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6. **Project Period:** 1 February 1986 - 31 July 1989
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9. Introduction

The principal aim of this program has been to study the electronic structure of insulating liquids of biological interest over a broad energy range from 0 to 30 eV. The studies basically consist of measuring the reflectance, transmittance, photoemission and photoionization of dielectric liquids in the vacuum ultraviolet spectral region as a function of angle of incidence and energy. These in turn may be interpreted in terms of the electronic structure of each liquid as it is excited by the passage of a charged particle. Optical data provides indirect evidence that collective effects occur in liquids. Direct observation of their existence is substantiated in studies of the energy distribution of electrons specularly scattered from the liquid surface.

The biological and chemical response of a system to incident radiation is partly determined by interactions with low-energy electrons. Energy losses in

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biologically related liquids are currently not fully understood. Critical parameters such as the photoemission yield and electron mean free path are needed for incorporation in radiation models. Our experimental data on the dielectric response of a medium, calculated stopping powers and low-energy electron attenuation lengths can provide the parameters used in electron transport calculations as well as in determining the appropriate photoelectric threshold of organic liquids. Data on the interaction of low-energy electrons in condensed matter and the dielectric response of these media are particularly important in the study of the early physical stage of charged-particle interactions. With the fundamental parameters provided by our studies, track structure calculations may yield relations between the primary excitation and the resulting localized excitations which lead to chemical and ultimately to biological effects.

Various techniques are used to study the electronic structure of liquids below 30 eV, a region of the vacuum ultraviolet accessible with a Seya monochromator and gaseous discharge light source. Reflectance and transmission measurements are made using both closed-cell and windowless techniques. The open-dish reflectance method employs a double ion chamber which measures photon intensities before and after reflection by gas ionization. The system is modified to permit absolute photoionization measurements. These data are analyzed to yield optical and dielectric parameters, photon mean free paths and electron attenuation lengths. Photoyields from these data

and a simple model are compared with those experimentally observed.

An alternate method for looking at electron attenuation lengths is provided by the scanning tunneling microscope. The STM provides direct control over distance and energy scales in liquids not easily attainable by any other means. It has been shown to be operable even at atomic resolution under water and saline solutions.

A more comprehensive description of the program goals and outcomes has been detailed in the Eighteen Year Progress Report (DOE/ER/60415-2) and the Annual Report for 1 February 1987 to 31 January 1988 (DOE/ER/60415-3).

10. Significant Accomplishments During the Budget Period:

(1) Studies were conducted on the optical and photoemissive properties of two dielectric liquids. Considerable efforts have been devoted over the past several years to photoemission measurements on dielectric liquids. Analysis of optical photoyield data provides information on electron mean free paths in the liquid. However, discrepancies between electron mean free paths derived from observed photoyields and a theoretical model for photoemission and those calculated using a modified Born approximation or empirical equation has long been a concern. Surface charging problems which may account for anomalous yields were considered.

To address the surface charging effect, preliminary studies were made on charge elimination through introduction of discharge needles and

submerged conductors near the active region, and on the effects of ionic doping of low ion-mobility liquids. Heptadecane, which had previously shown anomalously low photocurrents, showed normal photoemission behavior with discharge needles, as reported earlier in this project period. To further study surface charging effects, measurements were refined using submerged conducting grids to produce surface discharge in the irradiated heptadecane to confirm the earlier preliminary studies on this liquid. During the current budget year, similar measurements were made on the hydrocarbon pentadecane and refined on the silicone oil trimethylpentaphenyltrisiloxane, DC705. Measurements were also made on squalane and squalene in the 16 to 20 eV region. With the conductors, we were able to confirm normal photoemission behavior in these nearly zero conductivity liquids. However, to date, questions remain concerning the electron attenuation lengths derived from this photoyield data.

(2) An alternate method proposed for looking at electron attenuation lengths was provided by scanning tunneling microscopy. Electron energies involved in the STM operation range from ten's of eV to MeV. During the current budget period, design and assemblage was completed on a data acquisition system for the scanning tunneling microscope and successful imaging of an electrode surface submerged in liquid was demonstrated.

However, while progress was made with electrode imaging using the STM, we were not successful in getting higher voltage biases with satisfactory

signal to noise ratios for data in the eV range. Attempts to remedy this by proper choice of electrode materials and alternate signal sampling techniques were unsuccessful. The first year of research in this area was primarily exploratory. The STM has been shown to be operable even at atomic resolution under water and saline solutions. A major effort will be required, however, in the study of current-distance relationships as a function of bias voltage before the feasibility of using the STM to determine electron-attenuation lengths is demonstrated.

(3) One paper was published during the fiscal period. A copy of "Dielectric Response of a Silicone Liquid Excited By VUV Radiation and Electron Scattering," Journal of Electron Spectroscopy and Related Phenomena, 48 (1989) 435-447, is included with this report. Calculations on the dielectric response of the silicone oil Dow-Corning DC705 and revisions in the analysis of the data constituted a major effort for this project period.

The frequency-dependent dielectric properties of the liquid silicone were determined by two methods. First, the optical properties were redetermined from literature data and from new measurements taken in the vacuum ultraviolet to about 25 eV. Second, electron energy loss data were obtained by energy analysis of scattered electrons produced by incident beams of electrons with energies between 35 and 300 eV. As stated in the journal article, it appears from the data that many-body as well as single particle effects appear to have a role in the properties of this insulating dielectric liquid. Further

studies of low-energy interactions of electrons with liquids will be required to ascertain the role of multiple scattering, charging, and other effects on the interpretation of the data.

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