Vol. A54 (1978) ACTA PHYSICA POLONICA No 2

EXPERIMENTAL DEPENDENCE OF TWO-PHOTON ABSORPTION EFFICIENCY ON MODE SELECTION AND MODE LOCKING OF He-Ne LASER

By M. Głódź

Institute of Physics, Polish Academy of Sciences, Warsaw*

(Received February 15, 1978)

A He–Ne laser with a neon absorption cell inside the laser cavity was used as a source of either: (a) one mode-, (b) two mode-, (c) multimode free running-, or (d) multimode-locked -laser light. The enhancement factors obtained in the experiment of the two photon absorption efficiency in cases (b), (c), and (d) over the one mode case are generally in agreement with theoretical predictions.

1. *Introduction*

The dependence of non-linear optical processes on statistical properties of laser light has been investigated by many authors both theoretically and experimentally (e.g. [1–8]).

The present experiment was aimed at comparing the two-photon absorption coefficient in the case of a one mode laser used as a light source with that coefficient in the cases of either two modes-, multimode free running- or multimode-locked-laser.

Fig. 1 presents the experimental set-up. A $^3\mathrm{He}-^{20}\mathrm{Ne}$ laser ($\lambda = 632.8$ nm, TEM_{00n} mode) which contained in its cavity an absorption tube filled with $^{20}\mathrm{Ne}$ was used. The laser design was similar to that described in [9]. If the current of the discharge in the absorption tube is small or switched off, the free running action takes place. An increase in current results in mode selection and/or self locking of modes [9].

The laser modes spectrum was displayed on the first oscilloscope by means of a scanning spectrum analyser. The laser light intensity time dependence detected by a fast photodiode was displayed on the second oscilloscope.

Either α-NPO saturated solution in toluene or a solid solution of 3,4-benzopyrene in methyl polymetacrylate was used as a two photon absorber; the concentration of 3,4-benzopyrene in PMMA was 10^{-3} g/cm^3.

---

* Address: Instytut Fizyki PAN, Al. Lotników 32/46, 02-668 Warszawa, Poland.

The time-averaged fluorescence intensity $\langle I_F \rangle_t$ detected by a photomultiplier and a lock-in nanovoltmeter was a measure of the two-photon absorption. The signal should be proportional to the square of the time-averaged laser light intensity $\langle I_L \rangle_t$

$$\langle I_F \rangle_t = a g^{(2)} \langle I_L \rangle_t^2, \tag{1}$$

where factor $g^{(2)} = g^{(2)}(0)$ is a second order autocorrelation function and factor a depends on the molecular two-photon absorption coefficient of the sample, on the concentration of the absorbing molecules, and on certain geometrical features of the set-up.

For one coherent mode $g_1^{(2)} = 1$ (e.g. [3]).

For two modes of equal intensities $g_2^{(2)} = 1.5$ [8].

For multimode free-running laser light (infinite number of independent modes) as for a Gaussian field $g_{\text{chaotic}}^{(2)} = 2$ [e.g. [3]).

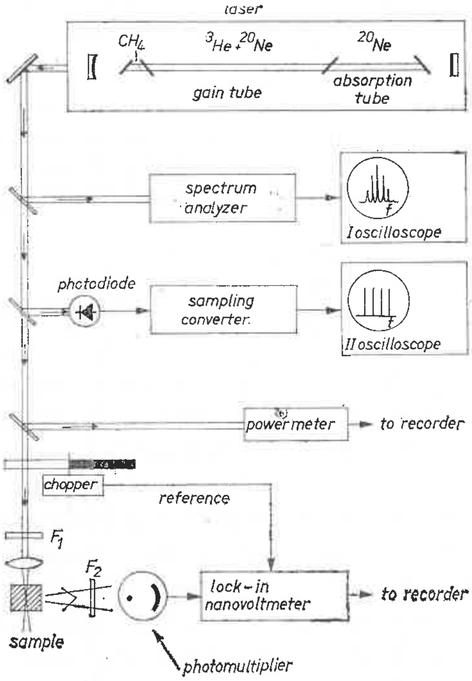

Fig. 1. Experimental set-up. The laser mirrors: distance $L = 5$ m, the radius of curvatures: $R_1 = 5$ m and $R_2 = \infty$, transparencies $T_1 = 4.7\%$, $T_2 = 0\%$; F_1 — Schott OG 5/2 mm filter; F_2 — Schott BG 12/4 mm filter; Sample — α-NPO solution in toluene or 3,4-benzopyrene in PMMA; photodiode — rise time less than 1 ns; photomultiplier — EMI 9789 QB

For perfectly mode-locked laser light, the $g_{\text{mode-lock}}^{(2)}$ value was calculated taking from Davis' paper [10] the formula for field amplitude $E(t, z)$ of a mode-locked laser when the modes spectrum envelope is of Gussian shape and assuming that each term in the series of equation (14) in Davis' paper gives the shape of the envelope of one of the electric field pulses. In simplified form and situating the maximum of such a pulse at $t = 0$ a pulse shape can be expressed:

$$\mathscr{E}(t) = A \exp\left(-\tfrac{1}{4}\,\Delta n^2 \Omega^2 t^2\right),$$

where A is a constant for a certain Δn; Δn — number of modes in half $1/e$ width of the spectrum envelope of modes amplitudes; $\Omega = 2\pi/T$ and $T = 2L/c$ the period of the pulses.

$$g^{(2)}_{\text{mode-lock}} = \frac{\langle I^2 \rangle_t}{\langle I \rangle_t^2} = \frac{\dfrac{1}{T}\displaystyle\int_{-T/2}^{T/2} I^2(t)\,dt}{\dfrac{1}{T}\left(\displaystyle\int_{-T/2}^{T/2} I(t)\,dt\right)^2}.$$

Taking for light intensity $I(t) \sim \mathscr{E}^2(t)$ and replacing the integration limits $[-T/2, T/2]$ by $[-\infty, \infty]$, one obtains

$$g^{(2)}_{\text{mode-lock}} = \Delta n \sqrt{\pi} = \sqrt{\frac{\pi}{2}}\, M, \tag{2}$$

where $M = \sqrt{2}\,\Delta n$ is the number of modes in full $1/e$ width of the spectrum envelope of their intensities.

2. *Experimental results*

In the experiment the $\langle I_F \rangle_t$ and $\langle I_L \rangle_t$ values were measured and then the coefficients $\alpha = g^{(2)} a$ were calculated from formula (1). Normalizing a given coefficient α to a coefficient corresponding to single mode laser light, one should obtain the $g^{(2)}$ value in this given case providing that we ensure for the factor a to be the same constant in the case of interest and in the one mode case and providing our single mode laser gives coherent light. The experimental results of such a normalization will be called enhancement factors and are summarised together with corresponding theoretical $g^{(2)}$ values in Table I and Fig. 3.

The enhancement factor 1.55 ± 0.03 obtained in the case of two modes of equal intensities exceeds the value 1.5. This might be due to an observed fact that two other modes of small intensities — the higher less than 2% of the intensities of the main modes — interfered with the two modes spectrum.

The free running laser light gave enhancement factors between 1.81 ± 0.10 and 2.53 ± 0.10. The first value was obtained when about 50 magnets were situated along the gain tube forming a nonhomogeneous magnetic field of about 300 Gauss. As these magnets were removed, the enhancement factor gradually increased up to 2.53 when all the magnets were removed.

The free running laser light spectrum contained over 20 modes (Fig. 2a). If more than 20 modes oscillate independently the $g^{(2)}$ value should be nearly 2 (1.95 for 20 modes of equal intensities — after [5]).

TABLE I

Experimental values of enhancement factors and corresponding values of second order correlation functions $g^{(2)} = g^{(2)}(0)$

Theory		Experiment	
case	$g^{(2)}$ values	enhancement factors	case
single mode	1		
two modes of equal intensities	1.5	1.55±0.03	two modes of equal intensities
many independently oscillating modes or chaotic light	2	1.81±0.10 ⋮ 2.53±0.10	with magnets } free running laser without magnets } free running laser
M locked modes in full $1/e$ width of the spectrum envelope of modes intensities (Gaussian shape)	$\sqrt{\frac{\pi}{2}}M$	versus M at the Fig. 3	M locked modes

Fig. 2c shows that additional pulsing trains due to self-locking of modes added to the stochastic fluctuations of the free-running laser light. As a result of this the corresponding enhancement factor was greater than two, namely about 2.5. The periodical pulse structure diminished if several magnets were placed along the laser tube (Fig. 2b). With magnets however the obtained enhancement factor was only about 1.8. It may be that the $g^{(2)}$ value for single mode laser in this experiment significantly exceeded 1 (for the laser was not well stabilized and certain fluctuations even of single mode intensity were observed) and/or perhaps not all of the over 20 modes oscillated simultaneously.

These results may serve as one more piece of evidence (another: see e.g. [6, 11]) that one should be very careful in considering multimode free running lasers as a source of light with the same fluctuations as the thermal source whose light is transmitted through a suitable narrow-band optical filter [12].

Fig. 3 presents the enhancement factors versus M — the number of locked modes in full $1/e$ width of modes intensity spectrum envelope. The error in M values is estimated as ± 1 and is mainly due to certain intensity fluctuations of each mode. The experimental data — points — are compared with theoretical straight line $\sqrt{\frac{\pi}{2}}M$ from the formula (2). For $M > 26$ the experimental points lay generally below the straight line. In this region the modes intensities fluctuations increased and were particularly strong in the vicinity of $M = 30$. This could cause the decrease in the real $g^{(2)}$ value [3, 4]. Most of the other points show a tendency to lie above the straight line, although it was expected that they would have fallen rather below the straight line due to some intensity fluctuations which

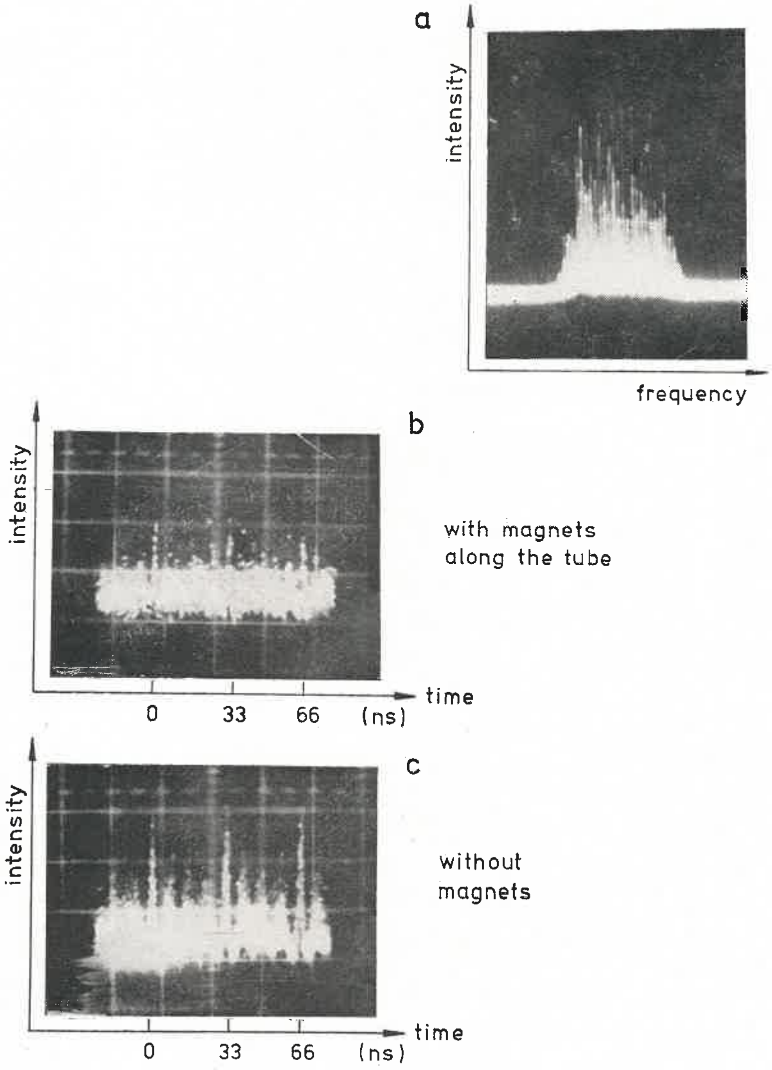

Fig. 2. The free running laser: (a) time averaged spectrum of modes (oscilloscope I — the exposition time ca 0.5 s); (b) the fluctuations of the laser light in the case when magnets are situated along the gain tube; (c) the fluctuations when magnets are removed

are always observed [3, 4]. Perhaps the assumption that the modes spectrum envelope is of Gaussian shape is not sufficiently valid here. This assumption was roughly checked for a few shapes — a certain asymmetry of some shapes was observed.

In the experiment two kinds of two-photon absorbers were used: α-NPO solution in toluene and a 3,4-benzopyrene solid solution in PMMA. The test on the nonlinearity order of the absorption process for α-NPO gave for this order the value 2.10±0.03. This value for 3,4-benzopyrene in PMMA is known from [13] as 2.02±0.05 ($\lambda = 630$ nm). No significant discrepancies were observed among the equivalent enhancement factors obtained with either α-NPO or 3,4-benzopyrene. Thus the enhancement factors values in Table I are averaged results from repeated experiments regardless of which of the two absorbers was actually used.

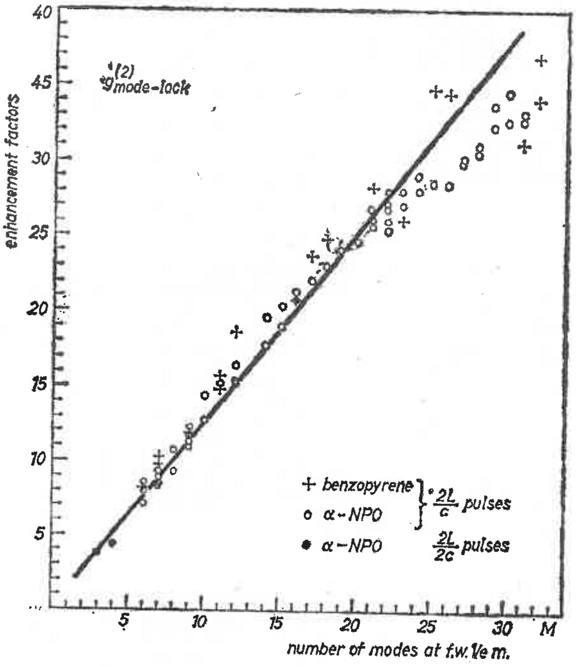

Fig. 3. Enhancement factors versus M — the number of modes in full $1/e$ width of the spectrum envelope of modes intensities (experimental points) compared with corresponding $g^{(2)\cdot}_{\text{mode-lock}} = \sqrt{\frac{\pi}{2}}\,M$ (straight line)

3. *Conclusion*

Although the $g^{(2)}(0)$ values do not define necessarily the type of light fluctuations, an experiment like this two-photon absorption may also be regarded as a source of information concerning the absorbed light itself.

The author wishes to thank Dr. J. Krasiński, W. Majewski and Dr. F. Sanchez for inspiring the idea of the experiment, Dr. J. Krasiński and Dr. L. Lis for many helpful discussions, Dr. A. Kujawski for valuable suggestions and his continuous encouragement, and L. Chałupka for technical assistance.

REFERENCES

[1] P. Lambropoulos, C. Kikuchi, R. K. Osborn, *Phys. Rev.* **144**, 1081 (1966).
[2] B. R. Mollow, *Phys. Rev.* **175**, 1555 (1968).
[3] A. A. Grütter, H. P. Weber, R. Dändliker, *Phys. Rev.* **185**, 629 (1969).
[4] R. Dändliker, A. A. Grütter, H. P. Weber, *IEEE J. Quantum Electron.* **QE-6**, 687 (1970).
[5] I. V. Tomov, A. S. Chirkin, *Sov. J. Quantum Electron.* **1**, 79 (1971).
[6] C. Lecompte, G. Mainfray, C. Manus, F. Sanchez, *Phys. Rev. Lett.* **32**, 265 (1974); *Phys. Rev.* **A11**, 1009 (1975).
[7] J. Krasiński, S. Chudzyński, W. Majewski, M. Głódź, *Opt. Commun.* **12**, 304 (1974).
[8] J. Krasiński, B. Karczewski, W. Majewski, M. Głódź, *Opt. Commun.* **15**, 409 (1975).
[9] Y. M. Beterov, V. N. Lisitsyn, V. P. Tchebotaev, *Opt. Spektrosk.* **30**, 932 (part I); 1108 (part II) (1971).
[10] L. W. Davis, *Am. J. Phys.* **38**, 918 (1970).
[11] T. Uchida, A. Ueki, *IEEE J. Quantum Electron.* **QE-3**, 17 (1967).
[12] L. Mandel, E. Wolf, *Mod. Phys.* **37**, 231 (1965).
[13] J. Krasiński, W. Majewski, M. Głódź, *Opt. Commun.* **14**, 187 (1975).