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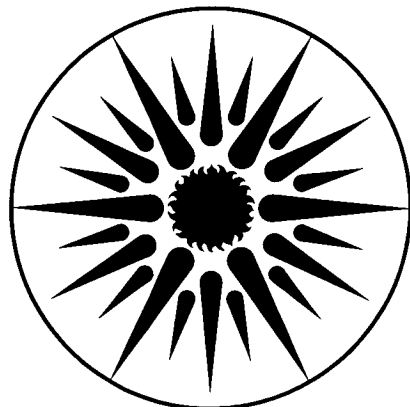
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MOLECULAR BEAM SAMPLING

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April 1983

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LBL-15799
WSS/CI 83-24

EFFECTS OF TRANSIENT COMBUSTION PHENOMENA ON MOLECULAR BEAM SAMPLING

D. Lucas, R. Peterson, F.C. Hurlbut, and A.K. Oppenheim
Energy and Environment Division
Lawrence Berkeley Laboratory
University of California
Berkeley, CA 94720

Western States Section/The Combustion Institute Paper No.83-24
Spring Meeting
Pasadena, CA

LBL Report No. 15799
April, 1983

This work was supported by the Director, Office of Energy Research,
Office of Basic Energy Sciences, Chemical Sciences Division of the
U.S. Department of Energy under Contract No. DE-AC-03-76SF00098.

ABSTRACT

The measurement of transient events with a molecular beam mass spectrometer is often difficult due to the rapid and large changes in temperature, pressure, and chemical composition encountered in a reacting mixture during combustion processes. We are studying how results arising from varying the source parameters under steady-state conditions can be used in interpreting data from a flash-initiated combustion mixture.

Time of arrival curves from heated argon beams are measured and compared to curves calculated using an algorithm based on a physical modeling of the time-of-flight process. Results indicate that the source temperature determines the peak arrival time, and that the beam half-width is essentially constant during a typical combustion event. These results are then applied to a reacting $\text{H}_2/\text{O}_2/\text{NO}_2/\text{Ar}$ system.

INTRODUCTION

During the past decades modulated molecular beam mass spectrometers (MBMS) have provided much new and valuable information about the chemical species present in a wide range of combustion systems. Examples include the pioneering work of Deckers and Van Tiggelen¹, laminar flame measurements of Milne and Green^{2,3}, low pressure flame studies of Peeters and Mahnen⁴ and of Biordi⁵, sooting flame investigation of Bittner and Howard⁶, and rocket exhaust probing of Goshgarian and Solomon⁷. Most of the studies have employed steady-state conditions to facilitate data acquisition and analysis, while just a few were concerned with transient combustion reactions. Young et al.⁸ sampled gases from an internal combustion engine, Milne et al.⁹ measured rapid pyrolysis, and Sloane and Ratcliffe¹⁰ investigated a spark ignited methane-oxygen flame. The most prominent theoretical limitations inherent in time resolved measurements were discussed by Smith¹¹.

We have constructed a direct sampling molecular beam system for the study of transient processes associated with ignition of lean homogeneous gas mixtures. The phenomena under study include thermal and chemical kinetic processes, including auto catalysis, electrical excitation, and photochemical effects. In the course of our studies we must not only determine a number of parameters such as kinetic rates, diffusivities, and conductivities, but also take into account a variety of perturbations introduced by the sampling method on the reacting mixture and the effects of rapidly changing temperature, pressure, and chemical composition on the formation of the molecular

beam itself. In this paper we address some of the problems associated with the temperature changes during transient combustion events, using time-of-flight (TOF) results obtained under steady-state conditions to help understand and interpret results in unsteady systems.

EXPERIMENTAL

The Molecular Beam Mass Spectrometer (MBMS) apparatus used in this investigation is shown schematically in Fig. 1. It is characteristic of such devices in that three stages of differential pumping are used to isolate the detection chamber from the sampling region. First stage pumping is provided by a 10" NRC oil diffusion pump (rated speed of 5300 l/s) backed by a Welch Duo-Seal 7 l/s mechanical forline pump. The second and third stages are each pumped by individual Varian 6" oil diffusion pumps with a rated speed of 2400 l/s. Each diffusion pump is backed by a 5 l/s mechanical pump. The third stage also contains a water baffle to minimize back streaming into the detection chamber. Pressures in the three stages are approximately 4×10^{-4} , 5×10^{-5} , and 2×10^{-7} torr, respectively, during experiments.

The sampling orifice is circular with a hole diameter of 100 micrometers. It is in the apex of a 120 degree included angle quartz cone which is made by a vacuum forming process followed by selective polishing. This procedure produces a sampling orifice with a small channel length with respect to the orifice diameter. A Pyrex skimmer is located 1.15 cm from the sampling orifice, or 115 x/d (skimmer distance to orifice diameter). It has an included internal angle of 60 degrees and a skimmer orifice diameter of 762 micrometers.

The mechanical chopper is an aluminum disk 0.165 cm thick and 5.08 cm in diameter, with eight 0.165 cm wide slots evenly spaced around the periphery of the disk. It is located 8.7 cm from the source. A timing aperture is placed inside the slots which allows a photodiode-detector assembly to signal each revolution. The chopper is spun by a synchronous motor (Globe Industries Model #53A1000-2) lubricated for vacuum service. The two phase drive circuitry allows operating speeds in excess 200 Hz. The distance from the chopper to the detector plane is 20.5 cm.

A 0.114 cm diameter collimating aperture is located between the second and third stage vacuum chambers. The detector is an Extra-Nuclear quadrupole mass filter, equipped with a cross beam electron impact ionizer. Filtered ions are detected by a 21 stage copper-beryllium particle multiplier tube (PMT). The current output from the PMT is dropped across a 100 K Ω resistor to ground, and the resulting voltage is amplified by a Pacific Photometric amplifier (model # 60A-2).

Signal acquisition is performed by a 10-bit, 4-channel, high speed analog-to-digital converter (ADC) with internal buffer memory (Lecroy model # 8210). A DEC LSI 11/2 computer controls the data acquisition process by using the photodiode-detector signal from the beam chopper to initiate the ADC. Once the amplifier signal has been digitized the computer down loads the buffer memory and the data is displayed and stored appropriately. Typical one shot data acquisition runs included digitizing 500 points for each channel with a sampling rate of 200 Khz. The maximum digitizing rate is 1 Mhz.

Argon TOF experiments were performed with the experimental setup

schematically portrayed in Fig. 2. Argon was passed through an electrically heated furnace positioned directly under the MBMS sampling orifice. A chromel-alumel thermocouple was placed near the sampling orifice to allow temperature measurements of the argon in the vicinity of the sampling region.

The analysis of the experimental data requires extremely accurate temporal positioning of the shutter function. Detection of the position of the shutter function ideally would be concurrent with the detection of the TOF signal. This can be accomplished by directing a laser through the molecular beam path with detection by a photodetector. Thus both the shutter function position and the resulting TOF curve can be obtained simultaneously with little, if any, timing error. In our particular experiment the presence of the thermocouple immediately in front of the sampling orifice precluded this signal acquisition technique. However, measurements of the laser gate timing were taken throughout the experiments, and no variations were observed within the uncertainty of the apparatus (~ 2 usec).

The shape of the shutter function is evaluated at a slow chopping speed, where molecules of virtually all velocities reach the detector within a time period that is short compared to the time required for the chopper to move a significant distance, and by optical detection of a HeNe laser beam that is aligned along the molecular beam path. The laser method should be compared to the beam chopping method to insure that no unexpected optical effects are present. Such a comparison is shown in Figure 3. The molecular beam trace shows that even at a chopper speed of 5 Hz, there is an asymmetry introduced on the falling edge of the signal by the velocity distribution of the molecules. The two curves are not superimposable because of the

finite flight time of the argon molecules, which results in a delay of ~400 usec. However, the agreement observed between the curve shapes validates the optical method, and allows its use even at the high chopping rates used during combustion experiments. The advantage of the optical technique is that it provides a reliable reference time for the shutter function for each slot in the chopper wheel, independent of mechanical imperfections and chopping rate. Also, since the shutter function is a purely geometric entity, it is valid at any chopping speed as long as its width is scaled properly. This scaling is accomplished by comparison with the chopped laser timing pulse obtained during the high speed TOF experiments.

THEORETICAL BACKGROUND

Prediction of the shape and arrival time of the chopped molecular beam is accomplished using an algorithm based on a physical modeling of the time-of-flight process.

Passage of a slot across the collimated beam of molecules allows molecules to continue on to the plane of the detector between the opening and closing times of the chopper (or between t_o and t_c). These times and the shape of the slot determine the shutter function $A(\tau)$. Molecules reaching the detector at a particular time t will have various velocities

$$v(t) = \frac{L}{t - \tau} \quad (1)$$

where L is the distance from the chopper to the detector. The number

of molecules in the range of velocities v to $v + dv$ is given by

$$dn(t, v) = KA(\tau)v^2 \exp\left(-\frac{m}{2kT}(v - U_b)^2\right) dv \quad (2)$$

where K is a normalizing constant and U_b is the local particle velocity. Substituting for v and dv in equation (1) and integrating over yields the time of arrival distribution

$$n(t) = \int_0^{\tau_{\max}} \frac{KA(\tau)L^3}{(t-\tau)^4} \exp\left(-\frac{m}{2kT}\left(\frac{L}{t-\tau} - U_b\right)^2\right) d\tau \quad (3)$$

with integration limits $\tau_{\max} = \tau_c$ for $\tau_c < t$ and $\tau_{\max} = t$ for $\tau_c > t$. The beam temperature T and the local particle velocity, U_b , are unique functions of the Mach number and are calculated from standard gas dynamic flow relations for perfect gases.

To evaluate the time of arrival curve, the shutter function is modeled by breaking the measured function into finite rectangular segments, the time width of each being small compared to the molecular flight times. Each segment is treated as a shutter "delta function", with the molecules originating from one segment having a velocity distribution

$$f(v) = A' v^2 \exp\left(-\frac{m}{2kT}(v - U_b)^2\right) dv \quad (4)$$

Summing the contribution from every segment at each time increment at the detector yields the calculated time of arrival distribution.

To compare the calculated values with experimental curves, two additional factors must be included: the time the ions take to traverse the mass filter, and the response of the detector and associated electronics. The first is determined by fitting a ambient temperature source calculation to an experimental measurement of the peak arrival time, yielding a constant value (Δt) which is added to

the arrival times of all subsequent calculations. The second is determined by a method described by Alcalay and Knuth¹². Detector response is assumed to have an exponential form. The trailing edges of experimental curves are measured at various chopper speeds, and the slope of the logarithm of the measured signal is plotted against the reciprocal of the chopper speed. Extrapolation to infinite chopper speed results in a detector response time, which can then be convoluted with the calculated curve to yield a more realistic time of arrival curve.

RESULTS AND DISCUSSION

1. Preliminary Calculations

Table I lists calculated time-of-arrival curves for an Ar beam at temperatures expected during a combustion event. These calculations use a shutter function that is fit to a measured curve, but assumes no detector delay or response time. The importance of these calculations is that they establish reasonable limits on the time resolution that can be achieved with our apparatus. For Mach numbers that are routinely measured by other researchers (in the range of 10 to 15), it can be seen that the beam half width is not a strong function of source temperature. This is a direct consequence of the beam velocity increasing as the temperature increases. The beam is indeed spreading more rapidly at the higher temperatures, but there is less time for this to occur before the molecules reach the detector plane. It can be concluded that with sufficiently fast chopping, the

problems associated with beam spread due to changing temperatures in combustion systems are not insoluble.

The other significant result of these calculations is that the arrival time of the peak signal is a strong function of the source temperature, but not of the Mach number. A change in the Mach number from 5.0 to 15.0 changes the arrival time of the peak by no more than 6 usec over a range of temperatures from 300 to 2100 K. In contrast, a change in source temperature from 600 to 900 K changes the peak arrival time by ~52 usec. It should be noted that the difference in arrival times does depend on the temperature range, with the higher temperatures exhibiting less of a difference. For a steady-state system or for one where the source temperature changes slowly, the peak arrival times may be used to determine the temperature of the gas being sampled by the mass spectrometer, with a ultimate resolution of ~10 K near ambient temperatures and ~50 K at the higher temperatures. Some of the problems associated with higher temperature gradient systems have been discussed previously^{8,11}, and we will discuss them in more detail later.

2. Heated Argon Beams

To determine how well our experimental system compares to the theoretical expectations determined in the previous section, the time-of-flight curves for heated argon beams under steady-state conditions were then measured. Some of these measurements are shown in Figure 4. The curves were obtained at identical chopping rates, and each curve was recorded without signal averaging. However, the stability of the system was such that numerous sweeps could be superimposed with no measurable change in the curve shape. The

location of the thermocouple for the source temperature determination had no effect on the measurements. The curves are normalized, and thus do not reflect the differences in intensity due to their varying source density. As expected, the hotter beams arrive at the detector more rapidly than the cooler beams, and the half widths of all the curves are not greatly different. There is evidence for a finite detector response on the falling edge of the curves, especially at the higher temperatures.

Fitting the calculated curves to the measured curves employed the room temperature beam results to determine the detector delay time and response. For argon and the mass spectrometer settings used in this study, a delay time of 48 usec and a response time of 55 usec were obtained. These values were then used as constants in the other calculations, where the source temperature and Mach number could be varied to obtain the best fit.

An example of how the calculated curves match the experimental curves is shown in Figure 5. This figure is for the highest temperature beam measured, ~880 K. The best fit is a beam temperature of 855 K and a Mach number of 12.0. The values used to match the other measurements are shown in Table II. The calculated temperatures match the experimental values to within the estimated uncertainty in the measured values. The Mach numbers vary smoothly from 15.0 at 305 K to 12.0 at 880 K. This trend can be interpreted as a viscous perturbation of the centerline Mach number in which the increased viscosity and the reduced density of the argon at the higher temperatures reduce the effective Reynolds number at the source¹³. Thus both the temperature and the Mach number we determined in the

steady-state argon beams reflect the lack of significant unexplained perturbations by the sampling system, and indicate that chopped beam measurements may be useful in interpreting transient combustion events.

3. Implications for Transient Combustion Events

Molecular beam sampling of time varying systems is much more difficult than in steady systems. Changes in temperature, pressure, and chemical composition of the sampled gas can alter the resulting molecular beam. If such changes are not well characterized, the end results of combustion experiments, the determination of species concentrations as a function of time, will be significantly in error. It is for this reason that we are utilizing a chopped molecular beam to study systems in which chopping will ultimately be eliminated or reduced in order to increase the signal strength of the species of most interest, the free radicals and other reactive chemical species.

Our results are to be used for the determination of the limits of our measuring system. For these estimates we assume that the reactive mixture under study is predominantly argon, and that homogeneous combustion occurs in a constant volume vessel. Such assumptions simplify the analysis in that the average molecular weight of the mixture is essentially constant during the reaction, and the density at the source does not vary significantly as the temperature increases. Thus the source Reynolds number remains reasonably high during the combustion event, changing only with the viscosity and acoustic velocity of the gas mixture.

In discussing time resolution, it should be noted that there are two regions in the beam path that need to be evaluated: the region

from the source orifice to the chopper, and the region from the chopper to the detector. We will first discuss the region from the chopper to the detector.

There are several factors determining chopping rates and resolution. Two important factors, the beam Mach number and the chopper to detector distance, are not easily altered. We thus fix these values for our system and examine the effects of changing the shutter function and the chopping rate.

The length of sampling time provided by the chopper does not have a significant impact on the resolution as long as this time is not much greater than the half width of the detected beam resulting from an extremely short shutter function. As an example, the half width resulting from a chopped argon beam at a source temperature of 300 K, a Mach number of 15.0, a detector response time of 55 usec, and a shutter open time of 2 usec is 84 usec. Increasing the shutter open time to 80 usec increases the half width to only 94 usec. The same calculations using a zero detector response time yields values of 42 and 58 usec.

The rate of chopping cannot be faster than the time necessary to resolve adjacent peaks. In a steady-state system this is determined solely by the half-width of the signal. In a combustion system, however, a rapid increase in temperature can cause a hotter beam segment, moving at a higher velocity, to overlap a cooler, previously chopped beam segment. Assuming that two adjacent chopped segments differ by 1800 K, the two beams will arrive at the detector at the same time if they originate at the chopper 220 usec apart (a chopping rate of ~4500 Hz). To achieve essentially complete resolution, the

chopping rate can be no greater than ~ 2500 Hz (with a subsequent limit on the rate of temperature increase of $\sim 5 \times 10^6$ K/sec, certainly a reasonable figure for combustion events). In our experiments we limit the chopping to 2000 Hz to remain safely within this constraint. At this rate our slots have a width of ~ 65 usec, a value which gives good time resolution with reasonable signal strength. Of course, measurements may be obtained at intervals much shorter than the chopping frequency if the transient event is reproducible enough to allow repeated experiments with differing delays between the chopper phase and the combustion event.

An additional complication arises from the same velocity differences existing between the source and the chopper. The effective width of the chopper will be larger than actually measured, and depends on the rate that the temperature rises within the reaction chamber. A simple model using only the peaks of the velocity distributions, a source to chopper distance of 8.7 cm, a rate of temperature rise of 4.5×10^6 K/sec, and a chopper open time of 65 usec results in an estimate of the time window that molecules entering the source can pass through the chopper. For a system starting at 300 K this time is 113 usec, and for 600 K it is 85 usec. The time approaches that of the chopper open time as the initial temperature rises due to the decreasing velocity difference of the molecules. Such calculations give no clue as to the shape of the chopped beam, and much more rigorous calculations are currently being done in order to better model the source/chopper/detector interactions. However, it appears possible that we will be able to obtain a time resolution in our system of ~ 100 usec throughout a combustion event.

4. Transient combustion

We applied the chopped molecular beam technique to study the combustion in a hydrogen-oxygen system. The reactants are diluted with argon, and the mixture is seeded with nitrogen dioxide, which is photolyzed by high energy flash lamps to initiate the reaction. The mixture ($H_2/O_2/Ar/NO_2 = 0.23/0.12/0.61/0.04$) is initially at 400 torr in a constant volume cell attached to the molecular beam system (Fig. 1.).

Some of the preliminary results using this system are shown in Figure 6. The top half of the figure shows the chopped argon signal. The lighter tracing is from a mixture that has not been ignited, but containing a noise spike at ~ 1.0 msec from the triggering circuit, which serves as a reference mark. The peaks are equally spaced, as expected from a source that remains at room temperature. The bolder trace is from a reacting mixture. The peaks are seen to vary both in time and intensity. However, the two events have been normalized separately, so the peak heights should not be compared directly. The lower half of the figure is a pressure trace from a Kistler Model 211B piezotron recorded during the same combustion event.

From its shape and time behavior of the pressure curve we conclude that the mixture is not reacting homogeneously. The first two peaks are recorded prior to the flash lamp discharge, so they coincide with the room temperature peaks. We interpret the changes in the remaining argon peaks by assuming that a radical-enhanced combustion wave is propagating towards the sampling nozzle. The gases in front of the combustion wave are being compressed, but their temperature is not rising quickly. Thus the density, and the

resulting signal intensity, increases. When the combustion wave passes the nozzle (between peaks 5 and 6), the density drops sharply, and the temperature increases to its maximum value.

The temperature of the sampled argon molecules were estimated by assuming that the average mass and heat capacities of the molecules in the reacting mixture are essentially constant, and that the source Reynolds number was high enough to prevent pressure diffusion in the nozzle¹⁴. Calculations of the peak arrival times yield temperatures for peaks 3 through 6 of 300 ± 25 , 475 ± 25 , 700 ± 100 , and 1900 ± 200 K. The remaining peaks are all near 1900 K. These results tend to confirm our assumption that the combustion wave passes the nozzle at ~ 3 msec.

CONCLUSIONS

We have shown that time-of-arrival curves from a chopped molecular beam system can be used to evaluate the quality of the resulting molecular beam, even for combustion systems in which the source conditions are rapidly varying. The half-width of the signal is not a strong function of the source conditions, and the time of arrival of the signal peak may be used to determine the source temperature. With proper design and characterization of the experimental apparatus, time resolution on the order of 100 usec can be achieved throughout a combustion event.

ACKNOWLEDGEMENTS

We would like to thank Ken Hom for help in performing the experiments, and Dr. Nancy Brown and Prof. Frank Hurlbut for helpful discussions. This work was supported by the Director, Office of Energy Research, Office of Basic Energy Sciences, Chemical Sciences Division of the U.S. Department of Energy under Contract No. DE-AC-03-76SF00098.

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TABLE I

Calculated Argon Arrival Curves

Chopper to detector plane distance = 20.5 cms

<u>SOURCE TEMPERATURE</u> <u>(K)</u>	<u>MACH</u> <u>NUMBER</u>	<u>BEAM VELOCITY</u> <u>(m/sec)</u>	<u>TIME OF PEAK</u> <u>ARRIVAL (usec)</u>	<u>HALF WIDTH</u> <u>(usec)</u>
300	5.0	541	404	134
	10.0	566	400	76
	15.0	572	398	58
600	5.0	765	296	98
	10.0	801	296	60
	15.0	809	294	52
900	5.0	937	252	82
	10.0	982	248	54
	15.0	991	248	50
1500	5.0	1210	206	68
	10.0	1267	202	52
	15.0	1280	202	48
2100	5.0	1432	180	64
	10.0	1500	178	50
	15.0	1513	176	48

Notes: Gating function for all calculations begins at t=0 and ends at t=80 usec.

Time resolution is 2 usec.

TABLE II

Comparison of Experimental and Theoretical Results¹

	<u>TEMPERATURE</u> (K)	<u>MACH</u> <u>NUMBER</u>	<u>TIME OF</u> <u>PEAK (usec)</u>	<u>HALF WIDTH</u> <u>(usec)</u>	<u>SOURCE REYNOLDS</u> <u>NUMBER²</u>
Experimental Beam	305	-	546	92	2230
Calculated Beam	305	15.0	546	93	
Experimental Beam	461	-	488	92	1380
Calculated Beam	435	14.0	488	91	
Experimental Beam	623	-	440	91	910
Calculated Beam	623	13.5	440	91	
Experimental Beam	880	-	404	90	640
Calculated Beam	855	12.0	404	88	

1. All calculated beams have $t = 55$ usec and a delay time = 48 usec.

2. Based on stagnation conditions.

Figure Captions

Figure 1. Schematic diagram of the molecular beam mass spectrometer (MBMS).

Figure 2. Schematic diagram of sampling system and furnace.

Figure 3. Shutter function measured by time-of-flight and laser detection. Solid line is the molecular beam.

Figure 4. Argon time-of-flight curves measured at source temperatures of 880, 623, 461, and 305 K.

Figure 5. Comparison of measured and calculated argon beam at 880 K.

Figure 6. a) Argon beam measured during combustion and non-combustion events.

b) Pressure trace recorded during combustion event.

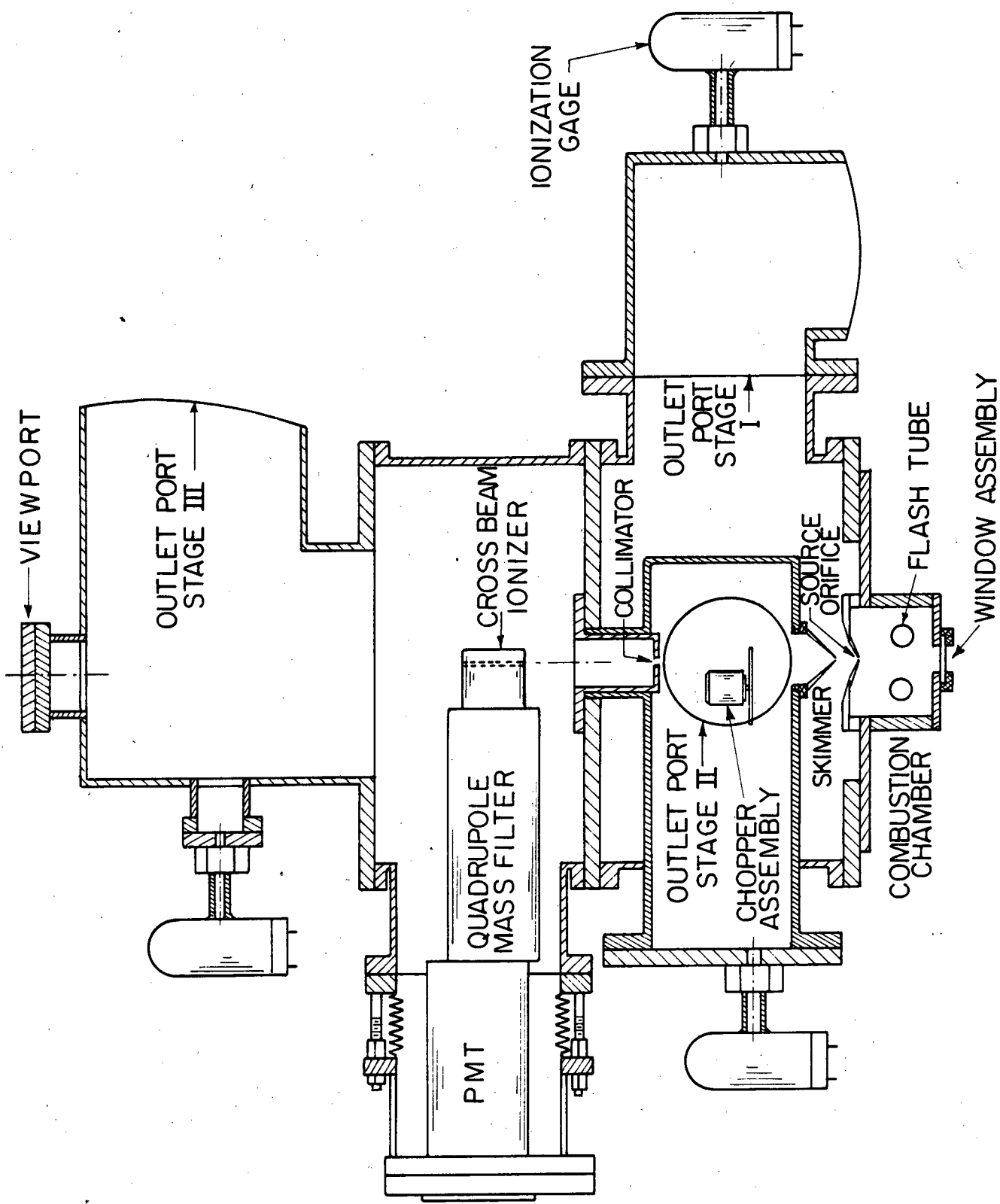
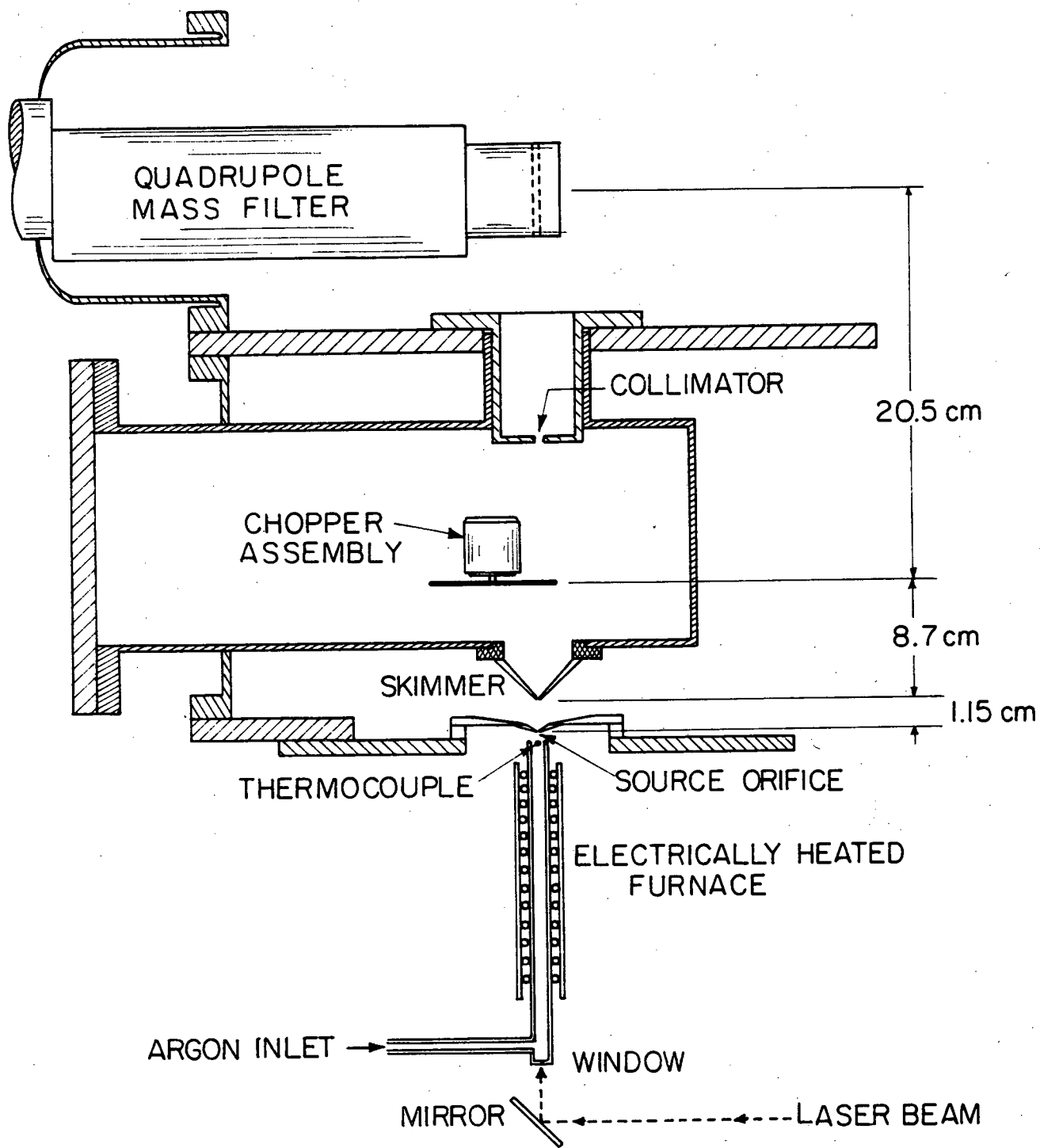
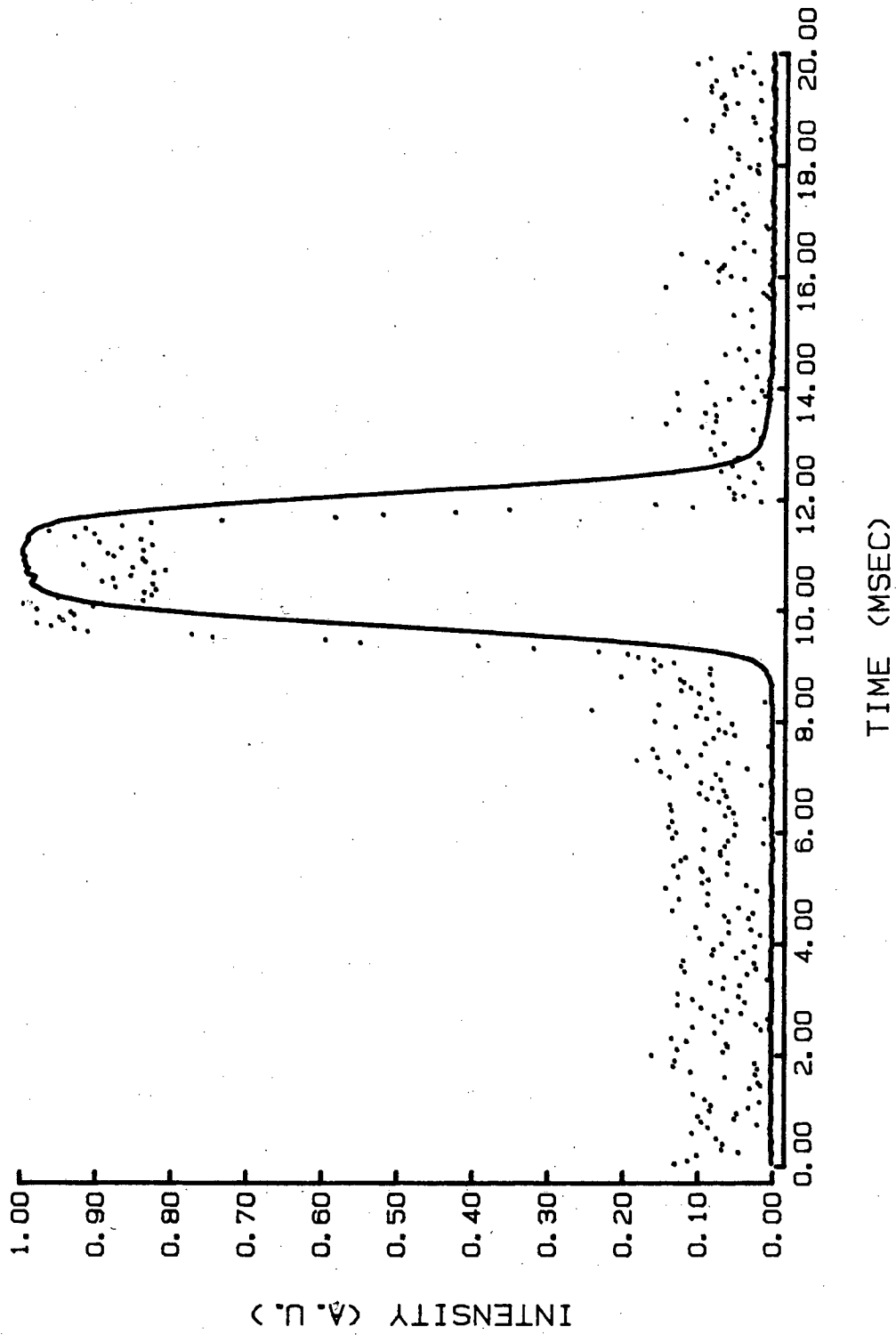


Figure 1



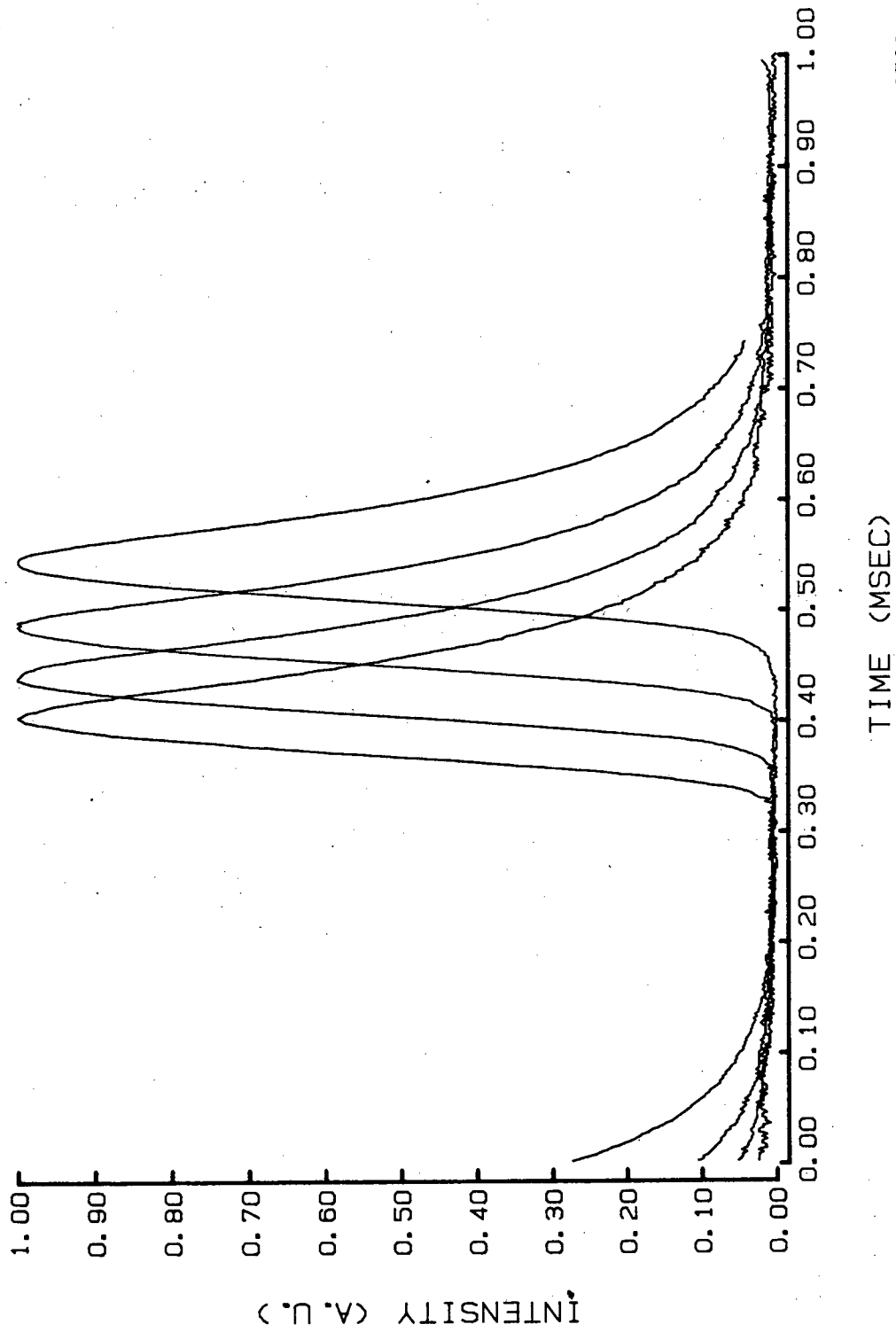
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Figure 2



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Figure 3.



-- XBL 833-8780 --

Figure 4.

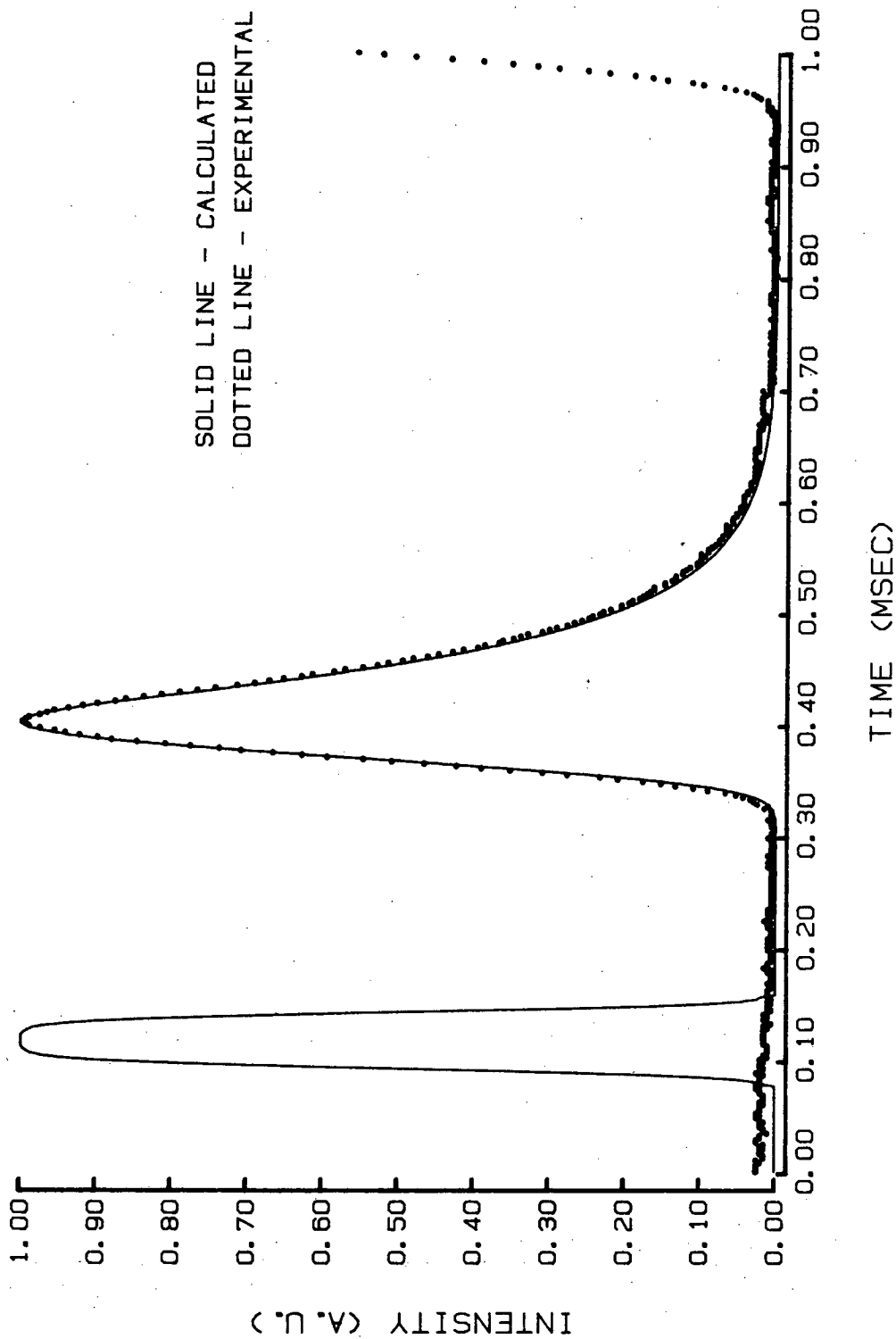
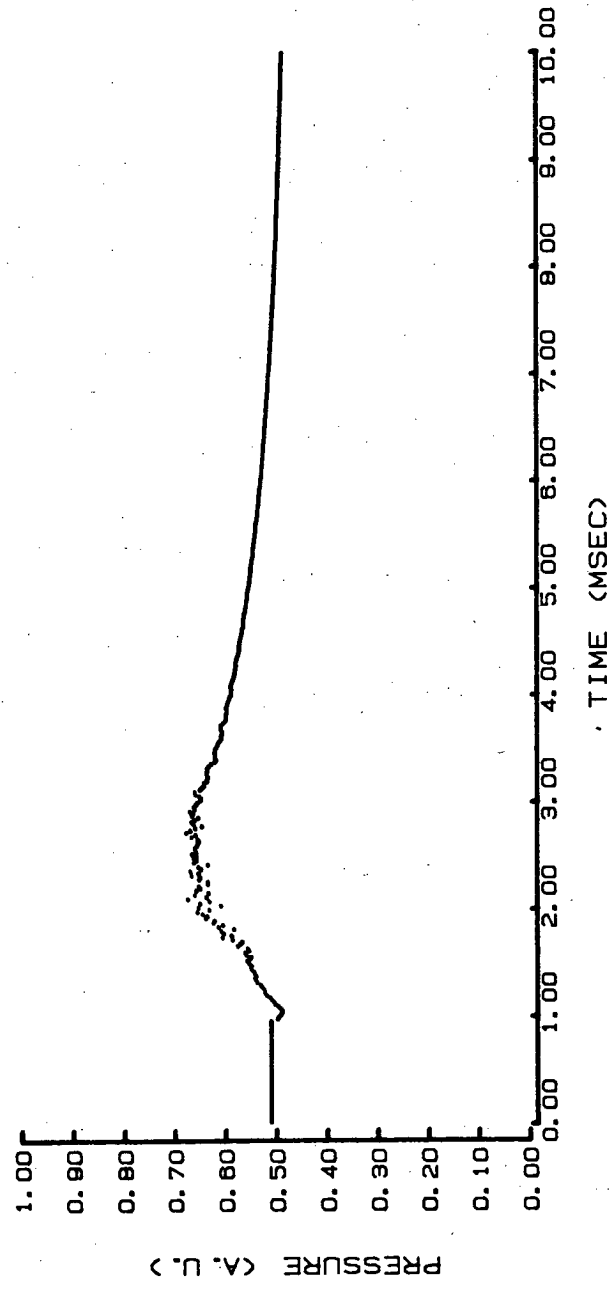
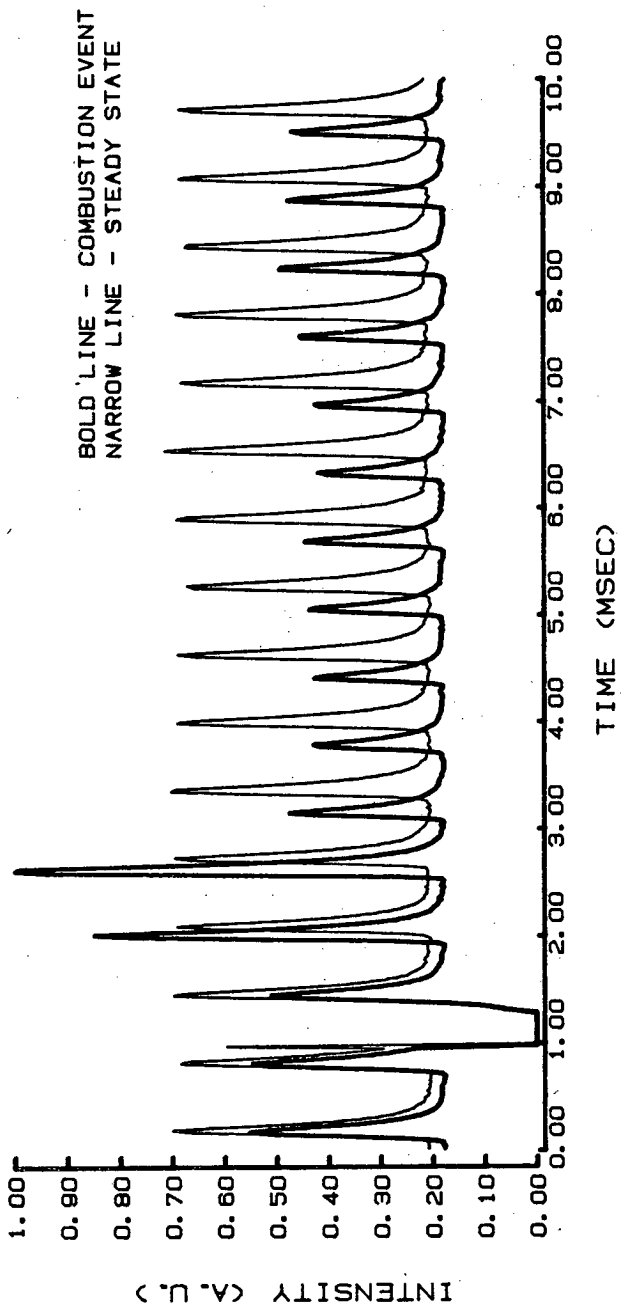


Figure 5.



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Figure 6.

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