on v_0 . The tendency toward the assumption at high τ values of τ -independent values is clearly visible.

4. STOCHASTIC EXCITATION

In the preceding section the analysis was based on the assumption that the characteristics of the radiation pulses are completely defined. In this section we consider the occurrence of the LID of gases under the action of pulses whose characteristics undergo random fluctuations. Here we shall assume that the field autocorrelation time τ_c is significantly shorter than the pulse duration τ .

The system of equations (2.3) can be reduced to an integro-differential equation for the difference between the populations of the states m and n (see, for example, Ref. 6):

$$\langle n(v, t) \rangle = -4 \operatorname{Re} \int_0^t \langle G^*(t-t') G(t) n(v, t-t') \rangle e^{-i\alpha t'} dt'. \quad (4.1)$$

Here $G(t) = -\epsilon(0, t) d_{mn} / 2\hbar$, where $\epsilon(0, t)$ is a random function of the time, and the symbol $\langle \dots \rangle$ denotes averaging over t greater than τ_c . Let us, following Burshstein,¹³ "uncouple" the variables:

$$\langle G^*(t-t') G(t) n(v, t-t') \rangle \rightarrow \langle G^*(t-t') G(t) \rangle \langle n(v, t-t') \rangle.$$

The "uncoupling" procedure is justified if no transitions occur between the states m and n of the particles during the period τ_c , which is assumed. In this approximation we can, by allowing for the fact that $\tau_c \ll \tau$, reduce Eq. (4.1) for $t \leq \tau$ to the form

$$\langle n(v, t) \rangle = -4|V|^2 \langle n(v, t) \rangle \operatorname{Re} \int_0^t \Phi(t') e^{-i\alpha t'} dt'. \quad (4.2)$$

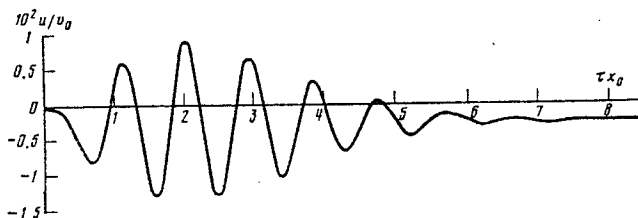


FIG. 1. Dependence of the drift-velocity component u along the radiation wave vector on the pulse duration τ under conditions when $\beta = -10^{-1}$ and $2|V| = \Omega = 5x_0$.

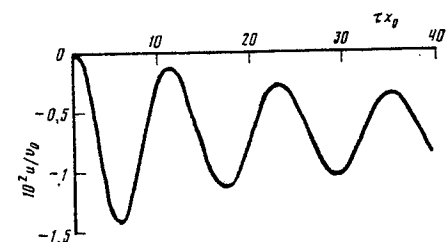


FIG. 2. Dependence of the drift-velocity component u along the radiation-wave vector on the pulse duration τ under conditions when $\beta = -10^{-1}$ and $2|V| = \Omega = x_0/2$.

Here
 $V^2 = \langle | \varepsilon(0, t) d_{mn} |^2 \rangle / 4\hbar^2$, $\Phi(t') = \langle \varepsilon^*(0, t-t') \varepsilon(0, t) \rangle / \langle | \varepsilon(0, t) |^2 \rangle$
 is the field autocorrelation function, which is connected
 with the radiation line shape $g(\omega' - \omega)$ by the relation

$$g(\omega' - \omega) = \frac{1}{\pi} \operatorname{Re} \int_0^\infty \Phi(t') e^{-i(\omega' - \omega)t'} dt', \quad \int_{-\infty}^\infty g(\omega' - \omega) d\omega' = 1.$$

From here we finally find that

$$\langle n(v, t) \rangle' = -A \langle n(v, t) \rangle, \quad A = 4\pi |V|^2 g(\Omega'). \quad (4.3)$$

The quantity A has the meaning of a transition probability per unit time. Thus, the requirement, formulated in Sec. 2, that the field-induced processes predominate over the relaxation processes is met in the present case when

$$A \gg \nu_q, \nu_h, \Gamma \text{ for } |x| \leq x_0.$$

The solution to Eq. (4.3) has the form

$$\langle n(v, t) \rangle = n(v, 0) e^{-At}.$$

With allowance for the initial conditions (2.4), we find the following expressions for $\rho_m(v, \tau)$ and $\rho_n(v, \tau)$:

$$\rho_m(v, \tau) = W(v) (1 - e^{-A\tau})/2, \quad \rho_n(v, \tau) = W(v) - \rho_m(v, \tau). \quad (4.4)$$

It is easy to see from the expressions (4.4) that the particles that (with allowance for the Doppler effect) are at resonance with the most intense spectral component of the radiation are the ones that are mostly excited. This indicates excitation selectivity with respect to the velocity components v_h in the present case. A similar situation obtains in the case of stationary particle excitation by a nonmonochromatic field.⁶ This circumstance makes the LID effect occurring under conditions of stochastic PPE and the effect in the SE regime essentially similar (see below).

Combining (4.4) and (2.10), we find for the drift velocity component along the wave vector the following expression:

$$u = -\frac{\beta v_0}{2\pi^{1/2} x_0} \int_{-\infty}^\infty \exp \left[-\left(\frac{x}{x_0} \right)^2 - A\tau \right] x dx. \quad (4.5)$$

For spectra that are symmetric about the carrier frequency ω , the relation $g(a) = g(-a)$ is valid. Taking this into account, and using (4.5), we can easily show that in this case u changes sign when the sign of Ω is changed. Thus, it is clear that, as in the SE regime, the direction of the drift is determined by the sign of the combination $\Omega(\nu_n - \nu_m)$.

If the radiation spectrum $g(\omega' - \omega)$ has a smooth envelope, and its halfwidth δ is significantly greater than the halfwidth x_0 of the Doppler contour, then it follows from (4.5) that up to terms of the order of $(x_0/\delta)^2$

$$u = -\beta |V|^2 (dg(\Omega)/d\Omega) \tau x_0 \exp \{-4\pi |V|^2 g(\Omega) \tau\} v_0. \quad (4.6)$$

In deriving the formula (4.6) we assumed that

$$4\pi |V|^2 x_0 \tau dg(\Omega)/d\Omega \ll 1.$$

The derivative $dg(\Omega)/d\Omega$ attains its maximum absolute values at $|\Omega| \sim \delta$. In this case the estimates $g(\Omega) \sim \delta^{-1}$ and $|dg(\Omega)/d\Omega| \sim \delta^{-2}$ are valid, and, consequently,

$$|u| \sim \pi |\beta| \frac{|V|^2 x_0 \tau}{\delta^2} \exp \left\{ -\frac{4\pi |V|^2 \tau}{\delta} \right\} v_0. \quad (4.7)$$

From (4.7) and the fact that $|\Omega| \sim \delta$, it is clear that, from the point of view of the maximization of $|u|$, the optimum conditions are

$$4\pi |V|^2 \tau / \delta \sim 1, \quad |\Omega| \sim \delta \sim x_0. \quad (4.8)$$

The first condition in (4.8) indicates that only the particles that are at resonance with the most intense spectral components lying in the interval of the order of the width of the radiation spectrum are efficiently excited. It is similar in meaning to the condition for appreciable saturation of the transition in the SE case when the field-induced broadening, which lowers the excitation selectivity, is not too large. The second condition in (4.8), which is entirely analogous to the condition that obtains in the SE regime, together with the first conditions, implies that the process of strong selective excitation with respect to v_h involves a significant fraction of the particles.

In the case of a "rectangular" spectrum of arbitrary width 2δ , i.e., for

$$g(\omega' - \omega) = \begin{cases} 1/2\delta & \text{for } -\delta \leq \omega' - \omega \leq \delta \\ 0 & \text{for other } \omega', \end{cases}$$

we find from (4.5) that

$$u = \frac{\beta v_0}{4\pi^{1/2}} \left(1 - \exp \left\{ -\frac{2\pi |V|^2 \tau}{\delta} \right\} \right) \left[\exp \left\{ -\left(\frac{\Omega - \delta}{x_0} \right)^2 \right\} - \exp \left\{ -\left(\frac{\Omega + \delta}{x_0} \right)^2 \right\} \right]; \quad (4.9)$$

$|u|$ attains its maximum value $|\beta| v_0 / 4\pi^{1/2}$ under the conditions $|V|^2 \tau / \delta \rightarrow \infty$, $|\Omega| = \delta \rightarrow \infty$. But when $2\pi |V|^2 \tau / \delta \sim 1$ and $|\Omega| \sim \delta \sim x_0$ the modulus of the drift velocity has the same order of magnitude.

5. APPLICATION TO MOLECULAR AND ATOMIC SYSTEMS

For many reasons⁴ it is important to obtain streams of drifting molecules in an infrared-radiation field. In this case the drift is accompanied by vibrational and rotational relaxation processes. These processes cannot, generally speaking, be described in the two-level approximation. But if, besides (2.2) (ν_q is the quenching rate of the excited vibrational state m), the conditions

$$\tau \ll \tau_{n,j} (j=m, n) \ll \nu^{-1}$$

($\tau_{n,j}$ is the characteristic time of the rotational relaxation in the j -th vibrational state) are satisfied, we need not take the rotational relaxation into account explicitly in the kinetic equations. All the expressions found above for the drift velocity then remain valid to within introduction in the right-hand sides of the factor $k_{n0} \sim 10^{-2} - 10^{-1}$, which is the Boltzmann weight of the resonant rotational sublevel of the lower vibrational state n . This is due to the fact that the field of each pulse interacts not with all the molecules, but with only those whose rotational state satisfies the resonance conditions.

For allowed electronic transitions in atoms the decay of the excited state is due largely to spontaneous emission. The decay rate is usually a quantity of the order of 10 MHz. In this case pulse repetition rates $\nu \sim 1$ MHz are necessary for the attainment of high drift velocities. If by chance the LID process occurs with the

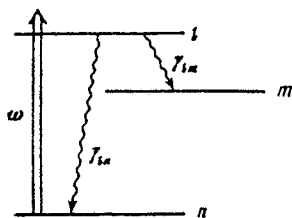


FIG. 3. The level m is metastable; γ_{ln} and γ_{lm} are respectively the spontaneous $l \rightarrow n$ and $l \rightarrow m$ transition rates. The pulsed periodic radiation is at quasiresonance with the $l \rightarrow n$ transition. The level m is populated selectively with respect to the velocities as a result of spontaneous $l \rightarrow m$ transitions.

participation of a metastable atomic state (see Fig. 3), then lower ν values are sufficient for its strong manifestation. In this case, if instead of the first of the conditions (2.2) we require that

$$\tau < \gamma_{lm}^{-1}, \gamma_{ln}^{-1}, \Gamma_{ln}^{-1} < \nu_q^{-1}, \nu_j^{-1} < \nu^{-1}$$

(Γ_{ln} is the homogeneous width of the $l \rightarrow n$ transition and ν_q is the decay rate of the state m), then all the formulas obtained above for u remain valid to within the introduction in the right members of the factor $\gamma_{lm}/(\gamma_{lm} + \gamma_{ln}) \leq 1$ and the replacement of the subscript m by l in the definitions of V and Ω .

Let us estimate the pulse-field intensities I_l and the mean pulsed-periodic-pump intensities

$$I = \tau \nu I_l = \tau \nu \hbar^2 |V|^2 / 2\pi |d_{ln}|^2, \quad (5.1)$$

necessary for the realization of the effects under discussion. As shown above, for the drift velocities obtained to be high, the pulse repetition rate ν should (with allowance made for the assumption that $\nu \ll \nu_q, \nu_j$) be as close as possible to the relaxation rates ν_q and ν_j . The frequencies ν_j are bounded from below by the requirement that the particle mean free path l_0 be short compared to the cell dimensions. Values of $l_0 \sim 1$ cm correspond to $\nu_j \sim 10^4$ Hz. The order of magnitude of the decay rate ν_q can be the same for molecules⁴ and atoms (metastable states).¹⁴ Thus, the optimal values of $|\beta|$ ($\sim 10^{-1}$) can be attained with $\nu \sim 1$ kHz. Let us use these values of ν to make estimates. Let us note that, as follows from (5.1), the possibility of the use of moderate pulse-repetition rates allows us to reduce the mean radiation intensity. At the same time this possibility is in itself an attractive one.

It can be seen from Fig. 2 that in the case of dynamic PPE with $2|V| = \Omega = x_0/2$ the maximum values of the drift velocity are realized if $\tau \approx 5x_0^{-1}$. Under these conditions we obtain from (5.1) the estimates $I_l \sim 1$ W/cm² and $I \sim 10^{-3}$ W/cm² for molecular gases and $I_l \sim 10^2$ W/cm² and $I \sim 10^{-3}$ W/cm² for atomic gases. These estimates were obtained with the use of the following char-

acteristic parameter values: $x_0 \sim 10$ MHz, $|d_{ln}| \sim 0.1$ D for molecules and $x_0 \sim 1$ GHz, $|d_{ln}| \sim 1$ D for atoms. In the case of stochastic excitation under the optimal conditions $|\Omega| \sim \delta \sim x_0$ the mean intensities I should be of the same order of magnitude as in the preceding case, but the pulse-field intensities I_l can be somewhat lower. Under the conditions corresponding to Fig. 1, similar estimates yield $I_l \sim 10^2$ W/cm², $I \sim 10^{-1}$ W/cm² for molecules and $I_l \sim 10^4$ W/cm², $I \sim 10^{-1}$ W/cm² for atoms. Thus, the requisite radiation intensities are entirely attainable with the use of existing infrared and optical lasers.

In conclusion the authors express their gratitude to G.V. Mironov for his help in the numerical analysis.

¹⁾ The quasiresonant, transfer of excitation by the absorbing particles to the buffer particles can also give rise to the LID phenomenon.⁵

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