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INSTRUMENTATION FOR TIME-RESOLVED ELECTRON DIFFRACTION

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Abstract We describe an apparatus for using electron diffraction as the probe technique in short-pulse time-resolved excite-and-probe experiments.

INTRODUCTION

Within recent years, a variety of experimental techniques have been developed to characterize short-lived states or species in material systems, to study time-resolved structural changes, and to measure rates of population change. Purely optical techniques now exist in wide variety, typically using pairs of light pulses derived from a single laser system. The first pulse of a pair is employed to place a material system in an excited electronic state, or otherwise to prepare it for study. The second, usually delivered through a variable time-of-flight delay line, is used to inquire subsequently about the instantaneous condition of the material. Visible or ir absorption are representative approaches.

A number of other time-resolved probe techniques, even with far poorer temporal resolution than optical methods, also provide valuable information of highly specific character in various respects. Examples which attest to this include 2D Fourier transform ESR spectroscopy,[1] neutron powder diffraction,[2] short-pulse x-ray diffraction,[3,4] flash EXAFS,[5] electron energy loss spectroscopy,[6] and low-energy electron diffraction.[7]

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PROBING WITH ELECTRON DIFFRACTION

Electron diffraction (ED) enables precise determination of gas-phase molecular structures, or of thin film polycrystalline or single crystal structural data. Recently we have developed a novel technique for recording ED intensities, replacing the customary photographic detector by a photodiode array (PDA).[8,9] In the new instrument, the scattering pattern is made visible on a phosphor screen. Light from the screen is imaged onto the surface of the PDA, which records, on-line and with very brief processing time, up to 1024 independent intensity points along a diameter of the diffraction pattern. Development of a fiber optic image transfer scheme has considerably enhanced detector sensitivity and data quality.[10,11]

The new ED procedure makes operationally feasible studies of transient molecular phenomena which never have been attempted with photographic ED. Specifically, it is possible to attempt time-resolved ED studies by combining on-line data recording with diffraction from pulsed electron beams. Our apparatus combines short-pulse optical excitation, pulsed-beam ED probing, and PDA detection. This may provide greatly improved signatures of various species for kinetic studies. It should yield highly specific information on the geometric structure of transient species. Conceivably we expect to witness structural changes as they proceed.

Ischenko et al.[12] have performed ED experiments with short electron pulses, but the structural results were rather indefinite due to detector problems. Recently Mourou et al.[13-15] employed electron pulses of a few ps duration to obtain diffraction patterns from thin films and from surfaces. Their experimental setup was based upon a modified streak camera system.

In order to develop a more versatile pulsed beam instrument, one specifically designed for time-resolved ED studies with our on-line ED procedure, we have constructed a pulsed electron gun using a photocathode driven by an excimer laser. It is the purpose of this paper to describe this new device.

Design of Apparatus for Time-Resolved Electron Diffraction

The system which we have constructed incorporates (1) a pulsed optical beam of variable frequency for electronic excitation of materials under study, (2) a synchronously pulsed source of 30 to 50 keV electrons in a space-charge limited (SCL) beam, (3) the necessary vacuum environment and sample-handling capabilities, and (4) detection and signal processing equipment. There are a number of engineering issues which impinge upon the design of this system.

Generation of the optical and electronic beams entails two excimer lasers, operated in tandem under control of a synchronizing circuit. Excimer 1 is employed to photoexcite molecular gases or polycrystalline films. It may be operated at 193 nm (with an ArF gas fill), at 248 nm (KrF), 308 nm (XeCl), or 351 nm (XeF). Any of these wavelengths may be delivered directly to a small volume of sample material within the path of the electron beam. Alternatively, the 308 nm output may be employed to drive a dye laser, for output tunable from 332 to ca 1000 nm.

The Electron Source

Excimer 2 is committed to generation of ca 100 mJ pulses at 193 nm (ArF). The intrinsically rectangular beam is directed through a splitting-and-recombining optical path to provide reasonably uniform illumination of a circular titanium photocathode within a Pierce-type electron gun. Other photocathode materials exist which might provide higher quantum efficiency and good response to less energetic photons, but these were considered chemically too delicate for the working environment. The requisite negative potential is introduced to the vacuum system through a 60 kV insulated feedthrough and a length of 50 ohm coax cable, acting as a charged line to provide a well defined driving impedance to the photocathode. The electron beam is further formed by a magnetic focusing lens and several apertures. A schematic representation of the electron source is given in Fig.1.

The choice of excimer lasers, which produce pulses of about 15 ns duration, clearly constrains the time resolution. Other laser types might provide shorter pulses. Our choice reflects an emphasis on the net rate of signal gathering and the flexibility of excitation wavelength.

High signal-gathering rate requires high electron fluence/pulse (current density x beam cross-sectional area x duration) and high repetition rate. To simplify certain interacting considerations,[16] the practicable current density is limited by space charge effects whereas a large beam area degrades the angular resolution of the diffraction pattern. Since as much as 20 A/cm^2 may be drawn through the 50 ohm driving impedance, a voltage drop of about 1 kV must occur. The intention is that the combination of rectangular current pulse and well defined driving impedance should prevent erratic excursions of the potential during the pulse and consequent degradation of the electron beam monochromaticity. The capacitance, C,of the photocathode assembly and the characteristic impedance R of the driving cable define a time constant RC. Fortunately this is 0.3 ns or less, so that the potential settles almost instantaneously; the fixed value is determined by calibration against a molecular standard.[8-11]

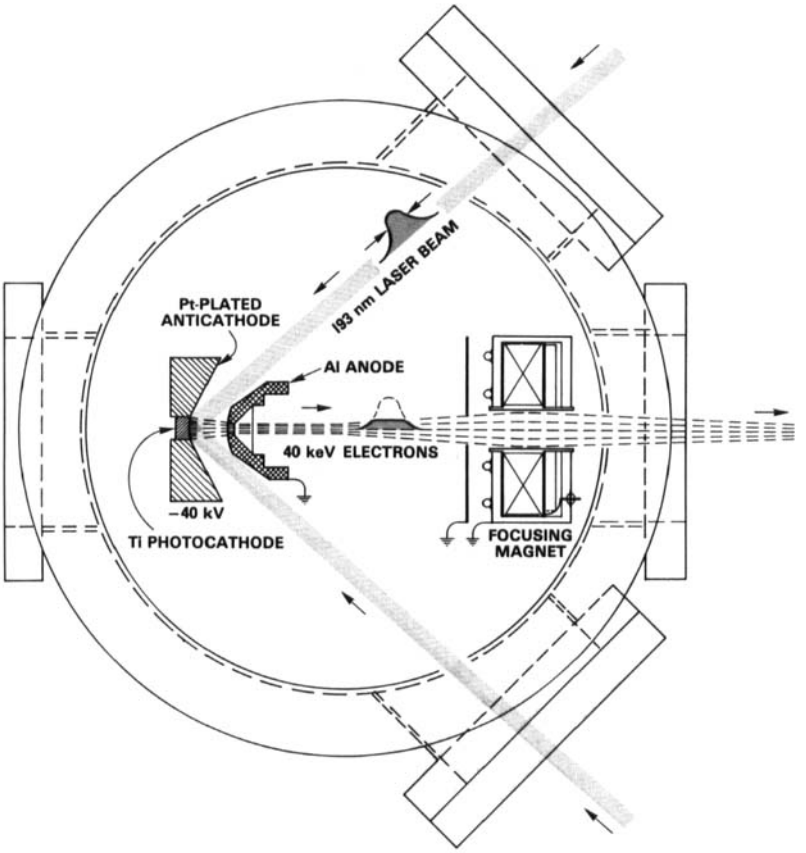

FIGURE 1 Pulsed electron source for diffraction studies of transient species.

First Trial Experiments with the Pulsed Electron Source

The very energetic photons from Excimer 2 drive the titanium photocathode with reasonable quantum efficiency. From direct measurements of current, we have estimated a value of 2×10^{-3}. The onset of SCL operation is found at approximately 1 mJ incident energy. Thus 100 mJ drives the gun well into the SCL region. Oscilloscope traces show that the shape of the current pulses is nearly rectangular. The laser pulses are approximately 15 ns FWHM; the electron pulses are somewhat longer.

As would be expected intuitively, the angular resolution of the ED pattern can be enhanced by use of a relatively small beam current. There exists a strategy to retain adequate signal-to-noise ratio by a compensatory high repetition rate and/or long runs of data gathering (unfortunately the required photolysis fluence/pulse remains fixed). There is, however, only disadvantage in deliberate

retreat from SCL current density. The ideal axially symmetric SCL beam exhibits laminar flow. It has, over any cross-section, uniform current density out to a definite edge where it drops abruptly to zero and the rays in any such section respect a common center of curvature. Any beam having these properties in some section is laminar and is an SCL beam everywhere. In its dependence upon axial position it resembles a lowest-order laser mode, first converging to a waist ($z = 0$, where the radius attains a minimal value r_w), then diverging in a symmetrical fashion.

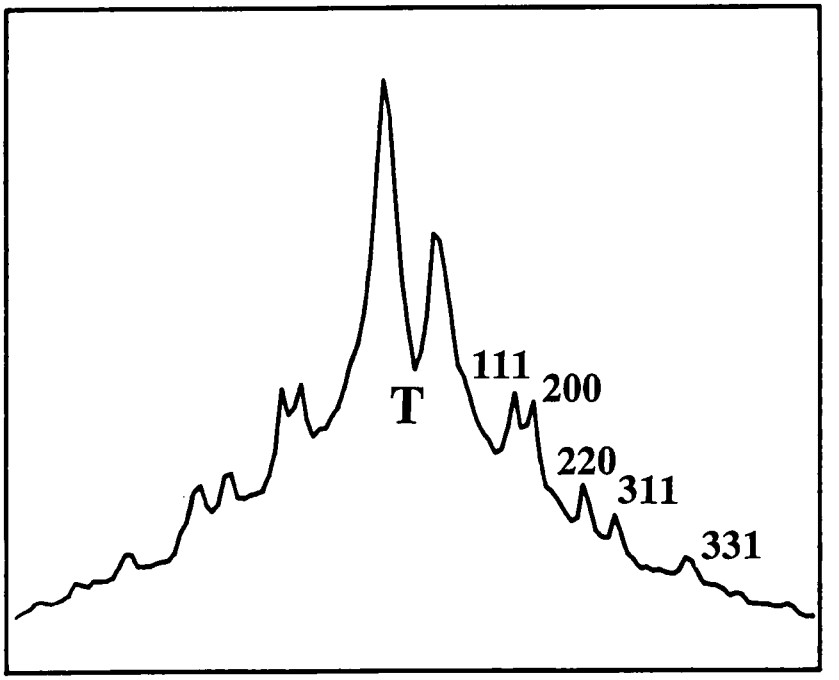

FIGURE 2 Diffraction pattern of polycrystalline gold obtained with a pulsed electron beam. Intensities were measured along a pattern diameter using the UoA real-time procedure.[8,9] (Beam trap at T.)

The quantitative theoretical treatment[16] expresses the issues in terms of the beam perveance P and a focal length fl(z_s). The perveance is the beam current divided by the 3/2 power of the beam potential. The perveance density is analogous to current density. The focal length is identified with the distance between the scattering plane (z_s) and the detector plane (z_d) in the experimental setup. Though there is some latitude (connected with the position of the scattering plane), choice of a specific fl value constrains the perveance density to a small range of values,

anywhere within the interval of reasonable setup configurations. Choice of a beam potential then fixes the current density, and the waist radius is the only free parameter. Because the flow is laminar, this radius can be fixed by an aperture stop anywhere over a broad range of axial positions. The signal-gathering rate then varies as r_w^2 and the space-charge limit to angular resolution varies as fl/r_w. This describes the expected conflict referred to in the prior paragraph.

Trial patterns, with a waist radius apertured to < 1 mm^2 were taken of polycrystalline gold. A sample picture is shown in Fig. 2. Preparations for time-resolved experiments, both with polycrystalline films and molecular gases, are underway in our laboratory and will be described elsewhere.

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