AD-A183 907 TIME-RESOLVED ELECTRON DIFFRACTION(U) ARKANSAS UNIV 1/1
FAYETTEVILLE DEPT OF CHEMISTRY AND BIOCHEMISTRY
R S MILLER 1987 N00014-85-K-0757

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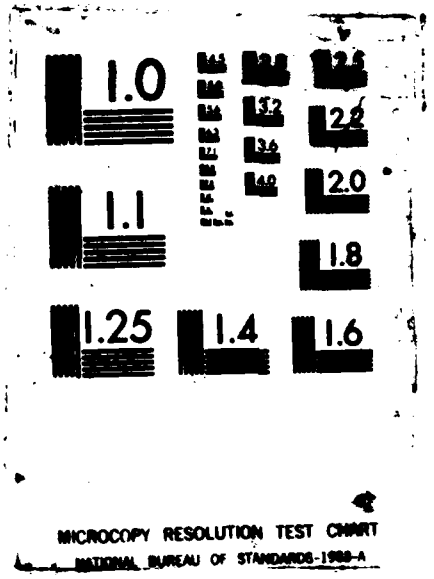
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Arkansas Univ. Chemistry + Biochemistry

ONR CONTRACT INFORMATION

Contract Title: Time-Resolved Elec Diff

Contract Number: N00014-85-K-0757

Work Unit Number: 4327-804

Scientific Officer: R.S. Miller

Annual technical rept.

Enclosure (1)

a. Description of Scientific Research Goals.

It is the goal of this research to construct an instrument for Time-Resolved Electron Diffraction (TRED) studies of the structures of short-lived, energetic molecular states and of the dynamics of primary dissociation processes found in molecules of energetic materials.

The TRED experiments will be performed by focusing the 193 nm output of a pulsed ArF excimer laser (pulse duration 10-20 nanoseconds) onto the cold photocathode of a high voltage electron gun, where electron pulses will be generated by field-assisted photoelectric emission. These electron pulses will be scattered off the gas-phase molecules in an effusive molecular beam after some fraction of the beam population has been excited by a pulse from a XeCl excimer-pumped dye laser. Electron diffraction intensities will be recorded using the real-time gas electron diffraction (GED) technology developed at the University of Arkansas[1,2]. By synchronizing the dye laser excitation pulse and the diagnostic electron pulse, intensity data can be obtained from molecules which all have the same age relative to the time of excitation. Thus, time-resolved observations of excited molecular states and of transient reaction products should be possible.

b. Significant Results of the Past Year.

b.1. Design and Construction of the Pulsed Photocathode.

Construction of the photocathode (PC) unit, begun last year, was completed. That is, PC vacuum chamber was assembled and put into operation; electron gun construction for space-charge limited pulsed electron emission was completed after thorough theoretical analysis; a second excimer laser was installed, tested, and the first excimer laser overhauled (converted to 3-phase electrical power with an improved thyratron); laser synchronization mode was selected and the synchronizer obtained; PC chamber was HeNe laser-aligned with the ArF excimer and the optical system for delivery of the 193 nm laser light onto PC emitting surface was installed; PC vacuum system was successfully tested and the first photoemitted pulsed electron beam was obtained.

Preliminary results indicate 1) that space-charge limited electron pulses can be easily obtained by our procedure; 2) that the pulsed electron beam can be readily focused with a magnetic lens to a diameter suitable for GED experiments; and 3) that the duration of the electron pulses is comparable to that of the ArF excimer-pump laser pulses.

b.2 Attempts to Improve Angular Detection Range and Detector Sensitivity.

To improve the existing detection mode, as described last year, a fiber optic detector system was designed and constructed. The system consists of a vacuum fiber window, a fiber coupler (1:1 or 2:1 imaging), and a Tracor Northern fiber optic covered photodiode array. The new detector was interfaced to an IBM personal computer, and its characteristics are currently being tested.

A considerable part of the funds needed for the ongoing detector improvement research is covered by an NSF grant ($115,000, starting date 4/1/87) which supports attempts to employ our real-time GED data recording technique as a detector for gas chromatography. High detector sensitivity is needed for this application as for the TRED studies in preparation.

b.3 GED Studies of the Products of Pyrolytic Reactions.

During the last year the first successful identification was reported [3] of photochemical reaction products observed by using our procedures. In these experiments, cis-1,2-dichloroethene (CDCE) and trans-1,2-dichloroethene (TDCE) were irradiated with 193 nm excimer laser light shortly before their GED patterns were recorded. TDCE was found in the diffraction pattern of irradiated CDCE, and CDCE was found in the pattern of irradiated TDCE. Experiments of this kind, albeit time-unresolved and involving stable molecular species, are directly useful for the TRED studies in preparation, because they provide us with the oportunity to gather experience in the analysis of GED data of mixtures containing several components.

In a variation of these experiments, a CO_2 laser (on loan from NRL) was restored. A special quartz inlet system was built for the electron diffraction instrument (in collaboration with W.Braun at NBS). A laser delivery system (contained beam) was designed and installed by which the CO_2 laser output can be focused onto the tip of the GED inlet system, thus heating a small area to temperatures of up to 1500K. In the diffraction chamber, the heated tip is less than 1 mm away from the electron beam. Thus, GED patterns can be obtained of gases immediately after they have traversed the high temperature region, and pyrolysis products can be observed before they can undergo collision-induced secondary reactions. First experiments were performed with CF_3I heated in this way. Temperature dependent changes in the diffraction intensities are clearly visible. Attempts at quantitative analysis are underway.

c. Plans for Next Year's Research.

During the next year we plan to continue the project by taking as many of the following steps as time will allow:

1.)Build a collimating and focusing system for the PC pulsed electron beam. Characterize the electron pulses (current and duration), measure laser and electron pulse temporal correlation, and determine correlation between irradiation intensity and photoemission intensity. Design and construct a gas inlet system for the PC-connected diffraction chamber, and expand the existing pumping system of the latter to make gas-phase experiments possible. Record first GED patterns with the new PC beam (solid state and gas phase samples). Construct the dye laser delivery system, implement and test the prepared excimer laser synchronization procedure, and initiate first studies of transient species.

2.)Develop data acquisition software for the fiber optic detector system, integrate the new detector with a diffraction chamber, and perform test GED experiments.

3.)Perform pyrolysis experiments using the CO_2 laser-heated inlet system in attempts to record GED data of energetic and reactive molecular species.

References:1.J.D.Ewbank,L.Schafer,D.W.Paul,O.J.Benston,and J.C. Lennox, Rev.Sci.Instrum.,55(1984)1598. 2.J.D.Ewbank,L.Schafer, D.W.Paul,D.L.Monts,and W.L.Faust, Rev.Sci.Instrum.,57(1986)967. 3.D.L.Monts,J.D.Ewbank,K.Siam,W.L.Faust,D.W.Paul,and L.Schafer, Appl.Spectrosc.,41(1987)631

List of Publications/Reports/Presentations

1. Papers Published in Refereed Journals:

1. D.L.Monts, J.D.Ewbank, K.Siam, W.L.Faust, D.W.Paul, and L. Schafer, "Gas Electron Diffraction Study of the 193 nm Laser-Induced Interconversion Between cis- and trans-1,2-dichloro ethene", Applied Spectroc., **41** (1987) 631.

2. Technical Reports: none.

3. Presentations:
a.Invited:

L.Schafer: "Structural Studies by Real-Time Gas Electron Diffraction and Ab Initio Calculations", University of Lille, France, March 1987.

John D. Ewbank, W.L.Faust, D.L.Monts, D.Paul, and Lothar Schafer:"Experiments to Exploit Real-Time Gas Electron Diffraction in Structural Studies of Transient Molecular Species and Time-Resolved Kinetics", Energetic Material Initiation Fundamentals Workshop, Los Alamos National Lab, Oct.1986.

b.Contributed:

D.L.Monts, J.D.Ewbank, K.Siam, D.W.Paul, L. Schafer, and W.L.Faust: "Real-Time Gas Electron Diffraction Studies of Laser-Transformed Species", 193rd ACS National Meeting, Denver, Col, April 1987.

4. Books: none

Enclosure (2)

LIST OF AWARDS

Name of Person Receiving Award.	Institution	Name of Award	Sponsor

none

Enclosure (3)

h. List of Participants.

At the University of Arkansas:

1. Dr. John D. Ewbank, promoted to tenure-track Assistant Professor of Chemistry, as of August 1987.
2. Dr. David L. Monts, Assistant Professor.
3. Dr. David W. Paul, Associate Professor.
4. Dr. Lothar Schafer, Professor.
5. Dr. Khamis Siam; postdoctoral research assistant.
6. Susan Kuip, graduate student
7. Stephen Bowling, undergraduate research participant

At NRL:

Dr. W. L. Faust, Optics Division.

i.Other Sponsored Research:

1. L. Schafer and D.L. Monts, "Picosecond Time-Resolved Electron Diffraction Studies of Unimolecular Decomposition", DoD-URIP, 8/1/84 to 8/31/86, $257,000.

2. D.L. Monts and L. Schafer, "Time-Resolved Studies of Transient Molecular Species by Synchronously Pulsed Optical Excitation and Diagnostic Electron Scattering", ACS-PRF, 7/1/86 to 8/31/88, $35,000.

3. D.L. Monts, "Investigation of the Use of Heavy Bridging Atoms as Energy Flow Blockers for Laser-Selective Chemistry", Arkansas Science and Technology Authority, 5/15/86 to 9/14/87, $14,898.

4. D. Paul, "Refinement of a Transferase Analyzer", NIH-BRSG, 11/15/85 - 10/31/86, $4,200.

5. D. Paul, "A Fundamental Study of the Reaction Between Organic Halides and Lithium Metal", Research Corporation, 11/1/82 - undefined, $9,300.

6. L. Schafer, J.D. Ewbank, D.W. Paul, and D.L. Monts, "Research Toward the Application of Real-Time Gas Electron Diffraction as a Detector for Gas Chromatography", NSF, $115,504, 4/1/87 -9/30/89.

7. D.L.Monts and L.Schafer, "ACS-PRF Summer Research Fellowship", ACS-PRF, 5/15/87-8/31/87, $2000.

Proposals Submitted: none.

PUBLICATIONS/PATENTS/PRESENTATIONS/HONORS REPORT
(Number Only)

Papers Submitted to Refereed Journals (and not yet published): none

Papers Published in Referenced Journals: one

Books (and sections thereof) Submitted for Publication: none

Books (and sections thereof) Published: none

Patents Filed: none

Patents Granted: none

Invited Presentations at Topical or Scientific/Technical Society Conferences: one

Contributed Presentations at Topical or Scientific/Technical Society Conferences: one

Honors/Awards/Prizes: none

Enclosure (4)

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