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Subnanosecond Pulse Measurements of 10.6 μm Radiation with Tellurium

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Abstract. Subnanosecond infrared pulses have been measured by noncollinear second-harmonic generation in tellurium. The method is very practical because due to the high refractive index the fine tuning of the phase matching is easily obtained by rotating the crystal around the optic axis.

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Second-harmonic generation has proved to be a very effective technique to measure mode-locked pulses. In principle, a short pulse is split into two pulses which pass collinearly or noncollinearly through a nonlinear crystal. The second-harmonic signal depends on the overlap of the two pulses in the crystal. By varying the time delay between these pulses a correlation function of the pulse envelope by means of second-harmonic generation can be obtained. To obtain background-free measurement of the correlation function it is essential that phase-matched second-harmonic generation is produced only when the fields of both pulses are present i.e. when the two pulses overlap in time. This technique is well known for the visible region of the spectrum [1]. However, in the infrared region it has not been explored in a comparative way, mainly due to the lack of optical material of sufficient quality. Collinear measurement of CO_2 laser radiation has been described in the past using proustite or GaAs as the nonlinear material [2, 3]. The presence of background radiation limits in this method the accurate measurement of the energy in the wings of the pulses.

In the present paper we will describe the noncollinear method for phase matching in a tellurium crystal to measure short pulses in the 10 μm region.

The second-harmonic radiation intensity, proportional only to the product of the two fundamental beam intensities, is detected in this method at an angle bisecting the angle between the two fundamental beams. Because of the different direction of the second harmonic radiation no filtering of the detected signal is required.

Although the non-collinear method may seem experimentally complicated because of the accurate adjustments of both the angle between the fundamental beams and their bisector along the y-axis it turns out that due to the high refractive index of tellurium the rotation of the crystal around the z-axis allows an accurate tuning of the phase match conditions.

Phase Matching of Noncollinear Beams

We consider the fundamental beams as two extraordinary waves of which the propagation vectors make the angles ψ and $\pi-\psi$ with the z (optic) axis of the uniaxial Te-crystal. The k vectors of the propagating waves are in the $z-r$ plane where the r-direction makes an angle θ with the positive x axis of the crystal, as shown in Fig. 1. Thus we have the following two fundamental waves

$$\mathbf{E}_1(z,r,t)=\tfrac{1}{2}(\mathbf{E}_1\exp[\mathrm{i}(\omega t-zk_e\cos\psi -rk_e\sin\psi)]+\mathrm{c.c.}), \quad (1)$$

$$\mathbf{E}_2(z,r,t)=\tfrac{1}{2}(\mathbf{E}_2\exp[\mathrm{i}(\omega t+zk_e\cos\psi -rk_e\sin\psi)]+\mathrm{c.c.}), \quad (2)$$

where k_e is the propagation constant of the extraordinary wave at the angle ψ. The field components in the

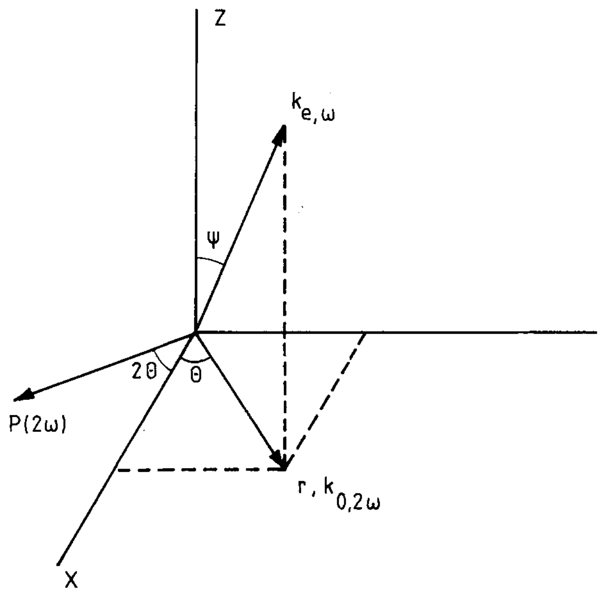

Fig. 1. Orientation of the propagation vectors of the fundamental and second-harmonic beams, the nonlinear polarization and their angles with respect to the crystal coordinates

Cartesian coordinates of the crystal are

$$\mathbf{E}_1=\begin{bmatrix}E_1\cos\psi^*\cos\theta\\E_1\cos\psi^*\sin\theta\\-E_1\sin\psi^*\end{bmatrix}\quad\text{and } \mathbf{E}_2=\begin{bmatrix}E_2\cos\psi^*\cos\theta\\E_2\cos\psi^*\sin\theta\\E_2\sin\psi^*\end{bmatrix}$$

respectively, where ψ^* and ψ differ slightly because in general $\mathbf{E}$ is not perpendicular to $\mathbf{k}_e$.

The trigonal Te crystal of point group 32 has the following non-zero matrix elements in the nonlinear susceptibility tensor: $d_{11}, d_{12}, d_{14}, d_{25}$, and d_{26} with the relations $d_{11}=-d_{12}=-d_{26}$ and $d_{14}=-d_{25}$ [4, 5].

The elements d_{14} and d_{25} are relatively small and will be neglected.

The nonlinear polarization at the frequency 2ω has the components

$$P_x=\tfrac{1}{2}d_{11}E_1E_2\cos^2\psi^*\cos^2\theta\exp[\mathrm{i}(2\omega t-2rk_e\sin\psi)], \tag{3}$$

$$-\tfrac{1}{2}d_{11}E_1E_2\cos^2\psi^*\sin^2\theta\exp \times[\mathrm{i}(2\omega t-2rk_e\sin\psi)]+\text{c.c.}, \tag{4}$$

$$P_y=-d_{11}E_1E_2\cos^2\psi^*\cos\theta\sin\theta\exp \times[\mathrm{i}(2\omega t-2rk_e\sin\psi)]+\text{c.c.}, \tag{5}$$

$$P_z=0, \tag{6}$$

where the non-phase matched terms have been left out, or

$$P_x=\tfrac{1}{2}d_{11}E_1E_2\cos^2\psi^*\cos2\theta\exp \times[\mathrm{i}(2\omega t-2rk_e\sin\psi)]+\text{c.c.}, \tag{7}$$

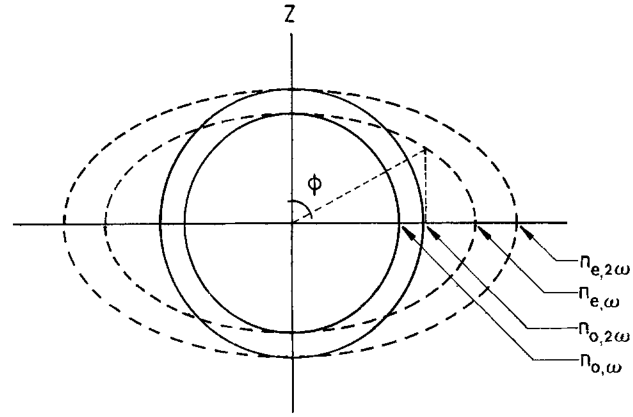

Fig. 2. Schematical representation of the ordinary and extraordinary indices of refraction in a uniaxial crystal. The values are given by the distance between the origin and the intersection between the considered ellipse or circle and the straight line defining the angle of propagation with respect to the z axis

$$P_y=-\tfrac{1}{2}d_{11}E_1E_2\cos^2\psi^*\sin2\theta\exp \times[\mathrm{i}(2\omega t-2rk_2\sin\psi)]+\text{c.c.}, \tag{8}$$

$$P_z=0. \tag{9}$$

It is seen that the polarization $P(2\omega)$ makes an angle -2θ with the x-axis. The absolute value of $P(2\omega)$ is independent on θ. However only that part of $P(2\omega)$ contributes to second-harmonic power that has the ordinary direction and thus lies in the $x-y$ plane and is perpendicular to r. So the production goes with $\sin3\theta$. Maximum production will occur for $\theta=\pi/6$ or $\pi/2$. Thus for $\theta=\pi/2$ the second-harmonic radiation with field E_3 propagates along the y-axis, i.e. the direction of r is along the y-axis. If we assume no absorption the field generated by this polarization field is then, using Maxwell's equations, given by [5]

$$\frac{dE_3}{dy}=-\mathrm{i}\omega d_{11}\cos^2\psi^*\left(\frac{\mu_0}{\varepsilon_{2\omega}}\right)^{1/2} \times E_1E_2\exp[-\mathrm{i}(2k_e\sin\psi-k_3)y]. \tag{10}$$

It is seen that phase matching is fulfilled for $2k_e\sin\psi_m=k_3$, where ψ_m is the phase-matching angle. Using the refractive indices for the ordinary and extraordinary rays in the uniaxial crystal the phase matching condition can be calculated. This is indicated in Fig. 2. For the extraordinary ray we obtain $n_e(\psi)$ from the relation

$$\frac{1}{n_e^2(\psi)}=\frac{\cos^2\psi}{n_0^2}+\frac{\sin^2\psi}{n_e^2}. \tag{11}$$

The numerical values at $\lambda=10\,\mu$m are: $n_{0,\omega}=4.795$, $n_{0,2\omega}=4.856$, and $n_{e,\omega}=6.24$.

For phase matching we have the condition

$$n_{e,\omega}(\psi_m)\sin\psi_m=n_{0,2\omega}. \tag{12}$$

Substituting (12) into (11) and using the numerical values of n we obtain $\psi_m = 58.196°$.

Next we integrate (10) over a length L and taking the intensity $I(2\omega)$ we get

$$I(2\omega) = 2\omega^2 d_{11}^2 \cos^4\psi^* \left(\frac{\mu_0}{\varepsilon_{2\omega}}\right)^{3/2} I_1 I_2 L^2 \frac{\sin^2(\Delta kL/2)}{(\Delta kL/2)^2} \tag{13}$$

with

$$\Delta k = \frac{2\pi}{\lambda_2}[n_{0,2\omega} - n_{e,\omega}(\psi)\sin(\psi)]. \tag{14}$$

$d_{11} = 5.7 \times 10^{-21}$ in MKS units and λ_2 is the second-harmonic wave length in vacuum.

If the two pulses of the fundamental beams are delayed with respect to each other by a time τ the second-harmonic power will be proportional to $I_1(t)I_2(t-\tau)$.

For light pulses that are much shorter than the time constant of the detecting system the response R will be proportional to the time integral of the second harmonic power, i.e.

$$R = C\int I_1(t)I_2(t-\tau)dt \tag{15}$$

which is for $I_1 = I_2$ the autocorrelation function of the original laser pulse.

Experimental Requirements

In the above analysis it was found that phase matching for the second-harmonic signal is obtained for the angle $\pi - 2\psi_m$ between the two fundamental beams and for their bisector oriented along the y axis of the crystal. To satisfy these two conditions the question arises how accurately this has to be fulfilled in the experimental situation. The answer has a direct meaning to the usefulness of the method in practice. For this reason we first calculate the required accuracy of the angle ψ_m in the case of $L = 1.3$ mm.

It is seen from (13) that for a mismatch of $\Delta kL = \pi$ the second-harmonic power drops to 40% of its maximum. From (14) we get for a small deviation

$$\Delta k = \frac{2\pi}{\lambda_2}[-n_{e,\omega}(\psi_m)\cos\psi_m \Delta\psi - \sin\psi_m \Delta n_{e,\omega}(\psi_m)]. \tag{16}$$

Using (11) we obtain

$$\Delta k = \frac{2\pi}{\lambda_2} n_{e,\omega}(\psi_m) \times \cos\psi_m \left[-1 + n_{e,\omega}^2(\psi_m)\left(\frac{1}{n_{e,\omega}^2} - \frac{1}{n_{0,\omega}^2}\right) \times \sin^2\psi_m\right]\Delta\psi. \tag{17}$$

Substituting the numericals for λ_2, ψ_m, $n_{e,\omega}$, and $n_{0,\omega}$ the maximum allowable deviation of ψ_m is then given by $|\Delta\psi| = 4.8 \times 10^{-4}$ rad. For the incident beam the corresponding value is obtained by multiplying with $n_{e,\omega}(\psi_m) = 5.7139$. So we have $\Delta\psi_0 = 2.7 \times 10^{-3}$ rad or 0.16° which is from the experimental point of view a high requirement.

Next we calculate the accuracy of the orientation of the bisector. This can be considered by changing the direction of one fundamental beam by $\Delta\psi$ in the yz-plane and the other one by $-\Delta\psi$. The variation of Δk is then given by

$$\Delta(\Delta k) = \frac{2\pi}{\lambda_1}\{-[f(\psi_m + \Delta\psi) - f(\psi_m)] - [f(\psi_m - \Delta\psi) - f(\psi_m)]\}, \tag{18}$$

where λ_1 is the fundamental wave length in vacuum, and

$$f(\psi_m) = \sin\psi_m n_{e,\omega}(\psi_m),$$

and $n_{e,\omega}(\psi_m)$ given by (11).

For small variation of Δk we write

$$\Delta(\Delta k) = -\frac{2\pi}{\lambda_1} f''(\psi_m)(\Delta\psi)^2. \tag{19}$$

After some algebra and substituting the numericals we obtain $\Delta\psi = 1.9°$. With respect to the incident orientation of the fundamental beams the maximum allowable deviation between the bisector and the y-axis is 11° which is quite acceptable from a practical point of view.

In conclusion, we mention that so far only the adjustment of the angle between the two fundamental beams will be a very critical operation. However, it can be shown that the experimental solution to this problem is found by simply rotating the crystal around the z axis. It turns out that in this way an accurate tuning of the proper phase matching can be obtained.

Fine Tuning of the Phase Match Condition

The effect of rotating the crystal around the z-axis can be calculated by means of Fig. 3. It shows how the crystal has been cut. The oblique planes are necessary because of the high refractive index. The fundamental beams enter the crystal more or less perpendicularly so that the correct orientation within the crystal can be adjusted externally. As clearified in the previous section the field vectors of the fundamental incident beams lie in the yz plane of the crystal. The second harmonic has its field along the x axis whereas it propagates along the y-axis. The dependence of $I(2\omega)$ on the external angle ψ_0 between the two beams is very

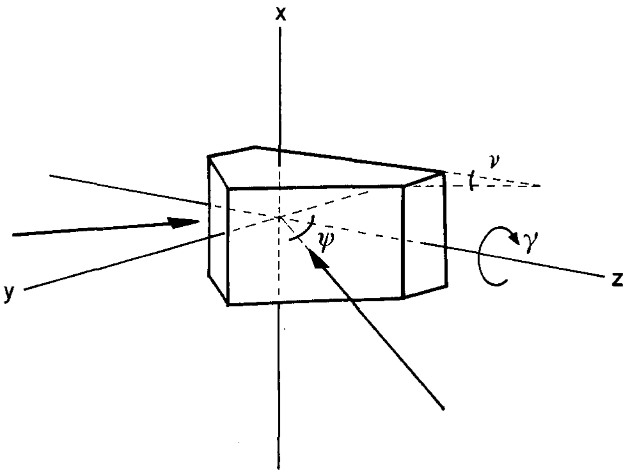

Fig. 3. Input parameters of the computer model

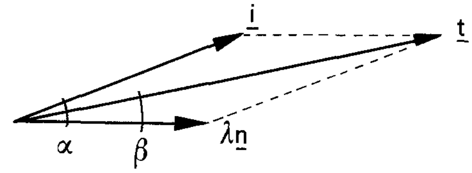

Fig. 5. See text

critical as can be seen in Fig. 4 for an interaction length $L = 1.3$ mm. The narrow range of the angle ψ_0 is in agreement with the estimate given in the previous section. The squares represent the measured values and the curve a computer calculation assuming plane waves. The larger width of the experimental data might be due to the (small) inherent beam divergence. Moreover, it is difficult to obtain an exact value for the interaction length. It is seen that the adjustment of ψ_m must be within a few tenths of a degree and becomes even smaller for larger L values. Therefore an important aspect in the design of the autocorrelator is the accuracy with which the phase matching in the crystal can be obtained. For this reason we investigated the relations between a number of parameters and their effect upon the second-harmonic signal.

Especially we investigated the possibility of correcting an error in the phase matched direction by rotation of the crystal around the z axis. The results are compared with a computer model which will be described as follows. It is assumed that the crystal is first rotated around the x axis for optimum second-harmonic generation, so that the y axis coincides with the bisector of the incoming fundamental beams. A symmetric situation is obtained with respect to the xy plane and only one half of the crystal has to be considered, let us say the right hand side of Fig. 3. The unit vectors in the direction of the incoming beam and the normal of the oblique plane may be expressed by the vectors **i** and **n**, respectively:

$$\mathbf{i} = \begin{bmatrix} 0 \\ \sin\psi_0 \\ \cos\psi_0 \end{bmatrix} \quad \text{and} \quad \mathbf{n} = \begin{bmatrix} \sin\gamma\cos\nu \\ \cos\gamma\cos\nu \\ \sin\nu \end{bmatrix}, \tag{20}$$

where ψ_0 is the angle between the z axis and the direction of propagation of the incident beam outside the crystal. ν is the angle between the oblique plane and the xz plane, which ideally should be equal to $\pi/2 - \psi_m$. The angle γ represents the rotation around the z axis. For $\gamma = 0$ the incident beam lies in the yz plane.

In order to find **t**, which represents the unit vector of the transmitted beam, we write (Fig. 5):

$$\mathbf{t} = \frac{\mathbf{i} + \lambda\mathbf{n}}{(1+\lambda^2)^{1/2}}. \tag{21}$$

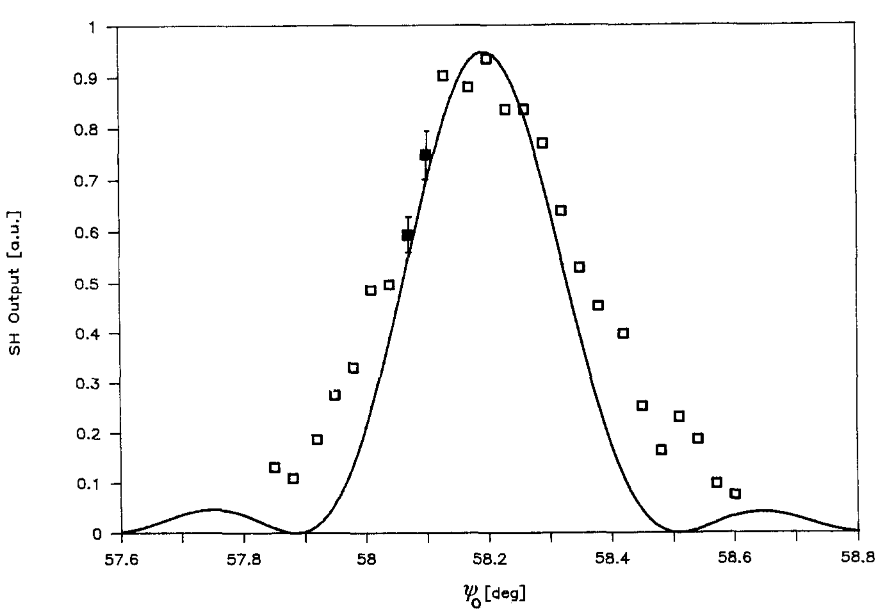

Fig. 4. Second-harmonic intensity as a function of the external angle ψ_0 between the fundamental beams for an interaction length of 1.3 mm

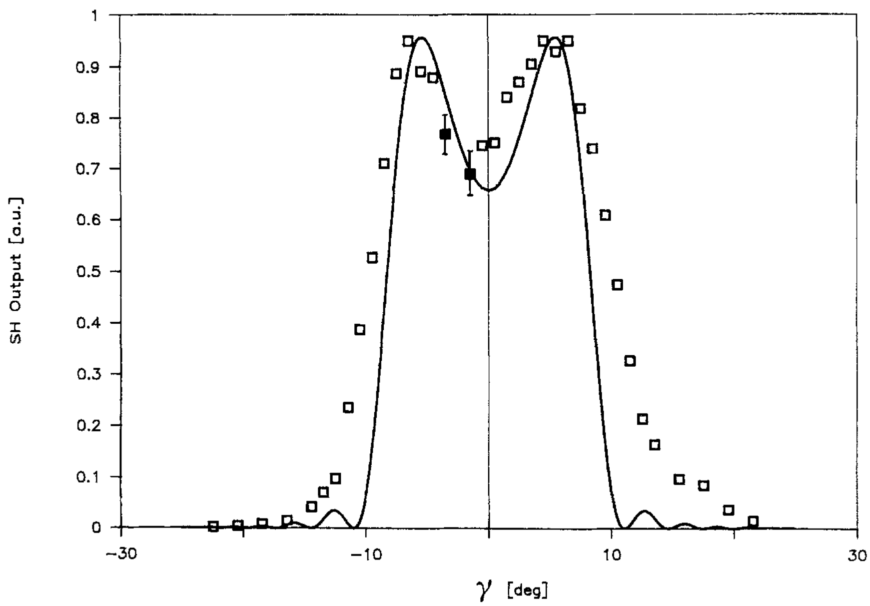

Fig. 6. The output signal as a function of γ for $L = 1.3$ mm, $\psi_0 = 58.200$ deg

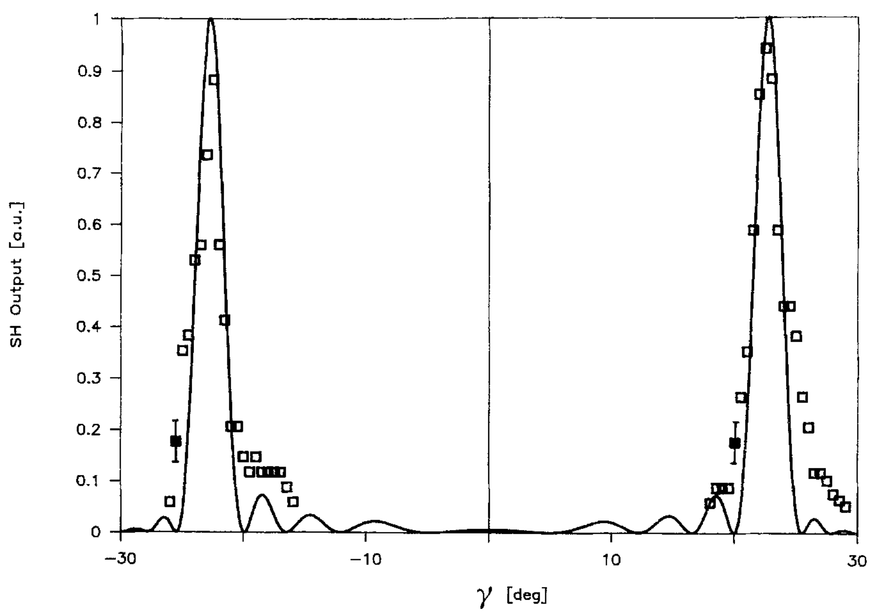

Fig. 7. The output signal as a function of γ for $L = 1.3$ mm, $\psi_0 = 59.85$ deg

A little algebra and Snell's law then yields

$$\lambda = [n_{e,\omega}^2(\psi) - 1 + (\mathbf{i} \cdot \mathbf{n})^2]^{1/2} - (\mathbf{i} \cdot \mathbf{n}), \tag{22}$$

where ψ represents the angle between the z-axis and $\mathbf{t}$. Cosψ is given by

$$\cos\psi = \frac{\cos\psi_0 + \lambda \sin\nu}{(1+\lambda^2)^{1/2}}. \tag{23}$$

The last equation can only be solved in an iterative way because λ is a function of ψ. By taking for instance $n(\psi) = 5$ as a starting value, λ is obtained by (22) and the first approximated value of ψ can then be found with (23). The obtained value of ψ will give a new value of $n(\psi)$ by means of (11) which can be used to obtain a new approximated value of ψ through (22) and (23), and so on.

To determine the influence of rotation on the intensity $I(2\omega)$ of the second-harmonic beam one also has to take into account the fact that the direction $\mathbf{r}$ of the second-harmonic beam is no longer along the direction of the y-axis. So (13) has to be multiplied by $\sin^2 3\theta$, where θ is the angle of $\mathbf{r}$ with respect to the x-axis. This angle θ can be calculated from the in-product of the x-axis and the unit-vector $\mathbf{r}$, which is determined by the projection of $\mathbf{t}$ on the xy plane.

The results of the calculations are shown by the curves of Figs. 6 and 7. In Fig. 6 there is a phase mismatch of only 0.004° and fine tuning is obtained by rotating γ over about 8°. In Fig. 7 the mismatch is

1.654° and the fine tuning is obtained by rotating γ over 23°. The experimental results are also plotted. Each square corresponds to an average of 25 shots of a pulsed CO_2 laser. Thus we find that a small mismatch given by $\Delta\psi_0$ can be corrected with a relatively large rotation γ around the z-axis.

Autocorrelator

A schematic drawing of the noncollinear optical system for measuring the autocorrelation is shown in Fig. 8.

The entrance mirror IV allows adjustment of the incoming beam, without moving the table. The mirror holder is mounted on a translation stage. The translation stage VII provides a maximum displacement of 15 cm, yielding an effective delay of 30 cm or 1 ns. This is sufficient for short pulses (several hundreds of ps), but not long enough for pulses of about 1 ns. We therefore attached a rail beside the translation stage, allowing displacement of the entire device itself. The position can easily be measured by any ordinary scale.

We generated mode-locked pulses with a 3 atmosphere CO_2 laser. Since the spacing between the pulses in the "pulse train" is much longer (12 ns) than the duration of the individual pulses (<1 ns) we fired the entire pulse train, thus measuring the averaged pulse length, weighted over the intensities of the pulses. Figure 9 represents the result obtained by the autocorrelator. The FWHM is about 9.0 cm, which corresponds to $0.090 \times 2/c = 0.60$ ns. The length of the original pulse (assumed to be Gaussian) then is $\frac{1}{2}\sqrt{2} \times 0.60 = 0.42$ ns.

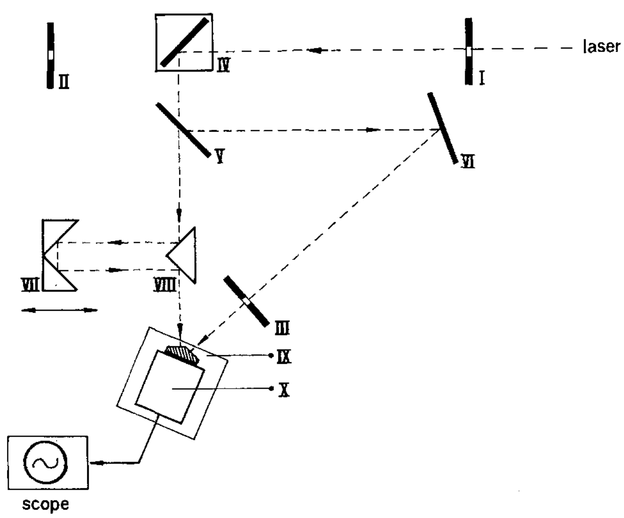

Fig. 8. I, III adjustable diaphragms, II diaphragm, IV entrance mirror (translation/rotation stage), V beam splitter, VI mirror holder, VII copper coated glass prisms mounted on translation stage, VIII copper coated glass prism mounted on prism table, IX adjustable mirror mount, X photoconductive cell

Conclusions

The very critical phase match conditions for CO_2 laser pulse measurements by means of second-harmonic generation in tellurium is feasible. The constructed device appeared to be rather easy in use, and is estimated to function well down to pulses with a FWHM of about 50 ps. The delay allows displacements of 0.01 mm, which implies a (theoretical) resolution of about 0.1 ps. This would make the autocorrelator suited for pulses down to about 1 ps, but we expect the resolution to be restricted by the limited stability of the optical components. The conversion efficiency was estimated to be about 1% (which appeared to be a very rough estimate) and corresponded to a maximum signal of about 0.5 V.

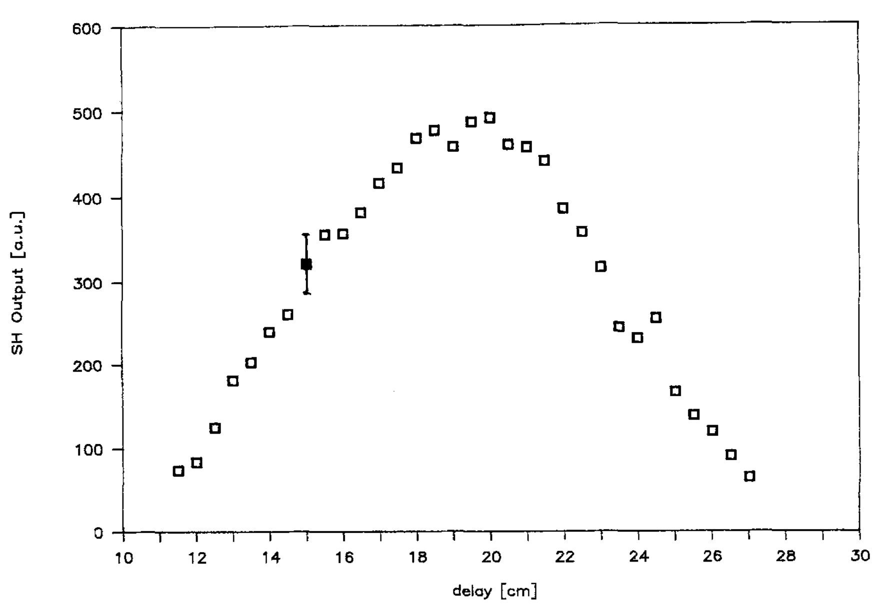

Fig. 9. Autocorrelation function of the laser pulses, as measured by the SHG autocorrelator. The FWHM of the laser pulse assumed to be Gaussian appears to amount to 0.42 ns

Installation and adjustment of the device appeared rather easy. Adjustments in order to obtain a maximum signal have to be performed by means of the entrance mirror and rotation of the crystal. If adjustment should be difficult, it can be made easier by decreasing the length of the overlap volume, i.e. by decreasing the beam diameter. This however will also decrease the output signal.

During the experiments, the experimental error appeared to be mainly caused by fluctuations in laser output. We dealt with this problem by dividing the second harmonic signal by the square of the laser intensity and by averaging over about 20 shots for each measurement. When the autocorrelator is to be used intensively, we suggest to have it entirely controlled by a microcomputer, which triggers the laser, averages the measurements and adjusts the delay by means of a stepping motor.

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