Vol. A52 (1977) *ACTA PHYSICA POLONICA* No 1

PARAMETRIC UPCONVERSION OF IR-RADIATION IN LiJO$_3$ WITH PULSED RUBY- AND CW-ARGON-LASERS

By D. Malz, K. Schindler and J. Bergmann

Sektion Physik, Friedrich-Schiller-Universität, Jena*

(Received November 12, 1976)

The possibility of using LiJO$_3$-crystals in a nonresonant arrangement for detecting of thermal radiation by optical mixing was investigated. By means of Q-switched ruby- and cw-argon lasers (pump lasers) the ir-photon conversion efficiencies (quantum efficiencies) and the phase matching angles are experimentally determined in the range of 1.98 μm $\leqslant \lambda \leqslant$ 2.67 μm. In LiJO$_3$ the stronger absorption sets in for $\lambda > 3$ μm, followed by a small transmission peak near 5 μm. In this range the second harmonic ($\lambda = 5.4$ μm) of a Q-switched C^{13}O$_2$-laser can be upconverted to $\lambda = 0.448$ μm with the help of a cw-argon laser. The possible quantum efficiencies and noise equivalent powers (NEP) are discussed.

In recent years we have investigated phase-matched parametric upconversion of infrared radiation in the birefringent nonlinear crystal LiJO$_3$ [1, 2]. Since radiation detectors and especially imaging systems in the visible region are most sensitive and also have low rise times ($\tau \leqslant 10^{-9}$ sec) as a rule, such a system of parametric upconversion followed by direct detection in the visible region was considered as a possibility for the detection of ir-radiation. Reviews for example are given by Kleinman and Boyd [3], Warner [4] and Fenner Milton [5].

In this paper we report on the sum frequency mixing $\omega_s = \omega_i + \omega_p$, where ω_s is the frequency of the upconverted output photon, ω_p the frequency of the coherent pump laser, and ω_i the frequency of the input ir-photon.

Sum frequency mixing avoids the noise connected with the spontaneous Stokes-fluorescence and the decay of the pump photons. In this paper we wish to give some information on the use of LiJO$_3$ for upconversion of thermal (see also [6]) and coherent ir-radiation in a non-resonant arrangement with the help of pulsed ruby- and cw-argon-lasers. In particular we are interested in upconversion of pulsed coherent ir-radiation in the range where the absorption of LiJO$_3$ begins [$\lambda \approx 5$ μm]. In this region we had to

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[*] Address: Sektion Physik, Friedrich-Schiller-Universität, Max-Wien-Platz 1, 69 Jena, Deutsche Demokratische Republik.

transform the frequency of the second harmonic (SHG) of a Q-switched CO_2 laser into the spectral range of visible light.

Since the parametric upconversion caused by nonlinear electron-photon interaction is a very fast process ($\tau \lesssim 10^{-14}$ s) it becomes possible to transform very short ir-pulses.

1. Optical properties of $LiJO_3$ and ir-power conversion efficiency

$LiJO_3$ is a negative uniaxial crystal of point group 6. Corresponding to measurements of Meisner [7] the second-order polarizability tensor $(\hat{d}_{ik})$ for $LiJO_3$ is given by

$$(\hat{d}_{ik}) = \begin{pmatrix} 0 & 0 & 0 & (0.09) & 1.7 & 0 \\ 0 & 0 & 0 & 1.7 & (-0.09) & 0 \\ 1.7 & 1.7 & 1.3 & 0 & 0 & 0 \end{pmatrix} \cdot 10^{-8}\ \text{esu}$$

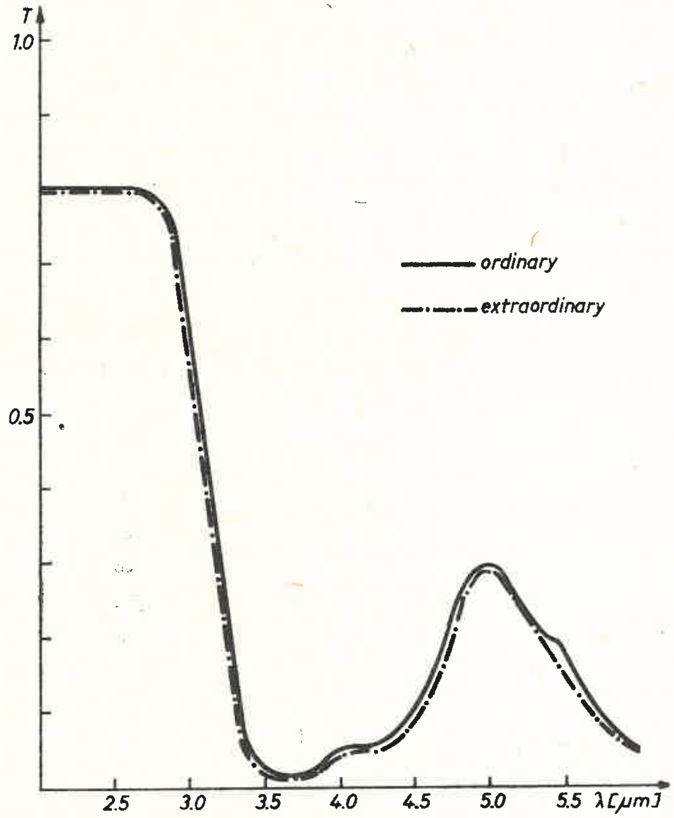

Fig. 1. Optical transmission of $LiJO_3$ ($l = 1.5$ cm)

In our experiments we used nearly collinear critical phase-matching for the waves in all cases. This allowed us to use the negative uniaxial crystal at the phase-matching condition of type I: $(\omega_i)_{ord} + (\omega_p)_{ord} = (\omega_s)_{extraord}$ for the parametric three wave interaction in $LiJO_3$. This interaction type caused an effective nonlinear coefficient which can be written as $d_{\text{eff}} = 2d_{31} \sin \vartheta_m$, where ϑ_m is the phase-matching angle between the optical axis (c) and the direction of the propagating pump beam. Fig. 1 shows the optical transmission of the $LiJO_3$-crystal ($l = 1.5$ cm) in the ir-region. In the visible range the transmission is warranted up to 0.29 μm. The slopes of ordinary and extraordinary polarized light have no essential differences. In Fig. 2 the calculated phase-matching curves for the used ruby ($\lambda = 0.694$ μm) are plotted and argon ($\lambda = 0.488$ μm) pump lasers. Fig. 3 shows the dispersion of $LiJO_3$.

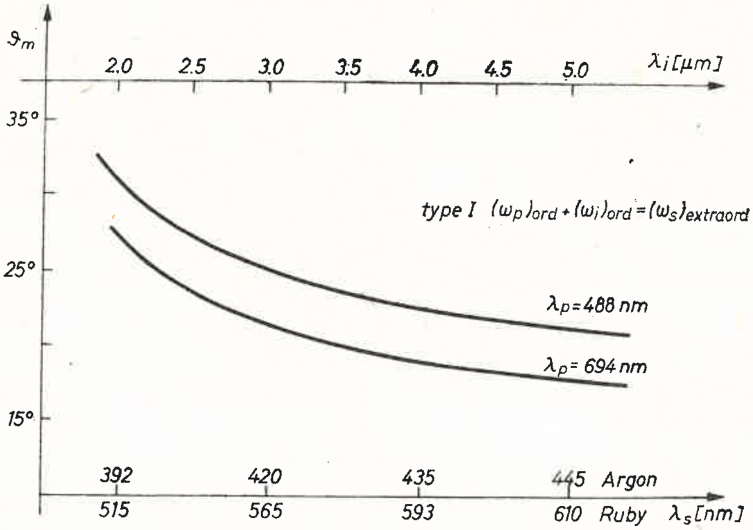

Fig. 2. Calculated phase-matching curves for ruby- and argon-laser in $LiJO_3$

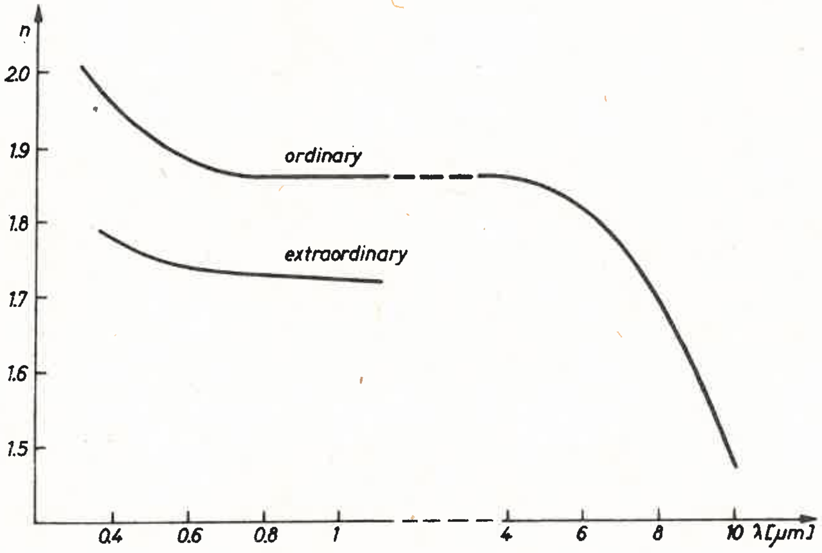

Fig. 3. Dispersion of $LiJO_3$

1.2. Quantum conversion efficiency and bandwidth

For collinear planewave interactions in a nonabsorbing, nonlinear dielectric crystal the instantaneous ir-power efficiency inside an upconverter material in the case of phase-matching and with neglecting of pump power deplation can be expressed as

$$\eta = \frac{L(\omega_s)}{L(\omega_i)} = \frac{\omega_s}{\omega_i} \sin^2 \left[\frac{2\pi}{c} d_{\text{eff}} \sqrt{\frac{\omega_s \cdot \omega_i}{n_s n_i}} \sqrt{\frac{8\pi L(\omega_p)}{c n_p A}}\, l \right], \tag{1}$$

where $L(\omega)$ is the radiation power at the frequency ω (watts), $L(\omega_p)/A$ — the instantaneous pump power density (watts $\cdot$ cm^{-2}), l — effective interaction length in the nonlinear material, n_s, n_i, n_p are refractive indices at ω_s, ω_i, ω_p, respectively.

The quantum efficiency is than defined as $N = \eta \dfrac{\omega_i}{\omega_s}$. To maximize conversion efficiency reasonably sized crystals and large pump power densities are needed (10^6 watts $\cdot$ cm^{-2} $\lesssim L(\omega_p)/A \lesssim 10^9$ watts $\cdot$ cm^{-2}). We also experimentally studied the case of the beginning of absorption (d_i) for small conversion of ir-power and slight pump power deplation $\left(\dfrac{dA_p}{dz} \approx \dfrac{dA_i}{dz} \approx 0\right)$. Therefore for Gaussian profiles $A_j \sim e^{-r^2/W_j{}^2}$ of interacting laser beams and for walkoff angle β_s (angle between the Poynting vector and the wave vector k_s of the signal wave ω_s in an anisotropic optical medium) we have as a sufficient approximation

$$\eta = \frac{32\pi^2\omega_s^2}{n_s \cdot n_p \cdot n_{ir} c^3} \cdot \frac{W_s^2}{W_p^2 \cdot W_i^2} \cdot d_{\text{eff}} \frac{L(\omega_p)}{\cos^4 \beta_s} \cdot \frac{1}{|\alpha_i|^2} \{e^{-\alpha_i z} - 1\}^2, \tag{2}$$

where W_j is the Gaussian beam radius for ω_j (j is s, p, i, respectively). If we wish to know the extent of the upconverted infrared passband $\delta\omega_i$, we must take into consideration the phase-matching condition for the wave vectors $|\Delta k| = |k_s - k_p - k_i| \leqslant 2\pi/z$, where z = interaction length. Since for a monochromatic pump wave $\delta\omega_i = \delta\omega_s$, we have

$$\delta\omega_s \leqslant 2\pi/z \left| \frac{\partial(\Delta k)}{\partial \omega_i} \right|^{-1}_{\Delta k = 0} \tag{3}$$

with

$$\frac{\partial(\Delta k)}{\partial \omega_i} = \frac{1}{c} \left(n_s - n_i + \frac{\partial n_s}{\partial \omega_s} \cdot \omega_s - \frac{\partial n_i}{\partial \omega_i} \omega_i \right).$$

2. *Experiments*

2. 1. Upconversion of thermal infrared radiation (2 μm $\leqslant \lambda \leqslant$ 3 μm)

The experimental set-up is depicted schematically in Fig. 4. The device is an angle-tuned 1.5 cm-LiJO$_3$ single crystal cut for the phase-matching of 24° to the c-axis and pumped in the first experiment by a Q-switched ruby laser ($\lambda = 0.694$ μm) emitting 25 nsec pulses with peak power densities of about 10^6 watts · cm^{-2}. For signal detecting we used a photomultiplier followed by an 100 MHz oscilloscope.

In the second experiment the mixing crystal is pumped by a cw-argon laser ($\lambda = 0.488$ µm) with a power density of about 7 watts · cm^{-2} within the crystal. The continuously radiating incoherent infrared source is modulated by a chopper for phase-

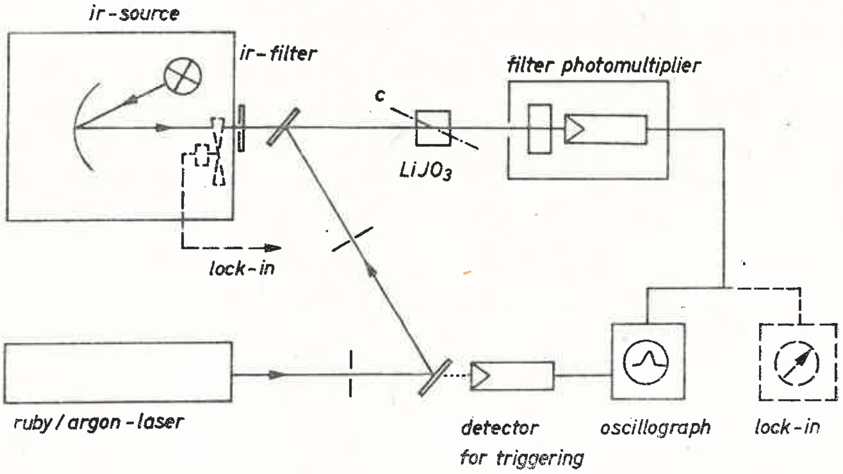

Fig. 4. Experimental setup for up-conversion of thermal ir-radiation

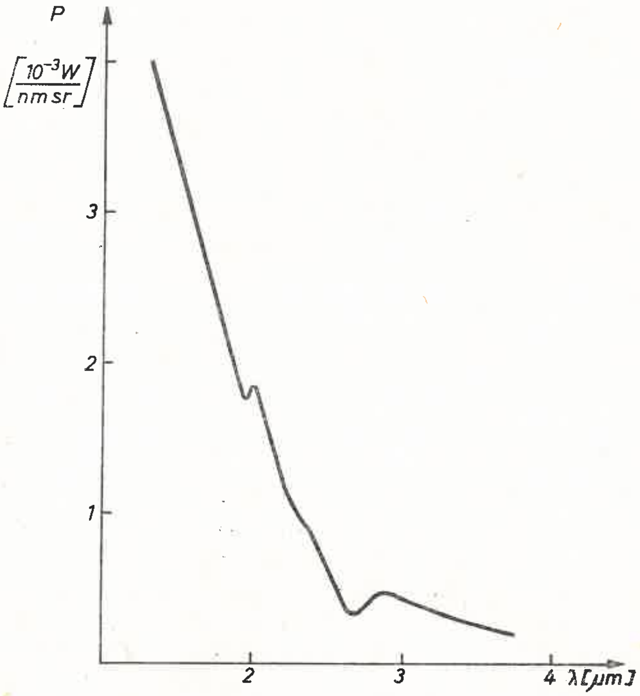

Fig. 5. Radiation power of the thermal ir-source

-sensitive detecting (lock-in amplifier) in this case. The measured spectral ir-radiation power with reference to 1 steradian and a spectral range of 1 nm is plotted in Fig. 5. The upconverted signal radiation is isolated by means of several optical narrow-band filters, which for instance cause a reduction of the ruby laser pulse of about $10^{-15} - 10^{-17}$. These filters, however, also considerably decrease the upconverted ir-signal.

2.2. Upconversion of 5.4 µm SHG-pulses

In Fig. 6 we present the experimental set-up for upconversion of infrared pulses generated by second harmonic generation (SHG) of Q-switched $C^{13}O_2$ laser pulses (10.8 µm) in a tellurium single crystal [8]. By means of a germanium flat and a NaCl-lens the SHG- and argon laser beams are focused into the same volume domain of the $LiJO_3$ crystal in the phase-matching direction. A reflecting diffraction grating is used to separate the signal beam and the argon laser beam. The upconverted SHG-pulse is detected with the help of a photomultiplier and a 60 MHz-oscilloscope.

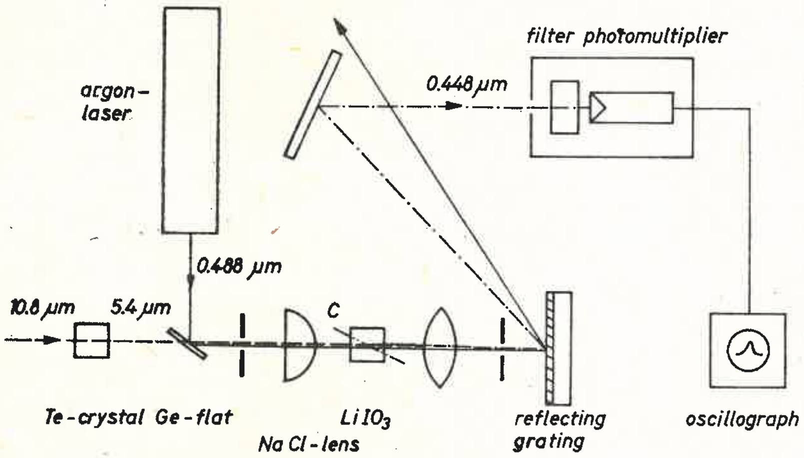

Fig. 6. Experimental setup for up-conversion of ir-pulses

3. *Results, upconversion efficiencies and noise equivalent powers*

3.1. Upconversion of thermal ir-radiation

The infrared spectral range $\Delta\lambda_i$ which can be upconverted is limited by the transmission of optical narrow-band filters ($\Delta\lambda_F$ = resultant spectral bandwidth) placed in front of the detector to filter out any ruby or argon laser radiation. For frequency mixing we have to fulfil the condition $|\Delta k| \leqslant 2\pi/z$ of the three interacting waves for the various, relatively small incidence angles of the pump- and ir-rays entering the crystal. This effects the upconverted spectral range and we get a reduction of $\Delta\lambda_i$, to $\delta\lambda_i$, which approximately corresponds to the value of $\delta\omega_i = \delta\omega_s$ analogous to formula (3). Numerical estimations of $\delta\lambda_i$; were demonstrated in our earlier papers [1, 2]. This selection of angle dependent narrow-band spectral regions out of $\Delta\lambda_i$; and out of the corresponding solid angles and the use of only one polarizing direction leads to a substantial decrease in upconversion efficiency in comparison to the result computed with the help of formula (1).

Because we have uniformly filled the entrance face of the crystal with ir-light, the walkoff angle β_s is negligible. Under these circumstances we derived the following experimental results summarized in Table I and II.

TABLE I

Summary of experimental data

λ_i [μm]	λ_s [μm]	$\vartheta_{m,ex}$ [degrees]	$L(\omega_i)_{in}$ [watts]	$L(\omega_s)_{out}$ [watts]	η_{ex}
Ruby laser ($\lambda_p = 0.694$ μm, $\Delta t = 25$ nsec) $L(\omega_p)/A = 1.1$ MW · cm^{-2}					
1.98	0.514	27.4	$1.2 \cdot 10^{-3}$	$3.3 \cdot 10^{-6}$	$2.8 \cdot 10^{-3}$
2.22	0.528	26.0	$1.4 \cdot 10^{-3}$	$2.9 \cdot 10^{-6}$	$2.1 \cdot 10^{-3}$
2,67	0.550	24.4	$0.9 \cdot 10^{-3}$	$1.3 \cdot 10^{-6}$	$1.4 \cdot 10^{-3}$
argon laser ($\lambda_p = 0.488$ μm, $\Delta t \to \infty$) $L(\omega_p)/A = 7$ watts · cm^{-2}					
2.38	0.405	30.5	$2.4 . 10^{-4}$	$2.3 \cdot 10^{-12}$	$1 \cdot 10^{-8}$

where $L(\omega_i)_{in}$ — ir-radiation power for $\Delta\lambda_i$; and the solid angle used immediately in front of the crystal; $L(\omega_s)_{out}$ — signal radiation power immediately behind the crystal; $\vartheta_{m,ex}$ — measured phase-matching angle.

It was impossible to measure the signal power $L(\omega_s)_{out}$ directly, because the pump- and signal beams for Type I phase-matching travel in the same direction, thus we also have strong losses of signal frequencies by filtering out the pump. By reason of the unfavorable properties of the filters we had loss factors V as follows:

$$V = 135\,(\lambda_s = 0.514\ \mu\text{m}), \quad V = 215\,(\lambda_s = 0.528\ \mu\text{m}),$$
$$V = 450\,(\lambda_s = 0.550\ \mu\text{m}), \quad \text{and} \quad V = 24\,(\lambda_s = 0.405\ \mu\text{m}).$$

For $L(\omega_s)_{out}$ divided by V we derived the following signal-to noise-ratios (S:N) and noise-equivalent powers NEP (W · Hz$^{-1/2}$) in the detecting system (see Table II).

TABLE II

S:N ratios and NEP

λ_i [μm]	S:N	Bandwidth [Hz]	NEP {S:N = 1} [watts · Hz$^{-1/2}$]	NEP {V = 1, $L(\omega_p)_{opt}$}$_{id}$ [watts · Hz$^{-1/2}$]
1.98	800	10^8	$1.5 \cdot 10^{-10}$	$2.2 \cdot 10^{-14}$
2.22	450	10^8	$3.1 \cdot 10^{-10}$	$2.8 \cdot 10^{-14}$
2.68	90	10^8	$1 \cdot 10^{-9}$	$4.4 \cdot 10^{-14}$
2.38 (argon)	64	1	$3.7 \cdot 10^{-6}$	$1.5 \cdot 10^{-9}$

NEP $\{\,\}_{id}$ means that we have a lossless pumpfilter and pump intensities of about 50 MW $\cdot$ cm^{-2}.

It seems impossible to obtain $V \approx 1$, but a factor $V = 10$ is certainly attainable. We, therefore, obtain about 10^{-13} watts $\cdot$ Hz$^{-1/2}$ for NEP$_{\text{min}}$. In our arrangement we failed to suppress the pump radiation completely. Therefore, the S:N-ratio can be enhanced (factor $10^1 - 10^2$). That means that NEP$_{\text{min}}$ may be decreased to $10^{-14} - 10^{-15}$ watts $\cdot$ Hz$^{-1/2}$.

3.2. Upconversion of 5.4 µm-pulses

For monochromatic, nearly collinear phase-matched upconversion it seems possible to obtain a sufficient correlation between the calculated (Eq. (2)) and measured upconversion efficiencies.

The experimental set-up shown in Fig. 6 was used to verify the predictions of the analysis. We used the following measurement conditions: Power of the argon laser: 70 mW, $\alpha_i \approx 0.6$ cm^{-1}, averaged argon laser intensity in the beam focal spot size

$$\left(L(\omega_p) \cdot \frac{W_s^2}{W_p^2 \cdot W_i^2}\right) = 35 \text{ watts} \cdot \text{cm}^{-2},\ l = 1.5 \text{ cm},\ \cos\beta_s \approx 1\ (\beta_s \approx 2^\circ).$$

Therefore, we get $\eta = 3.6 \cdot 10^{-6}$.

Table III shows the experimental data obtained for an ir-pulse power of 10^{-1} watts and a pulse width of 350 nsec.

TABLE III

λ_i [µm]	λ_s [µm]	$\vartheta_{m,\text{ex}}$ [degrees]	$L(\omega_i)_{\text{in}}$ [watts]	$L(\omega_s)_{\text{out}}$ [watts]	η_{ex}
5.4	0.448	(21 ± 0.5)	10^{-1}	$1.7 \cdot 10^{-7}$	$1.7 \cdot 10^{-6}$

Caused by the pumplight filtering system, the loss at becomes $V = 50$. Thus, the photomultiplier detects only $3.4 \cdot 10^{-9}$ W signal power with an output magnitude of 250 mV for a noise level of about 10 mV at 60 MHz bandwidth.

By this simple arrangement the minimal detectable ir-power $L(\omega_i)_{\text{min}}$ is given by $4 \cdot 10^{-3}$ W for 60 MHz.

The possibility exists to increase the pump intensity under simultaneous reduction of the losses V. Therefore by a minimal detectable ir-power of about $10^{-4} - 4 \cdot 10^{-5}$ watts for 60 MHz bandwidth seems to be a realistic value in our arrangement. The pertinent NEP is than given by $(1.3 - 0.5) \cdot 10^{-8}$ watts $\cdot$ Hz$^{-1/2}$.

In comparision to the use of infrared detectors based on the semiconductor photoconductive effect we can

1. avoid the cooling of the detector,
2. upconvert the true pulse shape of very short infrared pulses connected with the use of sensitive high-speed pulse techniques in the visible spectral region.

In spite of the small upconversion ir-power efficiencies for cw-laser in comparison to the use of Q-switched pump laser, we have here the advantage that we do not need any synchronization of the interacting pulses.

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