Resonant coherent transient process in a gas subjected to a standing wave field

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A report is given of a theoretical and experimental investigation of the decay of polarization in the presence of a standing electromagnetic wave field interacting with a Doppler-broadened transition. It is shown that in the direction opposite to the propagation of an exciting pulse the polarization decays, contrary to the usual case, not at the frequency of the exciting field but at the transition frequency. This makes it possible to use a transient process to study relaxation and to apply this process in ultrahigh-resolution spectroscopy without the Doppler broadening. A resonant coherent transient process was used to determine the relaxation constants of the process of decay of the dipole moment in SF_6 collisions ($\lambda = 10.6\ \mu$). This method was employed to resolve reliably the structure of the $Q(38)$ transition ($\lambda = 10.6\ \mu$) giving rise to $F_1^0 + E_0 + F_2^0$ lines of the $\rightarrow\nu_3$ band of SF_6 inside the Doppler absorption line.

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Earlier investigations[1,2] of resonant transient processes have revealed a new phenomenon in the interaction of optical fields with a gas. After the end of the interaction between a traveling wave pulse in a medium subjected also to a standing wave field, a decaying polarization wave appeared in the direction opposite to the propagation of the pulse. Beats between the decaying polarization wave and one of the components of the standing wave were observed at a frequency equal to the detuning of the field frequency from the center of a Doppler-broadened line. This has become known as the resonant coherent transient process (RCTP). The properties of the RCTP have some features in common with the optical free induction decay,[3] coherent emission in separated optical fields,[4] and wavefront reversal.[5]

The phenomenon in question is undoubtedly of interest in spectroscopy. The two most important tasks in spectroscopy are as follows. The first is to investigate the relaxation processes, and the second is to study the structure of levels and transitions. We shall show that the method utilizing the RCTP is suitable for investigating relaxation processes and for use in ultrahigh-resolution spectroscopy. In relaxation studies the potentialities of this method are analogous to those of the methods based on the use of coherent transient processes.[6]

The use of the RCTP in ultrahigh-resolution spectroscopy is based on the decaying polarization at the center of a Doppler-broadened line. This makes it possible to determine the position of the line center and to resolve Doppler-broadened lines. As in other nonlinear spectroscopy methods, the resolution is limited to the homogeneous width of a line.

We investigated transient processes in a gaseous medium subjected to radiation pulses and discovered a large number of phenomena due to different physical processes. We cannot go into details of all of them, so that in Sec. 1 we shall describe briefly the main phenomena observed experimentally. A theoretical treatment of the appearance of the RCTP is given in Sec. 2. Spectroscopic applications of the RCTP are described in Sec. 3 and, finally, Sec. 4 is devoted to the relationship between the RCTP and other coherent phenomena.

1. TRANSIENT PROCESSES OBSERVED EXPERIMENTALLY

We observed earlier[7,8] coherent emission in separated fields in a standing wave, to which attention was drawn in Ref. 9. We carried out detailed theoretical investigations of this effect.[10,11] The results reported below represent continuation of our studies of resonant coherent emission in time-separated standing wave fields.[7,8] We used the experimental arrangement shown in Fig. 1. Radiation from a CO_2 laser was split by a mirror 1 into two beams of equal intensity. Both beams passed through electro-optic modulators M_1 and M_2 and were directed by mirrors 4–6 so as to arrive from opposite sides in an absorbing cell containing SF_6. The modulators were controlled by electronic apparatus which provided opportunities for investigating all possible types of laser interaction with the absorbing medium under pulsed and cw conditions. The radiation pulses could be applied in series of up to three pulses of different duration and delay time, either simultaneously from both modulators or from each separately. The pulse duration τ and the delay time T were continuously variable from 0.3 μsec to a few microseconds. The apparatus made it possible to study the interaction of various combinations of traveling and standing wave pulses with the absorbing medium. We shall consider below the observed effects, beginning from the simplest phenomena associated with relaxation of the population.

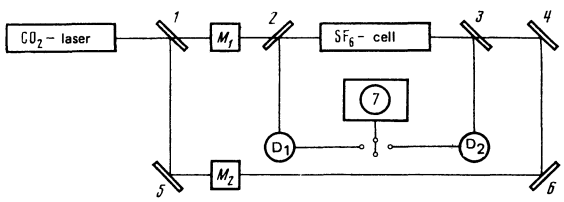

FIG. 1. Experimental arrangement: M_1 and M_2 are electro-optic light modulators; D_1 and D_2 are photodetectors; 7 is an oscilloscope; 1–6 are mirrors.

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When an absorbing medium is acted upon by two traveling-wave pulses, the effects observed are associated with saturation of the medium. If the pulses travel in the same direction, the transmission of the medium for the second pulse increases. This transmission is a function of the delay time between the pulses and is governed by the population relaxation rate. In the case of opposite pulses a similar effect is observed at the line center. This makes it possible to study the dynamics of relaxation of the Lamb dip.

In addition to the appearance of coherent radiation at moments $2T$, $3T$, etc. after the first pulse, when pulses were applied in the presence of a weak standing wave, we observed various transient processes in the form of periodically damped signals. These transient processes occurred at the leading or trailing edges of long pulses (i.e., of pulses of duration greater than the relaxation time of the dipole moment of the medium) and after the action of a short pulse.

Periodically damped signals were observed in the direction of propagation of the traveling wave pulse and in the opposite direction. When several short pulses were used, the transient processes did not appear immediately after each of the pulses but at moments corresponding to the photon echo signals. All these transient processes were coherent, since they were observed in the form of beats between the nonlinear polarization of the medium and the laser radiation. Moreover, they were resonant, because their amplitude and frequency were a function of the detuning of the field from the center of the absorption line. More complex transient processes representing combinations of those mentioned above were also observed.

The most remarkable among all the transient coherent processes was the RCTP,[1,2] discussed in detail below. The RCTP was similar to the free induction decay. A significant distinguishing feature of the RCTP was that we were not dealing with free decay of the induction, but with decay in the presence of a standing wave field. This altered the spectral composition of the radiation emitted by damped dipoles and the spatial characteristics of the radiation. Brewer and Shoemaker[12] put forward the Stark-cell method. The frequency of these damped dipoles, relative to the frequency of the probe radiation, was varied by switching on or off the electric field. The majority of our experiments were carried out by this method. Rapid frequency switches, which made it possible to investigate the relaxation processes in the optical range, were constructed relatively recently.[13] In the cases described the additional field could be regarded as the probe necessary solely for the detection of the decaying polarization. The nonlinear interaction of this wave with the damped dipoles was ignored. Therefore, the phenomenon itself represented free induction decay. The new class of phenomena could be attributed to the study of damped dipoles in the presence of other fields.

2. THEORETICAL ANALYSIS OF TRANSIENT PROCESSES IN A STANDING WAVE FIELD

Figure 2 shows a simplified scheme for observing the RCTP. A cell with an absorbing gas was illuminated with a weak standing wave of frequency ω, which differed from the frequency ω_{21} by the amount Ω. In our experiments the wave traveling to the right was modulated in amplitude. In the opposite direction we observed damped oscillations of the absorption traveling in the direction opposite to the incident pulse and the frequency of these oscillations corresponded to the detuning of the field frequency from the center of the transition line. The decay corresponded to Γ, i.e., to half the homogeneous line width 2Γ. An oscillogram of the observed RCTP signal is shown in Fig. 3.

We shall now consider theoretically the transient processes in a standing wave field. Let us assume that a gas of two-level atoms (molecules) interacts with the fields of two opposite waves:

$$\begin{aligned} \mathbf{E}(\mathbf{r}, t) &= \mathbf{e}E(\mathbf{r}, t) + \text{c.c.}, \\ E(\mathbf{r}, t) &= E(t)e^{ikz-i\omega t} + \tilde{E}(t)e^{-ikz-i\omega t}, \end{aligned} \tag{1}$$

whose amplitudes $E(t)$ and $\tilde{E}(t)$ vary slowly compared with $\cos\omega t$. For simplicity, we shall assume that the waves are polarized in the same direction $\mathbf{e}$. Our task is to determine the polarization of the gas

$$\begin{aligned} \mathbf{P}(\mathbf{r}, t) &= \mathbf{e}P(\mathbf{r}, t) + \text{c.c.}, \\ P(\mathbf{r}, t) &= d_{12}\int d^3v\, \rho_{21}(\mathbf{r}, \mathbf{v}, t), \end{aligned} \tag{2}$$

where d_{12} is the dipole moment of a $1 \rightarrow 2$ transition and $\rho_{21}(\mathbf{r}, \mathbf{v}, t)$ is the nondiagonal element of the density matrix considered in the Wigner representation.

The equations for the density matrix of an atom tra-

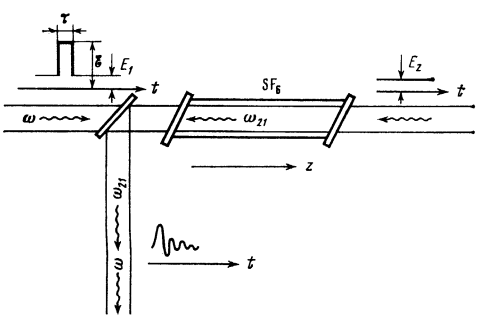

FIG. 2. Schematic representation of the observation of the resonant coherent transient process.

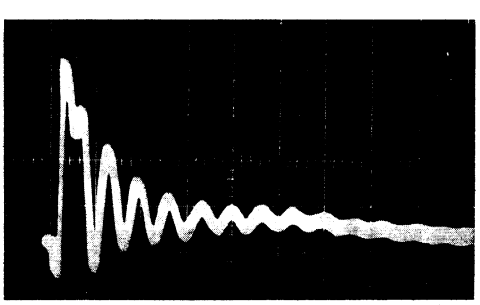

FIG. 3. Oscillogram of the RCTP signal. Each horizontal division represents 1 μsec.

veling at a velocity v are

$$\left(\frac{\partial}{\partial t}+\gamma_1\right)\rho_{11}=-\frac{i}{\hbar}[E(\mathbf{r}',t)d_{21}\rho_{12}-\rho_{21}d_{12}E^*(\mathbf{r}',t)]+\gamma_1 n_1^{(0)},$$
$$\left(\frac{\partial}{\partial t}+\gamma_2\right)\rho_{22}=\frac{i}{\hbar}[E(\mathbf{r}',t)d_{21}\rho_{12}-\rho_{21}d_{12}E^*(\mathbf{r}',t)]+\gamma_2 n_2^{(0)}, \qquad (3)$$
$$\left(\frac{\partial}{\partial t}+i\omega_{21}+\Gamma\right)\rho_{21}=\frac{i}{\hbar}E(\mathbf{r}',t)(\rho_{11}-\rho_{22}), \quad \rho_{12}=\rho_{21}^*,$$

where γ_2 and γ_1 are the reciprocal lifetimes of the upper and lower levels; $n_2^{(0)}$ and $n_1^{(0)}$ are the populations of the upper and lower levels in the absence of an electromagnetic field; $\mathbf{r}'=(x, y, z+vt)$. The equations (3) are written down in reference system moving, relative to the laboratory system, at a velocity v along the z axis, where v is the projection of the velocity of the investigated atom along the z axis in the laboratory reference system.

We shall solve the system (3) by the methods of successive approximations assuming that the field $E(\mathbf{r}, t)$ is weak and we shall confine ourselves to the third order in E. This is the lowest order in which the interaction of the opposite waves of interest to us is observed. The saturation effects will be considered in Sec. 3. In the Doppler limit $kv_0 \gg \Gamma$, where v_0 is the thermal velocity, the exponential function in the Maxwellian distribution can be replaced with unity in the course of averaging with respect to v, since we shall be interested in the frequency range $\Omega \ll kv_0$. If we find ρ_{21} from Eq. (3), return to the laboratory reference system, and bear in mind that

$$\int_{-\infty}^{\infty} dv \exp[ikv(t_1-t_2-t_3+t)]=\frac{2\pi}{k}\delta(t_1-t_2-t_3+t),$$

we obtain after averaging with respect to the velocities

$$P(\mathbf{r},t)=-\exp(ikz-i\omega t)i\alpha_0\int_{-\infty}^{t}dt_3\int_{-\infty}^{t_3}dt_2 E(t_3)E^*(t_2)E(t_2+t_3-t)$$
$$\times\exp[2(\Gamma-i\Omega)(t_3-t)]\{\exp[\gamma_1(t_2-t_3)]+\exp[\gamma_2(t_2-t_3)]\}; \qquad (4)$$

$$\bar{P}(\mathbf{r},t)=-\exp(-ikz-i\omega t)i\alpha_0\int_{-\infty}^{t}dt_3\int_{-\infty}^{t_3}dt_2 E(t_3)E^*(t_2)E(t_2+t_3-t)$$
$$\times\exp[2(\Gamma-i\Omega)(t_3-t)]\{\exp[\gamma_1(t_2-t_3)]+\exp[\gamma_2(t_2-t_3)]\}, \qquad (5)$$

where $\alpha_0=2\pi^{1/2}\Delta n^{(0)}|d_{21}|^4/kv_0\hbar^3$, and $\Delta n^{(0)}=n_1^{(0)}-n_2^{(0)}$.

Equations (4) and (5) do not include the terms describing the interaction of the waves E and $\bar{E}$ with one another, because these interactions are of no interest to us.

In the case of the transient processes under consideration here, it is necessary to assume that $\bar{E}(t)=E_2$ = const and $E(t)=E_1+\mathscr{E}(t)$. To be specific, we shall also assume that $\mathscr{E}(t)$ differs from zero only in the time interval $t\in[\tau_1, \tau_2]$. Separating the terms describing the interaction of the pulse $\mathscr{E}(t)$ with two waves of constant amplitude, we can reduce Eqs. (4) and (5) at $t > \tau_2$ to the form:

$$P(\mathbf{r},t)=-i\alpha_0\exp(ikz-i\omega t)|E_2|^2(\gamma_1^{-1}+\gamma_2^{-1})D\exp(-2(\Gamma-i\Omega)t); \qquad (6)$$
$$\bar{P}(\mathbf{r},t)=-i\alpha_0\exp(-ikz-i\omega t)E_2\{A_1(\gamma_1)+A_2(\gamma_1)+A_1(\gamma_2)+A_2(\gamma_2)\}; \qquad (7)$$
$$A_1(\gamma)=\tfrac{1}{2}E_1^*\{[B_1(\gamma)-C_1(\gamma)\exp[(\Gamma-\gamma-i\Omega)\tau_2](\Gamma-\gamma-i\Omega)^{-1}]$$
$$\times\exp[-(\Gamma-i\Omega)t]+C_1(\gamma)(\Gamma-\gamma-i\Omega)^{-1}e^{-\gamma t}\},$$
$$A_2(\gamma)=E_1\{[B_2(\gamma)-\tfrac{1}{2}C_2(\gamma)\exp[2(\Gamma-\gamma/2-i\Omega)\tau_2](\Gamma-\gamma/2-i\Omega)^{-1}]$$
$$\times\exp[-2(\Gamma-i\Omega)t]+\tfrac{1}{2}C_2(\gamma)(\Gamma-\gamma/2-i\Omega)^{-1}e^{-\gamma t}\}.$$

Here, the time dependence is explicit and the pulse shape $\mathscr{E}(t)$ influences only the coefficients D, B_i, and C_i which are independent of time:

$$D=\int_{\tau_1}^{\tau_2}dx\,\mathscr{E}(x)\exp[2(\Gamma-i\Omega)x]; \qquad (8a)$$

$$B_1(\gamma)=\int_{\tau_1}^{\tau_2}dy\int_{\tau_1}^{y}dx\,\mathscr{E}(x)e^{\gamma x}\exp[(\Gamma-\gamma-i\Omega)y]; \qquad (8b)$$

$$B_2(\gamma)=\int_{\tau_1}^{\tau_2}dy\int_{\tau_1}^{y}dx\,\mathscr{E}^*(x)e^{\gamma x}\exp[2(\Gamma-\gamma/2-i\Omega)y]; \qquad (8c)$$

$$C_1(\gamma)=\int_{\tau_1}^{\tau_2}dx\,\mathscr{E}(x)e^{\gamma x}; \quad C_2(\gamma)=C_1^*(\gamma). \qquad (8d)$$

The transient processes during a pulse ($\tau_1 \leqslant t \leqslant \tau_2$) depend largely on the pulse shape and if necessary can be determined from Eqs. (4) and (5). The time interval $[\tau_2, \tau_2+\tau]$, where $\tau=\tau_2-\tau_1$ is the pulse duration, is somewhat special. In this time interval we can expect, in addition to the transient processes described by Eqs. (6) and (7), also a photon echo signal due to different parts of a pulse ($\sim\mathscr{E}(t)|\mathscr{E}(t)|^2e^{ikz}$) and a similar signal traveling in the opposite direction ($\sim E_2|\mathscr{E}(t)|^2e^{-ikz}$). The latter signal can be found from Eq. (5). In experiments it is usually convenient to use a short pulse such that $\tau \ll \Gamma^{-1}$. Then, a large proportion of the transient process takes place during the time $t > \tau_2+\tau$.

We shall now discuss and interpret the results. Equation (6) describes a transient process at a frequency $\omega-2\Omega=2\omega_{21}-\omega$, which decays at the rate 2Γ. This transient process is due to atoms which are in resonance with the wave E_2. The width of the distribution of the velocity of this group of atoms (i.e., the width of the Bennett dip) is $\Delta v=\Gamma(1+\varkappa_2)^{1/2}/k$, where $\varkappa_2$ is the saturation parameter of the field E_2. When the field $E(t)$ ceases to act, the polarization decays at a rate $\Gamma+\Gamma(1+\varkappa_2)^{1/2}$ [the first term represents the rate of decay of a single atom and the second term is associated with the Doppler phase advance $k\Delta v$ (Δv is related to the width of the Bennett dip)]. In a weak field ($\varkappa_2 \ll 1$) the polarization decay rate is twice the rate of decay of the dipole moment of an atom. The average polarization frequency coincides with the center of the Bennett dip and, consequently, it is equal to $\omega_{21}-\Omega=2\omega_{21}-\omega$. This transient process would be called the decay of polarization in the field of an opposite wave. This decay is analogous to optical free induction decay.[3] In both cases the macroscopic polarization of the medium is governed by the atoms participating in the formation of a Bennett dip, which is manifested by a characteristic field broadening. However, the following important difference should be stressed: in the case of optical free induction decay the polarization decays at the frequency of the field, whereas in the field of the opposite wave the polarization decays at the frequency $2\omega_{21}-\omega$, which depends on the transition frequency. This makes it possible to use the transient process under discussion to study the structure of a transition which is masked by the Doppler broadening.

In the direction opposite to that of the propagation of the pulse, the transient process involves oscillations at the field frequency ($\sim C_1, C_2e^{-i\omega t}e^{-\gamma t}$), at the frequency

$2\omega_{21}-\omega$ (the term in A_2 analogous to the decay of polarization in the field of the opposite wave), and also at the frequency $\omega-\Omega=\omega_{21}$. We shall consider the mechanism of polarization at the transition frequency ω_{21}. The dipole moment of an atom induced by a short pulse $\mathscr{E}$ continues to oscillate its natural frequency:

$$d^{(1)}(t)\sim\mathscr{E}\tau\exp(-(\Gamma+i\omega_{21})t)e^{ikz} \tag{9}$$

(in a reference system linked with the atom). The dipole-moment wave (9) interacts with the spatial structure representing the energy density in the field of opposite waves: $\sim(E_2E_1^*e^{-2ikz}+\text{c.c.})$. Since the frequencies of the waves E_1 and E_2 are equal, this structure is at rest in the laboratory reference system. The interaction reverses the wave vector of the dipole moment wave. The nonlinear dipole moment of an atom in the laboratory system is

$$d^{(n1)}(v,t)\sim E_2E_1^*\mathscr{E}\tau e^{-ikz}\times\frac{\exp(-(\Gamma+i\omega_{21}+ikv)t)-\exp(-(\Gamma+i\omega_{21}-ikv)t)}{2ikv(\Gamma-\gamma-i\Omega+ikv)} \tag{10}$$

and it emits radiation in the $-z$ direction at two frequencies: $\omega_{21}-kv$ and $\omega_{21}+kv$. The former frequency is governed by the usual Doppler shift, whereas the latter appears because of modulation, at a frequency $2kv$, when an atom moves across the spatial structure formed by opposite waves.

The polarization of the gas can be found by averaging Eq. (10) over the velocities with the distribution $F(v)$:

$$P^{(n1)}(\mathbf{r},t)\sim E_2E_1^*\mathscr{E}\tau\exp(-ikz)\exp(-(\Gamma+i\omega_{21})t)I(t), \tag{11}$$

$$I(t)=\int_{-\infty}^{\infty}\frac{\sin kvt}{kv}\frac{F(v)\,dv}{\Gamma-\gamma-i\Omega+ikv}+f(t). \tag{12}$$

The emission of the gas is governed by the interference between the radiations of the atoms characterized by different projections of the velocity along the z axis. It is clear from Eqs. (11) and (12) that at a moment t the contribution is made to the emitted radiation by the atoms with $v\leqslant(kt)^{-1}$. At any arbitrary moment in time only those atoms are in phase which move across the light beam ($v=0$). All the other atoms particpate in the emission of radiation only to the extent that they do not manage to change their phase up to the selected moment. This is why there is no influence of thermal motion: the polarization decays in the same way as the dipole moment of an atom at rest. The radiation emitted by the gas at other frequencies [described by the function $f(t)$ in Eq. (12)] is determined by atoms with nonzero values of v corresponding to peaks and troughs in the distribution function $F(v)(\Gamma-\gamma-i\Omega+ikv)^{-1}$.

The appearance of a decaying polarization wave in the direction opposite to that of the exciting pulse is not associated with the population-difference spatial modulation with which the wavefront reversal is usually associated.[5] The difference between the populations at the lower and upper levels responsible for the RCTP is spatially homogeneous:

$$\Delta n\sim E_1^*\mathscr{E}\tau\frac{\exp(-(\Gamma-i\Omega+ikv)t)-1}{\Gamma-\gamma-i\Omega+ikv}+\text{c.c.} \tag{13}$$

The above analysis makes it possible to identify two basically new features of the observed effect: a) the RCTP decays at a rate Γ and this rate is independent of the applied fields; this makes the RCTP very attractive for investigating relaxation processes and collisions; b) a macroscopic polarization appears at the transition frequency ω_{21}. This makes it possible to use the RCTP in ultrahigh-resolution spectroscopy. The resolution of the method is governed by the homogeneous line width 2Γ.

The RCTP is observed experimentally by recording beats of the decaying polarization wave as the field E_2 is varied. Clearly, the frequency of these beats will be equal to the detuning Ω of the field frequency from the transition frequency. At the same time there are oscillations of the absorbed power at the frequency 2Ω, which correspond to the polarization at the frequency $2\omega_{21}-\omega$. Therefore, a very important question is the relative magnitude of the signals at these two frequencies. The ratio of the signal amplitudes of frequencies 2Ω and Ω can be calculated for a rectangular pulse on the assumption that $\gamma_1=\gamma_2=\gamma$:

$$W_2/W_1=\tfrac{1}{2}\exp(-\Gamma(t-\tau_2))\{(1-2e^{-2\Gamma\tau}\cos 2\Omega\tau+e^{-4\Gamma\tau})[(\Gamma-\gamma)^2+\Omega^2]\}^{1/2}\times\{(1-2e^{-\Gamma\tau}\cos\Omega\tau+e^{-2\Gamma\tau})[(\Gamma-\gamma/2)^2+\Omega^2)\}^{-1/2} \tag{14}$$

[$W=\mathbf{P}(\mathbf{r},t)\mathbf{E}(\mathbf{r},t)$ is the absorbed power].

If $\gamma,\ \Gamma\ll\tau^{-1},\ \Omega$, Eq. (14) simplifies greatly to

$$W_2/W_1=e^{-\Gamma(t-\tau_2)}\cos(\Omega\tau/2),$$

i.e., the expression (14) corresponding to $\Omega\tau=\pi(2k+1)$ ($k=0,\pm1,\pm2,\ldots$) is close to a minimum. Because of selection $\Omega\tau\approx\pi$, the ratio W_2/W_1 in our experiments was 0.1–0.2 already at the beginning of the transient process, so that the RCTP could be observed practically without admixture of the 2Ω signal.

3. SPECTROSCOPIC APPLICATIONS

a. Determination of the relaxation constants of SF_6–SF_6 collisions

Relaxation of the dipole moment in SF_6–SF_6 collisions was investigated during decay of the RCTP at various gas pressures in an absorption cell. The observations were made using the $P(33)$ transition of the A_2^1 line in the $0\rightarrow\nu_3$ band of the SF_6 molecule, coinciding with the $P(18)$ active transitions of a CO_2 laser in the 00^01–10^00 band. The CO_2 laser was operated continuously in the single-frequency regime and the width of the emission line was of the order of 20 kHz. Estimates showed that the advance of the radiation phase because of various perturbations did not exceed 10^{-1} rad during a typical observation time.

Figure 4 shows the experimental results converted to the half-width in accordance with the expression $\Gamma=(2\pi T_2)^{-1}$, where T_2 is the decay time of the dipole moment. We can see that the curve is nonlinear. The initial slope is considerably higher than the values reported earlier[14–17] and it amounts to 26 MHz/Torr. Curve 2 in Fig. 4 represents the line widths measured by the method of saturated absorption in the relevant spectral range.[17] At high pressures the slopes of curves 1 and 2 are identical within the limits of the experimental error.

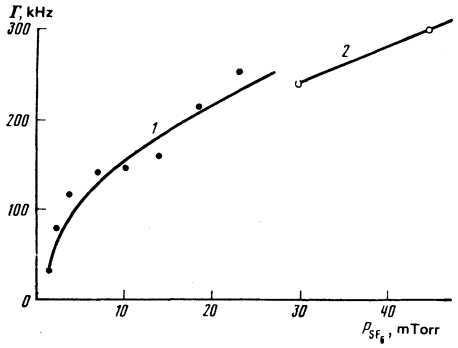

FIG. 4. Dependence of Γ on the gas pressure.

The nonlinear pressure dependence of the reciprocal of the relaxation time is due to the fact that at low pressures the intensity of the RCTP is influenced not only by the dephasing and quenching collisions, but also by the velocity-changing collisions. The condition under which the Doppler phase advance because of a change in the velocity as a result of a collision is significant can be represented in the form

$$\Delta v_z \geqslant \lambda/2\pi T_2, \tag{15}$$

where Δv_z is the change in the velocity in the scattering through small angles. At high pressures, corresponding to the condition

$$\Delta v_z \ll \lambda/2\pi T_2, \tag{16}$$

the phase advance during the relaxation time T_2 becomes insignificant. Therefore, the collisions resulting in scattering through small angles do not contribute to the rate of relaxation of the dipole moment. The conditions (15) and (16) are equivalent to $\theta k v_0 \gtrsim \Gamma$ and $\theta k v_0 \ll \Gamma$. It is shown in Ref. 18 that at low pressures, when $\theta k v_0 > \Gamma$, the broadening is governed by the total scattering cross section $\Gamma = n v_0 (\sigma_e + \sigma_i)$, where σ_e and σ_i are the cross sections of the elastic and inelastic scattering processes. In the range of pressures where $\theta k v_0 < \Gamma$ the broadening is entirely due to the inelastic processes. This pressure dependence of the line width is typical of molecular vibrational-rotational transitions, since the upper and lower states in such transitions belong to the same electronic state. Consequently, the amplitudes of the scattering by the upper and lower levels of vibrational-rotational transitions are sufficiently close and there is practically no dephasing as a result of collisions. This was demonstrated experimentally for the SF_6 molecule in Ref. 17.

The slope of curve 1 in Fig. 4 gives the scattering cross section $\sigma = (15 \pm 3) \times 10^{-14}$ cm^2. The difference between the initial and final slopes of curve 1 give the elastic scattering cross section $\sigma_e = (13 \pm 3) \times 10^{-14}$ cm^2. An estimate of the characteristic scattering angle θ gives 6.4×10^{-3} rad and it corresponds to a velocity change by 110 cm/sec as a result of collisions.

Relaxation of the dipole moment in SF_6 has been investigated by the resonance[14-17] and photon echo[19-21] methods. The results obtained by the photon echo method vary considerably with the conditions. Clearly, this variation is due to the nonlinear pressure dependence of the relaxation constant resulting from the influence of weak collisions. A theoretical estimate of the elastic scattering cross section given in Ref. 21 for the SF_6–SF_6 collisions is 11.9×10^{-14} cm^2. The total scattering cross section obtained in the present study is 1.3 times greater than this value. In view of the approximate nature of the calculations, the agreement between the measured and calculated values can be regarded as satisfactory.

The reported investigations have demonstrated that the RCTP can be used to investigate collisions between molecules. In our opinion, it is very important that the results obtained from the time measurements with the aid of the RCTP agree with the spectral data obtained by nonlinear laser spectroscopy.

b. Spectroscopy within the Doppler profile

An equally important application of the RCTP is the feasibility of resolving complex spectral structures within Doppler-broadened profiles. The effect under discussion is strongly resonant and, which is particularly attractive, it gives information on the detuning of the field frequency from the natural frequency of a transition in the form of the beat signal frequency. An experimental investigation carried out using this effect made it possible to resolve reliably the structure of the $Q(38)$ transition responsible for the $F_1^0 + E_0 + F_2^0$ line in the $0 \rightarrow \nu_3$ band of the SF_6 molecule, coinciding with the $P(16)$ active transition in the 00^01–10^00 band of the CO_2 laser.

The RCTP signal at the frequency of the $Q(38)$ line consisted of three components (Fig. 5). This signal had a complex spectral composition. Its spectrogram is shown in Fig. 6. It demonstrates clearly three frequency components separated by intervals of ~0.5 MHz, i.e., each of the components of the transition structure gives rise to its own RCTP at a frequency equal to the detuning of the field from the natural frequency of the component. This result is in agreement with the data of Ref. 22 obtained by investigating saturation resonances.

c. Stabilization of the laser emission frequency and measurements of shifts

The resonant properties of the transient processes in the field of a standing wave can be used to stabilize the emission frequency by locking to the center of an absorption line. When such processes are used in fre-

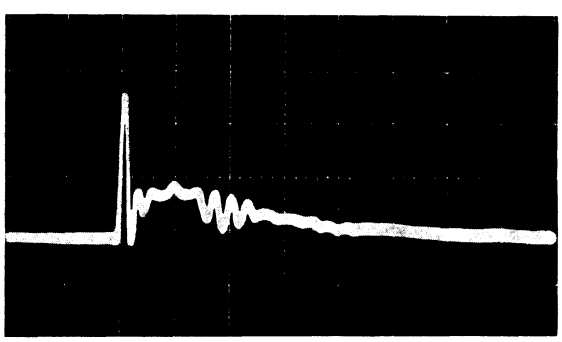

FIG. 5. Oscillogram of the RCTP signal for a line consisting of three components. One horizontal division represents 1 μsec.

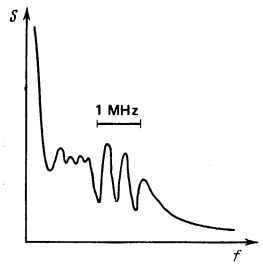

FIG. 6. Spectrogram of the RCTP signal.

quency discrimination, they are found to be characterized by a steep slope, because for a high-intensity signal they ensure resolution approximately equal to the homogeneous width 2Γ. Moreover, the information itself is obtained in an explicit frequency form. Of the two transient processes—resonant coherent and decay of polarization in the field of the opposite wave—the latter is to be preferred for frequency stabilization. This is due to the different influence of the intensity of the standing wave field on these transient processes. As shown in Sec. 2, the spectral line of the signal due to the decay of polarization in the field of the opposite wave experiences field broadening equal to the broadening of a Bennett dip. If the transition does not have a structure masked by the homogeneous width, the field broadening does not shift the average frequency of the signal due to the decay of polarization in the field of the opposite wave.

The situation is different in the case of RCTP. The influence of a strong field on the RCTP signal should be analogous to the influence of transient processes in the case when an atom is at rest. It is well known that a transient process in a two-level system subjected to a strong resonant field occurs at three frequencies: ω, $\omega \pm [\Omega^2 + (d_{21}E/\hbar)^2]^{1/2}$. The RCTP signal corresponds to the frequency $\omega - [\Omega^2 + (d_{21}E/\hbar)^2]^{1/2} = \omega_{21} + \Omega[1 - (1 + (d_{21}E/\hbar\Omega)^2)^{1/2}]$. In a weak field the expression in the brackets tends to zero. A field shift $\Delta\omega = -(d_{21}E)^2/\hbar^2\Omega$ appears when the field is increased, because the rate of decay is not accelerated. In our experiments the RCTP was observed after switching off a strong field pulse, so that the field shift was minimal in the study of the structure of the $Q(38)$ transition.

The decay of polarization in the opposite wave field can be used to determine the shift of the center of an absorption line as a function of pressure. If radiation from the same laser interacts with two absorption cells characterized by different pressures, it is possible to carry out a correlation comparison of the signals due to the polarization decay in each of the two cells. This experimental procedure increases the resolution and accuracy of the measurements, and makes it possible to relax the requirements in respect of the frequency stability of the laser employed.

4. RELATIONSHIP BETWEEN TRANSIENT PROCESSES IN THE FIELD OF A STANDING WAVE AND OTHER COHERENT PHENOMENA

The RCTP discussed here has certain properties common with other coherent phenomena. We shall now consider this point in greater detail.

a. Free induction decay

The decay of the polarization of a medium in the presence of a standing wave is governed by the homogeneous line width, exactly as in the case of free induction decay.[3] We have shown already that the relationship is closer when we consider a transient process in the direction of propagation of an exciting pulse (i.e., the decay of the polarization in the field of the opposite wave). This transient process is governed, like free induction decay, by a group of atoms which are in resonance with an electromagnetic field. The difference lies, on the one hand, in the methodological aspect, because the transient process is excited in our case not by the Stark field but by a light pulse. Such excitation of a transient process can sometimes be more interesting. On the other hand, the polarization decays at a frequency $2\omega_{21} - \omega$ which depends on the transition frequency and this readily distinguishes this transient process from free induction decay, since it allows us to investigate the line structure, measure shifts, etc.

The difference between the transient process observed in the direction opposite to the propagation of a light pulse (RCTP) is fundamental. This phenomenon cannot be called free induction decay but the decay of induction in the field of a standing wave. We have shown above that the presence of a standing wave suppresses completely the influence of the thermal motion of gas atoms. Consequently, the polarization decays at the frequency of the transition in a time $1/\Gamma$ irrespective of the field.

We excited the transient process with a short light pulse, but is is easily shown that the RCTP appears also when the field of one of the traveling waves is turned on. The method of a light pulse against the background of a standing wave makes it possible to separate easily the influence of a pulse as a source of both the transient process and the standing wave that excludes the Doppler broadening. The RCTP is closest to the results obtained by Levenson[23] who considered the influence of transient processes on the Lamb dip in the case of switching of a standing-wave field. Levenson used also a different approach to the phenomena under discussion. Clearly, the more complex pattern of the transient processes prevented Levenson[23] from separating clearly the process of formation of a polarization wave at the center of a transition in the direction opposite to the propagation of an induction wave.

b. Resonances in separated fields

The RCTP cannot be used directly in the optical range to study resonances in two fields, contrary to the situation in microwave measurements. It is known that in the optical range the influence of the Doppler effect in the observation of resonances in separated fields requires three fields.[24] In the microwave range the Doppler broadening is insignificant and, therefore, the polarization of a gas excited by an electromagnetic wave field decays at the transition frequency in a time $1/\Gamma$ and the energy absorbed from the second pulse behaves with the delay time T as $e^{-\Gamma T}\cos\Omega T$. A similar experiment in the optical range using the RCTP may be carried out as follows: a gas in a standing wave field is subjected to

two consecutive pulses traveling in the opposite directions. Since after the first pulse in the presence of a standing wave the polarization decays at the transition frequency and the influence of the Doppler effect is suppressed, the energy acquired from the second pulse is also proportional to $e^{-\Gamma T}\cos\Omega T$, which follows readily from our discussion in Sec. 2.

c. Wavefront reversal

We have considered so far the case when all three waves $\mathscr{E}$, E_1, and E_2 governing the RCTP are plane and travel along the z axis. We shall now consider how the wavefront of the RCTP behaves. The polarization wave vector $\mathbf{k}_p$ in the RCTP case is governed by the spatial matching condition: $\mathbf{k}_p=\mathbf{k}-\mathbf{k}_1+\mathbf{k}_2$. Since $|\mathbf{k}_p|\approx|\mathbf{k}| = |\mathbf{k}_1|=|\mathbf{k}_2|$, this condition is satisfied in just two cases: 1) $\mathbf{k}=-\mathbf{k}_2, \mathbf{k}_p=-\mathbf{k}_1$; 2) $\mathbf{k}=\mathbf{k}_1, \mathbf{k}_p=\mathbf{k}_2$ (here and later, we bear in mind the fact that one of the waves, namely E_2, travels in the opposite direction). In the former case, we can speak of reversal of the wavefront of E_1. In addition to the spatial phase matching, the occurrence of the RCTP requires that the temporal phase matching should also be satisfied: the phase of $\exp[i(\mathbf{k}t_1-\mathbf{k}_1t_2+\mathbf{k}_2t_3-\mathbf{k}_pt)\mathbf{v}]$ should be the same for all values of $\mathbf{v}$, i.e., $\mathbf{k}t_1-\mathbf{k}_1t_2+\mathbf{k}_2t_3-\mathbf{k}_pt=0$. This condition is obeyed rigorously only in the $\mathbf{k}=\mathbf{k}_1=-\mathbf{k}_2$ case. A small deviation from this condition is permissible provided it does not alter significantly the phase over the width of the Maxwellian distribution: $\Delta k \ll \Gamma/v_0$, $\alpha = \Delta k/k \ll \Gamma/kv_0$.

The angle α governs the permissible angle between the wave vectors of the interacting waves. Thus, the RCTP may be accompanied by reversal of the wavefront of the wave E_1 and this generalizes the resonant wavefront reversal in the four-photon interaction[25] to the case of pulsed fields.

CONCLUSIONS

Our investigation shows that the decay of induction in the presence of a standing wave field has a number of new qualitative singularities interesting from the scientific and practical points of view. A decaying polarization wave appears in the direction opposite to the direction of the polarizing wave. Irrespective of the frequency of the standing wave, the frequency of the decaying polarization wave coincides with the center of the Doppler-broadened transition. There is also a change in the decay kinetics compared with the usual free decay of induction. The RCTP opens up new avenues in spectroscopy, since it makes it possible to investigate effectively not only the relaxation processes, but also the fine structure of the lines masked by the Doppler broadening, shifts of the spectral lines, etc.

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