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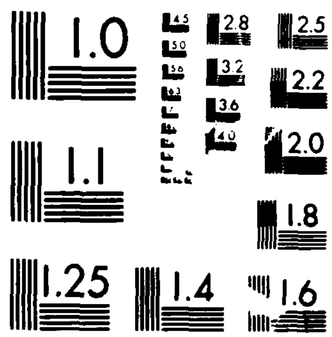

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The overall research program described in this report is to investigate the interaction of low (keV) and hyperthermal (10-100 eV) ions with clean and adsorbate-covered metal surfaces; in particular, ion-surface charge exchange processes. We report on the operation of the UHV scattering apparatus for the ion scattering measurements, ongoing development and upgrade of the apparatus, the use of two ion scattering simulations which together cover the entire energy range of interest, the further development of these simulations, and, most importantly, the ongoing experiments to determine charge transfer probabilities. The data we present in this report are representative of the scattering spectra we measure. Completed analysis of these and similar spectra will be presented in forthcoming papers. To date, we have measured spectra for Li^+, Na^+, K^+, He^+, Ne^+, and Ar^+ beams, ranging in energy from 50 eV to 4 keV, scattered from the <001> and <1-10> azimuths of Cu(110). Important features of these spectra have been identified with the scattering simulations.

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CHARGE EXCHANGE IN LOW ENERGY (keV) AND HYPERTHERMAL ENERGY (10-100 eV) ION SCATTERING

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I. SUMMARY

The overall research program described in this report is an investigation of the interactions of hyperthermal energy (10-100eV) and low energy (keV) ions with clean and adsorbate-covered metal surfaces: in particular, ion-surface charge exchange processes. This report covers research under grant AFOSR-86-0086, which was exclusively for personnel support. Instrumentation development for this research was, and continues to be, paid for in part by an AFOSR instrumentation grant (AFOSR-87-0048). A report on the overall research program must necessarily include both the scientific progress supported by this grant as well as instrumentation development under grant AFOSR-87-0048.

This report covers the following areas:

1) Completion and performance of the basic apparatus for low and hyperthermal energy ion scattering.
2) Ongoing development of apparatus for the project.
3) The development of simulation capabilities for hyperthermal and keV ion scattering.
4) Ongoing research on the fundamental mechanisms of charge transfer processes.

II. RESEARCH OBJECTIVES

The scientific objectives of the overall program are to investigate the interactions of hyperthermal energy ion beams (10-100eV incident energy) and low energy ion beams (keV energies) with clean and adsorbate-covered metal surfaces; in particular, to investigate ion-surface charge exchange processes.

Specific experimental goals are:

- To isolate the ion-surface charge exchange process as cleanly as possible, in order to probe the mechanisms of resonant charge exchange.
- To develop a model of ion-surface scattering which includes trajectory-dependent electron exchange processes.
- To use this model to develop a trajectory-dependent charge exchange spectroscopy sensitive to lateral variations in surface electronic properties.

During the period covered by this grant we have completed the apparatus for this project. We have recently obtained energy- and angle-resolved spectra of 50 eV to 4 keV alkali and noble gas ions scattered from a Cu(110) surface. Representative spectra will be shown.

After general features of the scattering dynamics at these different energies have been studied, we will begin measurements on charge exchange systems. The general approach that will be used for studying ion-surface charge exchange is to scatter well-characterized ion beams from surfaces and measure the energy and angular distributions of the scattered particles. Measurements of scattered ion distributions and peak intensities will be used to monitor charge exchange cross sections as a function of different physical parameters of the ion-surface systems (i.e. different incident ion species, different crystal surfaces, different scattering geometries, addition of surface adsorbates, etc.).

The scattering trajectories, even from single crystal surfaces, can be complex and difficult to interpret. It is often necessary to rely on computer simulations for identifying different peaks in an energy spectrum. At Cornell we have developed a simulation, called SAFARI, for hyperthermal energy ion scattering and report on the status and use of that simulation. For the keV energies, we use MARLOWE, a code developed by Mark Robinson from Oak Ridge National Labs. Both of these simulations are discussed in this report.

III. STATUS OF RESEARCH

1) Completion and Performance of Apparatus

We have recently completed construction of a UHV scattering chamber for ion beam analysis of single crystal surfaces. The beams range in energy from ≈10 eV to 10 keV. Available beams include alkali ions, noble gases, and other gas atomic and molecular species. Full energy- and angle-resolved spectra of scattered ions are measured using single-channel electrostatic analyzers and pulse-counting electronics; these detectors are being upgraded to the multi-channel mode. Additional capabilities include Auger electron spectroscopy, low energy electron diffraction, sputter cleaning and annealing, and hardware for gas and alkali atom deposition. We are adding to the chamber a Kelvin probe for making work function measurements and hardware for thermal desorption spectroscopy. More detail on the ion scattering hardware is given below.

a) <u>Beamline and source</u>

Making use of two different ion optics and transport programs, we have designed a differentially pumped UHV system to produce well-characterized singly-ionized beams of alkali and noble gas ions in the energy range from ~10eV to 10 keV [1]. Beam transport was designed to minimize the effects of space charge spreading for the hyperthermal energy beams. Beams of a few to tens of nanoamps in a 1-4 mm diameter spot, with ± 1° to 2° angular divergence and 1% energy resolution have been achieved at the sample position. For producing the alkali ion beams, we have recently completed construction of source with high efficiency extraction [2]. The extraction electrodes are based on a modified Pierce geometry. The source uses a solid state alkali ion emitter (made by Spectra Mat, Inc.) and two stages of extraction. A wire mesh in the first stage produces a more uniform current density across the beam profile. Using this source we can produce beams of Li, Na, K, Rb, and Cs ions.

b) Single-channel analyzer and electronics

The detector for the scattered ions is a hemispherical electrostatic analyzer with a 50 mm mean radius. The gap is 35% of the mean radius. One mm input and output apertures define the energy resolution to be 1% of the pass energy. A Channeltron electron multiplier is used for either pulse counting or analog measurements of the particles transmitted through this detector.

Two sets of electronics are used with this analyzer. The first is a set of low voltage electronics which is used for the hyperthermal energy beams (for pass energies of 0-170eV). The other is a set of high voltage electronics used for pass energies of ~100eV to 10keV. The analyzer is designed so that the desired pass energy is chosen by putting equal and opposite voltages on the two hemispheres. An energy scan is taken by sweeping the potential on the hemispheres under computer control with programmable dwell time and a programmable number of repetitive sweeps. Counts are accepted from the electron multiplier using a computer interfaced scaler with programmable dwell times. This analyzer will soon be equipped with a resistive anode multi-detector.

2) Ongoing Development of Apparatus

a) Spherical analyzer designed for multiple-energy channel detection

Due to the relatively large size of our present analyzer and due to space constraints inside the scattering chamber, the largest scattering angle currently accessible with the present arrangement is 128°. In order to detect scattered ions at angles near 180°, we have built a spherical analyzer which is a half-size version of the analyzer discussed above. Grooves cut into the spherical surfaces help to keep particles which scatter off the inner surfaces of the analyzer from being counted in the electron multiplier. The new analyzer is equipped with a resistive anode for multi-energy detection.

b) Toroidal analyzer

The toroidal analyzer is the next planned upgrade of the detector. Using the toroidal geometry we will be able to perform multiple-energy and multiple-angle detection simultaneously. Due to the increased complexity of the toroidal geometry, parts of this design must wait until we have tested the spherical multi-detector.

In collaboration with our machine shop, we have begun planning the design of this analyzer and how the machining of the electrodes will be done.

3) Present Simulation Capabilities for keV and Hyperthermal Scattering

We have developed a computer simulation, called SAFARI, currently running on the Cornell supercomputer, to simulate scattering of hyperthermal energy ions (a few eV to 100 eV) from periodic surfaces [3]. For simulations of keV energy ion scattering we use MARLOWE [4]. For the purposes of discussion here, the two most important distinctions between SAFARI and MARLOWE are 1) the way in which collision partners are chosen along the trajectory of the scattered ion, and 2) the technique used for choosing impact parameters for the incident ions. Both of these features and their relative merits are discussed more fully below.

a) <u>SAFARI</u>

SAFARI was written explicitly for simulations of hyperthermal energy ion scattering (in the energy range of a few hundred eV or less) where the collisions of the incident ion with individual surface atoms are no longer in the binary regime. Rather, at any given point along the trajectory, the simultaneous interactions of the incident ion with several surface atoms must be considered.

Because the scattering potential includes simultaneous interactions with several surface atoms, the calculation of each trajectory is time consuming. In order that SAFARI be made useful for running large numbers of spectra for comparison with data, the number of trajectories required to generate a spectrum is reduced by using the so-called adaptive grid size method for choosing impact parameters. This method is designed to efficiently simulate an experiment with a specified range of outgoing energies and angles, and with specified experimental energy and angular resolutions. For such applications this technique is considerably more efficient than Monte Carlo simulations in which impact parameters are chosen at random.

Using the adaptive grid size method, a grid is drawn which overlays the surface unit cell. The initial impact parameters coincide with the corners of all the grid subcells. Trajectories with these initial impact parameters are calculated. It is decided which energies and angles are of interest for a particular experiment. Further subdivision is performed only on regions of the surface unit cell for which the corresponding trajectories will contribute to that experiment. In [3] we give an example for which the adaptive grid size method required an order of magnitude fewer trajectories than Monte Carlo to converge to a statistically meaningful spectrum.

b) <u>MARLOWE</u>

MARLOWE is a simulation program developed by M.T. Robinson from Oak Ridge National Labs. MARLOWE makes use of the "binary collision approximation" in which the incident ion interacts with one surface atom at any given time (except for the case of nearly equidistant collision partners), and multiple collisions are treated as sequential binary collisions. By comparing simulated spectra with experimental ones, this assumption is found to be generally valid in the several hundred to kilo-electron volt range. Because of the relative simplicity of the scattering potential in the binary regime, the calculation of each trajectory is fast. We have used MARLOWE to simulate spectra of scattered keV alkali and noble gas ions.

MARLOWE uses a Monte Carlo technique for choosing impact parameters.

4) Ongoing Development of Simulation Capabilities

We have run simulations with SAFARI in which we compare the Monte Carlo versus adaptive grid size techniques of choosing impact parameters. As stated above, in reference 3 we give an example for which the adaptive grid size technique required an order of magnitude fewer trajectories than Monte Carlo to generate spectra of equivalent resolution. We are investigating changing the impact parameter selection routine in MARLOWE from Monte Carlo to the adaptive grid size technique.

Also, we are continuing to search for new algorithms which reduce calculation times for individual trajectories.

5) Ongoing Experiments on Charge Exchange Mechanisms

We have only recently begun to collect ion scattering spectra with the apparatus described in this report. Charge exchange probabilities are experimentally determined by measuring peak intensities of the scattered ions. For a detailed analysis of the charge transfer mechanism, it is useful to be able to correlate peaks in the experimental spectra with specific trajectories; i.e. single scattering trajectories or double and multiple scattering trajectories. It is for this reason that the scattering simulations are essential to this project.

Figures 1a and 1b show representative spectra of 1 keV K^+ scattered from Cu(110). The beam is incident 45° from grazing and the detector is located 45° from grazing in the plane defined by the incident beam and the surface normal. We refer to this scattering geometry as 90° specular scattering. Spectra are shown for scattering along the $\langle 001 \rangle$ (figure 1a) and $\langle 1\text{-}10 \rangle$ (figure 1b) azimuths.

Using trajectory analysis we have identified the major peaks in the two spectra. In figure 1a, the lowest energy peak at $E/E_0 \approx 0.25$ represents single scattering through 90° from individual surface atoms (where E_0 is the energy of the incident beam). The broad peak centered at $E/E_0 \approx 0.3$ includes zig-zag trajectories and multiple collision trajectories which involve collisions with second layer atoms. The highest energy peak at $E/E_0 \approx 0.48$ represents double scattering from two adjacent top layer atoms along the $\langle 001 \rangle$ azimuth. Note that in the $\langle 1\text{-}10 \rangle$ azimuth (figure 1b) there are two dominant peaks in the spectrum. The low energy peak is single scattering from individual atoms. The higher energy peak at $E/E_0 \approx 0.47$ is double scattering from two adjacent surface atoms along the close-packed rows of the Cu(110) surface.

Figure 2a shows a comparison of 1 keV scattered K^+ (solid curve) and Ar^+ (dotted curve) beams along Cu(110)$\langle 001 \rangle$ with a 90° specular scattering geometry. Two important differences between the spectra are: 1) The ion survival probability for K^+ is more than an order of magnitude larger than that for Ar^+ (The left-hand scale represents counts in the K^+ spectrum and the right-hand scale represents counts in the Ar^+ spectrum. The incident beam on target was 6 nA for the K^+ spectrum and 12 nA for the Ar^+.), and 2) the pure single scattering peak intensity ($E/E_0 \approx 0.25$) is relatively larger in the Ar^+ spectrum (compared to the multiple scattering peaks) than in the K^+ spectrum.

Both features 1) and 2) are due to the fact that the Auger neutralization cross section for Ar^+/Cu(110) is large whereas the K^+ ions are expected to survive as ions due to the low ionization potential of K relative to the Fermi level of Cu(110). Were the ionization potential of K larger than or equal to the Fermi level of Cu(110), we would expect to see resonant neutralization of the K^+ ions at the surface. The fact that the single scattering peak is somewhat enhanced in the Ar^+ spectrum demonstrates that the Ar^+ ions undergoing multiple collisions with Cu surface atoms have higher neutralization probabilities than those undergoing single atom collisions.

Figure 2b is a comparison of 1 keV K^+ (solid curve) and Ar^+ (dotted curve) scattering from Cu(110)⟨001⟩ for 70° specular scattering (35° from grazing along the incident and outgoing trajectories). Notice that for the K^+ spectrum, the pure single scattering peak at $E/E_0 \approx 0.4$ is buried under the multiple collision peak (maximum intensity at $E/E_0 \approx 0.45$). In the Ar^+ spectrum, due to preferential neutralization of the multiple scattering trajectories, this single scattering peak becomes clearly visible above the background.

Figures 3a and 3b show spectra for scattered 1 keV Na^+ ions along the Cu(110) ⟨001⟩ and ⟨1-10⟩ azimuths, respectively. The scattering geometry for both spectra was 90° specular. Identification of the peak structures are similar to those of the K^+ spectra in figures 1a and 1b, except that the pure single scattering along the ⟨001⟩ azimuth is hidden under the multiple collision peak at $E/E_0 \approx 0.48$.

Figures 4a and 4b show Na^+ spectra for 56 eV Na^+ from the Cu(110) ⟨001⟩ and ⟨$1-10$⟩ azimuths, respectively, again with 90° specular scattering. Here again the spectra for the two azimuths are quite different. SAFARI can be used to identify the peaks in these spectra. The peak positions and widths can be compared with those in figures 3a and 3b to illustrate some of the differences between keV and hyperthermal scattering. An extensive discussion of these differences will be included in a forthcoming paper.

We are currently in the process of measuring peak intensities relative to incident beam intensities for alkali ions (Li^+, Na^+, K^+) scattered from Cu(110) surfaces as a function of sample azimuth, scattering geometry, beam energy (10 eV to 4 keV), etc. Ion survival probabilities for the different alkali ions will be compared. These, in turn, are compared to scattered noble gas spectra (He^+,Ne^+, Ar^+). Beams of O^+ ions will also be used; in this case spectra of scattered O^+ and O^- ions will be measured. These spectra will be compared to spectra of similar beams and scattering geometries for the Ni(110) surface. Our goal in this case is to compare resonant neutralization probabilities for Cu(110) and Ni(110) surfaces. For our purposes, the principal difference between these two surfaces is an increased density (by an order of magnitude) of d-like electrons at the Ni(110) surface. We expect to learn from these experiments how the charge exchange probabilities depend on the density and spatial extent of the Fermi level electrons at the metal surface.

IV. REFERENCES

[1] D.L. Adler and B.H. Cooper, Rev. Sci. Instr., to be published (1988).

[2] D.R. Peale, D.L. Adler, B.R. Litt, and B.H. Cooper, in preparation.

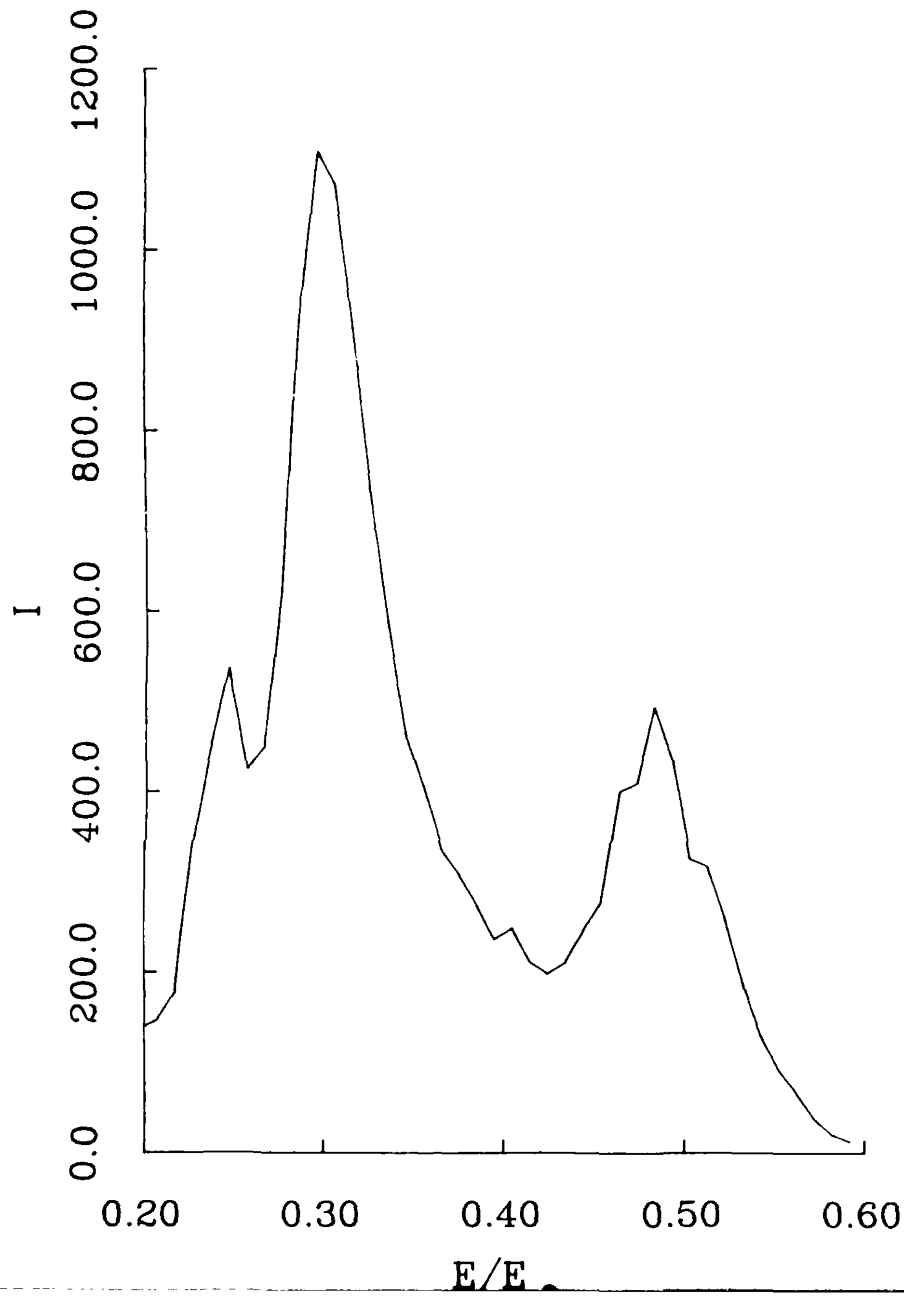
[3] D.M. Goodstein, S.A. Langer, and B.H. Cooper, J. Vac. Sci. Tech., to be published (1988).

[4] M.T. Robinson and I.M. Torrens, Phys. Rev. **B9**, 5008 (1974).

V. FIGURES

Figures 1 through 4 are representative spectra of alkali and noble gas ions scattered from the Cu(110) surface. Each of the figures is discussed in section III-5 of the text.

Fig. 1a



I
0.0
200.0
400.0
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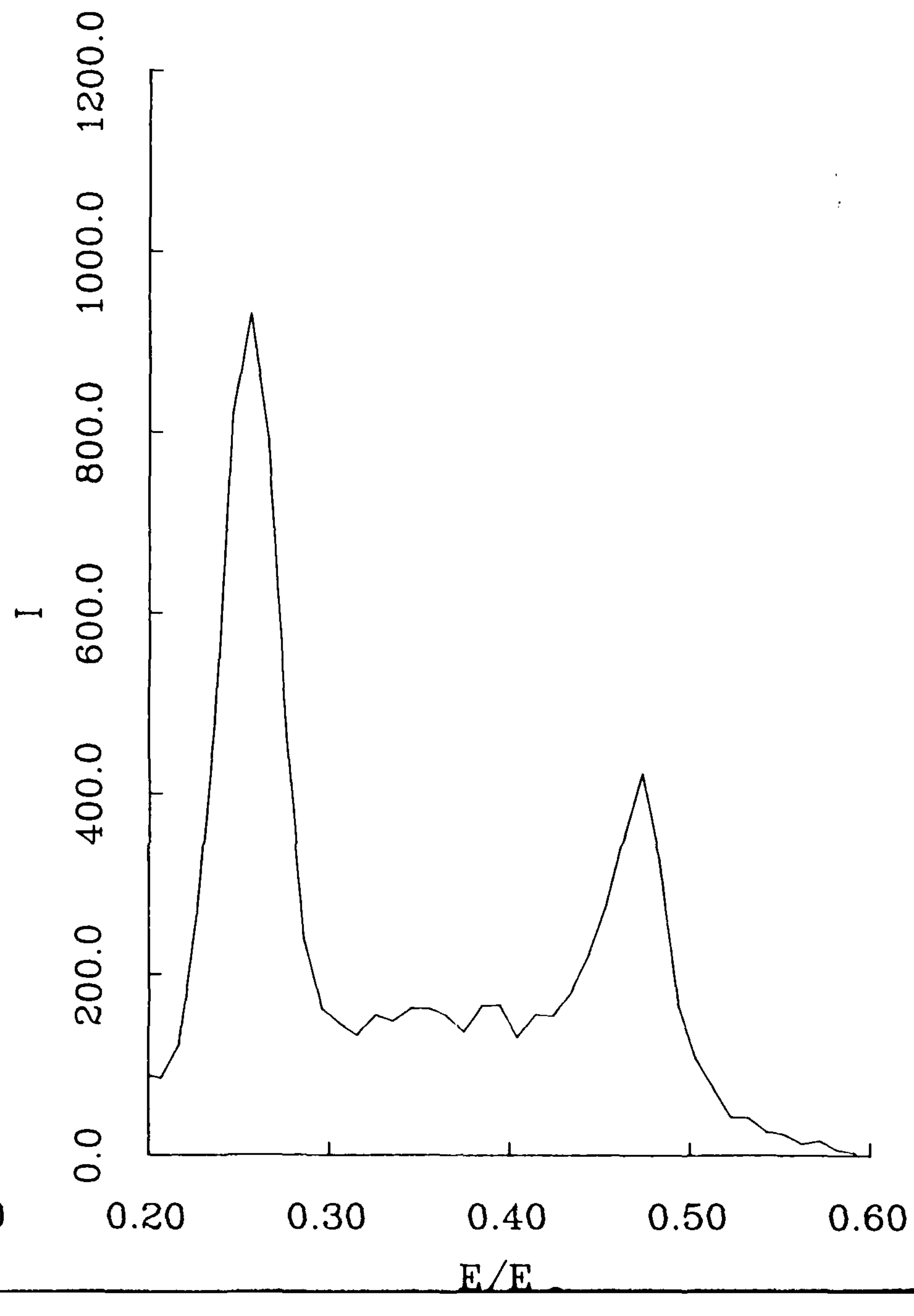
E/E

Fig. 1b



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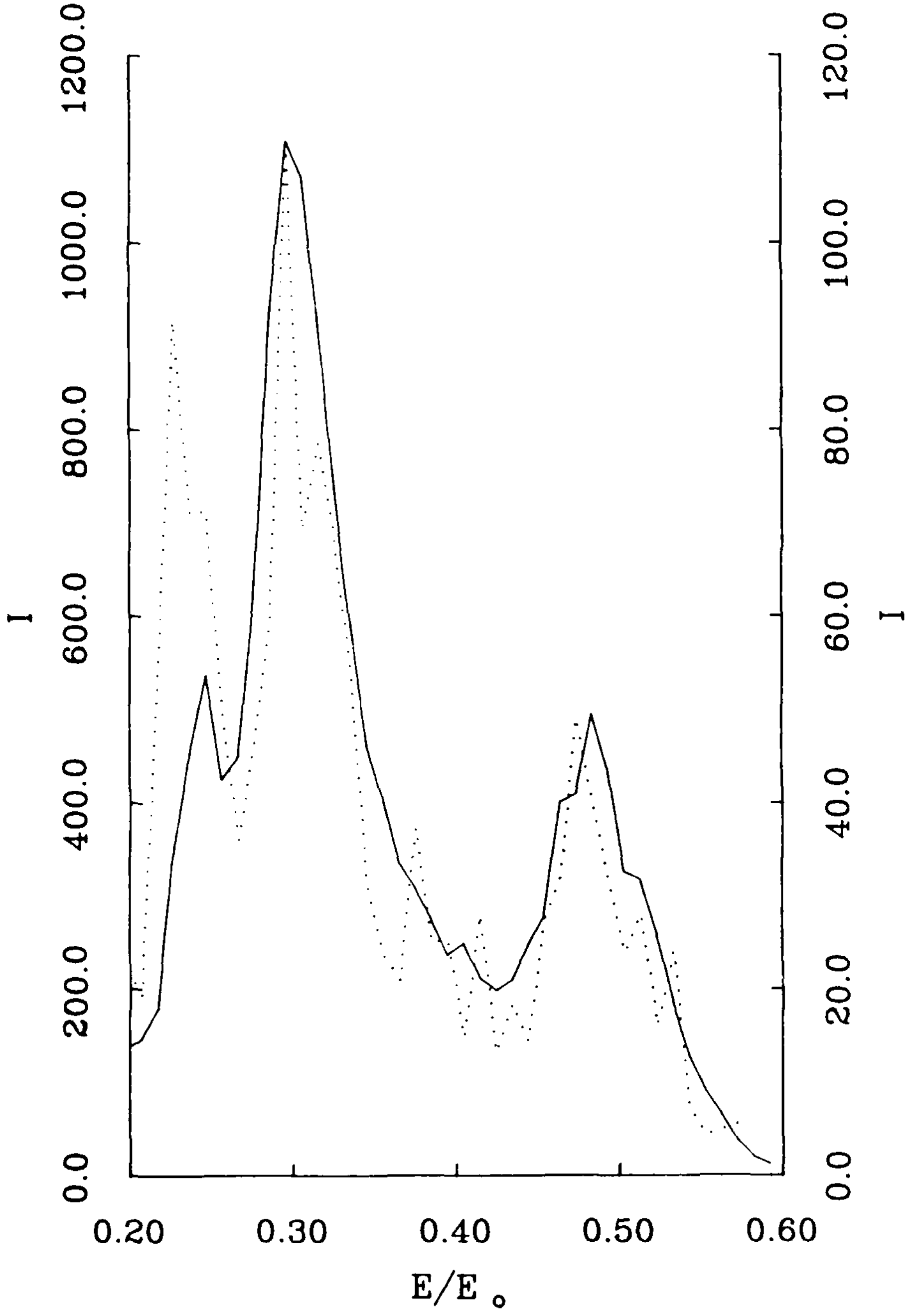
E/E

Fig. 2a



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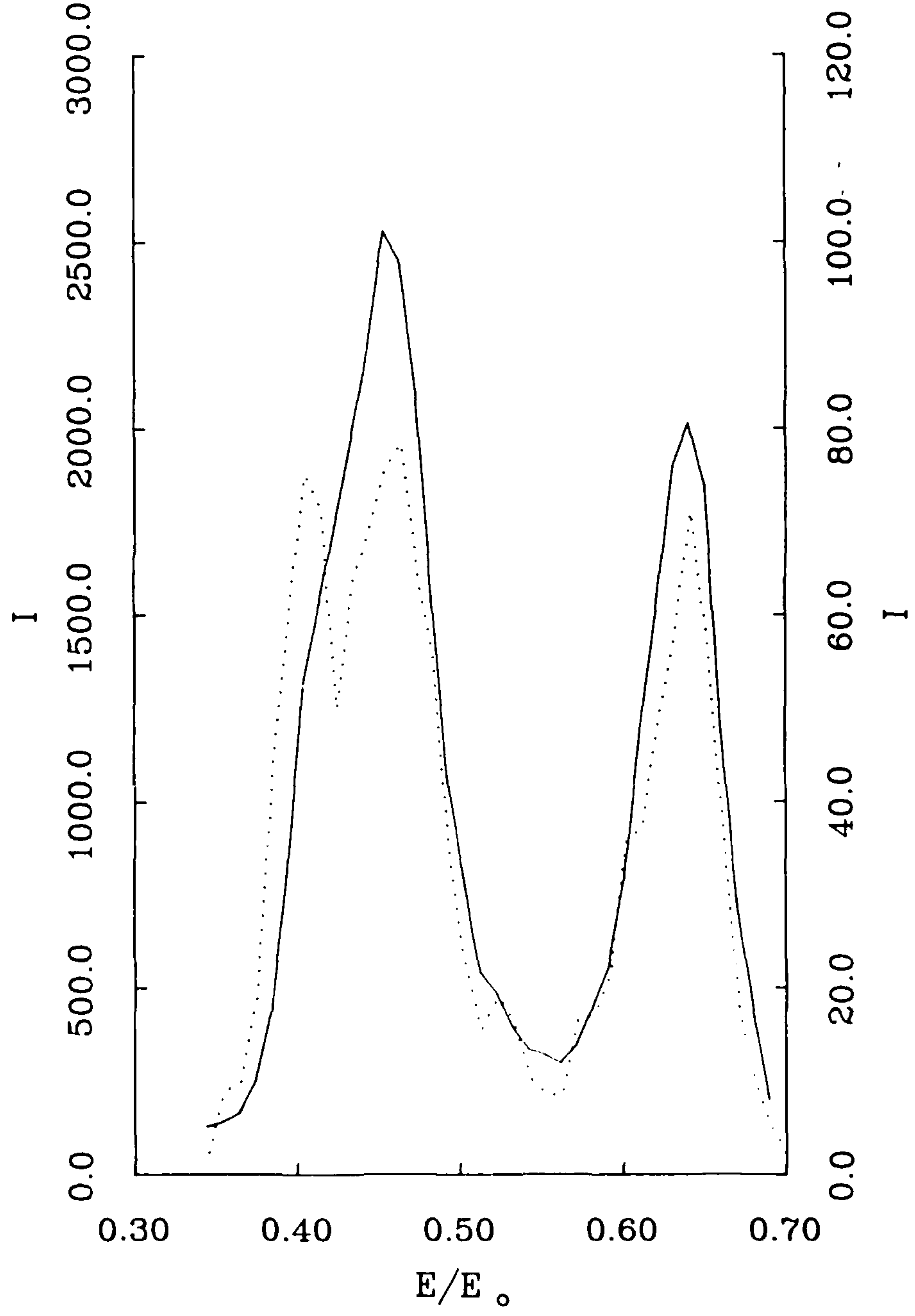
0.40
0.50
0.60
E/E o

Fig. 2b



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100.0
80.0
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40.0
20.0
0.0
I
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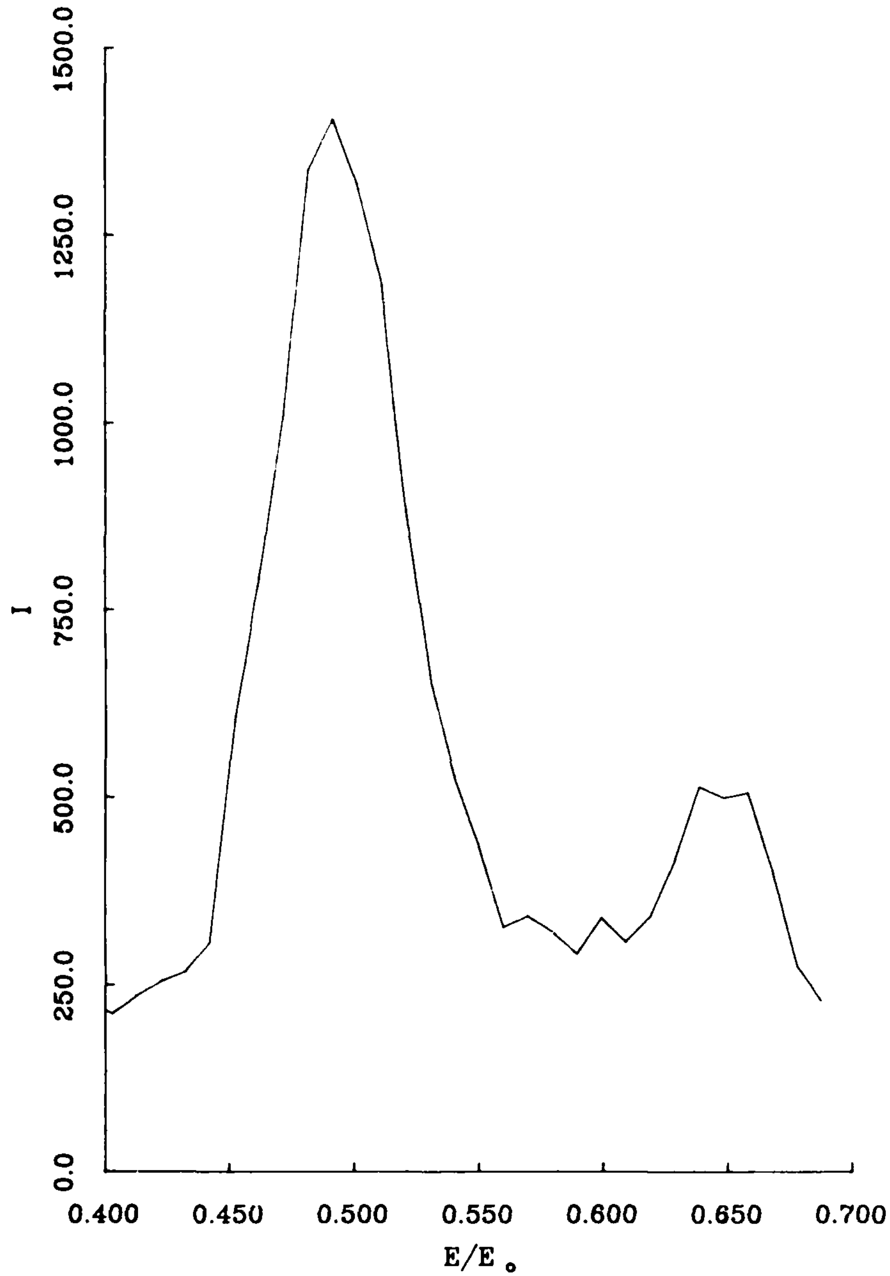
0.40
0.50
0.60
0.70
E/E o

Fig. 3a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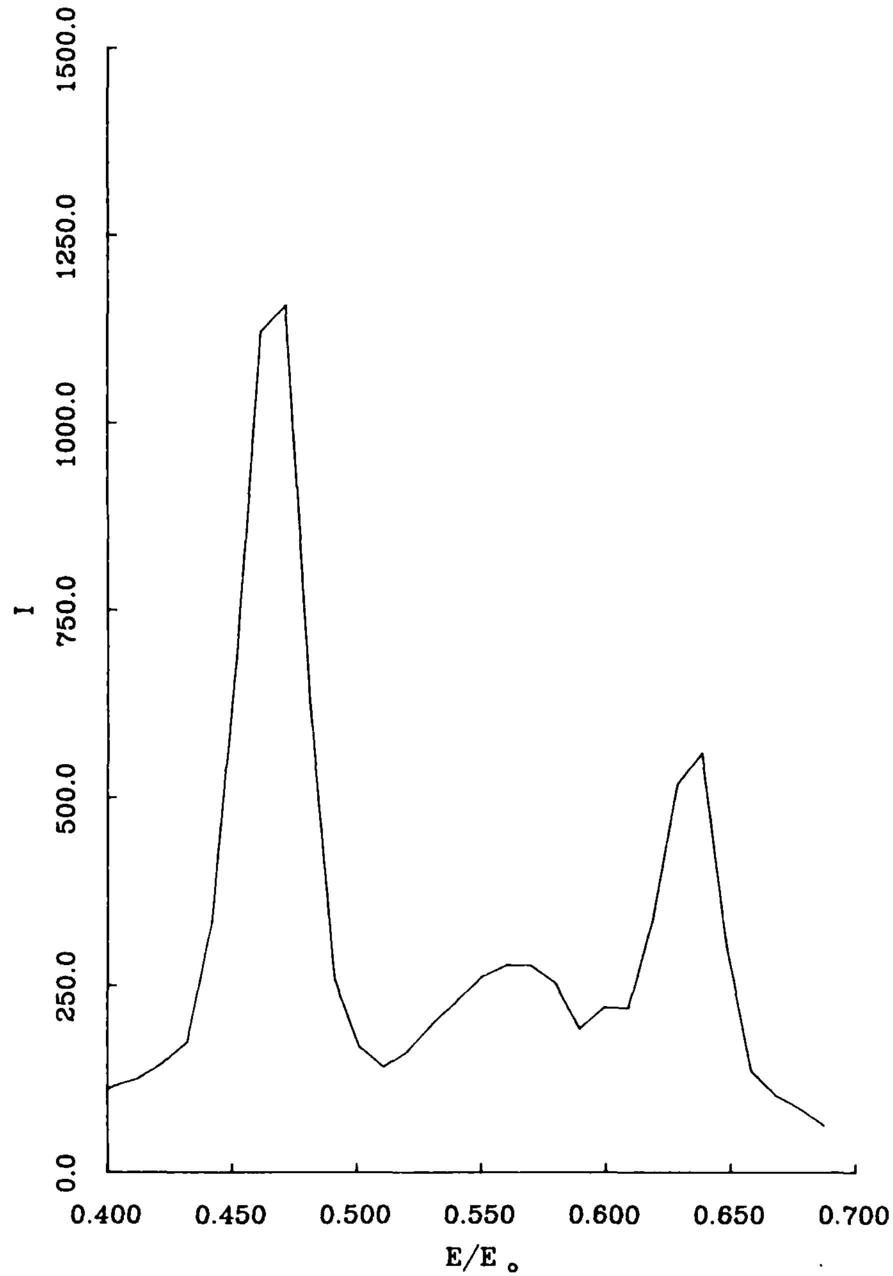
0.650
0.700
E/E o

Fig. 3b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500.0
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I
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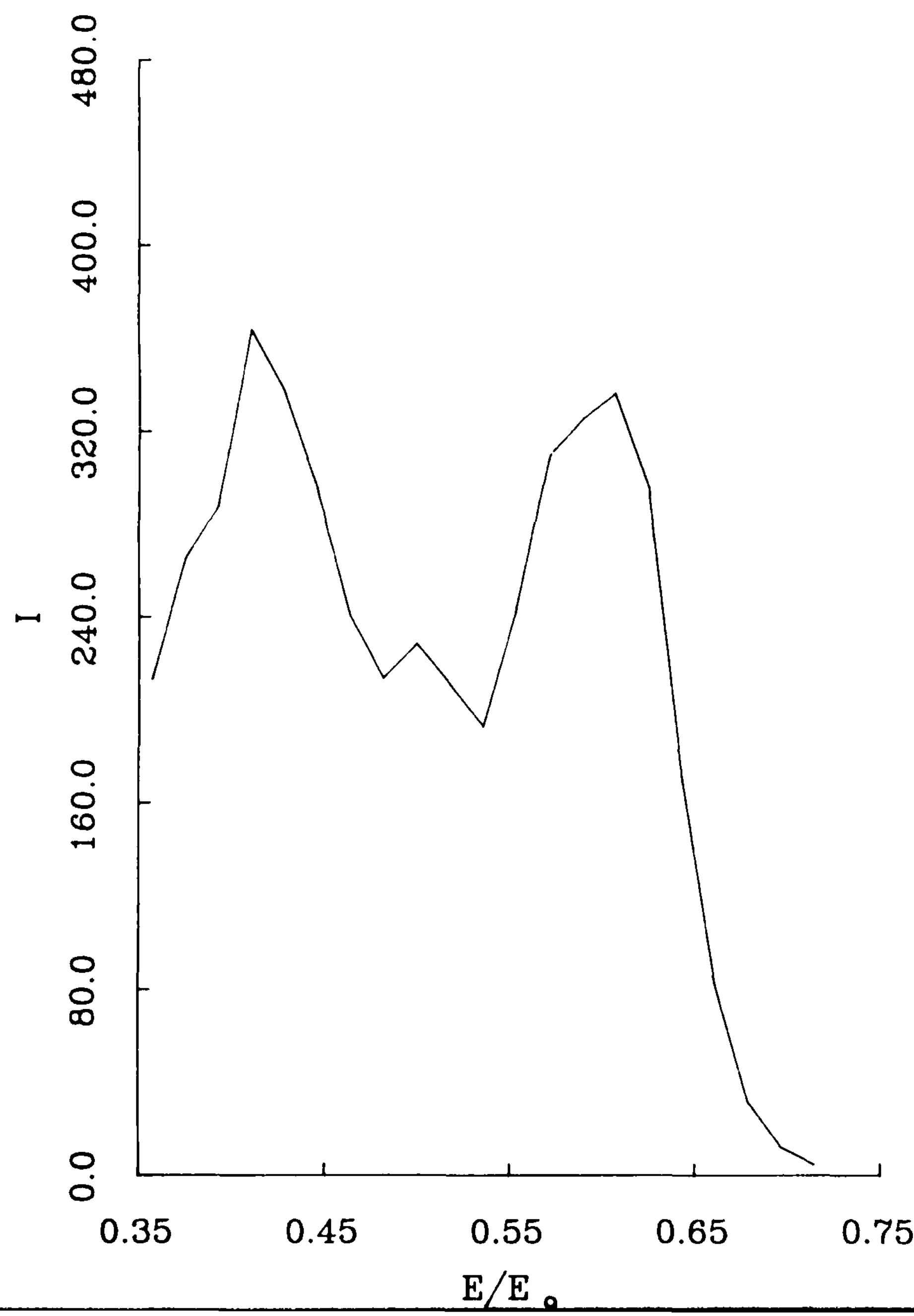
E/E o

Fig. 4a



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240.0
160.0
80.0
0.0
I
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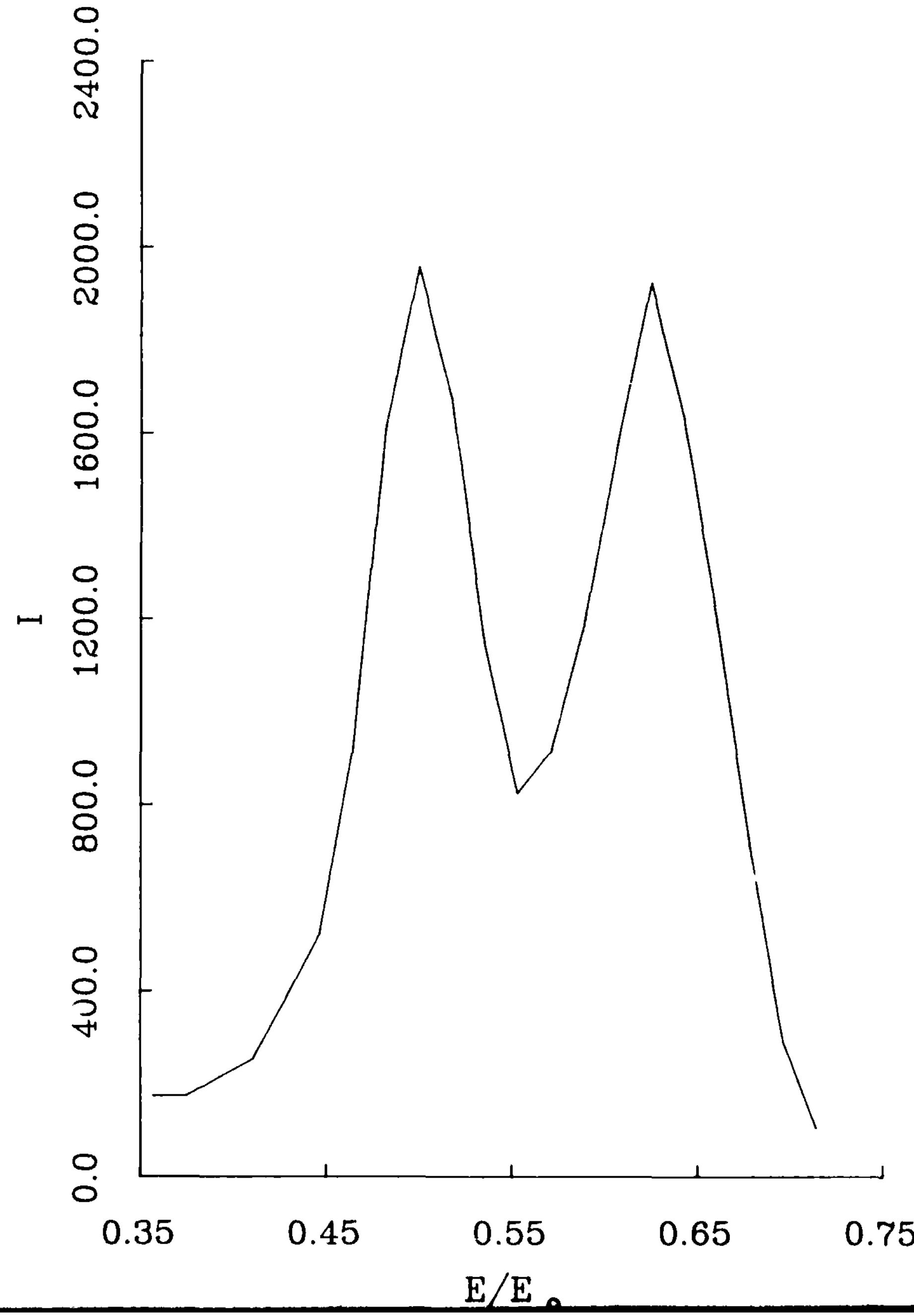
0.45
0.55
0.65
0.75
E/E$_o$

Fig. 4b



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1200.0
800.0
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0.0
I
0.35
0.45
0.55
0.65
0.75
E/E$_o$

VI. RESEARCH PARTICIPANTS

Participating in the charge exchange research program are myself (partial summer salary from AFOSR-86-0086) and graduate students D.M. Goodstein and D.R. Peale (both supported by AFOSR-86-0086). Other students working on the apparatus and involved in related research programs are D.L. Adler (supported by MSC/NSF-DMR-8516616) and R.L. McEachern and G.A. Kimmel (both supported by PYI/NSF-DMR-8451979).

VII. PUBLICATIONS

The following papers have been accepted for publication:

1) An Efficient Algorithm for the Simulation of Hyperthermal Energy Ion Scattering
   David M. Goodstein, Stephen A. Langer, and B.H. Cooper, J. Vac. Sci. Tech., to be published.

2) The Design and Performance of a UHV Beamline to Produce Low and Hyperthermal Energy Ion Beams,
   D.L. Adler and B.H. Cooper, Rev. Sci. Instr., to be published.

3) Design and Performance of Ion Optics for Hyperthermal (10-100 eV) and keV Ion Scattering
   D.L. Adler, B.H. Cooper, and D.R. Peale, J. Vac. Sci. Tech., to be published.

The following papers are in preparation:

1) A Versatile Apparatus for Low Energy and Hyperthermal Energy Ion Scattering Spectroscopy,
   R.L. McEachern, D.L. Adler, D.M. Goodstein, G.A. Kimmel, B.R. Litt, D.R. Peale, and B.H. Cooper, to be submitted to Rev. Sci. Instr.

2) The Design and Performance of a Source for Producing Alkali Ion Beams,
   D.R. Peale, D.L. Adler, B.R. Litt, and B.H. Cooper, to be submitted to Rev. Sci. Instr.

3) Charge Exchange in Low Energy Ion-Surface Collisions: Relation to Surface Chemistry
   B.H. Cooper, to be published in Solid State Communications

VIII. PRESENTATIONS

1) Computer Simulations of Hyperthermal Ion Scattering: Measuring Short-Range Surface Order in Crystals
   B .H. Cooper and D.M. Goodstein, presented at the Sixth International Workshop on Inelastic Ion Surface Collisions, Argonne National Laboratory, August 1986.

2) Hyperthermal Ion-Surface Scattering Simulation: Alternatives to Monte Carlo
D.M. Goodstein, S.A. Langer, and B.H. Cooper, presented at the New York Meeting of the American Physical Society, March 1987.

3) A Versatile Apparatus for Low Energy and Hyperthermal Ion Scattering,
R.L. McEachern and B.H. Cooper, presented at the New York meeting of the American Physical Society, March 1987.

4) An Ion Scattering System for the Energy Range 10eV to 10keV,
D.L. Adler and B.H. Cooper, presented at the New York meeting of the American Physical Society, March 1987.

5) Ion-Surface Scattering at Low and Hyperthermal Energies: Scattering Dynamics and Charge Exchange in Relation to Surface Chemistry
B.H. Cooper, invited talk at the American Chemical Society Meeting, New Orleans, September 1987.

6) Ion Surface Scattering at Hyperthermal Energies: Scattering Dynamics and Charge Exchange
B.H. Cooper, talk at the AFOSR Surface Chemistry Contractor's Conference, Colorado Springs, September 1987.

7) Interactions of Hyperthermal Ion Beams with Metal Surfaces.
D.M. Goodstein, R.L. McEachern, and B.H. Cooper, presented at the 34^{th} National Symposium of the American Vacuum Society, Anaheim, California, November 1987.

8) Design and Performance of Ion Optics for Hyperthermal (10-100 eV) and keV Ion Scattering
D.L. Adler, B.H. Cooper, and D.R. Peale, presented at the 34^{th} National Symposium of the American Vacuum Society, Anaheim, California, November 1987.

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