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ELECTRON EMISSION DURING MULTICHARGED ION-SURFACE INTERACTIONS

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Recent measurements of electron spectra for slow multicharged N ion-surface collisions are presented. The emphasis is on potential emission, i.e. the electron emission related to the neutralization of the ions. When using N ions that carry a K shell vacancy into the collision, characteristic K Auger electron emission from the projectiles is observed, as well as, for specific surfaces, target atom Auger transitions (resulting from vacancy transfer). Measurements of the intensity of these Auger transitions as a function of the time the ions spend above the surface can serve as a useful probe of the timescales characterizing the relevant neutralization processes. This technique is elucidated with the help of some computer simulations. It is shown that neutralization timescales required in the atomic ladder picture, in which neutralization takes place by resonant capture followed by purely intra-atomic Auger transitions, are too long to explain our experimental results. The introduction of additional neutralization/de-excitation mechanisms in the simulations leads to much better agreement with the experiments.

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1. Introduction

The interpretation of electron spectra induced by highly charged ions scattering from metal surfaces is, in general, not straightforward [1–3]. Generally, these spectra contain contributions from both kinetic emission and potential emission. Studying the potential emission part of the spectra provides information on the neutralization of the highly charged ions in front of metal surfaces. A model for this neutralization, frequently used in the past, was first introduced by Arifov et al. [4]. The model assumes resonant neutralization of the highly charged ions by metal electrons from the valence band. This leads to highly excited atoms that are assumed to de-excite by intra-atomic Auger transitions. Recently we performed some computer simulations [5] showing that the de-excitation rates used in this model are too slow to explain our experimental results. Additional neutralization/de-excitation mechanisms were shown to be required in order to obtain the faster de-excitation rates inferred from the measurements.

In the following, we will summarize the experimental results and the computer simulations leading to the above mentioned conclusions. We will also discuss the application of these conclusions on an additional analysis of our measurements.

2. Experimental Results

The highly charged ions used in the experiments were provided by the Oak Ridge National Laboratory Electron Cyclotron Resonance (ORNL-ECR) ion source. A well collimated beam of m/q selected ions was directed on a crystal at various angles of incidence. Emitted electrons were observed using a hemispherical analyzer (3% energy resolution) with a biased channel plate. The analyzer was rotatable in the plane defined by the incoming beam and the crystal normal. This set-up is shown schematically as an inset of Figure 1.

Also shown in Figure 1 is a set of measurements [5] taken for 24 keV N^{q+} ions ($q = 2, 4, 6$) incident on Cu(100) at 10° relative to the surface and a detection angle of 130° relative to the incoming beam. The spectra were normalized to the respective total electron yields which are given in the Figure caption. Assuming kinetic emission to be independent of the charge state of the incoming projectiles [6], distinction between kinetic emission and potential emission can be made. As discussed elsewhere [5], 95% of the N^{2+} electron spectrum consists of kinetic emission. We can therefore use this spectrum as the kinetic emission contribution in the N^{4+} and the N^{6+} spectra. Besides this kinetic contribution, the N^{4+} spectrum contains some additional intensity at the low energies (see Figure 1), part of which has been ascribed to LMM Auger electron emission from the moving projectiles [5].

For the N^{6+} spectrum in Figure 1, on the other hand, additional intensity is observed in a broad energy range. Most of this intensity (> 75%) consists of low energy electrons up to 30 eV [5]. These low energy electrons are probably emitted due to Auger mechanisms leading to the de-excitation of the highly excited atoms created close to the surface. Around 50 eV a discrete structure is observed which has been previously ascribed to LMM Auger electron emission from the moving projectiles [5,7]. The average energy of this structure is slightly higher than the comparable one in the N^{4+} spectrum (Figure 1), which is consistent with the expected shift in LMM Auger electron energies due to the presence of the K shell vacancy in the case of N^{6+}. The rest of the additional intensity starting at about 100 eV results from KLL Auger electron emission from the moving projectiles. The energy range of the discrete structure around 360 eV is consistent with calculated KLL Auger electron energies [5,8] that are appropriately Doppler shifted. This structure may contain contributions due to the emission of KLL Auger electrons prior to surface penetration on the incident trajectory, from ions travelling just below the surface and from reflected ions (however, only reflected

over small scattering angles). When the ions penetrate deeper than the mean free path of such 360 eV electrons in the target (at least on the order of 5 Å), inelastic scattering of the electrons will give rise to a much broader energy distribution. Such a broad energy distribution underlying the discrete structure is indeed observed experimentally (see Figure 1).

Similar results were obtained for another metal surface, namely Au(110) [1,9]. The dependence of the projectile KLL Auger electron intensity on the angle of incidence for this metal surface is shown in Figure 2 (after the subtraction of the broad energy distribution). The results were obtained for 60 keV N^{6+} ions and an observation angle of 90° with respect to the incident beam direction. An increase of the angle of incidence results in a decrease of the discrete KLL Auger electron intensity as was also found for Cu(100) [5]. This decrease in intensity can be explained by a decrease of the total time available for the neutralization of the incident ions. This was also found by de Zwart [2], who measured the LMM Auger electron intensity resulting from Ar^{9+} ions incident at 45° on polycrystalline W as a function of the energy of the incident ions. In the case of Au(110), interesting additional discrete structures were observed at 69 eV and at 220 eV [1] for both 60 keV N^{6+} ions and 70 keV O^{7+} ions. These structures were explained in terms of Au inner shell transitions, made possible subsequent to vacancy transfer from the projectiles to the target atoms. The intensity of the 69 eV target Auger transition is shown as a function of the angle of incidence of the N^{6+} ions in the inset of Figure 2. The behaviour of this structure is opposite of the behaviour of the KLL Auger electron intensity; an increase of the angle of incidence results in an increase of the target transition intensity. The reason for this increase is not straightforward and is related to different aspects of the neutralization problem as, for instance, the number of close collisions between the projectiles and the target atoms, the production mechanism for the

target vacancies and the fraction of projectile K shell vacancies available once the projectiles have penetrated the solid. This problem is discussed elsewhere in detail with the help of Monte Carlo simulations of the ion trajectories inside the solid [9].

3. Computer Simulations and Discussion

Recently we have described a computer simulation program calculating the de-excitation cascade of a highly excited atom formed in front of a metal surface by the resonant neutralization of a highly charged ion [5]. In brief, we assumed that a N^{6+} ion approaches a metal surface with a constant perpendicular velocity. At 30 a.u. above the surface six electrons were assumed to be resonantly transfered from the valence band of the metal surface to the n=7 shell of the ion. For the resulting highly excited atom, purely intra-atomic Auger processes were assumed to be the only de-excitation mechanisms. Furthermore, only those Auger mechanisms leading to electrons as low in energy as possible were taken into account. After each Auger step, a fast electron transfer from the metal valence band to the n=7 shell of the ion was assumed to keep the particle neutral. The intra-atomic Auger rates were estimated using Cowan's Hartree Fock codes [8]. The results obtained for this simulation are shown in Figure 3 where the number of electrons occupying the principal quantum shells are displayed as a function of the ion-surface distance (the perpendicular velocity used was comparable to 24 keV N^{6+} ions incident at 10°). The n=6 and the n=4 shells are not populated because such small Auger steps would not give rise to the emission of electrons. In contrast to the measurements (see Figures 1 and 2), in the simulation (see Figure 3) no KLL and even no LMM Auger electron emission is observed. This discrepancy between the measurements and the simulation is too large to be ascribed to the crudeness of the simulation [5]. Rather, it indicates that the assumption that the ion is resonantly neutralized to

a highly excited atom and subsequently de-excited only by intra-atomic Auger transitions may not be correct.

Therefore we performed [5] a modified simulation, assuming again resonant neutralization of the ion into n=7, but now followed by faster de-excitation mechanisms. We used the atomic rates multiplied by a factor of 20 for the transitions between n=7 and n=5 and for the transitions between n=5 and the M shell. In addition, we assumed the occurrence of Auger de-excitation directly into the L shell. The corresponding rate was taken to be 0 for large ion-surface distances and 10^{15} s^{-1} for distances smaller than 8 a.u. The results for this simulation are shown in Figure 4. As shown in the figure, this simulation leads to significant LMM and KLL Auger electron emission. As discussed in reference [5], with this modification satisfactory agreement with the measurements was obtained not only for the KLL and the LMM Auger electron yields, but also for the total electron yields.

As shown in the Figures 3 and 4, the simulations were stopped once the projectiles reached the surface. The measurements, on the other hand, might contain contributions from subsurface emission. However, our simulations can also be interpreted as starting and stopping at a distance shifted with respect to the surface plane. For example, starting the simulations at 25 a.u. above the surface and stopping at 5 a.u. below the surface, then corresponds to a 5 a.u. shift towards the surface.

A plausible explanation of our calculations is as follows. At a certain distance above the surface, the highly charged ions are neutralized leading to highly excited atoms. De-excitation of these atoms takes place by rates much faster than purely intra-atomic Auger rates. Close to the surface, fast Auger de-excitation directly into the L shell occurs, followed by KLL Auger electron emission from above and below the surface. As deduced from the

agreement between the simulations and the measurents, the loading of the L shell takes place during the last 8 a.u. This distance of 8 a.u. is probably related to the depth below the surface from which KLL Auger electron emission can be observed and thus to the mean free path of the KLL Auger electrons in the solid. Due to this fast loading of the L shell, the KLL Auger electron emission itself is reduced to, essentially, a one step emission problem (see Figure 4).

Such a scenario opens promising possibilities for the additional analysis of some of our experimental results. For instance, for the analysis of electron spectra of hydrogen like projectiles incident on Cu(100), assuming that the L shell is loaded close to the surface and using Monte Carlo simulations of the ion scattering inside the solid, it is possible to determine the effect of Doppler broadening on the Auger lineshape. Comparison with the measurements yields information on the relative contributions from incident, from penetrated and from reflected projectiles [9]. Studying these calculated projectile Auger electron intensities as well as calculated target Auger electron intensities as a function of the angle of incidence, and comparing them to the measured values (see Figure 2), provides more information on the neutralization problem.

Another possibility for such an analysis has recently been carried out on the electron spectra induced by bare ions like N^{7+} on Cu(100) [10]. In these measurements two discrete structures are observed, separated in energy by roughly 60 eV. These two structures are ascribed to the sequential filling of the K shell. Assuming that the L shell is loaded close to the surface reduces analysis to a simple two step problem. In the first step, the first K shell vacancy is filled, in the second step the remaining one. The ratio between the two intensities leads to an estimate of the total time available for this sequential decay and thus of the distance above the surface at which the "loading" of the L shell occurs. The measurements

and analysis will be presented elsewhere [10].

Finally, we mention the analysis of X-ray spectra obtained for hydrogen like ions incident on a metal surface. As observed recently by Briand et al. [11], such spectra contain detailed information on the electronic configuration of the atoms at the time the X-rays are emitted. Presently, we are working on a high resolution detection of such X-rays using a Bragg crystal in combination with a position sensitive detector.

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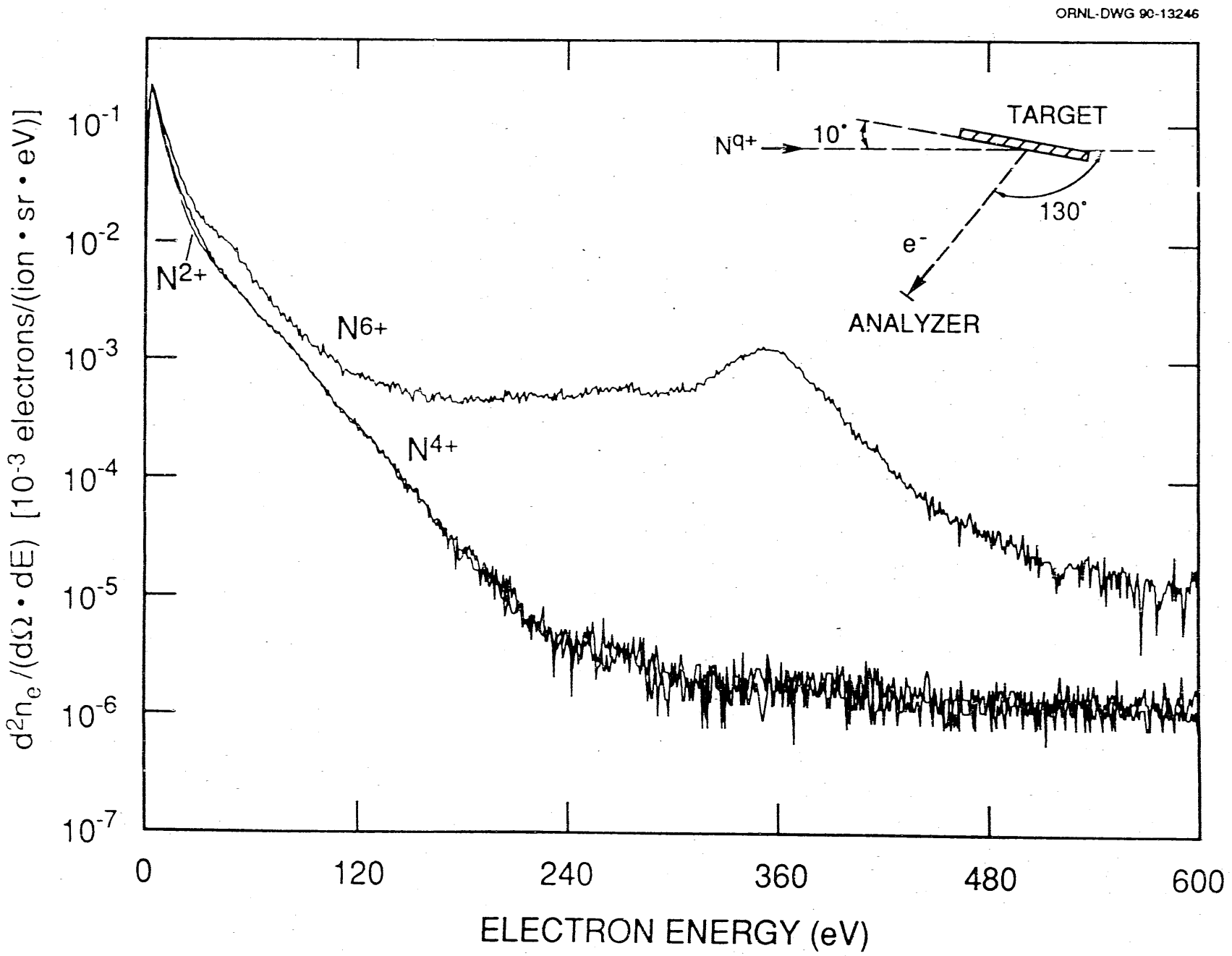
FIGURES

FIG. 1. Electron energy distributions for 24 keV N^{q+} ($q = 2, 4, 6$) ions incident on Cu(100) at 10°. The spectra were normalized to the respective total electron yields which were found to be 7.2, 8.1, and 12.5 electrons per ion in order of increasing charge states. A schematic view of the experimental set-up is shown in the inset.

FIG. 2. Dependence of the normalized KLL Auger electron intensity on the angle of incidence for 60 keV N^{6+} ions incident on Au(110). The dependence of the normalized Au line at 69 eV on the angle of incidence is shown in the inset. Both spectra shown are background subtracted.

FIG. 3. The calculated distribution of the electrons over the principal quantum shells as a function of the ion-surface distance deduced from the atomic ladder picture for 24 keV N^{6+} ions incident at 10°. The atomic rates which were used in the simulations, were estimated using Cowan's Hartree Fock codes [8].

FIG. 4. Same as Figure 3, but with the rates for the population of the n=5 shell and the M shell increased by a factor of 20 and assuming that Auger de-excitation directly into the L shell starts at a distance of 8 a.u.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TARGET
N^{q+}
10°
130°
e^-
ANALYZER
N^{2+}
N^{6+}
N^{4+}
10^{-1}
10^{-2}
10^{-3}
10^{-4}
10^{-5}
10^{-6}
10^{-7}
$d^2n_e/(d\Omega \cdot dE)$ [10^{-3} electrons/(ion • sr • eV)]
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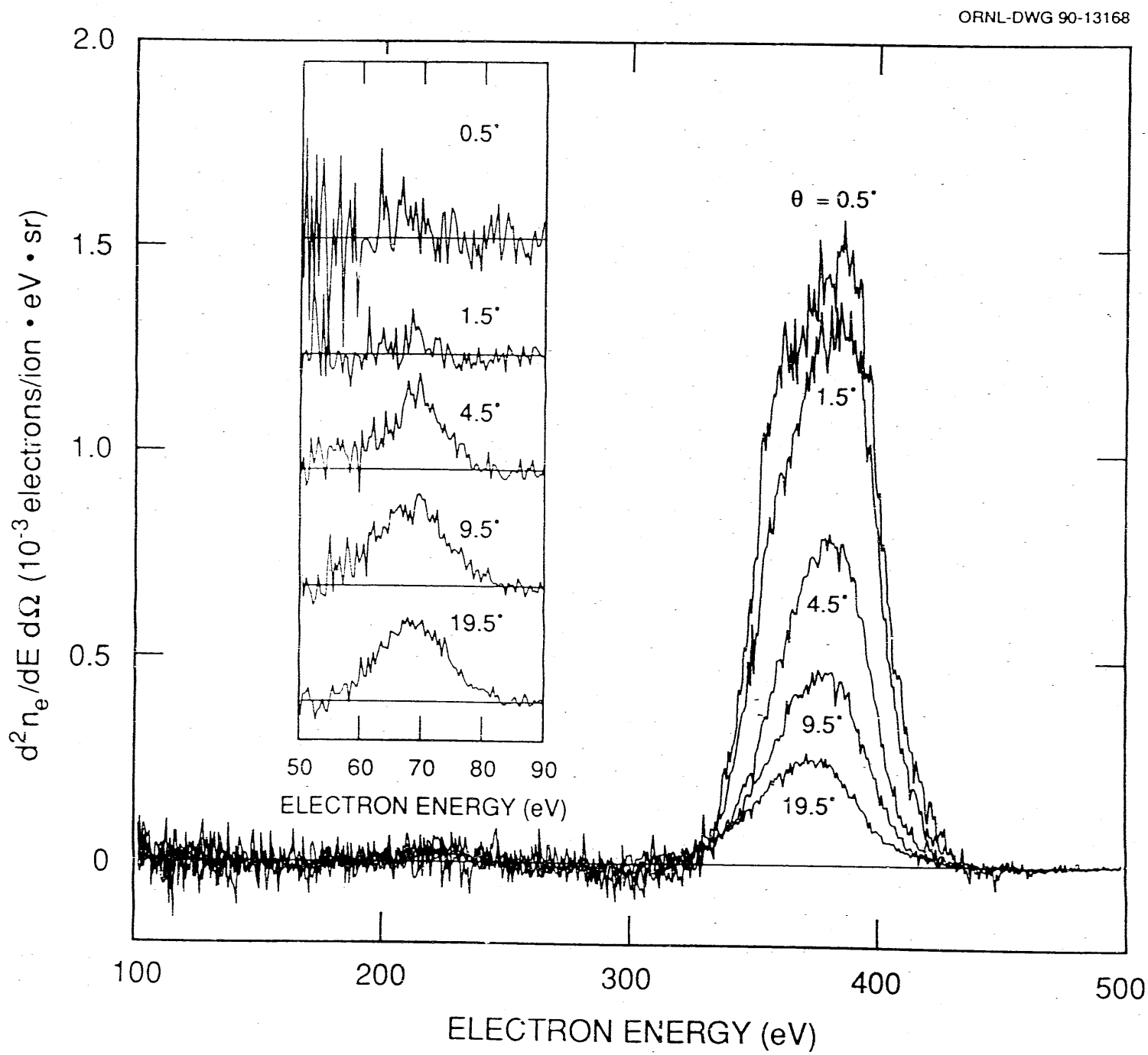
240
360
480
600
ELECTRON ENERGY (eV)

Fig. 1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1.0
0.5
0
$d^2n_e/dE\,d\Omega$ (10^{-3} electrons/ion • eV • sr)
100
200
300
400
500
ELECTRON ENERGY (eV)
θ = 0.5°
1.5°
4.5°
9.5°
19.5°
0.5°
1.5°
4.5°
9.5°
19.5°
50
60
70
80
90
ELECTRON ENERGY (eV)

Fig. 2

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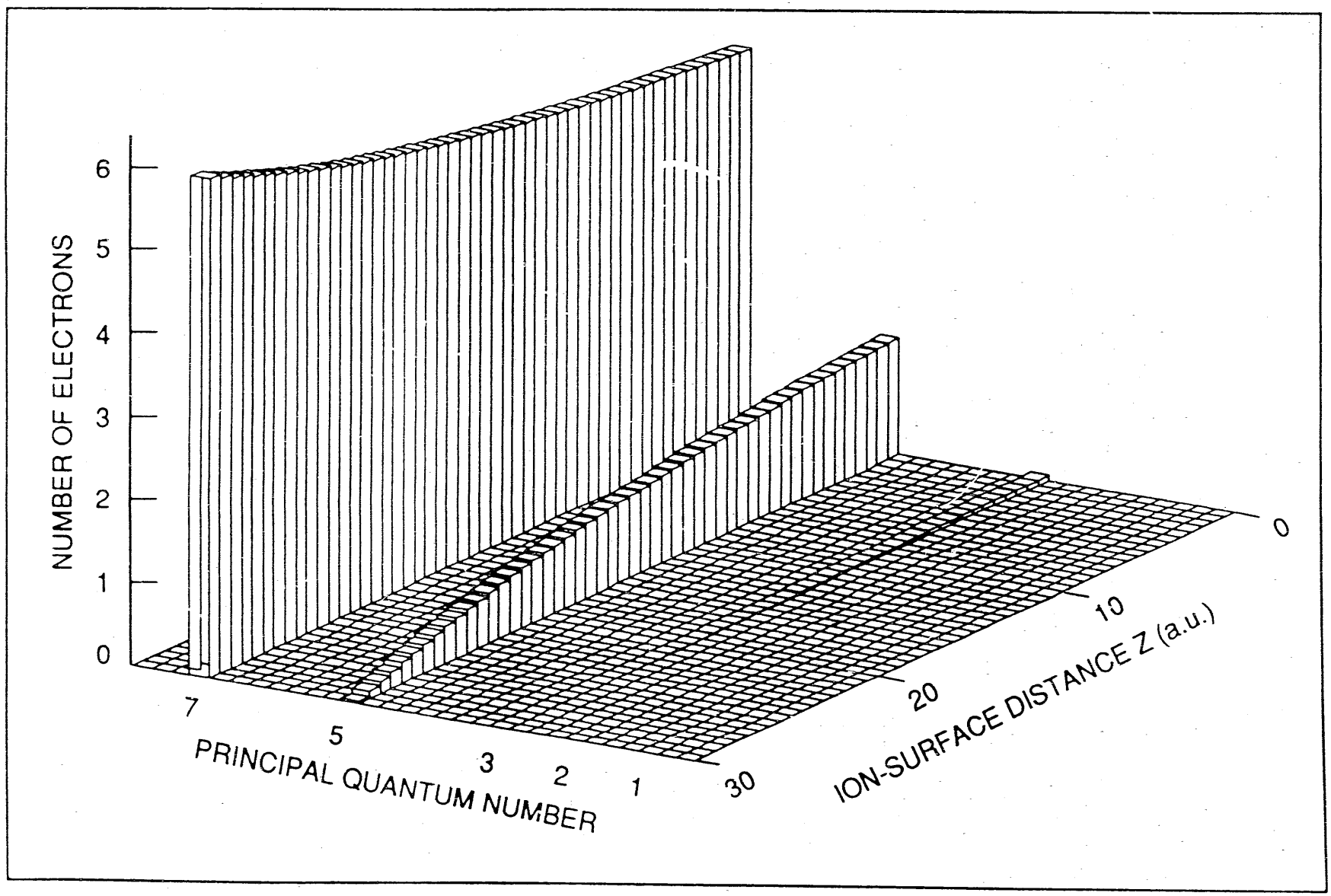

Fig. 3

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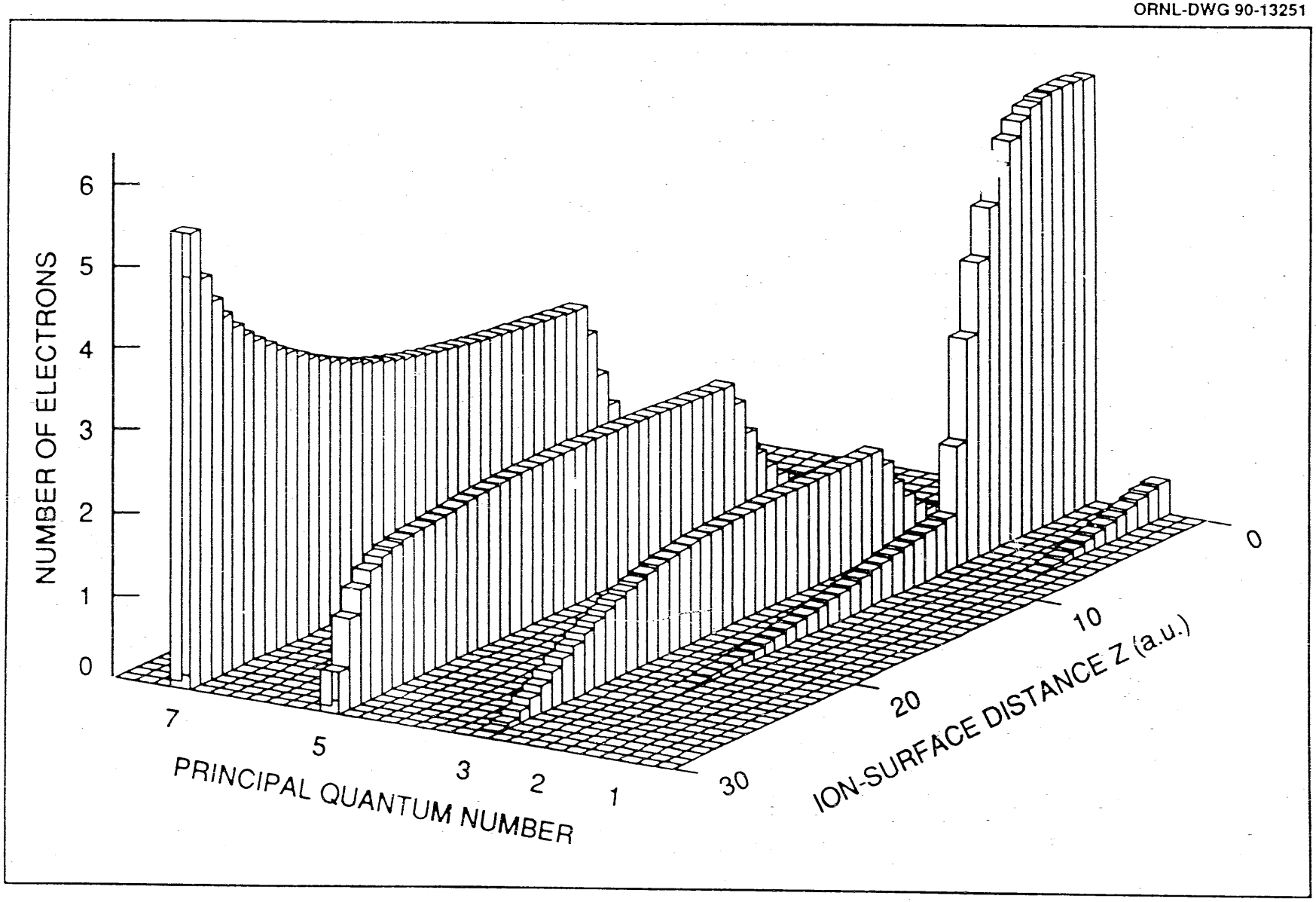

Fig. 4

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