Nonequilibrium mixing of optical frequencies

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A study was made of nonequilibrium mixing of ultrashort laser pulses. The theory was developed for the Gaussian and Lorentzian distributions of the original waves; the latter case was analyzed in detail. Nonequilibrium generation (by a cascade method) of the third harmonic of a laser emitting locked longitudinal modes was observed experimentally. It was predicted theoretically and confirmed experimentally that the width of the spectrum of the harmonic frequency reached saturation under nonequilibrium mixing conditions. The measured spectra of the second and third harmonics generated under quasistatic conditions and the ratio of their widths indicated that the spectrum of a train of ultrashort laser pulses was Lorentzian. Calculations were also made of the characteristic parameters of the nonequilibrium generation of sum frequencies in KDP and $CaCO_3$ crystals.

INTRODUCTION

Nonequilibrium interactions between optical waves are a subject of constant interest. This interest is primarily due to the availability of lasers emitting picosecond pulses. A considerable number of theoretical and experimental investigations have been made of the stimulated scatter- A considerable number of theoretical stimulated scattering of laser radiation and of the multiplication of optical frequencies under nonequilibrium conditions (see, respectively, refs. 1, 2 and 3, 4 as well as the literature cited in these references). Among the multiplication processes the greatest attention has been paid to the frequency doubling of ultrashort pulses when an ordinary laser wave excites an extraordinary second-harmonic wave in a nonlinear crystal, i.e., a two-wave nonequilibrium interaction is observed. On the other hand, the generation of higher harmonics (used frequently to raise the optical frequency) is in most cases a three-wave process: The frequencies or polarizations of the waves being mixed are usually different.[5-9] However, there have been hardly any investigations of nonequilibrium mixing of optical frequencies, although this method has produced the fifth harmonic.[5-6] The width of the coherent mixing spectrum is estimated in refs. 7-9 and elsewhere, subject to the phase matching of the waves.

We shall show that this approach is unsuitable in the nonequilibrium mixing case. We shall give a theoretical analysis of nonequilibrium mixing of optical frequencies and report an experimental study of the cascade generation of the third harmonic. We shall show that in the strong-nonequilibrium mixing the energy and width of the sprectrum of the third harmonic reach saturation. A comparison of the measured widths of the spectra of the second and third harmonics excited under quasistatic conditions confirms the Lorentzian form of the spectrum of ultrashort laser pulses, deduced from spectral analysis.

1. THEORY OF NONEQUILIBRIUM OPTICAL MIXING

For the sake of simplicity we shall consider nonequilibrium mixing of plane waves. In this case the generation of the sum frequency $\omega_3 = \omega_2 + \omega_1$, considered on the assumption that the fields of the exciting frequencies are fixed, is described by

$$\frac{\partial A_3}{\partial z}+\frac{1}{u_3}\frac{\partial A_3}{\partial t}=-i\gamma A_{1_0}\left(t-\frac{z}{u_1}\right)A_{2_0}\left(t-\frac{z}{u_2}\right), \tag{1}$$

where A_j and u_j are the complex amplitudes and group velocities of the waves of frequencies identified by the subscript; $A_{j_0}(t)$ is the amplitude at the input of a nonlinear crystal in the plane z = 0; γ is the nonlinear interaction coefficient; the direction of the z axis is assumed to be the same as the phase-matching direction in the crystal.

The solution of Eq. (1) can be analyzed conveniently by representing the amplitudes $A_j(t)$ in the form of their Fourier spectra; in this way we find that

$$A_3(\Omega, z)=i\frac{\gamma}{2\pi}\int_0^z \exp(i\nu_{32}\Omega z_1)\times\int_{-\infty}^{+\infty}A_{1_0}(\Omega_1)A_{2_0}(\Omega-\Omega_1)\exp(i\nu_{21}\Omega_1 z_1)\,d\Omega_1 dz_1. \tag{2}$$

Here, $\nu_{jn} = 1/u_j - 1/u_n$ is the detuning of the group velocities;

$$A(\Omega)=\frac{1}{2\pi}\int_{-\infty}^{+\infty}A(t)\,e^{-i\Omega t}dt. \tag{3}$$

We shall consider the spectral density $S_3(\Omega, z) = |A_3(\Omega, z)|^2$ of the sum frequency for the Lorentzian and Gaussian forms of the exciting Fourier spectra. Then,

$$A_{j_0}(\Omega)=\sqrt{S_j(0)}\frac{\Delta\omega_j}{\Delta\omega_j+i\Omega} \tag{3a}$$

or

$$A_{j_0}(\Omega)=\sqrt{S_j(0)}\exp\left[-\frac{1}{2}\left(\frac{\Omega}{\Delta\omega_j}\right)^2\ln 2\right], \tag{3b}$$

where $\Delta\omega_j$ is the half-width of the spectrum at the 0.5 level.

In the (3a) case, we obtain

$$S_3(\Omega, L)=\gamma^2\left[\frac{2\Delta\omega_1\Delta\omega_2}{(\Delta\omega_1+\Delta\omega_2)\nu_{32}}\right]^2\times\frac{\sin^2(\mu L\Omega/2\Delta\omega_1)+\mathrm{sh}^2 L/2}{\Omega^2+(\Delta\omega_1/\mu)^2}e^{-L}S_1(0)S_2(0)\tilde{S}_3^{(q)}(\Omega), \tag{4}$$

where

$$\tilde{S}_3^{(q)}(\Omega)=\Delta\omega^2/[\Delta\omega^2+\Omega^2];\ \Delta\omega=\Delta\omega_1+\Delta\omega_2; \tag{5}$$

$$\mu=\nu_{32}/\nu_{21};\ L=z/l_{21};\ l=(\nu_{21}\Delta\omega_1)^{-1},\ \nu_{21}>0. \tag{6}$$

The normalized spectral density of the sum frequency is given by the expression

$$\tilde{S}_3(\Omega, L)=\frac{(\Delta\omega_1/\mu)^2}{(\Delta\omega_1/\mu)^2+\Omega^2}\frac{\sin^2(\mu L\Omega/2\Delta\omega_1)+\mathrm{sh}^2 L/2}{\mathrm{sh}^2 L/2}\tilde{S}_3^{(q)}(\Omega). \tag{7}$$

In general, the spectrum of the frequency being excited, Eq. (4) or (7), depends on the parameters μ and $L = z/l_{21}$. The expressions (4) and (7) are asymmetric under the transposition of the subscripts $1 \rightleftarrows 2$, which is due to the structure of the interacting pulses. The spectrum (3a) corresponds to pulses with a steep leading edge. There-

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fore, the spectrum and, consequently, the intensity of the sum frequency practically cease to change when the delay time between the exciting pulses $T = z/u_2 - z/u_1$ becomes equal to the duration of the pulse traveling at the higher velocity. In the case under consideration we have $u_1 > u_2$ ($\nu_{21} > 0$) and the characteristic pulse separation length is $l_{21} = (\nu_{21}\Delta\omega_1)^{-1}$. If $\nu_{12} > 0$, we obviously have $l_{12} = (\nu_{12}\Delta\omega_2)^{-1}$.

Under quasistatic frequency mixing conditions (in this case the length of a crystal z is shorter than either of the pulse separation lengths, $z \ll l_{21}$ and $z \ll l_{32}$), it follows from Eq. (7) that

$$\tilde{S}_3(\Omega, L) = \tilde{S}_3^{(q)}(\Omega) \tag{8}$$

and, consequently, the half-width of the sum frequency spectrum is

$$\Delta\omega_3^{(q)} = \Delta\omega = \Delta\omega_1 + \Delta\omega_2.$$

Under nonequilibrium conditions we can distinguish two cases. If the exciting pulses do not separate in the length of the crystal ($z \ll l_{21}$), we have

$$\tilde{S}_3(\Omega, z) = \frac{\sin^2(\nu_{32}z\Omega/2)}{(\nu_{32}z\Omega/2)}\tilde{S}_3^{(q)}(\Omega). \tag{9}$$

This mixing case is analogous to the nonequilibrium frequency doubling discussed in detail in ref. 3, so that we shall not consider it any further. However, if $z \gg l_{21}$, we find that

$$\tilde{S}_3(\Omega, z) = \frac{(\Delta\omega_1/\mu)^2}{(\Delta\omega_1/\mu)^2 + \Omega^2}\tilde{S}_3^{(q)}(\Omega). \tag{10}$$

Thus, in contrast to Eq. (9), in the present case the sum frequency spectrum is smooth and its width is independent of the crystal length z. The expression for the width of the spectrum can be written in the form

$$2\Delta\omega_3^S = 2K(\mu)\Delta\omega_3^{(q)}, \tag{11}$$

where the coefficient of proportionality is

$$K(\mu) = \left\{\frac{1}{2}\left[\sqrt{[1+(q\mu)^{-2}]^2 + 4(q\mu)^{-2}} - 1 - (q\mu)^{-2}\right]\right\}^{1/2} \tag{12}$$

and

$$q = \Delta\omega/\Delta\omega_1.$$

Equation (11) represents the minimum nonequilibrium width of the sum frequency spectrum; if $|q\mu| \gg 1$, we find that $\Delta\omega_3^S = \Delta\omega_1/\mu$. Numerical calculations show that the minimum width $\Delta\omega_3^S$ of Eq. (11) is effectively reached when $L \approx 3$. In the $L < 3$ range the value of $\Delta\omega_3$ naturally lies between $\Delta\omega_3^S$ and $\Delta\omega_3^{(q)}$. In the cascade generation of higher harmonics the parameter q is independent of the spectral width of the original radiation if the process of multiplication of the optical frequencies in the preceding cascades is quasistatic. In this case the coefficient $K(\mu)$ of Eq. (12) represents the maximum narrowing of the spectrum under nonequilibrium excitation conditions and is governed solely by the dispersion properties of a nonlinear crystal (values of μ) and by the nature of the mixing process.

Table 1 gives the values of the characteristic parameters of the nonequilibrium mixing of frequencies in the generation of optical harmonics in KDP and $CaCO_3$ crystals.

Integrating Eq. (4) with respect to Ω, we obtain the following expression of the sum frequency energy:

$$W_3 = \frac{32}{cna^2}\frac{\Delta\omega_2\gamma^2 l_{21}^2}{q(1+|q\mu|)}F(l)W_1W_2, \tag{13}$$

where a is the radius of the exciting beams; W_1 and W_2 are the energies of the exciting beams;

$$F(L) = 1 + \exp(-2L) - 2\frac{|q\mu|\exp(-2L) - \exp[-(1+|q\mu|)L]}{|q\mu| - 1}. \tag{14}$$

It follows readily from Eqs. (13) and (14) that if $L < 1$ the energy is $W_3 \propto z^2$, whereas for $L > 1$, we have $W_3 \propto l_{21}^2$ [$F(L) \approx 1$]. The sum frequency energy reaches its maximum value in the latter case. The maximum energy conversion coefficient of the nonequilibrium mixing process is

$$\eta_{max} = \frac{W_{3,max}}{\sqrt{W_1W_2}} = \frac{2\sqrt{2}}{q(1+|q\mu|)}\left(\frac{l_{21}}{l_\gamma}\right)^2. \tag{15}$$

Here, $l_\gamma = [\gamma\sqrt{A_1(0)A_2(0)}]^{-1}$; $A_j(0)$ are the maximum values of the pulse amplitudes.

These characteristics of the nonequilibrium mixing of optical frequencies in the case of a Lorentzian spectrum (3a) are also basically valid in the case of a Gaussian spectrum (3b). The normalized spectral density of the sum frequency is now given by

$$\tilde{S}_3(\Omega, L) = \exp[-2(\varkappa p\Omega/\Delta\omega)^2]\,|\Phi(iL/2p)|^{-2}\,|\Phi(\varkappa p\Omega/\Delta\omega + iL/2p) - \Phi(\varkappa p\Omega/\Delta\omega)|^2\,\tilde{S}_3^{(q)}(\Omega), \tag{16}$$

TABLE 1. Parameters μ, K, and l_{21}, Representing Nonequilibrium Mixing of Optical Frequencies*)

Crystal	No. of harmonic	Type of interaction	$\lvert\mu\rvert$	$K = \frac{\Delta\omega_3^S}{\Delta\omega_3^{(q)}}$	$3l_{21}$, cm
KDP	2	$o_1E_1 \to E_2$	0.42	0.70	1.3
	3	$E_1o_2 \to E_3$	0.37	0.62	1.0
	3	$o_1o_2 \to E_3$	2.1	0.15	3.0
	5	$o_1o_4 \to E_5$	0.25	0.87	0.2
$CaCO_3$	3	$o_1o_1E_1 \to E_3$	0.72	0.40	1.0
	3	$o_1E_1E_1 \to E_3$	0.09	0.92	0.5
	5	$o_1o_1o_3 \to E_5$	3.3	0.08	0.3

*) These parameters are given for $\lambda_1 = 1.06\,\mu$; the characteristic lengths l_{21} are calculated for pulses of $\tau_1 = 1$ psec duration.

where $\Phi(x)$ is the error function;

$$\tilde{S}_3^{(q)}(\Omega) = \exp\left(-\frac{\Omega^2}{\Delta\omega_1^2 + \Delta\omega_2^2}\ln 2\right);$$

$$\Delta\omega = (\Delta\omega_1^{-2} + \Delta\omega_2^{-2})^{1/2};$$

$$2x = \frac{\Delta\omega_1^2 - \Delta\omega_2^2}{\Delta\omega_1^2 + \Delta\omega_2^2} + \frac{\nu_{32} + \nu_{31}}{\nu_{21}};\ L = \frac{z}{l_{21}'};$$

$$l_{21}' = (\nu_{21}\Delta\omega)^{-1}(\nu_{21} > 0);$$

$$p = (0.5\ln 2)^{1/2}. \qquad (17)$$

An analysis of Eq. (16) can be made in the same way as that of Eq. (7). However, in contrast to Eq. (7), Eq. (16) is symmetric relative to the exciting waves. This is reflected in the definition of the characteristic length l_{21}' for the Gaussian pulses [see Eq. (3b)].

2. EXPERIMENTAL RESULTS. CASCADE GENERATION OF THIRD HARMONIC BY PICOSECOND LASER PULSES

We used a mode-locked neodymium laser as the master oscillator. The reflection coefficients of the resonator mirrors were 98 and 4%. The resonator was 1.5 m long. A cell, 0.3 mm thick and containing a nonlinear absorber, was placed next to the 98% mirror; the initial transmission of the cell was 75–80%. The thermal lens effect, which appeared in the neodymium rod, was compensated by introducing a spherical lens (f = 4.5 m) into the resonator. The laser radiation was in the form of a train of 15–20 pulses. The energy of the train was 0.1 J and the average pulse duration was 3.3 psec (these results were obtained by a two-photon method whose capabilities in the case of ultrashort pulses are considered in refs. 3 and 10).

The laser frequency was tripled by the cascade method using two KDP crystals (one acted as the doubler and the other as the mixer). The second harmonic was excited in the first crystal (4 cm long) under quasistatic conditions. The mixer was 0.5, 4, or 8 cm long, so that we could observe the frequency tripling under quasistatic and nonequilibrium conditions. The $o_1o_1 \rightarrow E_2$ interaction was used in the doubler and the $E_1o_2 \rightarrow E_3$ interaction in the mixer.

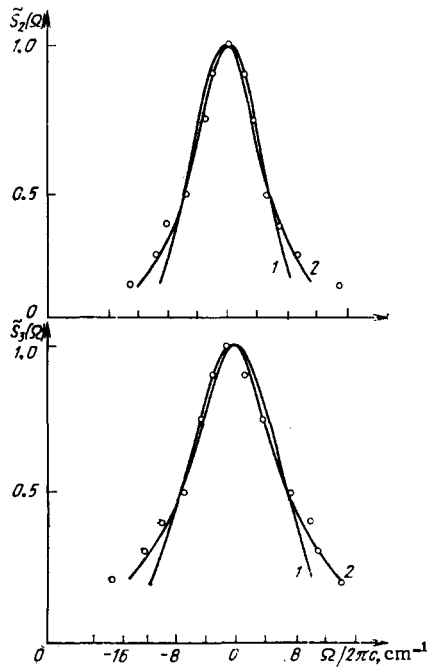

Fig. 1. Spectra of the second (a) and third (b) harmonics excited under quasistatic conditions: O) experimental results; 1) Gaussian distribution; 2) Lorentzian distribution.

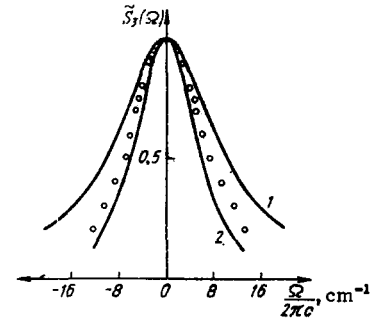

Fig. 2. Spectrum of the third harmonic generated under nonequilibrium conditions: O) experimental results; 1, 2) curves calculated for the quasistatic and nonequilibrium condition, respectively (in both cases the Lorentzian form is assumed).

In mixing the frequencies of light pulses the output depended on the delay time between the pulses at the mixer input. The group detuning between the exciting waves was $\nu_{21} = 0.8 \cdot 10^{-3}$ sec/cm in a KDP crystal and $\nu_{21} = 0.4 \cdot 10^{-15}$ sec/cm in air. Therefore, under our experimental conditions (when the doubler was 4 cm long and the distance between nonlinear crystals was 10 cm) the delay between the pulses was of little importance.

The second-harmonic spectrum was recorded with a UF-89 camera and a diffraction grating, whereas the third harmonic was recorded with an STÉ-1 spectrograph (the measurements were carried out simultaneously in the course of each laser flash). Spectrograms of a series of flashes were subjected to a microphotometric analysis and the results were then analyzed statistically. It was found that the experimental error did not exceed 5%.

The quasistatic frequency tripling was studied using a crystal 0.5 cm long ($z \ll l_{21}$, $z \ll l_{31}$, and $z \ll l_{32}$). The experimentally obtained spectra of the second and third harmonics are shown in Fig. 1. The continuous curves represent the theoretical dependences for the Gaussian and Lorentzian spectra. We can see that the experimental results fit better the Lorentzian distribution. This is confirmed by the ratio of the widths of the spectra of the second and third harmonics. The experimental value of the ratio $K = \Delta\omega_3/\Delta\omega_2$ was $K = 1.43 \pm 0.05$, whereas the theoretical value obtained under quasistatic mixing conditions was $K = 1.5$ for the Lorentzian spectrum [see Eqs. (6) and (8)] and $K = 1.2$ for the Gaussian spectrum [see Eq. (17)].

The third harmonic was also excited under nonequilibrium conditions using crystals 4 and 8 cm long (these values were greater than all the characteristic lengths listed in Table 1). Figure 2 shows the spectrum of the third harmonic obtained experimentally under nonequilibrium excitation conditions employing laser radiation of width $\Delta\nu_1 = 6.5$ cm^{-1} and the second harmonic of this radiation ($\Delta\nu_2 = 13$ cm^{-1}). We can see that the experimental distribution agrees better with curve 2, which is plotted

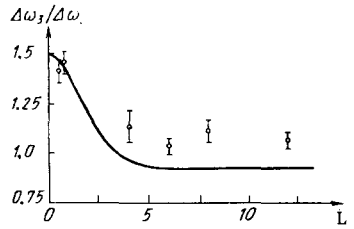

Fig. 3. Dependence of the relative width $\Delta\omega_3/\Delta\omega_2$ of the spectrum of the third harmonic on the reduced length $L = z/l_{21}$ of a nonlinear crystal (the continuous curve shows the dependence for the Lorentzian form of the exciting radiation; the points are the experimental values).

for the nonequilibrium conditions.

Figure 3 shows the experimentally determined dependence of the width of the third harmonic spectrum on the reduced length L of a crystal. The narrowing of the spectrum of the third harmonic stops in the range $L > 3$. The experimental values corresponding to the quasistatic mixing conditions ($L < 1$) are in good agreement with the calculated results, whereas the values obtained under the nonequilibrium conditions diverge somewhat from the theoretical curve. This may be due to the fact that the lengths of the nonlinear crystals used in the nonequilibrium case are such that they are affected by the aperture effects which are ignored in our theory; moreover, the width of the spectrum of laser pulses may vary along a train (the time structure of picosecond pulses is discussed in ref. 11). However, it should be pointed out that in the case of pulses of identical intensity the fluctuations in their duration do not affect the ratio of the average widths of the spectra of the sum and laser frequencies in the cascade generation process. This follows from Eq. (11) and from the observation that the coefficient $K(\mu)$ is independent of the width of the original spectrum.

Spectral measurements indicated that the energy conversion coefficient of the third harmonic generation did not exceed 0.5%. The maximum energy efficiency of the cascade tripler was ~10% when the maximum conversion efficiency of the second harmonic was ~30% and the laser radiation power was $\sim 10^9$ W.

2. CONCLUSIONS

The results of the present investigation can be summarized as follows.

1. Detailed measurements of the form of the spectra of the second and third harmonics of a train of ultrashort pulses make it possible to determine the spectrum of the original laser radiation. Ultrashort pulses are found to have the Lorentzian spectra. This is in agreement with the conclusions reached in ref. 12. However, in some earlier investigations (see, for example, refs. 13 and 14) the spectra of ultrashort pulses have been found to be peaks of ~ 5 cm^{-1} width superimposed on a wide pedestal (> 100 cm^{-1}). The difference between our spectra and those reported in refs. 13 and 14 is evidently due to the lower nonlinear losses in our experiments because of the higher minimum transmission of the cell containing a nonlinear absorber (the phenomena associated with the nonlinear losses in picosecond lasers are discussed thoroughly by Basov and others in refs. 15 and 16).

2. Nonequilibrium mixing of optical frequencies does not give rise to an unlimited improvement in the monochromaticity of the output radiation: The detuning of the group velocities results in the spatial separation of the pulses so that after a certain distance the sum frequency is no longer generated. This process differs considerably from the nonequilibrium frequency doubling[3] in which the width of the harmonic spectrum can be made as small as we please. It should be pointed out that the nonequilibrium mixing results cannot be deduced from the theory of mixing of monochromatic waves [$A_3 \approx \sin(\Delta z/2)/(\Delta/2)$] if allowance is made for the detuning $\Delta = k_1 + k_2 - k_3$ away from the output frequency (this procedure is widely used in analyses of the nonequilibrium doubling of optical frequencies). This is due to the fact that the nonequilibrium mixing mechanism is subject to the simultaneous influence of the group detuning between the original waves as well as between the original and excited waves (usually the detunings ν_{21} and ν_{32} are of the same order of magnitude as ν_{31}, as shown in Table 1). Consequently, the condition of phase matching gives an incorrect estimate of the width of the spectrum of the radiation which can be transformed effectively into the sum frequency.

The results of the present study are applicable not only to the generation of the sum frequency, but they can be applied also (in the case of small conversion coefficients) to the nonequilibrium generation of the difference frequency.

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