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FORMATION OF SUBPICOSECOND UV PULSES BY MULTIPLE NONLINEAR CONVERSION

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1. The purpose of this article is to describe an ultrashort-pulse generator that emits in the wavelength band $\lambda = 0.26 - 0.28\ \mu$ pulses of duration to 0.5×10^{-12} sec and power $\sim$10 MW. Such pulses were generated by twofold doubling of the emission of a neodymium-glass picosecond laser. Discrete frequency variation in the UV band was attained through SRS of the fourth-harmonic radiation ($\lambda_4 = 0.26\ \mu$) in liquid nitrogen. The energy efficiency of the conversion into the fourth harmonic reached 5%.

A major advantage of the method used by us to generate UV picosecond pulses is the improvement, through multiple nonlinear conversion, of the structure of the wave train (suppression of satellites, filtering of the wings of the non-synchronized spectrum) and reduction of the pulse width.

2. Picosecond pulses in the UV band are of considerable interest as a means of pumping UV lasers, in the determination of the relaxation times of electronic levels, and in the investigation of nonstationary nonlinear effects.

Cascade frequency conversion is at present practically the only way of obtaining sufficiently powerful picosecond pulses in the region $\lambda = 0.26\ \mu$.

3. The apparatus (Fig. 1) consisted of a picosecond-pulse generator (LGS-1 glass) and two frequency multipliers. In the main laser, the cell with the dye was made integral with the total-reflection mirror; the dye solution was replenished in the cell after each flash.

The generator operated in the lowest transverse mode, which was separated by a diaphragm of 2.2 mm diameter. Typical data on the picosecond pulse train were: total energy of train of 20 pulses $W_1 = 5 \times 10^{-2}$ J; pulse duration (determined from the two-photon luminescence track) $\tau_p = 4 \times 10^{-12}$ sec; peak intensity of unfocused beam $I_1 = 4 \times 10^{10}$ W/cm^2. The first frequency doubler (output frequency $\lambda_2 = 0.53\ \mu$) used in our experiment was a KDP, ADP, or CDA crystal; in the second doubler (output wavelength $\lambda_4 = 0.26\ \mu$) we could only use KDP or ADP. An effective quasistatic frequency doubling accompanied by narrowing of the pulse takes place if the length ℓ of the nonlinear crystal is

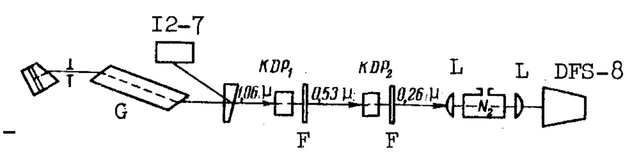

Fig. 1. Experimental setup: G - picosecond pulse generator, $KDP_{1,2}$ - frequency doublers using KDP crystals, F - filters, N_2 - liquid-nitrogen cyrostat, I2-7 - oscilloscope and other measuring apparatus.

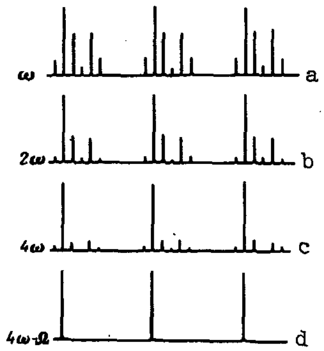

Fig. 2. Schematic diagram explaining the improvement of the structure of a train of picosecond pulses following triple nonlinear conversion. Figure 2a shows one of the realizations of the sequence of pulses with a substructure; Figs. 2b, 2c, and 2d show the results of their conversion in the doubler and by SRS in liquid nitrogen.

chosen such that $\ell \gtrsim \ell_{nl} \sim (k\chi E)^{-1}$ (ℓ_{nl} is the so-called nonlinear-interaction length) and $\ell < L_{ij} = \tau_i |u_i^{-1} - u_j^{-1}|^{-1}$ (L_{ij} is the group-delay length of the interacting pulses).

For a KDP crystal at $\tau_{1,2} = (3 - 4) \times 10^{-12}$ sec we have $L_{12} = 15$ cm and $L_{24} = 1$ cm. Accordingly, the nonlinear lengths at $I = (2 - 4) \times 10^{10}$ W/cm^2 are $\ell_{nl}^{(12)} = 0.2$ cm and $\ell_{nl}^{(24)} = 0.1$ cm. Starting from these estimates, the length of the KDP crystal was chosen to be $\ell_1 = 4$ cm in the first doubler and $\ell_2 = 0.2$ cm in the second. In a number of experiments we used a CDA crystal in the first doubler; its use is beneficial if the divergence of the main laser beam is large.

Characteristics of generator of UV picosecond pulses

Wavelength μ	Train energy J	Spectr. width cm^{-1}	Peak power W/cm^2
1.060	$5 \cdot 10^{-2}$	8	$4 \cdot 10^{10}$
0.530	$1 \cdot 10^{-2}$	25	$1.7 \cdot 10^{10}$
0.265	$1 \cdot 10^{-3}$	50	$5 \cdot 10^{9}$
0.282 (1st Stokes) SRS in N_2	$2 \cdot 10^{-4}$	—	$1 \cdot 10^{9}$

The characteristics of the KDP-crystal frequency converter are listed in the table. The total spectrum widths were measured at half-height of the intensity distribution.

The spectral measurements (as well as the oscillograms of the trains) show an appreciable improvement of the train structure (suppression of satellites etc.) by cascade multiplication of the frequency and by SRS (see Fig. 2). The fourth-harmonic spectra were smooth and symmetrical (there was no self-focusing and self-modulation in the KDP crystals); as a result, the duration of the fourth harmonic pulses was close to the reciprocal width of the spectrum.

4. Estimates of the power of the laser emission and its harmonics can be obtained not only by measuring the pulse energy and duration, but also indirectly, from saturation effects in the frequency doubler and from threshold

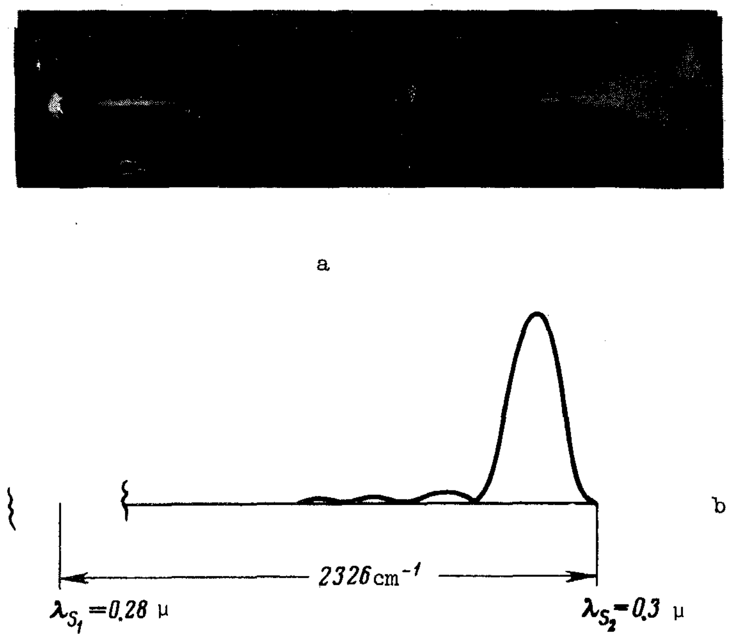

Fig. 3. Spectrogram and microgram of the spectrum of the first Stokes component (λ_s = 0.28 μ) and its broadening due to phase modulation.

effects such as SRS and self-focusing; these data were used in the table.

Nonlinear phenomena in liquid nitrogen were used also to estimate the duration of the picosecond pulses formed as a result of triple nonlinear conversion. Figure 3 shows a microgram and spectrogram of the spectrum of the first Stokes component of the SRS excited by the fourth harmonic in liquid nitrogen. The shape of the spectrum agrees with good accuracy with that of the spectrum due to phase self-modulation of a single pulse of duration $\tau = 0.5 \times 10^{-12}$ sec [1]. This value agrees with the estimate obtained from data on the width of the fourth-harmonic spectrum ($\Delta\nu_4 = 50$ cm^{-1}, $\tau_4 \simeq (\Delta\nu_4)^{-1} \simeq 0.6 \times 10^{-12}$ sec). The additional compression of the Stokes pulse is due to nonstationary effects that occur in SRS (cf., e.g., [2]).

5. The duration and shape [3] of the fourth-harmonic pulse could be varied in controllable manner in our setup by increasing the length of the crystal in the second doubler. Calculation [3] shows that the fourth-harmonic pulse duration can be varied in the range 0.5 - 5 psec by increasing the length of the second KDP crystal from 0.2 to 4 cm.

The presence of duration (and shape) modulation was confirmed by experiments in which the duration and shape of the pulse were determined from the self-action characteristics and SRS in liquid nitrogen. At pulse durations ∿0.5 psec, superbroadening of the spectrum was observed at fourth-harmonic powers lower than the threshold power of the nonstationary SRS; in a field of long pulses, the situation was reversed, and SRS (with several Stokes components) appeared much earlier than superbroadening

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