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NEW DEVELOPMENTS IN NON-LINEAR OPTICS

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1. INTRODUCTION

THE 3rd All-union Symposium on Non-linear Optics was held in Erevan at the end of October, 1967. It was organized by the Section of General and Applied Physics of the USSR Academy of Sciences, Moscow State University, Erevan State University, and the Academy of Sciences of the Armenian SSR. Representatives of practically all the Soviet and foreign research groups actively working in the field of non-linear optics participated in the symposium.

A simple comparison of the proceedings of the Erevan symposium with those of the First Symposium (held in Minsk in the summer of 1965, with the proceedings published in[1]) and the Second Symposium (held in Novosibirsk in the summer of 1966, with the proceedings published in[2]) indicates without doubt a considerable expansion in the volume of studies. The 230 Soviet and 40 foreign participants presented about 200 papers at Erevan in the different branches of experimental and theoretical non-linear optics.

Thus quantitative growth is at hand. However, what can we say on the qualitative side? This question was rather actively discussed in the corridors at the symposium. The diagram shown in Fig. 1 gives a certain picture of the viewpoint of the participants on this topic. The graph characterizes the qualitative progress (the tempo of accession of ideas, the development of new methods, etc.) of non-linear optics as a function of time. The origin of the curve is clearly dated at 1961 (the first experiments on frequency-doubling of light). The participants placed points to mark the position of the year 1967 on this curve.

While the conditions of inquiry apparently did not satisfy the necessary requirements usually imposed on studies of this type, and the curve in Fig. 1 cannot, strictly speaking, be acknowledged even as a quite reliable experimental fact, we think that the position of the optimists, who placed the points on the rising branch of the curve, is justified.

Before proceeding to argue this assertion in essence, we shall give some more data on the number of articles on non-linear optics published in the last two years in such journals as Pis'ma ZhÉTF, Physical Review Letters, Physics Letters, and Applied Physics Letters. As we know, these journals are characterized by high standards on novelty of ideas in the published material.

Assuming that the editors of all the cited journals are carrying out the programs printed on their covers, the cited figures undoubtedly indicate an accelerated accession of new ideas in non-linear optics. However, without restricting ourselves to the statistical data, we shall attempt below, on the basis of the proceedings of the Erevan symposium and the articles published in 1967, to characterize briefly and in essence the new

June-July	Pis'ma ZhETF	Phys. Rev. Letters	Phys. Lett.	Appl. Phys. Letters
1965–66	25	23	12	25
1966–67	25	31	14	40

ideas that have entered into non-linear optics in the last year.

We can conveniently classify the theoretical and experimental studies on non-linear optics into four groups (this division has already become traditional in classifying papers at symposia on non-linear optics):

a) Non-linear properties of material media.

b) Interaction and self-action of light waves.

c) Stimulated scattering.

d) Application of methods of non-linear optics in laser technology.

We shall discuss these four divisions separately.

2. NON-LINEAR PROPERTIES OF MATERIAL MEDIA

One of the most significant advances of the past year is the study of non-linear properties of semiconductors in the far infrared. The first step here was taken by Patel,[3] who found a new mechanism of non-linearity. He found (see also[4]) that the fourth-order non-linear susceptibility χ_{ijkl} at long radiation wavelength is determined by intraband energy transitions. This contrasts with the visible and near infrared ranges, where the non-linear properties of semiconductors are determined by interband quantum transitions.

We recall that the susceptibility tensors of different orders are formed from the coefficients in the expansion

$$P_i = \chi_{ij}E_j + \chi_{ijk}E_jE_k + \chi_{ijkl}E_jE_kE_l + \ldots, \qquad (1)$$

where P_i is the i-th component of the wave of polarization per unit volume, and E_i is a component of the light field.

In the classical approach, we can describe the non-linear susceptibility involved in intraband transitions as follows.[5] The Hamiltonian of a current carrier in a band has the form

$$H = \frac{p^2}{2m^+} - \frac{p^4}{4E_G m^{+2}}, \qquad (2)$$

where m^+ is the effective mass, and E_G is the width of the conduction band. The second term in (2), which determines the non-linear susceptibility, describes the anharmonicity of the band. The velocity, which equals $\partial H/\partial p$, has the form

$$v = \frac{\partial H}{\partial p} = \frac{p}{m^+} - \frac{1}{E_G m^{+2}} p^3. \qquad (3)$$

The polarization per unit volume is related to the

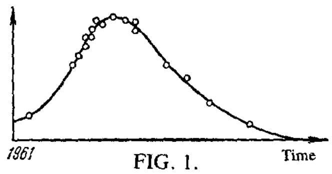


FIG. 1.

velocity as follows:

$$\dot{P} = e \int f(\mathscr{E}) v \, d\mathscr{E}, \tag{4}$$

where $f(\mathscr{E})$ is the energy distribution function of the carriers. The momentum p of the carriers entering into Eq. (3) is determined from the equation of motion

$$\dot{p} = eEe^{i\omega t}, \tag{5}$$

where E is the amplitude of the field and ω is its frequency. If we substitute (5) into (3), and (3) into (4), we can easily obtain the term that is cubic in the field in the expression for the polarization. This term involves four-photon processes, including the self-action effect, since the cubic term in the polarization corresponds to the quadratic term in the refractive index. In this mechanism, the non-linearity is determined by the concentration of carriers, as is not the case in the mechanism involving interband transitions.

However, the non-linearity mechanism set forth in[3-5] is dominant only at long enough wavelengths. If the radiation frequency approaches the frequency of the interband transitions, the contribution of the latter becomes more and more perceptible. In particular, as was shown in[6], combination transitions (interband and intraband) begin to play a large role. It would seem that experimental study of these problems is a matter for the immediate future.

It would seem that not only the cubic non-linearity is determined in the far infrared by intraband transitions. So is the quadratic non-linearity, which is responsible for second-harmonic generation and three-photon parametric effects. This involves the fact that the energy is not an even function of the momentum in the bands of semiconductors, owing to spin-orbit interactions, and there is a cubic term in the expression for H(p):

$$H(p) = \frac{p^2}{2m} + \gamma p^3 - \frac{p^4}{4E_g m^{+2}}, \tag{6}$$

Here the coefficient γ characterizes the spin-orbit interaction. However, in line with Patel's[7] experiments, no relation is observed between the amplitude of the second harmonic and the concentration of carriers. The contradiction here will apparently be resolved in further studies.

Studies on non-linear magnetooptics in the infrared have some bearing on the studies on non-linear properties of semiconductors. In the former, non-linear properties involving anharmonicity acquire a number of specific features, owing to the cyclotron motion of the carriers in the magnetic field. In particular, the cyclotron motions of the carriers give rise to Raman scattering.[8] When the light frequency coincides with one of the harmonics of the cyclotron frequency, a resonance peak in the non-linear susceptibility occurs, etc.[9]

Faĭn and his associates[69] have carried out some important studies on non-linear properties of crystal structures and ferromagnetic materials in the infrared. They estimated the contributions to the non-linear polarizability from optical and acoustic phonons, magnons, processes involving two quasiparticles, etc.

While noting the role of free carriers in determining the non-linear properties of semiconductors, we must not fail to mention cooperative effects involving interaction of carriers with one another. Thus, an experimental study has recently been made of spontaneous Raman scattering by the plasma vibrations of the carriers.[8,10] We must expect that very soon the stimulated Raman scattering, both by the cyclotron motions of carriers in a magnetic field and by plasma vibrations, will be achieved experimentally, In particular, the powerful dysprosium laser built by Prokhorov and his associates, having an emission wavelength $\lambda = 2.36\ \mu$, should be suitable for this. The results of a study of non-linear effects in Ge using this laser were reported at the Erevan symposium.[11]

Feofilov[14] has developed some original studies of cooperative phenomena due to interaction of two or more excited real states. These phenomena constitute a new and interesting branch of non-linear optics.

Bonch-Bruevich, Khodovoĭ, and their associates have made a detailed investigation of the Stark effect in strong light fields; the results of these studies were summarized in a review[53] last year.

An important line of study in this field of non-linear optics is the search for new-nonlinear materials. Appreciable progress has been made here in the past year. In particular, a number of new crystals having quadratic non-linearity have been found, and they show promise for use in non-linear optics. As we know, necessary requirements for such crystals are a large non-linear susceptibility and the existence of a direction of phase-matched interactions. These requirements are fulfilled by crystals of proustite.[12] They have a non-linear polarizability coefficient several tens of times larger than that of the crystal KDP, they possess matching directions, and they have a transmission band from 0.6 to 13 μ, which is a record high for non-linear crystals. We should note that in principle one could use proustite to build lag-free and low-noise radiation transformers from the far infrared (e.g., the 10-μ range) to the visible. This would be of great importance for infrared technology.

Another new crystal is ammonium oxalate.[13] Resembling the KDP crystal in its non-linear properties and transparency, it is biaxial, a fact that opens up new possibilities in non-linear optics. In particular, it can show simultaneous phase-matching of waves of frequencies ω, 2ω, and 3ω in the visible. The latter can be used for effective frequency-tripling and for building parametric light generators with low-frequency pumping. We should note also an entire set of new crystals of the KDP group and of the niobates, which have grown and studied recently, and which have definite advantages (see[55,65]).

Finally, the list of crystals that can be used for transforming the CO_2-laser frequency has also been extended. Besides the already well-known tellurium, selenium, and their derivatives, the crystals HgS (cinnabar) and CuCl studied in[15] are worthy of note.

Thus, successful work in searching for new non-linear materials is considerably expanding the region of practical use of the methods of non-linear optics.

3. INTERACTION AND "SELF-ACTION" OF LIGHT WAVES

One usually includes in this branch of non-linear optics such effects as the interaction of light waves in material media that takes place without participation of the internal motions of the medium (harmonic generation, mixing, parametric amplification, and laser action; these effects involve waves having widely differing frequencies), and "self-action," or effects involving variations in the angular and frequency spectra of quasimonochromatic waves in a non-linear medium (self-focusing and self-defocusing, self-contraction and self-expansion of wave packets).

The problems comprised in the first group have already become somewhat traditional in non-linear optics. Furthermore, the set of problems can be considered to be exhausted, at least in principle. Nevertheless, a number of new physical results have been obtained also in this field in the past year.

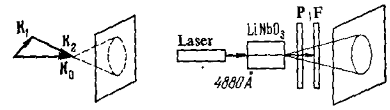
We are thinking primarily of the study of statistical effects in the interaction of light waves in transparent media, and in particular, of the direct detection of quantum fluctuations in a transparent non-linear medium. Non-linearity causes the quantum fluctuations to give rise to spontaneous decay of photons passing through the given medium. Experimentally, the effect is manifested as a distinctive luminescence of an optically transparent medium, in which a quantum of radiation of frequency ω_0 decays on being scattered into two quanta of frequencies ω_1 and ω_2 satisfying the relation $\omega_1 + \omega_2 = \omega_0$.*

Naturally, the emission is greatest in directions for which the law of conservation of momentum is satisfied:

$$\mathbf{k}_1 + \mathbf{k}_2 = \mathbf{k}_0. \qquad (7)$$

This effect is also called in the literature parametric luminescence (one can easily see that it is a spontaneous analog of the process of parametric amplification). It was found experimentally almost simultaneously (within a month) under differing conditions by three different groups: by Harris and his associates[16] (Stanford University, USA), by Mahr and Magde[17] (Cornell University, USA), and by us at the Moscow University.[18,19] It is essential to point out that the process of parametric luminescence occurs at intensity levels of the decaying photons far below the threshold of self-excitation of parametric oscillations. Hence the effect can be observed even in gas-laser beams. Figure 2 shows the basic diagram of an experiment to observe parametric luminescence. A beam of laser radiation impinges on a non-linear crystal in which phase-matching can occur. Spontaneous radiation at the frequencies ω_1 and ω_2 arises in the crystal. The angles at which radiation at the given frequencies is scattered are determined by the condition (7) and by the dispersion properties of the crystal. Figure 3 shows a

FIG. 2. Diagram of an experiment to observe parametric luminescence.



photograph of the luminescence in lithium niobate excited by an argon gas laser. The photograph shows a set of colored rings; the width of the rings and the spectral intensity distribution are determined by the details of the dispersion characteristics of the non-linear crystal (the radiation at the supplementary frequency lies in the infrared; in the experiments of Klyshko and Krindach[19] luminescence in $LiNbO_3$ was observed as far as $\lambda_2 \cong 5\ \mu$).

The phenomenon of parametric luminescence is of interest as a method of studying the non-linear susceptibility of a transparent medium by using sources that are not very intense. Practical interest in the effect involves the fact that the spontaneous decay of photons determines the level of intrinsic noise in parametric amplifiers and light generators.

Interest in studying statistical effects in non-linear interacting light waves is also stimulated by the results of successful experiments on harmonic generation and optical mixing using non-laser sources. It is thus a question on the theoretical level of generalizing the theory of non-linear interactions to random fields. Not only the classical analysis is of interest here (this theory has fundamentally been worked out by now), but also the quantum treatment (among the new studies along this line, we note[20]). The latter is especially interesting, since the methods of non-linear optics can provide new information on the coherent properties of light fields (see[21]), which have been studied so intensively in recent times. Finally, it is of undoubted theoretical interest to set up experiments on non-

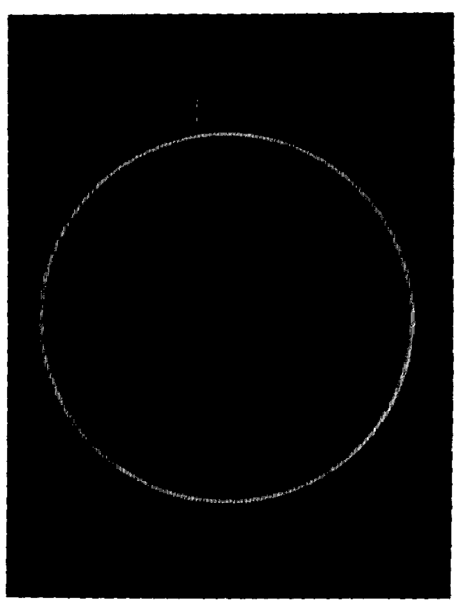


FIG. 3. Angular structure of the parametric luminescence in an $LiNbO_3$ crystal excited by an argon laser. Only the radiation in the visible is recorded; the supplementary frequencies lie in the infrared. (Photograph by Krindach, Moscow State University).

*Of course, one can have analogous effects of higher orders, in particular a decay of the type $2\omega_0 = \omega_1 + \omega_2$; they prove to be much weaker.

linear optics at low radiation intensities, in which one might detect such quantum characteristics of the fields as the photon statistics.

The search for new methods for matching phase velocities is continually urgent in non-linear optics. Until recently, one could compensate for the normal dispersion only in optically anisotropic media posessing a considerable birefringence. A new method of compensation is discussed in[22-24]: using circular birefringence in optically-active media (the idea of compensation itself is the same as in an anisotropic medium, but one uses different normal waves). From the practical standpoint, as was shown in[22,23], the problem amounts to finding media having a high enough constant optical rotation. Liquid crystals might be of some interest from this standpoint. However, their non-linearity is apparently very small. An important feature of this method of matching is the fact that one is dealing here with phase-velocity matching in an isotropic medium. In principal, in such media one can eliminate aperture effects, which restrict the possibilities of obtaining cumulative interactions in cases in which velocity matching is attained through anisotropy. Thus, if we could successfully realize phase-matching through circular birefringence, we could expect to attain coherence lengths of non-linear interaction of the order of the free path $L\delta$ of a photon, i.e., $\sim 10^2-10^3$ cm, in a transparent medium*($\delta \cong 10^{-2}-10^{-3}$ cm^{-1}). We recall that coherence lengths of ~10 cm are considered large in current non-linear optics. We should note besides that study of non-linear effects in optically-active media is also of undoubted independent interest. Using powerful sources of visible and ultraviolet radiation, as was shown[23,58,59], one can observe such new effects as non-linear rotation of the plane of polarization, non-linear circular dichroism, etc.

Very recently, non-linear optics has had at its disposal new sources of powerful radiation: generators of picosecond ($\tau_p \cong 10^{-11}-10^{-12}$ sec) light pulses. Such sources can be built either from lasers with synchronized modes, or by using the stimulated Raman scattering phenomenon at an angle of 180°.

Use of these generators permits one to proceed to a systematic experimental study of non-stationary non-linear effects. It suffices to say that one should observe considerable non-quasistatistical behavior at $\tau_p \cong 10^{-12}$, as a rule, even in such a "fast" effect as harmonic generation.[73] It is of considerable interest to study the passage of powerful picosecond pulses through a resonant medium when $\tau_p \ll T_{1,2}$ (the relaxation times).[70-72,74]

Study of Kerr self-focusing of picosecond pulses might help in revealing the self-action mechanisms. When the pulse duration τ_p is shorter than the relaxation time τ, the self-focusing length increases, characteristic temporal aberrations appear, etc.

This problem has been treated theoretically in[57], and an interesting experiment has been described very recently by Giordmaine and his associates.[60]

*The possibility of matching phase velocities in an isotropic medium by introducing a real anomalous dispersion (absorbing centers) has been recently demonstrated in [25]. Unfortunately, the value of L_δ necessarily proves to be rather small here.

The fact is also fundamental that work with picosecond pulses can make it possible to study the behavior of matter in extremely strong light fields.[75,73] The threshold for breakdown of the medium by electronic avalanches must increase sharply in going to picosecond pulses.[76]

Finally we note that continually more attention is being paid to higher-order effects (optical-harmonic generation in gases, four-photon parametric amplification and generation, etc.).

Hardly the least interesting field of non-linear optics in the last two years has been the study of self-action of light waves. The abundance of experimental and theoretical studies in this field is explained not only by the natural interest in a new physical effect; it also concerns the fact that it is precisely the self-actions that apparently determine the main features of the behavior of powerful light beams in material media. Here we should emphasize that, while even a year ago the basic attention among the self-action effects was being paid to spatial self-focusing, it has been shown very recently that self-defocusing of wave beams and self-contraction of wave packets play a significant role in many important situations.

In the field of study of self-focusing, we must mention first of all the experimental studies concerned with the dynamics of the process. Experimental studies[26,27] have shown that the self-focusing process occurs as follows. At first the beam contracts, and "thick" filaments of diameters of the order of 50 μ are formed. Then each of these filaments breaks up in turn into "thin" filaments of several microns diameter (Fig. 4). There is no reproducibility of the spatial distribution of filaments from flash to flash. In the discussion at the Erevan symposium, Chiao (University of California, Berkeley, USA) spoke of new experiments showing that the duration of the light pulse in each of the thin filaments amounts to 10^{-10} sec or even less. The reasons for the appearance of short-lived high-field regions ("short-lived filaments") may be highly varied, and are thus far unclear.

Perhaps it involves saturation of the non-linearity and the finite relaxation time of the non-linear susceptibility (and consequently, a finite rate of growth of the optical waveguide), and the strong forward scattering of the radiation (e.g., arising from Mandel'shtam-Brillouin scattering).

In the opinion of Prokhorov and his associates, who

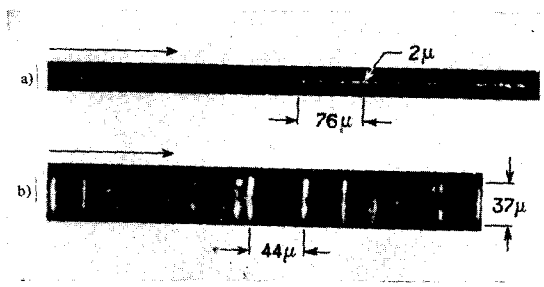


FIG. 4. The "thin filaments" (lateral view) into which a ruby-laser beam has broken down in carbon disulfide (according to Brewer and Townes [68]). The discrete luminescence involves excitation of standing waves in the filaments. a) A thin filament of diameter 2μ; b) a thick filament of diameter 37μ.

presented an interesting paper at the Erevan symposium on the theory of aberrational self-focusing of Gaussian beams,[28] the thin filaments are foci of radiation near the axis that shift in time.

The authors of[28] explain the results of the experiment described in[27] with their theoretical estimates.

In addition, some authors have adduced new physical mechanisms to explain the structure of a self-focusing beam. In a paper given at Erevan, Grob and Wagner (Stuttgart, West Germany) discussed the contribution of striction effects and, in particular, the non-local character of the non-linear echo, which involves spatial dispersion of sound. In a recent study, Shen and his associates[56] proposed a model of phase transitions in the strong light field to explain the fine filaments. It is also possible that the fine temporal structure of the laser radiation itself plays a substantial role in the behavior of a self-focusing beam, as Basov noted in the symposium.

The theory of the self-focusing of the thick filaments is in a relatively satisfactory state: it explains quantitatively at least the first stage in the process of filament formation, in which the beam begins to constrict to a focus owing to the non-linearity of the medium. We note that theory and experiment are in good quantitative agreement on this point, not only for Gaussian beams, but also for substantially inhomogeneous beams.[32] However, as for explaining the formation of thin filaments, only the first steps have been taken here. The advantage in the final solution of this problem, as well as many other obscure problems involving self-focusing,* must lie with detailed experiment. Apparently, under actual conditions the problem is a matter of a complex interaction of many factors (saturation and relaxation of non-linearity, stimulated scattering, many-photon absorption, etc.) whose interrelations are not obvious a priori.

We should mention at least two of the theoretical problems having independent importance: 1) The limits of applicability of the quasioptical approach to studying the phenomena near the focus and in the hyperthin filaments are not fully clear at present. One can elucidate this problem only with a more general theory of self-focusing than that using the parabolic equation. 2) There is a need for further development of the theory of self-focusing of complex beams, beams modulated simultaneously in space and time. Apparently it is important to take into account simultaneously both the self-action involving change of the angular spectrum and the self-action involving change of the frequency spectrum. Perhaps the mutual influence of these effects will explain certain features of self-focusing.

In media in which the refractive index declines with increasing intensity, the effect arises of self-defocusing of a beam. One of the most effective mechanisms of self-defocusing is heating of the medium. The features of thermal self-defocusing have recently been studied experimentally and theoretically (see, e.g.[29,30]).

Apparently the effect of thermal self-defocusing is fundamental in the self-action of continuous laser beams. One must also not fail to take it into account in systems employing long enough (>100 nsec) laser pulses. Strong self-defocusing can also occur in saturating laser amplifiers when the frequency of the signal is higher than the resonance frequency of the material. The problem of how important this effect is in actual situations is being currently studied.

In closing the discussion of the studies pertaining to this section, we note finally that the self-action effects (dependence of the refractive index on the wave intensity) are precisely what are involved in the anomalous spectral broadening of laser pulses in liquids. According to data given at the Erevan symposium, two types of broadening are observed in different situations: symmetric broadening (with respect to the mean frequency) ($\sim$100–200 cm^{-1}) (such data were given in[32]; see also[35]), and asymmetric, extending mainly in the Stokes direction (see, e.g.[34,36,37]). The size of this broadening can also be quite significant, and can amount to hundreds of cm^{-1} (Fig. 5). Several possible models were proposed at the symposium for the process of anomalous broadening of the frequency spectrum. In one way or another, all of them are based on taking into account a non-linear increment to the refractive index.

Apparently, symmetric broadenings are explained by an effect in which amplitude modulation of the intense wave is transformed into phase modulation (the envelope pulse is practically not altered here).* If we consider that this process occurs in the hyperthin filaments, which carry a flux intensity of $\sim10^{12}$ watts/cm^2, this mechanism can explain a broadening of $\sim$100–200 cm^{-1} (see[31,33]). The situation is more complex in the asymmetric broadenings. Indeed, at first glance they have a simple explanation involving stimulated scattering of the wing of the Rayleigh line. However, the nature of the broadening (according to Brewer,[36] each self-focusing filament has its own practically-discrete frequency shift) and its size cast doubt on this mechanism.

Townes and his associates[34] have proposed an explanation based on the fact that the envelope of the wave packet tends to acquire the form of a shock wave in a medium having a refractive index depending on the light amplitude, as shown in Fig. 6 (thus we are concerned with strong modifications of the envelope, in contrast with the first case). We should note that Ostrovskiĭ first developed a theory of this effect in 1963 without considering the problem of spectral broadening (see[33]). The numerical calculations that Kelley (Massachusetts Institute of Technology, USA) presented at Erevan show that the proposed mechanism can explain extremely large asymmetric broadenings in self-focusing liquids.

Nevertheless, as we see it, there are not enough experimental data obtained under strictly controlled conditions to solve the problem finally. However, in designing such experiments, one should not restrict one's self to recording spectra alone. Separate information on the amplitude and phase structure of the

*)We shall not bother to list them in detail here; they have been discussed recently in Usp. Fiz. Nauk. [31]

**Unfortunately, no detailed comparison of theory and experiment on this point has been performed yet. The situation is complicated in a number of cases by the fact that the experiments were performed with multimode lasers, for which additional broadening mechanisms exist.

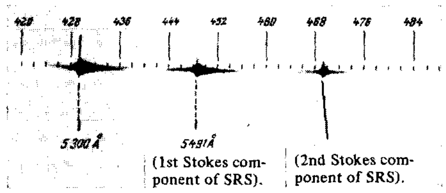


FIG. 5. Spectra of laser radiation and the Stokes components of SRS in carbon disulfide. A single-mode laser was used for excitation. Considerable broadening toward the Stokes region is visible. The photographs were obtained by Bol'shov and Venkin (Moscow State University), who were studying self-focusing of second-harmonic radiation from a neodymium laser in CS_2.

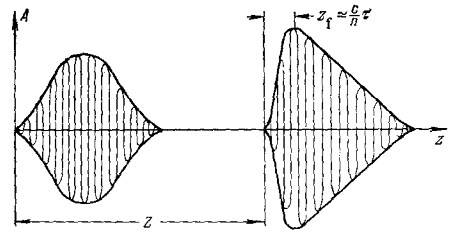


FIG. 6. Deformation of the envelope of a light pulse in a medium whose refractive index depends on the field ($n = n_0 + n^2E^2, n_2 > 0$). A steep drop is formed at the trailing front within a distance Z. The width of the front is related to the relaxation time τ of non-linearity.

radiation is of considerable interest. As we see it, a detailed study of the frequency spectrum of the second harmonic excited by the given radiation would be suitable from this standpoint. The process of harmonic generation is very sensitive to the form of phase modulation. Hence, it can be used in some cases for a sort of phase detection; concrete estimates along this line have been made in[38]. Apparently, there are no reasons for ruling out of consideration other broadening mechanisms, in particular, such as the effect of cross modulation of backward-traveling pulses, the appearance of which involves intense stimulated Raman scattering at an angle of 180°.

In connection with discussing the problem of anomalous broadening, we should also consider the practical aspect of the discussed effects: the problem is that of shaping of hypershort ($\tau_p = 10^{-11}-10^{-12}$ sec) powerful pulses in a passive non-linear medium. Thus, the effects of spatial self-focusing and self-contraction of packets can jointly lead to attainment of record highs in light fields in laser technology.

4. STIMULATED SCATTERING

The most interesting studies in this field in the past year have been those searching for stimulated scattering at the original frequency. The view is now generally acknowledged that every spontaneous scattering should transform into stimulated scattering at sufficiently high intensities of the scattered radiation.*

Study of the undisplaced component of the spontaneous Rayleigh scattering constitutes a major chapter in molecular optics. Purposeful searches for the corresponding stimulated scattering have been begun only very recently. Apparently, the first study systematically treating scattering of the undisplaced component in strong fields was that of Bespalov and Kubarev.[39] They reported observation of stimulated Rayleigh scattering of the undisplaced component in liquid mixtures.

A simple theoretical model was proposed in[39] to explain stimulated Rayleigh scattering in mixtures. The essence is a stimulated alteration of the concentrations of the components in the mixture. (The corresponding scattering is also called stimulated concentration scattering. Several groups have almost simultaneously undertaken searches for stimulated thermal Rayleigh scattering in liquids. Stimulated thermal scattering (STS) can arise for two reasons. The first is the electrocaloric effect, which consists in variation of the entropy of the material with varying electric field:

$$\Delta S = \frac{1}{4\pi}\left(\frac{\partial \varepsilon}{\partial T}\right)_p E\,\Delta E. \tag{8}$$

The second reason is variation of entropy due to absorption of light by the medium

$$\Delta S = \frac{1}{4\pi}\frac{\varepsilon''}{T} E\,\Delta E. \tag{9}$$

In turn, the entropy change produces a change in the polarization per unit volume, according to the relation

$$P = \frac{\partial \varepsilon}{\partial S} SE, \tag{10}$$

As in all other forms of stimulated scattering, this leads to a change in the electromagnetic field.

The first of stimulated thermal Rayleigh scattering mechanism occurs in pure liquids, and has been studied by Fabelinskiĭ and his associates.[40] The second mechanism, which is realized in absorbing liquids, has been observed by Rank and his associates.[41] The theory of these phenomena has been taken up in[40,42,43]. Interestingly, both mechanisms leads to small, oppositely-directed shifts in the frequency of the scattered radiation, the one in the Stokes direction, and the other in the anti-Stokes. This fact makes it possible to distinguish experimentally the contributions of each of the mechanisms. Stimulated scattering close to the undisplaced component is of undoubted interest also in applications (in particular, in connection with the defocusing problem. We should expect that this interesting effect will be subjected very soon to an all-sided investigation.

The past year has also brought important results in studying such already well-studied forms of stimulated scattering as SRS and SMBS.* The most important feature of the new studies in these fields is that quantitative results were obtained on the amplification coefficients, line widths, etc.

As for SRS, we must acknowledge the results of experimental study of SRS in gases as being the most

*In the overwhelming majority of cases, spontaneous scattering was studied in detail before the appearance of lasers, while stimulated scattering has been studied in recent years. Nevertheless, there are exceptions. The most salient example is parametric luminescence, which was discussed above, The stimulated process corresponding to it (parametric amplification) was detected experimentally as early as 1965, i. e., two years before the spontaneous process was discovered. There are also examples of situations in which the search for spontaneous and stimulated scattering were carried out almost simultaneously (e. g., scattering by spin waves, the above-mentioned scattering by plasmons, etc.).

*We use the standard abbreviations: SRS for stimulated Raman scattering, and SMBS for stimulated Mandel'shtam-Brillouin scattering.

significant. We refer to the quantitative studies of the Stokes components under different conditions,[44] the determination of the generation threshold,[44] and the study of brief SRS pulses in gases excited at an angle of 180°.[45]

The absence in gases of such sources of instability as self-focusing and self-contraction has made it possible to compare theory and experiment in detail, and to get an almost complete picture of the phenomenon. The results of the cited studies permit us to reconsider the currently rather widespread skeptical attitude concerning the prospects of spectroscopic applications of SRS.

In this regard, we should mention the study of Lallemand and Simova,[44] who used the narrowing of an SRS line in hydrogen for a quantitative study of the displacement of the Q(1) line as a function of the pressure, temperature, and admixture of foreign gases. Thus, the SRS phenomenon is beginning to justify the hopes put on it in its first stage of investigation.

In closing the review of new studies on SRS in gases, we note finally the studies of Sorokin and his associates[66] and of Movsesyan and his associates,[67] who observed interesting stimulated effects in potassium vapor. In particular, they studied Raman scattering from the electronic levels.

A number of new results have been obtained in studying SRS in crystals. Here the era of quantitative measurements permitting a correct estimate of the limits of applicability of the existing theories is apparently just beginning. Several papers were presented at the Erevan symposium on the intensities of lines and scattering patterns in calcite and diamond. We are referring to the papers by Ataev and Lugovoĭ,[61] Mayer (France),[62] and Stoicheff (Canada).[63]

Tannenwald (Massachusetts Institute of Technology, USA) presented some very interesting results at Erevan; he observed SRS in quartz at low temperatures.[49] He was apparently the first to observe SRS by a transition simultaneously active in Raman scattering and infrared absorption. These facts open up interesting possibilities in the field of designing optically-pumped lasers for the infrared. By the way, this problem has attracted the attention of a number of research groups in the Soviet Union and abroad over the last few years. In the studies published thus far, they have discussed varied approaches to solving it, the use of gases in external electric fields or at high pressures, or of non-totally symmetric infrared-active vibrations in crystals (e.g., $LiNbO_3$). We can expect that successful experiments on excitation of infrared radiation by SRS will be performed in the immediate future, and not only on quartz.

Sushchinskiĭ and his associates[50] have discovered an interesting phenomenon. They were studying stimulated Raman scattering in powders, and found that the SRS threshold depends on the grain dimensions, and decreases to a certain limit with decreasing grain size. These authors interpret the results of their experiments on the basis of ideas of generation in a statistically inhomogeneous medium.

SRS studies in liquids are closely interwoven with the study of self-focusing. The distinctive feature of the studies in the past year is the fact that their authors do not restrict themselves only to adducing self-focusing in situations in which the theory and experiment decisively disagree, but try to obtain a quantitative interpretation of the experimental data. Here we should note, in particular, the paper presented at the Erevan symposium by Townes, Sacchi, and Lifsitz (Massachusetts Institute of Technology, USA). They explained the angular structure of the anti-Stokes SRS components by a model taking account of the anomalous broadening of the spectrum of the Stokes components, and of the waveguide properties of the filaments. Some studies on SRS excitation by powerful picosecond pulses are extremely interesting from the standpoint of studying the contribution of self-focusing effects.[57,60]

In the SMBS field, one significant advance is the quantitative study of the effect in gases and certain liquids. This has made it possible to fix the limits of applicability of the steady-state theory (Bret (France)[46] and Kaiser (West Germany)[47] gave papers at Erevan on this problem). We should note that the theory of SMBS itself has been substantially refined in the past year (see[64,48]). Hypersonic waves were directly detected for the first time in studying SMBS in quartz crystals.[48]

Finally we note the very interesting perspectives that are opened up by using the stimulated-scattering effects as mechanisms capable of stabilizing laser radiation, of modulating the Q-factor of a resonator, etc. This set of problems ("stimulated-scattering-loaded lasers") is drawing more and more attention at present. Perhaps new possibilities are being opened up here for studying the scattering effect itself.

5. APPLICATION OF METHODS OF NON-LINEAR OPTICS IN LASER TECHNOLOGY

It is traditional to use the methods of non-linear optics to extend the range of frequencies covered by coherent-radiation generators. The past year has brought some definite achievements in this field.

We note first of all that the field of application of optical-harmonic generators has expanded considerably in the past year. New physical results have been obtained by using these generators not only to study SRS and SMBS, but also to exert powerful effects on matter.

Here we should mention the results reported at Erevan by Zverev and his associates, who used harmonic generators along with ruby and neodymium lasers to study the characteristics of the breakdown of solids by light; the results of Delone and his associates, who used harmonic generators in experiments on many-photon ionization of gases; and the paper[52], in which harmonic generators were used to pump semiconductor lasers in the ultraviolet range.

The latter line of work seems very promising to us, since thus it becomes possible to study recombination radiation in dielectrics having a wide forbidden band, and probably to build lasers for the vacuum ultraviolet (another way to solve this problem is to use electron excitation of dielectrics, as was discussed recently by Basov (see, e.g.[51])).

As for the technique of frequency multiplication itself, we should mention the studies on harmonic generators in the ultraviolet range. The limiting efficiency of an optical frequency doubler has been determined experimentally and theoretically, and amounts to ~50–60%. This fact is very important in the technique of cascade frequency transformation.

Considerable progress has been made in the field of developing pulsed parametric light generators. Such generators are now operating not only in the laboratories that began these studies, but also in a number of other Soviet and foreign laboratories.

The working characteristics of these generators are continually being improved, and new designs are being proposed for resonator systems and for control of the oscillation frequency. The work of Marennikov (Novosibirsk) is worthy of note. He has built a generator from KDP with electrooptic tuning. Kreutzer (Bell Telephone Laboratories, USA) has achieved the same tuning system, but with a $LiNbO_3$ generator.

A new advance in developing parametric generators is a mirrorless generator emitting parametric superluminescence pulses. The work on these generators is being conducted by the group of Harris (Stanford University, USA) and by us at Moscow University. Recently in our laboratory we have obtained superluminescence pulses of powers up to 10 kW in KDP crystals with pumping at $\lambda = 0.53\ \mu$.

Considerable attention is being paid to the problem of building continuous or quasicontinuous parametric generators. Studies are intensively being conducted in a number of laboratories in the Soviet Union and abroad on three concrete possibilities: KDP or $LiNbO_3$ generators pumped by the second harmonic or directly by a laser based on Nd^{+3} in garnet; a $LiNbO_3$ generator pumped by an argon laser; and a generator based on a tellurium crystal, pumped by a CO_2 laser.

The studies on parametric systems in the optical range are not limited solely to developing generators continuously tunable in frequency. Important perspectives in applications are being opened up in line with the invention of parametric frequency raisers. A paper by Warner (Malvern, England) was heard with interest at the Erevan symposium. He spoke of experiments on transformation of CO_2-laser radiation into the visible range. Such a transformation using a proustite crystal and a ruby laser can be very efficient (with an efficiency up to 30–50%), while the noise coefficient of the transformer is unity.

There also appeared some new applications of the methods of non-linear optics in 1966–67. First of all, we should note here the use of methods of non-linear optics to generate and study picosecond pulses. We recall that when SRS is excited in liquids at an angle of 180°, the duration of the Stokes radiation pulse $\cong 10^{-11}$ sec, while the instantaneous power is an order of magnitude higher than the pumping power. This is explained by the reversible peculiarities of the interaction of the backward-traveling component of the Stokes radiation with the pumping radiation. These peculiarities are such that the reflected backward-traveling pulse moves as though in a medium having an inverted population density, and contracts and becomes stronger as it propagates. An analogous effect has also been observed in SRS in gases.[45] It is apparently possible, in particular by the methods of non-linear optics, to form light pulses of extremely short duration ($\sim 10^{-14}$ sec, see[73]),

On the other hand, the methods of non-linear optics can be very effective in measuring the characteristics of picosecond pulses. Along with the methods already well known, which use the frequency-doubling process, we note a method recently proposed by Giordmaine,[60] using two-photon absorption, and suitable for detecting pulses of durations down to 10^{-13}–10^{-14} sec.

Finally, as Gerritsen[54] has recently demonstrated, the methods of non-linear optics open up interesting possibilities in holography.

In closing this brief review, which is mainly based on the materials of the Erevan symposium, we emphasize again that we have tried primarily to distinguish the new lines of investigation that have arisen in the past year. We must hope that the wealth of material given in the review will not arouse doubts in the reader as to the correctness of the assertion made at the beginning of this article.

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