Raman scattering of light by an excited medium

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We consider the Raman scattering in the general case of a medium which is displaced from its equilibrium state. We calculate the intensity of the Stokes and the anti-Stokes components of the spectrum for different scattering regimes (spontaneous RS, quasistatic and nonstationary SRS regimes). We apply the results to evaluate various schemes of active spectroscopy when the scattering of the probing pulse occurs in a medium which beforehand (or simultaneously) has been excited by coherent fields. In the second part of the paper we show that it is possible that there exists a self-induced transparency effect when short pulses undergo Raman interactions in a lossless equilibrium dispersive medium. We show that stationary self-transmission pulses (SRS solitons) have a Lorentzian shape. We establish that nonstationary incoming pulses of a well-defined form must break up into solitons as they propagate through the medium. We obtain similar results for the case of two-photon resonance absorption of pulses with unequal frequencies.

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Recently methods for studying the optical characteristics of substances (determination of radiative transition constants, of transverse and longitudinal relaxation times, identification of transitions) by means of a coherent pulse action have been developed intensively. Apart from methods using absorption and emission processes (photon echo, self-induced transparency, nutation effect) the method of active Raman scattering spectroscopy has been developed in refs. 1 to 3 and other papers. Its idea is to scatter the probing pulse by the molecular or quasi-particle vibrations of the medium which are active in RS and which are amplified by exciting pulses. In this connection it is of interest to study various cases of the scattering of a laser pulse in a medium excited by coherent fields. We give in the present paper a calculation of the intensity of the scattered light in the general case of a medium displaced from its equilibrium state (Sec. 2). The role of the excitation of the medium consists not only in a deviation of the level occupation from the equilibrium values. In those cases where this excitation leads to a non-vanishing spatial distribution of an off-diagonal element of the density matrix of the medium an additional coherent scattering appears with an intensity which exceeds the level of the usual scattering, both in the spontaneous and also in the amplified regimes. We find below formulae which are suitable for an analysis of the scattering when the medium is excited by various methods. In particular, they are applicable for evaluating various schemes of active spectroscopy which use (for the excitation) resonance and scattered pulses (Sec. 3).

We consider in Sec. 4 the excitation of a medium, which is initially in equilibrium, which is directly connected with an appreciable change in the populations when strong short scattering pulses propagate. We show the possibility of the existence of stationary pulses of stimulating and Stokes radiation (SRS solitons) which propagate through the medium without distortion of its shape. The presence of an effect similar to the self-induced transparency effect [4] is important both for determining the optical properties of a substance from the changes of self-transmission pulses (cf. [5]) and also from the point of view of obtaining ultra-short pulses and of other applications. [6]

1. BASIC EQUATIONS

We shall consider the scattering in the case of an arbitrary ratio of the length τ_i of the pumping pulse and the transverse relaxation time T_2. Such a discussion has been given in [7,8] for an equilibrium medium. In that case the amplification of the intensity of the Stokes wave I_S over a length z satisfies in the quasi-static regime the relation

$$I_s = I_s(0)\exp(\Gamma_0 z), \tag{1}$$

where Γ_0 is the static amplification coefficient. The amplification is appreciably smaller in the non-stationary regime ($\tau_i \ll T_2$):

$$I_s \sim \exp[2(2\Gamma_0\tau_i T_2^{-1}z)^{1/2}]. \tag{2}$$

We now assume that initially the medium is displaced from its equilibrium state in such a way that its polarization is non-vanishing and is a function of z, and that there is some initial given distribution of the molecules over the levels which is a function of z. This means corresponding deviations from the equilibrium values of the off-diagonal and diagonal elements of the density matrix of the medium. We shall assume that the scattering occurs at one pair of levels of the scattering molecules. The Hamiltonian of the scattering by a separate molecule is of the form (for the sake of simplicity we consider a one-dimensional problem and the case of parallel polarizations)

$$\mathcal{H} = \hbar\omega_v\sigma^3 - {}^1/_2\lambda(E_i^- E_s^+\sigma^+ + E_i^+ E_s^-\sigma^-), \tag{3}$$

where σ^3 and $\sigma^\pm$ are Pauli matrices, ω_v the frequency of the molecular transition, λ the scattering matrix element; the quasi-monochromatic fields for the pumping and of the Stokes wave are equal to

$$E_{i,s} = {}^1/_2(E_{i,s}^+ + E_{i,s}^-) = {}^1/_2\mathcal{E}_{i,s}(z,t)\exp[i(\omega_{i,s}t - k_{i,s}z - \varphi_{i,s}(z)] + \text{c.c.},$$

$\mathcal{E}_{i,S}(z, t)$ are slowly changing real amplitudes. The polarizations at the Stokes and the pumping frequencies are equal to

$$P_s^+ = {}^1/_2\lambda N_v\langle\sigma^-\rangle E_i^+, \quad P_i^+ = {}^1/_2\lambda N_v\langle\sigma^+\rangle E_s^+$$

N_V is the density of molecules. In the envelope approximation, with phenomenological account taken of the irreversible relaxation processes and of the dephasing due the inhomogeneous broadening, the Maxwell equations and the equations of motion for the mean values of the polarization give us an initial system that describes the forward scattering:

$$\frac{du}{dt} = -v\Delta\omega - \frac{u}{T_2}, \quad \frac{dv}{dt} = u\Delta\omega - \frac{v}{T_2} + \frac{\lambda}{\hbar}\mathcal{E}_i\mathcal{E}_s W + f(z,t),$$
$$\frac{dW}{dt} = -\frac{\lambda}{\hbar}\mathcal{E}_i\mathcal{E}_s v - \frac{W - W_0^{eq}}{T_1},$$

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$$\frac{\partial \mathscr{E}_s}{\partial z}+\frac{1}{c_s}\frac{\partial \mathscr{E}_s}{\partial t}=-\frac{1}{2}\lambda\mu_0 c_s\omega_s N_V \mathscr{E}_i v,$$
$$\frac{\partial \mathscr{E}_i}{\partial z}+\frac{1}{c_i}\frac{\partial \mathscr{E}_i}{\partial t}=\frac{1}{2}\lambda\mu_0 c_i\omega_i N_V \mathscr{E}_s v, \qquad (4)$$

where

$$u\pm iv=\langle\sigma^{\pm}\rangle\exp\{\mp i[(\omega_i-\omega_s)t-(k_i-k_s)z-(\varphi_i-\varphi_s)]\},$$

$W = \langle\sigma^3\rangle$, W_0^{eq} is the equilibrium value of $W(u_0^{eq} = v_0^{eq} = 0)$,

$$\Delta\omega=\omega_0-\omega_i+\omega_s, \quad c_{i,s}=c/\eta_{i,s},$$

T_1 the longitudinal relaxation time, η_i and η_s the refraction coefficients, and $\mu_0 = 4\pi/c^2$. In accordance with the statement of the problem we shall assume that initially the following quantities are given:

$$u|_{t=0}=u_0(z), \quad v|_{t=0}=v_0(z), \quad W|_{t=0}=W_0(z).$$

In deriving (4) we used the synchronism condition: $k_{i,s} = \omega_{i,s}/c_{i,s}$. In the equations for the polarizations we introduced a random δ-correlated force f(z, t):

$$\langle f(z,t)f(z',t')\rangle=g_s N_1(z)\delta(t-t')\delta(z-z'),$$

where $N_1(z)$ is the density of molecules in the lower level. The source f(z, t) takes into account the priming spontaneous emission in those cases where the natural fluctuations of the medium (when there is no preliminary excitation of the medium and no external Stokes emission) play a decisive role. In what follows we neglect the effects of group retardation, considering the scattering over a length $l \ll \tau_1|1/c_i - 1/c_s|$. We also neglect in (4) the motion of the populations $W = W_0(z)$—the case of not too strong fields (see the estimate in[8] and Sec. 4). In the approximation of a given stepped pumping field ($\mathscr{E}_i(t - z/c_i) \equiv \mathscr{E}_i$ at $|t - z/c_i| \le \tau_1/2$) we have at resonance ($\Delta\omega = 0$) the following equation for the amplitude of the Stokes signal

$$\frac{\partial^2 \mathscr{E}_s}{\partial x\,\partial y}+\beta\frac{\partial \mathscr{E}_s}{\partial y}-a^2\mathscr{E}_s-\frac{\alpha f}{W_1(y)}=0, \qquad (5)$$

where

$$\beta=1/T_2, \quad a^2=\lambda^2\mu_0 c_s\omega_s\mathscr{E}_i^2/2\hbar, \quad \alpha=-\lambda\mu_0 c_s\omega_s N_V\mathscr{E}_i/2,$$
$$x=t-\frac{z}{c_s}, \quad y=-\int_0^z W_1(z')dz', \quad W_1(z)=W_0(z)N_V=\frac{1}{2}[N_2(z)-N_1(z)].$$

The function $W_1(z)$ is half the difference between the densities of molecules in the upper and the lower levels (assumed in what follows to be of constant sign).

2. SCATTERING REGIMES

For a study of the different scattering regimes we solve first the general Cauchy problem for Eq. (5), putting the initial conditions in the form

$$\mathscr{E}_s|_{x=0}=G_0(y), \quad \mathscr{E}_s|_{y=0}=\bar{G}_0(x), \quad \bar{G}_0(0)=G_0(0), \quad \frac{\partial \mathscr{E}_s}{\partial y}\Big|_{x=0}=-\frac{F_0'}{W_1(y)}. \qquad (6)$$

We use the Riemann method to look for the solution at the point $x = \xi$, $y = \eta$ in the xy-plane.[9]

Making the substitution

$$\xi=\tau-\frac{\zeta}{c_s}, \quad \eta=-\int_0^{\zeta} W_1(\zeta')d\zeta',$$

we get

$$\mathscr{E}_s(\zeta,\tau)=\bar{G}_0(\tau')+{}^1/_2 G_0(\zeta)e^{-\beta\tau'}-{}^1/_2 G_0(0)e^{-\beta\tau'}I_0[2a\sqrt{\tau'\psi(\zeta)}]$$
$$+a\sqrt{\psi(\zeta)}\int_0^{\tau'}\bar{G}_0(\tau'-t')\frac{e^{-\beta t'}}{(t')^{1/2}}I_1[2a\sqrt{t'\psi(\zeta)}]dt'+\frac{e^{-\beta\tau'}}{2}\int_0^{\zeta}F_0(z)I_0(\varphi)dz$$
$$-\alpha\int_0^{\tau'}e^{-\beta r}dr\int_0^{\psi(\zeta)}I_0[2a(rs)^{1/2}]W_1^{-1}[\psi(\zeta)-s]f(\tau'-r,\psi(\zeta)-s)ds, \qquad (7)$$

where

$$\tau'=\tau-\frac{\zeta}{c_s} \quad \psi(\nu)=-\int_0^{\nu}W_1(\zeta')d\zeta' \quad \varphi=2a\left[\left(\tau-\frac{z-\zeta}{c_s}\right)(\psi(\zeta)-\psi(z))\right]^{1/2},$$

I_0 and I_1 are Bessel functions of an imaginary argument. Giving one or other form of the boundary functions (6) we get from (7) the value of the Stokes field $\mathscr{E}_S$ for the appropriate scattering regime. We must then in (7) take for the variables ζ and τ the coordinate and the time. According to (4) the function $F_0(z)$ is determined in terms of the initial values of the polarization

$$F_0(z)=-{}^1/_2\lambda\mu_0 c_s\omega_s N_V\mathscr{E}_i v_0(z). \qquad (8)$$

Spontaneous RS. In that case $\bar{G}_0(t) = G_0(z) = 0$. Moreover, in (7) we must take the limit as $a \to 0$ (no amplification). Then

$$\mathscr{E}_s(\zeta,\tau)=\frac{1}{2}e^{-\beta\tau'}\int_0^{\zeta}F_0(z)dz$$
$$-\alpha\int_0^{\tau'}e^{-\beta r}dr\int_0^{\psi(\zeta)}f[\tau'-r,\psi(\zeta)-s]W_1^{-1}[\psi(\zeta)-s]ds. \qquad (9)$$

The expression for $\mathscr{E}_S$ consists of a coherent part, caused by the excitation of the medium, and a noise term. Neglecting the noise part of the polarizability as compared to the regular part, we get

$$\mathscr{E}_s(\zeta,\tau)=-\frac{1}{4}\lambda\mu_0 c_s\mathscr{E}_i e^{-\beta\tau'}\int_0^{\zeta}N_V\omega_s v_0(z)dz. \qquad (10)$$

i.e., the intensity of the Stokes field is quadratic in the number of scattering particles, which is a consequence of the correlation coupling between the particles which occurs when the medium is coherently excited. For the sake of simplicity we consider the case $T_2 = \infty$:

$$\mathscr{E}_s(\zeta,\tau)=\frac{1}{4}\lambda\mu_0 c_s\omega_s\mathscr{E}_i\int_0^{\zeta}N_V[W_0^2(z)+v_0^2(z)]^{1/2}\sin\varphi_0(z)dz.$$

where

$$\sin\varphi_0(z)=-v_0/(W_0^2+v_0^2)^{1/2}.$$

If the excitation of the medium by a field is realized from the equilibrium state ($W = W_0^{eq}$), we have $-W_0^{eq} = (W_0^2 + v_0^2)^{1/2}$. The intensity of the radiation $I_S = c_S\mathscr{E}^2 / = c_S\mathscr{E}_S^2/8\pi\hbar\omega$ is hence equal to

$$I_s=\left(\frac{\lambda\mu_0 c_s\omega_s}{4}\right)^2\frac{\omega_s\eta_i}{\omega_s\eta_s}I_i|W_0^{eq}|^2(N_V\zeta)^2\sin^2\varphi, \qquad (11)$$

where

$$\sin\varphi=\frac{1}{\zeta}\int_0^{\zeta}\sin\varphi_0(z)dz$$

is the average degree of excitation of the molecules. Equation (11) was obtained in[10] by different means. When $F_0(z) \equiv 0$ we get from (9)

$$I_s=g\frac{\lambda^2\mu_0^2 N_V^2\omega_s\omega_i c_s^3}{8c_i}I_i T_2[1-e^{-\beta\tau'}]\int_0^{\zeta}N_1(z)dz. \qquad (12)$$

Quasi-static SRS regime ($\tau_1 \gg T_2$). In that case the amplification of the Stokes wave is described by the formula

$$\mathscr{E}_s(\zeta,\tau)=\bar{G}_0(\tau')\exp\left\{-a^2T_2\int_0^{\zeta}W_1(\zeta')d\zeta'\right\}, \qquad (13)$$

which follows immediately from (4) if we assume that $|\partial/\partial t| \ll 1/T_2$. We can also obtain it from the general solution (7), putting $\beta\tau' \gg 1$ and neglecting noise. For an equilibrium medium (13) goes over into (1).

Non-stationary SRS regime. In that case

$$G_0(\zeta) = \bar{G}_0(\tau - \zeta/c_s) \equiv 0. \tag{14}$$

The noise part of the scattering intensity is for large amplifications ($a \gg 1$) and arbitrary ratios of τ_i and T_2 equal to

$$I_s \approx -\frac{gN_1(0)N_v^2}{32\eta_s^2(\pi\gamma)^{1/2}W_1(0)} \exp\left[\frac{2a^2\psi(\zeta)}{\beta}\right]\left\{1+\Phi\left(\sqrt{2\beta\tau'} - a\sqrt{\frac{2\psi(\zeta)}{\beta}}\right)\right\}, \tag{15}$$

where

$$\gamma = 2\beta\tau' \text{ when } a\sqrt{2\beta^{-1}\psi(\zeta)} \geqslant \sqrt{2\beta\tau'},$$
$$\gamma = 2a^2\psi(\zeta)/\beta \text{ when } a\sqrt{2\beta^{-1}\psi(\zeta)} \leqslant \sqrt{2\beta\tau'}.$$

In the substantially non-stationary state ($\tau_i \ll T_2$) we have

$$I_s = \frac{\alpha^2 g c_s}{128\pi^2 a^3 \hbar\omega_s [\tau'\psi(\zeta)]^{1/2}} \exp\{4a\sqrt{\tau'\psi(\zeta)}\}. \tag{16}$$

When there is no excitation, Eq. (16) goes over into (2). An estimate shows that the coherent part of (7) for the case (14) equals

$$\mathscr{E}_s(\zeta,\tau) \approx \frac{F_0(0)e^{-\beta\tau'}}{4(a^3\tau'[\psi(\zeta)]^{1/2}[-W_1(0)])^{1/2}} \exp\{2a\sqrt{\tau'\psi(\zeta)}\}. \tag{17}$$

In deriving (17) we used the conditions

$$2a[\beta^{-1}\psi(\zeta)]^{1/2} > \tau - \zeta/c_s, \quad a \gg 1.$$

We note that the general solution (7) is valid also when the excitation of the medium leads to an inversion of the populations ($W_1 > 0$). However, in that case only Eqs. (9) to (12) for the spontaneous scattering and Eq. (13) for the regime of quasi-static absorption of the Stokes wave, if it is given for $\zeta = 0$, remain valid.

A similar consideration is also applicable to the description of the anti-Stokes scattering, if we formally in (7) make the changes

$$\mathscr{E}_s \to \mathscr{E}_{as}, \quad \omega_s \to \omega_{as}, \quad c_s \to c_{as}, \quad N_1(\zeta) \to N_2(\zeta), \quad N_2(\zeta) \to N_1(\zeta) \tag{18}$$

(the functions W_1 and $\psi(\zeta)$ change sign). In the case of an inverted population ($W_1 > 0$) the results (9) to (17) then remain valid if we make the substitutions (18). Equations (9) to (12) are applicable to the case $W_1 < 0$ for the spontaneous anti-Stokes scattering, while (13) and (18) hold for the quasi-static absorption of the anti-Stokes incoming field regime.

If the excitation of the medium is realized by fields with resultant wave vector $\mathbf{k}_{exc}$ (the expression for $\mathbf{k}_{exc}$ is determined by the actual scheme for the excitation—see below), which is not the same as the pumping field wave vector $\mathbf{k}_i$, the coherent scattering described by Eqs. (10), (13), and (17) can be observed only at an angle which is determined for the Stokes scattering by the relation

$$\mathbf{k}_s = \mathbf{k}_i - \mathbf{k}_{exc}, \tag{19a}$$

and for the anti-Stokes scattering by the relation

$$\mathbf{k}_{as} = \mathbf{k}_i + \mathbf{k}_{exc}. \tag{19b}$$

3. ACTIVE SPECTROSCOPY SCHEMES

We use the results of Sec. 2 for an evaluation of different cases of exciting the medium. We shall consider the following general scheme. Let the excitation of the medium be realized by one or several pulses. Simultaneously with their action, or with a certain time delay, a probing pulse is incident on the material which is scattered by an excited transition. In what follows we shall consider the following two variants of the general scheme: A) the scattering of the probing pulse is registered at the frequency of the excited transition, B) the scattered radiation is registered through another transition which has a common level with the excited one. We turn to a discussion of several concrete examples of schemes A (subsections I to III) and B (subsection IV).

I. Let the excitation of the medium be realized by a resonant coherent pulse of amplitude $\mathscr{E}_{res}(\zeta, \tau)$, of length $\tau_{res} \ll T_1$ and frequency ω_v (we assume that there is no alternative forbiddenness—molecules without an inversion center).

1. If $\tau_{res} \ll T_2$ (coherent interaction), after the pulse has passed we have (see, e.g., [11])

$$W_1(\zeta) = 1/2(N_2^{eq} - N_1^{eq})\cos\theta(\zeta), \quad v_1(\zeta) \equiv N_v v(\zeta) = 1/2(N_2^{eq} - N_1^{eq})\sin\theta(\zeta),$$

where N_1^{eq} and N_2^{eq} are the equilibrium values of the population densities,

$$\theta(\zeta) = \frac{2p}{\hbar}\int_{-\infty}^{+\infty}\mathscr{E}_{res}(\zeta,t')dt', \quad \operatorname{tg}\frac{\theta(\zeta)}{2} = \operatorname{tg}\frac{\theta(0)}{2}\exp\left(-\frac{\alpha_1\zeta}{2}\right),$$

p is the matrix element of the resonant transition, $\alpha_1 = 4\pi\omega_v^2 p^2/\hbar c\,\eta_{res} T_2^*$ is the absorption coefficient (η_{res} is the refractive index at the frequency ω_v, T_2^* the reversible transverse relaxation time). Here and henceforth we shall assume that the delay time $\tau_D \ll T_2$. We consider the scattering intensity at an angle satisfying the synchronism condition: $\mathbf{k}_s = \mathbf{k}_i - \mathbf{k}_{res}$, $\mathbf{k}_{res} = \mathbf{k}_{exc}$ is the wavevector of the resonance field. In the spontaneous RS regime

$$\mathscr{E}_s(\xi,\tau) = 1/8\lambda\mu_0 c_s\omega_s(N_1^{eq} - N_2^{eq})\mathscr{E}_i e^{-\beta\tau'} \times \int_0^{\xi}\sin\left\{2\operatorname{arctg}\left[\left(\operatorname{tg}\frac{\theta(0)}{2}\right)\exp\left(-\frac{\alpha_1 z}{2}\right)\right]\right\}dz.$$

Far from the self-transmission threshold ($\theta(0) \ll 2\pi$) the intensity of the Stokes emission is for a weakly absorbing medium ($\alpha_i\zeta \ll 1$)

$$I_s = Qe^{-2\beta\tau'}I_i(W_0^{eq})^2(N_v\xi)^2\sin^2\theta(0), \quad Q = \frac{\lambda^2\mu_0^2 c_s^3\omega_s\omega_i}{64c_i}, \tag{20}$$

which is quadratic in the total number of molecules (cf. (11)). When the condition $\mathbf{k}_s = \mathbf{k}_i - \mathbf{k}_{res}$ is violated or when $\tau_D \gg T_2$ the scattering intensity is given by Eq. (12). By virtue of (18) a result, analogous to (20), occurs also for the anti-Stokes scattering at an angle satisfying the condition $\mathbf{k}_{as} = \mathbf{k}_i + \mathbf{k}_{res}$. We note that one can use the τ_D-dependence of a measurement of the damping of the coherent spontaneous scattering to determine the time T_2 directly.

We give in Fig. 1 the total scattering intensity I_s^{tot} as a function of the synchronism angle (curve 1). The quantity I_s^{tot} consists of the coherent part I_s^{coh}, described by Eq. (20) (curve 2) and an incoherent isotropic part, described by Eq. (12) (curve 3). For comparison we have drawn as a dashed line the intensity of the noise scattering I_s^{noise} when the medium is not excited (Eq. (12) with $N_1(z) \equiv N_1^{eq}$). The curves given here correspond to the case of small $\theta(0)$ when $\sin\theta(0) \sim \theta(0)$. To get dimensionless quantities we used in Fig. 1 the quantities

$$A_1 = Qe^{-2\beta\tau'}I_i(W_0^{eq})^2, \quad A_2 = 8gA_1^{-1}QI_iT_2[1-e^{-\beta\tau'}]N_1^{eq}\left(\frac{1}{\theta^2(0)} - n\right),$$

$$n=\frac{1}{2}\left(1-\frac{N_V}{2N_1^{eq}}\right),\quad A_3=A_1(N_V\zeta)^2-A_2N_V^2N_1^{eq}n\zeta,$$

$$A_4=QI_i(N_V\zeta)^2(W_0^{eq})^2\theta^2(0).$$

In the amplification regime the Stokes intensities are described by Eqs. (13) and (17), where $\theta(0) \ll 2\pi$

$$\psi(\zeta)=\frac{N_2^{eq}-N_1^{eq}}{2}\left[\mathrm{ci}\,\theta(0)-\mathrm{ci}\left\{\theta(0)\exp\left(-\frac{\alpha_1\zeta}{2}\right)\right\}\right]. \qquad (21)$$

The total amplification in the quasi-static regime for $\alpha_1\zeta \ll 1$ is given in Fig. 2. For comparison we have indicated by dashed lines the amplification when there is no excitation which is given by Eqs. (1) and (13) with $W_1(\zeta) \equiv (N_2^{eq}-N_1^{eq})/2$.

2. If $\tau_{res} \gg T_2$, after the resonance pulse has passed through $F_0(\zeta) \equiv 0$ and $\psi(\zeta)$ equals[12]

$$\psi(\zeta)=\frac{N_1^{eq}-N_2^{eq}}{2}\int_0^{\zeta}\left\{1+\exp(-\sigma(N_1^{eq}-N_2^{eq})z)\cdot\right.$$
$$\left.\times\left[\exp\left(2\sigma I_0\left(\tau_{res}-\frac{z}{c_s}\right)\right)-1\right]\right\}^{-1}dz, \qquad (22)$$

where $\sigma = 4\pi\eta_{res}T_2\omega_V p^2/\hbar c$. The spontaneous scattering will then be incoherent. According to (12), (18), and (22) we must then observe amplification of the incoherent spontaneous anti-Stokes signal I_{as} due to the growth of the population in the upper level as compared with its thermal occupation. The maximum signal must be observed when $I_0 \gg (N_1^{eq}-N_2^{eq})l/2\tau_{res}$ when a brightening effect occurs and the populations become equal $(N_2(\zeta) = N_V/2)$. This is shown in Fig. 3. For comparison we show by the dashed line the level of the thermal scattering I_{as}^T when the medium is not excited, which is given by Eqs. (12) and (18) with $N_2(\zeta) = N_2^{eq}/2$. One can use the damping of this signal as function of τ_D to measure the time T_1 directly.

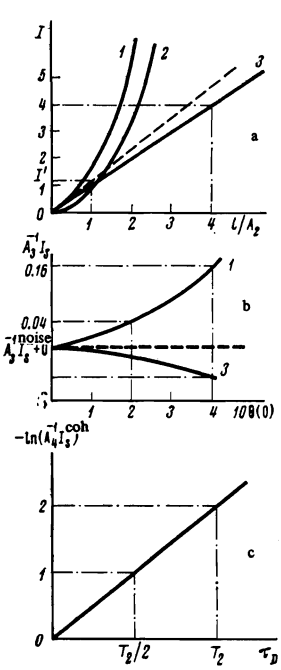

FIG. 1

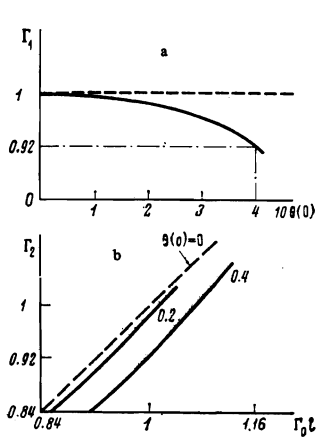

FIG. 2

FIG. 1. The intensity of the spontaneous Stokes scattering as function of (a) the length of the sample, (b) the cross section of the resonance pulse, and (c) the delay time (scheme with short exciting resonance pulse) $I = I_S/A_1A_2^2N_V^2\theta^2(0)$, $I' = \{1 - n\theta^2(0)\}^{-1}$.

FIG. 2. Stokes signal amplification as function (a) of the cross section of the resonance pulse and (b) of the sample length (scheme with short exciting resonance pulse). $\Gamma_1 = a^2T_2\psi(l)/\Gamma_0 l$, $\Gamma_2 = a^2T_2\psi(l)$.

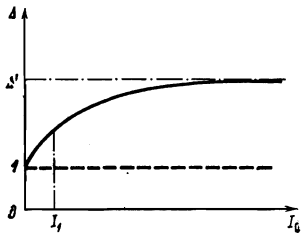

FIG. 3

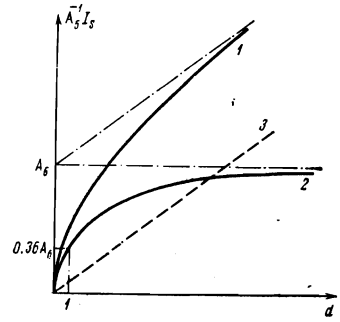

FIG. 4

FIG. 3. The anti-Stokes spontaneous scattering intensity as function of the exciting pulse intensity (scheme with resonance quasi-static exciting pulse. $\Delta = I_{as}/I_{as}^T$, $\Delta' = N_V/2N_2^{eq}$, $I_1 = (N_1^{eq}-N_2^{eq})l/2\tau_{res}$.

FIG. 4. The spontaneous Stokes scattering intensity as function of the sample length (scheme with simultaneous exciting resonance pulse), $d = \sigma|W_0^{eq}|l$.

The Stokes amplification regime is described by Eqs. (13), (16), (17), and (22)—the forward scattering intensity is down as compared to the case without excitation.

3. We consider the case of the simultaneous action of a resonance pulse of length $\tau_{res} \gg T_2$ and initial intensity $I_0 \ll \{2\sigma(\tau_{res}-l/c)\}^{-1}$ and of a probing pulse.[1)] Under those conditions we can neglect the change in the populations. For the most interesting case $\lambda\mathscr{E}_i\mathscr{E}_s \ll 2p\mathscr{E}_{res}$ we easily get, using the results of[12],

$$v=\frac{2p}{\hbar}T_2\mathscr{E}_{res}(0)\exp(\sigma W_0^{eq}\zeta), \qquad \mathscr{E}_{res}^2(0)\equiv\frac{8\pi\hbar\omega_c\eta_{res}}{c}I_0.$$

In the spontaneous scattering regime the intensities of the Stokes radiation when $\mathbf{k}_S = \mathbf{k}_i - \mathbf{k}_{res}$ and of the anti-Stokes radiation when $\mathbf{k}_{as} = \mathbf{k}_i + \mathbf{k}_{res}$ are given by Eqs. (10), (11), and (18) with $\beta = 0$ and $v_0 = v$. For instance, we have for the Stokes intensity

$$I_s=\left(\frac{\lambda p\mu_0\omega_c c_s T_2}{2\hbar\sigma}\right)^2\frac{c_s\omega_i}{c_i\omega_s}I_i\mathscr{E}_{res}^2(0)\{1-\exp(\sigma W_0^{eq}\zeta)\}. \qquad (23)$$

The scattering intensities are in that case independent of the ratio of τ_i and T_2 and, in view of the proportionality to the intensity of the resonance pulse, they can be much larger than the intensity of the usual spontaneous scattering. Curve 1 of Fig. 4 shows the spatial dependence of the total Stokes intensity consisting of the coherent part (23) (curve 2) and the intensity of the noise scattering when there is no excitation (curve 3). We have used in Fig. 4 the parameters

$$A_3=\frac{8gQ}{\sigma|W_0^{eq}|}N_V^2N_1^{eq}I_iT_2[1-e^{-\beta\tau}], \qquad A_4=\frac{2p^2\omega_v^2T_2\mathscr{E}_{res}^2(0)\,|W_0^{eq}|^2}{g\hbar^2\omega_s^2\sigma[1-e^{-\beta\tau}]}.$$

In the amplification regime the Stokes intensity at the synchronism angle is given by Eqs. (13) and (17) with $\psi(\zeta) \approx -N_VW_0^{eq}\zeta$,

$$F_0(0)=\frac{\lambda p\mu_0c_s\omega_s}{\hbar}N_VT_2\mathscr{E}_{res}(0)\mathscr{E}_i$$

and may be much larger than the forward scattering intensity (as $F_0 \sim \mathscr{E}_{res}(0)$).

II. Let the excitation of the medium be realized by an SRS pulse with frequency ω_i^{exc}, amplitude $\mathscr{E}_i^{exc}$, and wavevector $\mathbf{k}_i^{exc}$. We consider the case of a short ($\tau_{exc} \ll T_2$) exciting pulse guaranteeing a non-stationary excitation of the amplitude of the molecular vibrations (the case of a quasi-static excitation pulse is of no interest). This mode of excitation has been applied experi-

mentally.[3] We get analytical expressions for the spontaneous scattering intensity. After the exciting pulse has passed

$$v_0(\zeta)=\frac{\lambda}{\hbar}\int_0^{\tau^{exc}} W_0^{eq}\mathscr{E}_i^{exc}\mathscr{E}_s^{exc}(\eta)\,d\eta, \qquad (24)$$

where

$$\mathscr{E}_s^{exc}(\eta)=-\frac{\lambda\mu_0 N_V\omega_s^{exc}c_s^{exc}\mathscr{E}_i^{exc}}{2W_1^{eq}}\int_0^{\eta}dr\int_0^{-N_V W_0^{eq}\zeta}ds\cdot$$

$$\cdot f\left(\tau-\frac{\zeta}{c_s}-r,-N_V W_0^{eq}\zeta-s\right)I_0(2a^{exc}\sqrt{rs}),$$

$$\omega_s^{exc}=\omega_i^{exc}-\omega_c,\quad c_s^{exc}=c/\eta_s(\omega_s^{exc}),\quad a^{exc}=\mathscr{E}_i^{exc}\sqrt{\lambda^2\mu_0 c_s^{exc}\omega_s^{exc}/2\hbar}.$$

The spontaneous Stokes scattering intensity of the probing pulse $\mathscr{E}_i$ at an angle satisfying the condition $\mathbf{k}_S = \mathbf{k}_i - (\mathbf{k}_i^{exc} - \mathbf{k}_S^{exc})$ is according to (11) equal to

$$I_s=\frac{c_s}{32\pi\hbar\omega_s}\exp\left[-2\beta\left(\tau-\frac{\zeta}{c_s}\right)\right]\int_0^{\zeta}\int\langle F_0(z)F_0(z')\rangle\,dz\,dz',$$

where $F_0(z)$ follows from Eqs. (8) and (24). An estimate gives

$$I_s\sim\exp\left[-2\beta\left(\tau-\frac{\zeta}{c_s}\right)\right]\exp[4a^{exc}\sqrt{-W_0^{eq}N_V\tau_{exc}}\,\zeta], \qquad (25)$$

i.e., I_S can steeply exceed the level of the spontaneous noise scattering (12) due to the exponential dependence on the intensity of the exciting pulse $(\mathscr{E}_i^{exc})^2$. By virtue of (18) we have a result similar to (25) for the anti-Stokes scattering at an angle satisfying the condition $\mathbf{k}_{aS} = \mathbf{k}_i + (\mathbf{k}_i^{exc} - \mathbf{k}_S^{exc})$. In the amplification regime $I_S(\zeta, \tau)$ is given at $\mathbf{k}_S = \mathbf{k}_i - (\mathbf{k}_i^{exc} - \mathbf{k}_S^{exc})$ by Eqs. (13), (15), and (17) with $\psi(\zeta) = -N_V W_0^{eq}\zeta$, and exceeds the forward scattering intensity in the case of an unexcited medium, owing to the increase in the factor of the exponential, according to (8) and (24).

III. Let the excitation of the medium be realized by the simultaneous passage of L pulses with frequencies ω_l, $l = 1, 2, \ldots, L$, such that

$$\sum_{l=1}^{L_1}\omega_l-\sum_{l=L_1+1}^{L}\omega_l=\omega_c.$$

In that case the coherent Stokes and anti-Stokes scatterings are directed along the synchronism angle, satisfying conditions (19a) and (19b), where

$$\mathbf{k}_{exc}=\sum_{l=1}^{L_1}\mathbf{k}_l-\sum_{l=L_1+1}^{L}\mathbf{k}_l.$$

1. If the probing pulse is applied after the exciting pulses have passed, we have, neglecting the change in populations (λ_1 is the interaction matrix element)

$$v_0(\zeta,t)=\frac{\lambda_1 W_0^{eq}}{\hbar}\int_0^{\tau_m}\mathscr{E}_1(\zeta,t')\mathscr{E}_2(\zeta,t')\ldots\mathscr{E}_L(\zeta,t')\exp[-\beta(t-t')]\,dt',$$

$$\tau_m=\min\{\tau_l\}$$

The intensities of the spontaneous radiation at the synchronism angles are given by Eqs. (10), (11), and (18). For instance, for the Stokes radiation

$$\mathscr{E}_s(\zeta,\tau)=-\frac{\lambda_1^2\mu_0 c_s\omega_s N_V W_0^{eq}}{4\hbar}e^{-\beta\tau}\mathscr{E}_1\int_0^{\zeta}\int_0^{\tau_m}\mathscr{E}_1(\zeta',t')\mathscr{E}_2(\zeta',t')\ldots\mathscr{E}_L(\zeta',t')\,d\zeta'\,dt'.$$

We can use a measurement of the damping of the spontaneous coherent signal as function of τ_D, as in scheme II,[3] to determine directly T_2 for the case of molecules with an inversion center (cf. Fig. 1c). In the amplification regime the intensity of the coherent Stokes scattering is given by means of (13) and (17) where $\psi(\zeta) = -W_0^{eq}N_V\zeta$, and it exceeds the forward scattering intensity for the case of an unexcited medium.

2. When quasi-static excitation pulses ($\tau_l \gg T_2$) and the probing pulse act simultaneously we get for the spontaneous scattering amplitude at the synchronism angle

$$\mathscr{E}_s(\zeta,\tau)=-\frac{\lambda^2\mu_0 c_s\omega_s N_V W_0^{eq}}{4\hbar}T_2\int_0^{\zeta}\mathscr{E}_1\mathscr{E}_1(\zeta',\tau)\mathscr{E}_2(\zeta',\tau)\ldots\mathscr{E}_L(\zeta',\tau)\,d\zeta'.$$

Using the substitution (18) we get a similar result for the anti-Stokes scattering.

The case L = 2 (biharmonic pumping), $\mathscr{E}_l(\zeta, \tau)$ = const. corresponds to the coherent active RS spectroscopy scheme, treated theoretically and experimentally in[2].

IV. We studied in the cases I to III scattering at the frequency of the excited transition (scheme A). We now consider scheme B: the scattered radiation is registered at another transition which has a common level with the excited one. Such a scheme can be used for a study of transitions referring to the high-energy part of the spectrum which do not appear in the usual experiments because of the weak occupation of the corresponding levels. This possibility crops up in those cases where the exciting pulses produce an effective occupation of these levels by which the probing pulse can then be strongly scattered. Depending on the frequency of the exciting pulses such a scheme enables us to study electron-vibrational, vibrational-rotational transitions, transitions between anharmonic levels in the range of one type of normal vibrations, and also transitions between different kinds of normal vibrations. We do not give here the formulae for these cases, since one can easily obtain them through obvious modifications of the equations of Sec. 2 and subsections I to III. As the exciting pulses do not change the off-diagonal elements of the density matrix for the transition which is registered ($F_0 = 0$), the role of the exciting fields is now reduced to a change in the population of one of those levels by which the probing pulse is scattered. All excitation schemes are thus described by Eqs. (12), (13), (15), and (18) where $\psi(\zeta)$ is calculated for the different cases from the formulae of subsections I to III. We note further that the scattered radiation is now directed at the same synchronism angles as those at which it is observed when there is no excitation.

4. THE SELF-INDUCED TRANSPARENCY EFFECT. SRS SOLITONS

We considered in Secs. 1 to 3 the case, usually encountered in experiments, of not too strong fields when one can neglect the change in the populations. Under those conditions the propagation of the pumping pulses and of the Stokes radiation through the medium leads to a growth in the Stokes radiation and to the depletion of the pumping. We shall show that under the conditions of a coherent interaction (pulse lengths $\ll T_2$) a stationary scattering regime is possible when the pulses are sufficiently strong (we give estimates below)—the self-induced transparency regime when their amplitudes do not change. This means that the energy absorbed by the molecules from the pulses is later transferred coherently to the field because of induced scattering. For this we find stationary solutions of the complete SRS set of equations (3) and (4) taking into account the excitation of the medium during the scattering (change of populations and change in polarization) and the changes in both fields. We shall look for the solution of (4) for $T_1^{-1} = T_2^{-1}$

$= \Delta\omega = 0$ in the form $\mathscr{E}_s = \mathscr{E}_s(\tau - \zeta/V)$, $\mathscr{E}_i = \mathscr{E}_i(\tau - \zeta/V)$, where V is the group velocity of the pulses. One sees easily that only the equality of the group velocities guarantees the stationarity of the self-transmission pulses (solitons). Introducing the variable $t' = \tau - \zeta/V$ we have

$$\frac{du}{dt'} = 0, \quad \frac{dv}{dt'} = \frac{\lambda}{\hbar}\mathscr{E}_i\mathscr{E}_s W, \quad \frac{dW}{dt'} = -\frac{\lambda}{\hbar}\mathscr{E}_i\mathscr{E}_s v. \tag{26}$$

Solving (26) with the initial conditions

$$u|_{\tau=-\infty} = v|_{\tau=-\infty} = 0, \quad W|_{\tau=-\infty} = W_0^{eq}$$

and substituting the expressions for u, v, and W into the Maxwell equations we get

$$\left(\frac{1}{c_s} - \frac{1}{V}\right)\frac{d\mathscr{E}_s^2}{dt'} = -\frac{\Lambda\omega_s}{\eta_s}\frac{d\chi}{dt'}\sin\chi, \quad \left(\frac{1}{c_i} - \frac{1}{V}\right)\frac{d\mathscr{E}_i^2}{dt'} = \frac{\Lambda\omega_i}{\eta_i}\frac{d\chi}{dt'}\sin\chi, \tag{27}$$

where

$$\Lambda = \hbar\omega_0 c N_V W_0^{eq}, \quad \chi(t') = \frac{\lambda}{\hbar}\int_{-\infty}^{t'} \mathscr{E}_i\mathscr{E}_s\, dt''$$

is the angle over which the polarization vectors have rotated. Hence

$$\mathscr{E}_s^2/2a_s = \mathscr{E}_i^2/2a_i = \sin^2 {}^1/_2\chi, \tag{28}$$

where

$$a_s = \frac{|\Lambda|\omega_s c_s V}{\eta_s(V - c_s)}, \quad a_i = \frac{|\Lambda|\omega_i c_i V}{\eta_i(-V + c_i)}.$$

The existence of stationary pulses is thus possible under the conditions a_i, $a_s > 0$ or $c_s < V < c_i$, which requires the satisfying of the condition $c_s < c_i$, i.e., $\eta_s < \eta_i$. Neglecting the non-linear distortion of the dispersion curves we find that the condition $\eta_s > \eta_i$ can, for instance, be satisfied in the anomalous dispersion region or if ω_i and ω_s lie on different wings of the dispersion curve describing one or other linear absorption. When these conditions are satisfied by the solutions (27) and (28) the pulses have the Lorentz form

$$\mathscr{E}_s^2\left(\tau - \frac{\zeta}{V}\right) = \frac{2a_s}{1 + \tau_0^{-2}(\tau - \zeta/V)^2}, \quad \mathscr{E}_i^2\left(\tau - \frac{\zeta}{V}\right) = \frac{2a_i}{1 + \tau_0^{-2}(\tau - \zeta/V)^2} \tag{29}$$

$\tau_0 = \hbar/\lambda(a_i a_s)^{1/2}$. One sees easily that $\Omega \equiv \chi(\infty) = 2\pi$. The pulse energies equal

$$\Omega_s = \frac{\lambda}{\hbar}\int_{-\infty}^{+\infty}\mathscr{E}_s^2\, dt' = 2\pi\sqrt{\frac{a_s}{a_i}}, \quad \Omega_i = \frac{\lambda}{\hbar}\int_{-\infty}^{+\infty}\mathscr{E}_i^2\, dt' = 2\pi\sqrt{\frac{a_i}{a_s}}.$$

The relation between the pulse velocity V and their duration τ_0 is given by the expression

$$V = \frac{c_i + c_s \pm \sqrt{(c_i + c_s)^2 - 4c_i c_s(1 + \delta^2\tau_0^2 c_i^2 c_s^2)}}{2(1 + \delta^2\tau_0^2 c_i^2 c_s^2)}$$

where $\delta = \lambda\mu_0 N_V W_0^{eq}\sqrt{(\omega_i\omega_s)}$. Both values of V lie within the range $c_s < V < c_i$ (the problem of whether one or other value is realized can be elucidated only from the solution of the non-stationary problem). The condition that V be real leads to a limit on the pulse lengths $\tau_0^2 < (c_i - c_s)^2/4c_i^3c_s^3\delta^2$.

We note that in the general case of non-stationary pulses the "energy theorem" (cf. [4]) follows from Eqs. (4)

$$\frac{d\Omega_s(\zeta)}{d\zeta} = -\frac{\lambda\Lambda\omega_s}{\hbar\eta_s}\sin^2\frac{\Omega(\zeta)}{2}, \quad \frac{d\Omega_i(\zeta)}{d\zeta} = \frac{\lambda\Lambda\omega_i}{\hbar\eta_i}\sin^2\frac{\Omega(\zeta)}{2}, \tag{30}$$

where

$$\Omega(\zeta) = \frac{\lambda}{\hbar}\int_{-\infty}^{+\infty}\mathscr{E}_i(\zeta, t)\mathscr{E}_s(\zeta, t)\, dt, \quad \Omega_{i,s}(\zeta) = \frac{\lambda}{\hbar}\int_{-\infty}^{+\infty}\mathscr{E}_{i,s}^2(\zeta, t)\, dt,$$

while

$$\frac{d\Omega(\zeta)}{d\zeta} = \frac{\lambda^2\Lambda}{2\hbar^2}\int_{-\infty}^{+\infty} dt' \sin\chi\left[\frac{\omega_s\mathscr{E}_s^2}{\eta_i} - \frac{\omega_s\mathscr{E}_i^2}{\eta_s}\right] - \frac{\lambda}{\hbar}\int_{-\infty}^{+\infty} dt'\left[\frac{1}{c_s}\mathscr{E}_i\frac{\partial\mathscr{E}_s}{\partial t'} + \frac{1}{c_i}\mathscr{E}_s\frac{\partial\mathscr{E}_i}{\partial t'}\right]. \tag{31}$$

One shows easily that pulses with amplitudes proportional to $\mathscr{E}_i(\zeta, \tau)/\mathscr{E}_s(\zeta, \tau) = \text{const}$ and to $\Omega(0) = 2\pi m$, $m \neq 1$ satisfy Eqs. (30) and (31), while for arbitrary ζ we have

$$\Omega(\zeta) = 2\pi m, \quad d\Omega_s(\zeta)/d\zeta = d\Omega_i(\zeta)/d\zeta = 0,$$

i.e., $\Omega_i(\zeta)$, $\Omega_s(\zeta)$, and $\Omega(\zeta)$ do not change when the pulses propagate. By virtue of the uniqueness of the solution this means that pulses which have the properties $\mathscr{E}_i(0, \tau)/\mathscr{E}_s(0, \tau) = \text{const}$ and $\Omega(0) = 2\pi m$ when entering the medium, must remain proportional. However, the consideration given above shows that only pulses with $\Omega = 2\pi$ are stationary. Hence, such incoming pulses must break up when propagating into m pairs of solitons which remain proportional as they break up.

The formation of SRS solitons becomes impossible in a dispersionless medium when $\eta_i = \eta_s$. It is clear from (27) that in that case there do not exist such values of V that for them these equations simultaneously have a meaning for real $\mathscr{E}_i$ and $\mathscr{E}_s$. We note that in [13] Eqs. (4) were numerically integrated for the case $\eta_i = \eta_s$ ($T_1 = T_2 = \infty$, $\Delta\omega = 0$) and it was shown that as the strong pulses propagate they break up into separate "spikes."

It had also been established earlier [13] by numerical integration that strong incoming pulses break up when there is two-photon absorption. In contrast to the Raman scattering, the virtual level for this process lies between the levels of the operating transition of the molecules, i.e., $\omega_i + \omega_s = \omega_v$. Let us find the soliton solution in that case. One sees easily that one can obtain the description of the two-photon absorption by formally replacing ω_s by $-\omega_s$ in (4). As before, the stationary solutions are here Lorentz-shape pulses (29) with $V < c_s$ and $V < c_i$ (cf. [6], Sec. 12); all symbols have the same meaning as before. The relation between the group velocity V and the pulse length τ_0 is given by the formula

$$V = \frac{c_i + c_s + \sqrt{(c_i + c_s)^2 + 4c_i c_s(\delta^2\tau_0^2 c_i^2 c_s^2 - 1)}}{2(\delta^2\tau_0^2 c_i^2 c_s^2 - 1)}.$$

One shows easily that the inequalities $V < c_i$ and $V < c_s$ for which the solutions (29) exist are satisfied under the conditions

$$\tau_0 \geq \max\left\{\frac{2(c_i + c_s)}{\delta^2 c_i^2 c_s^3}, \frac{2(c_i + c_s)}{\delta^2 c_i^3 c_s^2}\right\},$$

giving a lower bound on the pulse length. In contrast to the Raman interaction in the case of two-photon absorption the solutions (29) for solitons exist in any region of the dispersion curve. When there is no dispersion the solitons can also exist. For proportional incoming pulses with $\Omega(0) = 2\pi m$, as in the RS case, one can establish the break-up into solitons.

In conclusion we give numerical estimates for the threshold amplitudes for self-transmission. For typical substances $\lambda \approx 10^{-23}$ to 10^{-25} cm^3. According to the earlier given formulae, we get, putting $\tau_0 \approx T_2/10$, the following estimate:

$$\mathscr{E}_i \sim \mathscr{E}_s \geq (2\pi\hbar/\lambda\tau_0)^{1/2} \approx (10 - 100)/(T_2)^{1/2}\ [\text{V/cm}].$$

In typical situations $T_2 \sim 10^{-11}$ s for fluids and $T_2 \sim 10^{-8}$ to 10^{-9} s for gases. This gives for fluids $\mathscr{E}_i \sim \mathscr{E}_s \geq 10^7$

to 10^8 V/cm and for gases $\mathscr{E}_i \sim \mathscr{E}_s \geq 10^5$ to 10^6 V/cm.

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[1)]Here and further we neglect in the cases of simultaneous action of exciting and probing pulses the energy exchange between the scattered wave and the exciting pulses.

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