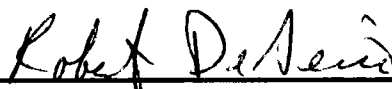


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Robert DeSerio, Major Professor

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7/28/89

**A PULSED LASER STUDY OF THE OPTOGALVANIC
EFFECT IN A NEON GLOW DISCHARGE**

A Thesis

Presented for the

Master of Science

Degree

The University of Tennessee, Knoxville

Glenn Harman

August 1989

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DEDICATION

To the memory of my brother, Tipton Willis Harman.

ACKNOWLEDGEMENTS

I would like to thank my major professor, Dr. R. DeSerio, for his guidance and technical assistance that made the completion of this thesis possible. I would also like to thank Dr. S. Georghiou and Dr. R. Lide for their support. Special thanks are in order to Dr. D.R. James of ORNL for supplying me with references on glow discharges and to Dr. R.J. Van Brunt of NIST for references on the optogalvanic effect. And suggestions by Dr. I. Sauers of ORNL were instrumental in the compilation of this thesis.

I humbly appreciate the emotional and physical support provided by my wife, Mary.

Finally, I thank the Physics department at the University of Tennessee, Knoxville for the interesting experience of graduate study in physics.

ABSTRACT

The optogalvanic effect in a neon glow discharge has been studied. Time-resolved voltage changes in the discharge induced by a 500 ps pulsed dye system were observed. The energy per laser pulse was varied from 20 - 150 μ J. The discharge current was varied from 0.2 - 1.5 mA at neon pressures of 4 and 20 torr. All dipole-allowed $1s^22s^22p^53s$ - $1s^22s^22p^53p$ transitions in the 585 - 700 nm wavelength range were observed. The signals observed were from all four levels $^1P_1$, $^3P_{0,1,2}$ of the first excited configuration. The largest signals came from the metastable $^3P_0$ and $^3P_2$ states demonstrating that they are important in the discharge. Those involving the $^1P_1$ state show possible indications of inverted populations. Time-resolved signals from transitions involving the $^3P_1$ states are presented for the first time. Their shapes may indicate a more dominant role in the ionization process for this ground state coupled state than has previously been suggested.

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INTRODUCTION

The optogalvanic effect (OGE) is a process in which a voltage change in an electrical discharge is induced by optical excitation. When the incident light is resonant with electronic transitions in the discharge, the changes in concentrations of the discharge species due to absorption or stimulated emission change the impedance of the gas and a voltage change takes place. The sign of the voltage change can be either positive or negative.

The phenomenon was first studied by Penning 60 years ago [Penning, 1931]. By optically exciting a neon glow discharge with the light from a second neon light he studied the effect of metastable neon atoms on discharge starting potentials. The study lapsed over the years but the advent of tunable lasers revived interest in the field. The ability to select many transitions enabled the OGE to be a powerful experimental method. The study has been combined with mass spectrometry [Smyth *et al.*, 1977] and with doppler-free spectroscopy [Lawler *et al.*, 1979]. Using chopped continuous wave (CW) lasers, Smyth and Schenck [Smyth and Schenck, 1978] and Zalewski *et al.* [Zalewski *et al.*, 1979] have studied the OGE in neon in detail. Their studies reaffirmed the importance of metastable atoms in a neon discharge. Enhancement of the metastable atoms lowers the impedance of the gas and the depletion of the metastable atoms increases the impedance of the gas. The chopped CW laser studies reveal that both positive and negative voltage changes are induced in a single excitation. The time dependence of the impedance change is best studied with a pulsed laser.

Erez, Miron, Shuker and others, all with the Nuclear Research Centre-Negev, Beer-Sheva Israel, have observed the OGE in neon with pulsed laser excitation [Erez *et al.*, 1979], [Miron *et al.*, 1979], [Shuker *et al.*, 1982]. They reported on OGE signals for transitions

originating in two states of the first excited level of neon. In the present study, original data is presented for transitions from all four states of the first excited level of neon. These signals are observed as functions of the discharge current and the laser intensity at different neon pressures.

CHAPTER 1

BACKGROUND INFORMATION

A study of the OGE covers several bodies of knowledge. This chapter provides rudimentary information on areas of interest. The properties of the states of the lower energy levels of neon are discussed along with the general aspects of a normal glow discharge. The radiative and collisional processes involved in maintaining equilibrium concentrations of atomic species in the discharge are outlined.

1.1 Neon

Neon is an excellent gas for a glow discharge. The ionization energy of this noble gas is large but the first and second excited shells, as shown in Figure 1.1, provide a good pathway for ionization. The red, orange and yellow colors of a neon light are mostly attributed to transitions between these states.

The ground state has a $1s^22s^22p^6$ configuration. The lowest excited configuration is $1s^22s^22p^53s$ with four levels denoted as $1s_i$ ($i = 2, 3, 4, 5$) in Paschen notation. In Russell-Saunders ($L-S$) notation, these levels are written as $^1P_1$, $^3P_0$, $^3P_1$ and $^3P_2$, respectively. The $^1P_1$ and $^3P_1$ are strongly mixed in neon and the spin quantum number implied by this notation is only approximate. Nonetheless, the $L-S$ notation will be used to refer to these lowest excited states. The $^3P_0$ and $^3P_2$ states are metastable (with radiative lifetimes on the order of seconds) [Smyth and Schenck, 1978] since transitions directly to the ground state are forbidden by electric dipole transition rules. The $^1P_1$ and $^3P_1$ states have radiative lifetimes on the order of nanoseconds [Smyth and Schenck, 1978]

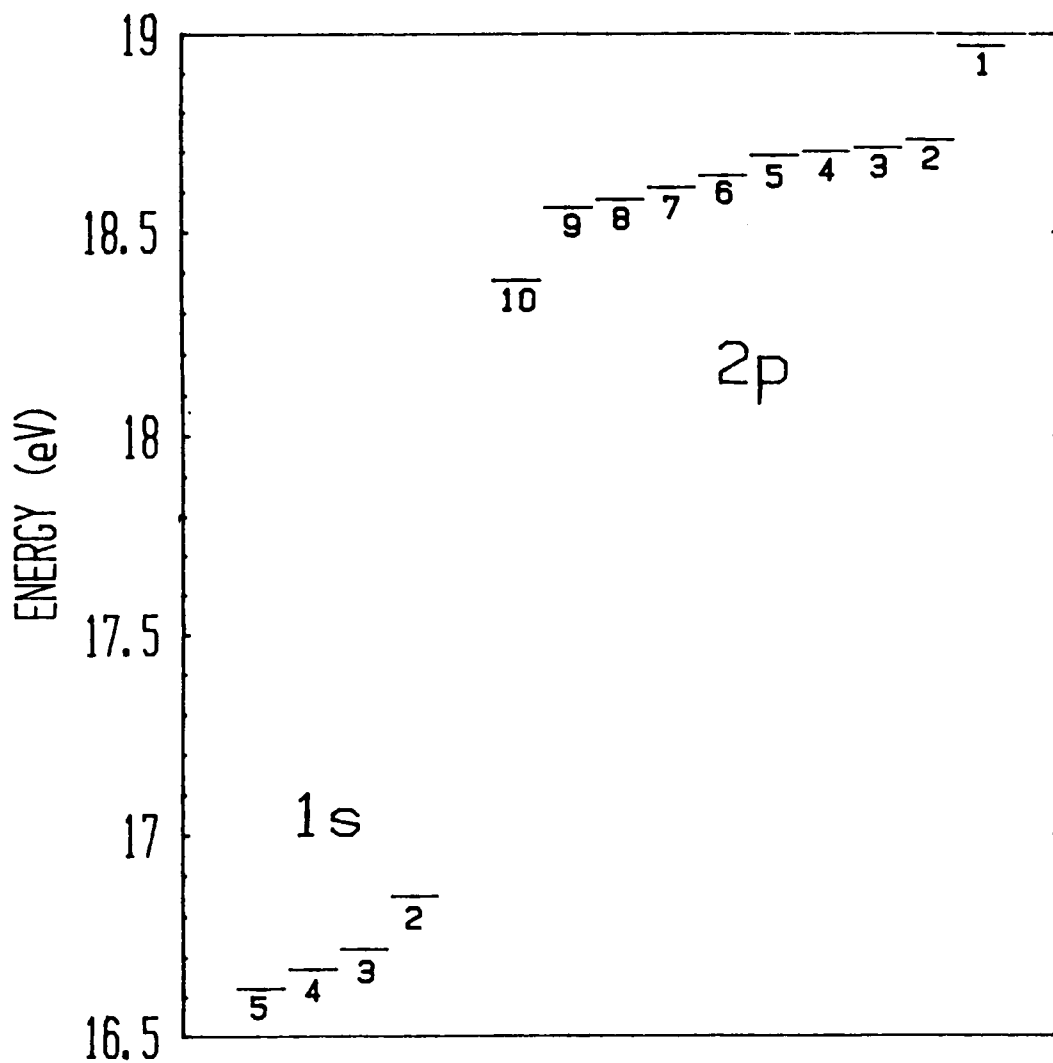


Figure 1.1: The first and second excited state energy levels for neon. The ionization potential for neon is 21.56 eV. Paschen notation is given. The 1s; ($i = 2, 3, 4, 5$) states are the $^1P_1$, $^3P_0$, $^3P_1$ and $^3P_2$, respectively, in Russell-Saunders notation.

but radiation trapping can effectively increase their lifetimes several orders of magnitude at normal discharge pressures [Kane, 1984].

The next excited configuration is $1s^2 2s^2 2p^5 3p$ resulting in ten levels which will be referred to in Paschen notation as $2p_i (i = 1, 2, \dots, 10)$. These $2p$ states have short radiative lifetimes of approximately 20 nanoseconds [Chang and Setser, 1980] and are not affected much by radiation trapping. Figure 1.2 illustrates the allowed transitions between the $2p$ and the $1s$ shells. Ten possible transitions are ruled out in the L-S coupling scheme.

1.2 Glow Discharge

The glow discharge in this experiment is clearly delineated into regions typical of normal glow discharges as seen in Figure 1.3. The details of the regions are covered in depth by others [Kunhardt and Luessen, 1983], [Nasser, 1971], [Loeb, 1965]. Basically, electrons boil off the cathode and collide with incoming positive ions. These ions are excited to higher energies and subsequently are neutralized with high energy photons being emitted to create the cathode glow. The Crook's dark space is a high field region where few electron-neon collisions take place. Electrons are accelerated to high energies here. Entering the negative glow region, the electrons are too energetic but with collisions they lose energy and begin to scatter and ionize. The brightest part of the entire discharge is located a small distance into the negative glow. The electrons lose energy throughout the negative glow region due to collisions and diminished ion space charge. The low energy electrons emerging from the negative glow do not produce ionization and the subsequent reduction in luminosity typifies the Faraday dark space. This is a high field region where electrons gain enough energy to keep the positive column supplied with current. The positive column is a nearly neutral region with only enough electrons and ions present to keep the column conducting. The region is characterized by a low

	$^1P_1$	$^3P_0$	$^3P_1$	$^3P_2$
$2p_1$	585.2	$J=0 \nleftrightarrow J=0$	540.1	$\Delta J \neq 2$
$2p_2$	659.9	616.4	603.0	588.2
$2p_3$	665.2	$J=0 \nleftrightarrow J=0$	607.4	$\Delta J \neq 2$
$2p_4$	667.8	$\Delta J \neq 2$	609.6	594.5
$2p_5$	671.7	626.6	612.8	597.6
$2p_6$	693.0	$\Delta J \neq 2$	630.5	614.3
$2p_7$	702.4	653.3	638.3	621.7
$2p_8$	717.4	$\Delta J \neq 2$	650.6	633.4
$2p_9$	$\Delta J \neq 2$	$\Delta J \neq 3$	$\Delta J \neq 2$	640.2
$2p_{10}$	808.2	743.9	724.5	703.2

Figure 1.2: Wavelengths for transitions between the first and second excited levels of neon. The excited states of the first level are situated across the top and the excited states of the second level are situated down the left hand side. The transitions for which OGE signals were recorded in this study are in bold face.

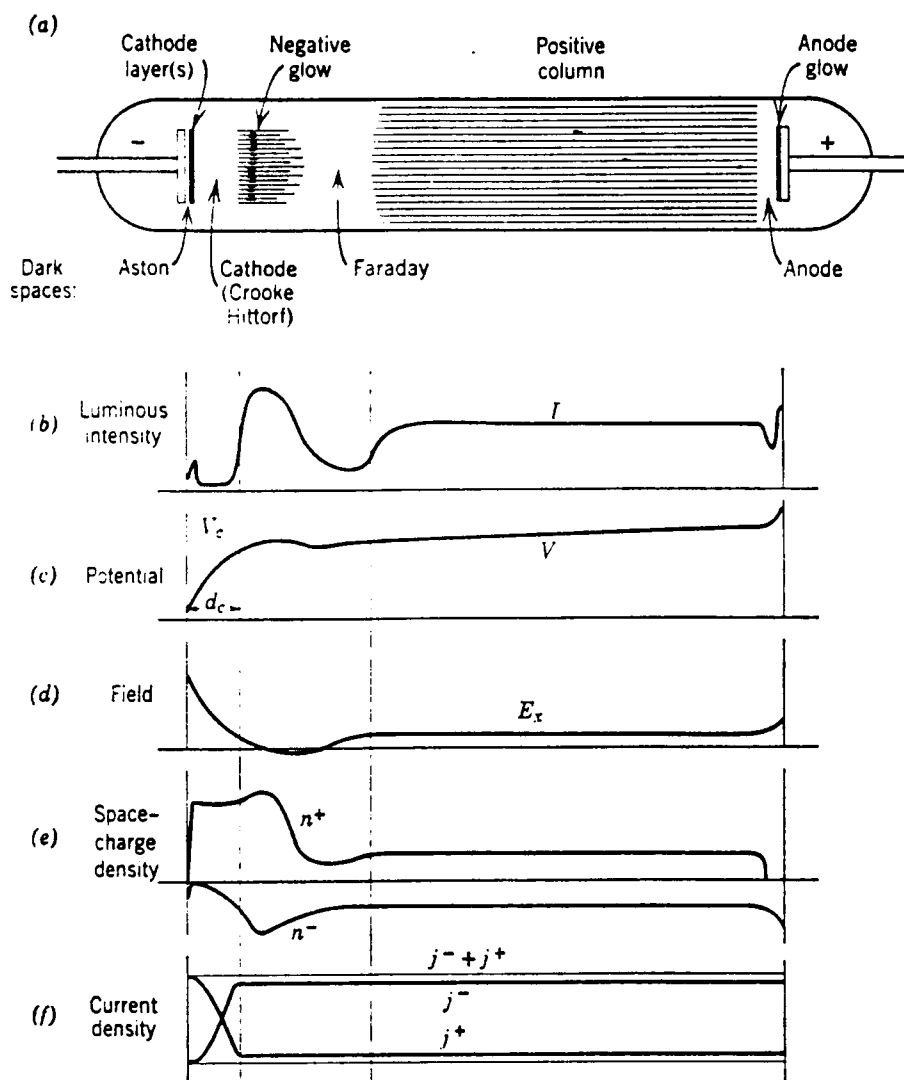


Figure 1.3: Normal glow discharge in neon in a 50 cm tube at $p = 1$ torr. The luminous regions are shaded. The 1 cm tube used in this work displayed a similar structure.

Source: Nasser, E., Fundamentals of Gaseous Ionization and Plasma Electronics

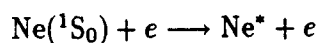
potential gradient and thus electron collision energies are low. Photoemission in this region is due to deexcitation of excited states and not recombination. The anode dark space is another high field region. Here the electrons are energized enough to cause ionization and create the anode glow. The ions created here supply the positive column.

Measurements are taken in the positive column for two reasons. First, the electron and ion concentrations are equal which helps in modelling. Second, measurements are more reproducible in the positive column. There is not much variation of OGE signals within the positive column due to the low potential gradient and the large size of the column makes it easy to isolate from the other regions of the discharge. Other areas of interest to the present study are the areas where electron and ion concentrations are high. The largest electron density is located in the brightest part of the discharge, the negative glow. Secondly, there are large numbers of electrons and ions in the cathode glow and the anode glow.

1.3 Discharge Processes

A rate equation analysis will not be attempted but a presentation of important processes involved in a glow discharge is in order. These are processes in which the atomic species, electrons and ions are created and destroyed. The processes considered are listed below. Sources for rate constants are listed with each process.

1. Electron impact excitation:



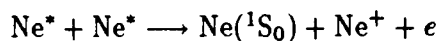
[Kane, 1984], [Lawler and Doughty, 1983]

2. Electron impact ionization:



[Kane, 1984], [Lawler and Doughty, 1983]

3. Atom - Atom collisions:

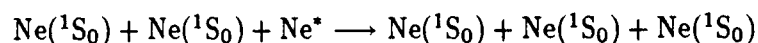


[Kane, 1984]



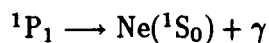
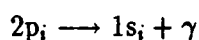
[Leichner *et al.*, 1975], [Cohen *et al.*, 1977]

4. Three body collisional quenching:



[Leichner *et al.*, 1975]

5. Spontaneous emission:



[Weise, 1980]

Radiation trapping should be considered for transitions involving the ground state. Rate constants have not been found for trapping involving the $^1\text{P}_1$ state but Kane [Kane, 1984] says trapping reduces the effective emission rate for the $^3\text{P}_1 \longrightarrow \text{Ne}(^1\text{S}_0) + \gamma$ process by over three orders of magnitude. Diffusion is considered to maintain neon pressure and

recombination is assumed to maintain charge balance. Atomic species populations are given by Kane [Kane, 1984] and Zalewski [Zalewski *et al.*, 1979].

CHAPTER 2

THE EXPERIMENT

The experimental setup, illustrated in Figure 2.1, consists of four parts: the vacuum chamber housing the discharge, the laser system, the discharge power supply, and the data acquisition system.

The chamber used to house the discharge is a stainless steel 6-Way "Del-Seal" cross with 2.73 inch diameter flanges. Two flanges contain glass windows to allow the laser beam to pass through the chamber. Two steel electrodes, 8 mm in diameter, 10 mm apart are situated within a glass tube to contain the discharge. The electrodes are held with an assembly of alumina rods such that the laser beam can pass between them. A diffusion pump evacuates the chamber to below 10^{-5} torr, measured with an ionization guage. The chamber is filled with research grade neon, having a purity of 99.999%. Gas pressures are varied from 4.0 to 20.0 torr and measured with a convectron gauge.

The pulsed dye laser system used is made by PTI Lasers, Inc. A model PL202 dye laser is pumped by a model PL2300 nitrogen laser. The laser pulse width is 0.5 ns with typical energies per pulse of 150 μ J. Repetition rates were typically 2 Hz. Rhodamine B dyes are used to scan the 570-700 nanometer wavelength range. The laser beam is directed with two front silvered mirrors to a cylindrical lens in front of the discharge cell. This lens directs a 1 mm wide, 1 cm tall beam transversely between the two plane electrodes. The laser beam is polarized axial to the discharge. For intensity variation, a pair of polarizers is inserted in the beam.

There are two 20 $k\Omega$ potentiometers in series with the fixed 500 V floating power

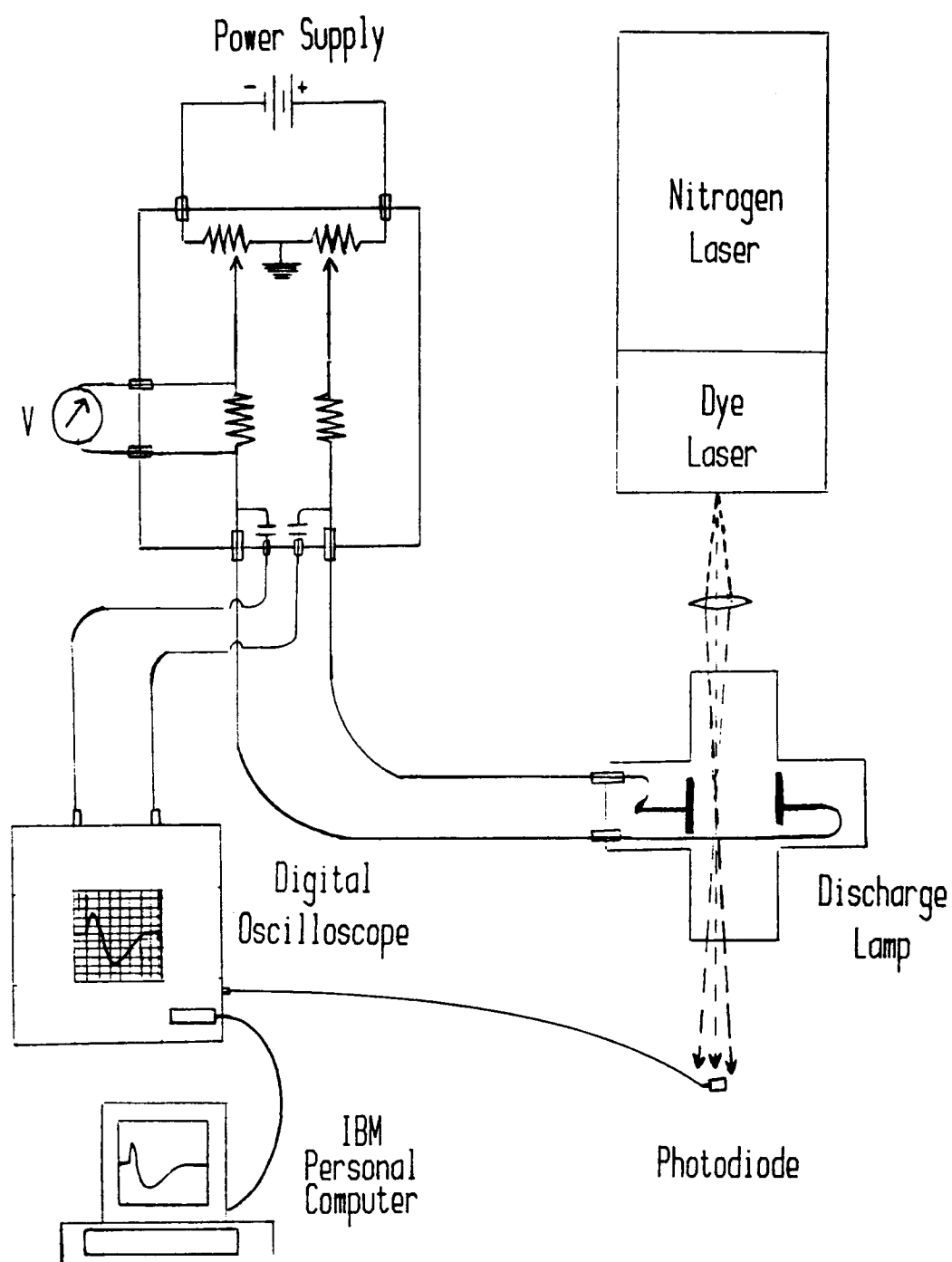


Figure 2.1: Experimental setup for observation of OGE.

supply. They were used to adjust the overall voltage, V , across the discharge and ballast resistors while keeping each electrode charged roughly equal and opposite so that neither arced to the chamber walls. Two $100\text{ k}\Omega$ ballast resistors stabilize the discharge. The voltage across one of the ballast resistors was used to determine the discharge current. The discharge is operated at the low end of the normal glow region: $0.2\text{--}1.5\text{ mA}$ and $210\text{--}240\text{ V}$ anode-cathode voltage. Figure 2.2 illustrates the steady state voltage-current relationship for a pressure of 20 torr.

The OGE signal was AC coupled with two $1\text{ }\mu\text{f}$ capacitors to a 7704a storage oscilloscope system by Tektronix including the 7D20 programmable digitizer. The effect of the ballast resistor in series with the discharge is that a small change in the impedance of the discharge is accompanied by a proportional change in discharge voltage. The voltage

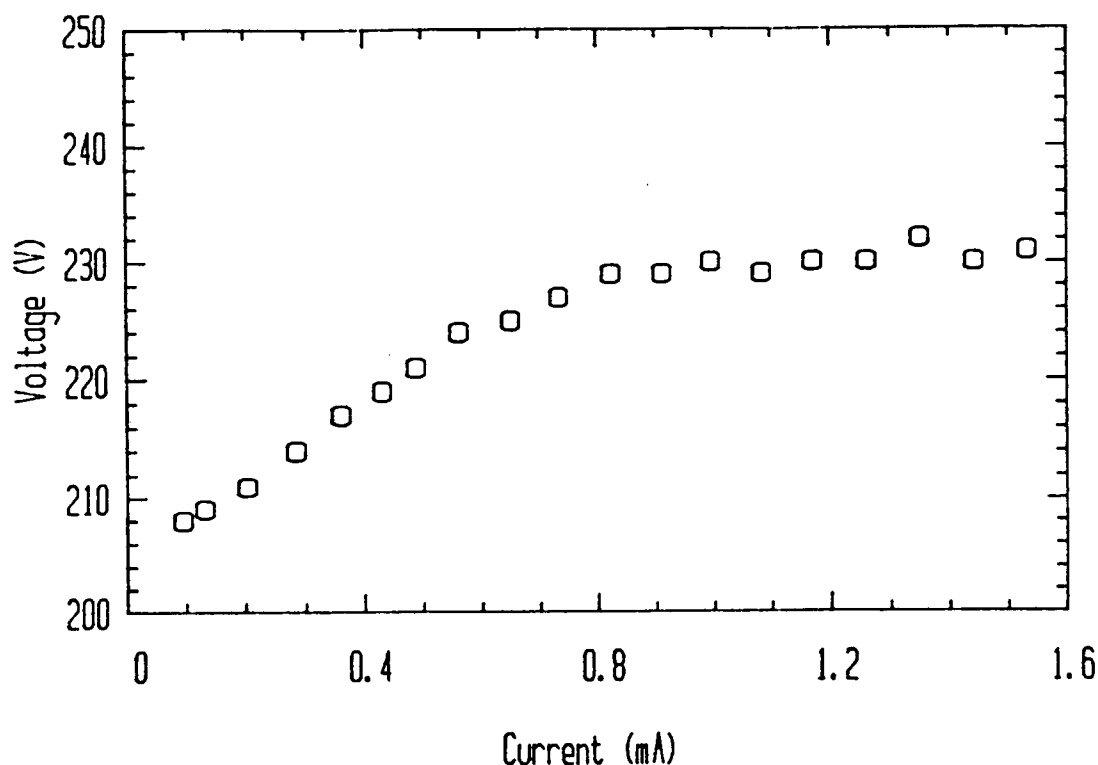


Figure 2.2: Measured voltage versus current relationship for neon glow discharge. Neon pressure: 20 torr.

signals resulting from laser irradiation were fed into the oscilloscope in a manner such that an increase in ionization in the discharge is seen as a drop in voltage. A photodiode, positioned at the exit window of the discharge chamber, triggered the oscilloscope. The digitized signals were transferred to an IBM personal computer with an IEEE 488 interface and Tektronix GURU software.

The signals were averaged for 256 laser pulses. The shape of the signals from different pulses was relatively constant but the magnitudes varied by as much as 30%. The shot-to-shot variation in laser intensity was measured to be less than 5%. The discharge itself was very stable with the noise accounting for far less than 1% of most signals. The bandwidth of the laser is 0.05 nm with wavelength jitter estimated at about 0.01 nm. This jitter is suspected to be the cause of the signal variation. The OGE wavelength sensitivity is shown in Figure 2.3, a shift in wavelength of 0.01 nm introduces a change in magnitude of 28%. Averaging the signals helped alleviate the problem. When the signals are averaged 10 times the magnitude varies less than 10%. All quoted signals were averaged 256 times and were found to vary less than 5% in magnitude and 1% in crossover point times. The data were taken in a manner such that all variations in discharge current, neon pressure, and laser intensity for a particular transition were made without changing the laser wavelength setting.

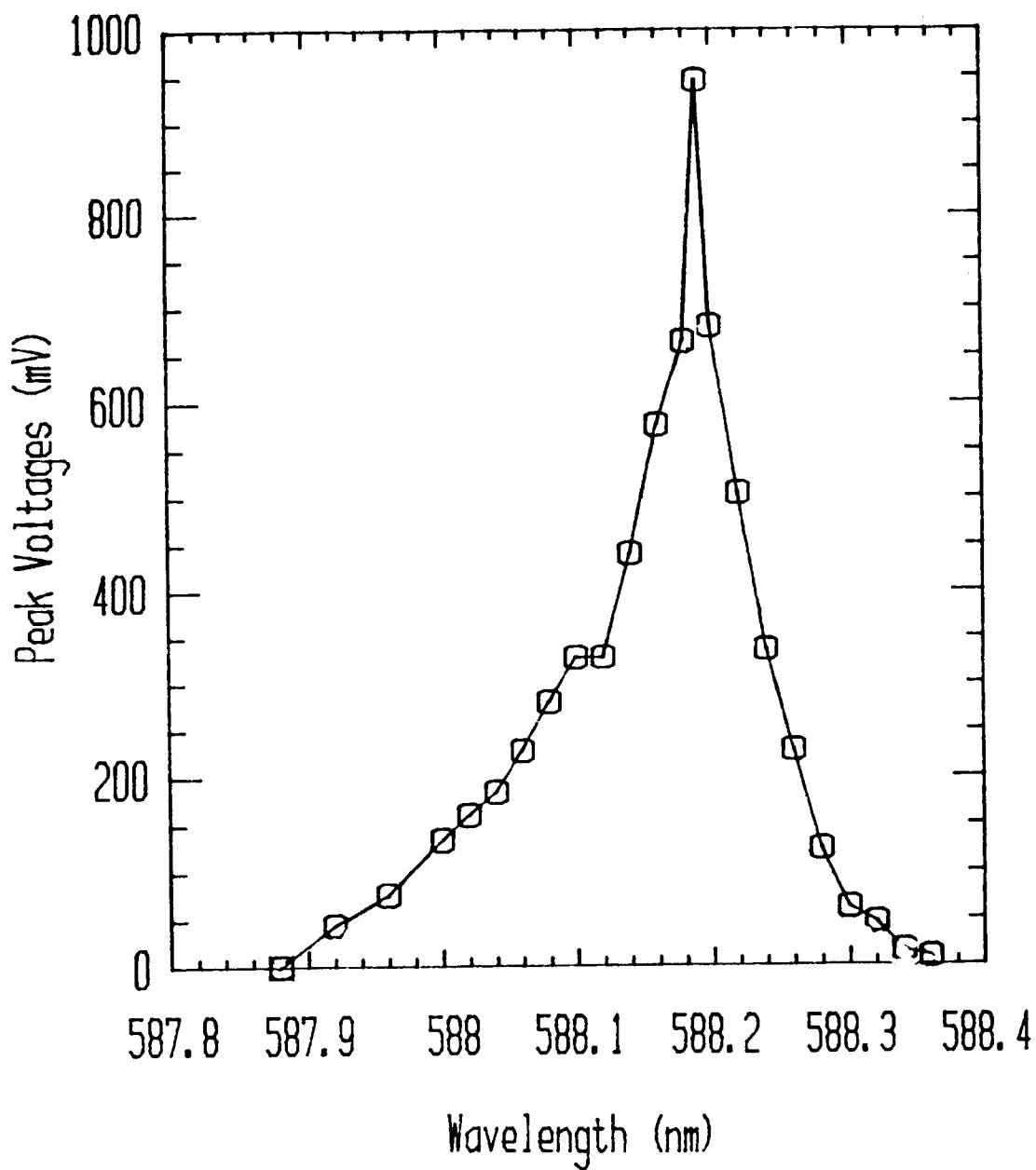


Figure 2.3: Variation of OGE signal magnitudes with laser wavelength for the 588.2 nm transition.

CHAPTER 3

OGE SIGNALS

In this experiment, the optogalvanic effect is seen as the change in a DC voltage with time. All voltage signals portray positive and negative components. Three different general signal shapes are observed for transitions between the 1s and 2p levels. These correspond to transitions involving the $^3P_2$ or $^3P_0$, the $^1P_1$, and the $^3P_1$ excited states. No OGE signals were observed for electric dipole forbidden transitions. With discharge conditions of 1.0 mA and 20 torr neon pressure, the voltage magnitudes range from 4 mV to 500 mV corresponding to voltage changes from 0.002% to 0.2%. The time for reequilibration, T_{eq} , ranges from approximately $50\mu s$ to $400\mu s$. T_{eq} is seen to decrease with increasing current or laser intensity. Also, times for peak maxima and minima lessen in a proportional manner. Generally, T_{eq} decreases with increasing pressure but this is not always true.

3.1 Metastable States

A typical OGE signal for a $^3P_2$ to 2p_i transition is seen in Figure 3.1. The neon pressure is 20 torr and the discharge current is 1.0 mA. The fast (typically $3 - 5\mu s$) initial drop in voltage is followed by an increase to a positive value and a decay back to equilibrium in approximately $100\mu s$. The signal is typical of previously published pulsed OGE signals in neon [Erez *et al.*, 1979], [Miron *et al.*, 1979].

All OGE signals originating in the $^3P_2$ and $^3P_0$ metastable states displayed a form similar to Figure 3.1. Sample signals of each line originating in a metastable state

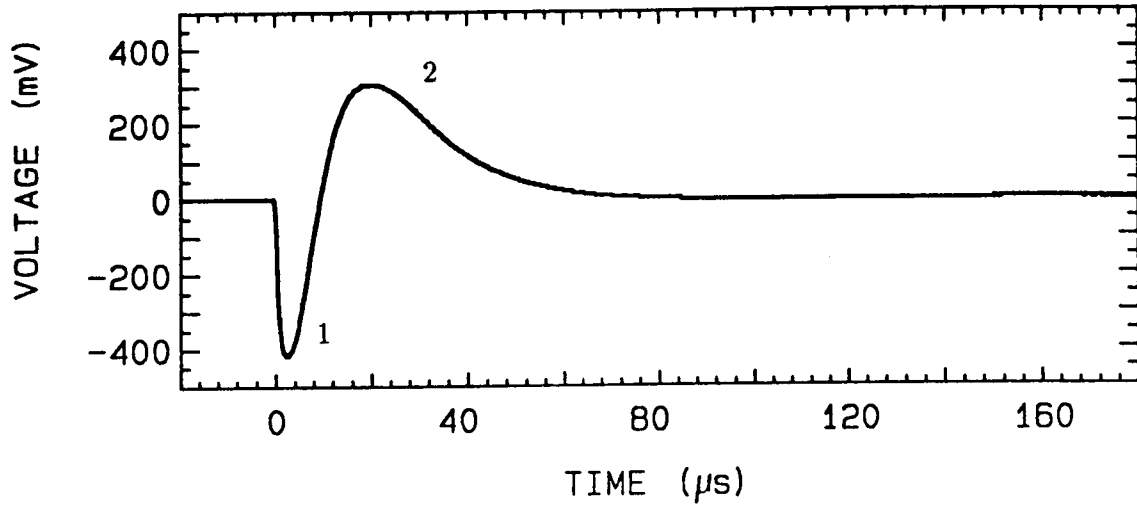


Figure 3.1: Voltage change with time due to 594.5 nm laser pulse. This is a strong OGE signal typical of transitions involving the metastable states of neon. Pressure : 20 torr, discharge current : 1.0 mA.

observed in this work at discharge conditions of $P = 20$ torr and $I = 1.0$ mA are shown in Appendix B and Appendix D. The signals retained their basic form when the discharge was run at different currents and pressures. Although, as is shown in Figure 3.2, at a low pressure of 4 torr and with a 1 mA current, the traces did show another feature: a small voltage drop was seen on the tail end of the signals. This feature disappeared at lower currents and higher pressures. This feature has not been reported before. The effect of increasing the current on OGE signals for the $^3P_2$ to $2p_2$ transition is seen in Figure 3.3. The initial voltage drop, V_1 , attributed to $2p_2$ ionization increases in magnitude relative to the positive voltage peak, V_2 , from a factor of 1.0 to 1.4. The signal is contracted in time also with T_{eq} dropping 31 % as the current is raised from 0.2 mA to 1.5 mA. Similar behavior was noted for all other metastable based OGE signals observed as a function of current.

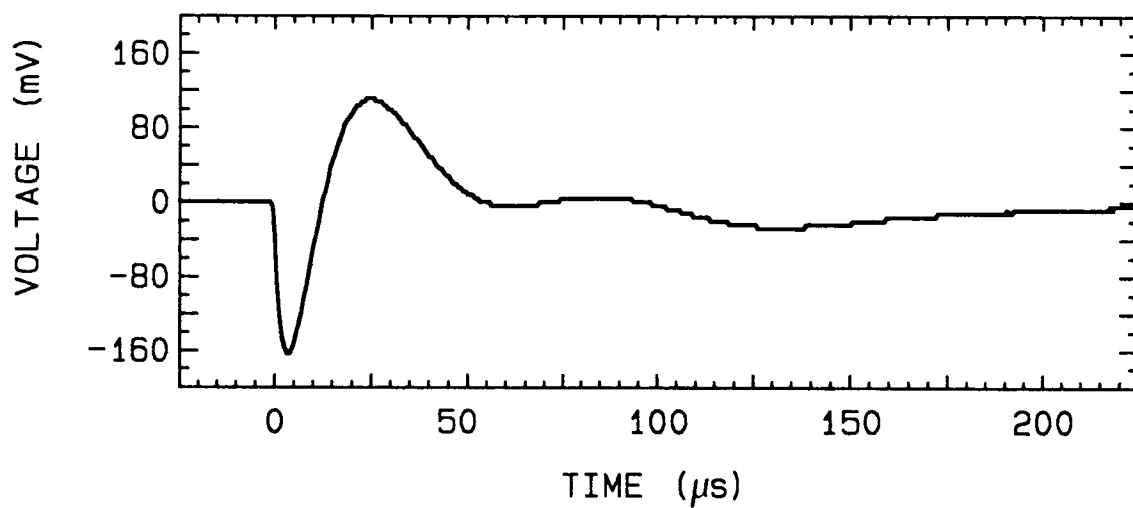


Figure 3.2: Voltage change due to 614.3 nm laser pulse. The neon pressure is 4 torr and the current is 1.0 mA. The second voltage dip is only seen with low pressure, high current discharges for signals involving metastable states.

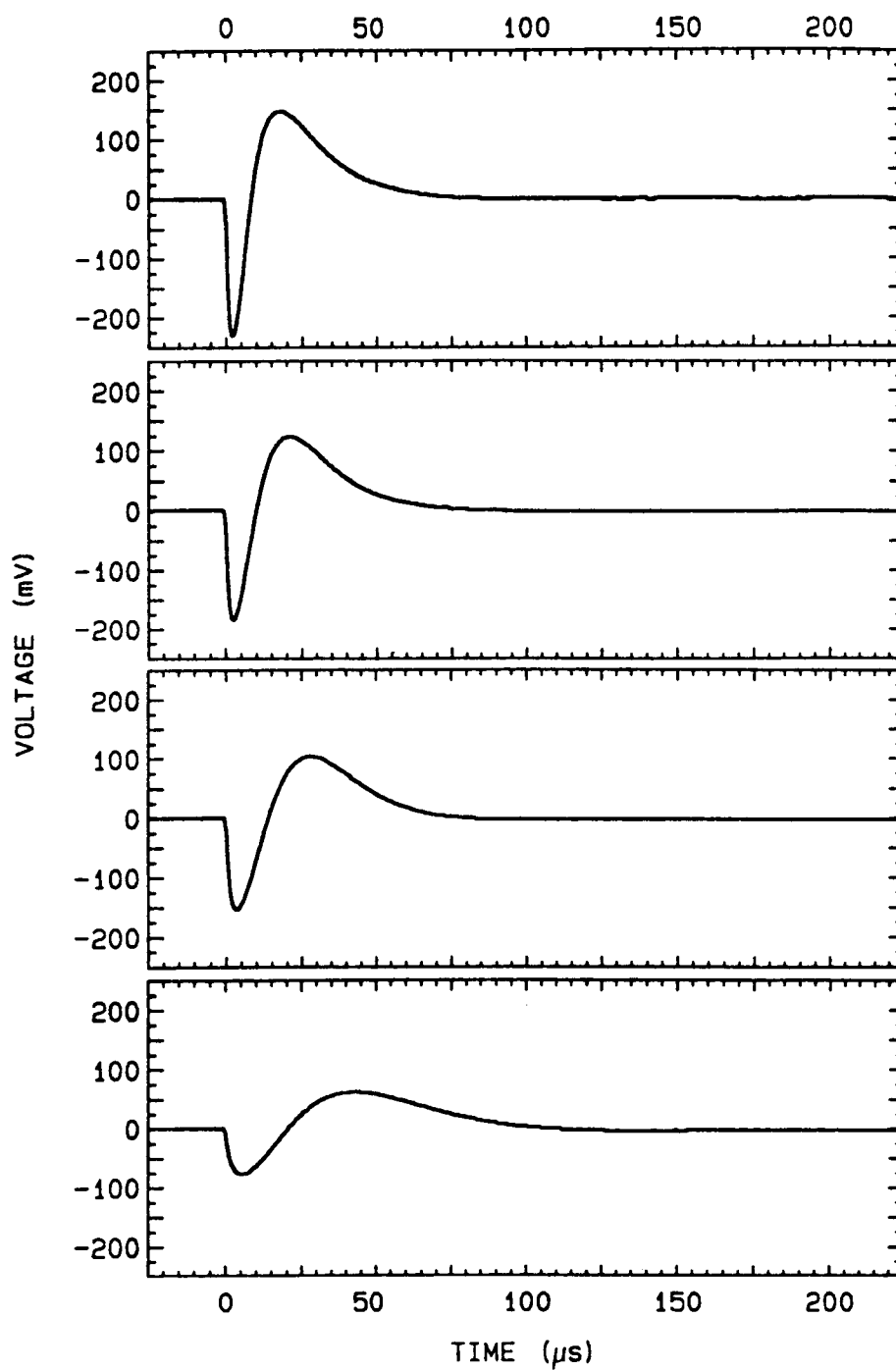


Figure 3.3: Voltage change with time due to 588.2 nm laser pulse for various discharge currents. From bottom to top, the currents are 0.2, 0.5, 1.0, and 1.5 mA. Pressure : 20 torr.

The temporal response of the OGE also varies with the transverse location of the beam in the discharge. This spatial variation is illustrated in Appendix D1. T_{eq} is relatively smaller when the beam is directed at either the end of the discharge than when directed at the positive column. For this low pressure, $T_{eq} = 355 \mu s$ for the signal in the positive column, while the anode glow and cathode glow have reequilibration times of $182 \mu s$ and $236 \mu s$, respectively.

The dependence of the OGE laser intensity for the 588.2 nm line is seen in Figure 3.4. The magnitude of the positive and the negative components of the signal vary with the laser intensity in a mild quadratic fashion.

3.2 Radiative Singlet

When the laser is tuned to a transition involving the $^1P_1$ state, an OGE signal different from those for the $^3P_0$ and $^3P_2$ states is seen. The signal is essentially the mirror image of a metastable signal, as is seen in Figure 3.5. The peak times and reequilibration times are comparable to those of the long lived triplet states. However, the largest voltage changes were only 15% as large as the strongest metastable signals. All six $^1P_1 \leftrightarrow 2p_i$ transitions observed in this work produced OGE signals similar in form. The effect of current, laser intensity, and beam location variation on the OGE signals involving the $^1P_1$ state can be seen in Appendix A2.

3.3 Radiative Triplet

Time-resolved data for $^3P_1 \leftrightarrow 2p_i$ optical excitation are presented for the first time. All transitions originating in $^3P_1$ states produce signals similar to those shown in

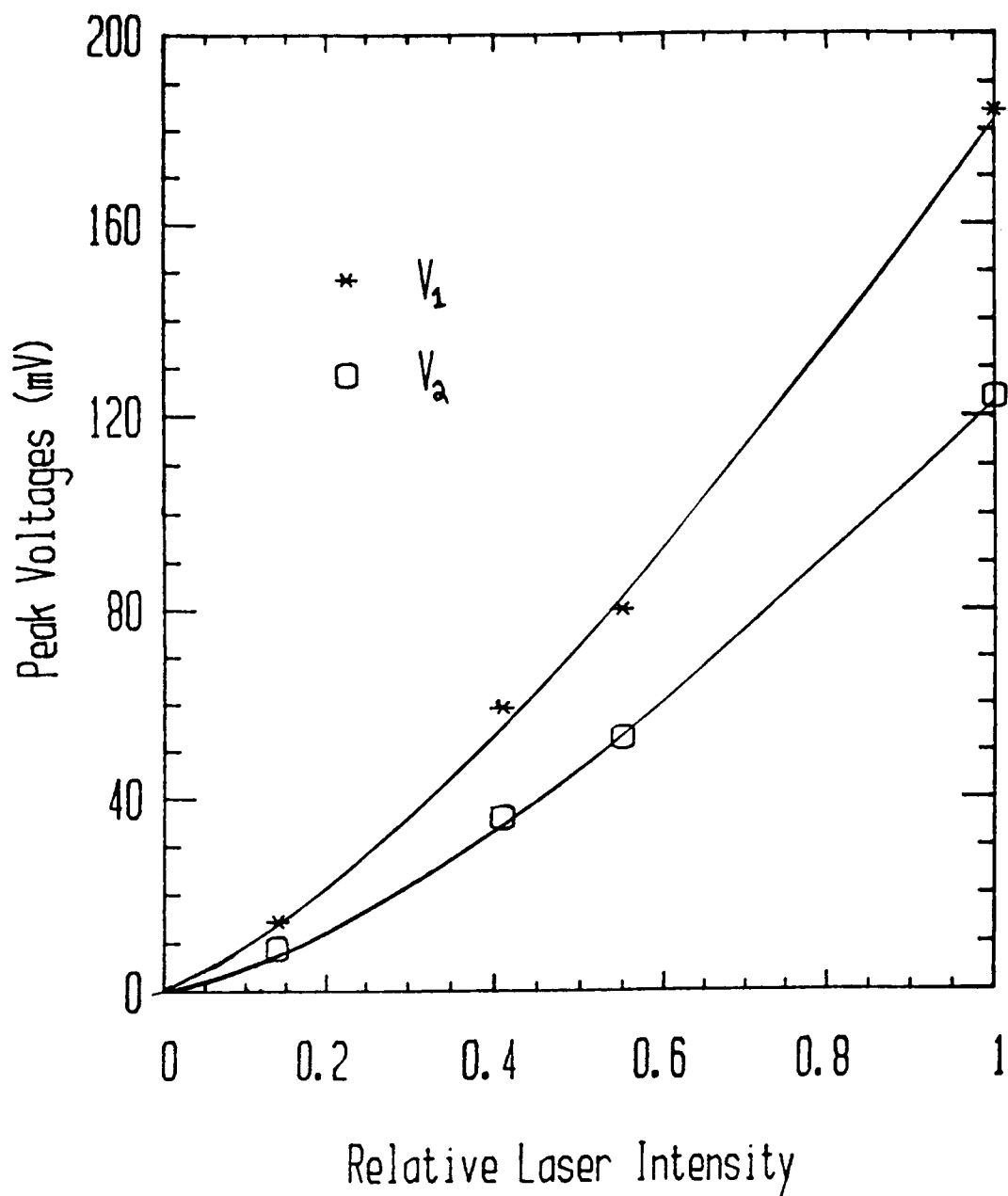


Figure 3.4: Voltage peak magnitudes for the 588.2 nm OGE signal as a function of laser intensity. V_1 and V_2 are the magnitudes of the negative and the positive voltage peaks, respectively. There is an uncertainty of 5% in both variables for each data point. Regression analysis reveals that $V_1 \propto I/I_0 + 0.76 (I/I_0)^2$ and that $V_2 \propto I/I_0 + 0.99 (I/I_0)^2$

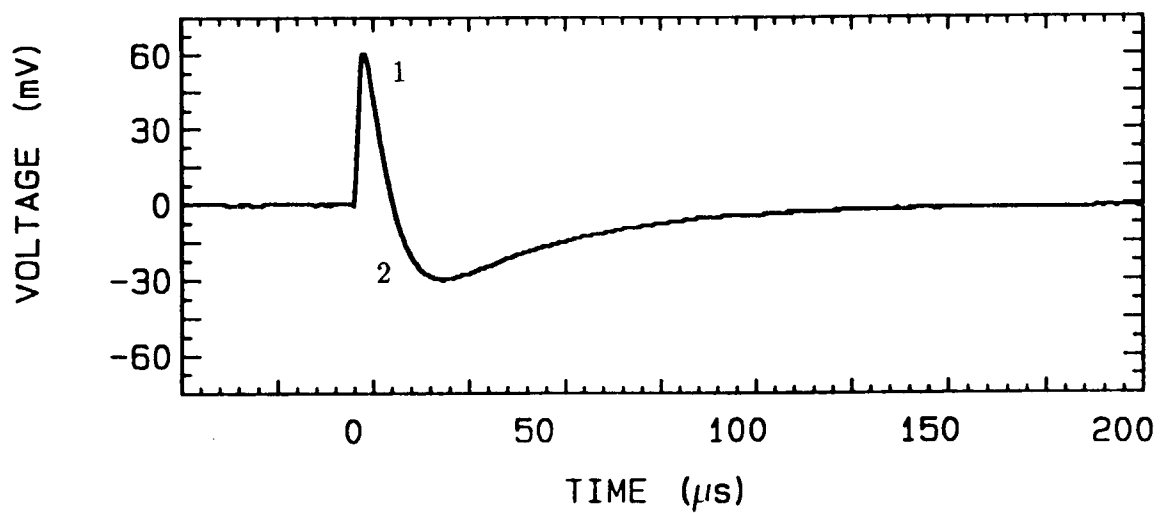


Figure 3.5: Voltage change with time due to 671.7 nm laser pulse. This is an OGE signal typical of transitions involving the $^1P_1$ state of neon. Pressure : 20 torr, discharge current : 1.0 mA.

Figure 3.6 although the relative size of the two voltage drops vary as is shown by Figure 3.7 and Figure 3.8.

The mechanisms of the neon response to this excitation from the $^3P_1$ state to the second excited level are unknown. Table 3.1 shows that explicit dependences of the voltage ratio, $|V_1/V_3|$, or the time of the signal's positive peak, T_2 , on the upper level state, the energy(wavelength), or the transition probability are not there.

$|V_1/V_3|$ is also seen to vary for any particular line with discharge current, laser intensity, or the transverse location of the beam. For the 630.5 nm line, $|V_1/V_3|$ is seen to decrease in Figure 3.9 as the current is lowered and in Figure 3.10 as the laser intensity is reduced. In Figure 3.11 the initial voltage drop is almost seen to disappear as the laser intensity is diminished for the 650.6 nm line. And the difference in the values of $|V_1/V_3|$ for the 603.0 nm line is dramatic for beams transmitted through the different parts of the discharge as illustrated in Figure 3.12.

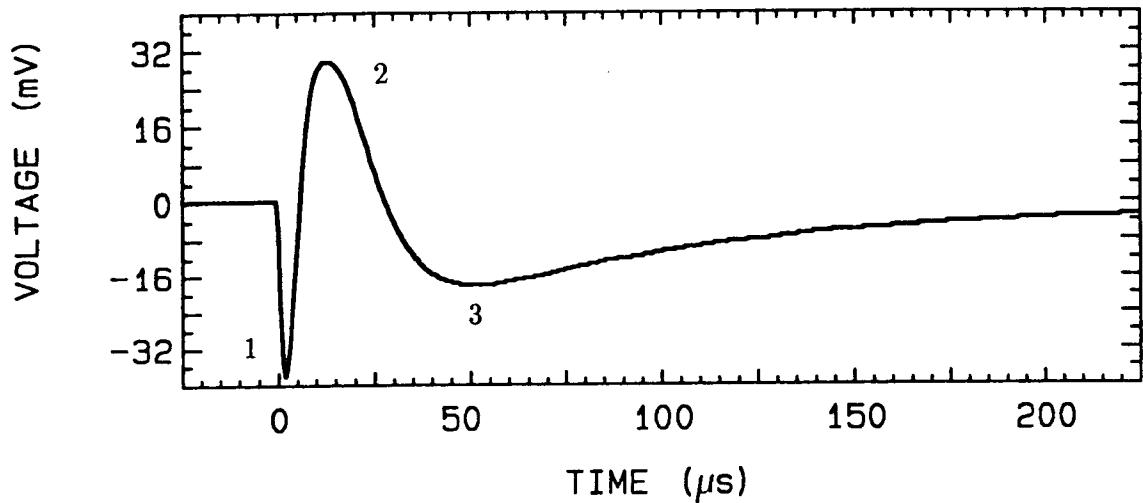


Figure 3.6: Voltage change with time due to 630.5 nm laser pulse. The 630.5 nm transition involves the $^3P_1$ state. Neon pressure : 20 torr, discharge current : 1.0 mA.

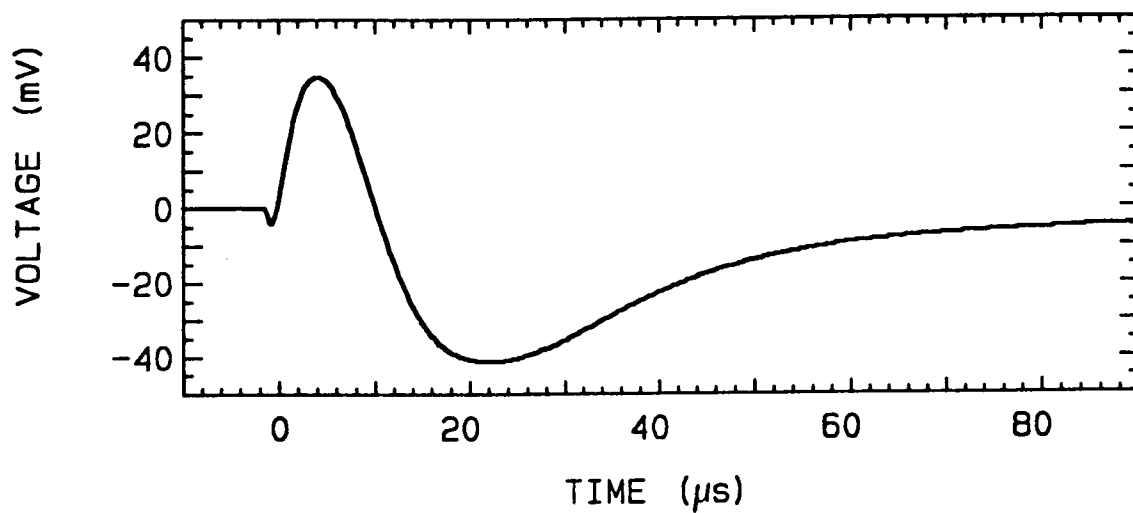


Figure 3.7: Voltage change with time due to 650.6 nm laser pulse. Neon pressure : 20 torr, discharge current : 1.0 mA.

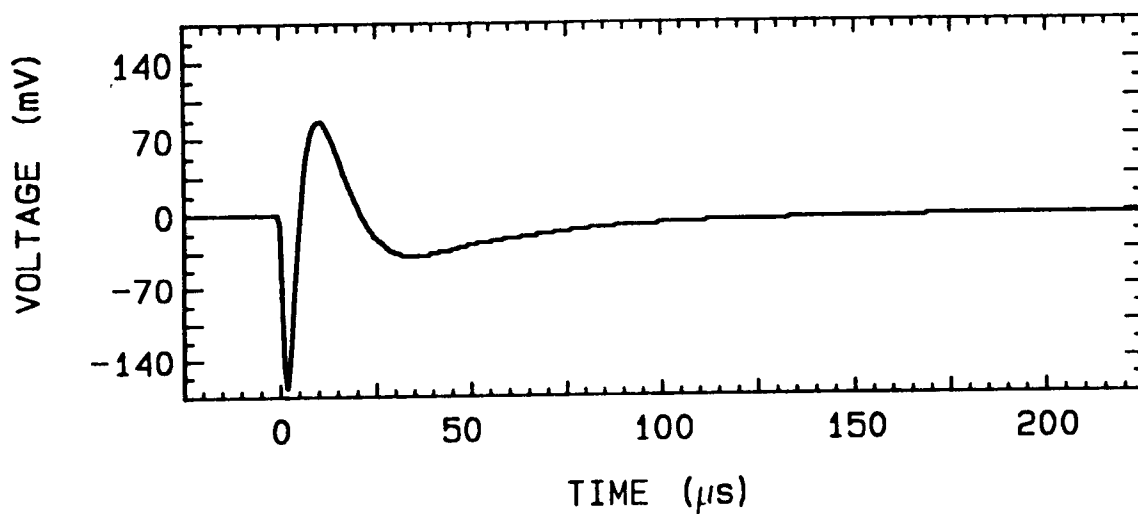


Figure 3.8: Voltage change with time due to 609.6 nm laser pulse. Neon pressure : 20 torr, discharge current : 1.0 mA.

Table 3.1: A comparison between the ratio $|V_1/V_3|$ and T_2 for transitions involving the $^3P_1$ state and various parameters. Note the lack of any complete dependences.

line (nm)	$ V_1/V_3 $	T_2	upper state	transition rate
		$[10^{-6} \text{ s}]$		$[10^8 \text{ s}^{-1}]$
603.0	1.7	9.4	$2p_2$	0.0561
607.4	3.2	11.2	$2p_3$	0.603
609.6	4.4	10.6	$2p_4$	0.181
612.8	3.0	9.2	$2p_5$	0.0067
630.5	0.8	11.2	$2p_6$	0.0416
638.3	0.02	6.2	$2p_7$	0.321
650.6	0.10	5.5	$2p_8$	0.300

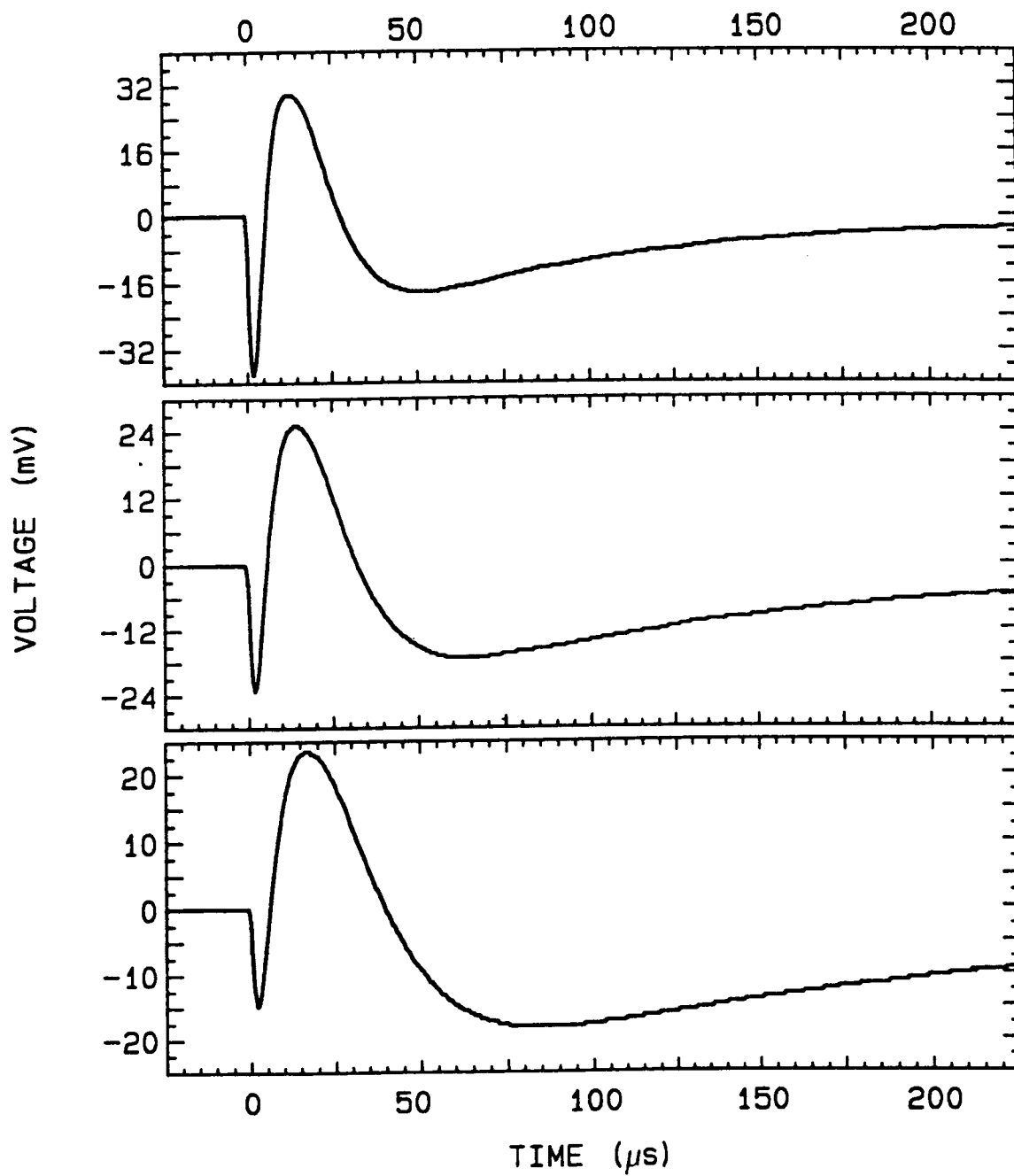


Figure 3.9: Voltage change with time due to 630.5 nm laser pulse for various discharge currents. The current was 1.0, 0.5, and 0.2 mA for the top, middle, and bottom plots, respectively. Neon pressure : 20 torr.

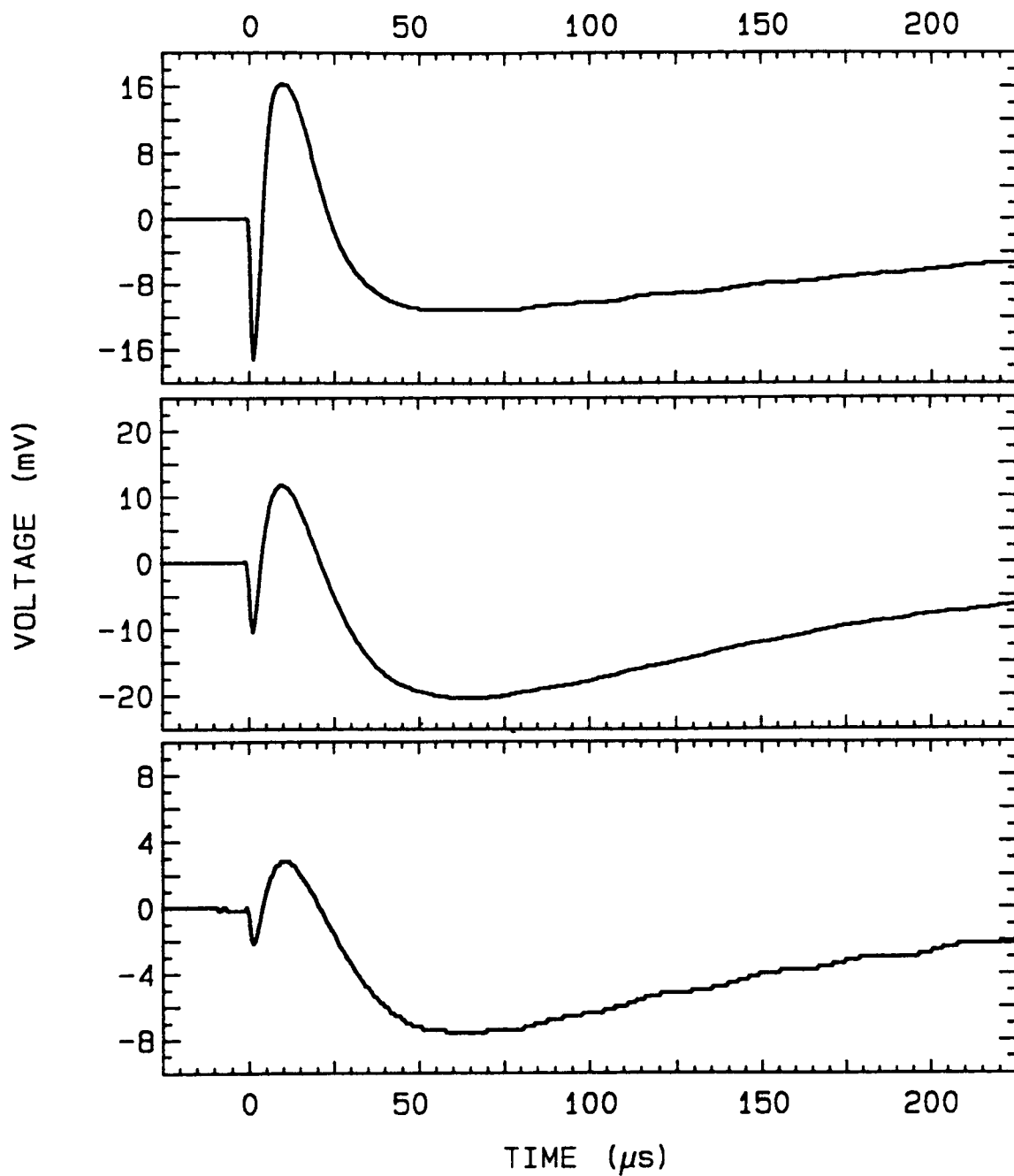


Figure 3.10: Voltage change with time due to 630.5 nm laser pulse for various laser intensities. From top to bottom the laser intensities were 55%, 41%, and 14% of full strength, respectively. Neon pressure : 20 torr, discharge current : 1.0 mA.

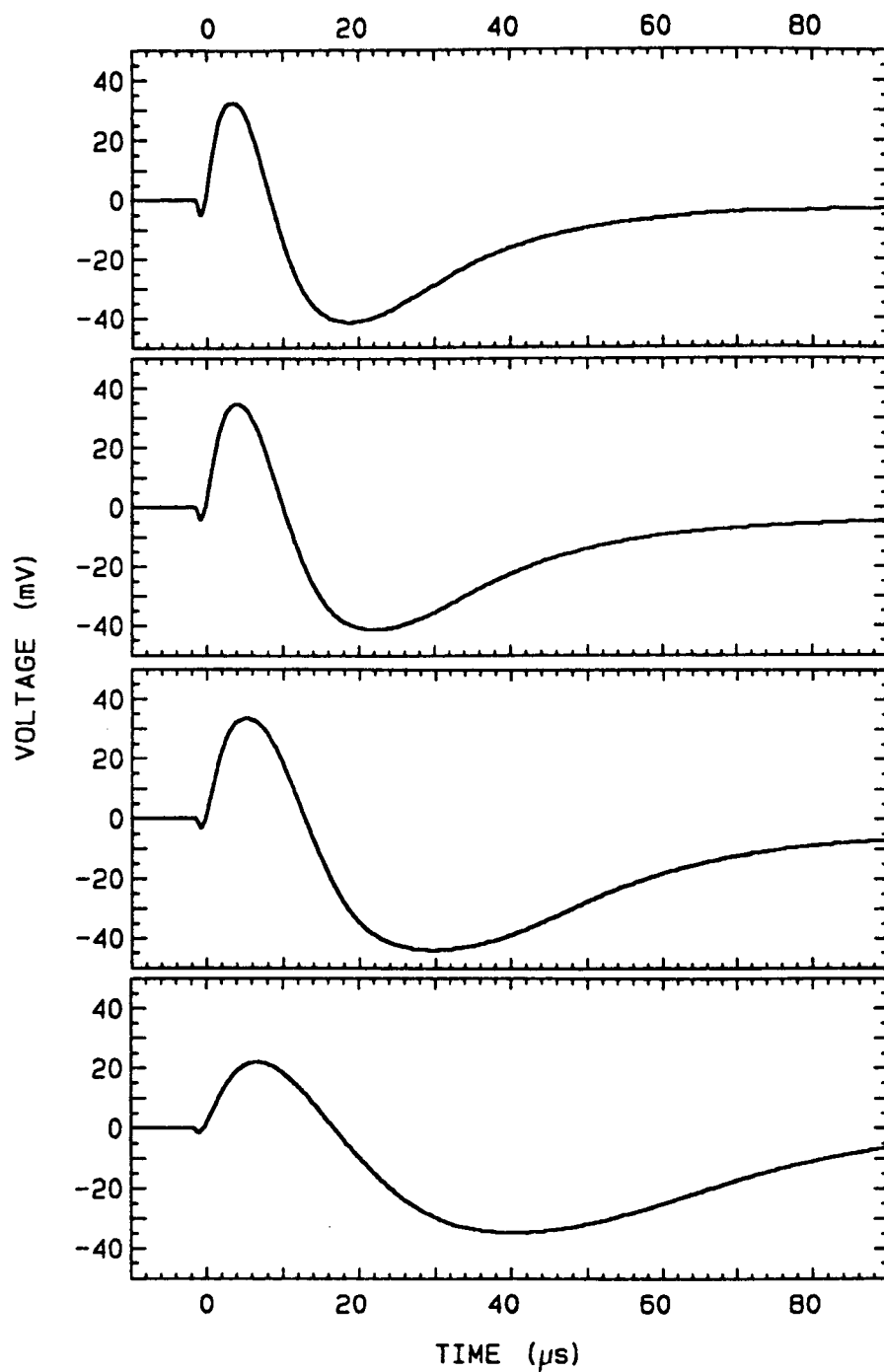


Figure 3.11: Voltage change with time due to 650.6 nm laser pulse for various discharge currents. The currents were 1.5, 1.0, 0.5, and 0.2 mA for the top through bottom plots, respectively. Neon pressure : 20 torr.

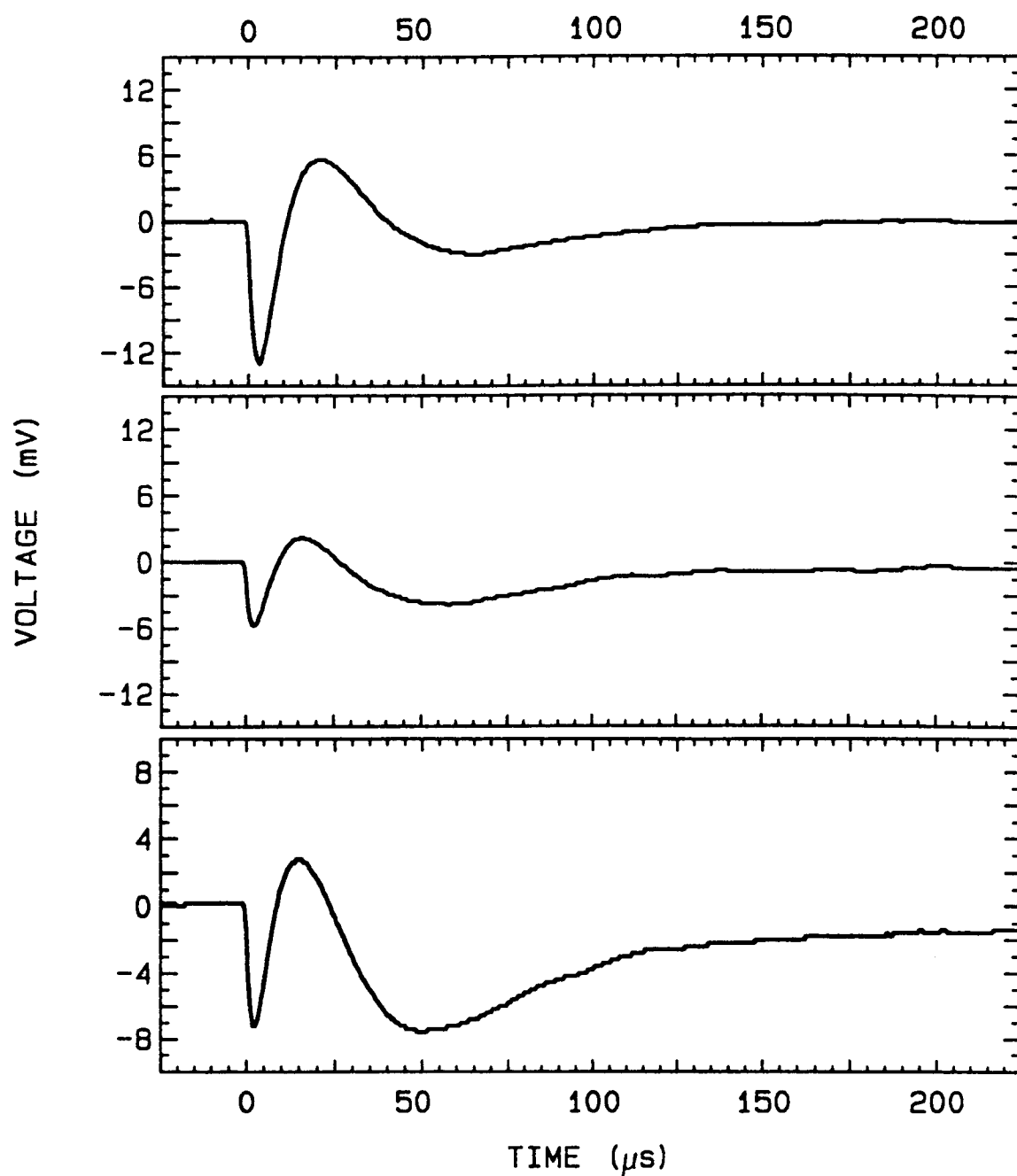


Figure 3.12: Voltage change with time due to 603.0 nm laser pulse for various transverse laser beam locations in the discharge. The signals from top to bottom were from beams passing through the negative glow, positive column, and anode glow, respectively. Neon pressure : 4 torr and discharge current : 0.5 mA.

CHAPTER 4

ANALYSIS

An analysis of the OGE in a neon glow discharge is difficult. Rates for many processes involved in maintaining the discharge are unknown. And the energy dependences of the rates of the collisional processes involving the atomic species are not known. Also, the manner in which a local impedance change is transmitted through a discharge is not well known. Though a theory utilizing a "collection efficiency" for a discharge that was excited by a chopped CW laser has been proposed by Lawler [Lawler, 1980]. Nevertheless, some aspects of the OGE in neon can be understood with some simplifying assumptions.

An explanation of OGE signals involving metastable neon states as proposed by others [Kane, 1984], [Erez *et al.*, 1979], [Kravis and Haydon, 1981] follows: Assume the excitation of $^3P_2$ states to a $2p_i$ level that can decay to any $1s$ level, such as the 588.2 nm transition to the $2p_2$ state. How will equilibrium be reached and what will happen until it does? Immediately after the pulse, the $2p_2$ state has a boosted population and the $^3P_2$ state has a depleted population. The $2p_2$ state has a higher ionization coefficient, $2 \times 10^{-8} \text{ cm}^3\text{s}^{-1}$, than the $^3P_2$ state, $6 \times 10^{-9} \text{ cm}^3\text{s}^{-1}$, [Kane, 1984] and thus an increase in ionization takes place and a voltage drop is seen. As the $2p_2$ state is ionizing, the process of spontaneous emission competes for the surplus states. In fact, spontaneous emission to the $1s$ levels is 10^5 times faster than electron impact ionization for the $2p_2$ state. The destination of this emission is any one of the four $1s$ levels with branching ratios [Weise, 1980] determining the individual decay rates. Thus, the metastable states will effectively lose a portion of their equilibrium concentration to radiative states which

decay to the ground state. The low metastable concentration increases the impedance of the gas and the voltage across the discharge rises accordingly.

The initial decrease in ionization for OGE signals involving the $^1P_1$ states is attributed by Shuker to an inverted population in which upper $2p_i$ populations are driven down to the $^1P_1$ level by the laser [Shuker *et al.*, 1982]. Smyth and Schenck, in CW laser studies, attributed their overall increase in ionization to an initial excitation of $^1P_1$ states to the $2p$ level and the subsequent enrichment of metastable populations by radiative decay [Smyth and Schenck, 1978]. These contradicting theories each correctly predict one part of the time resolved signal but not the other. Smyth and Schenck's "overall increase in ionization" does not explain the initial increase in the impedance of the gas. And Shuker's paper does not present a reason for increased ionization after the $2p_i$ states are deexcited to the $^1P_1$ level. The fact that the signals involving the metastable states and the $^1P_1$ states are mirror images of one another does not mean that the processes involved can simply be reversed. The later part of the metastable signals is attributed to a depletion of metastable states via radiative decay from the pumped $2p$ state. The fast initial voltage peak of the signals involving the $^1P_1$ states, supposedly indicative of an inverted $2p$ state population, was seen for all six $2p \leftrightarrow ^1P_1$ transitions studied. This would mean that each of the $2p$ levels had an inverted population since the laser isolated the states with nanosecond pulses while the mixing times among $2p$ states is on the order of microseconds. Also, there is some evidence to contradict $2p_i$ populations being larger than $^1P_1$ populations. Smyth [Smyth *et al.*, 1977] in CW laser studies found that signals for transitions originating in $2p$ levels were 50-100 times smaller than signals originating in $1s$ levels and attributed the fact to the $1s$ levels having far greater populations than the $2p$ levels. But again, if the populations are not inverted, the initial increase in impedance is unexplainable.

An explanation of the OGE signals for transitions involving the $^3P_1$ state has not been found. The signals from this ground state coupled triplet contain the initial voltage drop characteristic of the metastable signals and the later voltage drop seen in the signals of the ground state coupled singlet state. The positive component of the different $^3P_1$ signals seems to be shifted later in time for the higher lying 2p states such that they are very similar to the signals of the metastable triplets. For the transitions among the $^3P_1$ and lower lying states the positive component of the signal is almost immediately present in the voltage disturbance. Thus it is tempting to link the signals involving the $^3P_1$ and $^1P_1$ states together, but the triplet state is attributed with the second largest population of the 1s states [Zalewski *et al.*, 1979] and thus an inverted population with a $2p_i$ level is out of the question.

Besides the fact that the causes for the positive and negative components of the OGE signals are not completely understood there are other obstacles to understanding the OGE. A direct analysis of the signals is limited to measuring the magnitudes and temporal locations of peaks and crossover points. And since the signal is governed by a large set of differential equations the positive and negative components of the signal are not independent of one another. Thus whether the cause of a change in the ratios of peak heights to one another or even in the magnitude of an individual peak is due to factors that aid or inhibit ionization is unclear. Also, via collisions, the excited state populations are not isolated from one another.

Collisional mixing among the 2p levels is considerable. The OGE signal for the 640.2 nm line is evidence for this. This $^3P_2 \leftrightarrow 2p_9$ transition is unique in that the $2p_9$ state can only decay back to the $^3P_2$ state as is shown in Figure 1.2. But, the 640.2 nm signal is identical to the other metastable signals in which the positive component of the signal is attributed to metastable depletion by 2p state decay to radiative 1s

states. Fluorescence from the $2p_8$ and $2p_{10}$ states has been observed when the $2p_9$ state is pumped [Smyth and Schenck, 1978]. Collisional relaxation rates of $2p$ level states can be found in a paper by Chang and Setser [Chang and Setser, 1980] .

A theoretical study [Cohen *et al.*, 1977] and an experimental study [Leichner *et al.*, 1975] both propose theories of collisional mixing among $1s$ states. Their rates for $1s$ intratriplet mixing and spin flipping are large enough to make the processes worthy of attention.

The temporal contraction of metastable OGE signals with increasing discharge current has been predicted [Erez *et al.*, 1979]. In this work an increase in current contracted all signals for transitions originating in the $1s$ levels. The shorter time, T_{eq} , for metastable excitations could be attributed to a quicker reestablishment of equilibrium metastable state concentrations. A principal process in the production of metastable atoms is electron impact excitation of ground state atoms [Smyth and Schenck, 1978], [Kane, 1984]. Thus the depleted metastable states are repopulated sooner at higher currents. The contracted signals for the ground state coupled transitions cannot be simply explained in this manner. The latter part of their signal would be attributed to a surplus of metastable states and since collisions with ground state and other metastable atoms which do not directly depend on electron densities are the major factors in the depopulation of the metastable states, a direct dependence of the later negative component of this signal on current should not be expected. Perhaps the fact that an increase in intensity was also seen to quicken the reequilibration time will provide insight as to the cause of this trend. In both cases stronger signals accompanied the shorter times.

The results of the variation of laser intensity indicate that the laser pulses were not saturating the neon. Figure 3.4 on page 21 showed that the magnitude of the 588.2 nm signal did not saturate when the laser intensity was increased. In fact that figure portrays a slight quadratic dependence which would indicate the presence of atom-atom collisions.

Metastable-metastable collisions are considered by Kane to be a major factor in the 588.2 nm OGE signal [Kane, 1984].

The different concentrations of species in the various regions of a discharge would be expected to produce different OGE signals for different laser beam locations. And this is observed.

CHAPTER 5

CONCLUSION

OGE signals involving metastable states have been presented and show properties that are supported by theory. Transitions involving ground state coupled atoms produce OGE signals that are not completely understood. The OGE signals involving the radiative singlet state, $^1P_1$, are partially explained by an inverted population with the $2p_i$ states. But this theory would need to be expanded to account for the increased ionization seen on a longer time scale. Explanations for the causes of the OGE signals involving the $^3P_1$ state are purely speculative and demand further study.

Acuchem, a computer program that solves first order differential equations is being used to model the OGE in neon [Braun *et al.*, 1988]. Process reactions and rates and approximate species populations are input and equilibrium concentrations are established. Then the initial species concentrations are altered to simulate the transferred states resulting from laser irradiation. Using this method for metastable based transitions, the time-resolved electron and positive ion concentrations are seen to be crude facsimiles of OGE signals. In order to simulate the initial voltage drop of the experimental signals, the electron impact ionization coefficient of the pumped $2p$ state had to be increased by over four orders of magnitude. Furthermore, the quoted decay rates of the $2p$ states seem too fast to be able to account for the comparable sizes of the negative and positive components of the signals. For each $2p$ state that is ionized via electron impact, 10^5 will decay to $1s$ states of which over half will end up in the ground state coupled $^1P_1$ and $^3P_1$ states. Since the $2p$ electron impact ionization coefficient is less than an order of magnitude larger than the coefficient for the $1s$ states, any signal attributable

to perturbed 2p state populations would be about four orders of magnitude smaller than that attributable to perturbed 1s state populations. This circumstantial evidence leads one to believe that ionization of the 2p states is not responsible for the initial voltage changes with which it has been credited. A new theory that just uses the 2p states as a vehicle for changing the 1s state populations and that attributes the initial change in voltage of the OGE signals to ionization from the $^1P_1$ state is being studied. We expect that this theory will explain the inverted OGE signals seen for transitions based in the metastable states and the $^1P_1$ state. An attempt to model the OGE in neon with this theory is under way.

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APPENDICES

OGE Signals

These appendices contain OGE signals for all neon transitions observed in this experiment.

Appendix A contains signals for transitions involving the $^1P_1$ state.

Appendix B contains signals for transitions involving the $^3P_0$ state.

Appendix C contains signals for transitions involving the $^3P_1$ state.

Appendix D contains signals for transitions involving the $^3P_2$ state.

APPENDIX A

Signals for transitions involving the $^1P_1$ state

This appendix is subdivided into sections for individual transitions involving the $^1P_1$ state.

Appendix A1 contains signals for 585.2 nm laser pulses.

Appendix A2 contains signals for 659.9 nm laser pulses.

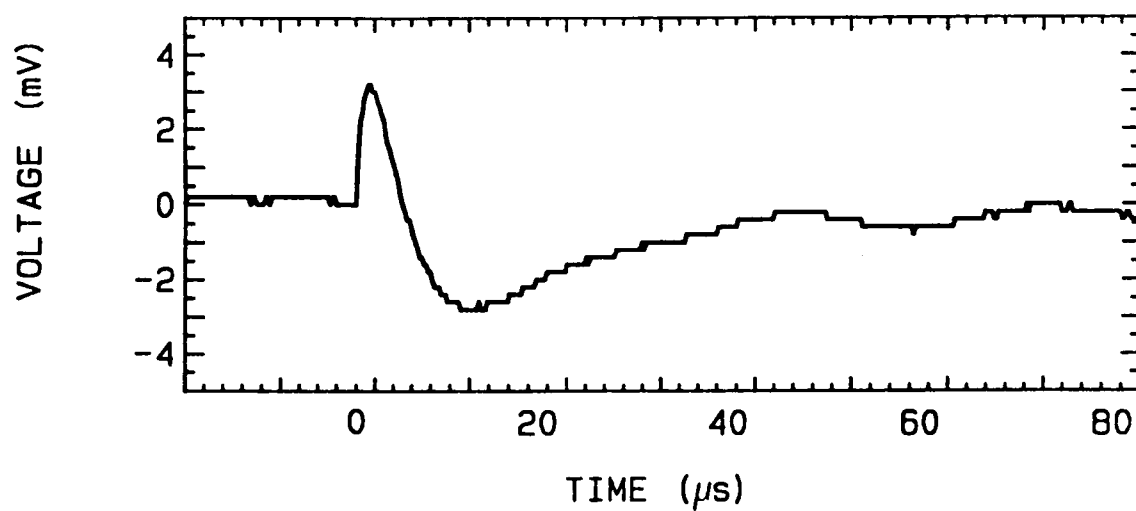
Appendix A3 contains signals for 665.2 nm laser pulses.

Appendix A4 contains signals for 667.8 nm laser pulses.

Appendix A5 contains signals for 671.7 nm laser pulses.

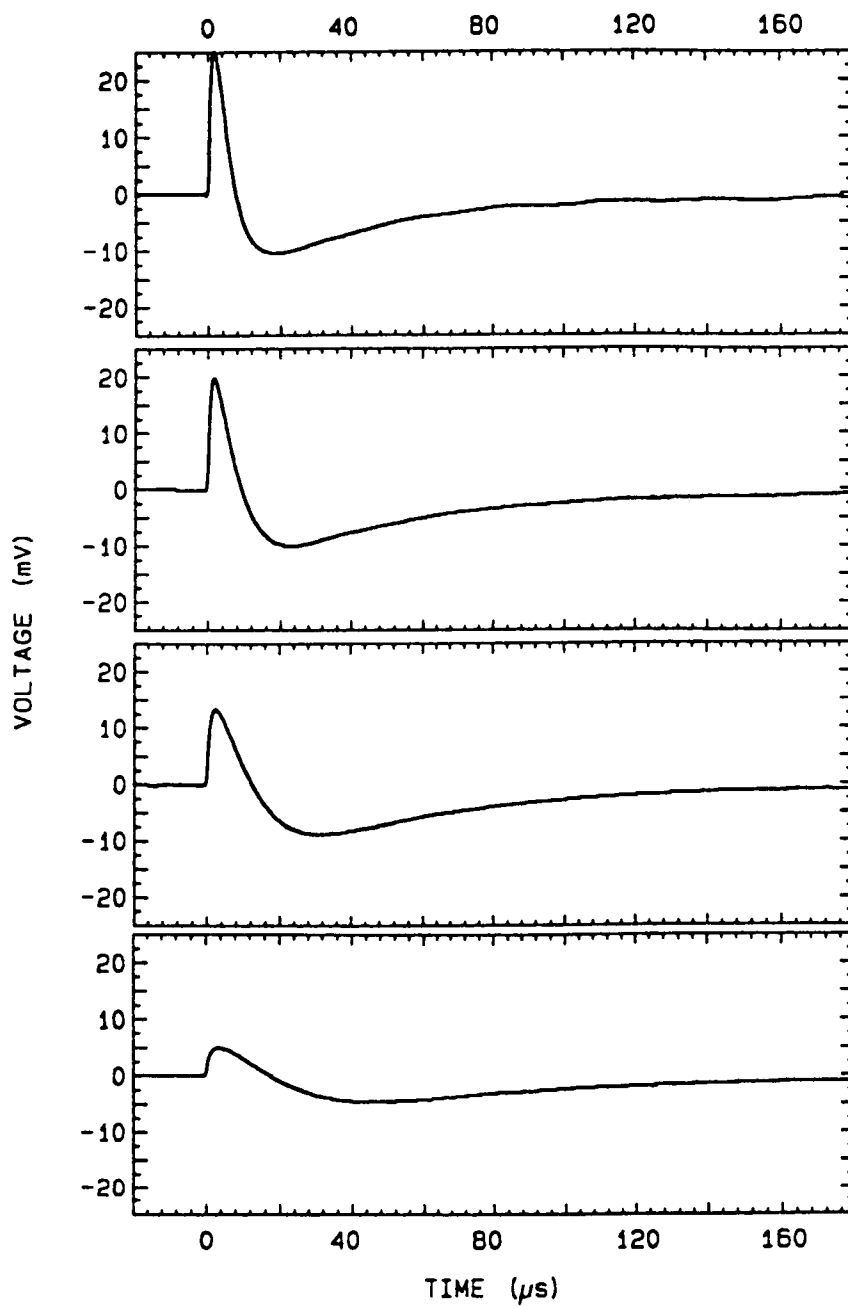
Appendix A6 contains signals for 693.0 nm laser pulses.

A1

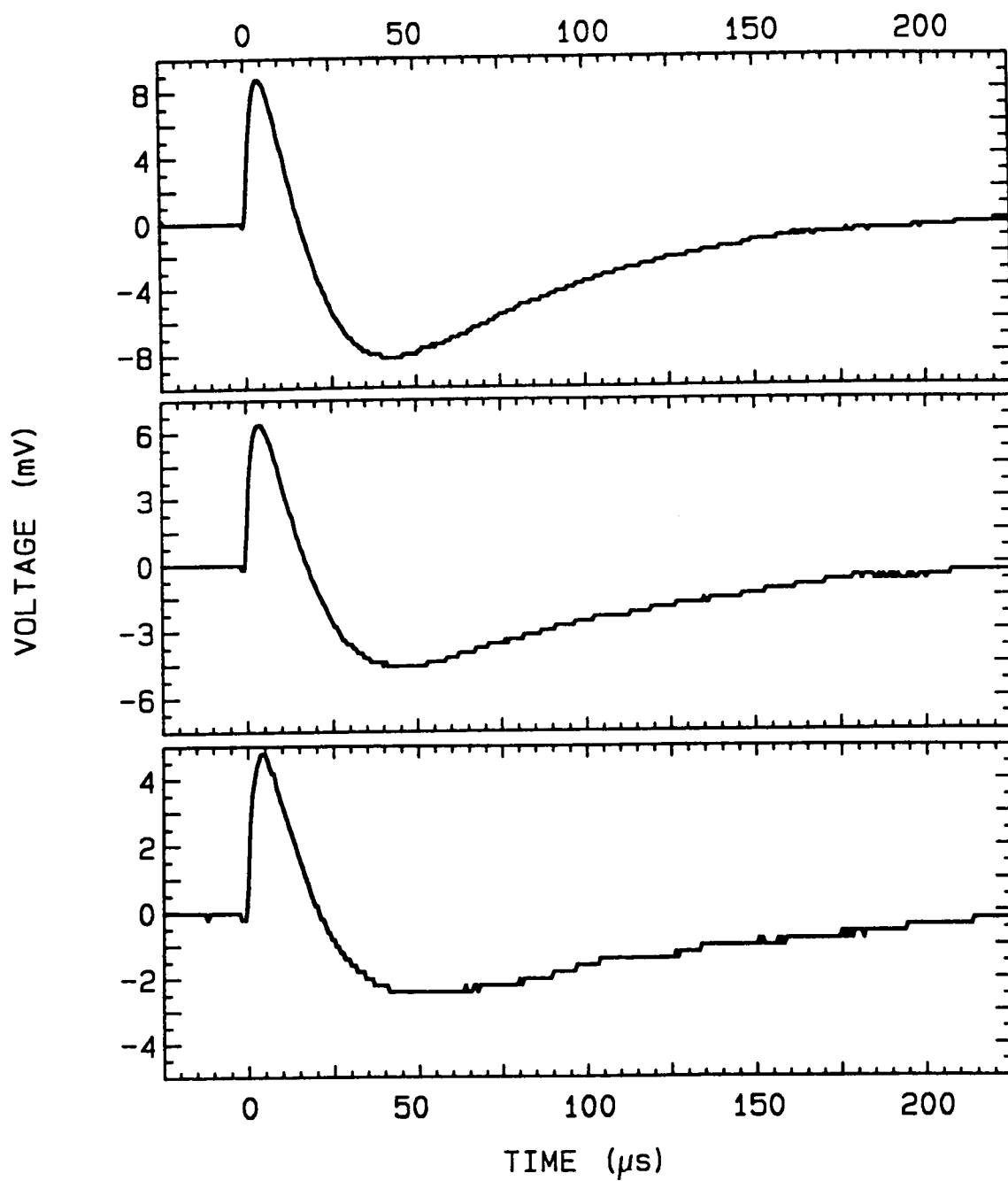


Voltage change with time due to 585.2 nm laser pulse. The neon pressure was 20 torr and the discharge current was 1.0 mA.

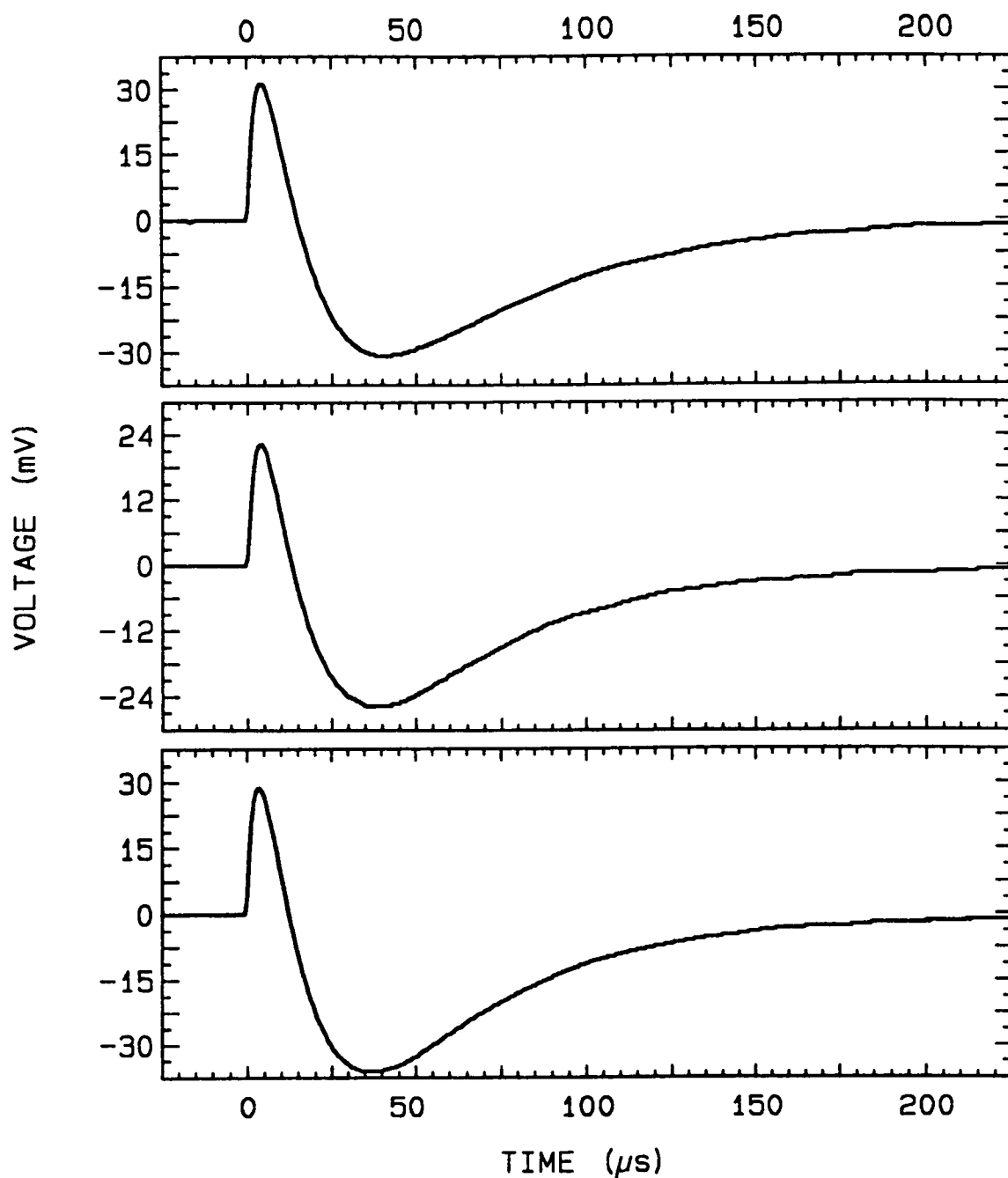
A2



Voltage change with time due to 659.9 nm laser pulse for various discharge currents. The current was 0.2, 0.5, 1.0, and 1.5 mA from bottom to top. The neon pressure was 20 torr.

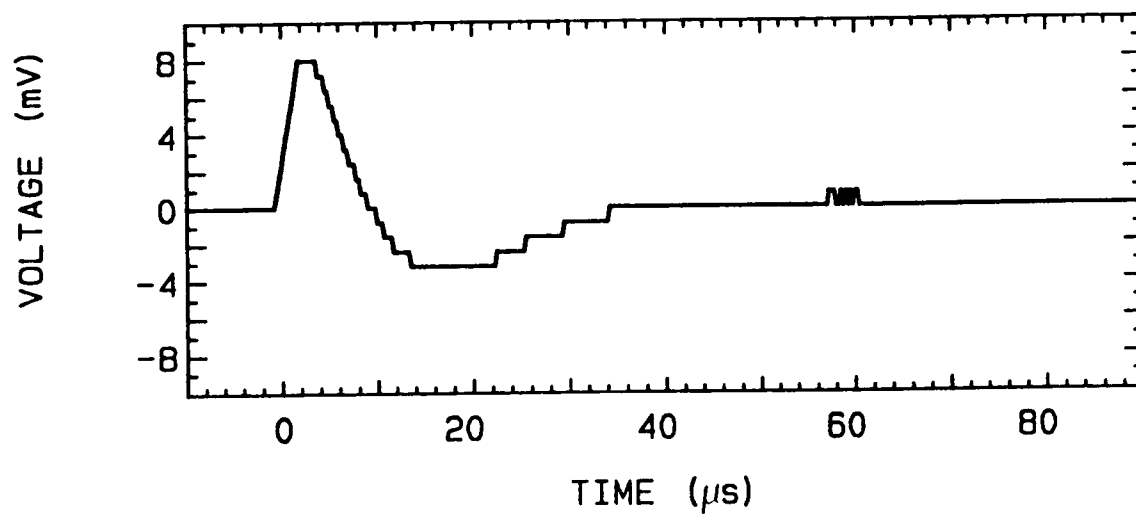


Voltage change with time due to 659.9 nm laser pulse for various laser intensities. From bottom to top the laser intensities were 14%, 41%, and 55% of full strength, respectively. The neon pressure was 4 torr and the discharge current was 0.5 mA.

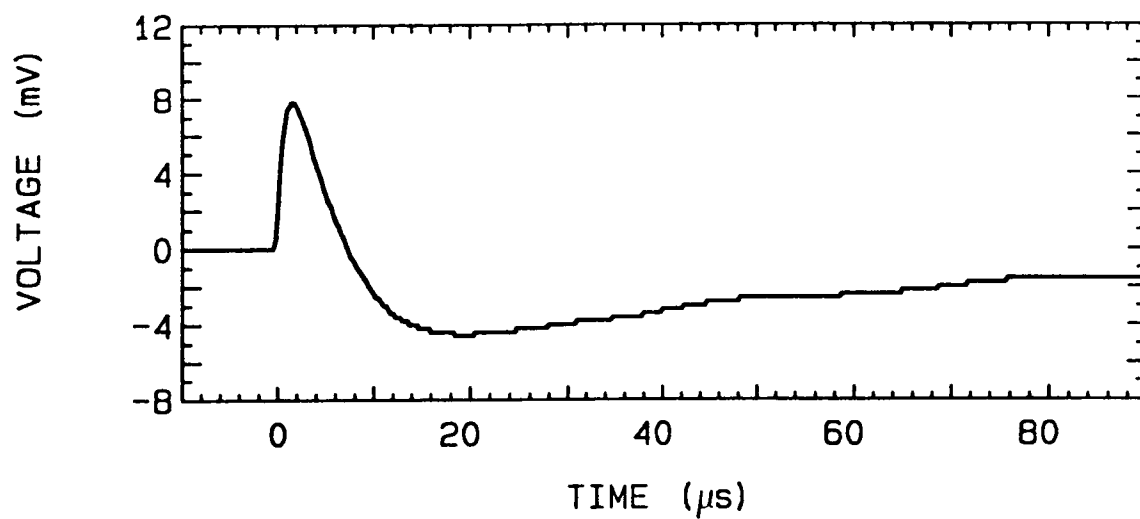


Voltage change with time due to 659.9 nm laser pulse for various transverse laser beam locations in the discharge. The signals from bottom to top were from beams passing through the anode glow, positive glow, and negative glow respectively. The neon pressure was 4 torr and the discharge current was 0.5 mA.

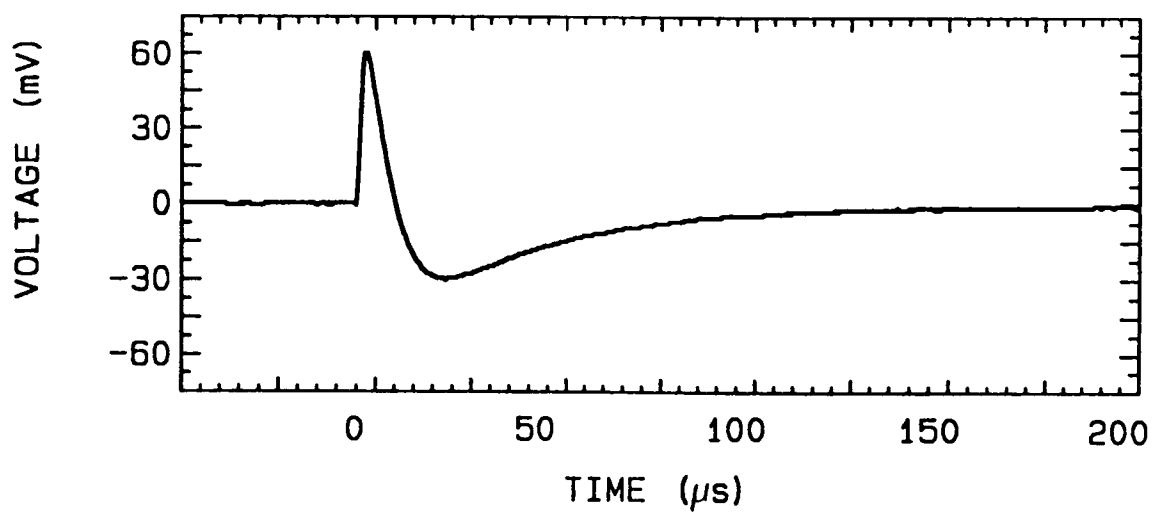
A3



Voltage change with time due to 665.2 nm laser pulse. The neon pressure was 20 torr and the discharge current was 1.0 mA.

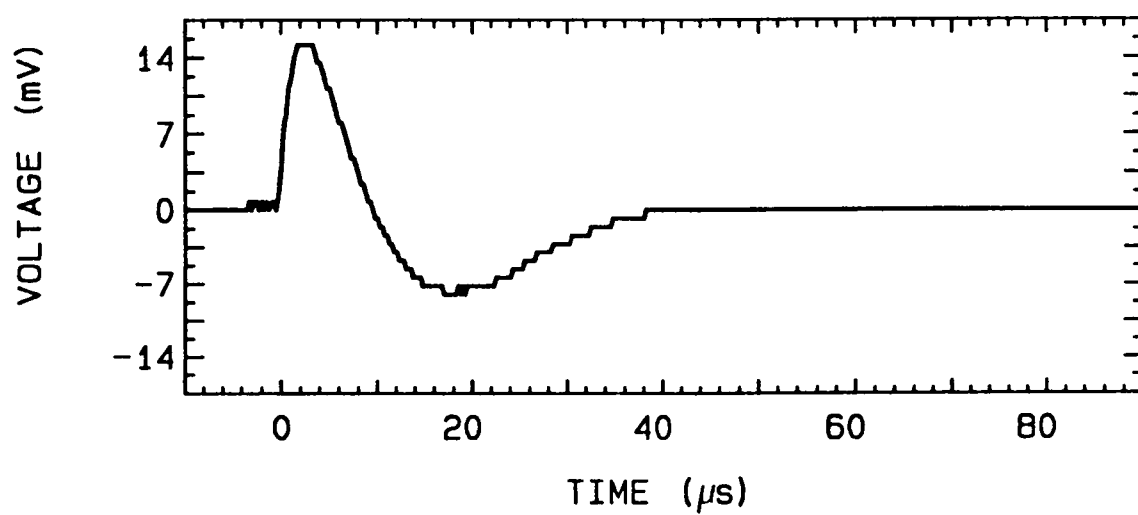


Voltage change with time due to 667.8 nm laser pulse. The neon pressure was 20 torr and the discharge current was 1.0 mA.



Voltage change with time due to 671.7 nm laser pulse. The neon pressure was 20 torr and the discharge current was 1.0 mA.

A6



Voltage change with time due to 693.0 nm laser pulse. The neon pressure was 20 torr and the discharge current was 1.0 mA.

APPENDIX B

Signals for transitions involving the $^3P_0$ state

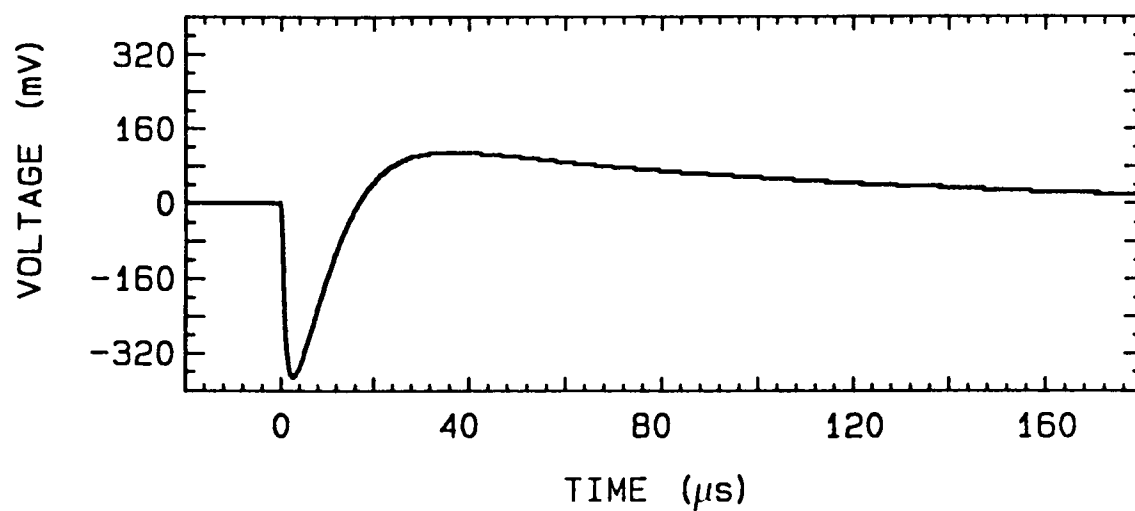
This appendix is subdivided into sections for individual transitions involving the $^3P_0$ state.

Appendix B1 contains signals for 616.4 nm laser pulses.

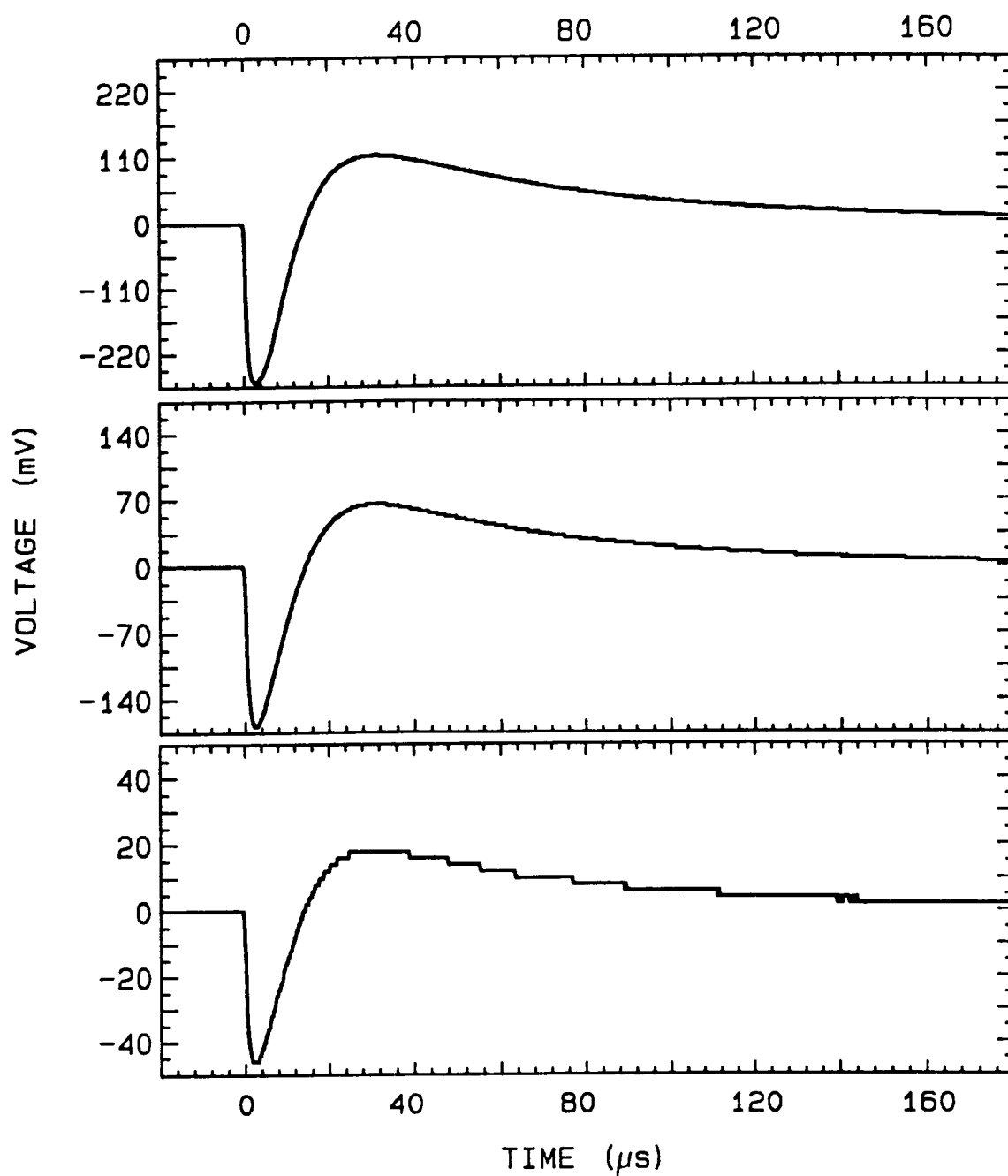
Appendix B2 contains signals for 626.6 nm laser pulses.

Appendix B3 contains signals for 653.3 nm laser pulses.

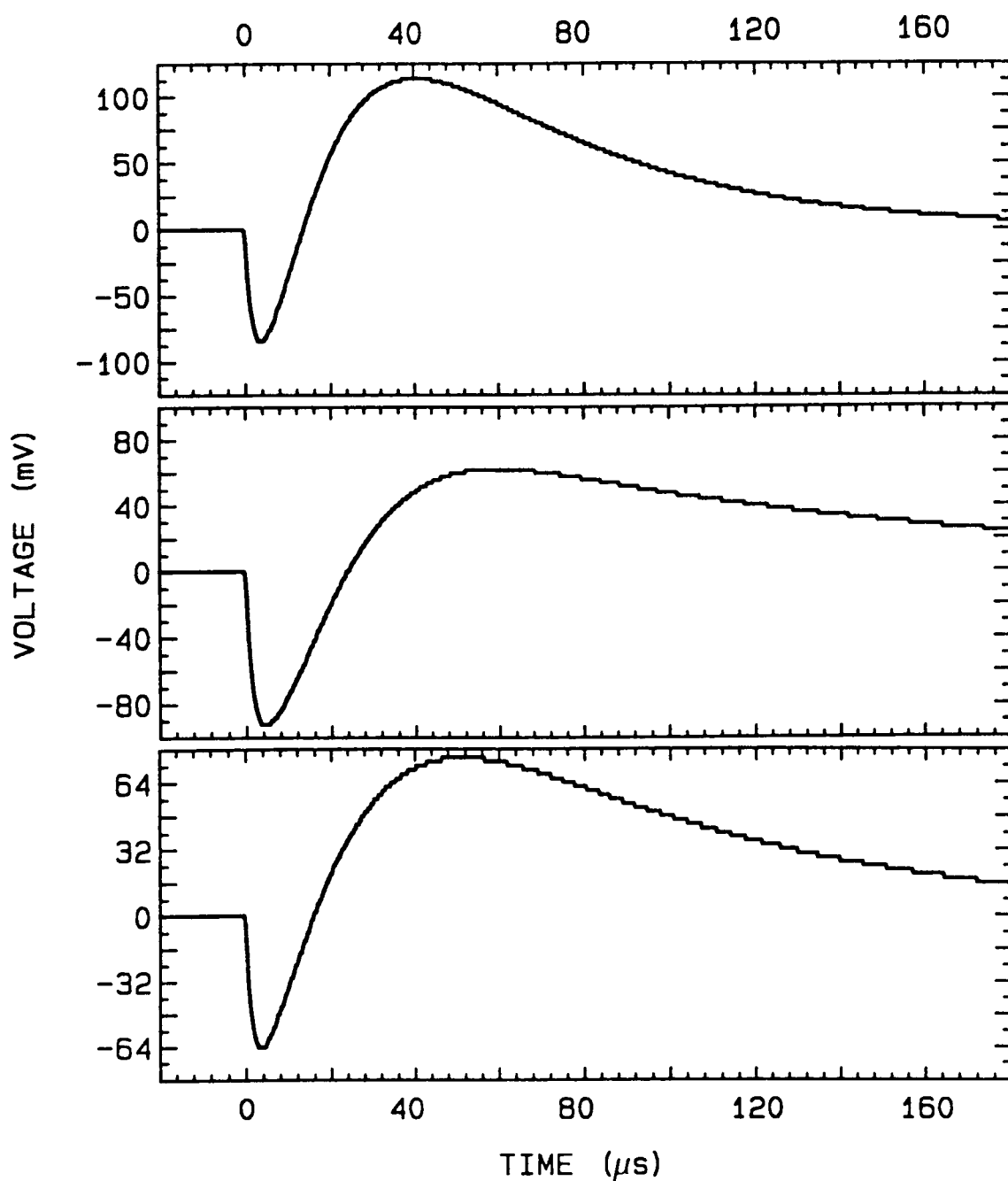
B1



Voltage change with time due to 616.4 nm laser pulse. The neon pressure was 20 torr and the discharge current was 1.0 mA.

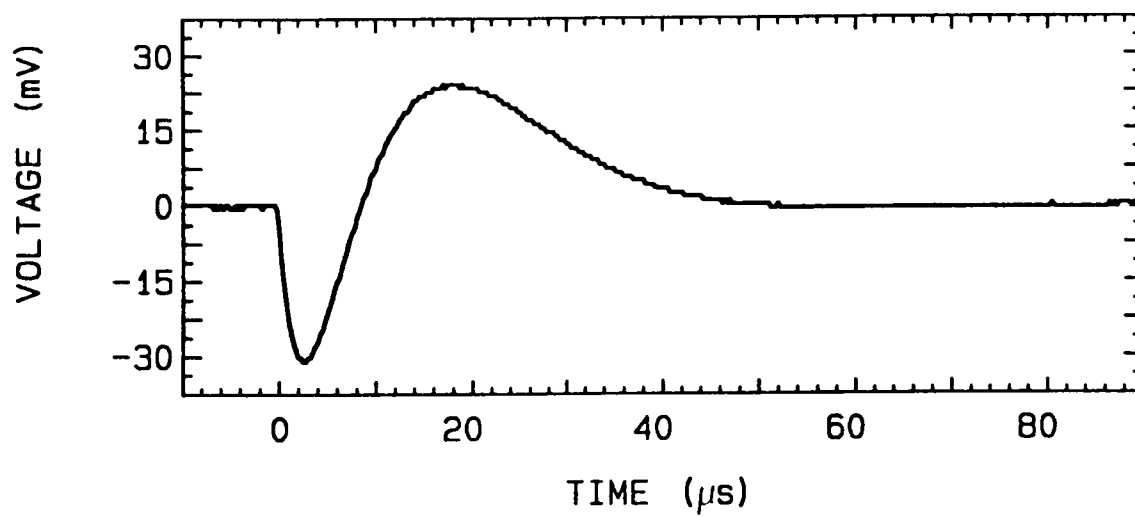


Voltage change with time due to 616.4 nm laser pulse for various laser intensities. From bottom to top the laser intensities were 14%, 41%, and 55% of full strength, respectively. The neon pressure was 20 torr and the discharge current was 1.0 mA.



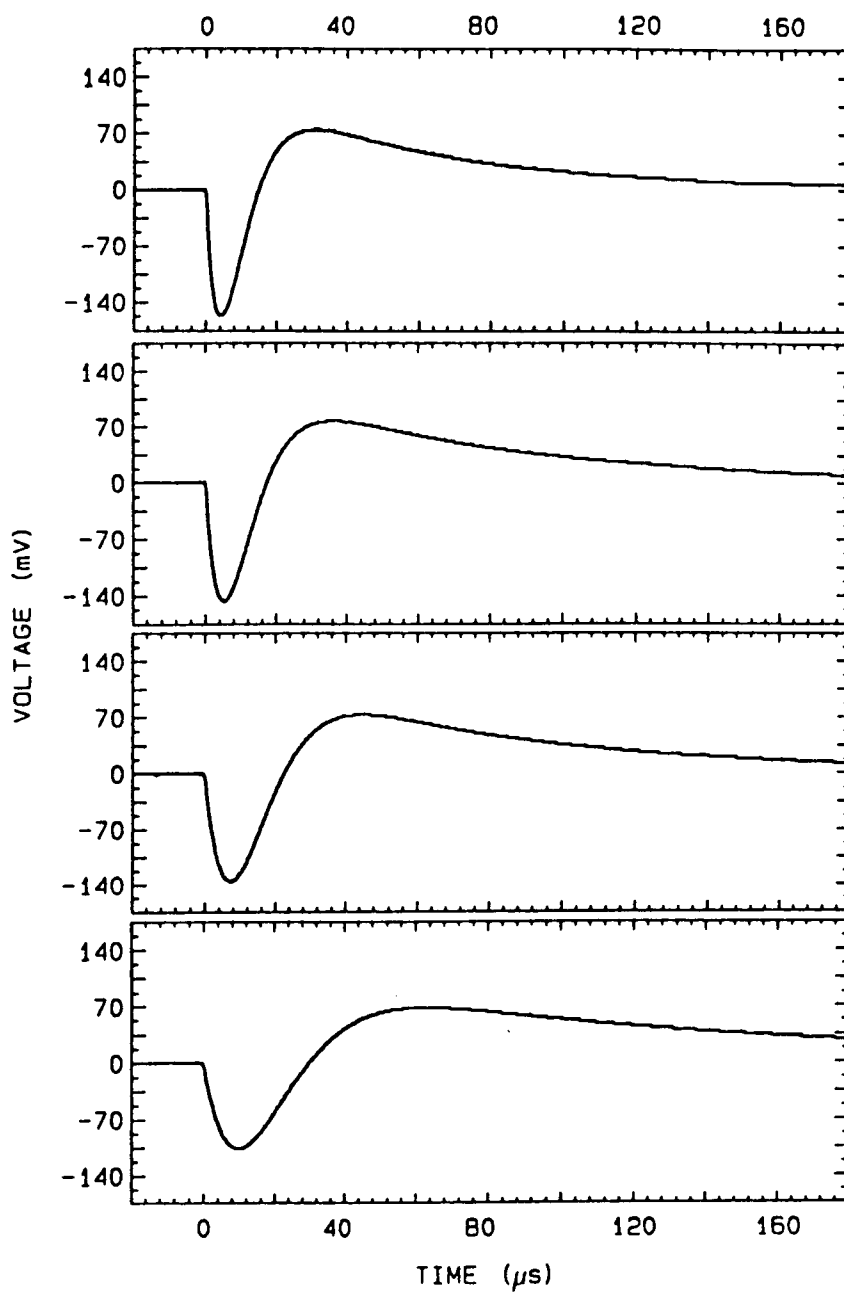
Voltage change with time due to 616.4 nm laser pulse for various transverse laser beam locations in the discharge. The signals from bottom to top were from beams passing through the anode glow, positive glow, and negative glow respectively. The neon pressure was 4 torr and the discharge current was 0.5 mA.

B2



Voltage change with time due to 626.6 nm laser pulse. The neon pressure was 20 torr and the discharge current was 1.0 mA.

B3



Voltage change with time due to 653.3 nm laser pulse for various discharge currents. The current was 0.2, 0.5, 1.0, and 1.5 mA from bottom to top. The neon pressure was 20 torr.

APPENDIX C

Signals for transitions involving the $^3P_1$ state

This appendix is subdivided into sections for individual transitions involving the $^3P_1$ state.

Appendix C1 contains signals for 603.0 nm laser pulses.

Appendix C2 contains signals for 607.4 nm laser pulses.

Appendix C3 contains signals for 609.6 nm laser pulses.

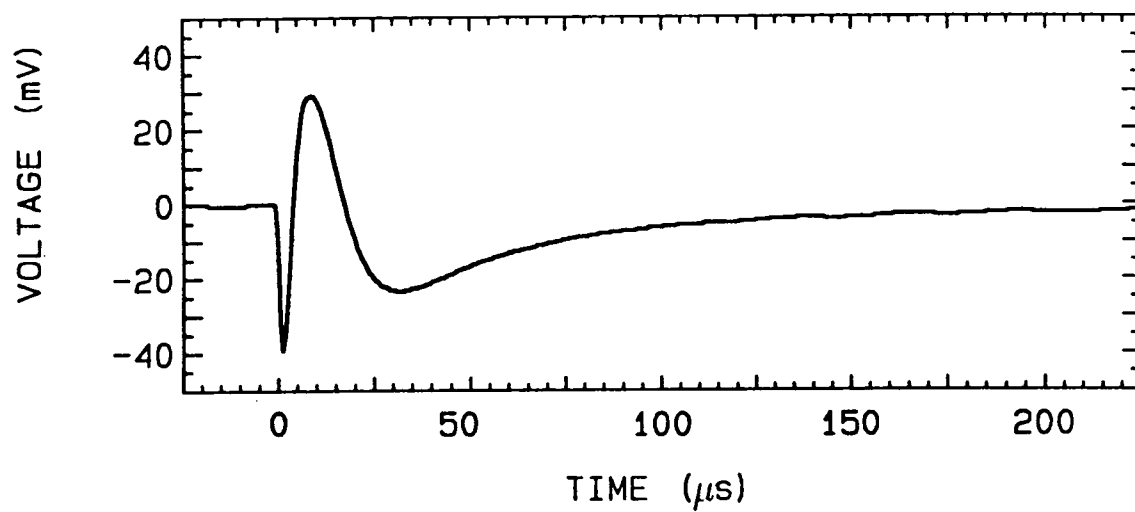
Appendix C4 contains signals for 612.8 nm laser pulses.

Appendix C5 contains signals for 630.5 nm laser pulses.

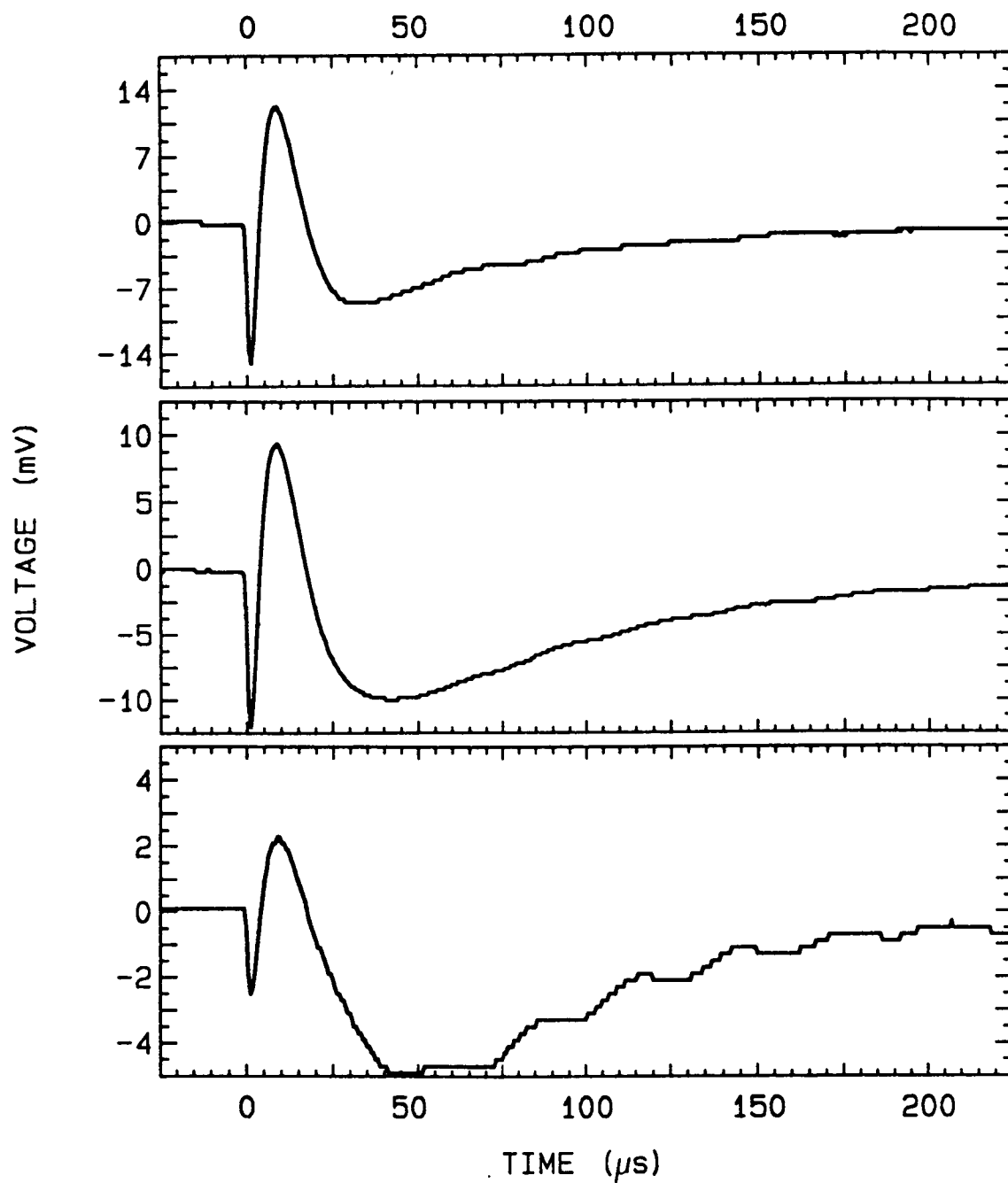
Appendix C6 contains signals for 638.3nm laser pulses.

Appendix C7 contains signals for 650.6 nm laser pulses.

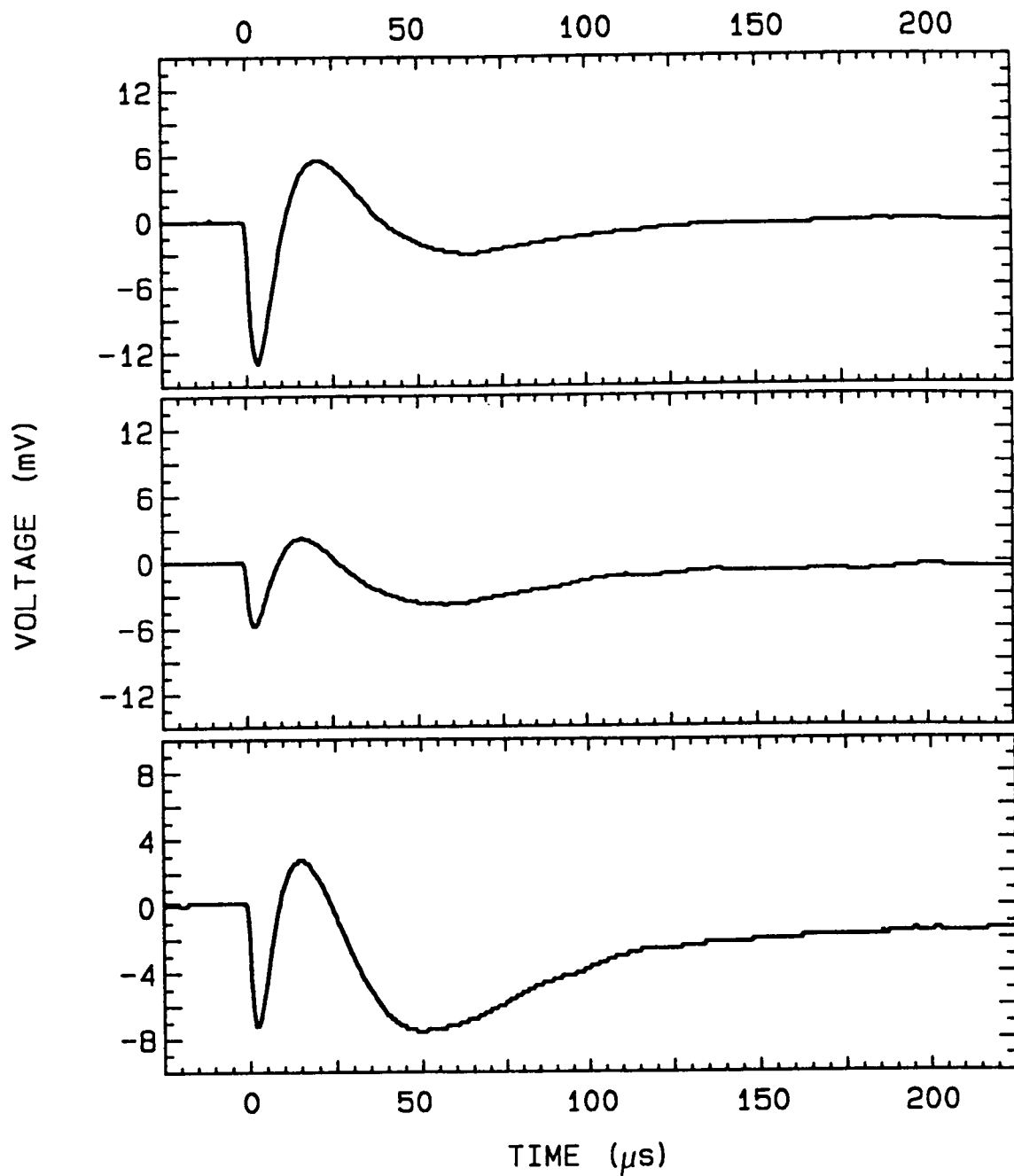
C1



Voltage change with time due to 603.0 nm laser pulse. The neon pressure was 20 torr and the discharge current was 1.0 mA.

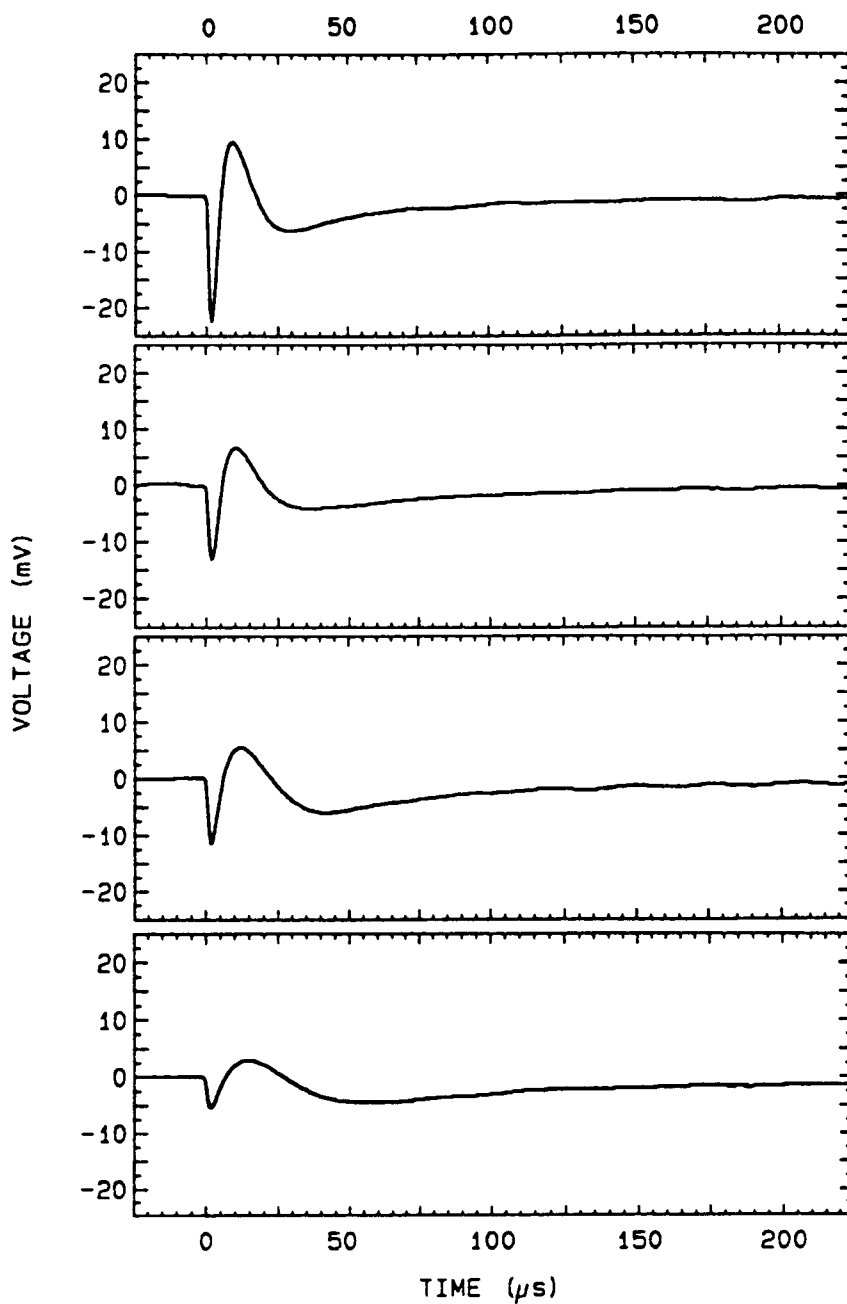


Voltage change with time due to 603.0 nm laser pulse for various laser intensities. From bottom to top the laser intensities were 14%, 41%, and 55% of full strength, respectively. The neon pressure was 20 torr and the discharge current was 1.0 mA.



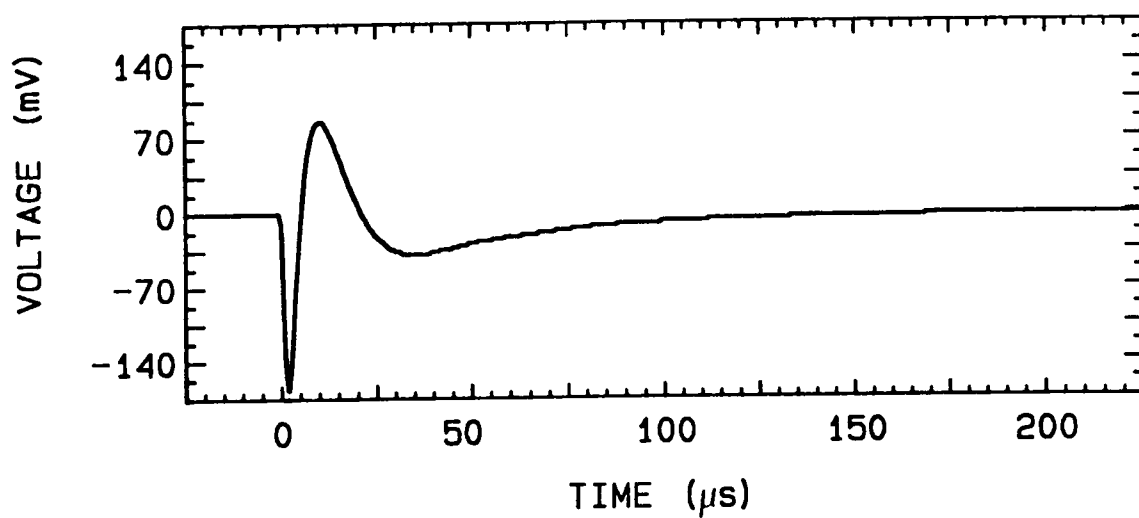
Voltage change with time due to 603.0 nm laser pulse for various transverse laser beam locations in the discharge. The signals from bottom to top were from beams passing through the anode glow, positive glow, and negative glow respectively. The neon pressure was 4 torr and the discharge current was 0.5 mA.

C2



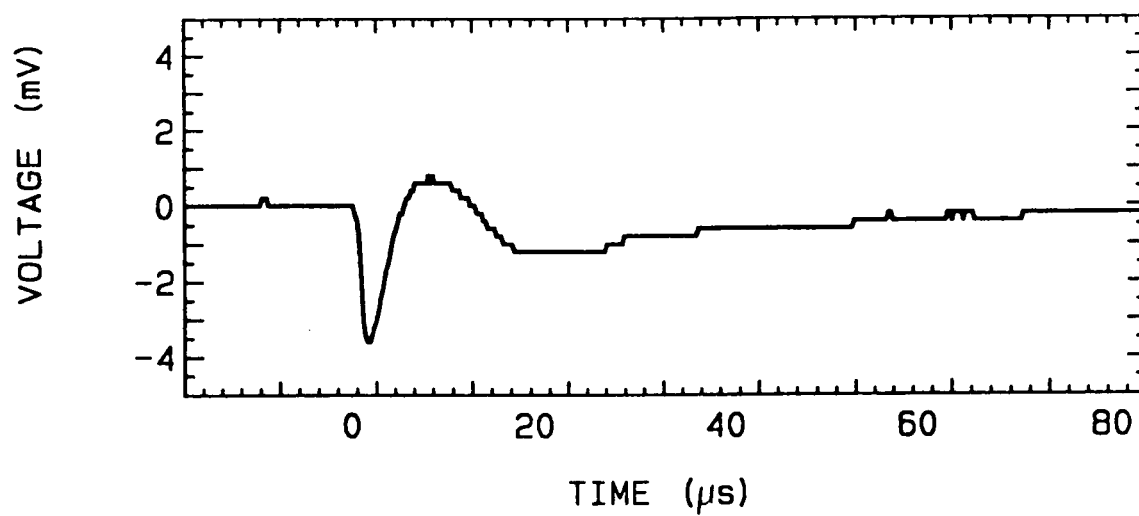
Voltage change with time due to 607.4 nm laser pulse for various discharge currents. The current was 0.2, 0.5, 1.0, and 1.5 mA from bottom to top. The neon pressure was 20 torr.

C3



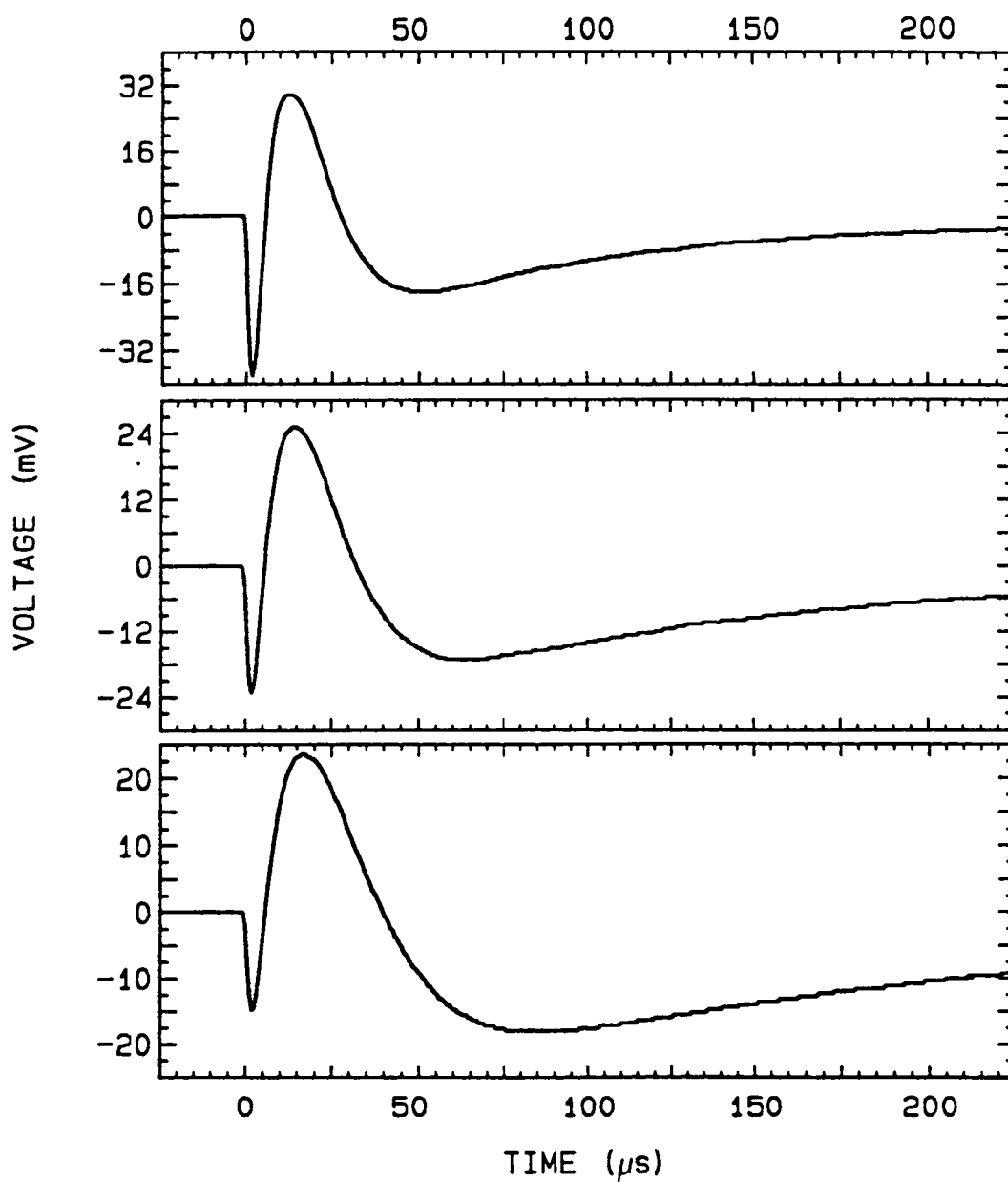
Voltage change with time due to 609.6 nm laser pulse. The neon pressure was 20 torr and the discharge current was 1.0 mA.

C4

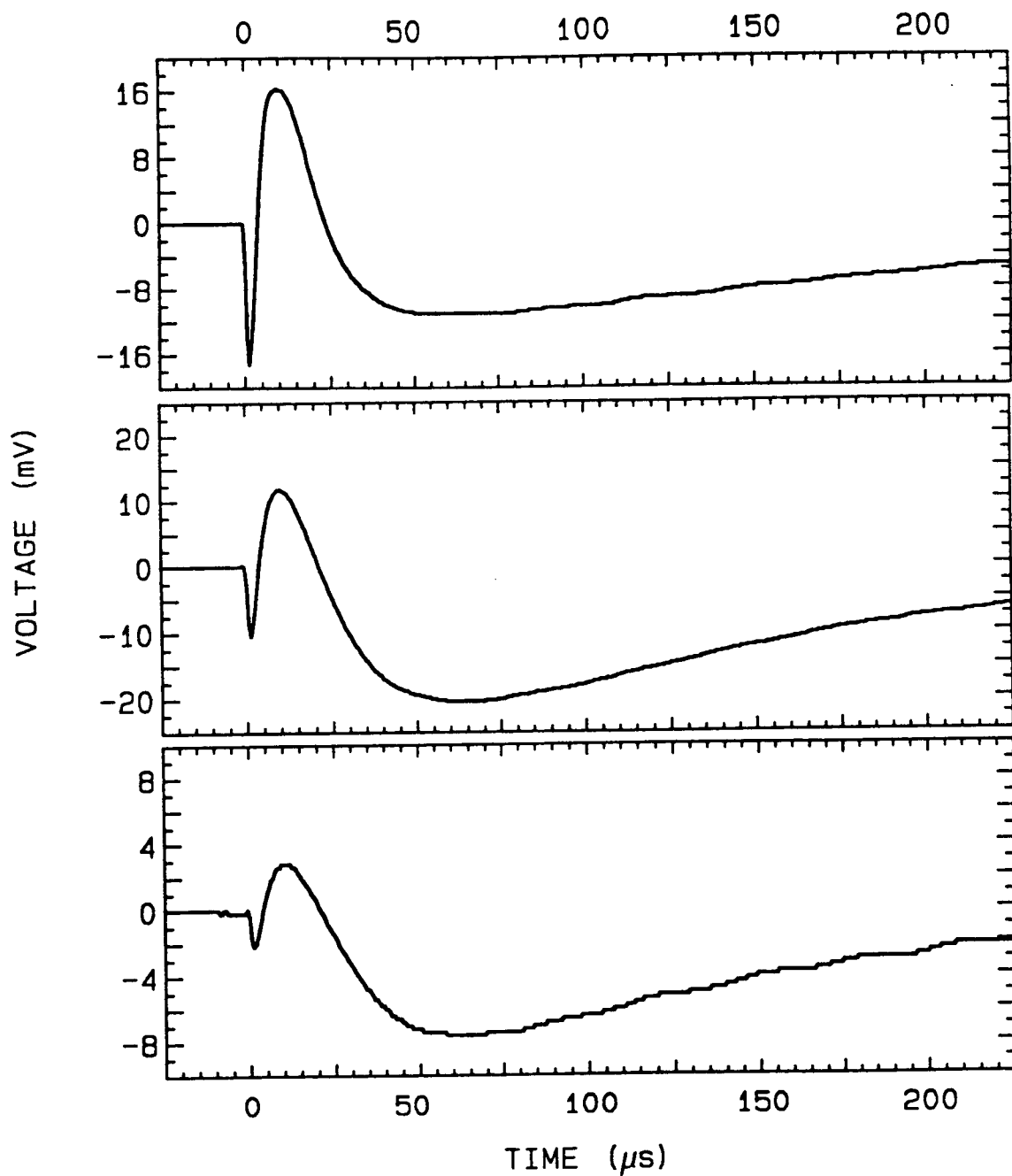


Voltage change with time due to 612.8 nm laser pulse. The neon pressure was 20 torr and the discharge current was 1.0 mA.

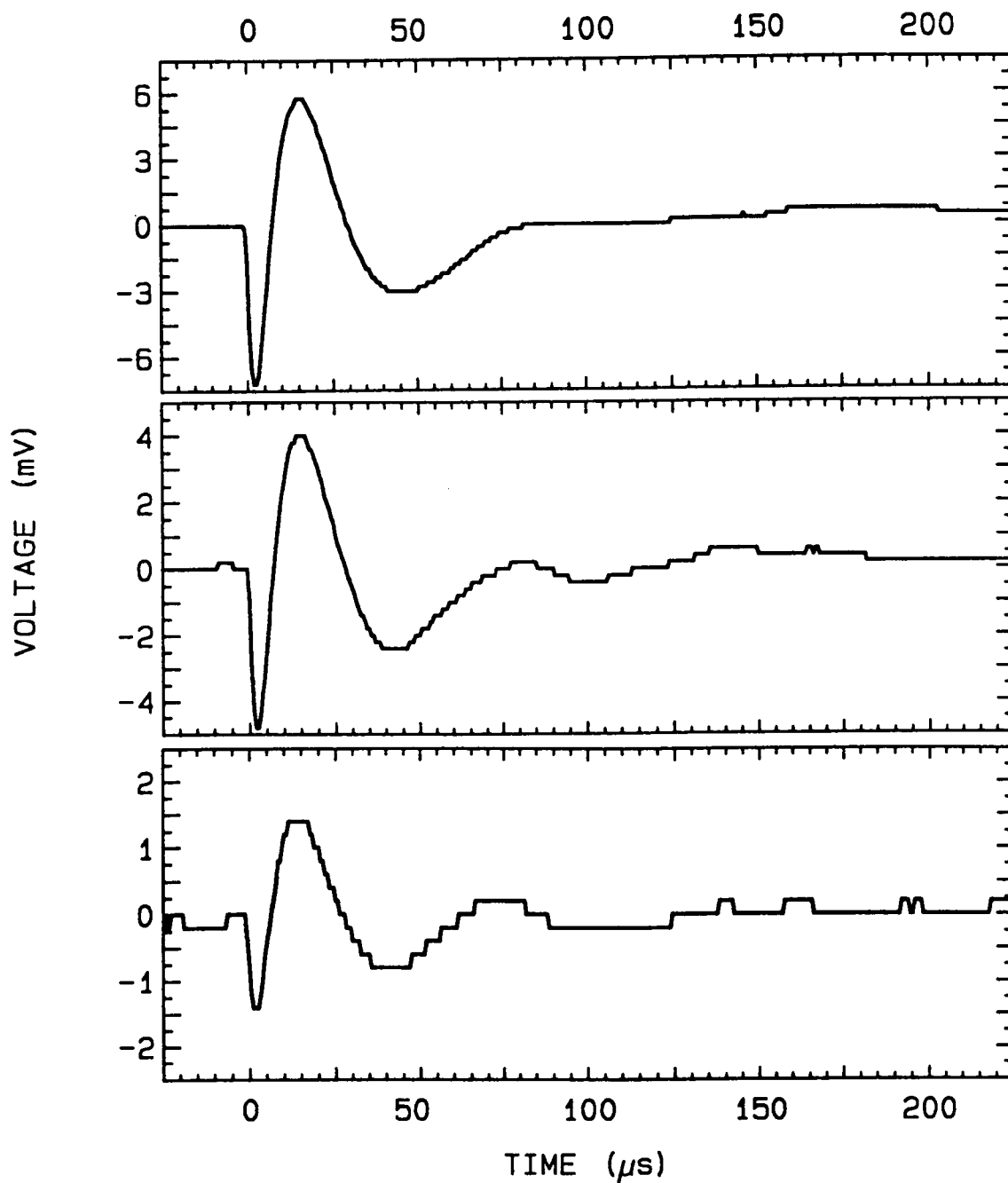
C5



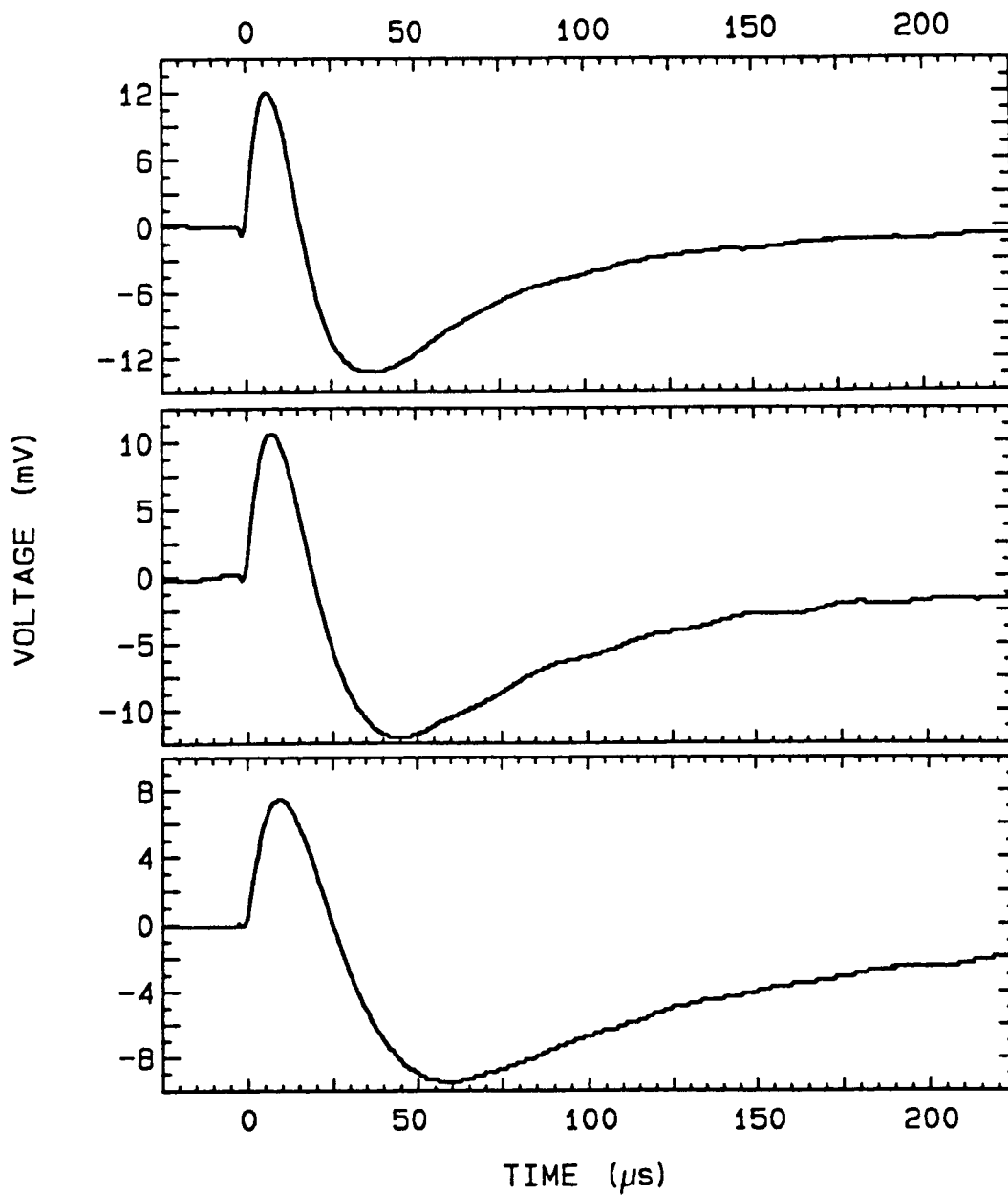
Voltage change with time due to 630.5 nm laser pulse for various discharge currents. The current was 0.2, 0.5, and 1.0 mA from bottom to top. The neon pressure was 20 torr.



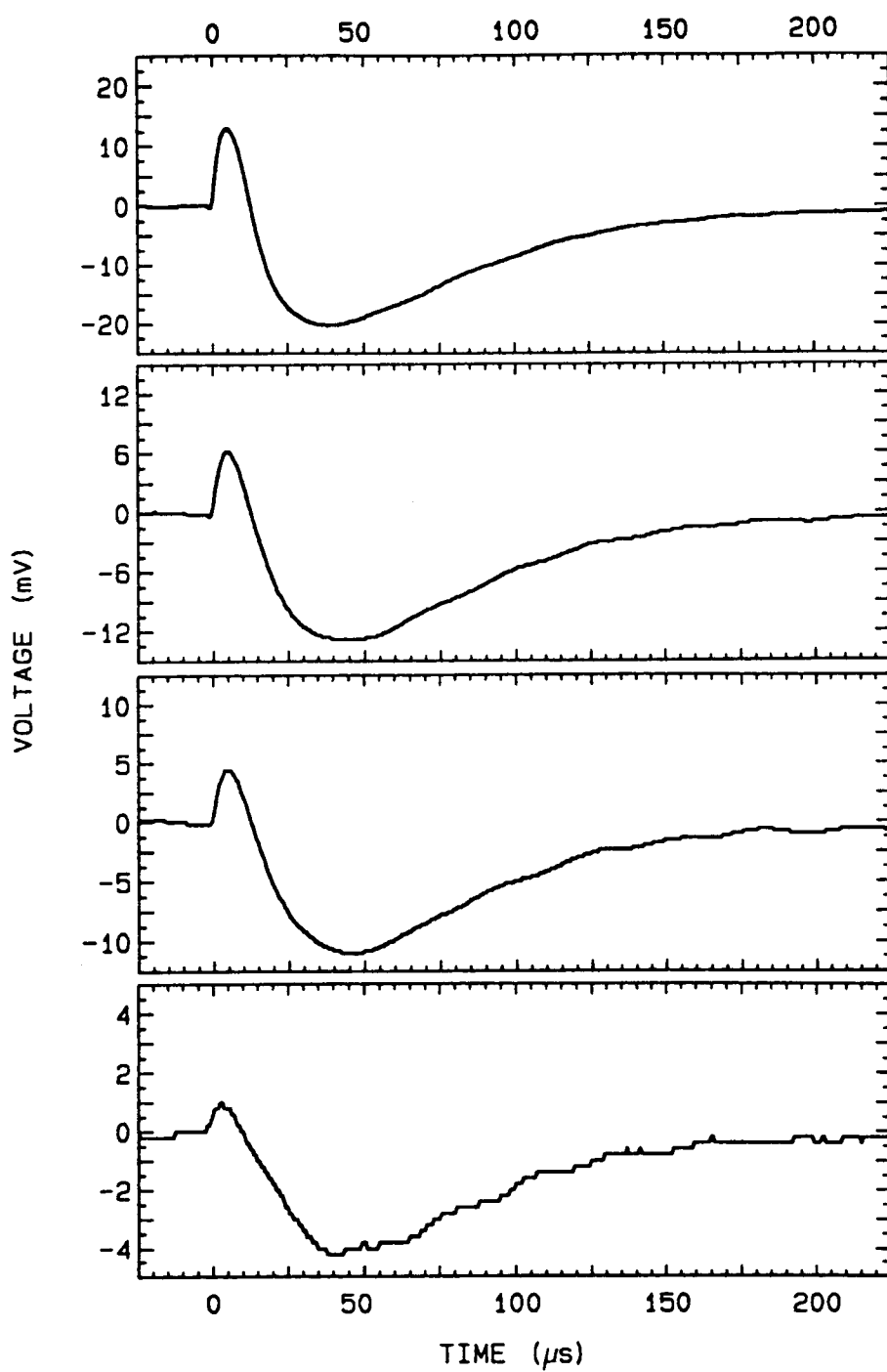
Voltage change with time due to 630.5 nm laser pulse for various laser intensities. From bottom to top the laser intensities were 14%, 41%, and 55% of full strength, respectively. The neon pressure was 20 torr and the discharge current was 1.0 mA.



Voltage change with time due to 630.5 nm laser pulse for various laser intensities. From bottom to top the laser intensities were 14%, 41%, and 55% of full strength, respectively. The neon pressure was 4 torr and the discharge current was 0.5 mA.

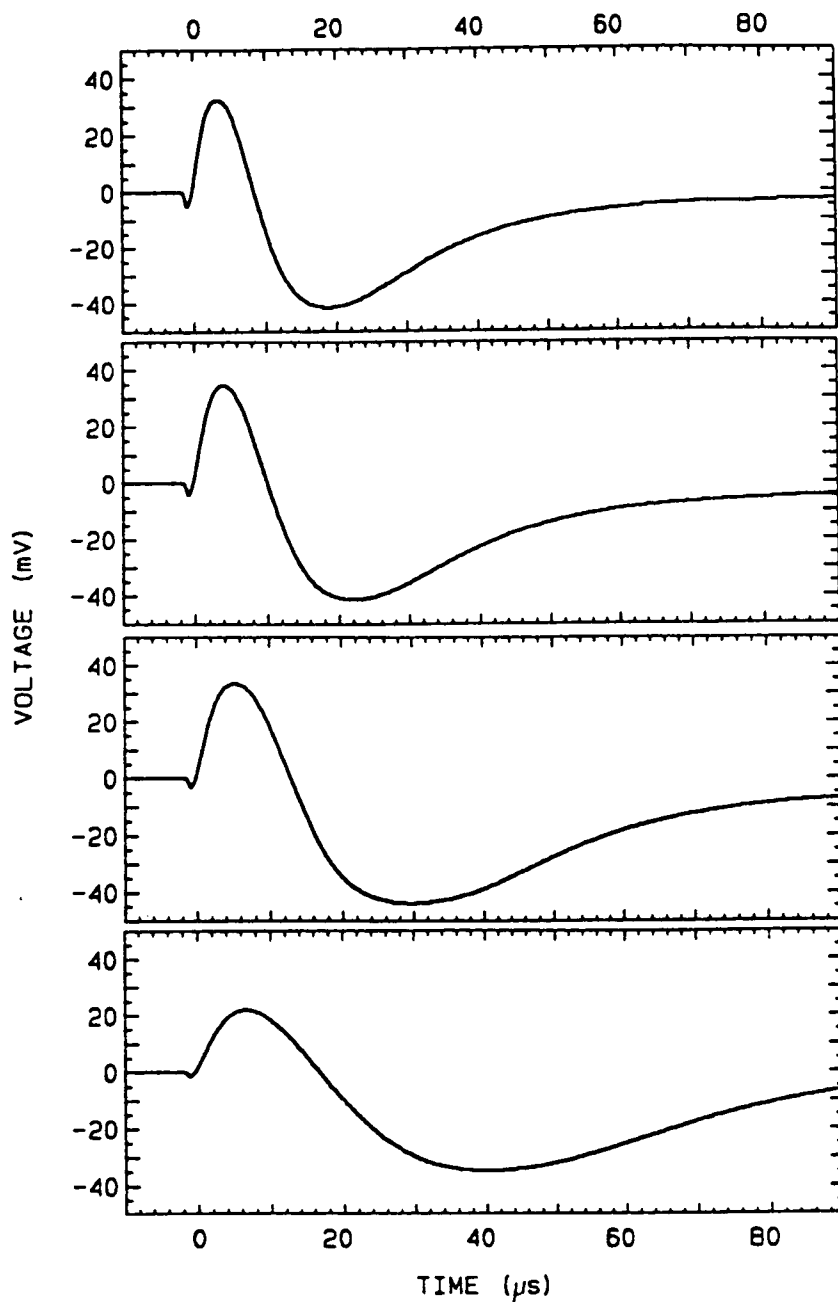


Voltage change with time due to 638.3 nm laser pulse for various discharge currents. The current was 0.2, 0.5, and 1.0 mA from bottom to top. The neon pressure was 20 torr.



Voltage change with time due to 638.3 nm laser pulse for various laser intensities. From bottom to top the laser intensities were 14%, 41%, 55%, and 100 and the discharge current was 1.0 mA.

C7



Voltage change with time due to 650.6 nm laser pulse for various discharge currents. The current was 0.2, 0.5, 1.0, and 1.5 mA from bottom to top. The neon pressure was 20 torr.

APPENDIX D

Signals for transitions involving the $^3P_2$ state

This appendix is subdivided into sections for individual transitions involving the $^3P_2$ state.

Appendix D1 contains signals for 588.2 nm laser pulses.

Appendix D2 contains signals for 594.5 nm laser pulses.

Appendix D3 contains signals for 597.6 nm laser pulses.

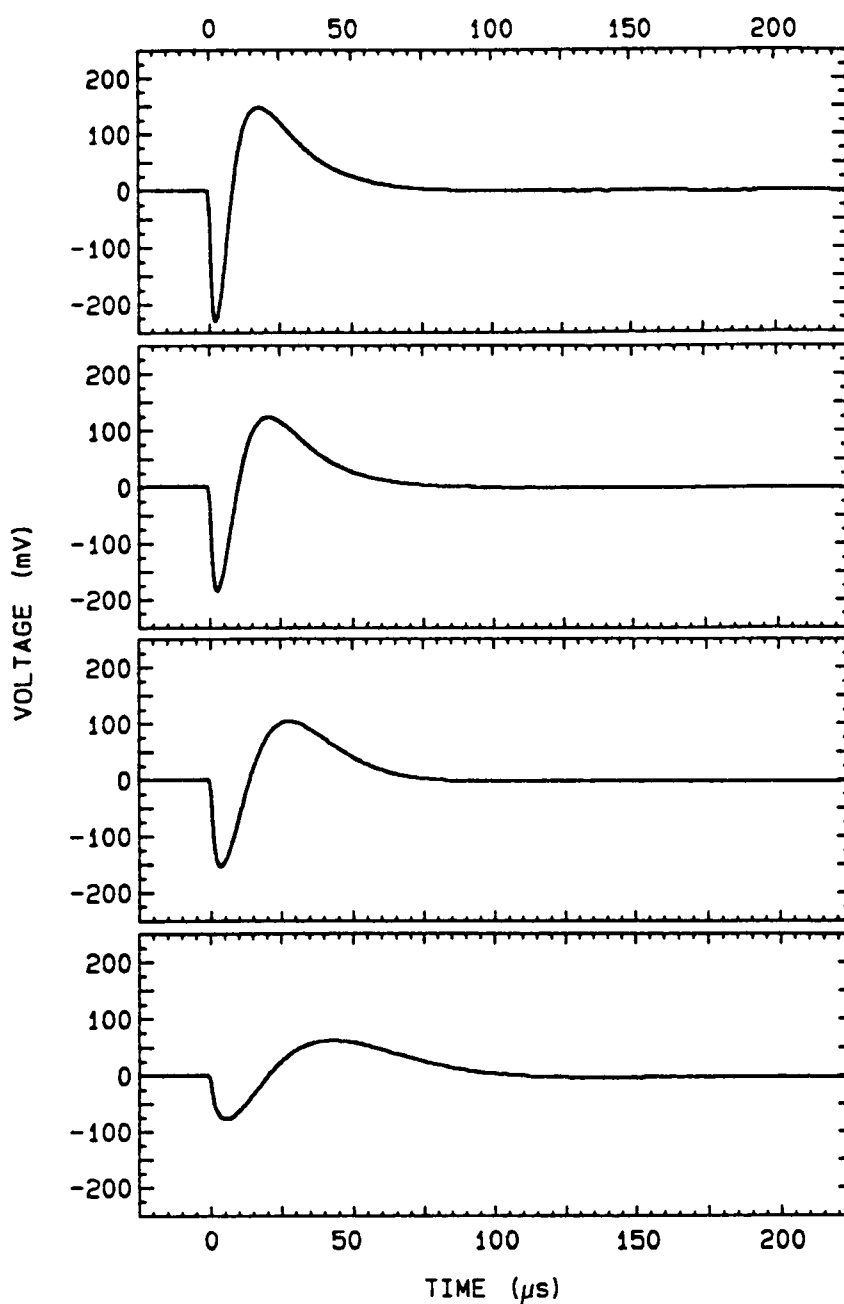
Appendix D4 contains signals for 614.3 nm laser pulses.

Appendix D5 contains signals for 621.7 nm laser pulses.

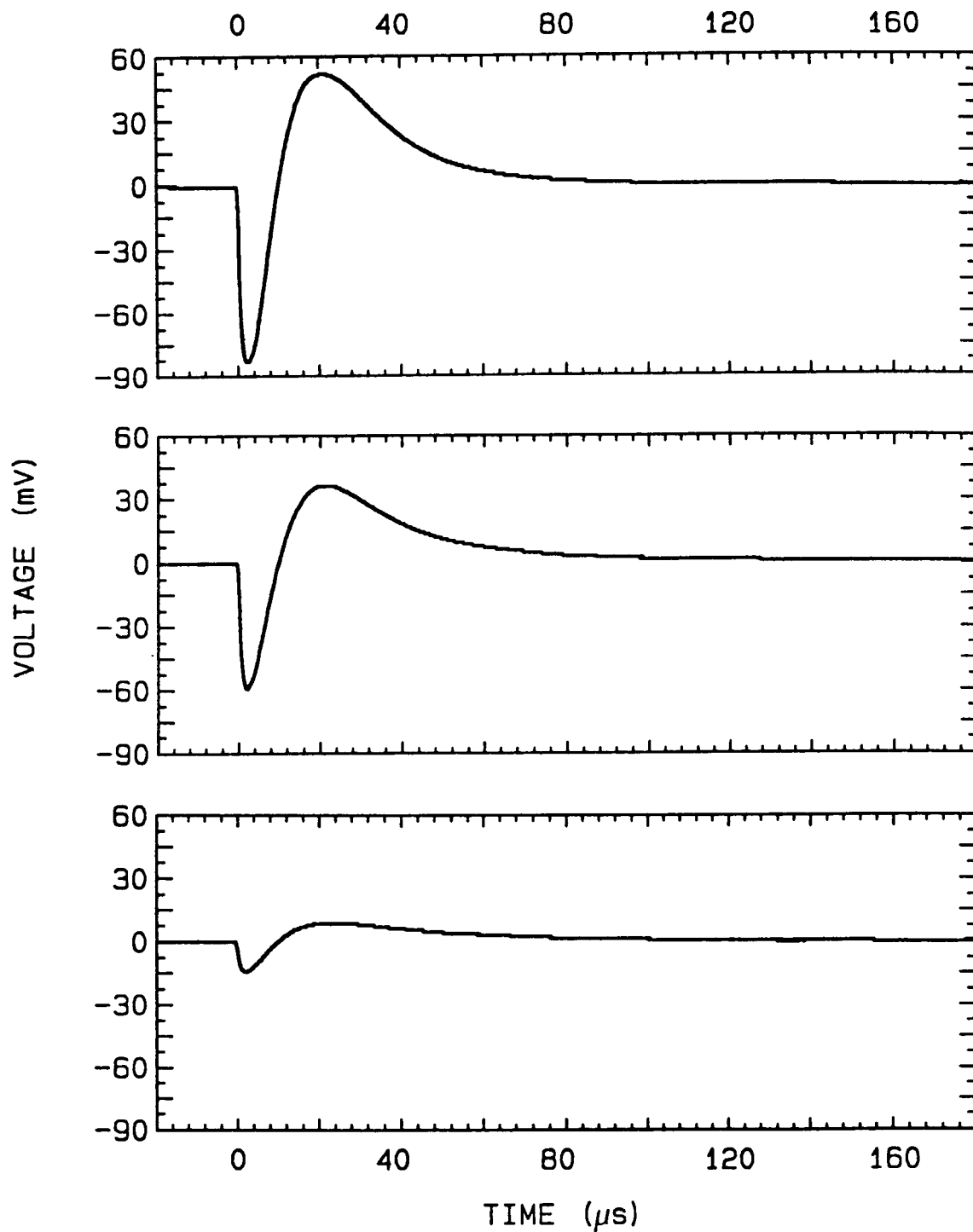
Appendix D6 contains signals for 633.4nm laser pulses.

Appendix D7 contains signals for 640.2 nm laser pulses.

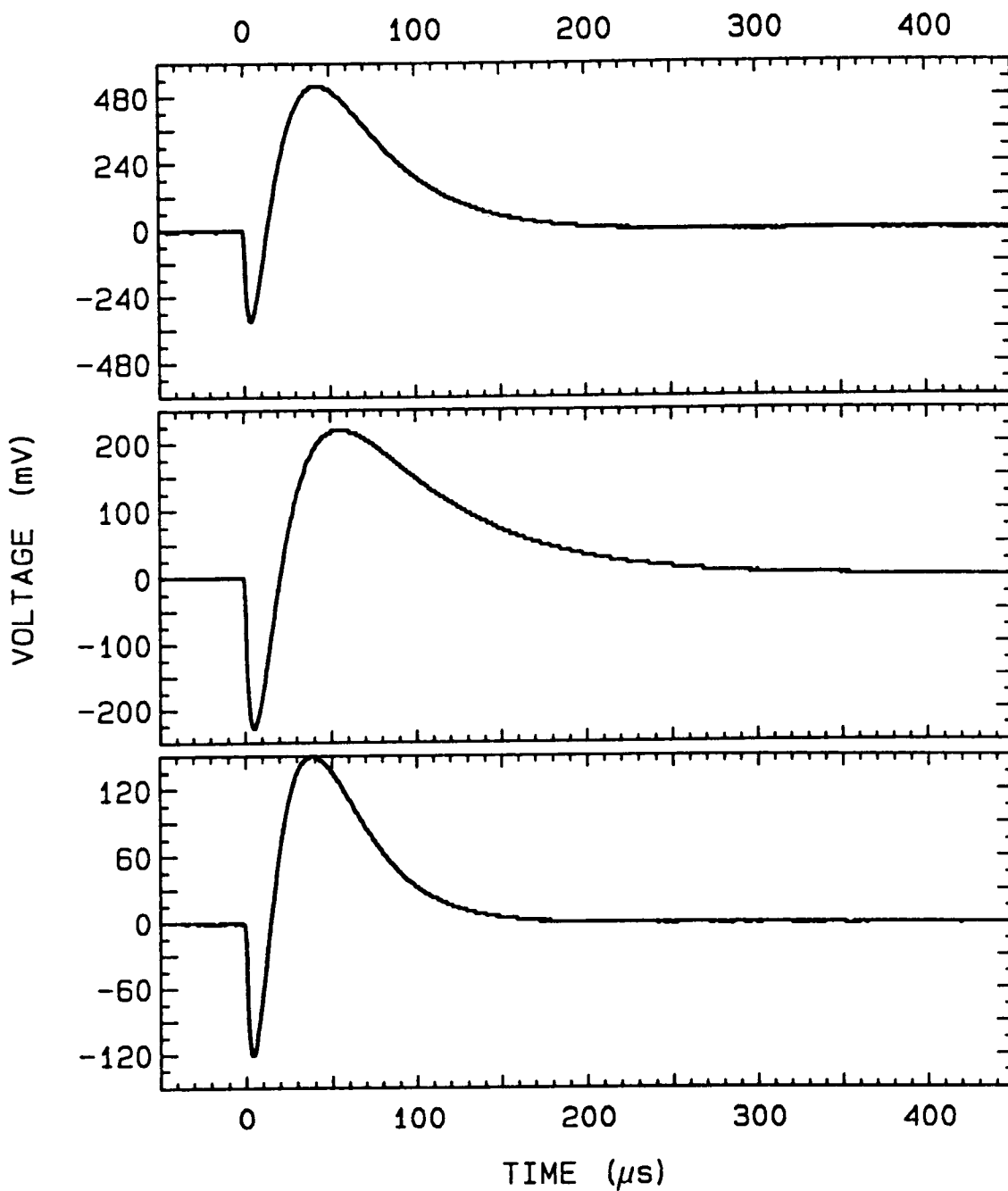
D1



Voltage change with time due to 588.2 nm laser pulse for various discharge currents. The current was 0.2, 0.5, 1.0, and 1.5 mA from bottom to top. The neon pressure was 20 torr.

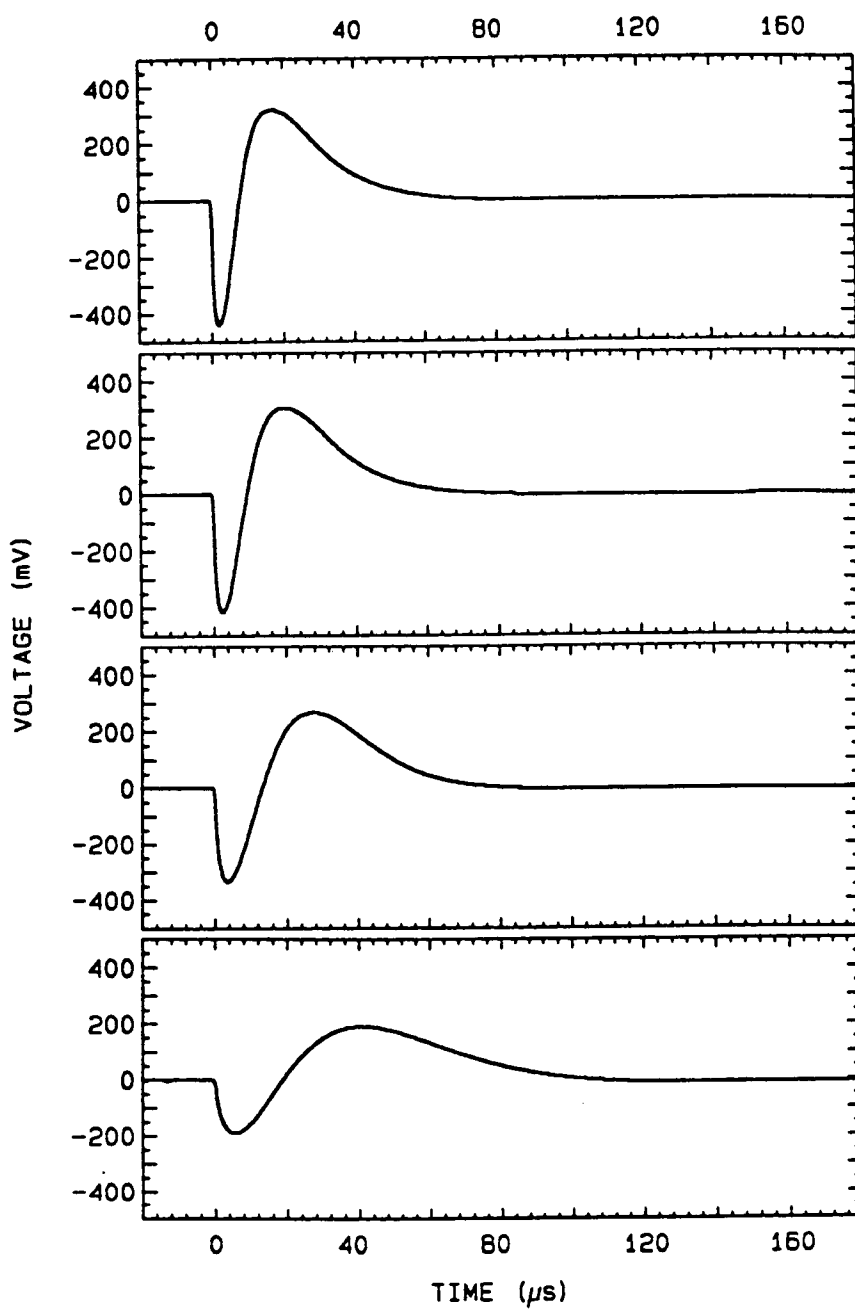


Voltage change with time due to 588.2 nm laser pulse for various laser intensities. From bottom to top the laser intensities were 14%, 41%, and 55% of full strength, respectively. The neon pressure was 20 torr and the discharge current was 1.0 mA.



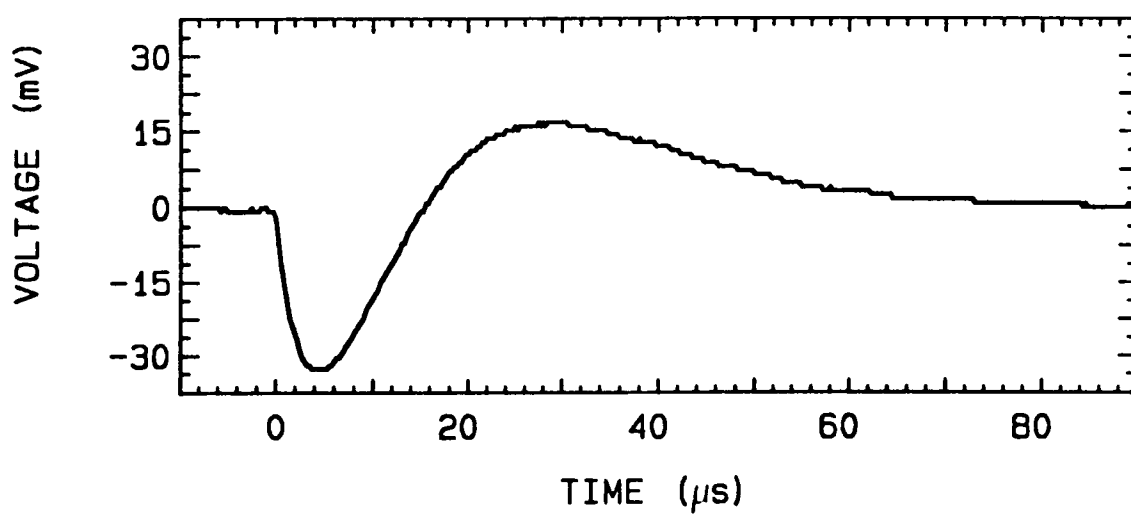
Voltage change with time due to 588.2 nm laser pulse for various transverse laser beam locations in the discharge. The signals from bottom to top were from beams passing through the anode glow, positive glow, and negative glow respectively. The neon pressure was 4 torr and the discharge current was 0.5 mA.

D2



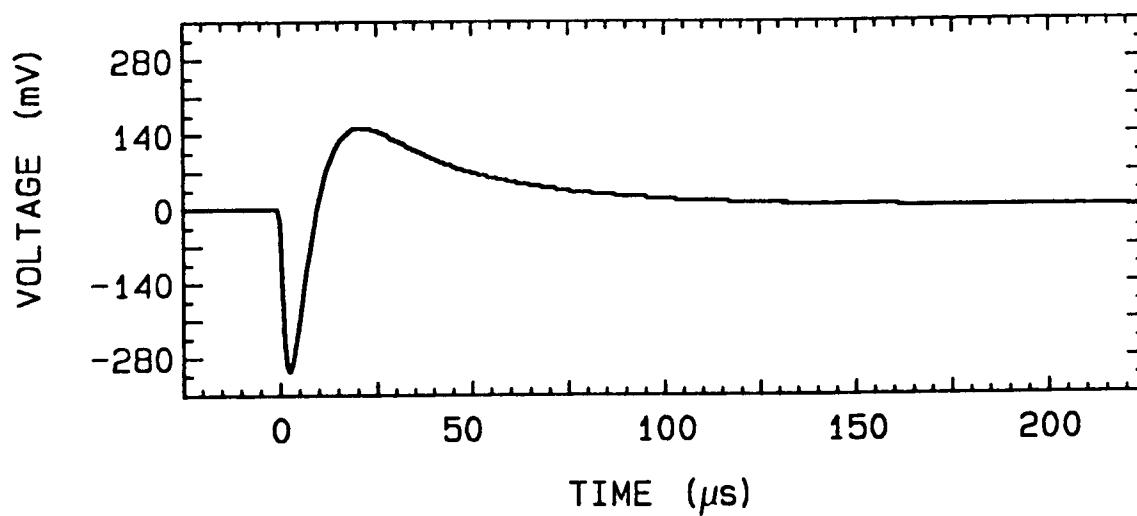
Voltage change with time due to 594.5 nm laser pulse for various discharge currents. The current was 0.2, 0.5, 1.0, and 1.5 mA from bottom to top. The neon pressure was 20 torr.

D3



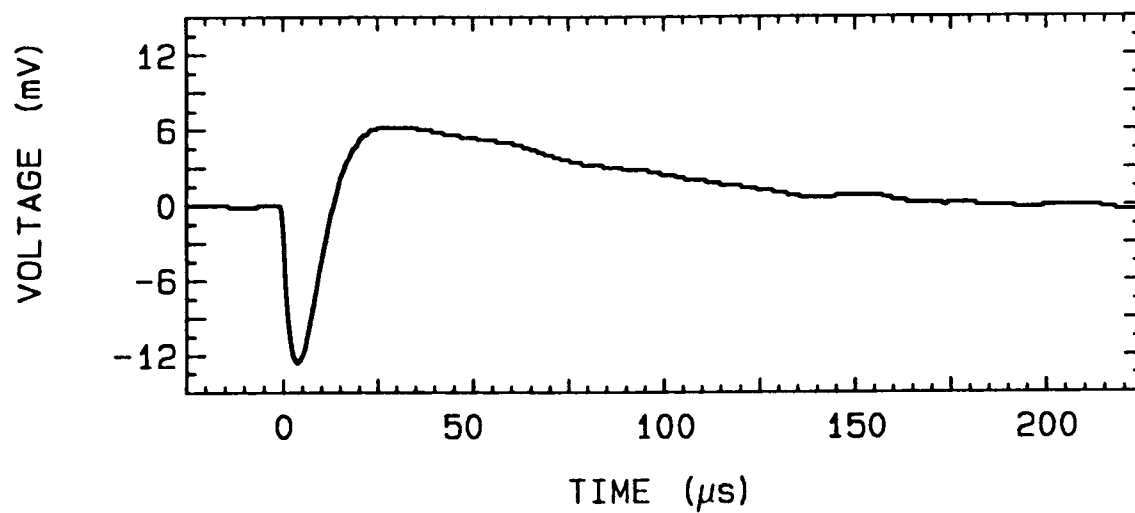
Voltage change with time due to 597.6 nm laser pulse. The neon pressure was 20 torr and the discharge current was 1.0 mA.

D4



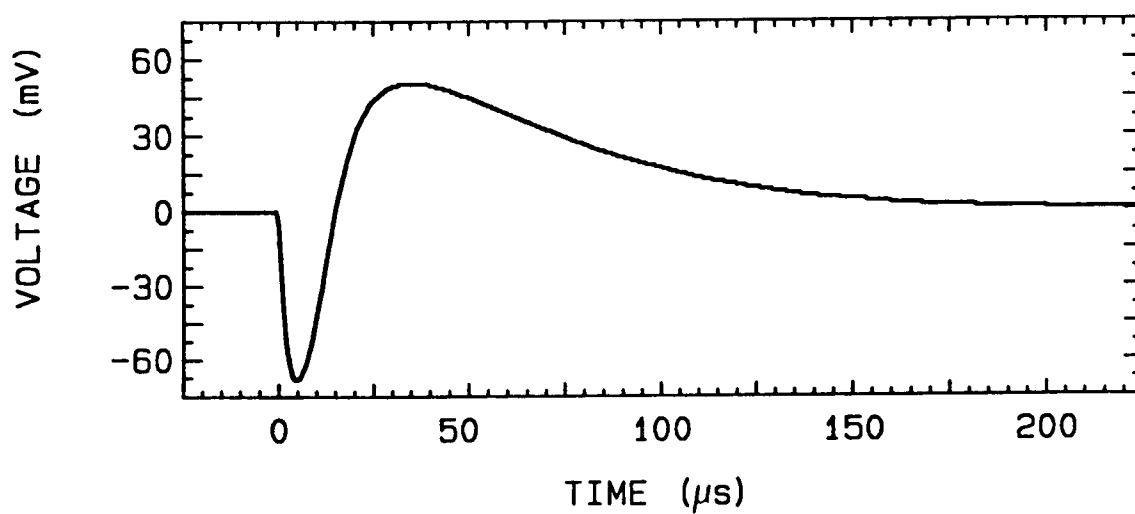
Voltage change with time due to 614.3 nm laser pulse. The neon pressure was 20 torr and the discharge current was 1.0 mA.

D5



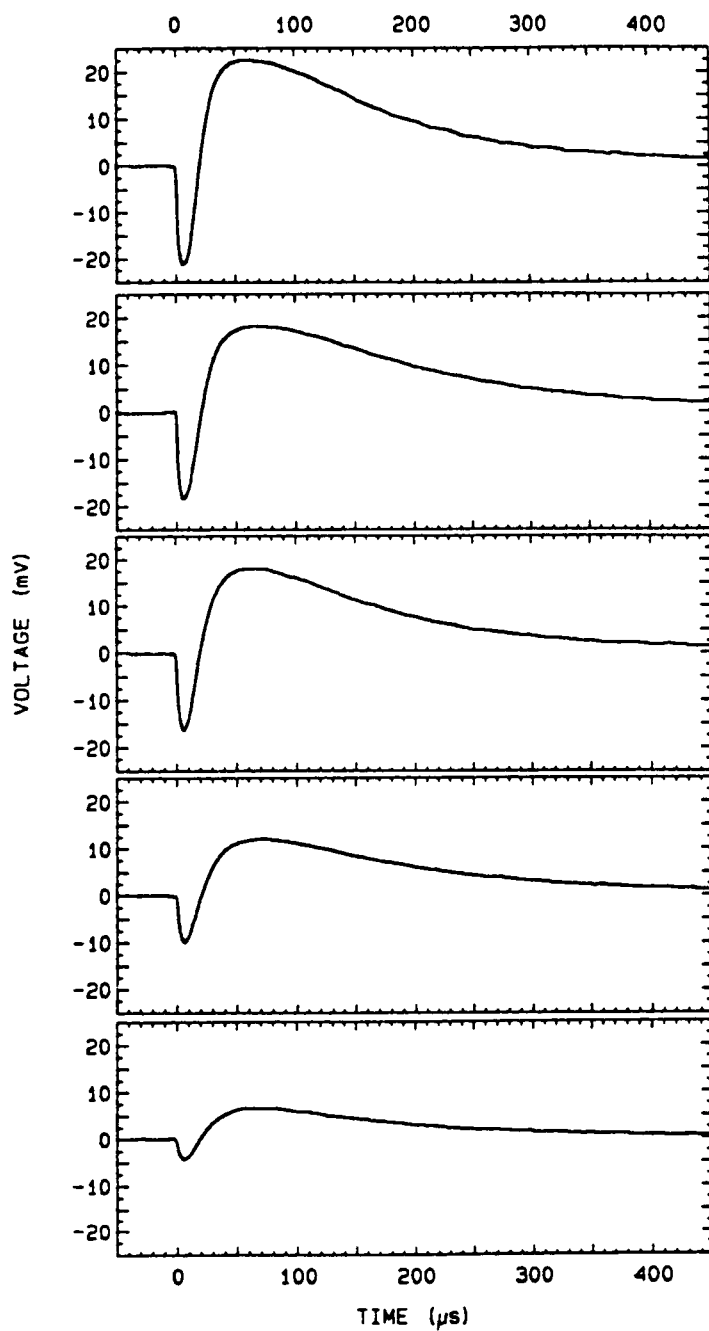
Voltage change with time due to 621.7 nm laser pulse. The neon pressure was 20 torr and the discharge current was 1.0 mA.

D6



Voltage change with time due to 633.4 nm laser pulse. The neon pressure was 20 torr and the discharge current was 1.0 mA.

D7



Voltage change with time due to 640.2 nm laser pulse for various laser intensities. From bottom to top the laser intensities were 14%, 28%, 41%, 51%, and 55% of full strength, respectively. The neon pressure was 20 torr and the discharge current was 1.0 mA.

VITA

Glenn Hughes Harman was born in Panama City, Florida on January 25, 1962. He grew up in Tallahassee, Florida and was graduated from Alfred B. Maclay High School in June 1980. He attended Florida State University, and in April 1985 he received a Bachelor of Science degree in Physics. In the fall of 1985 he accepted a teaching assistantship at the University of Tennessee and began study toward a Master's degree. This degree was awarded in August 1989. He is currently employed as a Research Associate at ORNL.