Dynamic self-diffraction of coherent light beams

V. L. Vinetskiĭ, N. V. Kukhtarev, S. G. Odulov, and M. S. Soskin

Institute of Physics, Academy of Sciences of the Ukrainian SSR, Kiev
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A review is given of the present state of theoretical and experimental work on the interaction of two coherent light beams of equal frequency in a nonlinear medium. It is shown that dynamic self-diffraction is substantially different for media with different types of response. Stationary energy transfer between interacting beams is possible in the case of nonlocal nonsymmetric response, whereas this type of transfer is forbidden in the case of local response. Different mechanisms for self-induced changes in the refractive index are examined together with the corresponding processes of energy transfer between the writing beams. The more important applications of dynamic self-diffraction are discussed.

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CONTENTS

1. INTRODUCTION

Self-interaction of laser beams in a medium that is due to changes in its properties induced by incident radiation is among the most interesting phenomena in nonlinear optics. Self-focusing and self-defocusing of a light beam, self-rotation of the plane of polarization, and so on, are examples.[1,2] Self-interaction between two or more light beams has attracted increasing attention in recent years. Suppose that two coherent light beams of equal frequency, i.e.,

$$E_l = C_l \exp i(\omega t - \mathbf{k}_l \cdot \mathbf{r}), \quad l = 1, 2, \tag{1.1}$$

are incident on a nonlinear medium and, without intersecting, form the interference field

$$|E|^2 = |E_1 + E_2|^2. \tag{1.2}$$

The result of this is a periodic variation in permittivity due, for example, to a cubic linearity of the form

$$\Delta\varepsilon = \varepsilon_2 |E|^2. \tag{1.3}$$

This variation in permittivity is, in effect, a diffraction grating which, in the case of complicated wave fields, is referred to as a holographic grating. The incident beams are diffracted by the grating and new beams appear, propagating in new directions (these are the diffraction orders). There is also a change in the intensity and phase of the writing beams themselves. This holographic effect is referred to[4] as self-diffraction.[1)]

Self-diffraction can also be described in a different way,[5] namely, as frequency-degenerate stimulated four-photon scattering of light by the oscillations of atoms and electrons in the medium that are induced by the interacting beams or as the "scattering of light by light."[2)] In both approaches, a quantitative description of the phenomenon reduces to the solution of the Maxwell equation containing the nonlinear increment $\Delta\varepsilon$. Since all crystals, liquids, and gases have nonzero cubic nonlinearity ε_2, self-diffraction is a very general effect that is produced in all media when the beam power is high enough.

Substantial literature has appeared in recent years on the properties associated with self-diffraction and on its utilization in nonlinear laser spectroscopy for the amplification and transformation of laser beams and for optical data-processing. In this review, we shall examine the physical principles of self-diffraction and the data obtained as a result of studies of this phenomenon and its applications. As a rule, we shall adopt the approach used by the respective authors of the original papers reviewed, but the interaction between beams will in general be interpreted as self-diffraction in a nonlinear medium.[3)] Many of the published papers on self-diffraction are devoted to studies of mechanisms responsible for nonlinearity in particular media. Such

[1)] In the nondynamic situation, the permittivity ε remains practically unaltered during the grating writing process so that there is no beam self-interaction. The change in the properties of the medium becomes effective after development, and subsequent reading does not erase the grating.

[2)] When $\omega_1 \neq \omega_2$, we have stimulated Raman scattering, stimulated Mandel'shtem-Brillouin scattering, intensity holograms, and so on.[10-13] Henceforth, we shall be concerned with the frequency-degenerate interaction unless stated to the contrary.

[3)] The topicality and complexity of the nonlinear diffraction problem was indicated in Ref. 2, where the importance of the development of basic concepts in this field was emphasized.

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questions require separate examination and lie outside the framework of the present review.

The phenomenon of self-diffraction is due to a process in which the writing and reading of the grating occur simultaneously and in a self-consistent manner. The permittivity change $\Delta\varepsilon$ induced by the radiation gives rise to a redistribution of the intensity and phase of the interference field and this, in turn, is reflected in the spatial distribution of $\Delta\varepsilon$. All this determines the complicated dynamic nature of self-diffraction, including the stage after the establishment of the stationary state, and the considerable difference between the properties of the dynamic and static (given) gratings. Thus, the equal-phase surfaces (the "lines" of the grating) become inclined and bend, and the modulation depth becomes a function of position in the grating.[6-9] When the writing process takes place under reflection geometry, new effects connected with the variation in the period of the grating for a given angle between the beams are found to occur.[14]

The fact that dynamic self-diffraction is nontrivial in character is most clearly seen in sufficiently thick targets where, as a result of interference quenching, the intensity of the higher-order diffraction beams becomes negligibly small (this will be referred to as Bragg self-diffraction). It turns out that two coherent beams writing the Bragg grating $\Delta\varepsilon$ in a medium with a local zero-inertia response[4)] do not take part in energy transfer for any ratio of the intensities[5,15,16] although this grating can be detected and investigated by examining the diffraction of an auxiliary test beam by it.[13] Energy transfer remains forbidden in the case of local nonzero-inertia response in the stationary state.[17,18] Moreover, the diffraction of two coherent beams by a given grating, including the unshifted grating, is always accompanied by a change in their intensity with the exception of beams of equal intensity.[19] The absence of energy transfer under dynamic conditions is due to the interaction between the dynamic grating and the light field mentioned above, namely, the grating adjusts itself in such a way that the energy transfer in the Bragg beams is the same and is mutually compensated for any intensity ratio.

As a result of an active search, a number of ways have been found for producing dynamic self-diffraction. They rely on a departure from any of the conditions forbidding energy transfer:

1) Nonlocal response. Bragg self-diffraction of two beams, as in Fig. 1a, in media in which dynamic scattering is shifted in phase relative to the interference field by an angle that is not zero or a multiple of π.

2) Noninstantaneous response. Nonstationary Bragg self-diffraction of two beams, as in Fig. 1b, in a medium with inertial nonlinear response.

3) More than two beams. Self-diffraction of three (or four) beams in a medium with zero-inertia local response, under the conditions of spatial synchronism, as in Figs. 1c and d.

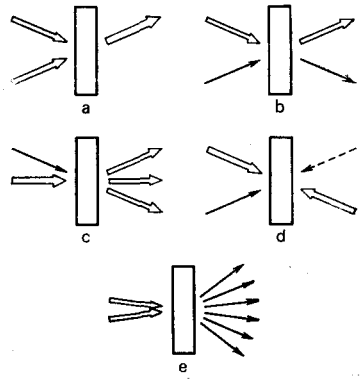

FIG. 1. Schematic illustration of dynamic self-diffraction in a nonlinear medium: a) Bragg self-diffraction of two beams in a medium with nonlocal response; b) nonstationary Bragg self-diffraction in media with local response; c) self-diffraction of three beams in a medium with local response in the presence of spatial locking; d) self-diffraction of four opposite beams, collinear in pairs.

4) Small-volume grating. Dynamic self-diffraction by a thin grating with the participation of higher diffraction orders (Fig. 1e).

Each of these cases will be examined in a separate Section of this review. The last Section will discuss results achieved through the utilization of the self-diffraction effect.

Let us consider briefly the history of the problem. It appears that self-diffraction was, in fact, first discussed in the course of an analysis of a totally different problem,[20] namely, the splitting of a powerful laser beam into "filaments" during self-focusing in a nonlinear medium.[5)] The analysis was, in fact, concerned with a plane wave modulated by a weak interference field equivalent to the degenerate four-photon interaction of Fig. 1c. It was shown that the plane wave was unstable against small field perturbations for $\Delta\varepsilon > 0$. The critical size of the instabilities and, correspondingly, the critical spatial frequency, determined by the nonlinear properties of the medium and the power carried by the field, were found.

All the leading features of Bragg self-diffraction, including the appearance of the dynamic grating, and the conditions for and size of energy transfer in the scheme shown in Fig. 1c were analyzed in Ref. 5 which appeared in the same year. The experimental realization of this scheme on the basis of ruby-laser beams interacting in nitrobenzene followed soon after.[21] This was followed by work on stimulated temperature scattering of the Rayleigh-line wing, and stimulated scattering due to absorption (detailed references are cited in Refs. 11 and 12). From the standpoint of self-diffraction, the initially theoretical and subsequently experimental verification of the possibility of self-diffraction of beams of

[4)] Local response of a medium is defined as that for which the extrema of the resulting grating and of the writing interference field are coincident (this is the so-called unshifted grating).

[5)] This is not a fortuitous coincidence because the two effects, i.e., self-focusing and self-diffraction, are due to cubic nonlinearity.

strictly equal frequency in media with local response under nonstationary conditions[17,22] is the most important result, although the reason for the effect was not established at the time (see Sec. 4).

By now, dynamic self-diffraction has become an independent branch of holography. The emergence of dynamic holography as part of the development of holography generally was recently reviewed in Ref. 9. It was noted that the starting point for the development of dynamic holography was the discovery of stationary energy transfer and its interpretation as a consequence of the writing of the shifted grating in lithium niobate crystals.[39] The first systematic theory of this effect was given in Ref. 8, and interest in studies in this field arose in connection with the possibility of holographic transformation of intensity as a means of correcting the laser wave front.[118,110]

Renewed interest in degenerate four-wave interaction arose in 1977-8 when it was shown that weak beams could be efficiently enhanced and complex conjugate waves could be generated in the stationary state under the conditions of spatial synchronism in a scheme involving crossed beams collinear in pairs.[23,24]

The description of self-diffraction from the standpoint of nonlinear optics is the most convenient in the case of the transparency region when the response can be regarded as being instantaneous and the increment $\Delta\varepsilon$ in the Maxwell equation is determined by the nonlinear polarizability $P^{(NL)}$ of the form[3]

$$P_i^{(NL)}(\omega, t) = \chi_{iklm}(\omega) E_k E_l E_m, \tag{1.4}$$

where the variable subscripts i,k,l,m correspond to the Cartesian components x,y,z, and χ_{iklm} is the cubic nonlinearity tensor whose values are standard characteristics of the material.[3,37]

Almost simultaneously with the emergence of the nonlinear optics approach (in 1967[25]), the other, holographic, approach was developed. The holographic approach provided a more graphic interpretation of the effect of self-diffraction and led to qualitatively new results in a number of cases. The example of self-diffraction of two beams of monopulse radiation from a ruby laser in thin films of a solution of a transmitting dye (cryptocyanine) was used to demonstrate that it was possible to form, read, and transform images in real time with the aid of dynamic holograms. In recent years, self-diffraction has been produced and investigated in a large number of reversible detecting media.[26-35]

In the general case of a medium with nonzero inertia, the nonlinear polarizability is given by

$$P^{(NL)}(\omega, r, t) = \hat{\chi}[E(\omega, r, r', t, t') E(\omega, r, r', t, t')] \times E(\omega, r', t'), \tag{1.5}$$

where $\hat{\chi}[E(\omega,\mathbf{r},\mathbf{r}',t,t')E(\omega,\mathbf{r},\mathbf{r}',t,t')]$ is an integro-differential operator whose form is determined by all the processes involving the migration of excitation in coordinate and energy spaces, i.e., by the change in level population in the medium. Accordingly, $\hat{\chi}$ is no longer a standard characteristic of the medium and depends, in addition, on the temporal and spatial characteristics of the writing fields. For example, in the case of pulsed excitation in a time interval that is small in comparison with the excited-state lifetime τ, the nonlinearity determined by $\hat{\chi}$ is independent of τ but, in the case of continuous illumination, $\hat{\chi}$ does depend on τ. This difference between the effects of self-interaction of beams in a medium with delayed or nonlocal response means that the holographic description of self-diffraction becomes more convenient, since it explicitly involves the phase mismatch between the field and grating, which plays a dominant role in the self-diffraction process.[7,8,18]

It is important to note that the terminology used to describe self-diffraction is not as yet finally established. The phrase temporary holograms has been widely used in foreign literature since the publication of Ref. 25 and is meant to indicate that the holograms are produced and read in the presence of radiation in the course of which the writing beams become modified and that the holograms decay after the end of the writing process. "Real time" holography has a very similar meaning. "Self-diffraction"[4] and "dynamic holography"[9,28,31] are most frequently used in Soviet literature and have the same meaning. If the length of the writing pulse is less than or comparable with the response relaxation time of the recording medium, one speaks of "transient holograms (nonstationary holograms).[27]

To so-called superposition-state holograms[114,115] which appear during the interference between excited atomic states[116] are a new and interesting form of dynamic holograms. Superposition holography has been studied mainly theoretically, but the first experimental results on superposition holograms in ruby have already been reported.[117]

2. ENERGY TRANSFER IN DYNAMIC SELF-DIFFRACTION. GENERAL PRINCIPLES

The theory of the phenomenon is based on the solution of Maxwell's equations that include the nonlinear dependence of the permittivity on the light-field amplitude [see (1.3)]. The specific form of this dependence in each particular case is deduced from the set of constitutive equations for the medium. Since the boundary conditions are periodic, the solution is usually sought in the form of a series in terms of the spatial harmonics of the original interference field acting on the medium. The result of this is the appearance of a set of discrete light beams—higher diffraction orders—and there is also a change in the amplitude and phase of the interacting beams. Depending on experimental conditions (thickness of nonlinear layer, angle between the beams, and size of nonlinearity), the higher diffraction orders may appear (thin dynamic hologram) or may be interference-quenched (volume holography and Bragg diffraction).

Let us begin by considering the solution for the general case when l-th order diffraction has nonzero intensity. If we write the l-th order light wave amplitude in the form $E_l = \sqrt{I_l}\exp(i\varphi_l)$, and the p-th component of the permittivity grating in the form $\varepsilon_p = |\varepsilon_p|\exp(i\Phi_p)$, we

obtain the following expressions for the intensities I_l and phases φ_l of the l-th beam (see the Appendix):

$$\frac{\partial I_l}{\partial z} = -\alpha I_l + k_0^2 k_z^{-1} \sum_{p+m=l} |\varepsilon_p| \sqrt{I_m I_l} \sin(\Phi_p - \varphi_{lm}), \quad (2.1)$$

$$\frac{\partial \varphi_l}{\partial z} = \frac{k_x^2 (l^2-1)}{2k_z} - k_0^2 (2k_z \sqrt{I_l})^{-1} \sum_{p+m=l} |\varepsilon_p| \cos(\Phi_p - \varphi_{lm}), \quad (2.2)$$

where α is the absorption coefficient, k_x, k are the components of the incident-wave vector along the x, z axes, and $\varphi_{lm} = \varphi_l - \varphi_m$ is the phase of the interference field due to the l-th and m-th components of the beams. The first term on the right-hand side of (2.1) describes the absorption of the l-th beam, and the second term takes into account the resultant change in its intensity due to the diffraction of partial beams by the corresponding grating components. The first term in (2.2) determines the change in the phase of the l-th beam relative to the Bragg beams that is connected with the difference between the paths traversed by the beams in the nonlinear medium (the geometric phase difference). The second term is due to the nonlinear "transfer" of phases during self-diffraction.

It follows from (2.1) that the contribution of an individual diffraction term to the intensity of the l-th beam is proportional to $\sin(\Phi_p - \varphi_{lm})$. In other words, the necessary condition for energy transfer between the l-th and m-th components of the light beam in the presence of a phase mismatch between the interference field due to these components and the p-th component of the grating is

$$\Phi_p - \varphi_{lm} \neq 0, \pi. \quad (2.3)$$

The change in the beam phase, in turn, appears as a result of diffraction [described by the sum over the diffraction orders in (2.2)] if $\Phi_p - \varphi_{lm}$ is not an odd multiple of $\pi/2$ and, in the case of non-Bragg beams, as a result of geometric mismatch in the direction of the original beams with $l = \pm 1$ [first term in (2.2)].

For the volume grating in the Bragg approximation ($C_{\pm 1} \neq 0, C_m = 0, |m| > 1$)[14] and when the absorption is weak, the expression given by (2.1) can be written in the form

$$\frac{\partial I_{\pm 1}}{\partial z} = \pm i k_0^2 (2k_z)^{-1} (\Delta\varepsilon^* C_1 C_{-1}^* - \Delta\varepsilon C_1^* C_{-1}). \quad (2.4)$$

For a medium without inertia but with local response, $\Delta\varepsilon = \varepsilon_2 C_1 C_{-1}^*$ [see (1.3)] and $\partial I_{\pm 1}/\partial z = 0$. Energy transfer in the case of dynamic Bragg self-diffraction in a medium with zero-inertia local response is thus found to be absent.

On the other hand, the phase $\varphi = \varphi_{\pm 1} - \varphi_{-1}$ is transferred from the strong beam to the weak in accordance with (2.2):

$$\frac{\partial \varphi}{\partial z} = \frac{k_0^2 \varepsilon_2}{2k_z} \frac{I_{+1} - I_{-1}}{\sqrt{I_{+1} I_{-1}}}. \quad (2.5)$$

This result was first obtained in Ref. 5 and then in Ref. 15 and in several of the subsequent papers. Finally, the exact integrals of Maxwell's equations in a nonlinear reactive homogeneous medium were used in Ref. 16 to derive this conclusion for two opposing waves of equal frequency under the law of conservation of energy and momentum.

Phase transfer, which is equivalent to a change in the phase velocity of the propagating beams, leads to a tilting of the equal-phase surfaces of the writing interference field toward the weak wave, and the absence of energy transfer is a consequence of the strict coincidence between the extrema of the interference field and the induced grating at all times. In the case of the response of a medium with inertia, the same result is obtained under stationary conditions. Experiments performed with different media have confirmed these theoretical conclusions.[22,31]

We emphasize that phase transfer in the absence of energy transfer[36] can be used in phase-sensitive devices to modulate laser radiation and for similar applications. It can also be used to determine the size of the cubic nonlinearity of different media.[37]

The characteristics of energy transfer and the properties of the dynamic grating in a particular nonlinear medium can be found for the first two cases of self-diffraction from the self-consistent solutions of the equations for the intensities and phases of the field and the complex grating amplitude ε_p in the Bragg approximation (see the Appendix):

$$\left.\begin{aligned} \frac{\partial I_{\pm 1}}{\partial z} &= \mp \frac{k_0^2}{k_z} \varepsilon \sqrt{I_{+1} I_{-1}} \sin(\varphi - \Phi), \\ \frac{\partial \varepsilon}{\partial t} + \frac{|\varepsilon| \cos\psi_\tau}{\tau} &= a \sqrt{I_{+1} I_{-1}} \cos(\varphi - \Phi + \psi_a), \\ \frac{\partial \varphi}{\partial z} &= -\frac{k_0^2 \varepsilon (I_{-1} - I_{+1})}{2k_z \sqrt{I_{+1} I_{-1}}} \cos(\varphi - \Phi), \\ \frac{\partial \Phi}{\partial t} &= \frac{\sin\psi_\tau}{\tau} - \frac{a \sqrt{I_{+1} I_{-1}}}{\varepsilon} \sin(\varphi - \Phi + \psi_a), \end{aligned}\right\} \quad (2.6)$$

where ψ_a, ψ_τ are the phase shifts associated with zero-inertia and finite-inertia writing mechanisms, the parameter a is determined by the particular response mechanism, and $\varepsilon = |\varepsilon_{\pm 2}|$ is the modulus of the amplitude of the fundamental component of the permittivity grating. In the stationary case, when $\partial\varepsilon/\partial t = \partial\Phi/\partial t = 0$, Eq. (2.6) assumes the form

$$\left.\begin{aligned} \frac{\partial I_{\pm 1}}{\partial z} &= \pm \frac{k_0^2}{k_z} a\tau I_{+1} I_{-1} \sin(\psi_\tau + \psi_a), \\ \frac{\partial \varphi}{\partial z} &= \frac{a\tau}{2} (I_{+1} - I_{-1}) \cos(\psi_a + \psi_\tau), \\ \Phi &= \varphi + \psi_\tau + \psi_a. \end{aligned}\right\} \quad (2.7)$$

It is clear from (2.7) that the stationary mismatch between the phases and the field at the grating ($\psi_\tau + \psi_a$) is determined by the drift component (ψ_τ) and the nonlinear response of the medium (ψ_a).

3. BRAGG SELF-DIFFRACTION BY THE SHIFTED GRATING

We must now consider the first method of producing Bragg-beam energy transfer, i.e., self-diffraction by the shifted phase grating. It follows from (2.4) that the energy transfer has a maximum when the grating is shifted by $\pi/2$, i.e., by a quarter of the period, relative to the interference field. A graphic explanation of this fact can be given as follows. When self-diffraction takes place, two collinear waves propagate in the direction of each of the interacting beams, namely, the transmitted zero-order wave from one of the beams and the first-order diffraction wave from the other. For the un-

shifted grating, these two waves are phase-shifted by $\pi/2$ relative to each other. When the grating is shifted by a quarter of the period, there is an additional phase difference between the waves, which is equal in magnitude but is different in sign ($\pm\pi/2$). For the acceptor beam, the waves are in phase and add constructively, whereas for the donor beam they are in antiphase and add destructively. This provides us with the possibility of complete interference quenching of one of the interacting beams during self-diffraction by the shifted grating (Fig. 1a).

In the case of the shifted grating ($\psi_\tau + \psi_a = \pi/2$), the initial set of equations given by (2.6) reduces to

$$\frac{\partial I_{\pm 1}}{\partial z} = \mp \frac{k_0^2}{k_z} a\tau I_{+1} I_{-1}, \qquad \frac{\partial \varphi}{\partial z} = 0; \tag{3.1}$$

so that the beam intensities are given by

$$I_{\pm 1}(z) = I_0 \{1 + [m \exp(\Gamma z)]^{\pm 1}\}^{-1}, \tag{3.2}$$

where $I_0 = I_{+1} + I_{-1}$ and $m = I_{-10}/I_{+10}$ is the intensity ratio of the beams at entry to a nonlinear medium of thickness z.

It follows from (3.2) that Bragg self-diffraction by the shifted grating leads to an enhancement of the beam toward which the grating is shifted, independently of the original beam intensity ratio. When the medium thickness z is large enough, the intensities of the two beams combine almost completely into a single beam at exit (Fig. 2).[8]

The gain is given by

$$\Gamma = -\frac{1}{2} \ln \left(\frac{I_{-1}}{I_{-10}} \frac{I_{+10}}{I_{+1}} \right) = \frac{k_0^2}{k_z} \frac{a\tau}{I_0}, \tag{3.3}$$

and characterizes the efficiency of energy transfer, which is proportional to the amplitude of the shifted grating but is independent of the thickness of the medium. We also note that the induced grating is very inhomogeneous in depth (Fig. 2). This is a consequence of the variation in the contrast of the writing field in the course of propagation through the medium. There is also the absence of phase transfer, so that the grating "lines" for plane writing waves are planes parallel to the bisectrix of the incident beams.

Nonlinear media with nonlocal response are quite rare. It is clear that these must be media without a center of inversion for which there is one or several special directions for which the variation of the optical properties is sensitive to the sign of the applied disturbance. Known detecting materials with this property include crystals with linear electrooptic effects, namely, the ferroelectrics $LiNbO_3$, $LiTaO_3$, and $BaTiO_3$, and semiconductors CdS, CdTe, $Bi_{12}SiO_{20}$, and $Bi_{12}GeO_{20}$.

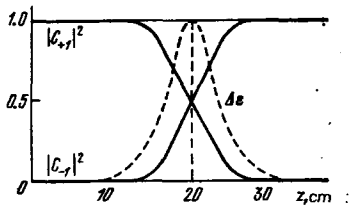

FIG. 2. Dynamic self-diffraction in a shifted stationary gratings. The normalized intensity is shown for two interacting beams as a function of the thickness of the nonlinear layer together with the nonlinear increment in the permittivity as a function of hologram depth.[8]

One of the possible mechanisms that can be used to write the shifted holograms in these crystals in connected with the diffusive redistribution of space charge over the crystal. When a periodically modulated light field is applied to the crystal, an inhomogeneous distribution of nonequilibrium electrons is set up in the conduction band. Diffusion then ensures that some of the electrons enter dark regions of the interference pattern and are captured by deep traps. The space-charge field that modulates the refractive index of the crystal appears as a result of this. In most real crystals, the screening length is much less than the period of the interference field, and the stationary distribution of free carriers is practically the same as the distribution of the pump radiation. The stationary field due to the space charge that results from this diffusion process is proportional to the logarithmic derivative of the distribution $n(x)$ of free carriers:[38]

$$E(x) \sim \frac{kT}{e} \frac{d}{dx} \ln n(x). \tag{3.4}$$

The refractive-index grating with the same spatial frequency as that of the pump radiation is thus shifted by a quarter of the period relative to the interference field.

The correct interpretation of stationary self-diffraction in lithium niobate in terms of the spatial mismatch between the interference field and the grating during the diffusion process was first given in Ref. 39. However, the enhancement of energy transfer during grating mismatch had been predicted much earlier.[5]

Diffraction by crossed gratings was subsequently considered in Refs. 8 and 40, where a number of important qualitative results is reported. Calculations for a specific model of an electrooptic crystal and a detailed comparison with experiment were reported in Ref. 41. A gain of $\Gamma \approx 10$ cm^{-1} was obtained in the case of writing in nominally pure reduced crystals of lithium niobate at the wavelength of the helium-cadmium laser. This is much greater than the gain recorded for most active media of solid-state lasers, which is of the order of the gain of liquid dye lasers. In complete accordance with the theory,[41] the gain increased linearly with the spatial frequency of the grating, and was practically independent of the absolute intensity or the intensity ratio (Fig. 3). The minimum amplified signal was 10^{-6} W/cm^2 and was determined by light scattering in the crystals. It has also been shown[41,42] that nonlocal response is possible during the redistribution of the space charge in electrooptic crystals in sufficiently strong external electric fields. Recent work[43,44] has shown that this phenomenon can be recorded experimentally in potassium niobate crystals.

In most of the detecting media used for dynamic writing, response is local so that the first Fourier component of the refractive-index distribution is the same as the distribution of light in the interference field. Stationary energy transfer is possible in these media but special techniques using the inertia of real non-

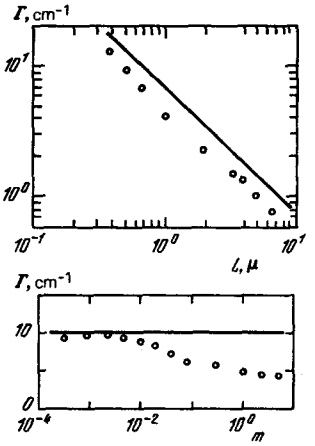

FIG. 3. Dynamic self-diffraction in a stationary shifted grating in an impurity-free lithium niobate crystal.[41] Gain Γ as a function of the period L of the interference field and the ratio m of the intensities of the interfering beams. Solid lines—calculated, open circles—experimental.

linearity have to be used.

The first proposal for an artificial mismatch between the holographic grating and the interference field was given in Ref. 45. In absorbing liquids, nonlinearity is of thermal origin: the refractive index changes as a result of the heat release during light absorption. This grating relaxes relatively slowly through thermal diffusion and the equalization of temperature through the cell. If the detecting medium is displaced during the writing process in the direction perpendicular to the interference field planes, the resulting refractive index distribution becomes mismatched relative to the field (Fig. 4).

The moving-medium method has turned out to be exceedingly useful for the writing of dynamic holograms with pulsed laser beams operated under free-generation conditions.[45-47] Thermal holograms in a moving cell containing a liquid have been written with a ruby laser.[46] Figure 5 shows the envelopes of the sequence of generation peaks for a modified acceptor beam as a function of the rate of displacement of the cell and the direction of its motion. It is clear that the sign of the shift determines the direction of energy transfer, and its velocity governs the magnitude of the effect. A thousandfold amplification of a weak acceptor beam was achieved in this experiment, and it was found that 10%

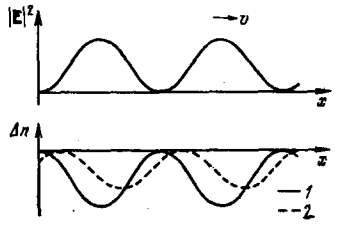

FIG. 4. Dynamic self-diffraction in a layer of absorbing liquid moving at right-angles to the planes of the interfering field.[45] The light intensity distribution $|E(x)|^2$ in the interference pattern and the variation in the refractive index $\Delta n(x)$ is shown for a stationary (1) and a moving (2) medium.

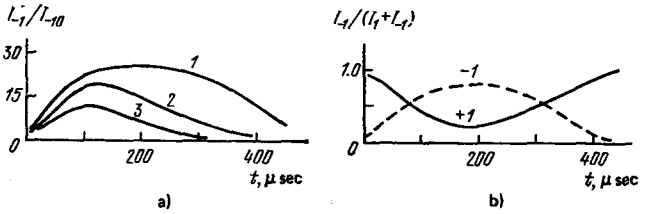

FIG. 5. Dynamic self-diffraction by the thermal grating with artificial phase mismatch:[46] a) time variation of the enhancement of the weak beam when the medium moves with velocities $v = 8$, 0, and -8 cm/sec for curves 1–3, respectively; b) time dependence of intensities leaving the hologram and normalized over the peaks for the amplifying (+1) and modified (−1) beams for $v = 8$ cm/sec and $m = 20$.

of the total laser energy could be concentrated.

In media in which writing of the hologram is connected with the appearance of charged particles, it is possible to use shift techniques based on the application of electric and magnetic fields.[48,49] When a semiconducting crystal is placed in crossed electric and magnetic fields whose directions are parallel to the planes of the interference field, the Lorentz force, parallel to the normal to these planes and having the same sign for electrons and holes, is found to appear. As a result, the stationary distribution of electrons and holes becomes shifted relative to the generating light field.

Shifted holograms of this kind have been obtained by exciting electron-hole pairs in silicon with a Q-switched neodymium-glass laser.[49] A 30% increase in the intensity of the acceptor beam was observed for $E = 10^2$–10^3 V/cm and $H = (2-5) \times 10^3$ Oe. The same beam is weakened in intensity when the polarity of the magnetic field is reversed (Fig. 6). When the writing is repeated on the same piece of the crystal, the effect becomes less well defined because of the accumulation of photochemical transformations. However, the properties of the crystal can be restored by annealing, and the relationship illustrated in Fig. 5 can be reproduced. Stationary enhancement is not observed in the absence of the fields.

We note that the phenomenon of self-diffraction by stationary shifted dynamic gratings has been analyzed within the framework of the holographic approach, and we know of no papers in which the traditional nonlinear-optics approach has been applied to this problem.

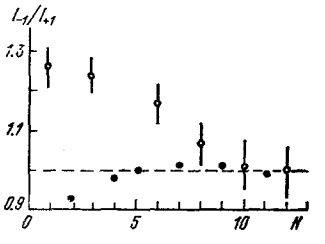

FIG. 6. Dynamic self-diffraction in a free-carrier grating in silicon, shifted by externally applied crossed electric and magnetic fields.[49] The figure shows the intensity ratio for the two beams leaving the crystal as a function of pulse number N. The beams intercepted by the crystal have equal intensity.

4. NONSTATIONARY ENERGY TRANSFER BETWEEN INTERACTING BEAMS

Investigations performed in recent years have shown that media with noninstantaneous local response, in which stationary energy transfer is forbidden, can exhibit efficient energy redistribution between Bragg beams in time intervals comparable with the nonlinearity relaxation time. Since there is no physically special direction in many media with local nonlinearity, the effect appears only when the intensity of the two interacting beams is different, and energy transfer always occurs from the strong to the weak beam.

Qualitative analysis of the phenomenon can be based on the original set of equations given by (2.6) with $\varphi_a = \varphi_\tau = 0$. It follows from the equation for the phase difference φ between the two beams that, when $I_{+1} \neq I_{-1}$, the position of the maxima in the interference field in the medium is a function of time.[18,46] (Fig. 7). At time $t=0$, there is no grating, $\varepsilon = 0$, and the planes of the interference field lie along the normal to the surface of the nonlinear layer ($\partial\varphi/\partial z = 0$). In the stationary case, $\varphi - \Phi = 0$ but $\partial\varphi/\partial z \neq 0$, i.e., the grating is inclined to its original position. Thus, the "lines" of the interference field in the medium are shifted (rotated) during the transient process. At the same time, the time mismatch between the writing and diffraction processes leads to a spatial mismatch between the recorded grating and the interference field produced by the interacting beam. This mismatch [nonzero argument $\varphi - \Phi$ in the formula for the change in the beam intensity given by (2.6)] is, in fact, responsible for the energy transfer between the two beams. The mismatch between the gratings depends on the depth of penetration of the nonlinear medium (Fig. 7b). In the case of stationary states, the holographic grating succeeds in catching up with the interference field, and energy transfer ceases.

It also follows from (2.6) that, initially, energy transfer proceeds from the strong to the weak beam. The transfer is oscillatory in character in the presence of strong phase modulation, $|\varphi - \Phi| > \pi/2$, and vanishes altogether when the beam intensities are equal.

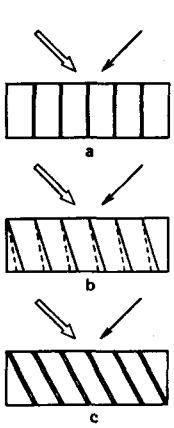

FIG. 7. Different cases of development of the nonstationary energy transfer effect: a) initial time $t=0$; b) time of interaction comparable with the characteristic nonlinearity relaxation time, $t \approx \tau$; c) stationary state, $t \gg \tau$. Solid lines represent surfaces of equal phase in the interference field; broken lines show the distribution of the refractive index in the medium.

Exact solution of (2.6) for arbitrary donor and acceptor beam intensities is difficult to obtain. The first theory of this was given in Ref. 17, where the effect of nonstationary thermal Rayleigh scattering was examined numerically on a computer in the approximation of a given pump-wave field. Analytic calculations have also been reported[18,50] without introducing the given-field approximation, but only for moderate gain. Finally, a computer simulation of the effect for arbitrary acceptor-beam intensity has been reported.[47]

The time dependence of the intensity of the transmitted beam is given by[18]

$$I_{\pm 1}(z) = I_{\pm 10} \pm (\tau z \Delta)^2 (I_{-10} - I_{+10}) I_{10} I_{-10} e^{-t/\tau} \left(\frac{t}{\tau} - 1 + e^{-t/\tau} \right), \tag{4.1}$$

where z is the thickness of the nonlinear layer, τ is the characteristic nonlinearity relaxation time, and the parameter Δ is determined by the ensemble of characteristic parameters of the medium that define the photorefraction process.[18,50] It is clear that the nonstationary increment on the acceptor-beam intensity is quadratic in the thickness of the medium and the depth of modulation of the refractive index, and that, initially, it increases as the square of the time.[17,18,50]

The nonstationary energy transfer effect was first observed during the writing of the thermal grating by waves moving in opposite directions in a solution of carbon tetrachloride containing iodine, and was interpreted as "stimulated temperature reflection."[51] The weak beam was reflected from the phase grating produced during its interaction with the strong beam and was found to increase its intensity by a factor of more than 200 over an interaction length of 2 cm. A similar phenomenon was subsequently observed during the interaction between two beams of light from a laser with self-locked oscillations in an absorbing liquid.[26]

The interpretation of the effect as a nonstationary process was first given in Refs. 17 and 22. Qualitative results on the amplification coefficients that followed from numerical calculations were examined, and a quantitative comparison was carried out for the kinetics of the amplified pulse and the calculated pulse obtained by assuming that the pump pulse was Gaussian (Fig. 8). This comparison resulted in an estimate for the half-width of the central peak in Rayleigh scattering by methyl alcohol containing iodine.

Nonstationary energy transfer can be explained qualitatively in the language of nonlinear optics as a change in the frequencies of the interacting beams[11] during the transient stage of hologram recording. In point of fact, if we recall that the grating that is responsible for diffraction is "unfolded" in space with a definite velocity, the diffracted beam must assume Doppler increments of different sign and frequency. The magnitude of these increments is determined by the reciprocal of the nonlinearity relaxation time, and may amount to hundreds of megahertz for the thermal non-

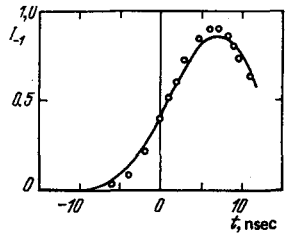

FIG. 8. Nonstationary self-diffraction in an absorbing liquid.[22] Open circles are experimental; solid curve is the result of a theoretical approximation. The figure shows the time variation of the weak-beam intensity.

linearity. However, it must also be remembered that the frequency shift varies with time from zero to some value, and then again becomes equal to zero. The integrated spectrum-broadening effect in self-diffraction in an absorbing liquid was observed in Ref. 12. For media with highly inertial nonlinearity, for example, lithium niobate, the nonstationary frequency shift may amount to a fraction of a hertz, which is practically undetectable. On the other hand, the fact that the position of the grating "lines" in space varies during the transient process is the basic presumption in the holographic treatment of the effect, and mischange is reliably detected experimentally.

Nonstationary energy transfer has also been investigated in studies concerned with dynamic holography aimed at determining the conditions for the observation of energy transfer.[46,52] Figure 5 illustrates the energy transfer effect during the writing of the thermal grating in a stationary cell, i.e., when stationary energy transfer is forbidden. Subsequent studies have shown[47] that, when the donor beam intensity is high enough, a virtually 100% conversion to the acceptor beam is possible. The characteristic feature of the process is the considerable stretching of the transient interval in comparison with the hologram relaxation time, and the departure from regular kinetics (Fig. 9).

Nonstationary energy transfer is also observed during the writing of holograms (based on free carriers in silicon crystals) by the monopulse radiation from a neodymium-glass laser. Pure amplification was found to occur at power levels in excess of 1 MW/cm^2, and the calculated threshold power was found to be in agreement with observations.[53] Amplification was observed only when the intensities of the interacting beams were different, and the energy was always transferred from the strong to the weak beam. Measurement of the gain Γ as a function of the nonlinearity relaxation time τ showed that the effect was, in fact, nonstationary in

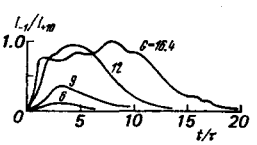

FIG. 9. Nonstationary self-diffraction in the case of thermal nonlinearity.[47] The calculated normalized weak-beam intensity I_{-1}/I_{+10} is plotted as a function of time. The parameter G is proportional to the initial intensity of the strong wave and the thickness of the material; $m=0.01$, $\alpha\tau=5$.

character, and was observed only when the length of the pump light pulse was $\Delta t \lesssim 10\tau$.

This type of phenomenon has also been observed during the writing of dynamic holograms in cadmium sulfide, based on the two-photon absorption of monopulse ruby laser radiation.[54]

Exceedingly strong nonstationary amplification occurs during the writing of holographic gratings in lithium niobate crystals placed in an external electric field. The drift writing mechanism, which relies on the redistribution of photoexcited carriers in the electric field, leads to the writing of the unshifted component of the holographic grating. Nonstationary energy transfer thus becomes possible.[50] The usual stationary diffusion amplification operates at the same time. Experiment shows that the gain associated with one effect is added to or subtracted from the other, depending on the orientation of the C-axis of the crystal and the beam intensity ratio. An oscillatory energy transfer process between the beams occurs when the external field exceeds 5 kV/cm, and practically the entire donor beam energy is transferred to the acceptor beam. As in most of the effects described above, the gain remains in the range 10–100 cm^{-1}.

Maximum nonstationary energy transfer (weak-beam intensity increases by a factor of 2500) occurs when the grating is written in lithium niobate crystals doped with iron,[55] owing to the photovoltaic effect. Figure 10 shows the relative increase in the weak-beam intensity during nonstationary energy transfer in $LiNbO_3$ doped with 0.05% by weight of iron.

5. THREE- AND FOUR-BEAM INTERACTION UNDER THE CONDITIONS OF SPATIAL SYNCHRONISM

It follows from the foregoing discussion that Bragg self-diffraction of two beams in the case of the unshifted spatial grating does not lead to energy transfer between the beams. At first sight, energy transfer in arrangements using the higher diffraction orders would seem to be impossible because of the difference between the geometric phase differences [see (2.1)], i.e., a departure from spatial synchronism with the Bragg beams. It is, however, possible to achieve energy transfer by using the phenomenon of phase transfer in dynamic gratings [see (2.5)]. Geometric dephasing of a complementary diffraction order can be compensated by changing the

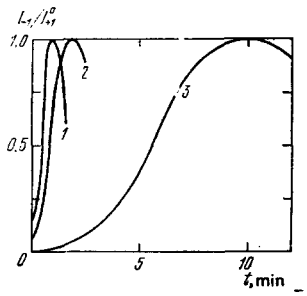

FIG. 10. Nonstationary self-diffraction during the interaction between light beams in lithium niobate crystals, showing the photovoltaic effect.[55] The initial beam intensity ratio is 1:10, 1:25, and 1:100 for curves 1–3, respectively.

weak-beam phase φ_{-1} by phase transfer. Since, in accordancw tih (2.5), φ_{-1} depends on the strong-beam intensity I_{+1}, this compensation is possible only for suitably chosen values of I_{+1}. This produces the interaction of all three beams under the conditions of spatial synchronism, and the corresponding energy transfer (Fig. 1c). Three-beam interaction of this kind was the first arrangement used for dynamic self-diffraction (see the Introduction).

We shall now follow Ref. 5 in considering the basic relationships for this process. Suppose that a medium with zero-inertia cubic nonlinearity $\varepsilon_2>0$ intercepts strong (E_0) and weak (E_1) waves. The resulting unshifted dynamic grating $\Delta\varepsilon$ [see (1.2)] with wave vector $q=k_0-k_1$ induces the polarization wave

$$\Delta P=\frac{\varepsilon_2}{8\pi}\left[|C_0|^2(E_0+2E_1)+\frac{1}{2}(|C_0|^2C_1^*e^{i(\mathbf{k}_0+\mathbf{q})\mathbf{r}-i\omega t})+\text{c.c.}\right] \quad (5.1)$$

with wave vector $\mathbf{k}_0+\mathbf{q}=2\mathbf{k}_0-\mathbf{k}_1$. The phase transfer is then equivalent to an increase in the refractive index for the weak wave, given by

$$\Delta n=\frac{\varepsilon_2|C_0|^2}{4n_0}. \quad (5.2)$$

Suppose that the weak wave E_2 with wave vector $\mathbf{k}_2=\mathbf{k}_0+\mathbf{q}=2\mathbf{k}_0-\mathbf{k}_1$ propagates through the medium at the same time, and that the wave vector corresponds to the first non-Bragg diffraction order. The spatial locking condition

$$2\mathbf{k}_0=\mathbf{k}_1+\mathbf{k}_2 \quad (5.3)$$

is then satisfied for the above three waves.

In accordance with (5.3), two pump-wave photons are scattered into two weak-wave photons during the four-photon interaction and are amplified equally.[6)]

The angle θ_{opt} between the beams for which (5.3) is valid is given by

$$\theta_{opt}=\pm\sqrt{\frac{2\Delta n}{n_0}}=\pm\sqrt{\frac{\varepsilon_2|C_0|^2}{2\varepsilon_0}}. \quad (5.4)$$

This situation occurs when the wave E_2 is absent at entry, since it appears during the self-diffraction process in a thin layer of the medium (see Sec. 6).

For arbitrary diffraction orders that are symmetric with respect to the strong wave, the weak wave gain is given by[57]

$$\Gamma=|l-1|k_0\sin\frac{\theta}{2}\sqrt{\frac{\Delta\varepsilon}{2}-\frac{(l-1)^2}{4}\sin^2\frac{\theta}{2}}. \quad (5.5)$$

It follows from this expression that $\Gamma>0$ for $0<\theta<\theta_{lim}$ where θ_{lim} is the angle for which the square root in (5.5) is equal to zero. At θ_{opt}, the gain reaches its maximum value of $\Gamma_m=\Delta\varepsilon k_0/2=\varepsilon_2|C_0|^2k_0/2$. For the diffraction orders nearest to the strong wave, $l=-1$; $+3$, and $\Gamma>0$ for

$$A=\frac{\varepsilon_2|C_0|^2}{8\varepsilon_0\sin^2(\theta/2)}>\frac{1}{4}. \quad (5.6)$$

Thus, in the given-field approximation, an exponential growth of the weak wave in the three-beam arrangement is possible in a medium with positive cubic nonlinearity. However, the strong-wave power threshold must be exceeded. This threshold is determined both by the medium parameters and by the angle between the beams.

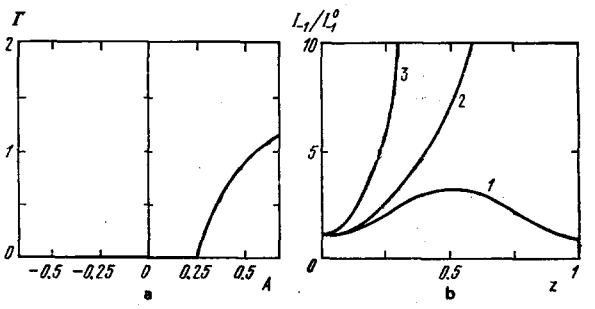

FIG. 11. Three-beam dynamic self-diffraction under the conditions of space matching: a) gain Γ for weak waves as a function of the parameter A; b) three types of dependence of weak-beam intensity on the thickness of the nonlinear layer[57] [$A<1/4$ (1), $A=1/4$ (2), and $A>1/4$ (3), respectively].

If the medium exhibits nonactive losses characterized by absorption length l_a, the strong-wave threshold power is

$$P_{th}=\frac{n_0c}{8\pi l_a}|C_0|^2. \quad (5.7)$$

Estimates performed for typical nonlinear media ($\varepsilon_2=7.5\times10^{-11}$ esu for the Kerr nonlinearity in CS_2) with $\lambda=6943$ Å and $l_a=10$ cm yield $P_{th}=9$ MW/cm^2.

There are also possible solutions with $\Gamma=0$, when the weak waves grow parabolically (Fig. 11, curve 2). When $\Gamma<0$, the weak-wave intensity oscillates with depth of propagation in the medium because of the violation of the space locking condition (this class of solutions was first examined in Ref. 8, where, however, it was erroneously concluded that it was the only class of solutions). Finally, a qualitative analysis has been carried out of the nonlinear problem of interaction between three waves without the given-field approximation,[57] and it has been shown that the transfer was always oscillatory in character. Conditions have also been found under which the weak wave has a complicated front and is amplified without appreciable distortion.

Three-beam self-diffraction was first carried out with monopulse ruby laser radiation of 900 ± 300 MW/cm^2. Two beams with an initial intensity ratio of 8:100 were allowed to cross in a cell containing nitrobenzene ($l=3$ mm). All other effects were either suppressed (stimulated Raman scattering) or did not succeed in developing (self-focusing, stimulated Mandel'shtam-Brillouin scattering). When $\theta_{opt}=8.3\pm0.8$ mrad, an additional beam of the same frequency (theoretical value $\theta_{opt}=7.2\pm1.3$ mrad) was observed at the angle θ_{opt} although the measured value of Γ turned out to be several times smaller than the calculated value. The theoretical prediction[20,5] of simultaneous and practically symmetric amplification of weak beams was confirmed for sufficiently long interaction regions ($l=75$ mm).

Three-beam self-diffraction of broad-bend dye laser radiation near the absorption lines of potassium and

[6)] If the medium is illuminated by two strong waves of equal intensity, self-diffraction will give rise to two additional symmetric diffraction orders corresponding to two coupled four-phonon interactions.[56]

rubidium was investigated in Ref. 56. In accordance with (5.6), the additional beams appeared only on the short-wave side of the absorption band, where $\varepsilon_2 > 0$, and the original weak beam, amounting to about 2% of the strong beam, was amplified by an order of magnitude. The appearance of symmetric weak beams for $I_{+10} : I_{-10} = 1 : 1$ was also noted.

It would appear that one of the most universal and promising arrangements for dynamic holography is that proposed in 1977 in Refs. 23 and 24, where a nonlinear medium with instantaneous local response was illuminated on opposite sides with two plane pump beams E_1 and E_2 propagating in opposite directions, and two signal beams E_3 and E_4 were present at a certain angle to the other two. The latter beams also propagated in opposite directions (Fig. 1d):

$$\mathbf{k}_1 + \mathbf{k}_2 = 0; \quad \mathbf{k}_3 + \mathbf{k}_4 = 0. \tag{5.8}$$

The spatial locking conditions are then satisfied automatically. Stationary energy transfer to the signal waves in a medium with local response thus becomes possible and, in contrast to the three-beam scheme, this can occur for an arbitrary angle between the pairs of beams, and any sign of the nonlinearity ε_2. One photon from each of the beams participates in the elementary event.

The standard procedure for solving the wave equation with nonlinear polarization [as in (1.4)] leads to the following equations for the signal waves in the given-field approximation:

$$\frac{\partial C_4}{\partial z} = i\varkappa^* C_3^*, \qquad \frac{\partial C_3^*}{\partial z} = i\varkappa C_4, \tag{5.9}$$

where $\varkappa = 2\pi\omega\chi C_1 C_2 / cn$ is the complex coupling constant between the waves.

For a clearer understanding of the four-beam interaction, we return to holographic language, i.e., to the consideration of the dynamic gratings produced as a result. The gratings that are written by the opposing beams C_1, C_2 and C_3, C_4 are excluded from consideration because Bragg diffraction of the second pair of beams cannot be produced by them and the opposing beams themselves do not exchange energy in accordance with Sec. 2. The self-diffraction process occurs as follows. Beams C_1 and C_3 write the transition grating, which can diffract the beam C_2 under the Bragg conditions, and generate wave C_4.[7)] Beams C_3 and C_4^* in turn write the grating that diffracts the pump wave C_1 and generates wave C_3. Reflecting gratings produced by beams C_2, C_3, and so on, can be described in a similar way.

We shall now solve (5.9) for the most interesting case of a single signal wave $C_3(0)$ [$C_4(L) = 0$]. We then have

$$C_4(0) = -i\frac{\varkappa^*}{|\varkappa|}\operatorname{tg}(|\varkappa|L)C_3^*(0), \qquad C_3(L) = \frac{C_3(0)}{\cos|\varkappa|L}. \tag{5.10}$$

If the gain Γ along the path L, determined by $|\varkappa|L$, is sufficiently large, the intensity of the resulting reflected wave $C_4(0)$ is greater than the incident-signal wave $C_3(0)$. When the condition $|\varkappa|L = \pi/2$ is satisfied, generation by waves C_3 and C_4 becomes possible without a cavity resonator although the introduction of a totally reflecting mirror perpendicular to C_3 reduces the generation threshold by a factor of two.[59] The results that we have obtained must be regarded as purely qualitative because the given-pump field approximation is violated for $|\varkappa|L = \pi/2$.

An important conclusion is that generation of the complex conjugate wave remains possible for signal waves with complicated fronts. In this case, (5.10) assumes the form

$$C_4(x, y, z<0) = -i\frac{\varkappa^*}{|\varkappa|}\operatorname{tg}(|\varkappa|L)C_3^* \quad (x, y, z<0), \tag{5.11}$$

i.e., the amplified reflected wave C_4 is the complex conjugate of the incident wave C_3 for $z < 0$, i.e., outside the nonlinear medium. This enables us to perform dynamic correction of wave fronts by the well-known holographic method. The possibility of beams with complex conjugate fronts in nonlinear-optics processes was first discovered and explained in the case of stimulated Mandel'shtam-Brillouin scattering.[61-62]

It has been shown[63] that the use of the four-wave interaction in beams traveling in opposite directions in fiberoptics systems containing a core with the necessary cubic nonlinearity results in the generation of an amplified complex-conjugate signal wave at moderate (1 W) pump levels and extended interaction lengths (10–100 m) with simultaneous automatic compensation of phase distortion due to the optical inhomogeneity of the fiber. Theoretical estimates have shown that other nonlinear effects do not develop at the same time. Fast media can be used to perform these operations in real time.

It is important to emphasize that the generated complex conjugate wave is a "time-reversed" modification of the direct signal wave C_3.[23] When the appropriate conditions are satisfied, e.g., the nonlinear medium is thin enough, it is possible to achieve the time compression of ultrashort phase-modulated pulses.[64]

The first experimental work with the four-beam arrangement, using beams traveling in opposite directions, was performed well before the proposals reported in[23,24] This arrangement was employed in Ref. 65 in an investigation of nonstationary holograms, using free carriers in silicon crystals, written by a neodymium-glass laser beam ($\lambda = 1.06\ \mu$, power in excess of 1 MW/cm^2). A wave that was the complex conjugate of the signal wave and was generated in the course of transmission through an image-bearing slide was observed.

At practically the same time, an analogous experi-

[7)] Wave C_4 is the complex conjugate of C_3 in accordance with the basic laws of holography[58-60] which are valid for dynamic gratings, since it can be reconstituted by the wave C_2 which is the complex conjugate of the reference wave C_1 (for plane waves, complex conjugation is equivalent to propagation in opposite directions). We note that our notation is different from that adopted in Refs. 24 and 63, where the signal wave is called C_4 and the resulting complex conjugate wave is called C_3.

ment was performed with a dynamic volume grating written by a ruby laser in a solution of a transparent dye.[66] Here again, it was shown that the complex conjugate wave was produced in the direction opposite to the signal wave, and was used to compensate the curvature of the wavefront produced by a positive lens. However, the stationary interaction of beams traveling in opposite directions was not considered in this work [condition (5.8)] and the experiments were carried out with nonstationary holograms.

The use of the complex conjugate wave for the correction of phase inhomogeneities in laser amplifiers was suggested in Ref. 45, where the complex conjugate wave was produced by reading a hologram with a beam having a plane wave front and produced by reflection of part of the reference beam in the reverse direction.

The four-wave interaction in liquid carbon disulfide between beams traveling in opposite directions and derived from neodymium and ruby lasers (10–100 MJ per pulse of 10^{-8} sec) was used in Refs. 67 and 68. Practically complete compensation of the nonuniformity in the reflected complex conjugate wave C_4 was achieved, and the properties of this wave were satisfactorily described by the theory for $|C_3|^2 < 0.1|C_1|^2$. The writing and reconstruction of the image of a binary transparency with a resolution of 40 lines/mm was reported in Ref. 67. The arrangement used to compensate optical distortions was found to be highly sensitive to any departure from strict coaxiality of the two pump waves C_1 and C_2.

The Kerr nonlinearity near the resonance doublet $D_1 - D_2$ of sodium[69] has been used to reach $n_2 \approx 10^{-8}$ esu and to reduce substantially the necessary pump power density (down to 40 kV/cm^2). At the same time, a reflected wave was produced that was stronger than the signal wave by a factor of 100. A tunable dye laser with $\Delta\nu_T = 0.03$ cm^{-1} was used to investigate the wavelength dependence of the intensity of the reflected wave (Fig. 12). A departure from the linear dependence of the intensity of the fourth wave on $|C_3(0)|^2$ was found to occur at high C_3 intensities (Fig. 13).

The scheme described above was realized by using saturated absorption in the Doppler line (sodium vapor at 2×10^{11} cm^{-3}) at $\lambda = 5890$ Å, pumped by a continuous dye laser of only 15 MW.[70] The contribution of the phase grating could be increased by departing from the line center at which the grating was of pure amplitude

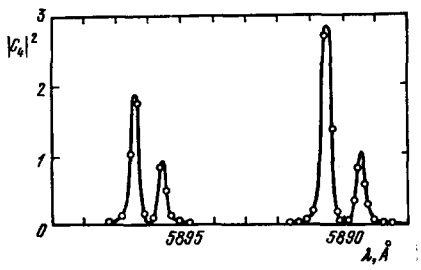

FIG. 12. Self-diffraction of four collinear beams propagating in opposite directions in sodium vapor.[69] The intensity of the fourth wave $|C_4|^2$ is shown as a function of the wavelength of the interacting beam.

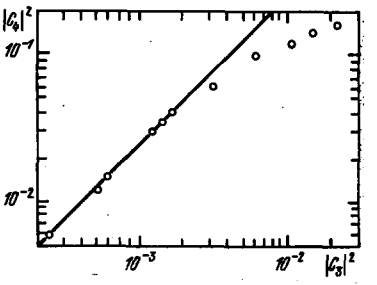

FIG. 13. Self-diffraction of collinear beams traveling in opposite directions in sodium vapor.[69] The figure shows the intensity of the fourth wave $|C_4|^2$, which is the complex conjugate of the signal wave C_3, as a function of the intensity of the latter.

type. Generation of the complex conjugate wave was observed above 30 mW/cm^2. The efficiency reached 0.2% at 600 mW/cm^2. A resolution of 12 lines/mm was achieved under these conditions in the complex conjugate wave in the real image of a test object placed in the path of the signal wave. The wavelength range in which the effect was observed, especially at low pumping levels, was much narrower than the Doppler linewidth. This opens up new possibilities for laser spectroscopy within inhomogeneously-broadened lines.

Four-wave interaction of beams from the CO_2 laser ($\lambda = 10.6$ μ) in germanium was investigated in Ref. 71. The generation of the complex conjugate wave and compensation of phase inhomogeneities was demonstrated.

The generation of the complex conjugate wave in a barium titanate system was reported in Ref. 72.

6. SELF-DIFFRACTION WITH THE PARTICIPATION OF HIGHER DIFFRACTION ORDERS

The characteristic manifestation of self-diffraction is the appearance of several higher-order diffraction beams propagating at angles satisfying the vector relation

$$\mathbf{k} = \mathbf{k}_0 \pm i\mathbf{q}, \tag{6.1}$$

where $\mathbf{q}$ is the grating vector (Fig. 1e) and $i = 1,2,3,\ldots$.

This is analogous to Raman-Nath diffraction by given periodic structures[73] and occurs when the higher diffraction orders are not interference-quenched within the nonlinear layer for one reason or another. Many researchers have used the appearance of higher diffraction orders to investigate the nonlinearity mechanism.[74,75]

Self-diffraction, including the appearance of the higher orders, is usually discussed on the basis of a simplified scheme without taking into account the reaction of the change in the refractive index on the writing light beams (this is the nonstationary phase transparency approximation[76–78]). The calculated characteristics of diffraction by a thin thermal grating[76,77] show that the intensity of the higher diffraction orders is described by a sum of the form

$$I_{\pm m} = TI_0(J_m^2 + J_{m+1}^2), \tag{6.2}$$

where J_m and J_{m+1} are Bessel functions representing

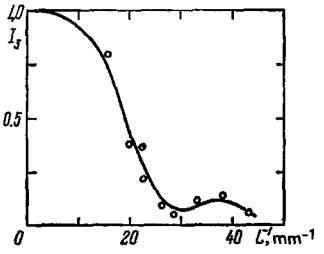

FIG. 14. Dynamic self-diffraction with the participation of higher diffraction orders.[87] The figure shows the intensity of the first non-Bragg diffraction order as a function of the spatial frequency of the interference field for the cadmium telluride crystal. Solid line—calculated,[86] open circles—experimental.

diffraction in a given direction from each of the writing beams, and T is the layer transmission.

Experimental studies of self-diffraction under the Raman-Nath conditions have been carried out for solutions of organic dyes[28,30,66] and semiconductors.[27,29,32,33,54,74,81-84] It was shown that, when the diffraction efficiency was low, reasonable agreement could be a achieved with different variants of the nondynamic theory.[76-78,84,85,88] This can be understood by recalling that the absolute change in the phase difference between the interacting beams becomes negligible when the overall phase change across the nonlinear layer is small.

Improved calculations referring to particular experimental situations appeared after this work. For example, nonstationary self-diffraction by thermal gratings was examined in Ref. 85 and by free-carrier gratings in Ref. 74. Raman-Nath self-diffraction was analyzed in Ref. 78 with allowance for nonlinear absorption.

The correct description of self-diffraction with allowance for dynamic feedback is difficult because of the increased number of equations. The first non-Bragg intensities $I_{\pm 3}$ have been calculated[49,86] for self-diffraction in semiconducting crystals produced as a result of the production of pairs of free carriers. Comparison with experiments performed with CdTe crystals in which stationary dynamic holograms were written by monopulse Nd^{3+}:YAG radiation showed that there was good agreement with theory[87] (Fig. 14).

7. APPLICATIONS OF THE SELF-DIFFRACTION EFFECT

The phenomenon of self-diffraction has found extensive applications in both physics and technology.

Image writing, reading, and transformation in real time were demonstrated in the very first paper on dynamic holography.[25] Proposals for logic elements based on dynamic holograms were subsequently investigated.[89] The same idea formed the foundation for a method of measuring the duration of ultrashort light pulses writing the grating. This was suggested and carried out in Ref. 91. The characteristics of self-diffraction or test-beam diffraction by a dynamic hologram can be used to determine the probabilities of different relaxation processes leading to the erasure of the grating (see the review paper given in Ref. 74). Here, we have the possibility of measuring the temperature diffusivity in liquids and solids,[92,93] the mobilities of free carriers, and the probabilities of recombination processes in semiconducting compounds, the depth of impurity centers participating in the writing and erasure processes, and the investigation of new mechanisms of nonlinearity. For example, the writing of dynamic gratings in record times with the aid of intraband absorption by the carriers was reported in [81,82].

Modern technology can be used to measure diffracted radiation of 10^{-5} of the intensity of the incident light, which corresponds to the modulation of the optical path difference by amounts of the order of $10^{-3}\lambda$, i.e., the situation has been pushed practically to the limit of the optical band. This high sensitivity of the method means that exceedingly weak effects, such as second sound propagation in crystals, can be investigated.[95,96] The coefficients of diffusion of excited molecules of a dye in liquid crystals[97] and the anisotropy of thermal conductivity in liquid crystals[98] have also been investigated.

Self-diffraction can also be used to determine the components of the nonlinear polarizability tensor, which is responsible for self-diffraction,[58] and to investigate internal inhomogeneities in a medium that give rise to the inhomogeneity of the nonlinear polarizability tensor.[68]

Four-wave paired collinear interaction can be used to achieve one of the variants of two-photon laser spectroscopy within the Doppler-broadened line with the elimination of background.[68,70]

On the other hand, dynamic holography has opened up new possibilities for real-time image processing. Self-diffraction in self-translucid liquids and gases has been suggested[99,101] as a means of correlational comparison between two continuously-varying specimens. Two beams of light passing through variable transparencies are used to write the Fourier hologram in the focus of a lens and the third, specially shaped beam of the same frequency is used to read and reconstruct the mutual correlation function for the two specimens in one of the diffraction orders.

Amplification of coherent light beams, including amplification of beams carrying optical information, is a possible application. The dynamic nature of the process is such that it can be used to amplify time-dependent signals.

Image enhancement during the writing of stationary shifted holograms in lithium niobate crystals[41] has been reported as well as nonstationary energy transfer in lithium niobate crystals in an external field[50] and in different four-wave arrangements for paired collinear interaction.[69,70] Figure 15 shows the image of a television testcard enhanced by a factor of ten during the writing of a hologram in a nominally pure lithium niobate crystal (stationary amplification).

Another possible application of dynamic self-diffraction is real-time holographic interferometry.[102,103]

Several theoretical treatments[40,104-108] have been re-

FIG. 15. Enhancement of the testcard image on a television screen as a result of dynamic self-diffraction by a stationary shifted grating written in a nominally pure lithium niobate crystal.

ported of possible applications of the energy transfer effect in determinations of the parameters of electro-optic crystals. The use of the energy transfer effect in the visualization of phase inhomogeneities in a nonlinear medium and in the correction of amplitude-inhomogeneous light beams has been discussed.[108]

In the case of stationary self-diffraction,[39] the direction of energy transfer depends on the sign of the mobile charge carrier.[39] It has recently been shown[109] that, when lithium niobate crystals are excited in the ultraviolet band, the main mobile carriers are holes and not electrons as in the case of excitation in the visible band.

Writing in dynamically nonlinear media can also be used to solve the problem of the transformation of complicated wavefronts to a given form and, in particular, to correct the angular divergence of real lasers.[8,45,60,110] Calculations have shown that, when the writing conditions are correctly chosen, considerable enhancement can be achieved for the acceptor beam with nearly plane wavefront without appreciable distortion.[111,112]

On the other hand, the generation of complex conjugate wavefronts in Raman-Nath self-diffraction and in the four-wave interaction[68] can be used as a means of compensating dynamic phase inhomogeneities of powerful laser amplifying sections[45] and fiberoptic schemes (this is a variant of adaptive optical systems with amplification).

Like many other nonlinear effects, self-diffraction can be used to control the duration of the diffracted beam. Compression resulting from the four-wave interaction, which depends on the pump-wave intensity, was reported in Ref. 67. However, the compression of ultrashort light pulses by "time inversion" of the beam with the complex conjugate wavefront is much more interesting.[64]

APPENDIX

The basic set of equations used to describe self-diffraction of light waves in a nonlinear medium includes the Maxwell equations

$$\nabla\times\mathbf{E}=-\frac{1}{c}\frac{\partial\mathbf{H}}{\partial t},\quad \nabla\times\mathbf{H}=\frac{1}{c}\frac{\partial\mathbf{D}}{\partial t}+\frac{4\pi}{c}\mathbf{j},\qquad \mathbf{D}=(\varepsilon+\Delta\varepsilon)\mathbf{E},\quad \mathbf{J}=\sigma\mathbf{E}, \tag{A.1}$$

where ε is the permittivity averaged over the volume, and $\Delta\varepsilon$ is the light-induced nonlinear increment in the permittivity. We shall confine our attention to two waves with polarization perpendicular to the plane of incidence xz [i.e., $E=(0,E,0)$], incident symmetrically on the crystal at an angle of 2θ to each other (see Fig. 1). We suppose that the medium is infinite in the x and y directions, so that we can put $\partial/\partial y=0$ in (A.1) and seek the solution in the form of the Fourier series

$$E_y(x,z,t)=\sum C_n(z,t)\,e^{i(\omega t-k_z z+nk_x x)},\qquad \Delta\varepsilon(x,z,t)=\sum \varepsilon_p(z,t)\,e^{ipk_x x}. \tag{A.2}$$

Substitution of (A.2) and (A.1) yields (for $k_z=k_0\sqrt{\varepsilon}\cos\theta$, $k_x=k_0\sqrt{\varepsilon}\sin\theta$):

$$\frac{1}{2k_z c^2}\frac{\partial^2 C_l}{\partial t^2}-\frac{1}{2k_z}\frac{\partial^2 C_l}{\partial z^2}+\frac{\omega}{c^2 k_z}\frac{\partial C_l}{\partial t}+\frac{\partial C_l}{\partial z}+\frac{i(1-l^2)k_x^2}{2k_z}C_l+\frac{2\pi\omega\sigma}{k_z c^2}C_l=-\frac{ik_0^2}{2k_z}\sum\varepsilon_p C_{l-p}. \tag{A.3}$$

Henceforth, we shall consider smooth variations of C_l with z and t, which is valid provided

$$\frac{\partial^2 C_l}{\partial t^2}\ll 2\omega\frac{\partial C_l}{\partial t},\quad \frac{\partial^2 C_l}{\partial z^2}\ll\frac{1}{2k_z}\frac{\partial C_l}{\partial z}.$$

These conditions correspond to characteristic time (τ) and space (l) scales of variation of C_l that substantially exceed the period $T=2\pi/\omega$ and the wavelength λ ($\tau\gg T$; $\lambda\ll 4\pi l\sqrt{\varepsilon}\cos\theta$).

Simultaneously with the Maxwell equations, we must consider the equation for the light-induced nonlinearity $\Delta\varepsilon$. This can be written in the following form that describes a sufficiently broad range of nonlinear media:

$$\frac{\partial}{\partial t}(\Delta\varepsilon)=D\frac{\partial^2}{\partial t^2}(\Delta\varepsilon)-v\frac{\partial}{\partial x}(\Delta\varepsilon)-\frac{\Delta\varepsilon}{\tau_0}+\beta I+\gamma\hat{P}\left(\frac{\partial I}{\partial x},\int I\,dx\right), \tag{A.4}$$

where the first two terms on the right-hand side describe the drift (v is the drift velocity) and the diffusion (D is the diffusion coefficient) components of the pump flux along the $0x$ axis, which are responsible for the variation in, for example, the number of current carriers or excitons in semiconductors and ferroelectrics, or the fluxes of heat and liquids, where $\Delta\varepsilon$ is proportional to the temperature, and so on. The third term in (A.4) describes the relaxation of excitations, and the last two the local and nonlocal response of the medium to the incident ration. In general, the nonlocal term can also be nonlinear; I is the intensity of light given by $I=\sum_{m,n}C_m C_n^* e^{i(m-n)k_x x}$.

Using the expansion (A.2) for the Fourier components of C_l and ε_p, we obtain

$$\frac{\partial C_l}{\partial z}=iQ_l C_l-\frac{\alpha C_l}{2\cos\theta}-\frac{ik_0^2}{2k_z}\sum\varepsilon_p C_{l-p}, \tag{A.5}$$

$$\frac{\partial\varepsilon_p}{\partial t}=a_p\sum_{m-l=p}C_m C_l^*-\frac{\varepsilon_p}{\tau_p}, \tag{A.6}$$

where $Q_l=(k_x^2/2k_z)(l^2-1)$, $\alpha=4\pi\sigma/c\sqrt{\varepsilon}$ is the absorption coefficient, and $\tau_p^{-1}=\tau_0^{-1}+Dk_x^2p^2+ipvk_x$; the function $a_p(\beta,\gamma,p)$ is determined by the particular mechanism responsible for the light-induced variation of ε. Equations (A.5) and (A.6) are the basic equations for self-diffraction in media with nonlinearity given by (A.4). The first term on the right-hand side of (A.5) describes the change in the phase of the l-th beam component due to its deviation from the Bragg direction. The second term describes absorption in the medium,

and the last term is the contribution of diffraction to the l-th order of different beams. Transforming to real variables $\varepsilon_p = |\varepsilon_p| e^{i\Phi_p}$; $C_l = \sqrt{I}\, e^{i\varphi_l}$, where $|\varepsilon_p|$, Φ_p are the amplitude and phase of the p-th Fourier component of the holographic grating and I_l, φ_l determine the intensity and phase of the l-th beam of light, we obtain, instead of (A.5), the following expressions for the case of weak absorption:

$$\frac{\partial I_l}{\partial z} = -\alpha I_l + \frac{k_0^2}{k_z} \sum_{p+m=l} |\varepsilon_p| \sqrt{I_m I_l} \sin(\Phi_p + \varphi_m - \varphi_l),$$
$$\frac{\partial \varphi_l}{\partial z} = Q_l - \frac{k_0^2}{2k_z \sqrt{I_l}} \sum_{p+m=l} |\varepsilon_p| \cos(\Phi_p + \varphi_m - \varphi_l). \qquad \text{(A.7)}$$

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