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TWO PHOTON TRANSITIONS IN THE OPTOGALVANIC SPECTRUM OF NEON

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Abstract

Seventeen two-photon transitions for neon have been observed in the 580-635 nm spectral region for use in the spectroscopic study of its higher excited levels, which are not accessible by one-photon absorption. To compare the two and one-photon absorption signals originating from the same lower level, an effort was made to record single-photon optogalvanic spectrum in the available wavelength region. The dependence of optogalvanic signals on the laser power has also been studied. The optogalvanic signals were obtained from a commercial neon-filled, hollow-cathode lamp, which was irradiated with pulsed dye laser pumped with N_2 laser.

Introduction

Two-photon absorption is the simultaneous absorption of two photons by a molecule which undergoes a transition $E_i \rightarrow E_f$ with $(E_f - E_i) = \hbar(\omega_1 + \omega_2)$. The photons may either come from a single laser beam passing through the absorbing sample or they may be provided by two beams emitted from one or two lasers.

The first detailed theoretical treatment of two-photon processes was given in 1931 by Guppert-Mayer [1], whereas the experimental realization had to wait for sufficiently intense light sources provided by pulsed lasers [2]. The first two-photon absorption in the molecular gas phase was observed on benzene by Hochstraser *et al.* [3], in which the absorption is monitored by the fluorescence emitted from the upper excited level.

Following our experimental studies on the optogalvanic effect (OGE) of neon [4, 5], it was decided to use this technique for detection of two-photon absorptions in the atom to shed light on the dynamics of OGE and its utilization as a source of standard line.

The optogalvanic signal is observed as a change in the impedance of a discharge when irradiated with a laser light tuned to a transition frequency of the species in the discharge. Small changes in the impedance can be easily detected with a high signal to noise ratio [6, 7].

Using OGE, seventeen two-photon absorption lines were recorded in the spectral region from 580 nm to 635 nm, for transitions between the first group of neon excited states in the $1s^2 2s^2 2p^5 3s$ configuration and its higher excited levels. Variations in the optogalvanic signals which are due to the discharge current changes were also studied. To compare the two and single-photon absorption signals originating from the same lower level, an effort was made to record the single-photon optogalvanic spectrum in the available wavelength region. The dependence of optogalvanic signal intensity on the laser power has also been studied.

Keywords: Multiphoton transitions in atomic spectra; Optogalvanic spectroscopic methods; Laser interactions with plasma

Experimental Section

A schematic representation of the experimental setup is shown in Figure 1. Two-photon absorption

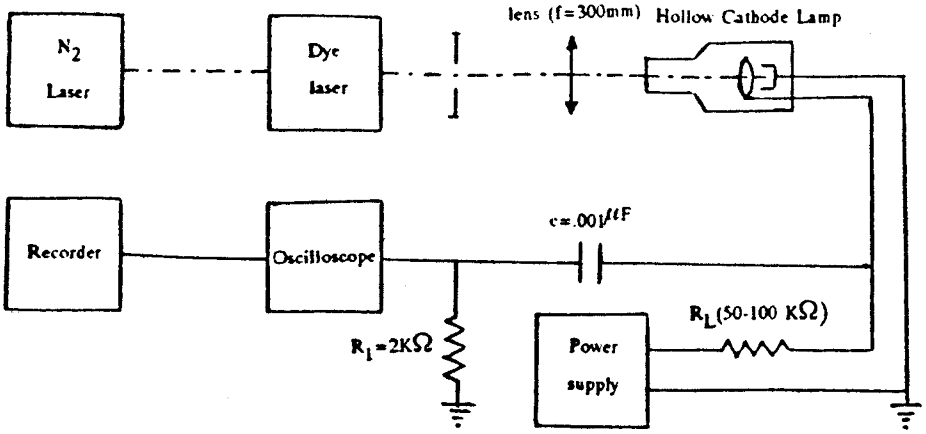


Figure 1. Schematic representation of experimental setup

was studied on a WL22937A hollow-cathode neon lamp. The laser from a pulsed dye laser (PRA, LN107) with power of 4 kw and 0.4 A° line-width was focused with a 30-cm focal lens on the negative glow region of the lamp. The dye laser was pumped by the output of a N_2-laser (PRA, LN-1000).

The neon pressure in the discharge tube was specified at 4 torr. The tube was operated at currents between 1 and 8 mA supplied by a regulated dc power supply. The optogalvanic signals, measured with the RC differentiating circuit, were sent to a storage oscilloscope (Le Croy 9400) and then fed into a chart recorder (HP-7475A).

Experimental Results

Seventeen two-photon absorptions were observed in neon hollow cathode discharge in the wavelength range between 580 to 635 nm. These are tabulated in Table 1 and the full spectrum of these transitions is shown in Figure 2. All of the observed two-photon absorptions in this work originate from the four levels of first excited state configurations which have been designated as $1s_m$ (m=2,3,4,5) by Paschen [8]. The $1s_3$ and $1s_5$ levels are metastable with a radiative life time of the order of one second having an important role in the two-photon transitions. Two-photon transitions in the neon terminate at higher excited states and their intermediate virtual levels are $2p_n$ (n=1 to 10) elec-

Table 1. Two-photon transitions in neon at a discharge current I=6mA

Transitions	$E_i(cm^{-1})$ - $E_f(cm^{-1})$	Standard Transition Wavelengths (A°)	Laser Wavelengths (A°)
$1s_5 \rightarrow 4s_1'$	134044-167819	2960.74	5921.9
$1s_5 \rightarrow 4s_1''$	134044-167808	2961.69	5923.8
$1s_5 \rightarrow 4s_1'''$	134044-167807	2961.78	5924.3
$1s_5 \rightarrow 4s_1''''$	134044-167806	2961.86	5924.6
$1s_4 \rightarrow 4s_1'$	134461-167819	2997.80	5995.9
$1s_4 \rightarrow 4s_1''$	134461-167808	2998.77	5997.7
$1s_4 \rightarrow 4s_1'''$	134461-167807	2998.87	5997.9
$1s_4 \rightarrow 4s_1''''$	134461-167806	2998.95	5998.2
$1s_5 \rightarrow 4d_5'$	134044-166986	3035.58	6071.6
$1s_5 \rightarrow 4d_6$	134044-166978	3036.28	6073.2
$1s_4 \rightarrow 4d_5$	134461-166986	3074.55	6149.7
$1s_3 \rightarrow 4d_1''$	134821-167058	3101.94	6203.3
$1s_4 \rightarrow 3s_2$	134461-166667	3104.99	6208.5
$1s_3 \rightarrow 4d_3$	134821-167022	3105.41	6211.9
$1s_3 \rightarrow 4d_6$	134821-166978	3109.65	6220.8
$1s_5 \rightarrow 3s_4$	134044-165923	3136.78	6273.8
$1s_5 \rightarrow 3s_5$	134044-165829	3145.13	6290.7

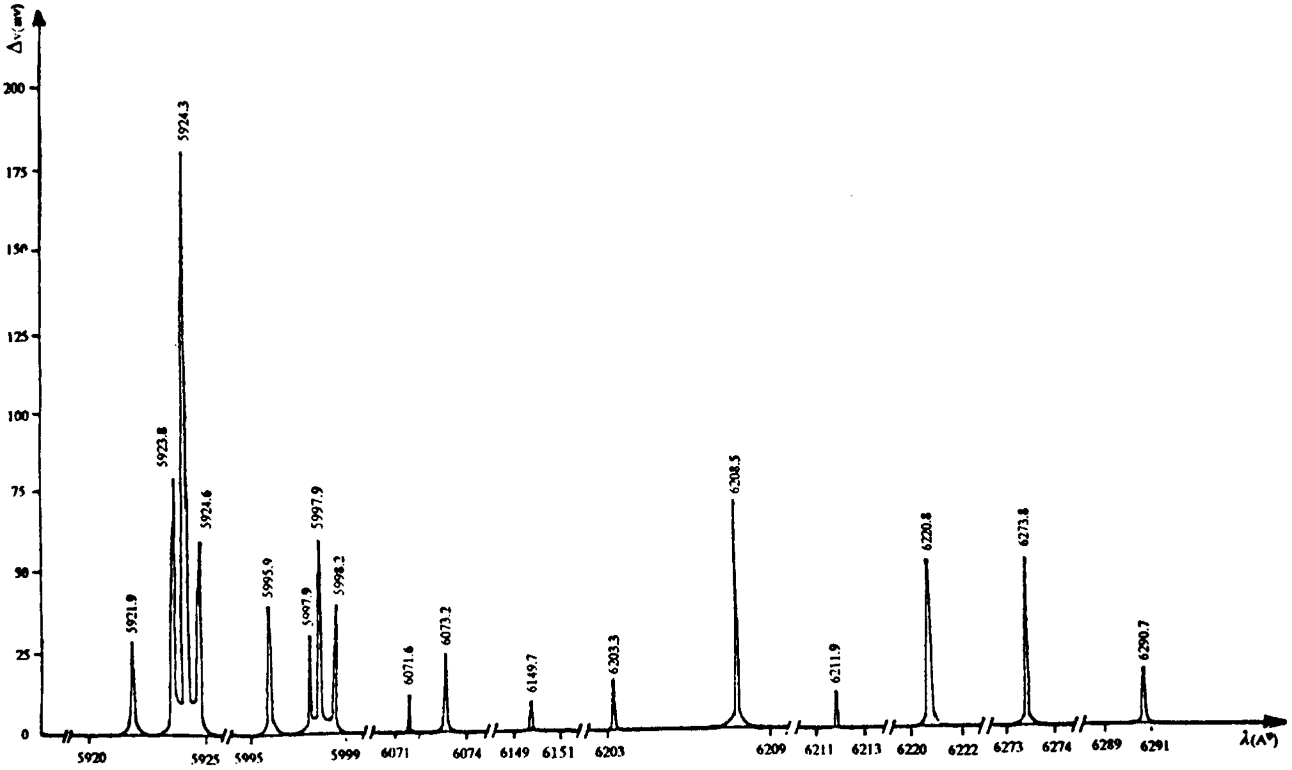
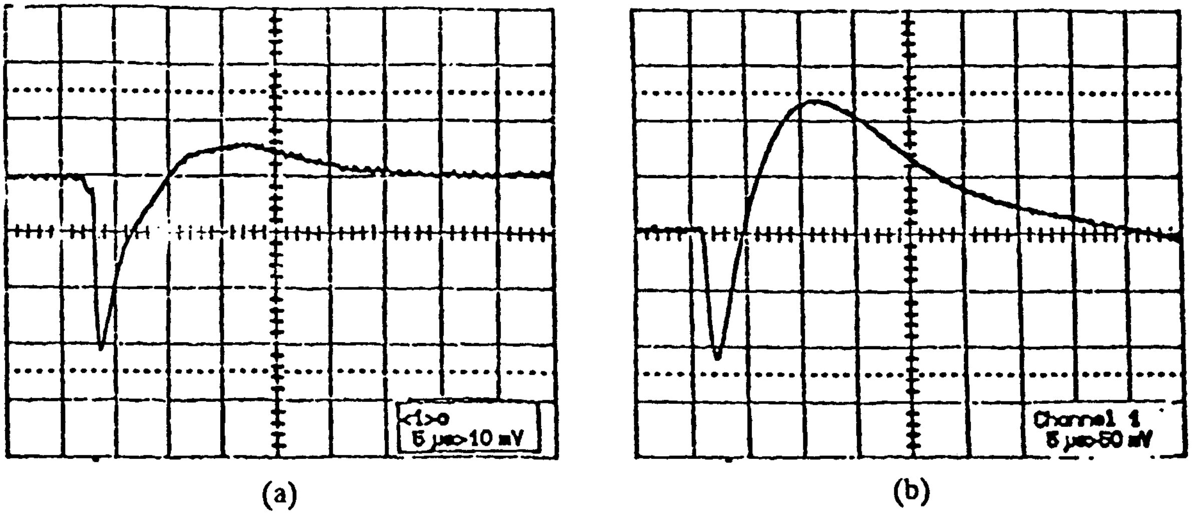


Figure 2. Two-photon OGE spectrum of neon

tronic states arising from the second excited configuration. All the observed lines were matched with the possible two photon transitions which can be calculated from the term values for neon [9]. The selection rules for the two-photon transitions are $\Delta J=0, \pm 1, \pm 2$ and $J=0 \nleftrightarrow J=1$ [10].

The general structure of the optogalvanic signal involves two main parts as shown in Figure 3. The first part is a fall in the signal which appears about 1 μsec after the laser pulse. This is generally the predominant part of the signal and corresponds to drop in the impedance of the discharge relative to the steady-state



(a) (b)

Figure 3. Optogalvanic spectrum for two-photon absorption (a) and one-photon absorption (b)

value. Here the drop in the impedance is caused in part by the increased electron temperature from fast electrons formed through superelastic collisions with atoms in laser-populated excited states. A further part is caused by the increased charge density due to the electrons and ions produced through ionization of laser-populated excited states. The second part of the signal is an increase in the impedance begining ~ 5μsec after the laser pulse, which is dominant only for transitions from metastable levels. There are profound effects on the discharge characteristics when metastable levels are depopulated by the laser, and we observe an increased impedance until the steady-state population of the level is attained. All the observed two-photon optogalvanic signals in the neon had only negative part at the discharge currents less than 1.2 mA, but they showed positive part as well by increasing the current above it. The time dependence of optogalvanic signals on the two-photon transition of $1s_5$ (J=2) 59243A° 4s‴ (J=3) are shown in Figure 4 for different discharge currents.

To compare the two and single-photon absorption signals, an effort was made to record the one-photon optogalvanic spectrum in the available wavelength region. Thirteen one-photon optogalvanic absorptions were observed, in the range between 580 to 635 nm for transitions from the levels arising from the configuration of first excited state, $1s^22s^22p^53s$, and levels of configuration of second excited state, $1s^22s^22p^53p$.

The wavelength of these transition lines along with the energy of their lower and higher levels are tabulated in Table 2. The basic structure of optogalvanic signals for two and one-photon are the same, but the amplitude of two-photon transition signals (especially the positive part of the signal) is much lower than that of single-photon absorption lines although transitions may start from the same lower levels (Figure 3). The reason for this could be due to a greatly reduced cross section for absorption in the two-photon transition relative to one-photon absorption.

It has been shown theoretically that, at resonance, the two-photon transition rate, T, increases with the square of spatial power density of laser beam [11].

$$T(\mathrm{sec}^{-1}) \sim \left(\frac{P}{S}\right)^2 \ \mathrm{W/mm^2}$$

Also, the amplitude of the two-photon optogalvanic signal is proportional to both the illuminated volume V and the two-photon transition rate T:

$$F \sim V \times T \sim V\left(\frac{P}{S}\right)^2$$

To obtain detailed information concerning this dependency, the optogalvanic signal amplitude of $1s_5 \rightarrow 4s'''_1$ two-photon transition was detected for different lens-discharge separations. A plot of the above dependency (Figure 5) depicts that, at the beginning, the two-photon transition probability increases faster than pumped volume reduction by varying the lens-discharge separation followed by a saturation. Further increase in the power density, however, results in the production of uncompensated reduction on the pumped volume and thus decreases the signal amplitude. Then increasing the lens separation further, yields an increase in the amplitude of the signal again.

Conclusion

We have observed two and one photon optogalvanic lines of neon using a very simple detection scheme. The practical benefit of our experiments is the identification of new and useful standard lines. The comparison between two and single-photon tran-

Table 2. One-photon transitions in neon at a discharge current I=6mA

Transitions	$E_i(\mathrm{cm}^{-1})$ - $E_f(\mathrm{cm}^{-1})$	Standard Transition Wavelengths (A°)	Laser Wavelengths (A°)
$1s_2 \rightarrow 2p_1$	135890-152972	5852.48	-
$1s_5 \rightarrow 2p_2$	134044-151040	5881.89	5882.5
$1s_5 \rightarrow 2p_4$	134044-150860	5944.83	5944.8
$1s_5 \rightarrow 2p_5$	134044-150774	5975.53	5975.5
$1s_4 \rightarrow 2p_2$	134461-151040	6029.99	6031.0
$1s_4 \rightarrow 2p_3$	134461-150919	6074.33	6074.9
$1s_4 \rightarrow 2p_4$	134461-150860	6096.16	6096.6
$1s_4 \rightarrow 2p_5$	134461-150774	6128.45	6128.1
$1s_5 \rightarrow 2p_6$	134044-150318	6143.06	6143.0
$1s_3 \rightarrow 2p_2$	134821-151040	6163.59	6163.8
$1s_5 \rightarrow 2p_7$	134044-150124	6217.28	6217.8
$1s_3 \rightarrow 2p_5$	134821-150774	6266.49	6266.5
$1s_4 \rightarrow 2p_6$	134461-150318	6304.78	6304.6
$1s_5 \rightarrow 2p_8$	134044-149826	6334.42	6334.8

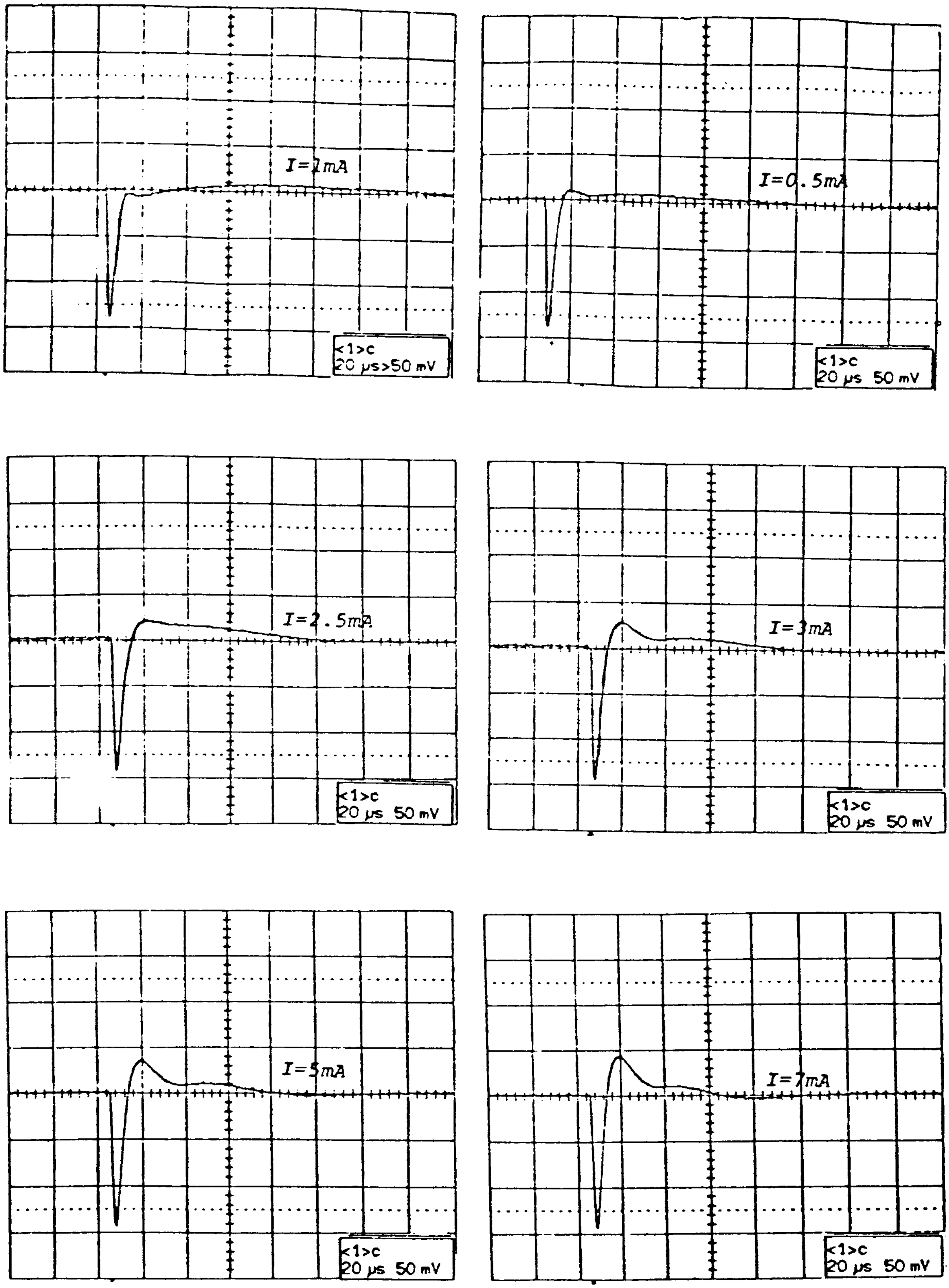


Figure 4. Temporal behavior of $1s_5 4s_1'''$ two-photon absorption optogalvanic signals at different discharge currents

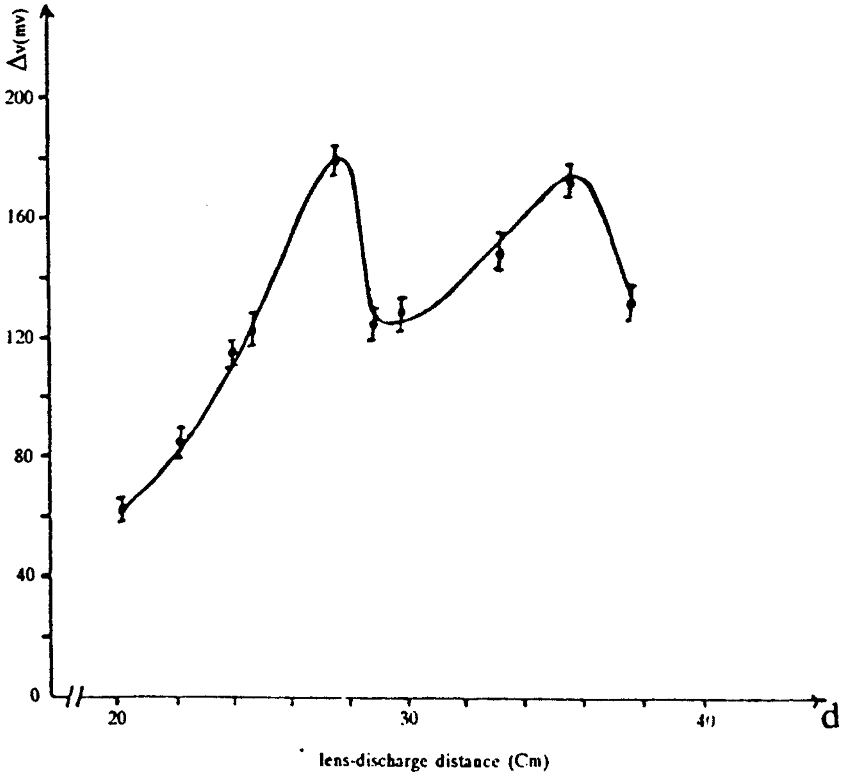


Figure 5. Two-photon signal amplitude ve lens (f=30 Cm)- discharge separation

sitions, particularly those originating from the same lower level, shed light on the dynamics of OGE. The results obtained in the visible region confirmed that our equipment is quite suitable for observation of neon two-photon laser induced absorption and allowed the prediction of the optimum power density for UV transitions.

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