INTENSITY DETECTION OF PICOSECOND RUBY LASER PULSES BY TWO PHOTON ABSORPTION

W. BLAU and A. PENZKOFER
Naturwissenschaftliche Fakultät II - Physik, Universität Regensburg, 8400 Regensburg, Fed. Rep. Germany

Received 8 December 1980

The peak intensity of single picosecond light pulses from a mode locked ruby laser and its second harmonic is detected by two photon transmission measurements. The two photon absorption coefficients $\alpha^{(2)}$ of the applied crystals CdS and KI are measured ($\alpha^{(2)}_{CdS}$ (14400 cm^{-1}) = 19 cm/GW, $\alpha^{(2)}_{KI}$ (28800 cm^{-1}) = 8 cm/GW) and calibration curves are presented.

1. Introduction

The intensity dependence of various nonlinear optical effects may be used to determine the intensity of picosecond light pulses [1,2]. Here the two-photon transmission technique [2] is applied to determine the peak intensity of picosecond light pulses from a ruby laser at the fundamental ($\tilde{\nu}_F$ = 14400 cm^{-1}) and second harmonic frequency ($\tilde{\nu}_{SH}$ = 28800 cm^{-1}). This method has several advantages. i) The measured energy transmission is independent of the pulse duration as long as two photon induced excited state absorption is negligible. ii) It is independent of the beam radius. iii) The energy transmission is only slightly dependent on the temporal and spatial pulse shape. iv) No absolute signal values have to be measured.

The two photon absorption coefficients $\alpha^{(2)}$ of the applied crystals CdS [3,4] (frequency ν_F) and KI [5,6] (frequency ν_{SH}) are determined and calibration curves $T_E(I_0)$ are presented.

2. Theory

The energy transmission $T_E = E_T/E_I$ through a two photon absorbing medium is given by [2,7]

$$T_E = \frac{1-R}{1+R}\int_0^\infty dr'\, r' \int_{-\infty}^{\infty} dt'\, s(r',t') \times \frac{\exp(-\alpha l)}{1+\alpha^{(2)}(1-R)I_0 s(r',t')[1-\exp(-\alpha l)]/\alpha} \times \left[\int_0^\infty dr'\, r' \int_{-\infty}^{\infty} dt'\, s(r',t')\right]^{-1}, \quad (1)$$

R is the reflection factor, $s(r', t') = s(r/r_0, t/t_0) = s(r')s(t')$ describes the spatial and temporal shape of the inpute pulses, α is the linear absorption coefficient and l is the sample length. In eq. (1) changes of the two photon absorption due to light reflected back to the sample at the excit face of the crystal are neglected. This omission is tolerable for short light pulses ($\Delta t\, c/n \ll l$) or weak reflections R. The effects of excited state absorption of generated electrons and holes [8] are not included in eq. (1).

In the experiments $\alpha^{(2)}$ is determined from eq. (1) by measuring $T_E, R, \alpha, l, s(r'), s(t')$ and $I_0 = E_I/[2\pi r_0^2 t_0 \int_0^\infty r' s(r')\,dr' \int_{-\infty}^{\infty} s(t')\,dt']$. Then eq. (1) is used to calculate calibration curves $T_E(I_0)$.

3. Experiment and results

The experimental setup for the intensity detection of the fundamental laser pulses is shown in fig. 1. A

0 030–4018/81/0000–0000/$ 02.50

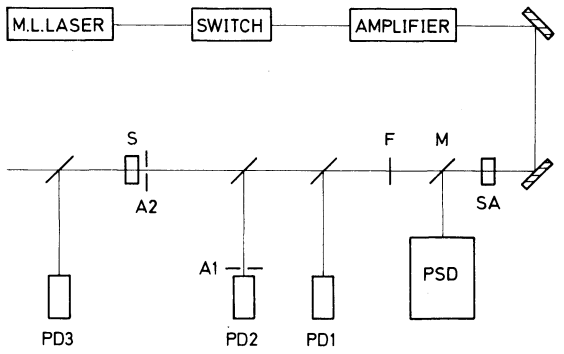

Fig. 1. Experimental setup. SA, saturable absorber, M, partially reflecting mirror; PSD, pulse shape and duration detection system; F, filters; S, sample; PD1–PD3, photodetectors; A1 and A2, apertures.

mode locked ruby laser generates a train of picosecond light pulses (saturable absorber: mixture of DDI and rhodamine 6G in methanol [9]). A single pulse is selected with a krytron triggered Pockels cell shutter and increased in energy by a ruby laser amplifier. The output pulse energy is about 5 mJ. The background energy content is reduced by a saturable dye cell SA (DDI in methanol).

The temporal shape and duration of the light pulses is measured with the saturable absorber technique described in ref. [10]. A 6×10^{-5} molar solution of DOTC-Iodide in DMSO (cell length 2 cm) was used as bleachable dye [11]. An average pulse duration of $\Delta t = 24$ ps (FWHM) was measured. The pulse shape was found to be asymmetric. It is best fitted by a leading gaussian and a trailing hyperbolic secant function $s(t') = [1-(\theta')t')]\exp(-t'^2) + \theta(t')/\cosh^2(t'/\kappa)$ with $\kappa = 1.8 \pm 0.3$ ($\Delta t = (0.83 + 0.88\kappa)t_0$).

The spatial profile of the input pulses is truncated by the apertures A_1 and A_2 of radius $r_A = 1.5$ mm. The shape is approximated by the truncated gaussian $s(r') = \exp(-r'^2)$ for $0 \leqslant r' \leqslant r_A/r_0$, and $s(r') = 0$ otherwise. The 1/e-radius r_0 is determined by comparison of the signal heights S of detectors PD1 and PD2 ($S_2/S_1 = 1-\exp[-(r_A/r_0)^2]$, $r_0 \approx 5$ mm).

The reflection R was determined by the linear transmission T through the transparent crystal ($\alpha \approx 0$) with a spectral photometer [$T = (1-R)/(1+R)$]. A value of $R = 0.036$ was obtained for the anti-reflection coated crystal ($T = 0.93$). The input energy E_I was measured with the calibrated photocell PD2. The calibration was performed with energy meters.

The energy transmission T_E through the CdS crystal (length 1 cm, dark resistivity 2.3×10^9 Ωcm from Cleveland Crystals, Inc.) is measured with photodetectors PD2 and PD3. The c-axis of the crystal was oriented parallel to the electric field vector of the input laser light. The input energy E_I to the crystal is varied with neutral density filters. For input energies $I_0 > 10^8$ W/cm^2 a lens of 1 m focal length was inserted in 50 cm distance from the crystal.

The measured energy transmission T_E versus input peak intensity I_0 is depicted in fig. 2. A two photon absorption coefficient of $\alpha^{(2)} = (1.9 \pm 0.2)\times10^{-8}$ cm/W ($\sigma^{(2)} = \alpha^{(2)}h\nu_F/N = (2.7 \pm 0.3)(\times 10^{-49}$ cm^{4}s, $N = 2\times10^{22}$ cm^{-3}) is obtained by comparing the experimental points with the theoretical curve 3. Even at the highest input peak intensity of $I_0 \approx 3\times10^8$ W/cm^2 ($T_E \approx 0.25$) no deviation of the experimental points from the calculated curve is observed. This fact

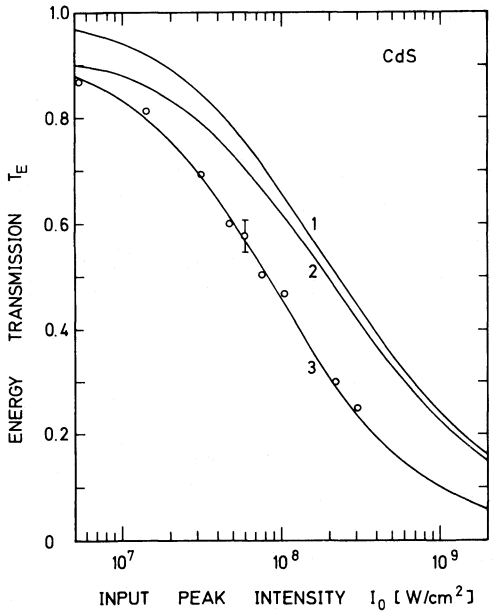

Fig. 2. Two photon transmission through CdS. Experimental points: Sample length $l = 1$ cm; reflection coefficient $R = 0.036$; c-axis parallel to input light field; frequency $\tilde{\nu}_F = 14400$ cm^{-1}; aperture $r_A = 1.5$ mm ($r_A/r_0 = 0.3$). Curves are calculated for $\alpha^{(2)} = 1.9\times10^{-8}$ cm/W, $l = 1$ cm and temporal asymmetric pulse profile with gaussian rise and hyperbolic secant decay ($\kappa = 1.8$): (1) spatial gaussian with $R = 0$; (2) spatial gaussian with $R = 0.036$; (3) spatial truncated gaussian with $r_A/r_0 = 0.3$ and $R = 0.036$.

indicates that the effect of excited state absorption is negligibly small in our experiments ($\sigma_{ex} = \sigma_{ex,e} + \sigma_{ex,h} \leqslant 5 \times 10^{-17}$ cm^2 is estimated by an analysis similar to ref. [8]). The observed two photon absorption coefficient is in good agreement with previously reported values measured with Q-switched ruby lasers [12–17]. In ref. [18] a larger two photon absorption cross-section is found with nanosecond ruby light pulses.

The curves 1 and 2 of fig. 2 represent calibration curves for the peak intensity detection. They belong to spatial gaussian and temporal asymmetric pulses (gaussian rise and hyperbolic secant decay with $\kappa = 1.8$). Curve 1 is calculated for reflectivity $R = 0$ while curve 2 belongs to $R = 0.036$. For pulses with temporal gaussian shape the same energy transmission is obtained at about 7 percent lower input peak intensity.

For the intensity detection at the second harmonic of the ruby laser the two photon absorption coefficient of KI at $\tilde{\nu}_{SH} = 28800$ cm^{-1} was determined in a similar procedure as described above.

The second harmonic was generated in an ADP crystal (1 cm long). The pulse duration and pulse shape was measured with a 2×10^{-4} molar solution of dimethyl POPOP in toluene. An average pulse duration of $\Delta t \approx 18$ ps was found. The pulse shape could be fitted with a gaussian rise and a hyperbolic secant decay ($\kappa \approx 1.3 \pm 0.2$).

The input intensity of the second harmonic light was varied with a lens of 1 m focal length (distance from sample 90 cm) and filters. The 1/e beam radius reduced to $r_0 \approx 0.7$ nm and apertures with a radius of $r_A = 0.35$ mm were used.

The KI crystal (length 1 cm, cubic axis parallel to the electric field vector) was inserted in a cell with Suprasil windows. Cyclohexane was used as index matching fluid (thickness ≈0.5 mm). The two-photon absorption of the windows [19] and cyclohexane is neglgible. A reflection factor of $R = 0.05$ was found ($T = 0.905$).

In fig. 3 the measured energy transmission versus input peak intensity is depicted. The experimental points fit well with the calculated curve 3. A two photon absorption coefficient of $\alpha^{(2)} = (8 \pm 2) \times 10^{-9}$ cm/W is deduced ($\sigma^{(2)} = \alpha^{(2)} h\nu_{SH}/N = (4 \pm 1) \times 10^{-49}$ cm^{4}s, $N = 1.135 \times 10^{22}$ cm^{-3}). The excited state absorption cross section is estimated to be $\sigma_{ex} \leqslant 5 \times 10^{-17}$ cm^2 [8]). Previously reported two photon absorption cross sections for KI at the same frequency are in good agreement with our measurements [19,20].

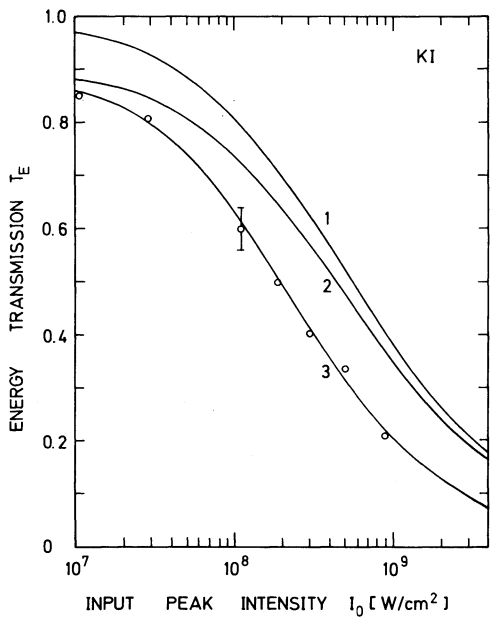

Fig. 3. Two photon transmission through KI. Experimental points: Sample length $l = 1$ cm; reflection $R = 0.05$; frequency $\tilde{\nu}_{SH} = 28800$ cm^{-1}; aperture $r_A = 0.35$ mm ($r_A/r_0 = 0.5$). Curves are calculated for $\alpha^{(2)} = 8 \times 10^{-9}$ cm/W, $l = 1$ cm and temporal asymmetric profile with gaussian rise and hyperbolic secant decay ($\kappa = 1.3$). (1) Spatial gaussian with $R = 0$; (2) spatial gaussian with $R = 0.05$; (3) spatial truncated gaussian with $r_A/r_0 = 0.5$ and $R = 0.05$.

The curves 1 and 2 are the intensity calibration curves for $R = 0$ and $R = 0.05$, respectively. A spatial gaussian and temporal asymmetric profile with gaussian rise and hyperbolic secant decay ($\kappa = 1.3$) is assumed.

References

[1] A. Penzkofer, D. von der Linde and A. Laubereau, Optics Comm. 4 (1972) 377.
[2] A. Penzkofer and W. Falkenstein, Optics Comm. 17 (1976) 1.
[3] R. Braunstein and N. Ockman, Phys. Rev. 134 (1964) A 499.
[4] P.J. Regensburger and E. Panizza, Phys. Rev. Letters 18 (1967) 113.
[5] K. Park and R.G. Stafford, Phys. Rev. Letters 22 (1969) 1426.
[6] J.J. Hopfield, J.M. Worlock and K. Park, Phys. Rev. Letters 11 (1963) 414.

[7] A. Penzkofer and W. Kaiser, Appl. Phys. Letters 21 (1972) 427.
[8] A. Penzkofer and W. Falkenstein, Optics Comm. 16 (1976) 247.
[9] R. Cubeddu, R. Polloni, C.A. Sacchi and O. Svelto, IEEE J. Quant. Electron. QE-5 (1969) 470.
[10] J. Wiedmann and A. Penzkofer, Optics Comm. 30 (1979) 107.
[11] J.P. Fouassier, D.J. Lougnot and J. Fauré, Optics Comm. 18 (1976) 263.
[12] V.K. Konyukhov, L.A. Kulevskii and A.M. Prokhorov, Sov. Phys. Doklady 10 (1966) 943.
[13] V.V. Arsenev, V.S. Dheprovksii, D.M. Klyshko and A.N. Penin, Sov. Phys. JETP 29 (1969) 413.
[14] M.S. Brodin, S.G. Shevel, F.F. Kodzhespirov and L.A. Mozharovskii, Sov. Phys. Semic. 5 (1971) 2047.
[15] M.P. Lisitsa, P.I. Sidorko, P.E. Mozol and N.I. Vitrikhovskii, Soc. J. Quant. Electron. 2 (1972) 432.
[16] I.M. Catalano, A. Cingolani and A. Minafra, Phys. Rev. B9 (1974) 707.
[17] R. Baltrameyunas, V. Gavryushin and Yu. Vaitkus, Sov. Phys. Sol. State 18 (1976) 660.
[18] G. Kobbe and C. Klinshirn, Z. Physik B 37 (1980) 9.
[19] P. Liu, W.L. Smith, H. Lotem, J.H. Bechtel, N. Bloembergen, and R.S. Adhav, Phys. Rev. B17 (1978) 4620.
[20] I.M. Catalano, A. Cingolani and A. Minafra, Phys. Rev. B5 (1972) 1629.