Coherent interaction between phase-modulated light pulses and an active medium of two-level particles with inhomogeneous broadening

É. M. Belenov, P. G. Kryukov, A. V. Nazarkin, and I. P. Prokopovich

P. N. Lebedev Physics Institute, USSR Academy of Sciences

(Submitted 5 November 1990)
Zh. Eksp. Teor. Fiz. **100**, 105–112 (July 1991)

It is shown that induced transitions of an ensemble of two-level particles with an inhomogeneously broadened line in the field of a phase-modulated pulse are analogous to Landau-Zener inelastic transitions in molecules when their terms cross. Even for $\tau \gg T_2^*$, where τ and T_2^* are, respectively, the pulse width and the inhomogeneous relaxation time of the medium, a transition of particles of the entire spectrum $(T_2^*)^{-1}$ from an upper to a lower level is possible, signifying the complete removal by the pulse of the energy stored in the active medium. Analysis of the dynamics of a phase-modulated pulse by means of the Maxwell-Bloch equations shows, in addition, the possibility of amplifying it without the development of field-amplitude oscillations.

1. INTRODUCTION

One of the most interesting trends in modern laser physics is the possibility of obtaining powerful ultrashort laser pulses (USP) of width <1 psec and energies up to several J.[1] The power in the laser beam reaches a level of several terawatts. After focusing, the intensity can reach 10^{20} W/cm^2, and the electric field strength of the light wave exceeds the electric field of the Bohr atom. It is assumed that obtaining such pulses will make it possible to set up experiments on nonlinear quantum electrodynamics.[2]

At the present time, two main approaches are used to obtain powerful USP. The first consists in using wide-aperture excimer lasers to amplify femtosecond pulses in the UV range.[3] The pulses being amplified are obtained from dye lasers, with subsequent conversion to the second harmonic. The amplification linewidth and pulsewidth are such that during amplification, coherent-propagation effects begin to play an appreciable role.[4] The second approach, used both in excimer systems and in near-IR solid state lasers,[5] consists in first producing phase modulation of the USP carrier frequency and stretching the pulse over time. After such a "chirped" pulse has been amplified, it is passed through an artificial medium with dispersion of the group velocity of such magnitude and sign that the pulse is compressed to a width <1 psec.

In both approaches, the amplification takes place either in a medium with an inhomogeneously broadened line, or in a medium whose properties are similar to those of an inhomogeneous ensemble of radiators because of the complex vibration-rotation structure of the working levels.[6] This naturally gives rise to the problem of removal of the energy stored in the active medium under coherent interaction conditions, i.e., when the pulse width τ is smaller than the time T_2 of irreversible polarization relaxation of the substance.[1)]

A remarkable manifestation of coherence effects in active two-level media with a homogeneously broadened line is the possibility of removal by the pulse of the entire energy stored in the medium. The energy is completely removed for pulses with a constant phase and an area equal to $\theta_0 = \pi(2m+1)$, $m = 0, 1, \ldots$. Thus, in an extended amplifier, an initial pulse of small area $\theta_0 \ll 1$ is shaped into a square-wave pulse dependent on self-similar variables, whose energy increases linearly and whose width is reduced in proportion to the path traveled.[10–12] All of the above also applies in the presence of inhomogeneous broadening in the pulse-shaping stage where its width becomes shorter than the time T_2^* of inhomogeneous dephasing of the radiators. However, the attainment by the electric field envelope of the self-similar amplification mode for an initial pulse width $\tau_0 \gg T_2^*$ requires comparatively extended amplifying media. The question therefore arises, should the pulse field in the case of practical interest,

$$T_2^* \ll \tau_0 < T_2, \tag{1}$$

be shaped so that the entire energy stored in the medium is removed at comparatively small (in the limit, infinitely small) amplification lengths? It will be shown below that the problem has a solution, and the determining pulse characteristics are not amplitudinal, but phase characteristics.

2. INTERACTION OF TWO-LEVEL PARTICLES WITH THE FIELD OF A PHASE-MODULATED PULSE AS THE LANDAU-ZENER PROBLEM OF THE PREDISSOCIATION OF A DIATOMIC MOLECULE

Let us consider an inhomogeneous ensemble of two-level particles which interacts resonantly with a pulse field

$$\mathscr{E}(t) = E(t)\cos[\omega t + \varphi(t)], \tag{2}$$

where ω is the carrier frequency, and E and φ are slowly changing amplitude and phase. Considering that the pulse is shorter than the homogeneous dephasing time T_2, we can describe the dynamics of an individual two-level particle with the aid of the probability amplitudes a_j of the upper ($j=2$) and lower ($j=1$) levels:

$$\begin{aligned} i\frac{\partial a_2}{\partial t} &= -\frac{1}{2}\frac{\mu_{12}}{\hbar}E(t)a_1, \\ i\frac{\partial a_1}{\partial t} &= -[\omega_{21}-\omega-\dot{\varphi}(t)]a_1 - \frac{1}{2}\frac{\mu_{12}}{\hbar}E(t)a_2. \end{aligned} \tag{3}$$

Here μ_{12} is the transition dipole moment; $\omega_{21} = \omega_{21}^0 + \Delta\omega$ is the eigenfrequency of the individual particle, detuned from the amplification line center ω_{21}^0 by $\Delta\omega$. Before the interaction with the field ($t \to -\infty$), all the particles are in the

0038-5646/91/070057-04$03.00

upper level, i.e., $a_2(\Delta\omega, -\infty) = 1$, $a_1(\Delta\omega, -\infty) = 0$.

The energy removal efficiency in the ensemble of particles after passage of the pulse ($t \to \infty$) is characterized by the quantity

$$\eta = \int_{-\infty}^{\infty} n_1(\Delta\omega, \infty) g(\Delta\omega) d\Delta\omega, \quad (4)$$

where $n_j(\Delta\omega, t) = |a_j(\Delta\omega, t)|^2$ is the population of the particle levels, and $g(\Delta\omega)$ is the particle distribution function as a function of the detuning ($\int g(\Delta\omega) d\Delta\omega = 1$).

Below we consider phase-modulated pulses. First, however, it is helpful to discuss the case of a pulse with a constant phase.

For $\dot{\varphi} = 0$, the effects of the phase memory of the medium will be manifested only for particles whose eigenfrequencies lie close to the pulse carrier frequency $\omega = \omega_{21}^0$ within the confines of the spectral pulsewidth: $|\Delta\omega| \lesssim \tau_0^{-1}$. From Eqs. (3), we have an approximate expression for the energy level populations of these particles:

$$n_1(t) \approx \sin^2 \frac{\mu_{12}}{2\hbar} \int_{-\infty}^{t} E(t') dt', \quad n_2(t) \approx \cos^2 \frac{\mu_{12}}{2\hbar} \int_{-\infty}^{t} E(t') dt'. \quad (5)$$

Particles that have detunings from resonance that exceed the spectral pulsewidth, $|\Delta\omega| > \tau_0^{-1}$, interact with the field in a quasistationary manner; the polarization and populations in this case are functions of the instantaneous value of the pulse field:[13]

$$n_1(t) \approx \frac{1}{2}\left\{1 - \left[1 + \left(\frac{\mu_{12}E(t)}{\hbar\Delta\omega}\right)^2\right]^{-1/2}\right\},$$
$$n_2(t) \approx \frac{1}{2}\left\{1 + \left[1 + \left(\frac{\mu_{12}E(t)}{\hbar\Delta\omega}\right)^2\right]^{-1/2}\right\}. \quad (6)$$

It follows from expressions (5) and (6) that after passage of the pulse ($E(\infty) \to 0$), only those particles of the ensemble that have memory will remain in the lower level and will thereby contribute to the energy removal of the medium. From the expressions (4) and (5) we obtain the following estimate for the energy removal efficiency:

$$\eta \sim (1 - \cos\theta_0)(\Delta\omega_i \tau_0)^{-1}, \quad (7)$$

where

$$\theta_0 = \frac{\mu_{12}}{\hbar} \int_{-\infty}^{\infty} E(t) dt$$

is the pulse area, and $\Delta\omega_i$ is the inhomogeneous linewidth $\Delta\omega_i \approx (T_2^*)^{-1}$.

We note that the above estimate can be rigorously substantiated when the pulse shape is of the hyperbolic secant form, for which parameter-dependent exact solutions exist.[14] An important consequence of the expression (7) is that for a fixed pulsewidth τ_0, an increase in pulse amplitude (which can be obtained, for example, by focusing the radiation into the active medium) does not lead to an increase in energy removal, so that the efficiency of the latter for $\Delta\omega_i \tau_0 \gg 1$ remains small, $\eta \ll 1$. It is pertinent at this point to emphasize the qualitative difference of this coherent interaction mode (1) from the incoherent, when the pulsewidth is substantially longer than the time of irreversible relaxation of polarization T_2. In the incoherent mode, the energy removal can be increased by means of field broadening, and in principle, it is possible to remove one-half of the total energy stored in an inhomogeneous ensemble.[15]

We now turn to phase-modulated pulses, for which $\dot{\varphi} \neq 0$. The efficiency with which the particle ensemble interacts with a field can be increased by continuously varying the instantaneous pulse frequency $\omega(t) = \omega + \dot{\varphi}(t)$ with time. In the simplest case of phase modulation linearly dependent on time, we have

$$\dot{\varphi}(t) = \beta t, \quad (8)$$

and in order for the instantaneous pulse frequency to pass through resonance with all the eigenfrequencies of the ensemble in a time τ_0, the magnitude of the chirp $\beta = \partial\dot{\varphi}/\partial t$ must satisfy the condition

$$|\beta| \gtrsim \Delta\omega_i \tau_0^{-1}. \quad (9)$$

By virtue of the assumption (1), this condition signifies that when the particles interact with the pulse field, what is significant is not the change in its amplitude, but the rapid phase change at the instant t' when it passes through resonance $\omega(t') = \omega_{21}$. Hence, one can neglect the change in pulse amplitude, assuming it to be constant in the course of the interaction and equal to $E(t')$. Substituting Eq. (8) in the system (3) and considering the foregoing remark, we can readily see that the equations which we have analyzed are analogous to those considered in the theory of Landau-Zener inelastic transitions in the crossing of molecular terms[16] (see also the adiabatic passage through resonance[15]).

Using the asymptotic expression for the transition probability,[16] we obtain the following estimate for the energy removal efficiency:

$$\eta = 1 - \exp\left[-\frac{\pi}{2}\left(\frac{\mu_{12}\bar{E}}{\hbar}\right)^2 |\beta|^{-1}\right], \quad (10)$$

where $\bar{E}$ is the characteristic magnitude of the pulse field strength. The applicability of this equation is determined by the condition that the Landau-Zener interaction time $\tau_{\text{int}} = |\beta|^{-1/2}$ be small in comparison with the pulse width τ_0 and is ensured by the conditions (1) and (9). According to Eq. (10), the energy removal reaches unity when

$$\frac{\mu_{12}\bar{E}}{\hbar} > \left(\frac{2}{\pi}|\beta|\right)^{1/2}. \quad (11)$$

The minimum value of the pulse field at which the entire energy stored in the ensemble is removed obviously corresponds to the minimum possible rate of frequency change $|\beta|_{\min} \approx \Delta\omega_i \tau_0^{-1}$, whence we have

$$\bar{E}_{min} \approx \frac{\hbar}{\mu_{12}}\left(\frac{2}{\pi}\Delta\omega_i \tau_0^{-1}\right)^{1/2}. \quad (12)$$

Thus, control of the pulse phase has the most significant effect on the energy removal in an inhomogeneous ensemble of two-level particles: In the absence of phase modulation at any field amplitude, the removal efficiency is less than $\eta \sim (\tau_0 \Delta\omega_i)^{-1} \ll 1$, and in the presence of a chirp and under the condition $\bar{E} > \bar{E}_{\min}$, the removal efficiency is $\eta \to 1$. It should be emphasized that this effect is essentially nonlinear in the pulse field. Indeed, estimating from the expressions (5) and (8) the energy removal for pulses of small area

$$\theta_0 \approx \frac{\mu_{12}}{\hbar} \bar{E}\tau_0 \ll 1,$$

one can readily ascertain that when the excitation of the transition is weak, chirping of the pulse does not produce a gain in energy removal, and at best (when $\beta \approx = \Delta\omega_i \tau_0^{-1}$), the energy removal efficiencies are found to be equal.

3. NUMERICAL MODELLING OF THE AMPLIFICATION OF A CHIRPED PULSE

Presented below are some results obtained by considering the propagation dynamics of a field pulse with initial phase modulation, specified by the law (8), in the halfspace $z > 0$ filled with a medium of two-level particles with an inhomogeneously broadened Gaussian shape of the amplification line.

The change in the reactive and active components $P_{1,2}$ of polarization of the particle and in the difference of the populations and levels was described by the equations

$$\frac{\partial P_1}{\partial t} = -(\Delta\omega + \dot{\varphi})P_2 - P_1/T_2,$$

$$\frac{\partial P_2}{\partial t} = (\Delta\omega + \dot{\varphi})P_1 - P_2/T_2 + \frac{\mu_{12}^2}{\hbar} En, \quad (13)$$

$$\frac{\partial n}{\partial t} = -\frac{E}{\hbar} P_2 - \frac{n-1}{T_1}$$

with the initial conditions

$$P_1(z, -\infty) = P_2(z, -\infty) = 0, \quad n(z, -\infty) = 1, \quad (14)$$

and the field amplitude and phase were described by the equations

$$\frac{\partial E}{\partial z} + \frac{\varkappa}{c}\frac{\partial E}{\partial t} = \frac{2\pi\omega N}{c\varkappa} \int g(\Delta\omega) P_2(\Delta\omega) d\Delta\omega,$$

$$E\left(\frac{\partial \varphi}{\partial z} + \frac{\varkappa}{c}\frac{\partial \varphi}{\partial t}\right) = \frac{2\pi\omega N}{c\varkappa} \int g(\Delta\omega) P_1(\Delta\omega) d\Delta\omega, \quad (15)$$

where N is the particle number density and $\varkappa$ is the nonresonant index of refraction of the medium.

It is assumed that a pulse with a Gaussian amplitude profile

$$E(t) = E_0 \exp\left[-\frac{1}{2}(t/\tau_0)^2\right], \quad (16)$$

whose carrier frequency ω coincides with the amplification line center ω_{21}^0, is supplied to the input of the medium ($z = 0$). The results of the numerical integration of the system (13), (15) are presented in dimensionless quantities: the z coordinate is measured in units of reciprocal linear gain $\alpha^{-1} = \hbar c\varkappa / 2\pi\omega N \mu_{12}^2 T_2^*$, time t is measured in units of initial pulsewidth τ_0, and the electric field $E(t)$ is normalized to the quantity $(\hbar/\mu_{12}\tau_0)$.

It is convenient to introduce the average efficiency of energy removal on the length L of the active medium according to the relation

$$\bar{\eta} = L^{-1} \int_0^L \eta(z) dz.$$

Figure 1 shows $\bar{\eta}$ as a function of the input-pulse amplitude at the maximum E_0 over relatively short lengths of the active medium ($L = 0.3$). It is evident that when the chirp is absent ($\beta = 0$), an increase in pulse amplitude does not result in an increase in energy removal efficiency. A characteristic feature is the oscillating form of the dependence $\bar{\eta}(E_0)$, caused by the effects of coherent saturation of the particles interacting with the field. As the chirp size increases for $\beta > 1$, first, an increasingly large number of particles enter into the interaction with the field, and second, a change to the mode of Landau-Zener interaction of the particles with the field takes place (Fig. 1). In this mode, an increase in field amplitude leads to a complete removal of energy. In contrast to the case $\beta < 1$, the particle ensemble behaves in an irreversible manner.

Figure 2 shows the dynamics of the field envelope $E(t)$ and of the function $\dot{\varphi}(t)$ of the chirped pulse as the latter propagates in an extended amplifier. Shown for comparison are calculated data for a pulse with the same initial profile of the envelope, but a constant phase $\dot{\varphi} = 0$. On entering the medium, the pulse is chirped in such a way that the frequency tuning during the pulse width τ_0 is approximately equal to the width $\Delta\omega_i$ of the inhomogeneous line, and the magnitude of the field strength is chosen from the condition (12) of complete energy removal. It is interesting to note that amplification in this case is not only more efficient, but substantially more uniform than in the case $\dot{\varphi} = 0$ (cf. Fig. 2a and 2b). This is because a chirped pulse at each fixed point of the active medium at different instants interacts with different particles of the inhomogeneous ensemble. As a result, at considerable amplification lengths, there is an absence of response accumulation effects, which are characteristic of the amplification of pulses without phase modulation and which usually lead to the development of an oscillating profile and to a stretching of the pulse trailing edge.

4. CONCLUSION

It has been shown that the process of coherent amplification of powerful light pulses in a medium with an inhomo-

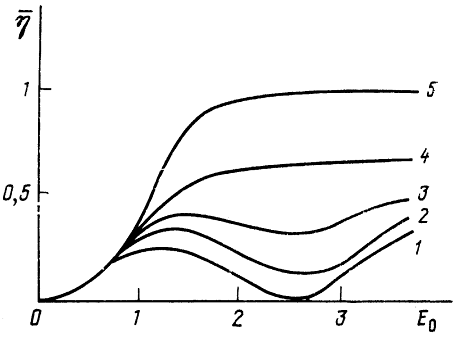

FIG. 1. Mean efficiency $\bar{\eta}$ of energy removal by a pulse in an active medium with an inhomogeneously broadened line vs pulse amplitude E_0 at the input for different values of β: 1—$\beta = 0$; 2—$\beta = 0.3$; 3—$\beta = 0.7$; 4—$\beta = 2$; 5—$\beta = 4$. Calculated parameters: $L = 0.3$; $\Delta\omega_i\tau_0 = 4$; $T_2 = 10$, $T_1 = \infty$.

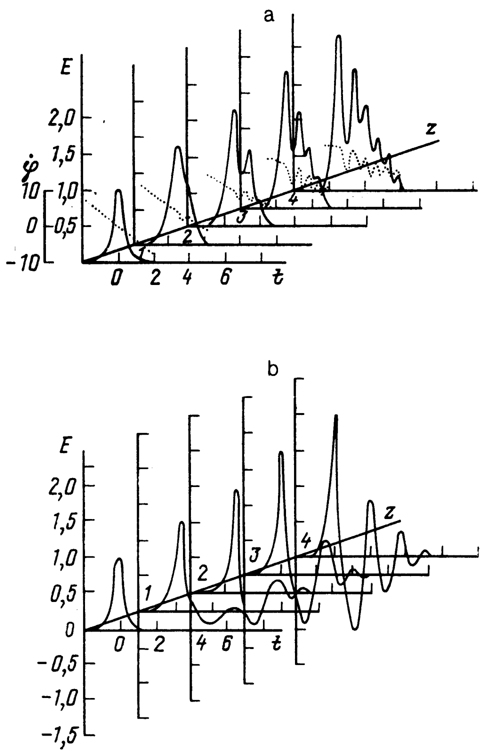

FIG. 2. Evolution of envelope E and of phase modulation $\dot{\varphi}$ of the pulse undergoing amplification in a medium with an inhomogeneously broadened line: a—input pulse is phase-modulated ($\beta = 3$), b—without phase modulation. Calculated parameters: $\Delta\omega_i\tau_0 = 3$; $T_2 = 10$, $T_1 = \infty$.

geneously broadened line can be optimized by modulating the carrier-frequency phase. When the field strength of the amplified phase-modulated pulse exceeds a certain critical value, complete exchange of the energy of the active medium takes place.

To estimate the parameters of the chirped pulse, we examine the amplification in the active medium of an XeCl excimer laser on the most intense vibronic transition $B(v=0)\rightarrow X(v=2)$, corresponding to the wavelength $\lambda = 308$ nm. At a pulse width $\tau_0 > 1$ psec, the contribution of other vibronic transitions can be neglected.[6] The contribution to the amplification on this transition is due to ~ 100 lines in the P and R branches of the rotational structure of the transition; the width of each of the lines is determined by the rotational relaxation time $T_2 = T_R = 10$ psec, and the total linewidth of the transition is $\Delta\omega_i \approx 30$ cm^{-1} (which corresponds to $T_2^* \approx 1$ psec). Assuming that the pulsewidth is $\tau_0 \lesssim 10$ psec and that the transition dipole moment is $\mu_{B\rightarrow X} = 0.5\times 10^{-18}$ esu,[6] we find from the expression (12) that the minimal chirped-pulse intensity providing for complete energy removal is $I \approx 10^7$ W/cm^2 and substantially below the maximum permissible value for the active medium in question.

In conclusion, we note the following. If dispersive elements are used to compress the shaped chirped pulse in an optimal manner, the pulsewidth obviously becomes of order $\tau \lesssim (\Delta\omega_i)^{-1}$, and the amplitude reaches $\overline{E} \gg \hbar\Delta\omega_i/\mu_{12}$. Then the area of the pulse is of order $\theta_0 \sim \pi$, and this pulse can also remove the entire energy stored in the medium. However, amplification of a frequency-modulated pulse is much more advantageous, since self-focusing and breakdown in the active medium limit the peak value of the intensity of the radiation being amplified.

[1)] Note that such aspects of the amplification of chirped pulses as amplitude-phase modulation and its influence on further compression of the pulse (but not on energy removal in the medium) were discussed in Refs. 7–9.

[1] S. A. Akhmanov, V. A. Vysloukh, and A. S. Chirkin, *Optics of Femtosecond Laser Pulses* (Nauka, Moscow, 1988).
[2] R. C. Fernow, H. G. Kirk, I. Bigio *et al.*, Preprint Brookhaven Nat. Lab. 1986 (July 30).
[3] J. H. Glownia, G. Arjavalingam, P. P. Sorokin *et al.*, Opt. Lett. **11**, 79 (1986).
[4] S. A. Akhmanov, V. M. Gordienko, M. S. Dzhidzhoev *et al.*, Kvantovaya Elektron. (Moscow) **13**, 1957 (1986) [Sov. J. Quantum Electron. **16**, 1291 (1986)].
[5] D. Strickland and G. Mourou, Opt. Commun. **56**, 219 (1985).
[6] V. T. Platonenko and V. D. Taranukhin, Kvantovaya Elektron. (Moscow) **14**, 62 (1987) [Sov. J. Quantum Electron. **17**, 34 (1987)].
[7] J. E. Rothenberg and D. Grishkowsky, J. Opt. Soc. Amer. B **2**, 626 (1985).
[8] V. D. Taranukhin and S. Yu. Ten, Zh. Eksp. Teor. Fiz. **95**, 1246 (1989) [Sov. Phys. JETP **68**, 718 (1989)].
[9] V. D. Taranukhin and S. Yu. Ten, Kvantovaya Elektron. (Moscow) **17**, 1162 (1990) [Sov. J. Quantum Electron. **20**, 1071 (1990)].
[10] P. G. Kryukov and V. S. Letokhov, Usp. Fiz. Nauk **99**, 169 (1969) [Sov. Phys. Usp. **12**, 641 (1969)].
[11] V. E. Zakharov, Pisma Zh. Eksp. Teor. Fiz. **32**, 603 (1980) [JETP Lett. **32**, 589 (1980)].
[12] E. M. Belenov, P. G. Kryukov, A. V. Nazarkin *et al.*, J. Opt. Soc. Amer. B **5**, 946 (1988).
[13] N. B. Delone and V. P. Krainov, *Atoms in Strong Light Fields*, Springer, New York, 1985.
[14] A. O. Melikyuan, Dokl. Akad. Nauk Arm. SSR **51**, 214 (1970).
[15] L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms*, Wiley, New York, 1975.
[16] L. D. Landau and E. M. Lifshitz, *Quantum Mechanics*, Pergamon, Oxford, 1977.

Translated by Adam Peiperl