Envelope shock waves on picosecond light pulses in isotropic liquids

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Envelope shock waves on picosecond light pulses were investigated experimentally in capillary fiber lightguides (FL's) filled with organic liquids. The peak power in the shock-wave pulse and the spectral width of the light continuum accompanying the shock waves in the FL were found to be substantially different depending on whether the capillaries were filled with CCl_4 or C_6D_6. The length of the envelope shock wave pulses did not exceed the limit of the time resolution of the electron-optical photodetector (2×10^{-12} s) in both cases. It is shown that the formation of the envelope shock waves occurs under conditions of effective stimulated Raman scattering by molecular vibrations in the liquid core of the FL. The experimental results are compared with existing models of envelope shock waves on ultrashort light pulses. It is suggested that the observed discrepancy between theoretical estimates and measured parameters of envelope shock waves is due to the neglect in the theoretical models of nonlinear losses of light and temporal broadening of light pulses due to waveguide dispersion in the FL, and the fact that the complex physical origin of the nonlinear response of such systems is not taken into account. Possible applications of envelope shock waves to the generation of high-power light pulses for the subpicosecond range and to the investigation of the dynamics of molecular reorientational and librational motion in the liquid phase with subpicosecond relaxation times are discussed.

1. INTRODUCTION

One of the most promising ways of producing powerful light pulses in the subpicosecond range in isotropic liquids exploits the self-compression of such pulses for given dispersion and nonlinearity of the liquid under investigation. In 1966, Ostrovskiĭ showed theoretically that it was possible to produce envelope shock waves on ultrashort light pulses in Kerr liquids with nonzero nonlinear-response inertia.[1] Numerical calculations on the envelope shock waves have resulted in a more detailed physical model of this phenomenon and have established that the most convenient object for the observation of shock waves may be a capillary fiber lightguide (FL) with a liquid core.[2] The formation of envelope shock waves on powerful picosecond light pulses in such systems had already been demonstrated experimentally.[3] It had been found that, in practice, when the FL was filled with pure organic liquid, the deformation of the temporal envelope of light pulses and the concentration of light energy on the leading edge of such pulses occurred when different nonlinear processes compete, and the shock-wave parameters were determined to a considerable extent by the complex physical nature of the nonlinearity of the nonlinear response that was not taken into account in the model described in Ref. 2. On the other hand, this dependence of the parameters of envelope shock waves on the characteristics of nonlinear liquid core governs all possible studies of the dynamics of molecular motion in the liquid phase that exploit the phenomenon of self-interaction of powerful light pulses in the form of envelope shock waves.[4]

In this paper, we report the results of an investigation of mechanisms producing envelope shock waves in FL's filled with liquid organic compounds with very different nonlinear-response inertia, and the results of a comparison between self-compression of light pulses in FL's with predominantly electron or nuclear (orientational) nonlinear susceptibility of core material. We paid particular attention to the effect of competition between self-interaction processes and parametric transformation of light energy, and to the effect of field distribution in the light wave over the modes in few- and many-mode FL's on the evolution of envelope shock waves.

2. THEORETICAL DESCRIPTION OF THE PROPAGATION OF POWERFUL LIGHT PULSES IN EXTENDED WAVEGUIDING MEDIA

The variation in the spatial, spectral, and temporal characteristics of powerful ultrashort light pulses is determined by the combined effect of nonlinearity and dispersion of the medium which, in our case, was the liquid core of a capillary FL. One of the simplifying assumptions used to describe the propagation of light pulses in the FL is the so-called paraxial approximation in which we consider only light rays in the paraxial modes of the FL. We shall suppose that a plane light wave of the form

$$E(r,\ z,\ t)=A(r,\ z,\ t)\ \cos\ (\omega_0 t-kz+\varphi)+\text{c.c.}, \tag{1}$$

where E is the field strength, ω_0 the frequency, k the wave vector, and φ the phase, propagates along the z axis of the FL. The wave satisfies the boundary conditions at $r=R$ (r and R are the radial coordinate and the radius of the FL core, respectively) and is a solution of the nonlinear wave equation

$$\nabla^2\mathbf{E}-\frac{1}{c^2}\frac{\partial^2\mathbf{D}}{\partial t^2}=\frac{2n_2n_0}{c^2}\frac{\partial^2}{\partial t^2}(|E|^2\mathbf{E}), \tag{2}$$

where c is the velocity of light, n_2 and n_0 are the nonlinear refractive index and the refractive index of the undisturbed FL medium, respectively, and D is the dispersion parameter

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of the FL core material.

In the nonlinear isotropic media that we shall consider below, the refractive index n_2 will be positive. The refractive index can be written in the form

$$n(E)=n_0+n_2|E|^2. \tag{3}$$

For light waves with slowly-varying amplitudes, we can obtain a general solution of (2) that describes the propagation of light pulses in FL's with a given degree of nonlinearity and given dispersion of core material. Special solutions of (2), obtained by neglecting either the nonlinearity or dispersion of the FL medium, followed by summation of the contributions of portions of the medium with purely nonlinear and purely dispersive properties, are used to describe the soliton regime of propagation of light pulses and the mechanism responsible for the evolution of envelope shock waves on these pulses.

The basic difference between the dynamic deformation of the shock-wave envelope and the static soliton regime is that the former involves considering the relaxation of the nonlinear medium, i.e., the finite nonlinear-response time. For the media examined in this paper, this parameter is the Kerr nonlinearity time τ_K. In accordance with Ref. 2 (see also Ref. 1), the relaxation equation for a nonlinear medium is

$$\tau_K\frac{d(\delta n)}{dt}=-\left(\delta n-\frac{n_2}{2}|E|^2\right). \tag{4}$$

The evolution of the envelope shock wave in which energy is concentrated on the leading edge of the light pulse can be described by solving Eqs. (2) and (4). The physical significance of this solution can be interpreted as follows. The field of a powerful light wave modifies the refractive index of the medium in the FL by the amount δn, so that a phase change $\delta\varphi$ is produced over a path dz:

$$d(\delta\varphi)=(\omega_0/c)\,dz\delta n. \tag{5}$$

The time variation of $\delta\varphi$ results in frequency modulation of the light wave, so that there is a negative frequency shift on the leading edge of the pulse and a positive shift on the trailing edge. When the nonlinear and dispersive parameters of the medium and the nonlinear response inertia determined by τ_K are exactly balanced, the maximum increase in the group velocity is associated with the top of the light pulse envelope. This is accompanied by a cascade process of energy concentration that results in a narrow high-intensity maximum on the leading edge of the pulse and a smoothing (trailing) of the rear edge. Fisher and Bishel[2] have reported numerical calculations on envelope shock waves in a model medium consisting of alternate segments with purely nonlinear and purely dispersive properties, but did not take into account the influence of linear loss or energy transformation through parametric processes such as stimulated Raman scattering. According to estimates reported in Ref. 4, the variation in the temporal envelope produced in real liquids is largely determined by the dispersion of the medium. In this approximation, the characteristic length of the nonlinear dispersive medium over which the envelope shock wave develops is given by[1,2]

$$z_{sh}=\left(\frac{\lambda}{D(\lambda)\delta n}\right)^{1/2}c\,\Delta t, \tag{6}$$

where $D(\lambda)\equiv\lambda^3d^2n/d\lambda^2$ describes the dispersion of the medium and λ and Δt are, respectively, the wave length of the radiation and the pulse length at entry to the medium. The values of $D(\lambda)$ have been tabulated for most common organic liquids and are of the order of 0.1–0.2 μm in the red region of the spectrum. The choice of the values of λ and Δt establishes a relationship between z_{sh} and the maximum change δn in the refractive index in the field of the light wave. Thus, according to the estimates given in Ref. 2, for a light pulse with $\Delta t=5$ ps, $\lambda=1.06$ μm, and energy density in the pulse of about 22 GW/cm^2, the magnitude of z_{sh} in liquid carbon sulfide is 20 cm.

Basically different conditions of self-compression of powerful light pulses arise in liquid media with electronic nonlinear susceptibility. The electronic nonlinear-response time is shorter than the light-wave period, so that the hyperpolarizability of the molecules in the field of the powerful light wave in the case of the electronic mechanism gives rise to modulation of the refractive index with the period of the light wave, and expression (3) assumes the form[5]

$$n(t)=n_0+n_2E_0^2f(t,z)\cos^2[\omega_0(t-n_0z/c)], \tag{7}$$

where E_0 is the maximum field strength in the light wave and $f(t, z)$ is the normalized envelope functions. Neglecting the difference between group and phase velocities and the change in $f(t, z)$ over the length l of the capillary FL, we can obtain an expression for the amplitude and phase of the light wave leaving the FL, which depend on the parameter

$$B=\tfrac{1}{2}\beta f^2(\tau,l)\quad(\tau=t-n_0l/c,\quad\beta=n_2\omega_0l|E_0|^2/c). \tag{8}$$

The numerical calculations in Ref. 5 can be used to conclude that, in a medium with electronic nonlinear susceptibility, the original light pulse splits into a series of subpulses whose number increases with increasing B. The concept of the temporal pulse envelope is thus seen to lose its meaning for large enough values of B and β. Interference between the subpulses causes an anomalous increase in the steepness of the wave fronts and the formation of a narrow central peak on the temporal structure of the pulse when the dispersion parameter $D(\lambda)$ and the length l of the medium are large enough. The degree of self-compression of the pulse and of the concentration of energy in the peak is then much greater than the analogous parameters of envelope shock waves in media with the Kerr mechanism of nonlinear response.

Pure organic liquids usually have both the electronic and the orientational components of nonlinear susceptibility with different ratios of these components. The most satisfactory model of envelope shock waves[2] is the self-compression of light pulses in liquids containing molecules with purely orientational nonlinearity. These media evidently include carbon sulfide and benzene and its derivatives,[6] whereas for liquid CCl_4 the electronic susceptibility exceeds[7] the nonlinear susceptibility due to the reorientation of molecules in the liquid phase by the light field. Our results on the temporal self-compression of picosecond light pulses in FL's contain-

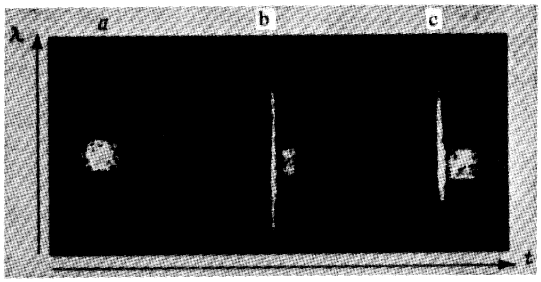

FIG. 1. Photographs of amplitude-time scans of light pulses: a—at exit from the capillary FL $\lambda = 532$ nm, $\Delta t = 35$ ps, $\delta\tilde{\nu} = 0.2$ cm^{-1} $P \approx 0.3$ kW); b—at exit from the FL filled with CCl_4 ($\lambda = 642$ nm, $\Delta t \sim 20$–25 ps, $P \approx 1$ kW); c—at exit from the FL filled with C_6D_6 ($\lambda = 595$ nm, $\Delta t \sim 25$ ps, $P \sim 0.7$ kW).

ing liquid C_6D_6 and CCl_4 are compared below with experimental data on the compression of analogous pulses in the quartz FL in which the nonlinearity is determined entirely by the electronic mechanism.[8]

3. EXPERIMENTAL METHOD

The experimental setup that we have used is described in one of our previous papers.[9] The pump was an Nd^{3+}:YAG laser operating in the lowest transverse mode. Light pulses corresponding to the second harmonic of this laser, with pulse length $\Delta t = 35$ ps, were introduced into the FL by a microobjective and left the FL for the Agat high-speed photorecorder (time resolution 2 ps) or the STE-1 spectrograph. The length and diameter of the quartz FL were 2 m and 40–50 μm, respectively. By varying the temperature of the FL liquid core, we were able to ensure single-mode propagation of light pulses in the CCl_4-containing FL in the temperature range 15–25 °C. The light-pulse energy was measured with the FPM-02 device.

The balance of dispersive and nonlinear parameters of the medium under investigation was achieved by varying the frequency of the pump by exciting stimulated Raman scattering by Raman-active vibrations of molecules in the liquid core of the FL. Thus, for the CCl_4 and C_6D_6 molecules, the Stokes shift was 459 and 944 cm^{-1}, respectively, and the number of SRS components varied from 2 to 10, covering the red region of the spectrum. By suitably choosing the SRS components and the pump power, it was possible to satisfy condition (6) for which z_{sh} is equal to the length of the FL.

The most difficult problem was to record simultaneously the spectral, amplitude, and temporal parameters of the light pulses, which was necessary to confirm the very existence of envelope shock waves. This was done by spectral resolution of the radiation leaving the FL, and by separately and synchronously projecting (1) the radiation at the resonance frequency of the SRS component and (2) the wings of the spectral background near the given component onto different portions of the entrance slit of the Agat camera.[3] The central portion of bands produced by the time scan then corresponds to the radiation at the resonance frequency of the SRS component, while the upper and lower portions of the scan correspond to the Stokes and anti-Stokes component of the spectral background, respectively. The relative intensity was estimated from the brightness of the individual portions of the time scan on the screen of the Agat camera.

4. EXPERIMENTAL RESULTS

Figure 1 shows photographs of the wavelength- and time-scanned light pulses at the end of the FL for the pump (a) and the Stokes SRS components, in CCl_4 (b) and C_6D_6 (c). Microphotometer examination of successive frames showed that the length of these pulses was $\Delta t = 35 \pm 2$ ps and that the time envelope of the pump pulses was almost Gaussian. Effective SRS was observed for the fully symmetric oscillations $\Delta\tilde{\nu} = 459$ cm^{-1} and $\Delta\tilde{\nu} = 944$ cm^{-1} of the CCl_4 and C_6D_6 molecules in the FL for pump power of about 0.3 kW. The number of purely Stokes SRS components varied depending on the pump power and geometric dimensions of the FL, and could be as high as 20–24 (Ref. 9) in the case of CCl_4, whereas only one or two components were recorded in the anti-Stokes region.

The advantage of using the extended waveguide medium in the FL is that we can continuously control the SRS conversion of pump energy. By successively projecting onto the entrance slit of the Agat camera pairs of radiations such as the pump and the first Stokes component, the first and the second components, and so on, we were able to use the above method (see, for examples, Fig. 2 in Ref. 9) to show that, in the FL's that we have investigated, the stimulated Raman scattering occurred without depletion of the pump field, and that pulses corresponding to the SRS components retained the Gaussian shape of the temporal envelope.

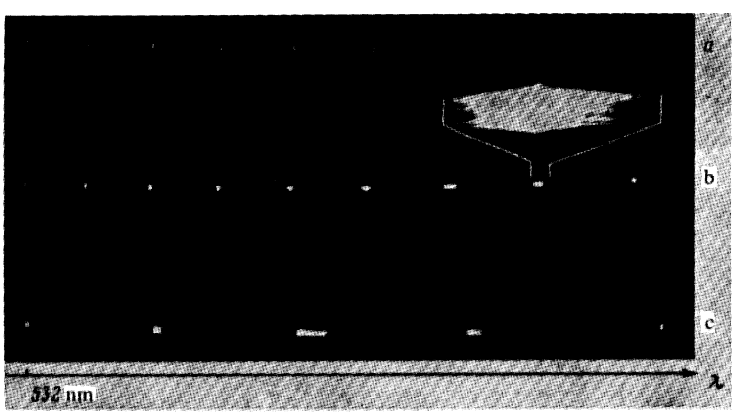

FIG. 2. Photograph of the spectra of nonlinearly transformed radiation at exit from the FL: a—in liquid CCl_4, $P \approx 0.3$ kW; b—liquid CCl_4, $P \simeq$ kW, c—liquid C_6D_6, $P \simeq 0.7$ kW.

In contrast to the quartz FL, for which the spectral width of the pulses was some tens of cm^{-1}, the SRS lines in capillary FL's containing CCl_4 and C_6D_6 were found to be narrow (Fig. 2a). The exact spectral width of these pulses was measured interferometrically and was found to be 1–2 cm^{-1} for the first Stokes components of SRS in the FL.

As the pump power was increased, the SRS components acquired a spectral pedestal that was symmetric for the FL containing CCl_4 and asymmetric for C_6D_6. Moreover, the distortion of the temporal envelope of pulses corresponding to the SRS components was not observed when the background was not extensive. However, when the pump power was of the order of 1 kW, and the width of the spectral continuum for the SRS components was several hundred cm^{-1}, the temporal envelope of the SRS pulses became sharply asymmetric, with a clearly defined intensity peak on the leading edge. The Stokes and anti-Stokes portions of the spectral continuum of the SRS components in CCl_4 can be seen in Fig. 1b to correspond to a maximum on the leading edge of the temporal envelope of length not exceeding the limiting time resolution of the Agat camera, whereas the pulse length at the resonance frequency of the Raman transition in the central portion of the time sweep is 20–25 ps. For the C_6D_6 molecules under analogous conditions, for the most part only the Stokes component of the spectral background develops (Fig. 2c) in which the intensity peak again occurs on the leading edge of the temporal envelope (Fig. 1c).

The pump power corresponding to the envelope of the pulse of the SRS Stokes component for the oscillation with $\Delta\tilde{\nu} = 459\ cm^{-1}$ (Fig. 1b) is $P = 1.0 \pm 0.2$ kW. We note that, for this pump-pulse power, the appearance of the spectral background and of the asymmetry of the temporal envelope was recorded for the series of SRS components due to the CCl_4 and C_6D_6 molecules in the FL's, but the most clearly defined narrow zone of concentrated light energy was found on the leading edge of pulses of SRS components shown in Fig. 1.

The limit in the increase in the radiation power is probably set by the self-focusing threshold in the liquid and, for very low losses, by competition between the parametric processes of nonlinear transformation and the effect of self-interaction of laser radiation. By measuring the area under the amplitude-time scan of pulses on the screen of the Agat camera, we showed that 40–70% of the pulse energy is concentrated on the leading edge of the pulse in the shock-wave regime.

The numerical calculations reported in Ref. 2 show that the optimum length of the FL for the observation of the envelope shock wave is 1.5–2.5 m. When the cross section of our FL's, the measured light pulse parameters, and the values of $D(\lambda)$ and n_2, which are about 0.1 μm and 2.5×10^{-13} cgs (Ref. 10), respectively, are taken into account, we find from (6) that z_{sh} is approximately 3 m, which is in satisfactory agreement with the length of our FL ($l = 2.4$ m). For given FL length, we were able to observe different stages in the evolution of the envelope shock waves because we were able to control the radiation power entering the FL and choose different SRS components and the corresponding dispersive parameters of the liquid under investigation. The dynamics of the evolution of envelope shock waves on light pulses was not examined.

The length Δt of the envelope shock wave pulse is of fundamental importance but it may also be of practical value for the development of a source of ultrashort pulses exploiting the shock-wave phenomenon. The estimated Δt based on the observed spectral width of these pulses (Figs. 2b) and (c), which amounts to some hundreds of cm^{-1}, yield a value of the order of some tens of femtoseconds. Possible reasons for the temporal broadening of the shock-wave pulse in the FL are considered below.

5. DISCUSSION OF RESULTS

Let us now examine the agreement between our experimental results on envelope shock waves on light pulses and predictions of theoretical models of this phenomenon. The essential shortcoming of existing models of envelope shock waves is that they neglect light losses in the medium. It is natural to suppose that the shock wave will not be formed when z_{sh} exceeds $1/\alpha$, where α is the total (linear plus nonlinear) energy loss in the medium. As a rule, carefully purified liquids absorb and scatter by an amount not exceeding $10^{-4}\ cm^{-1}$ and this amount of loss has practically no effect on the formation of shock waves in the FL. The main loss channel in such systems is the nonlinear loss resulting from the redistribution of light energy over the SRS components. In fact, envelope shock waves will always be produced in FL's in the absence of effective SRS conversion. Simple estimates show that the necessary magnitude of the exponential factor in the expression for the threshold SRS intensity[11] is $\exp(glP/\alpha) = 30$, where g is the SRS gain, which is readily achieved for light pulses with $P \simeq 1$ kW in the nonlinear medium of the FL containing CCl_4 or C_6D_6, so that a consistent theoretical model of envelope shock waves on picosecond light pulses in such systems must involve the consideration of parametric processes.

We have discussed in part the competition between nonlinear processes in the FL under picosecond excitation previously.[12] In the limiting case, there may be considerable depletion of the pump field and the SRS components under multiwave conversion, but the very fact that envelope shock waves are produced at the frequencies of several SRS components in our experiment indicates that high nonlinear losses are compensated in the FL's. We assume that the mechanism reponsible for this compensation of nonlinear losses is Raman amplification at the frequencies of the Stokes SRS components in liquid CCl_4 and C_6D_6 in the field of the phase-modulated pump.

The specific feature of the nonlinear transformation of light-pulse energy in waveguiding media such as the FL is that, in contrast to unbounded nonlinear media, here we have the generation of a very broad light continuum.[13] It is well-known that the spectral broadening of a pulse in a liquid medium is entirely due to the phase self-modulation of the light wave.[14] A different situation arises when there is competition between SRS processes and phase self-modulation, which is discussed theoretically in a general form in Refs. 15 and 16. Thus, it is established in Ref. 15 that anomalously large broadening of pulses of the Stokes SRS component is

due to both phase self-modulation of these pulses and the parametric amplification of quantum noise at the frequency of the Stokes component in the field of the phase-modulated pump, where the contributions of these physical mechanisms are not additive. Additional spectral broadening of the pulses by Raman scattering predetermines the possibility that a given z_{sh} can be attained for lower pulse intensity as compared with the unbounded-medium model and the capillary FL in which the spectral broadening is exclusively due to the phase self-modulation effect.

The approximate numerical calculation of the effect of SRS on the formation of shock waves, given in Ref. 4, is confined to the case where the SRS process in the medium results in oscillations on the temporal and spectral envelopes, which mask the shock-wave pulse. Moreover, the amplitude of such oscillations is set by the Raman gain g of the medium. The presence of the characteristic valley on the temporal envelope of the pulses, between the shock-wave peak and the signal at the resonance frequency of the Raman transition (Fig. 1), is not in agreement with the predictions of the theoretical model in Ref. 4. We may suppose that the observed differences between our experimental data and the theoretical model[4] are due to the discrepancy by more than an order of magnitude between the values of g for the liquid CCl_4 and C_6D_6 and those chosen in Ref. 4. Moreover, we cannot exclude the possibility that, in our experiments, a reduction occurred in the magnitude of g in the single- and few-mode FL in comparison with unbounded Raman-active media,[17] which was not examined in Ref. 4.

In view of the foregoing, it is important to note that the numerical calculations on envelope shock waves on pulses in capillary FL's, given in Ref. 2, were confined to single-mode FL's. To elucidate the role of mode composition in the evolution of shock waves in real FC's containing CCL_4 and, especially, C_6D_6, we must analyze the propagation of optical pulses in the FL. As a rule, the waveguide disperison of group velocities in multimode FL's exceeds by an order of magnitude the dispersion in core material,[18,19] so that the temporal broadening of pulses due to waveguide dispersion must be estimated for such systems. It was assumed in Ref. 2 that the difference beween the material and waveguide dispersion in capillary FL's was small and could be described within the framework of perturbation theory by introducing a correction term into the expression for $D(\lambda)$ given by (6). This is incorrect because estimates of the effect of waveguide dispersion have shown that z_{sh} depends strongly on the temporal broadening of light pulses in such systems. In the first approximation (neglecting mode coupling effects), we find that, for the capillary FL's used in our research and for a characteristic waveguide (intermode) dispersion of about 1000 ns/nm·km (Ref. 18), a 10% spread in the light pulse power produces a change of approximately 5% in $z_{sh} = 1$ m and, correspondingly, a broadening of the shock-wave pulse with parameters $\delta\tilde{\nu} = 500$ an^{-1}, $\Delta t = 30$ fs to $\Delta t \approx 0.5$ ps. Thus, in real capillary FL's, the value of z_{sh} will increase monotonically with Δt as a result of waveguide dispersion, whereas the change in the spectral composition of the radiation under the influence of waveguide dispersion and, correspondingly, the correction term in (6), can probably be neglected. On the other hand, possible practical applications of the envelope shock wave phenomenon to the generation of ultrashort light pulses must rely on searches for, and development of, laser-pump sources of high pulse-intensity stability.

In the theoretical models of the evolution of envelope shock waves cited above, the physical nature of the nonlinear-response inertia was not taken into account. When envelope shock waves are described, equation (4) contains the Kerr relaxation time τ_K of the nonlinear medium. We recall that, in media with purely electronic nonlinear response (for example, quartz FL's) for which τ_K is negligible, envelope shock waves do not form[2,20] and the very concept of a light pulse envelope loses its meaning.[5] More than 50% of the nonlinear susceptibility of liquid CCl_4 is due to the electronic mechanism,[7] whereas in liquid benzene, most of the contribution to the cubic nonlinear susceptibility is provided by the orientational mechanism with typical relaxation times of about 3 ps (the electronic contribution does not exceed 20%). It is probable that it is precisely this ratio of the electronic to orientational nonlinearities that determines the observed difference between the intensities of envelope shock wave pulses in capillary FL's containing CCl_4 and C_6D_6 (Figs. 1b and c).

It is clear from Figs. 1b and 2b that the nonlinear response of liquid CCl_4 produced broad-band coherent radiation in our experiments. For these capillary FL's containing CCl_4 and the single-mode quartz FL ($l = 2.4$ and 6 m, respectively), we found by direct measurement with the Agat camera that the coherent continuum from the quartz FL (with a width of several hundred cm^{-1}) exhibited a dispersive delay between the anti-Stokes part of the background and the Stokes wing.[8] At the same time, ultrashort light pulses did not exhibit dispersive broadening or delay in the capillary FL containing CCl_4 (Fig. 1b). We assumed that the instantaneous electronic response of the liquid CCl_4 in the capillary FL occurred only after the concentration of light energy on the leading edge of the pulse by the shock-wave mechanism due to the reorientation of CCl_4 molecules in the light field, so that the shock-wave peak coincided with the electronic response maximum at exit from the FL. We note that the orientational response of the CCl_4 molecules must probably be considered even for pulses that are partially deformed by the electronic response: the steepening of wave fronts, for example, reduces the length of the nonlinear medium or the pulse power at entry to the FL that is necessary for the formation of the envelope shock waves. An analogous conclusion follows from a consideration of the effect of fluctuations on the temporal envelope of a light pulse in the numerical simulation of envelope shock waves given in Ref. 2.

Recent measurements of the nonlinear-response inertia by stroboscopic and interferometric[22] methods, using light pulses with $\Delta t = 30$–60 fs, have established the complex nature of the curve representing the damping of induced birefringence in liquid media. For example, in CS_2, the calculated time for the reorientation of these molecules is 2.1 ps, whereas the faster nonlinear response with the characteristic time of about 0.33 ps was assigned to librational oscillations retarded by collisions between molecules in the liquid phase.

It is natural to assume that the formation of envelope shock waves on picosecond pulses depends on the ratio of the orientational to the librational components, whereas the amplitude and time parameters of shock waves should, in principle, contain information on the dynamics of molecular motion in the liquid phase with subpicosecond and femtosecond relaxation times.[4]

In conclusion, we turn to a comparison between the generation of ultrashort light pulses in the form of envelope shock waves in the form of envelope solitons. Light pulses of limiting length ($\Delta t = 30$ fs) have so far been produced in experiments on the spectral broadening of optical pulses in single-mode quartz fiber lightguides, followed by compression in a dispersive delay line incorporating two diffraction gratings.[23] For 13th order soliton pulses in the quartz FL, numerical estimates of pulse lengths in the anomalous dispersion region yield $\Delta t = 30$ fs when the light pulses at entry to the fiber have $P = 10$ kW and $\Delta t = 1$ ps (Ref. 24). The dynamics of higher-order solitons in glass FL's was also discussed in Ref. 25. Both z_{sh} and the fiber length necessary for the formation of higher-order solitons decrease with increasing light-pulse power. According to Ref. 25, an increase in the rate of frequency scanning of the light wave and a reduction in the relaxation time τ_K of the nonlinear medium in (4) in the shock-wave regime have an analogous effect for higher-order solitons. Possible modification of the light-pulse envelope by the corresponding frequency modulation of the pulse at entry to the FL was considered in Ref. 26. It may be possible to control the evolution of envelope shock waves by superimposing the initial frequency modulation of the light pulse on the change in the pulse frequency due to the nonlinearity of the medium in the FL, which has the same sign and a complicated (orientational and electronic) nonlinearity mechanism. In view of the above properties of self-compression of light pulses in media with electronic nonlinear susceptibility[5] for pulse power of about 10 kW, calculations of the dynamics of higher-order solitons in quartz fibers must be reexamined in the light of the particular features of the electronic response of such systems. For isotropic liquid media, there is very considerable current interest in the development of a self-consistent model of the evolution of envelope shock waves for different ratios of the electronic to orientational hyperpolarizability components with allowance for other competing nonlinear phenomena and the specific features of waveguide propagation of powerful picosecond light pulses in capillary FL's filled with liquid media.

CONCLUSIONS

1. We have determined experimentally the evolution of envelope shock waves on powerful picosecond light pulses in liquid media with very different nonlinear-response inertia in capillary FL's. The peak power of the envelope shock-wave pulse is of the order of some tens of kW for pulse lengths not exceeding 2 ps. When used in combination with stable picosecond laser pulses, the envelope shock wave mechanism is an important new way of generating powerful light pulses of subpicosecond duration.

2. The concentration of energy at the maximum of the envelope shock wave depends on the physical nature of the nonlinear response of the liquid medium so that, by analyzing the conditions under which the shock wave is formed, we can estimate the magnitude of the electronic and nuclear hyperpolarizabilities of molecules in the liquid phase. Theoretical and experimental studies of ultrafast relaxation of nonlinear systems due to the electronic nonlinear response in a time not exceeding the light-wave period are of fundamental importance. Possible applications of envelope shock waves to the study of the dynamics of molecular motion with subpicosecond and femtosecond relaxation times deserve particular attention.

3. The formation of envelope shock waves with maximum compression of the light pulse at the peak of the shock wave occurs in the presence of competition between nonlinear processes accompanying the appearance of the envelope shock waves. Existing models of the evolution of envelope shock waves on ultrashort light pulses in isotropic liquids predict only the qualitative picture of dynamic self-compression of the pulses. It will therefore be necessary to develop theoretical models that will take into account the competition between self-interaction and the essentially nonstationary parametric transformation of the energy of subpicosecond pulses in extended Raman-active waveguide media, and also the differences between the physical nature of the nonlinear response of different liquid media.

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