Up-conversion of microsecond CO_2-laser pulses

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Up-conversion of long, 3 to 50 μs, CO_2-laser pulses was studied experimentally. The conversion efficiency of 1 to 2% was achieved when using Nd:YAG laser radiation at 1.064 μm wavelength as a pump radiation and a 3.9-mm-long $ZnGeP_2$ crystal that is characterized by the absorption coefficient of 2.0 cm^{-1} at this wavelength and 2.5 cm^{-1} at the up-converted wavelength (0.9548 μm). It was shown that when using a thick silicon avalanche photodiode operating in the photon counting mode the signal-to-noise ratio can be increased by 450 times as compared with the direct detection of CO_2-laser signals by an mercury cadmium telluride (MCT) photodiode.

The up-conversion (upward frequency conversion) of IR radiation into the visible and near IR regions is very attractive for a many reasons. It proved possible to use high-sensitivity fast detectors and image recorders requiring no cryogenic cooling, usual photosensitive materials, and other well developed and, as a rule, less expensive technical means for recording the up-converted IR signals and images. Up-conversion of CO_2-laser radiation, which is widely used in many applied systems, attracts particular interest. Up-converters based on nonlinear $AgGaS_2$ and Ag_3AsS_3 single crystals are most widely used since recent times and provide the highest efficiency. Their quantum efficiency achieves 20–40% when using nanosecond Nd:YAG and lasers emitting shorter waves as pump sources. However, at up-conversion of long, 3 to 50-μs, CO_2-laser pulses the efficiency drops down to $4\cdot10^{-8}$ – 10^{-2}% (Ref. 1). This is caused by several factors. First, the values of phase matching angles of the known up-converters are typically within 20–55°. This restricts the efficient length of the crystals and, consequently, the up-conversion efficiency because of the optical walk off effect and impossibility of optimal focusing. Second, the widely used crystals are characterized by relatively low values of the quality coefficients $M = d^2/n^3$ (proportional to the conversion efficiency). Third, the increase in the pump pulse duration decreases the dielectric strength of crystals and, as a result the threshold pump intensity, what also leads to the corresponding decrease in the conversion efficiency. As a result, low efficiency of the up-conversion and low quantum efficiency of the detectors in the near-IR, which is much lower than that of the mid-IR detectors, (usually equal to 0.5) make it inappropriate using the up-converters of long CO_2-laser pulses, in particular, as a part of lidar systems.

The obvious ways to enhance the efficiency of up-conversion are to use more efficient nonlinear crystals of high optical quality, to accomplish the conditions of 90° matching, and to increase the dielectric strength of working surfaces of crystals. It is seen from Fig. 1 that the use of crystals with high quality coefficients in practice means the use of crystals whose shortwave boundary of the transparency spectrum is shifted toward longer waves as compared with the boundary of the $AgGaS_2$ and Ag_3AsSe_3 crystals.

Until recently the potential advantages of such a transition have been ignored because of imperfect technologies of manufacturing high-quality samples of such promising crystals as $ZnGeP_2$. These crystals are characterized by high nonlinear susceptibility d_{14} = 75 pm/V, their quality coefficient is at the third place among all known nonlinear crystals and at the first place among the crystals transparent all over the near IR region from 0.7 to 2.5 μm. All this potentially provides for a six times higher efficiency of the up-conversion than that in $AgGaS_2$ and 140 times higher than that in Ag_3AsS_3. Earlier it was shown[2] that accomplishment of phase matching of the II ($oe \to o$) type in $ZnGeP_2$ with the use of Nd:YAG-laser radiation as a converting one provides fulfillment of the matching conditions at the matching angles close to 90° (Ref. 2). This lifts the restriction on the length of crystals used and allows optimal focusing of radiation to be achieved. Nevertheless, the first two attempts to use $ZnGeP_2$ crystals for up-conversion of CO_2-laser radiation failed. The up-conversion of a continuous-wave CO_2-laser radiation demonstrated in 1971 allowed only to record this effect,[2] whereas in 1979 the up-conversion performed with the use of a nanosecond Nd:YAG laser yielded only 5% efficiency.[3] This was caused by low optical quality of the single crystals used. In the region of generation of CO_2 lasers, the optical loss factor α achieved 1.0 to 2.0 cm^{-1}, and at the wavelengths of a Nd:YAG laser (λ_1 = 1.064 μm) and up-converted radiation (λ_3 = 0.9548 μm) it was from 10–20 to 50 cm^{-1}. Advances in the development of solid-state matrix high-sensitive image visualizers also was a factor in the loss of interest in up-conversion.

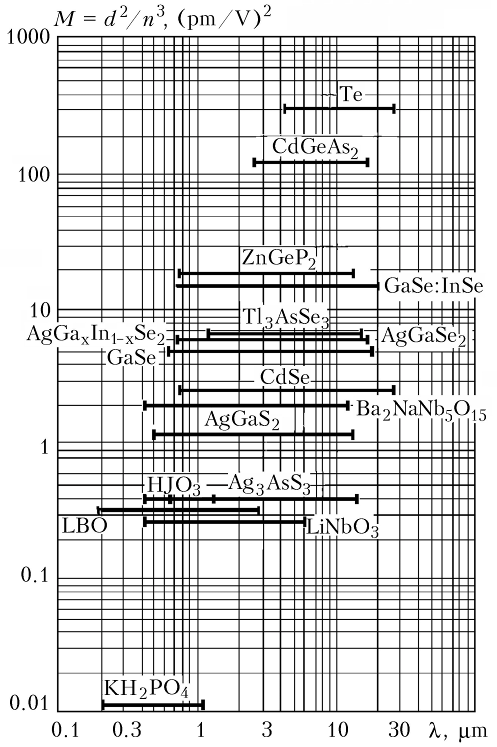

Fig. 1. Quality coefficients and regions of spectral transparency of different nonlinear crystals.

In our opinion, now the situation with the use and development of up-converters of long CO$_2$-laser pulses has changed because of two new circumstances.

The first circumstance is connected with the technological achievements in manufacturing of high-quality nonlinear crystals suitable for the up-conversion of CO$_2$-laser radiation into the near IR region. These crystals include, first of all, ZnGeP$_2$. As was reported in Ref. 4, the optical quality of ZnGeP$_2$ single crystals we have managed to manufacture using a modernized growing technology is characterized by the absorption coefficient $\alpha = 0.01$ cm^{-1} in the window of maximum transparency. The advances in the post growth processing allowed it to be decreased down to 2.5–2.0 cm^{-1} in the region $\lambda = 0.95$–1.06 μm.

The second circumstance is that recently developed uncooled detectors of the near-IR radiation based on Ge and A$_3$B$_5$ crystals and InGaAs and AgGaAsP compounds have shifted the long-wave boundary of the detector sensitivity to 1.6–1.7 μm. The quantum efficiency of signal recording for these detectors exceeds 50% in the region $\lambda = 0.84$ μm, equals roughly 30% at the sum frequencies of the CO$_2$ and Nd:YAG lasers, and exceeds 15-% level at $\lambda = 1.3$ μm and 10-% at $\lambda = 1.55$ μm with the capability of operation in the photon counting mode in all these cases. The development of a photomultiplier tube (PMT) with a photocathode having the quantum efficiency of 0.1% at the wavelength of 1.6 μm was also reported.[5,6] These detectors have sensitive areas of 0.1 to 0.5 mm in size, what is optimal for lidar systems and provides the minimum noise equivalent power (NEP). For a comparison let us note that the best PMT's with the photocathode of S–1 type, for example, FEU–83 model, have the quantum efficiency about 10^{-6}% at $\lambda = 1.3$ μm, 10^{-3}% at $\lambda = 1.06$ μm, and 0.1% at $\lambda = 0.958$ μm.

This paper is devoted to the experimental study of the feasibility of creating high-efficiency up-converters of long CO$_2$-laser pulses by mixing CO$_2$-laser radiation with radiation of Nd:YAG lasers in ZnGeP$_2$ single crystals grown using a modernized technology and subjected to post growth processing in order to improve its optical quality and to enhance the dielectric strength of its working surfaces.

Generally speaking, the upward frequency conversions of CO$_2$-laser radiation in ZnGeP$_2$ can be accomplished by sum or difference frequency generation of both type I ($e + e \to o$; $o - e \to e$) and type II ($e + o \to o$; $o - e \to o$) of three-frequency interactions. The law of conversion of energy states that the relation $1/\lambda_1 \pm 1/\lambda_2 = 1/\lambda_3$ must be fulfilled. Then the converted radiation has the wavelengths:

$$\lambda_3 = \frac{\lambda_1 \lambda_2}{\lambda_1 + \lambda_2} \quad \text{and} \quad \lambda_3 = \frac{\lambda_1 \lambda_2}{\lambda_1 - \lambda_2}\,. \tag{1}$$

The maximum conversion efficiency is achieved if the phase matching conditions are fulfilled. In the vector form these conditions can be written as:

$$\mathbf{k}_3 = \mathbf{k}_1 + \mathbf{k}_2, \quad \mathbf{k}_3 = \mathbf{k}_1 - \mathbf{k}_2, \tag{2}$$

where $\mathbf{k}_i$ are the wave vectors of the pump radiation ($i = 1$), CO$_2$-laser radiation ($i = 2$), and resulting radiation ($i = 3$). Actually, they express the law of conservation of momentum. In the scalar form for sum frequency generation of the type II in positive crystals we have from Eq. (2):

$$\frac{n_1^o\, n_1^e}{\lambda_1 \sqrt{(n_1^o \sin\theta)^2 + (n_1^e \cos\theta)^2}} + \frac{n_2^o}{\lambda_2} = n_3^o\, \frac{\lambda_1 + \lambda_2}{\lambda_1 \lambda_2}\,. \tag{3}$$

where $n_i^{o,e}$ are the main refractive indices for the ith o- and e-waves, respectively, determined by the Selmeier dispersion relations. Our experience shows that the best agreement with the experimental results can be obtained by estimating the refractive indices from the Selmeier relations:

$$(n^{o,e})^2 = A^{o,e} + B^{o,e}\lambda^2/(\lambda^2 - C^{o,e}) + \\ + D^{o,e}\lambda^2/(\lambda^2 - E^{o,e}), \tag{4}$$

where λ is in μm. The corresponding constants are given in Table 1. All other versions of the constants, we found in the literature, gave physically unrealistic results, most probably, because of the well known incorrectness of the Selmeier constants in the shortwave part of the transparency spectrum.[7]

Matching curves calculated with the use of the data from the Table are shown in Figs. 2*a* and *c*. As follows from the curves, almost all spectrum of CO_2-laser radiation can be up-converted by sum or difference frequency generation by mixing with the Nd:YAG-laser radiation ($\lambda = 1.064\ \mu m$) under conditions of 90° matching. Besides, most intense parts of the 9 and $10\ \mu m$ bands of CO_2-laser radiation can be up-converted with of the conditions of 90° matching fulfilled by use of type II interaction, that is, under optimal conditions. Among transformations under the type I interaction, difference frequency generation must be more efficient than sum frequency generation, and it is characterized by a wider spectral and more narrow angular matching widths. The temperature dependence of phase matching conditions at sum frequency generation in a crystal at room temperature can be determined as:

$$\frac{\partial\theta}{\partial T} \approx \frac{1}{\sin\,(2\theta)\,(n_1^e - n_1^o)} \times$$

$$\times \left\{\frac{\lambda_1}{\lambda_3}\frac{\partial n_3^o}{\partial T} - \frac{\lambda_1}{\lambda_2}\frac{\partial n_2^o}{\partial T} - \frac{\partial n_1^o}{\partial T} - \frac{f(n_1^e - n_1^o)}{fT}\sin^2\theta\right\}. \quad (5)$$

Upon substitution of the corresponding values we obtain $\partial\theta/\partial T = -0.006$ deg/K. As is seen from Figs. 2*a* and *c*, this value is not a constant all over the entire temperature range, but it decreases markedly at the temperature below 200 and above 400 K. In the latter case the birefringence gradient even changes its sign. The up-conversion efficiency for plane interacting waves in the approximation of a preset field can be determined as

$$\eta = \frac{I_3}{I_1} = \frac{8\pi^2\, d_{\mathrm{eff}}^2\, L^2\, I_2}{n_1\, n_2\, n_3\, c\, \varepsilon_0\, \lambda_3^2}\,\sin c^2\left(\frac{|\Delta k|\,L}{2}\right) \approx$$

$$\approx \frac{2.974 \cdot 10^4\, M_{\mathrm{eff}}\, L^2\, I_2}{\lambda_3^2}, \quad (6)$$

where I_1 and I_2 are the intensities of the pump waves; L is the crystal length; d_{eff} is the effective coefficient of nonlinear optical susceptibility of the second order, which has the form $d_{\mathrm{eff}} = (d_{14} + d_{36}) \sin\theta \cos\theta \cos 2\varphi \approx \approx d_{14} \sin 2\theta \cos 2\varphi$ and $d_{\mathrm{eff}} = -d_{36} \sin\theta \sin 2\varphi$, respectively, for the interactions of type I and type II. Here $M_{\mathrm{eff}} = d_{\mathrm{eff}}^2/(n_1\, n_2\, n_3)$ is the actual or effective value of the quality coefficient. The values of M for interactions of type I and type II in $ZnGeP_2$ at different crystal temperature are shown in Figs. 2*b* and *d*. Equation (6) is valid if the spectral width of the pump radiation does not exceed, at sum frequency generation, the values

$$\Delta\lambda_1 = \frac{\lambda_2^2}{L}\left|\left\{\lambda_3\frac{fn_3^o}{f\lambda_3} - \lambda_1\frac{fn_1^o}{f\lambda_1} - n_3^o + n_1^o\right\}^{-1}\right|, \quad (7)$$

where $\partial n/\partial\lambda$ is found from the Selmeier relations. Besides, the pump radiation must fall within the external field of view of a crystal

$$\Delta\theta = 2\sqrt{\frac{2\,\lambda_2}{L\, n_2^e\,|1 - (n_2^e/n_2^o)^2|}}. \quad (8)$$

If these conditions are fulfilled, the conversion efficiency, at sum frequency generation, in our case can be easily estimated as:

$$\eta = 1.8 \cdot 10^{-6}\, I_2. \quad (9)$$

Here I_2 is in MW/cm^2. It follows from Eq. (9) that we can expect the efficiency of several per cent, at the pre-breakdown pump intensity.

Table 1. Constants of Selmeier relations for $ZnGeP_2$ crystals at temperature from 100 to 500 K

T, K	Type	A	B	C	D	E
100	*o*	4.49338530	5.10900476	0.12778958	2.16855561	900
	e	4.53789703	5.28559310	0.13390490	2.10759312	900
150	*o*	4.52238331	5.10517584	0.12953514	2.16873708	900
	e	4.56209039	5.28876458	0.13562281	2.10527581	900
200	*o*	4.55320893	5.10583525	0.13153917	2.16893560	900
	e	4.59314629	5.29268202	0.13772522	2.10451540	900
250	*o*	4.58457517	5.11280186	0.13378216	2.16914685	900
	e	4.62177621	5.30603357	0.13993953	2.10214323	900
300	*o*	4.61511259	5.12797577	0.13623774	2.16936050	900
	e	4.69874418	5.27924444	0.14339365	2.09861247	900
350	*o*	4.64099213	5.14791199	0.13856847	2.16952994	900
	e	5.13295051	4.89674401	0.15576712	2.10317249	900
400	*o*	4.66484235	5.17782620	0.14107930	2.16968693	900
	e	5.09042780	4.99794548	0.15680542	2.09884054	900
450	*o*	4.68535622	5.21956368	0.14373622	2.16981674	900
	e	5.05767976	5.09918075	0.15816346	2.10218698	900
500	*o*	4.70134287	5.27485500	0.14650344	2.16991032	900
	e	5.19415933	5.04078045	0.16466963	2.09668743	900

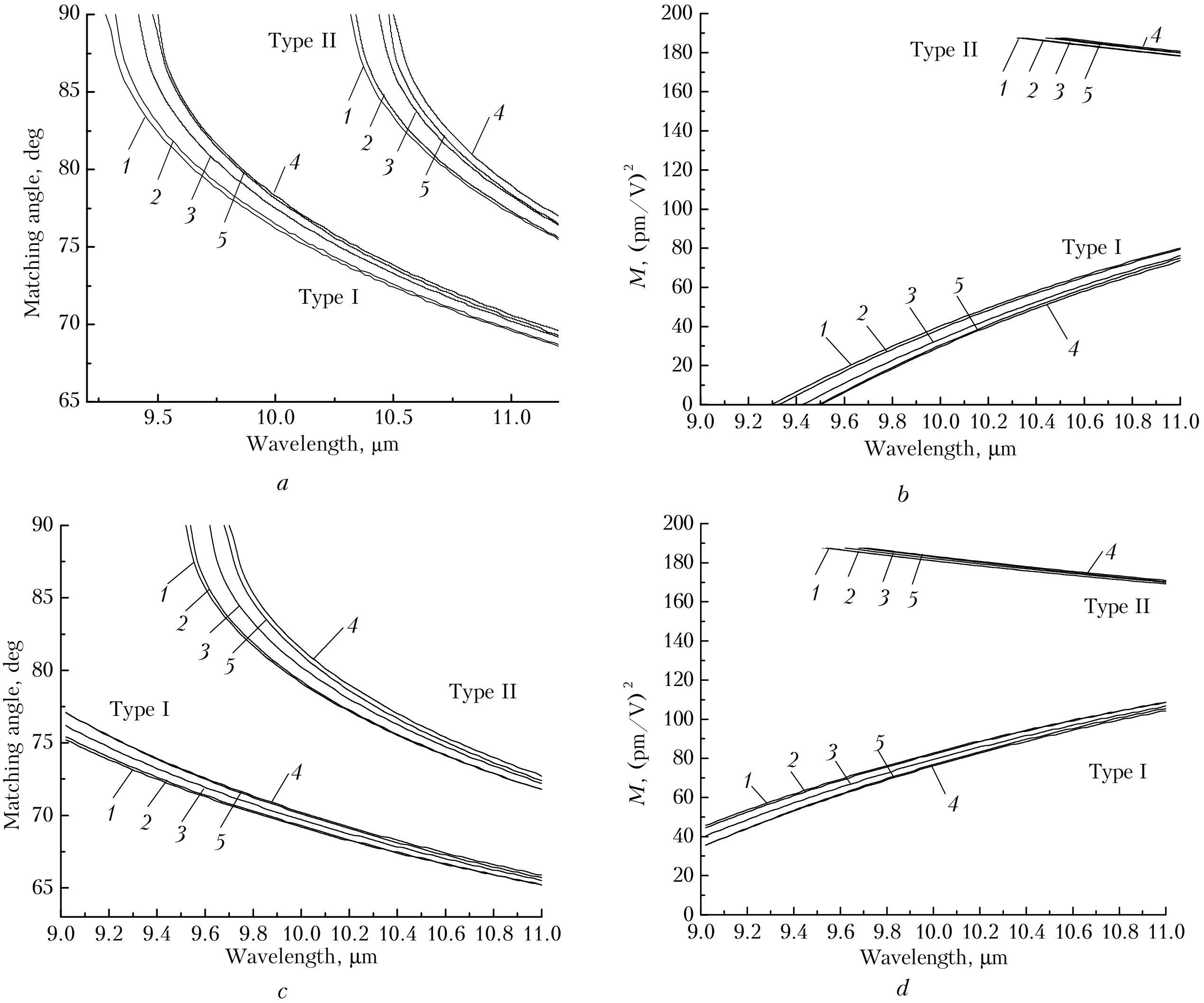

Fig. 2. Matching curves for sum (*a*) and difference (*c*) frequency generation of Nd:YAG-laser (λ_1 = 1.064 μm) and CO_2-laser (λ_2 = 9.2–10.8 μm) radiation in $ZnGeP_2$ crystals and corresponding values of the effective quality coefficients (*b* and *d*) at different crystal temperatures of 100 (*1*), 200 (*2*), 300 (*3*), 400 (*4*), and 500 K (*5*).

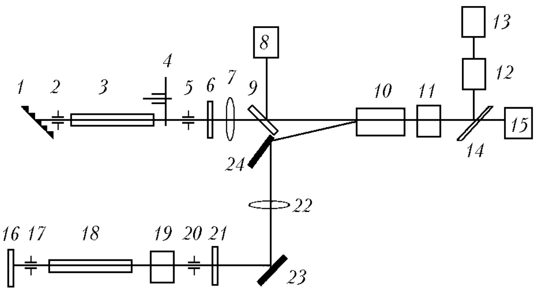

Fig. 3. Up-converter of CO_2-laser radiation: CO_2 laser: diffraction grating *1*, diaphragms *2* and *5*, active element *3*, and output mirror *6*; modulator *4*, antireflection coated focusing lenses from GaAs *7* and *22*, three-frequency LG-126 He–Ne laser *8*, beam splitters *9* and *14*, $ZnGeP_2$ crystal *10*, three bandpass interference filters *11*, oscilloscope *12*, FEU-83 PMT *13*, pyroelectric power and energy meter *15*; Nd:YAG laser: totally reflecting mirror *16*, diaphragms *17* and *20*, active element *18*, acoustooptical Q-switch *19*, output mirror *21*; beam-folding mirrors *23* and *24*.

The experimental setup is shown schematically in Fig. 3. Long pulses were generated by CO_2 lasers of two types: a modernized serial cw LG–704 laser and home-assembled TEA CO_2 laser. Both of the lasers were passively stabilized by being mounted on a rigid Invar frame. The cw laser radiation was amplitude-modulated inside or outside a cavity with a chopper at a frequency from 10 to 200 Hz. Output radiation pulses 1 to 20 μs long had the peak power from 10 W to 3 kW.

The mode structure of radiation was formed with irises, magnetostriction frequency trim, and the discharge current regulation. In this case, the regime of generation of a single longitudinal TEM_{00} mode with a beam about 5 mm in diameter was readily obtained. In the second case, the TEA CO_2 laser was used, which provided pulses with the leading peak of 250 ns duration, which is usual for lasers of this type, and a long (3–5 μs) tail. The peak output power was respectively 2.4–3.0 and 0.4–0.8 MW.

The CO_2-laser radiation was attenuated to the milliwatt level and microwatt with a set of plane-parallel CaF_2 plates. The commercially available Nd:YAG laser equipped with an acoustooptical switch provided matching of output pulses in their time shape and beam size. Its peak power varied from 0.2 to 3.0 MW. Technological advances allowed us to manufacture an optical element of $ZnGeP_2$ crystal with the optical loss factor $\alpha = 0.59$ cm^{-1} at $\lambda = 10.513$ μm (10P(12) line of a CO_2 laser), $\alpha = 2.0$ and 2.5 cm^{-1} at $\lambda = 1.064$ (Nd:YAG laser) and $\lambda = 0.9548$ μm (up-converted radiation), respectively, at the thickness of 3.9 mm and orientation $\theta = 85°$, $\varphi = 45°$.

Figure 4 shows the short-wave part of the transmission spectrum of two crystals after successful post growth annealing (curves *1* and *2*). This fragment indicates repeatability of the obtained result. Curve *1* corresponds to the crystal used in our experiment.

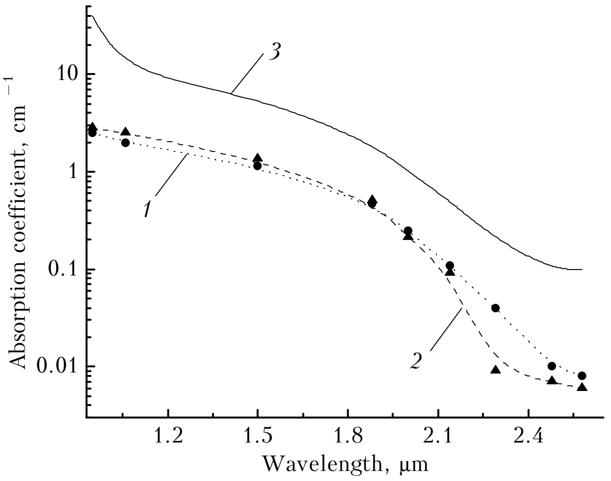

Fig. 4. Short-wave part of the spectral dependence of the absorption coefficients of two high-quality $ZnGeP_2$ crystals subjected to additional post growth annealing (*1*, *2*) and crystal typical for the early 80's–90's (*3*). Curve *1* corresponds to the crystal used in our experiment.

For a comparison, this figure also shows curve *3*, which characterizes typical crystals grown in the early 80's–90's. The up-converted radiation was selected by three bandpass interference filters with $T = 47\%$ for $\lambda < 0.96$ μm and $T = 1.7 \cdot 10^{-5}\%$ at $\lambda = 1.064$ μm. It was measured by the PVDTs pyroelectric power and energy meter, and the time shape of pulses was observed with a pre-selected FEU–83 PMT with a photocathode of S–1 type (Ag–O–Cs) having the quantum efficiency of $2 \cdot 10^{-3}\%$ at $\lambda = 1.064$ μm and 0.1% at the converted radiation wavelength $\lambda = 0.958$ μm. By slightly focusing the beam its diameter was decreased by 1.5–2 times in the case of conversion of TEA CO_2-laser radiation and by 2.5–3.0 times in the case of up-conversion of radiation of a Q-switched CO_2 laser. The maximum up-conversion efficiency at the peak of power about 0.1% in the stable regime and almost 0.2% at the appearance of signs of the crystal destruction during a whole working day was achieved for TEA CO_2-laser pulses with the total duration of 3 μs. The same efficiency was also observed at the up-conversion of pulses of the CO_2 lasers of the second type. However, up to 3 fourth of power of Nd:YAG-laser pulses was lost at matching the cross sections of interacting beams and for preventing the breakdown of the working surfaces of crystals.

The principal utility of the use of such an up-converter for recording signals of lidar type, without detailed consideration of all perturbing factors, can be deduced from the comparison of the noise equivalent power of a lidar as a measuring system with the use of direct detectors of CO_2-laser radiation and an up-converter with a near-IR detector. One should keep in mind that the main source of noise in detection of actual lidar returns with the up-conversion of the recorded radiation by sum frequency generation is the background radiation. This noise can be suppressed by the use of cooled diaphragms.[8] In that case, as in ours, the noise of photodetectors N(det) remains the main source of noise. Advantages of the considered methods of signal recording can be evaluated as follows.

The detectability D of the photosensitive material of the MCT photodiodes operating at the CO_2-laser wavelengths is equal to $2 \cdot 10^{11}$ cm · Hz$^{1/2}$ · W^{-1} (Refs. 6 and 8). When using an MCT photodiode with a photosensitive area S having the diameter $d = 0.5$ mm, which is typical of lidar systems, the corresponding NEP(MCT) $= \sqrt{S}/D$ is equal to $2.2 \cdot 10^{-12}$ W/Hz$^{1/2}$. For an earlier modification of the FEU–83 PMT (produced in the USSR), the PCAC31034C PMT produced by the PCA Company with an InGaAs photocathode, and a C30902S thick Si avalanche photodiode produced by the EG&G, having the same diameter of the photosensitive area (0.5 mm) and suitable for recording the up-converted radiation at the wavelength of 0.958 μm, NEP(det) is respectively (4, 2.5, and 1.9) $\cdot 10^{-16}$ W/Hz$^{1/2}$, according to specifications.[6,8] In the latter case, we consider the diode operating in the photon counting mode with the dark current count rate of 10^4 pulses per second at the operating power supply voltage of only 20 V. The NEP (in W) at the output of the two above-mentioned systems can be found taking into account the quantum efficiency of the detector η and up-conversion η_{up}, as well the pass band Δf and the noise equivalent power of the detector itself by the equation

$$\text{NEP} = \text{NEP(Det)}\,\sqrt{\Delta f}/(\eta\ \eta_{up}). \qquad (10)$$

The quantum efficiency of the MCT photodiode at the wavelength of 0.958 μm is 0.5, whereas for the other three of the above-mentioned detectors it is 0.002, 0.005, and about 0.3, respectively.[8] All the above-listed detectors have the pass band no less than 10 MHz, what provides the recording of lidar returns with the spatial resolution from the minimum of 50 m to quite suitable values of 150–450 m, which are typical for gas analyzers and lidars operating in the near infrared. Then the noise equivalent power at the output electronic system matched by the 10-MHz pass band is equal to $2.9 \cdot 10^{-9}$ W for the case of direct recording of signals with an MCT diode and $(12.8, 2.6, \text{and } 1.9) \cdot 10^{-13}$ W, for the recording system with up-conversion of CO_2-laser radiation into the near IR region and recording of the up-converted signals with the use of the other of the above listed detectors operating in the near IR region. Taking into account their quantum efficiency, at the up-conversion efficiency of 1%, we finally have the following values of NEP: $1.4 \cdot 10^{-8}$, $6.4 \cdot 10^{-8}$, $5.2 \cdot 10^{-9}$, and $6.3 \cdot 10^{-12}$, respectively. The use of FEU–83 useless, since we have a 4.6 times loss in the signal-to-noise ratio of a lidar as compared with the direct detection. However, using the RCAC31034C PMTs and the Si avalanche photodiode operating in the photon counting mode, we can obtain a 37 times and 450 times benefit in the signal-to-noise ratio, respectively. The further increase of the up-conversion efficiency in $ZnGeP_2$ is possible, if the losses at $\lambda = 0.9548$ μm are decreased down to the desirable level ≤ 0.1 cm^{-1} due to improvement of the technology of high-quality crystal growth. In recent time, foreign technologies already allowed obtaining crystals with the loss about 1 cm^{-1} (Ref. 9).

Technological advances in the growth and in the post growth processing of $ZnGeP_2$ crystals allowed the optical loss factor in the region of 0.95–1.1 μm to be decreased down to the level of 2.5–2.0 cm^{-1}, and the use of such crystals for up-conversion of long CO_2-laser pulses by mixing them with Nd:YAG-laser radiation ($\lambda = 1.064$ μm) allowed the conversion efficiency from 1 to 2% to be achieved. In particular, at the sum frequency generation the conversion efficiency of 1% was obtained in the stable regime. The achieved efficiency of the up-conversion of long (3–50 μs) CO_2-laser pulses makes urgent the study of applicability of up-converters and existing near-IR detectors, including current IR PMT's, in recording systems of CO_2-laser-based lidars. The benefit in the signal-to-noise ratio may reach 450 times when using current avalanche diodes operating in the photon counting mode. It is also worth studying the possibilities of application of new nonlinear crystals, for example, $HgGa_2S_4$, and alloyed and mixed crystals like GaSe:In and GaSe:InSe, for the up-conversion of CO_2-laser radiation.

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