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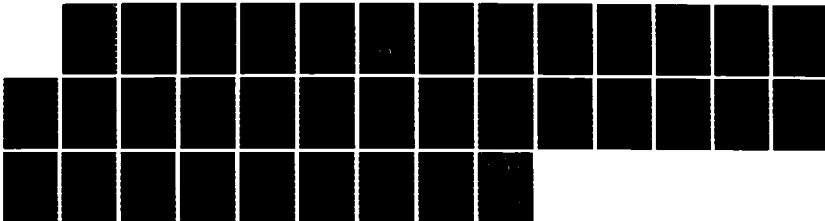
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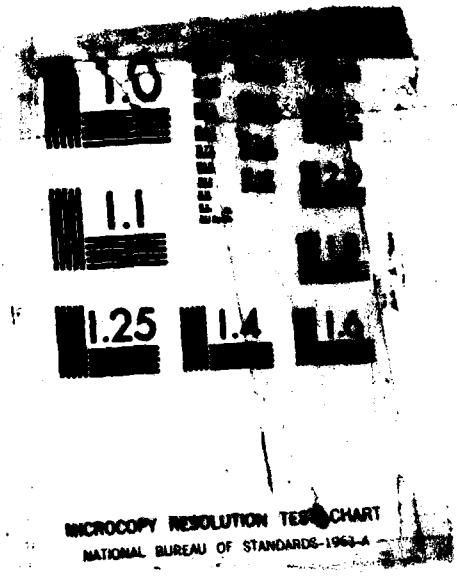
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TECHNICAL REPORT ARAED-TR-85006

**SINGLE-SHOT H(2) CARS SPECTROSCOPY OF PROPELLANT
IGNITION AND COMBUSTION UNDER PRESSURE**

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L. E. HARRIS

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number) A single-shot H(2) CARS spectrometer was developed, and experimental considerations for accurate thermometry were examined. The spectrometer is capable of synchronization with external events such as laser-initiated propellant ignition or ballistic compression, given an inherent delay time of 250 microseconds and a predetermined diagnostic time window approaching 1 second. Additional capability to operate in a double Q-switched mode (two diagnostic probes 60 to 90 microseconds apart) has been demonstrated, with inherent delay time of 190 microseconds. <p style="text-align: right;">(cont)</p>		

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20. ABSTRACT (cont)

fluctuations & intensity

Application to static samples of H(2) and to an H(2)/O(2) flame reveal unreferenced errors approaching 10% or more in temperature determination, and indicate the necessity for simultaneous standardization in accurate thermometry. Intensity determination in an unreferenced fashion reveals fluctuations of 30%, with contributions from YAG and dye laser intensity and phase fluctuations. Q-branch lines from the first vibrational state ($v = 1$) of hydrogen were observed for the first time in the H(2)/O(2) flame using this apparatus. These observations have been incorporated into more accurate spectroscopic constants for use in computer modeling, and concur with ab initio results and recent spectral determination in hydrogen discharge. A parallel reference system has been implemented in the apparatus, and performance evaluation has begun in single-pulse extraction experiments with the laser running at 10 Hz.

Facilities have been established with which to perform single-shot H(2) CARS diagnostics on propellant ignition and flame under pressure. Preliminary experiments indicate that choice of background gas, flow rates through the pressure cell, YAG laser fluence, and dye laser spectral width are critical factors in the development of a working protocol. CARS spectra of nitrogen product gas from propellant burn have been obtained, and spectra of hydrogen evolved in propellant flame have been observed. Furthermore, the effect of oxygen as a background gas has been demonstrated to critically alter the behavior of the propellant. Timing with Ruby laser-initiated propellant ignition has been accomplished, and experiments are anticipated to probe the feasibility of CARS application to this event.

CONTENTS

	Page
Introduction	1
Experimental	2
Laser Systems	2
Transmission and Detection Optics	4
Detection of CARS; Data Acquisition Logic	5
Test Procedures	5
Results and Discussion	7
Triggerable, Single-Shot CARS Spectrometry	7
Basic Advances in Hydrogen CARS Spectrometry	8
CARS Approach to High Pressure Propellant Combustion	8
References	19
Distribution List	21

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INTRODUCTION

The dynamic evaluation of propellant and igniter performance is of particular importance to charge design and quality control efforts of ARDC's Armament Engineering Directorate. Advances in laser diagnostic/visualization techniques, such as holography, laser-induced fluorescence and Coherent Anti-Stokes Raman Scattering (CARS), among others, have made possible extraction of chemical and physical flowfield information in combustion testing and model flame study. This information can be of various dimensionality (3-D, 2-D, pointwise), temporal resolution, and synchronicity, depending on the method chosen and laser sources applied to the problem at hand. The priority (and precision) of required information must first be established in choosing a laser diagnostic/visualization method for propellant and igniter evaluation. The temporal and optical characteristics of the appropriate flowfield and test scenario are factors just as critical in the planning and development of research facilities.

The work reported here is an approach to laser diagnostics of propellant flowfields under pressure using a single-shot CARS spectrometer. The theoretical basis of CARS and its applications in hostile combustion environments have been extensively reviewed (ref 1 through 3). Briefly, two incident laser beams at frequencies ω_1 (the pump beam) and ω_2 (the Stokes beam) will interact in a medium, through the medium's third-order electronic susceptibility (χ^3), to generate coherent radiation at the anti-Stokes frequency ω_3 ($\omega_3 = 2\omega_1 - \omega_2$). When the medium contains a molecule with a Raman-active transition (rotational or vibrational) at the difference frequency $\omega_1 - \omega_2$, the resonant CARS signal is observed. Significant CARS signal can also be generated from a non-resonant medium, creating an experimental interference which should be considered in analysis. The CARS power spectrum can be used to extract concentration and temperature data on the resonant molecule using spectroscopic data, the Boltzmann distribution assumption, and computation procedures of various sophistication.

The major advantage of CARS over other current diagnostic techniques is signal collection efficiency--hence its potential for application to single dynamic events and ease of application to steady combustion events. The CARS power generation is several orders of magnitude greater than spontaneous Raman and, most importantly, the CARS signal is collimated into a small solid angle that is readily collected for spectroscopic analysis. (Raman and emission techniques can only collect a small solid angle of the 4π steradians over which these signals are generated). The CARS technique offers the capability for high spatial resolution (1 cm to less than 1 mm, depending upon configuration) and temporal resolution for single-shot experiments. The potential for accurate thermometry (ref 4) and rapid analysis (ref 3) has been demonstrated by a number of groups.

Disadvantages of the CARS approach to combustion diagnostics have also been documented. The transmissive nature of the technique precludes its application to opaque or highly scattering media, and refractive index gradients can realign the experiment's optical axis. The magnitude of laser power necessary for CARS generation is relatively large, limiting the field of view possible for single-shot measurements and, to an extent, interfering with the flowfield chemistry (refs 5 and 6). The CARS approach to high pressure, confined combustion of propellant is a complex experimental undertaking.

Hydrogen gas has been chosen as the target molecule in these studies because of its large Raman cross section, its presence in the equilibrium products of propellant combustion, and its ubiquity in the laser-induced decomposition of propellant materials. Spectroscopic analysis at frequencies corresponding to the hydrogen Q-branch is facile because the spectrum is unambiguous (the individual rotational lines narrow and separate within a convenient contour for broadband dye lasers) and because no interference from other Raman-active molecules is expected. Previous modeling (ref 7) and experimental (ref 8) efforts have demonstrated the utility of thermometry using the rotational temperature of hydrogen measured by the CARS method. The narrowness of the Q-branch lines does, however, present a challenge for spectrometry using a broadband dye laser for the Stokes beam, a multimode Nd:YAG laser for the pump beam, and optical multichannel analyzers for detection of the resultant anti-Stokes beam.

EXPERIMENTAL

The experimental approach to CARS measurements in propellant combustion is detailed below; the electro-optical system is shown in figure 1. A CARS spectrum is generated by the focused interaction of pump and Stokes lasers in the sample. The gain contour of the Stokes dye laser is chosen (or manipulated) to correspond to the Raman-active frequencies of the target molecule. Using a colinear configuration of the two beams, affectionately known as "USED-CARS," the interaction volume is fairly large, the signal level is at its optimum, and the alignment procedure is reasonably straightforward. Exiting the sampled region, the pump and Stokes beam are blocked, and the anti-Stokes beam (the CARS signal) is diverted to an optical multichannel analyzer (OMA) manufactured by Princeton Applied Research) system for detection and recording.

Laser Systems

The pump laser used for these experiments is a vintage Quanta-Ray DCR 1-A Nd:YAG unstable oscillator-amplifier system whose output is frequency-doubled to produce 0.532 micron laser pulses of high peak power (up to 30 megawatts) and short duration (10 nanoseconds). The output of this laser is multimode and of a bandwidth approaching 1 cm^{-1} . Recently an electronic line narrowing option was installed and the output appears Gaussian a large percentage of the time (>75%).

The green output of the Nd:YAG is split by a dichroic mirror (DM in figure 1) in order to simultaneously pump a Quanta-Ray PDL-1 dye laser and serve as the pump beam in the CARS interaction. A grating operated in Littrow provides the dye laser end mirror; broadband operation is accomplished by aligning the zero-order reflection with the amplifier train optic axis. Higher-order operation yields a progressively narrower, tunable bandwidth. The zero-order output frequencies are those encompassed by the dye gain curve. This side-pumped dye laser produces 8 nanosecond pulses of Stokes-shifted radiation, with conversion efficiencies dependent upon the dyes employed and ranging from 2% to 15%. The amplifier can be pumped in an end-pumped or side-pumped fashion, with the resulting beam shape either toroidal or circular, respectively.

The Nd:YAG laser is the cornerstone of the experiment; control of its firing to within 200 microseconds after external trigger input is readily accomplished with the timing logic and circuitry shown in figure 2. While the mode structure of the laser output may depart from the thermally stabilized ideal, CARS spectra can be obtained in this triggerable fashion. The capability to extract two 10-nanosecond laser pulses from one lamp charge of the Nd:YAG rod has been demonstrated with the circuit shown in figure 3. The resultant time separation can be as low as 90 microseconds, creating the possibility for multiple CARS measurements in such shortlived events as propellant ignition or ballistic compression². Modification of the basic circuit for synchronization with propellant events is accomplished by placing the event within the "blank" time window and triggering the millisecond digital delay generator (DDG) with a voltage level transduced from some characteristic of the event of interest (e.g., pressure rise or light level). The firing sequence must be appropriately "primed" with a TTL level pulse first; the diagnostic laser pulse then occurs 200 microseconds after the event "flag" triggers the millisecond DDG. If the Nd:YAG is to be maintained at its thermally stabilized, 10-Hz repetition rate prior to its "blanking," the internal lamp and Q-switch time base is left operative in the case of single-shot control. For double-pulse control, the Q-switch must be switched from an external time base to the timing set by the microsecond DDG in figure 3.

Explorations of various dye combinations have yielded appropriate systems for use in the Stokes laser at the hydrogen Q-branch (Raman bandhead at $4,160 \text{ cm}^{-1}$, corresponding to a wavelength of 0.683 micron). Nile blue 690 perchlorate lases over the spectral region encompassing the ground state ($v = 0$) Q-branch of hydrogen, with efficiencies of 3% over a bandwidth of $<100 \text{ cm}^{-1}$. The dye system of rhodamine 640 perchlorate/oxazine 725 perchlorate has higher efficiencies, also lasing over the ground state and into the excited state ($v = 1$), with a bandwidth of $\sim 150 \text{ cm}^{-1}$. Addition of NaOH to this solution will shift the gain curve to higher ro-vibronic levels of the excited state. Most interestingly, this dye system can be adjusted to lase in both the hydrogen and nitrogen Q-branch Stokes regions³. One must be careful in utilizing this dye mixture in a dye laser with a side-pumped amplifier, as the amplifier cell can act as a laser

¹ Note that more recent Quanta-Ray models have been fitted with a high voltage relay board, triggering the laser with an intrinsic delay of ~ 3 milliseconds after external trigger input.

² A detection scheme for multiple measurement on this timescale would have to rely on electro-optical deflection of the two CARS beams on the detector array, or alternately on a means of detection which could resolve the two incoming beams in time (such as a fiber optic array fitted to the monochromator at the Q-branch positions, photomultiplier detection of the line intensities and multiple recording with fast digital oscilloscopes). Note also that a higher repetition rate dye laser may be necessary.

³ We have demonstrated a means of multiplexing CARS data, via dual detection, and using this two-color Stokes beam for nitrogen and hydrogen Q-branch recording. More sophisticated multiplex methods require the use of two distinct dye lasers (ref 9).

itself (generating an uncontrollable experimental interference). The energy transfer dye system of LDS 698/DCM has also been used in hydrogen ground and excited state CARS thermometry, exhibiting efficiencies of 8% over a broad bandwidth of 250 cm^{-1} . This system can be tuned over a wide range by altering the concentration of DCM, the donor in the energy transfer couple. The system has been used in identifying higher ro-vibronic levels of hydrogen in laboratory flames. An attempt to create an LDS 698/cycodextrin/water dye was successful in producing a laser at the appropriate wavelength, but efficiencies were prohibitively low. Alternate approaches to Stokes frequency selection, such as using a crude (~ 100 lines/mm) grating in first order as the dye laser end mirror, have not as yet been attempted.

Transmission and Detection Optics

The optical configuration for colinear CARS measurements is shown in figure 1. The pump and Stokes beams are concentrically phase-matched via the alignment of dichroic mirror (DM, 0.40 reflection and 0.60 transmission at 0.532 micron wavelength, 45 degree incidence) and long-pass dichroic mirror (CD, maximum reflectivity at 0.53 micron wavelength and maximum transmission at 0.60 micron wavelength, 45 degree incidence). The beams are then focused with lens L1 and recollimated with a magnification of 1 by an identical lens (L2). The pump and Stokes beams are blocked with a Shott color filter (BG-12, blue-violet glass, red and green absorbing); their reflections are absorbed by beam stops (which are not indicated in figure 1). The anti-Stokes CARS signal is diverted with a mirror tandem (M0, M1) to the condensing lens (CL), which matches the aperture of the 1/4-meter monochromator (Jobin-Yvon model M25M, dispersion $\sim 10 \text{ \AA/mm}$). A vidicon two-dimensional array (SIT, PAR Model 1254) fills the exit aperture of the monochromator, providing for multichannel detection.

Variations in CARS data on static hydrogen samples and hydrogen-containing flames pointed to the necessity of a simultaneous normalization procedure. A parallel reference system has been implemented in the apparatus to increase the accuracy of temperature determination (ref 8). A broadband beamsplitter (BS, 0.70 transmission, 0.30 transmission at both 0.532-micron wavelength and at wavelengths to the red) is inserted into the optical path. The pump and Stokes beam are turned with an equilateral prism (P, anti-reflection coated) to the reference sampling region. The beams are focused with lens L3 and recollimated by an identical lens (L4). The resonant CARS signal generated from a heated cell (Perkin-Elmer model 910952, temperature to 475 K) of hydrogen gas is diverted to the condensing lens via the mirror combination (M2, M3, M1). Mirror M0 in the sample leg is, of course, positioned for free transmission of the reference beam.

Alignment of the parallel reference system is surprisingly easy for colinear CARS, once the reference leg components are approximately in place. The colinear configuration of pump and Stokes beams is first aligned in near field (at CD) and then in far field (at L3) using the naked eye (with the lasers both safely attenuated). A hydrogen gas cell is then placed in each section for the next step. Fine tuning of the mirrors DM and CD is accomplished by optimizing the CARS intensity, observed by photomultiplier, in both the reference and sample sections. The Stokes laser is tuned (first order, 0.683 micron output, bandwidth $\sim 10 \text{ cm}^{-1}$) to the $J = 1$ resonance of the hydrogen Q-branch; and the CARS signal, now visible, is brought to the slit of the 1/4-meter monochromator by adjusting

mirrors MO-M4 and the condensing lens. Broadband operation is then resumed; fine tuning of the detection optics is minimized with this procedure. Reference cell and test section CARS signals are spatially separated on the monochromator slit, and hence on the vidicon face, by appropriate displacement of the mirrors. Flames tend to realign the CARS beam in the test section; therefore, a small adjustment of both transmission and detection optics is often necessary. In preparation for optical bomb studies of propellant combustion, a small hydrogen diffusion flame is inserted into the center of the chamber for realignment purposes.

Detection of CARS; Data Acquisition Logic

The reference cell and test section signals are, as stated above, brought to the slit of the monochromator, dispersed, and presented to a vidicon array for multichannel detection. The two-dimensional scanning capability of the OMA controller (PAR model 1216) is utilized to concurrently detect the two spatially separated beams. The OMA console (PAR model 1215) provides for an externally triggered data acquisition format which is exclusively used here. Data are recorded either in the memory of the 1215 console or in digital storage of the 1216 controller video-rate output with a Nicolet 4904 digital oscilloscope. Reference and test video signals are output in serial fashion for digital recording.

The data acquisition logic module of the electro-optical system ensures that initiation of multichannel scans is synchronized with the single laser shot of interest and that signals from previous laser pulses do not interfere with the measurement⁴. An electromechanical shutter (Uniblitz model 100-2B) is placed in front of the monochromator slit; it is opened slightly before the diagnostic pulse and remains open well over the pulse duration. Initiation of the vidicon scan then commences with the laser pulse. For external firing experiments, a trigger signal for this sequence is derived directly from the pulse generator (to 1 sec) in figure 2. Logic hardware, as shown in figure 4, has been developed for single-pulse extraction measurements with the laser running at 10 Hz. In this case, the previous laser pulse initiates the shutter opening and vidicon scanning sequence. Reliable results are obtained from both of these approaches.

Test Procedures

Preliminary evaluation of CARS thermometry in static hydrogen cell, hydrogen diffusion flames, surface-mixed hydrogen-oxygen torches (Bethlehem apparatus model PM-2C) and LOVA/nitramine propellant combustion (burning unconfined in air and ignited by match) has proceeded with both the unreferenced and referenced

⁴ The time constant of vidicon detection is long, as the photo-activated diodes hold charge for several scans of the recording electron beam. High-voltage gating of the beam can lower this time constant (while tending to affect response linearity); fast scanning of short vidicon sections can deplete the diodes at a quicker rate.

apparatus. The flames are burned directly in the dehumidified experimental area; LOVA combustion products are properly vented away from the area via a duct and blower.

Propellant combustion under a pressure of inert gas is accomplished with an optical bomb (Atlantic Research Corporation model 105) and sealed bomb head assembly. This optical bomb affords an aperture of 1.7 inches and an optical path length of 3.6 inches. The body of the bomb consists of the main body and two window assemblies. The main body is fabricated of 321 stainless steel. The window assemblies are sealed by leather-backed O-rings, and provide for a test pressure rating of 5,000 psig. Separate connections for gas inlets and outlets provide efficient purging of combustion products from the bomb cell. The window holder is fabricated from 304 stainless steel, and each window glass (Pyrex, 2 in. thick) is held tightly with O-rings and the internal pressure of the bomb. Each window assembly has a gas inlet for flushing solids from the area of the glass during the burn cycle. The bomb head, which suspends the sample in the middle of the bomb, is fabricated from type 347 stainless steel. Electrical feedthroughs provide for ignition of the propellant via nichrome wire or electric match. All parts directly in contact with the propellant are 304 stainless steel. The window assemblies and the bomb head are designed for easy removal from the bomb with a spanner wrench.

A high pressure gas handling system provides for considerable flow rates of the background gas. A single valve controls the inlet to a manifold which supplies three gas inlets on the bomb: two for each window purge and a large inlet at the bottom of the bomb body. Combustion products are vented through a large outlet at the top of the bomb body. A blower and duct carry these products away from the experiment area.

The bomb window assemblies have been modified to seat recessed, thinner windows with a smaller aperture. This modification increases the optical path length inside the bomb and protects the window glass from combustion products, thermal transfer, and laser damage. Working pressures can be as high as 800 psig. A high pressure fiber optic feedthrough, to guide laser radiation for ignition under pressure, is now at the design stage.

LOVA/nitramine propellant, ignited sympathetically with the flame of a NOSOL propellant grain (nichrome hotwired) was evaluated with the use of an unmodified bomb at moderate pressures (200 to 300 psig) of nitrogen, argon, and helium background gas. The flowfield was somewhat smokey; the burning rate was fast; and spatter of combustion products was observed on the windows under these conditions. Attempts at propellant combustion under modest pressures of oxygen (15 psig) led to catastrophic failure of the inlet window. An expansive flame, emitting a continuum of blue radiation much as a hydrogen-oxygen torch, was observed. Preliminary operation with the modified bomb at modest pressures of nitrogen (20-40 psig) reveals a more smokey flowfield for LOVA/nitramine, effectively opaque to the CARS approach. Tests with propellant JA-2 at similar pressure have shown a clearer flowfield, more amenable to the technique.

RESULTS AND DISCUSSION

Triggerable, Single-Shot CARS Spectrometry

Initial tests of the single-shot circuitry shown in figure 2 were completed on room temperature hydrogen gas contained in a cell at 19 psig and on a diffusion flame of hydrogen in air.

The cell studies measured the integrated CARS signal as a function of a preset "LAMP" delay--accomplished by dialing in various delays in the Digital Delay Generator (msec). The anti-Stokes signal passing through a Mercury line filter (transmission maximum 0.4358 micron, bandwidth of 0.0010 micron) was detected by a fast photomultiplier and recorded in a single sweep of a Tektronics oscilloscope (model 7104). The shot-to-shot variabilities of the pump Nd:YAG and Stokes dye lasers were concurrently detected by a fast photomultiplier terminated in the 1 megohm input of a Nicolet digital oscilloscope for recording. Two important results emerge from the data in table 1: the laser can be reliably triggered at "LAMP" delays greater than 300 milliseconds; and the propagation of error in the hydrogen CARS experiment must include beam parameters other than P_{green} and P_{red} . The primary result is encouraging, as it establishes a generous window in time within which an external event can be placed for study⁵. The second result, pointing to the large shot-to-shot errors in absolute intensity measurements with an unreferenced apparatus, is consistent with the findings of other groups (ref 8) using a similar system at 10 Hz, with data acquisition in single-pulse extraction mode. The agreement in the crudely normalized data ($P_{\text{CARS}}/P_{\text{green}}^3$) over the delay period studied is encouraging and points to the promise of a resonant reference for more accurate concentration measurements and thermometry.

Unreferenced single-shot spectra of a hydrogen diffusion flame were detected and recorded with the vidicon OMA system. With the use of a simple Boltzmann analysis, the Stokes dye laser contour and the ratio of the $J = 1$ line intensity to the $J = 3$ intensity, a temperature determination was carried out. A single "LAMP" delay of 421 milliseconds was selected, and a set of five shots was taken. The measured temperature was 870 K (± 120 K), which is modest for a flame but is consistent with the spatial resolution of the collinear CARS experiment (>1 cm with L1 having a focal length of 8 inches) and the spatial extent of the flame (<1 cm). The deviation of the ratios is of the order of 40%, as expected, but the shape of the Boltzmann curve in this temperature domain lowers the error in temperature measurement to that observed.

⁵ Later investigations show that the time lag of the pulse generator is 180 milliseconds, contributing to the negative result at short "LAMP" delays.

Basic Advances in Hydrogen CARS Spectrometry

The implementation of a parallel reference system has improved the precision of temperature measurement, and observations of higher rovibronic lines in the hydrogen Q-branch have confirmed the spectroscopic analysis used in data reduction (refs 10 and 11).

Single-pulse extraction measurements with the parallel, resonant reference system have proceeded with the apparatus detailed in figure 1. Encouraging results have been obtained with an uncomplicated Boltzmann analysis for the independent ratios of peak areas associated with the $J = 1$, $J = 2$, and $J = 3$ Q-branch transitions. Test samples include room temperature hydrogen gas, heated hydrogen gas (to 465 K), hydrogen diffusion flame, hydrogen-oxygen torch and LOVA/nitramine propellant combustion. Representative spectra are shown in figure 5; the test section hydrogen CARS (diffusion flame) is shown in the first scan and the reference cell (460 K hydrogen) is shown in the second scan. Quantitative analysis indicates the possibility of systematic errors in the apparatus due to a number of experimental factors, such as CARS signal misalignment and toroidal-beam aging; channel crosstalk and saturation of the vidicon array; Stokes dye laser divergence and axial stimulated emission (ASE); and pump Nd:YAG laser mode structure. Other CARS workers have noted unequilibrated flame spectra in this hydrogen Q-branch region to which our determination may be especially subjected. We have observed apparent non-equilibrium distributions, via CARS, in both hydrogen diffusion flame and hydrogen-oxygen torch. Confirmation of these results may emerge in the coming months with the implementation of a single-mode Nd:YAG laser (ref 12) and further experimental refinements.

Multiple-shot averaged spectra of hydrogen-oxygen torch and methane-nitrous oxide flames have revealed higher rovibronic levels of the Q-branch. At flame temperatures of 2,800 K in the methane-nitrous oxide flame, lines were observed for $J < 12$ in the $v = 0$ band (ground state) and for $J < 10$ in the $v = 1$ band (excited state). Line positions compare favorably ($\pm 2 \text{ cm}^{-1}$) with spectroscopic constants derived from spectral observations in hydrogen discharge (ref 10) and from the ab initio results of Ermler⁶.

CARS Approach to High Pressure Propellant Combustion

The focus of these studies is a characterization of LOVA/nitramine propellant, burning under a high pressure of inert gas. These conditions essentially mimic the ballistic environment within which the propellant is to perform. Results of post-flame temperature and flame dynamics hence may give some insight into the quality of propellant performance without actual gun test. The experimental complexity of the CARS approach to propellant combustion under pressure is manifold, including factors such as the grain burning rate (R), flame deflection

⁶ $B_e = 60.7922$, $D_e = 0.0448$, $\omega_e = 4400.39$, $\alpha_e = -3.0320$. Further details are available in an ARDC technical report published in December 1983 (ref 7).

of the transmitted beams, laser-induced breakdown in the flame zone at such elevated pressures, laser-induced breakdown of particles in the flowfield, and the effect of background gas on the burning rate and CARS efficiency.

The burning rate of propellants is empirically related to ambient pressure by a positive exponential (i.e., $R = A \exp(+KP)$). The time then available for spectroscopic study is short at elevated pressures, and the propagation of the grain away from the CARS interaction zone prohibits any signal averaging. The application of a steady-state strand burner, which would advance a longer grain at its burning rate, is the best of approaches (unfortunately we are limited to a fixed burner configuration).

In our preliminary experiments, deflection of the CARS signal is caused by the refractive index gradients associated with flame structures. Work with the bomb follows the procedure of first aligning the system using a hydrogen diffusion flame placed in the center of the bomb; then placing the propellant and ignition mechanism on the bomb head, sealing and pressurizing the vessel, igniting the grain, and then taking spectroscopic measurements. It is hoped in this fashion to avoid any deflection of the signal to be observed.

Dielectric breakdown within the flame zone has been observed for nearly all of our studies due to the particle-laden flowfield of the nitramine propellant (ref 13) burning under a moderate pressure of inert gas and the high laser fluences employed. This factor is a major reason for the infrequent successes that we have had in these attempts; amendment of our experimental procedures to increase the flow rate through the pressure cell may remedy this problem somewhat. Spectra of nitrogen product gas from combustion were obtained after a burn in helium background gas.

Exploration of background gases for the high pressure experiments has led to some interesting observations. First, there appears to be a diffusion of gas to the flame zone which affects the transfer of heat to the burning grain and, hence, the burning rate of the propellant. Employment of helium gas lead to an accelerated burning rate in comparison with nitrogen combustion. Laser-induced breakdown in the background gas is observed at all pressures to 300 psig with argon, and to a lesser extent with helium. Effective target/background combinations of gases for CARS spectroscopy have been identified: hydrogen target/nitrogen background and nitrogen target/helium background. Experimentation with lower pressures of oxygen gas as background have shown a dramatic change in the flame of the propellant, as one would expect.

Within the foreseeable future, a project will be initiated to study a propellant with a cleaner flowfield, JA-2 and other nitramine formulations (ref 13), and to move to progressively higher pressures and higher background gas flow rates. Concurrent development of apparatus and propellant experimentation should merge effectively in the measurement of flame temperatures and the definition of working procedures.

Table 1. Shot-to-shot variability of coherent powers critical to CARS generation, as a function of "LAMP" delay time. Each number represents 25 single-shot measurements; the number in parentheses is the standard deviation of the set. The relative error (standard deviation/quantity) is expressed as a percentage following the slash. P_{GR} is the Nd:YAG second harmonic generation power; P_{RED} is that of the dye laser.

"LAMP" DELAY	P_{GR} (REL.)	P_{CARS} (REL.)	P_{CARS}/P_{GR}^3 (REL.)	P_{RED} (REL.)
221 MS. 62 NS. JITTER 64% SUCCESS RATE	166(74) /45%	---	---	---
321 MS. 28 NS. JITTER 100% SUCCESS RATE	197(13) /7%	5.7(1.8) /32%	1.15(0.20) /18%	4.7(0.3) /7%
421 MS. 40 NS. JITTER 100% SUCCESS RATE	177(88) /5%	3.5(0.9) /26%	1.21(0.23) /19%	---
521 NS. JITTER 48 NS. JITTER 100% SUCCESS RATE	157(07) /5%	2.7(0.9) /33%	1.10(0.22) /22%	---

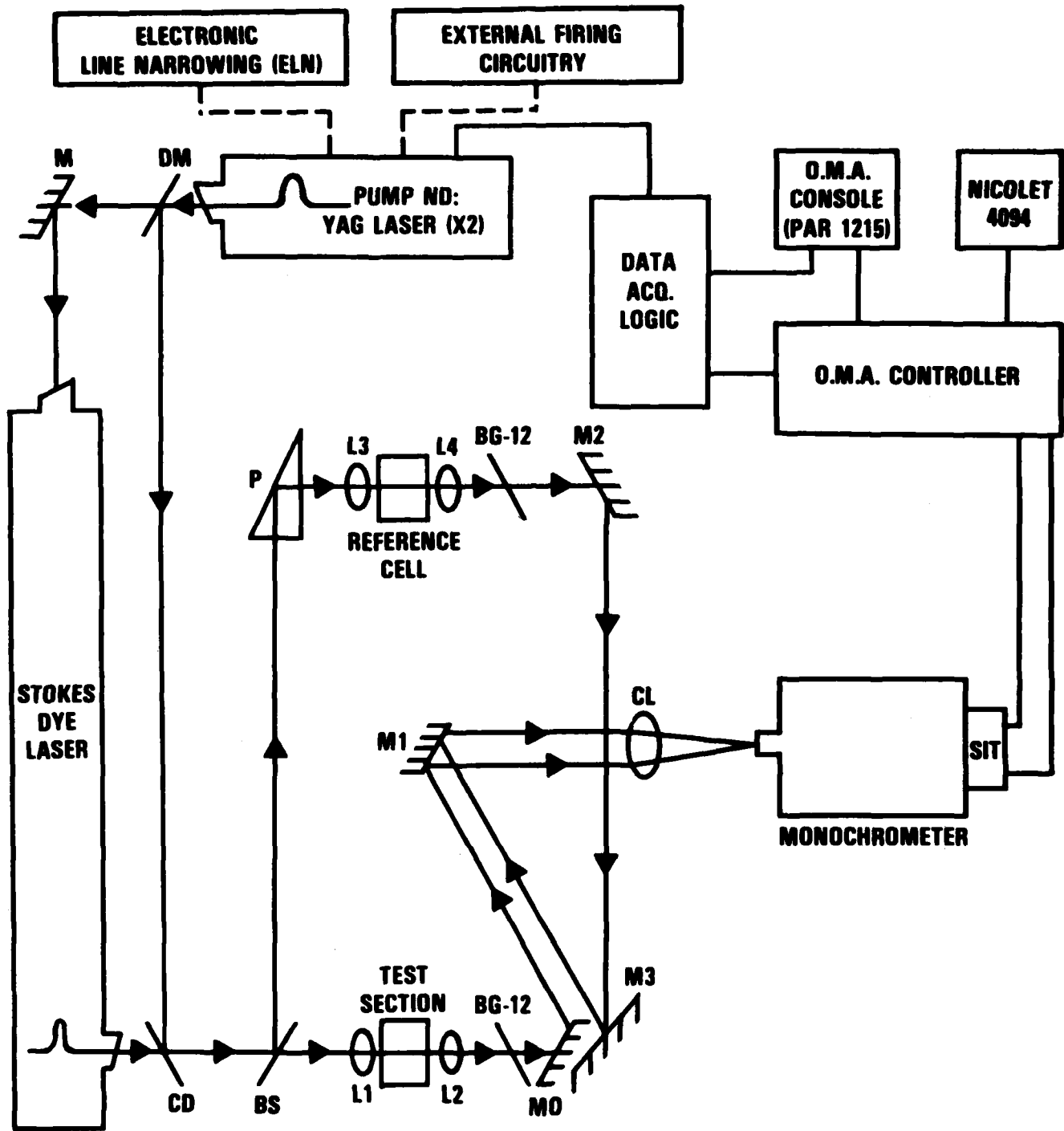


Figure 1. Electro-optical system for referenced CARS

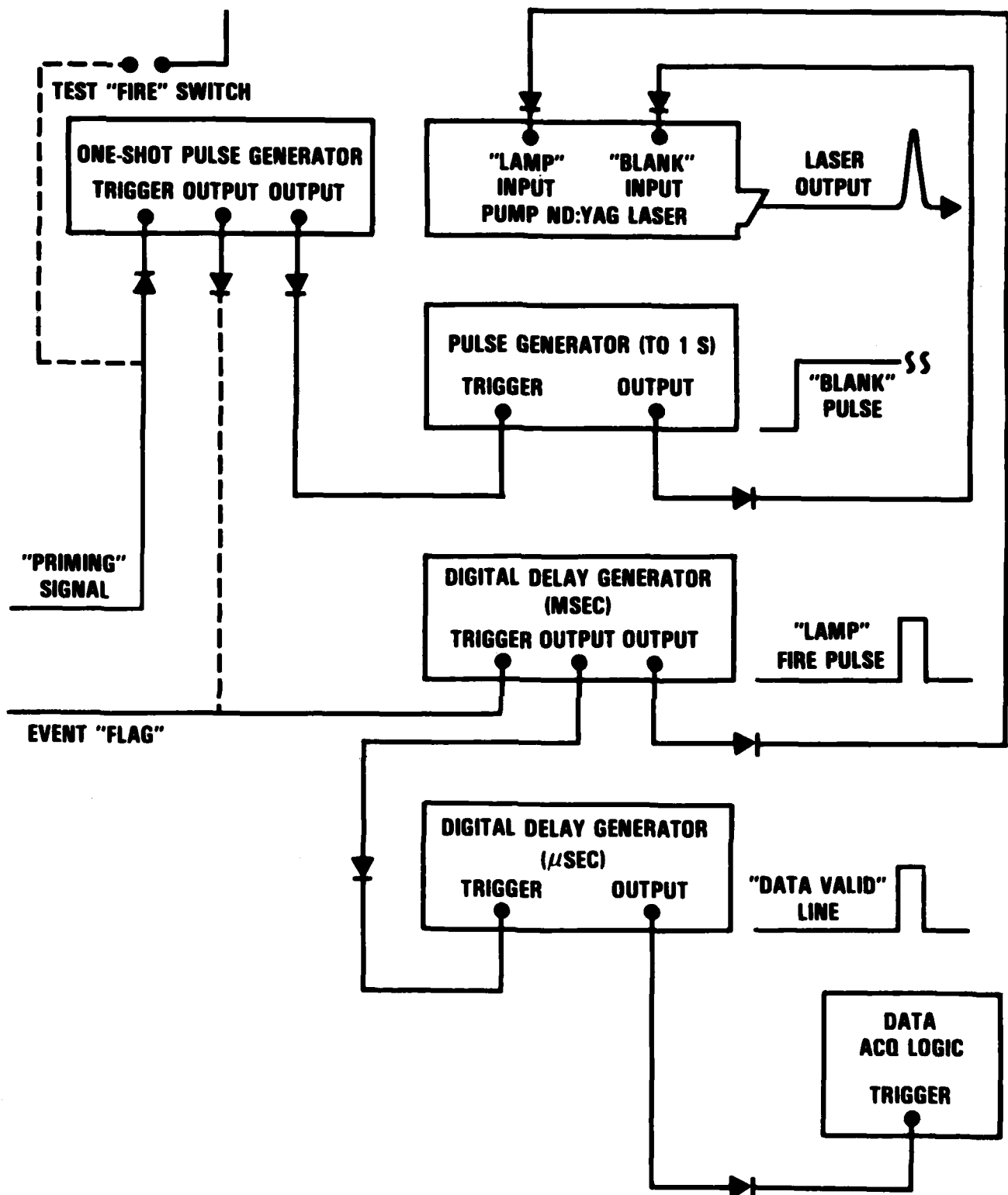


Figure 2. Single-shot circuitry for CARS spectrometry. Test circuit is indicated by dashed line (- -). Laser output occurs 200 μ sec after "LAMP" fire pulse.

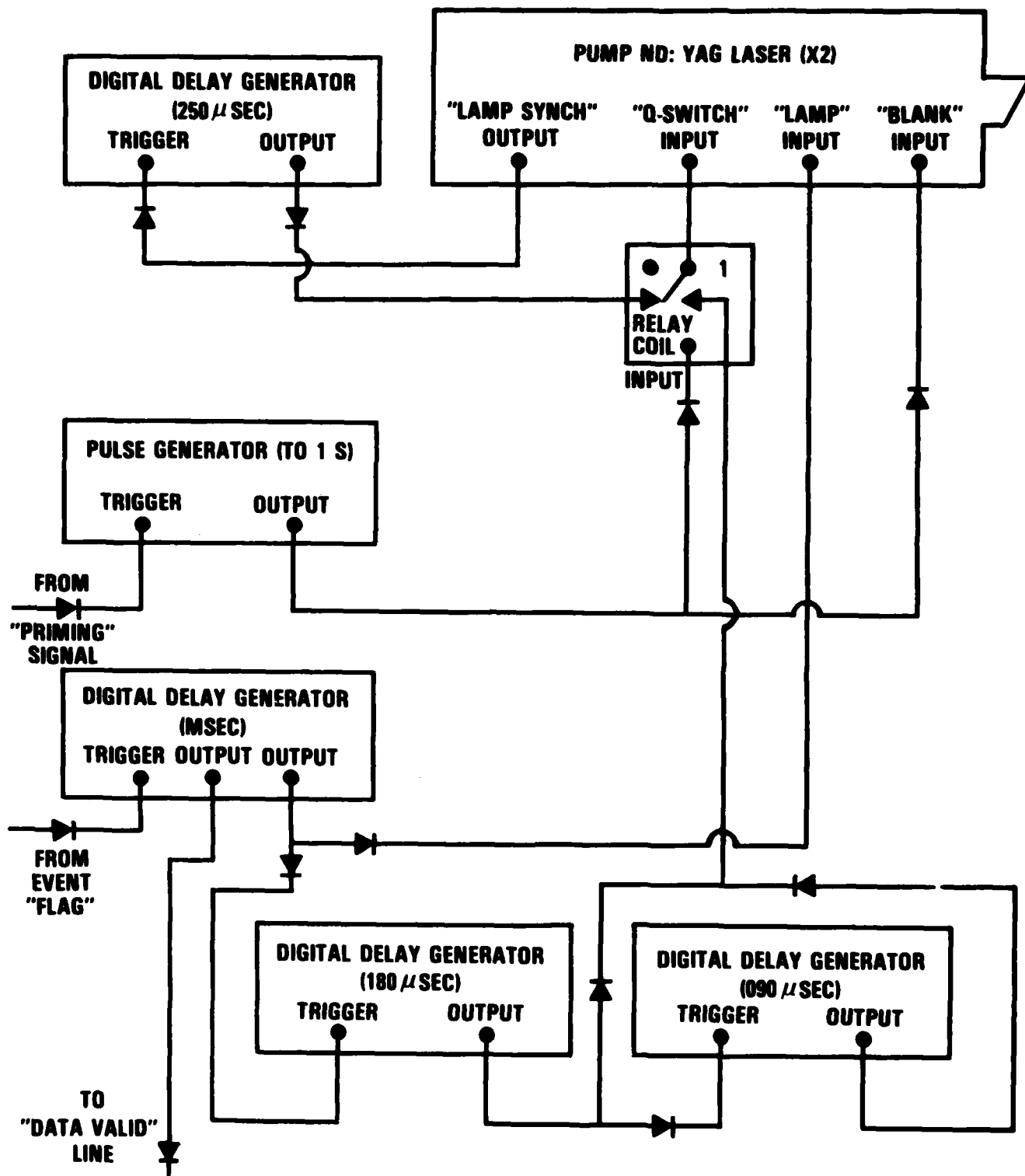


Figure 3. Double-pulse extraction hardware. The 250 μsec delay maintains the laser and SHG crystals for subsequent firing (laser setting 10 Hz repetition rate, external Q-switch mode).

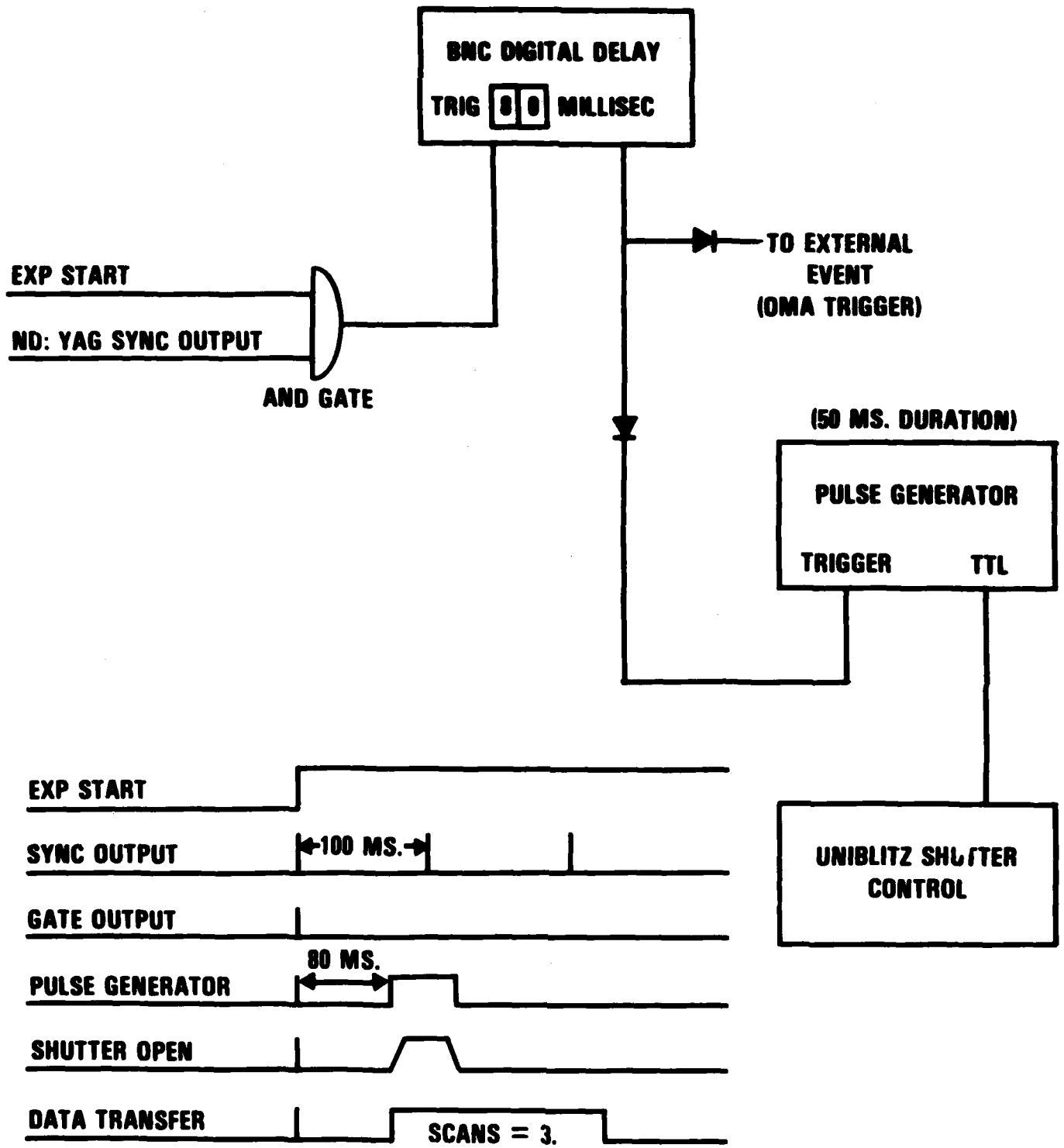


Figure 4. Timing diagram

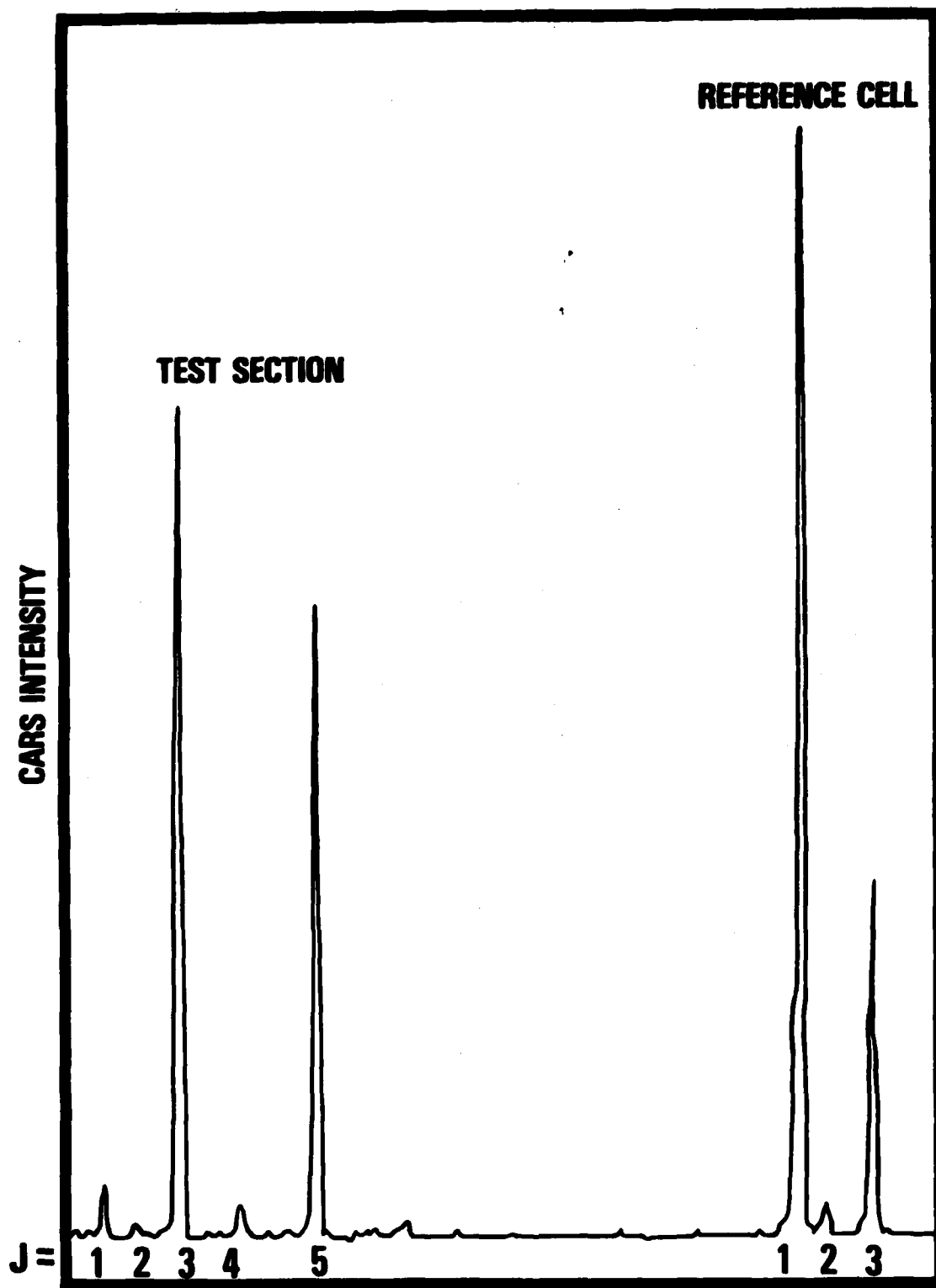


Figure 5. Referenced CARS spectra. Hydrogen Q-branch.

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