This chapter extends the discussion presented in the previous chapter to the energyloss and charge state distributions of oxygen projectiles scattered off an Al(110) surface. The energyloss spectra and charge state distributions were obtained as a function of incident charge state of the O projectiles.
10.1 Introduction

The previous chapter discussed the energy loss and charge state distribution of He projectiles scattered off an Al(110) surface. The relative simplicity of this collision system facilitated the application of a model in which energy loss is described in terms of friction coefficients. In this model, the magnitude of the friction undergone by the projectiles traveling through the target electron sea depends on the charge state of the projectile – i.e. neutral He atoms suffer much less friction than ionized projectiles. At some point along its trajectory, the charge state of the He projectile can be altered leading to different energy losses before and after the charge changing process. The distribution of pathlengths traveled before and after the point of charge exchange was modeled in terms of two ‘mean free paths’ – equal to the product of projectile velocity and Auger lifetime (for He$^+$) or dynamic resonant loss lifetime (for He neutrals). From this distribution a theoretical energy loss spectrum was derived which – after inclusion of straggling – was fitted to the measured spectra. The parameters used to fit the spectra were the friction coefficient $\gamma_{\text{neu}}$ of neutral He, the minimum energy loss $Q_0$ and the straggling width $\Omega$.

A similar study of charge state fractions and energy loss of multiply charged ions impinging on surfaces might – just as for the He case – shed some light on the history (the charge changing and scattering events it experiences at the surface) of those projectiles. However, a description of energy loss suffered by larger $Z$-projectiles scattered off a surface is much more complex than for the He case of the previous chapter. Firstly, the number of possible charge states of the projectiles – and thereby the number of charge-changing processes involved – is much larger than for the former system. Secondly, the magnitude of the charge-dependent friction terms is not well known for larger $Z$-projectiles. In this short chapter, a set of pilot experiments on the charge state distributions and energy loss resulting from multiply charged ion-surface collisions will be discussed.

In the experiments, the energy loss and charge state distributions of multiply charged O$^{q+}$ ($q \in \{2, 7\}$) projectiles scattered off an Al(110) surface were measured. For all charges, an energy of 16.3 keV ($v \approx 0.2$ a.u.) was chosen. The projectiles were incident along the $<1\overline{1}0>$ surface direction. A specular reflection geometry of $\psi = 2.5^\circ$ and $\theta = 5^\circ$ was chosen.

10.2 Scattered charge state fractions

The reflected charge state intensities as a function of incident charge state are shown in figure 10.1. The charge state distributions were measured in two steps. First, the intensities of the differently charged scattered ions
were obtained by integrating the energy distribution for each charge state measured using the electrostatic analyzer. The intensities obtained were corrected for incident beam intensity. Then, the time of flight tube mounted at $\theta = 10^\circ$ was used to measure the fractions $O^+ / O^+$ and $O^- / O^+$ of positive and negative ions with respect to the total scattered projectile yield for incident angles of 2.5, 5 and 7.5 degrees. For these incident angles, the measured + and - fractions were found to be practically angle independent. Therefore they can be used as a rough estimate of the $O^+$ and $O^-$ fractions one would measure for a $\psi = 2.5^\circ$, $\theta = 5^\circ$ geometry, facilitating an absolute calibration of the charge state fractions measured with the ESA.

Our results confirm the main findings of De Zwart et al.\textsuperscript{161} who measured the relative intensities of $1^+$, $2^+$ and $3^+$ scattered ions as a function of incident charge state ($q^+ = 1$ to 11) for 20 keV Ne, Ar and Kr ions incident at a polycrystalline W target. In their experiments, no dependence of the relative intensities for $q^+ = 1, 2, or 3$ on the incoming charge state was found. They could however not determine neutral scattered projectiles. More recently, Folkerts et al.\textsuperscript{162} measured scattered charge state intensities – including the neutral fraction – for 60 keV $O^{q^+}$ impinging on a Au surface. Again, they found the intensities of the low charge states to be virtually independent of the charge state of the primary ion. In their experiment, almost equal fractions of about 5% of $O^+$ and $O^-$ ions were found.

Figure 10.1: Scattered charge state fractions $O^{q^+} / O^{q^+}$ for 16.3 keV $O^{q^+} \rightarrow 7^+$ incident on Al(110) along the $<1\bar{1}0>$. Scattering geometry: $\psi = 2.5^\circ$ and $\theta = 5^\circ$. 
for all incoming charge states. The fractions of higher charge states were found well below the 1% level. For incident ions carrying a K-shell vacancy (O$^{7+}$ and O$^{8+}$), an increase roughly by a factor of 10 for the higher (2+ up to 5+) charge states was found.\textsuperscript{162}

In our experiments depicted in figure 10.1, the contribution of the O$^-$ fraction is about 3 times larger than the O$^+$ fraction (about 15% and 5% respectively). The larger yield of O$^-$ compared to the one reported by Folkerts et al.\textsuperscript{162} is attributed to the different electronic properties of the Al and Au targets used. The formation of negative ions will be discussed in more detail in the next chapter of this thesis.

For incident ions O$^{2+}$ to O$^{5+}$ a maximum 3+ charge state is found in the scattered projectile yield. The intensity of this fraction is - as the intensities of the -1, + and 2+ fractions - virtually independent on the primary charge state of the ion. The memory of the initial charge state therefore seems to be completely lost for these cases. However, for incident ions carrying a K-shell vacancy (O$^{7+}$ and a fraction of metastable O$^{6+}$ 1s2s) four-fold and five-fold charged ions are found among the scattered projectiles. These charge states show that a (small) fraction of K-shell vacancies survive the reflection from the surface. On the way out, these vacancies are filled by auto-ionization by which the charge state of the projectile is increased by at least one unit. The increase by a factor of $\sim 10$ of the ions scattered into a 4+ charge state for O$^{7+}$ primary ions compared to O$^{6+}$ ions is consistent with the expected 5% to 10% fraction of metastable 1s2s ions in the O$^{5+}$ beam (also see chapters 5 and 6). It is however important to emphasize that scattered projectiles in a charge state larger than 1+ make up a very small percentage ($\ll 1\%$) for all incident charges.

### 10.3 Energy loss

Figure 10.2 presents the energy loss spectra of scattered O$^{n+}$ ($r = -1 \text{ to } 3+)$ projectiles following collisions of O$^{n+}$ up to O$^{7+}$ ions on the Al target. All spectra are measured using the electrostatic analyzer. Since we are only interested in the shape of the spectra for different scattered species, the distributions are normalized to the maximum count rate for each individual spectrum. The abscissa is taken as the fraction $E/E_0$.

The energy loss spectra show virtually the same evolution for all incident charge states. This is exactly as one would expect in view of the constant yields of projectiles scattered into -1, +, 2+ and 3+ charge states. However, a significant difference in the maximum energy loss suffered by the different scattered ions is observed. The O$^-$ ions undergo the largest loss whereas the loss for the ions scattered into a positive charge state slightly decreases with increasing charge. Also the minimum loss (in the previous chapter denoted
Figure 10.2: Energy loss distributions of scattered $O^{r+}$ $r = 1, 2, 3$ for 16.3 keV $O^{2+}$ - $O^{5+}$ incident on Al(110) along the <110> crystal axis. The abscissa is normalized to the primary energy, the ordinate to the maximum count rate for each individual spectrum. The scattering geometry was $\psi = 2.5^\circ$ and $\theta = 5^\circ$. 
by \( Q_0 \) is slightly decreasing with increasing charge of the scattered projectile. The decrease in width of the spectra observed for increasing scattered charge state is attributed to a decrease in straggling (\( \Omega \)) undergone by the projectiles. The slopes of the low-energy tails of the spectra – proportional to the inverse of the friction undergone by the projectiles, see the previous chapter – become steeper with increasing scattered charge state.

These observations, decreasing friction \( \gamma \), straggling \( \Omega \), and minimum loss \( Q_0 \) with increasing exiting charge state can only be explained by different trajectories followed by the projectiles. The primary charge of the ion obviously does not play a role, since the spectra show the same evolution for all incident charge states. Shorter trajectories through the electron gas for higher final charge states do however imply less friction, less straggling and smaller minimum loss. Moreover – as discussed in the previous chapter – the survival probability of a certain charge state depends crucially on the length of the trajectory followed inside the electron gas of the target.

Finally, the question arises how the energy loss distribution of O projectiles scattered as neutrals relates to the losses measured here. The loss spectra of negative O\(^-\) ions probably closely resemble the loss spectra of projectiles scattered as neutrals. In order to form a negative O\(^-\) ion, it is necessary to populate the affinity level of oxygen which is bound by 1.46 eV. As the ion approaches the solid, the affinity level is shifted downwards due to the interaction of the electron with the negative ion’s image charge within the metal. However, once the projectile enters the electron sea at the surface, the O-affinity level is merged into the conduction band of the solid. The negative oxygen ions observed in the reflected charge state distribution are therefore formed on the way out of the projectiles. Since neutral O is a necessary precursor for the formation of negative O, the energy distribution of negative oxygen ions merely reflects the distribution one would measure for O\(^0\).

A detailed study of the formation of negative ions can therefore give additional information not only on the target electronic structure but also on the deexcitation of the primary ions leading to the formation of the precursor neutrals. The next chapter discusses a study of the formation of negative ions resulting from collisions of singly- and multiply charged C, O, and F ions on an Al(110) surface.