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"Holographic Particle Detection"**

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Holographic Particle Detection: Progress Report

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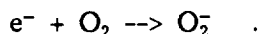
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INTRODUCTION

The aim of this project was to study the feasibility of developing a novel particle track detector based on the detection of $1p-1s$ emission radiation from electron bubbles in liquid helium. The principles, design, construction, and initial testing of the detection system have been described in previous reports. The main obstacle encountered was the construction of the liquid-helium-tight infrared windows. Despite numerous efforts in testing and redesigning the windows, the problem of window leakage at low temperature persisted. Due to limited time and resources, we switched our attention to investigating the possibility of using room-temperature liquid as the detection medium. A possible mechanism was the detection of de-excitation radiation emitted from localized electrons in common liquids where electrons exhibit low mobilities, as suggested in the previous report. The purity of the liquid is critical in this method as the dissolved impurities (such as oxygen), even in trace amounts, will act as scavengers of electrons. In this report, we discuss another mechanism whereby the formation of the superoxide ions (O_2^-) by electron scavenging behavior of dissolved oxygen is exploited to detect the track of ionizing particles. An experiment to measure the ionization current produced in a liquid by a pulsed X-ray beam in order to study properties of the ions is also reported.

DETECTION OF IONIZING PARTICLES BY SUPEROXIDE IONS

Electrons produced by the absorption of ionizing radiation are localized in many liquids and may be detected by their absorption or emission of photons. However, this method demands a very high degree of purity of the liquid. Dissolved oxygen is a well-known scavenger of electrons. If the liquid contains dissolved oxygen, as often is the case, the electrons liberated by the ionization process are readily attached to the oxygen molecules to form superoxide ions.¹



In aprotic solutions (without H^+ ions), these ions are practically stable. However, in aqueous solutions, the superoxide decomposes to form dioxygen (O_2) and hydrogen peroxide.² The reaction (known as "dismutation") proceeds via basically second-order decay kinetics with a rate constant depending on the acidity of the solution.³ At low pH, the rate constant is about $10^6 \text{ M}^{-1} \text{ s}^{-1}$, increasing to about $4 \times 10^7 \text{ M}^{-1} \text{ s}^{-1}$ for pH around 5 and then decreases to approximately $10^5 \text{ M}^{-1} \text{ s}^{-1}$ at a pH of 8.

The superoxide ions have some interesting properties that may be utilized to detect ionizing radiations. An absorption band in the near ultraviolet region has been observed.^{2,3} The absorption peak is located at about 250 nm with a maximum absorption coefficient of about $2400 \text{ M}^{-1} \text{ cm}^{-1}$ and a half-maximum-half-width of about 35 nm (Fig. 1). The absorption has been attributed to a transition between the $2p\sigma_g$ and $2p\pi_g^*$ orbitals. No report of the emission spectra from the excited superoxide ions has been found in the literature. The dismutation reaction of superoxide in aqueous solution is accompanied by a spontaneous light emission in the blue end of the visible spectrum with a peak at 450 nm and a half

width of about 50 nm (Fig. 2).⁴ The emission is found to be enhanced by the presence of carbonate ions.

Suppose that the light emission from the dismutation of superoxide in water is to be used in the detection of ionizing particles. We can then estimate the number of photons that would be emitted by a single minimum ionization particle. Such a particle would typically produce about 10^5 ion- pairs per cm of track with a core about 0.1 micron across, giving an initial electron density of about 10^{15} cm⁻³ or 2×10^{-6} M (Moles/liter). The solubility of oxygen in water at 1 atm, 25°C is 1.3×10^{-3} M and the rate constant for the attachment of electrons to oxygen molecules in water⁵ is 2×10^{10} M⁻¹ s⁻¹. Since the electron concentration is much smaller than the oxygen concentration, the decay of electron concentration with time is essentially first order, with a rate of

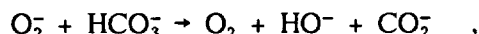
$$k_1 = (2 \times 10^{10} \text{ M}^{-1} \text{ s}^{-1}) \times (1.3 \times 10^{-3} \text{ M}) = 2.6 \times 10^7 \text{ s}^{-1} ,$$

and a mean life of about 40 ns. Thus, the electrons are converted into superoxide ions almost instantaneously. The superoxide ion concentration, N, is then almost equal to the initial electron concentration and, assuming a pH near 5, the rate of dismutation of the superoxide can be estimated by

$$-dN/dt = k_2 N^2 = (k_2 N)N = (4 \times 10^7 \text{ M}^{-1} \text{ s}^{-1}) \times (2 \times 10^{-6} \text{ M})N = \frac{N}{10^{-2} \text{ s}} ,$$

so the decay rate is rather slow.

Another approach might take advantage of the fact that luminescence is known to occur when superoxide ions oxidize certain reagents in aqueous solutions such as luminol (5 amino-2,3 dihydro-1,4-phthalazinedione) and luciferin (a glycoprotein of the boring clam *Pholas dactylus*).⁴ Carbonate ions stimulate luminol luminescence by a factor of 35. The mechanism for enhancement has been hypothesized as



then CO_2^- reacts with luminol. Since the study by chemists of luminol and luciferin seem directed toward developing a test for the presence of O_2^- ions produced by various reactions, we could not find data on the speed or luminescent efficiency of the reactions. If the concentration of luminol were $\sim 10^{-2}$ M and rate constant were $\sim 10^7$ M⁻¹ s⁻¹, the time constant would be ~ 10 μ s. It is not clear from the literature whether a free electron, e^- , can directly oxidize luminol or luciferin; if so, the reaction time constant would probably be much shorter.

If nearly every reaction produced a visible photon, there would be ~ 1 photon per 30 eV, to be compared with ~ 1 photon per 100 eV in the best plastic scintillators. Although this is not enough light to directly "see" tracks, this approach might revolutionize calorimeters which measure total energy in a cascade. High density liquids, such as some bromine compounds and certain water solutions, have not been made to scintillate, presumably because liquid scintillators depend upon a property of the solvent which allows excitons to travel far enough to reach solute molecules which fluoresce. Chemical luminescence activated by e^- or O_2^- might provide a means to make dense liquids "scintillate."

It would also be worthwhile to search for the de-excitation radiation emitted after the superoxide ion absorbs a near UV photon. This emission, if it exists, should be at a longer wavelength, possibly near to the blue end of the visible region.

DETECTION OF IONIZATION CURRENT IN LIQUID

We have investigated the detection of the ionization current produced in liquids by a pulsed X-ray beam with a view toward using the X-ray beam as the ionization source for future experiments. It is important to have a feel for the magnitude of charges produced by the source. The schematic of the experimental setup is shown in Fig. 3. An ionization chamber has been constructed out of a transparent polypropylene vial with two brass electrodes 1 cm apart. The cathode is connected to a variable high-voltage power supply, and the output from the anode is fed into a current-to-voltage converter followed by another stage of voltage amplification. The overall sensitivity of this current detector is 0.4 V/nA. The output voltage signal is then digitized by a CAMAC-controlled waveform digitizer and transported to a microcomputer (IBM XT clone) for processing and display. The X-ray output is triggered by a TTL pulse supplied by a pulse generator operated in the manual mode. Another output from the pulse generator supplies a simultaneous NIM pulse to trigger the waveform digitizer. The X-rays are collimated into a narrow beam by a 1-mm-diameter circular aperture on a 1-cm-thick lead plate. The beam can be directed onto any part between the two electrodes in the ionization chamber.

The X-ray tube voltage is adjustable from 40 to 120 kV. The dose per pulse is determined by the tube current and pulse width, which is adjustable from a minimum of 0.5 ms to a maximum of 2 s. The tube has been calibrated using an air ionization chamber. It was found that when operated at 100 kV and 500 mA, the exposure rate at a distance of 50 cm from the source is about 15 mR/ms, which is equivalent to about 2.4×10^{10} ion-pairs/g of air.

The system was first tested with air in the chamber. A typical current pulse is shown in Fig. 4. The X-ray tube was operated at 50 kV, 300 mA, 1 ms, and the high voltage across the electrodes was set at -50 V. The X-ray beam was directed just above the cathode surface. The observed charge collected seemed to agree with the predicted value. The exposure was estimated to be 2 mR, which was equivalent to 6×10^{-13} C/cm³ of air. The observed pulse height was 200 pA with a width of approximately 4.5 ms. The charge collected was approximately $(200 \text{ pA} \times 4.5 \text{ ms})/2 = 4.5 \times 10^{-13}$ C.

Pure ethanol was next placed into the chamber. Figure 5 shows a typical pulse obtained. Since ethanol has a high intrinsic conductance, a capacitive coupling was inserted between the anode of the chamber and the input into the current-to-voltage converter (Fig. 6) to isolate the dc component. The noise was large in this case, so that the average of 10 pulses was required to get a reasonably noise-free signal.

When FC72 fluid was used in the chamber, an unexpected signal was obtained. A positive pulse was obtained instead of a negative pulse as seen in the previous two cases (Fig. 7). When the polarity of the high-voltage supply was reversed, the signal pulse also reversed its polarity into a negative pulse. We measured the pulse height for different values of the high voltage applied at the electrode and obtained the result shown in Fig. 8. It seemed that the positive pulse existed even when the high voltage was set to zero and a voltage of a few hundred volts in the reverse direction was required to nullify the signal. At higher voltages, the polarity of the pulse was reversed. We were unable to explain this observation.

CONCLUSION

We have investigated several methods of detecting ionizing radiations using light emission from electrons or impurity ions in liquids. Further experiments still need to be done to establish feasibility of using these methods in detection of ionizing particles.

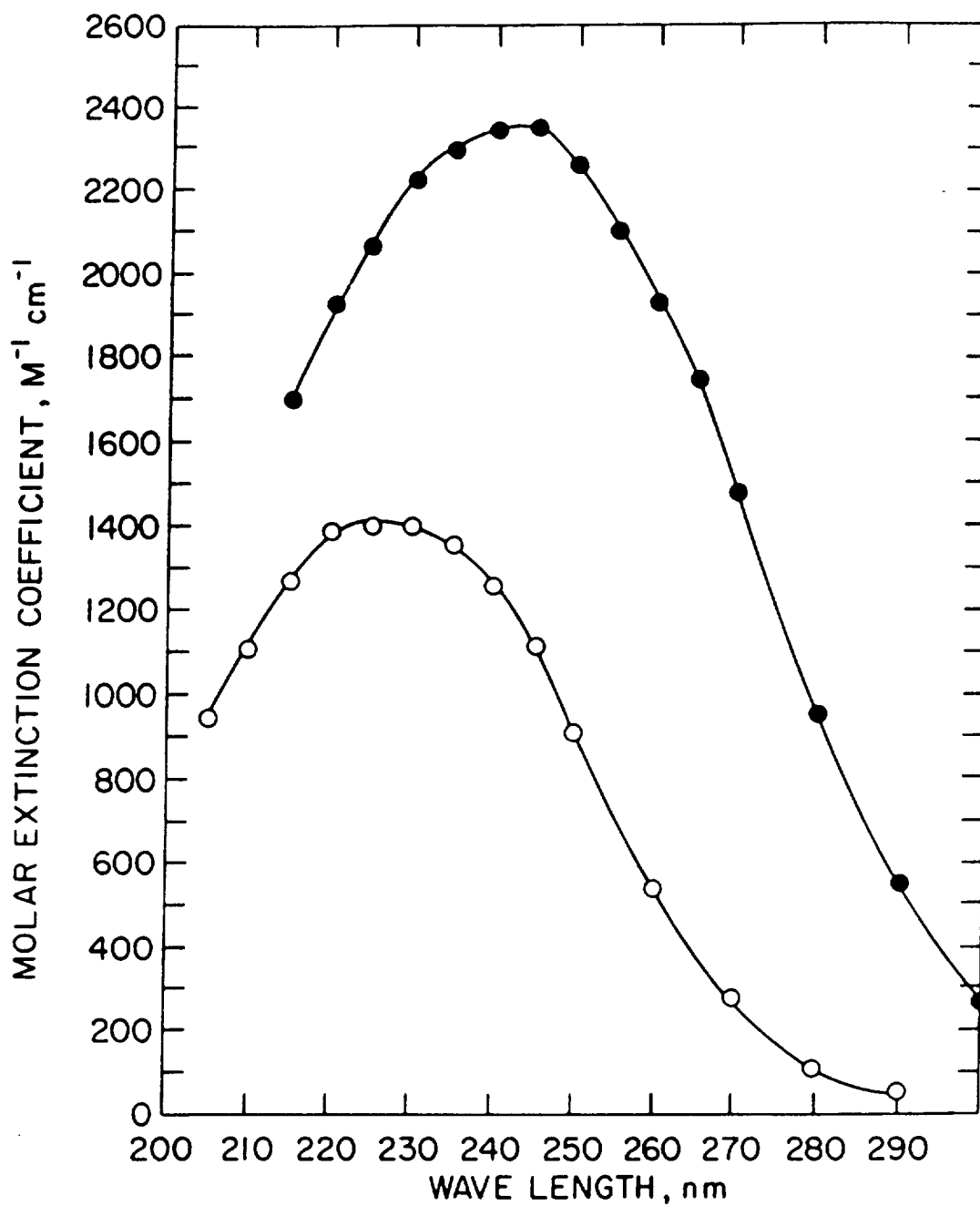
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FIGURE CAPTIONS

- Fig. 1. Absorption spectra of O_2^- at pH 7 (upper curve) and HO_2 at pH 2 (lower curve) (adapted from Ref. 3).
- Fig. 2. Emission spectrum of the system xanthine oxidase-acetaldehyde- O_2 -carbonate ions. Superoxide ions are produced in this system due to the oxidation of acetaldehyde by molecular oxygen catalyzed by xanthine oxidase. (adapted from Ref. 4).
- Fig. 3. Schematic of the experimental setup to detect the ionization current produced in liquid by a narrow X-ray beam.
- Fig. 4. A typical signal trace obtained with air inside the ionization chamber. The HV supply was set at -50 V and the X-ray tube was operating at 50 kV, 300 mA with a pulse width of 1.0 ms.
- Fig. 5. A typical signal trace obtained with pure ethanol inside the ionization chamber. The HV supply was set at -300 V and the X-ray tube was operating at 100 kV, 300 mA with a pulse width of 1.0 ms.
- Fig. 6. AC coupling circuit used to remove the DC component from the signal. This circuit was needed when the ionization chamber was filled with ethanol, as the DC current arising from the intrinsic conductance of ethanol would overload the current-to-voltage converter.
- Fig. 7. Signal traces obtained with FC72 inside the ionization chamber for various settings of the HV supply. The X-ray was operating at 100 kV, 500 mA with a pulse width of 1.0 ms. The HV value in each case was: 1: -2000 V, 2: -500 V, 3: 0 V, 4: +500 V, 5: +1100 V and 6: +2000 V. The traces have been displaced vertically for clarity of presentation.
- Fig. 8. Variation of signal pulse height with high voltage when FC72 was used as the ionization chamber liquid.

Fig. 1



Absorption spectra of O₂⁻ at pH 7 (upper curve) and HO₂⁻ at pH 2 (lower curve).⁴⁴

Fig. 2

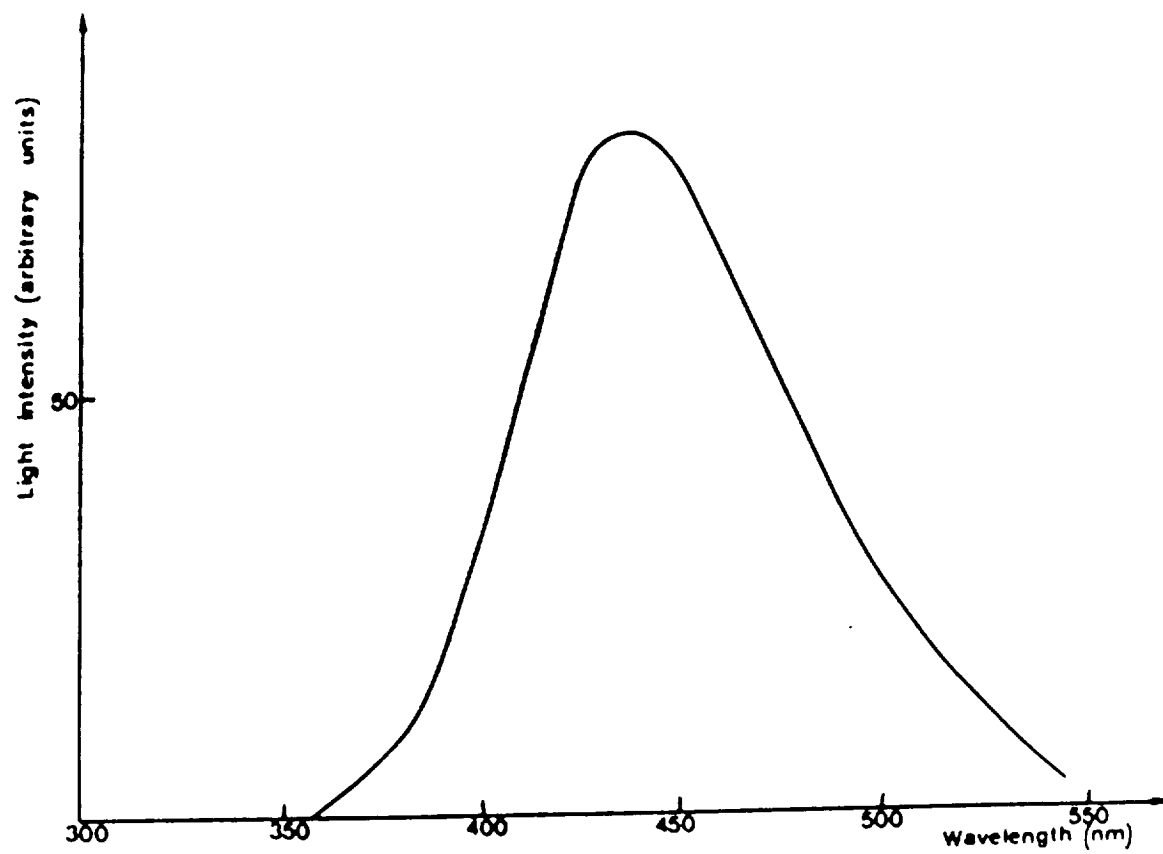


Fig. 3

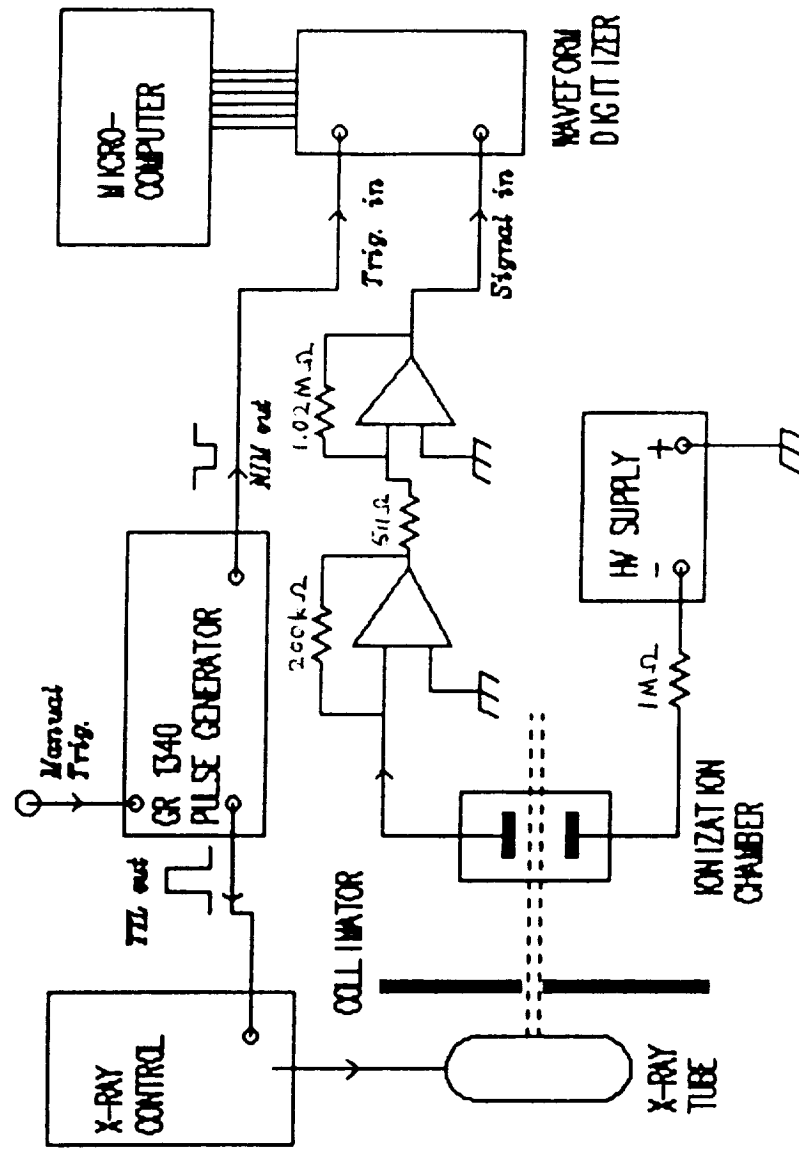


Fig. 4

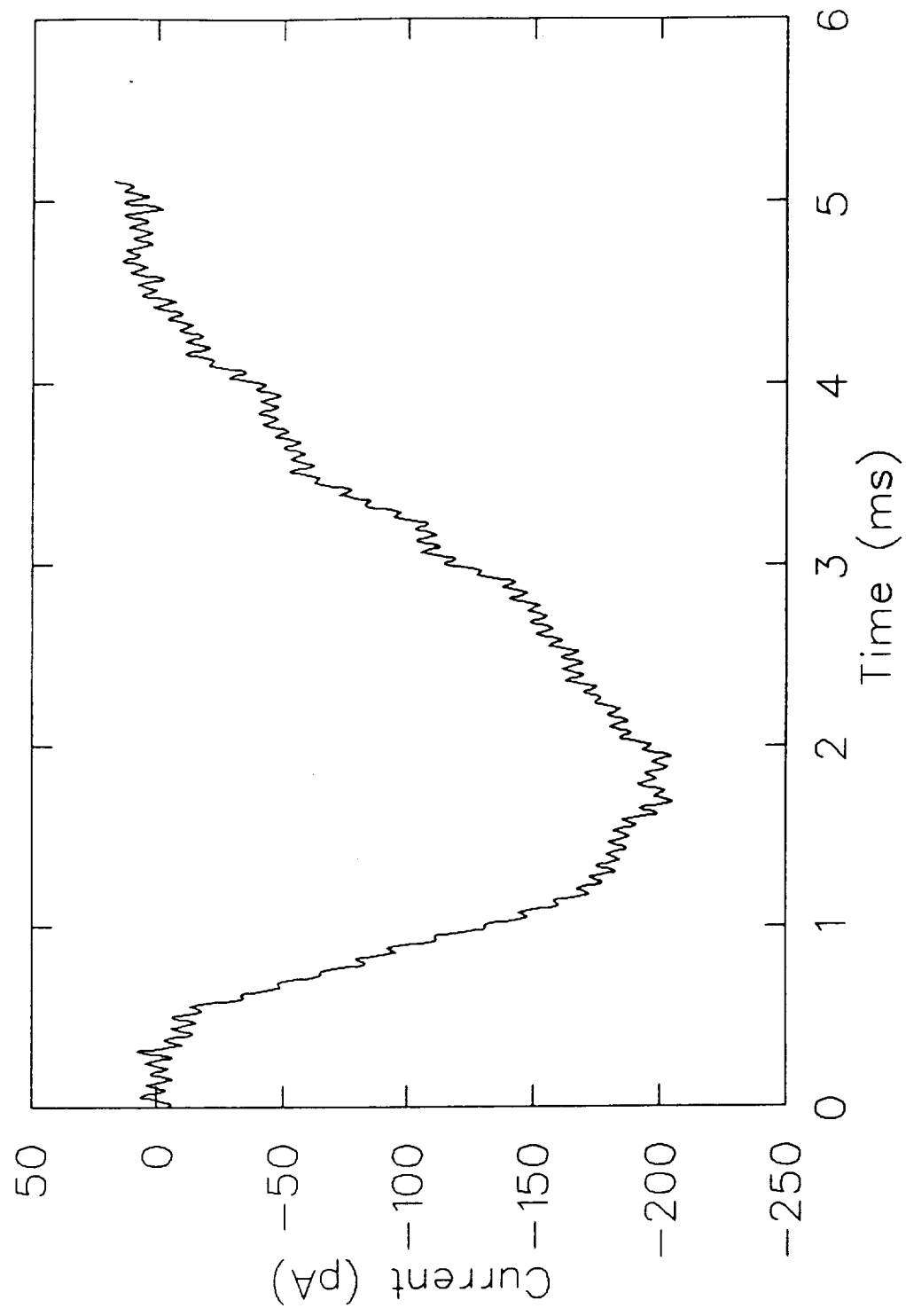


Fig. 5

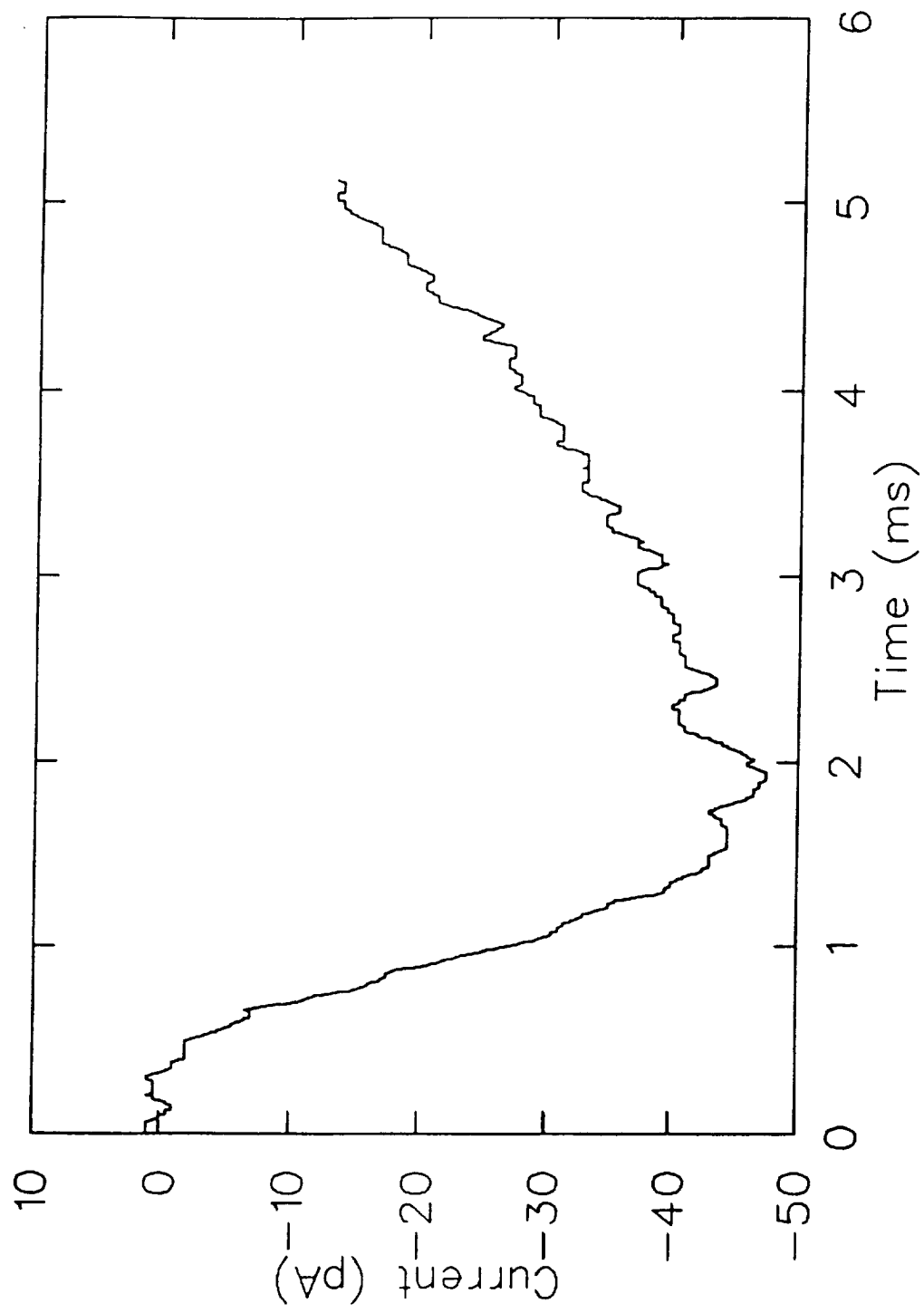


Fig. 6

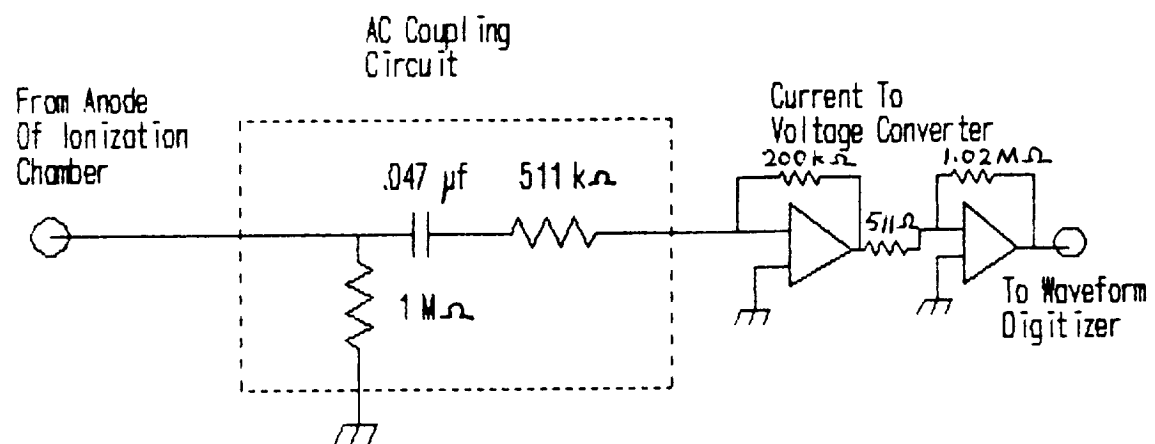


Fig. 7

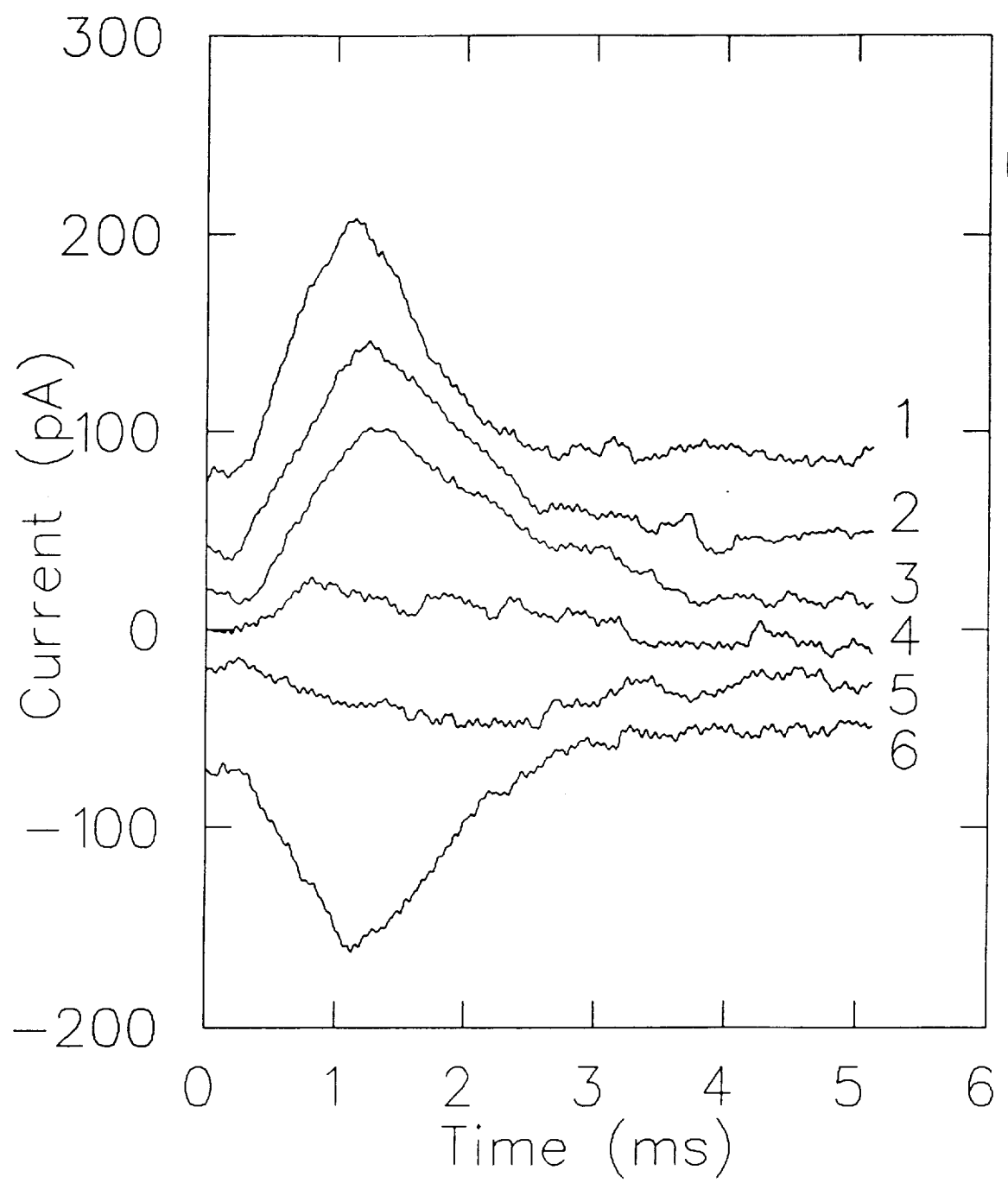


Fig. 8

