Energy-angle distribution of low-energy hydrogen ions in thin aluminum and gold foils


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Abstract

Energy and angular distributions of hydrogen ions in thin solid foils of elements with significantly different electronic structures and atomic masses, such as Al and Au, have been measured at 9 keV using the transmission technique. The results are compared with Monte Carlo simulations using a classical scattering approach and an impact-parameter independent electronic energy loss, as well as with a simplified model containing the main physical features. In both cases, the effect of foil roughness on angular dependence of the energy loss has been included and it has been found to be responsible for the variations observed at small angles. The measured angular distributions have also been compared with standard multiple scattering functions corresponding to Thomas–Fermi and Lenz–Jensen potentials and with calculations based on $1/r^6$ power potentials. © 2000 Elsevier Science B.V. All rights reserved.

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1. Introduction

The energy distributions of protons as a function of the exit angle after traversing thin solid foils are determined by physical factors such as the elastic and inelastic energy loss cross-sections (as a function of the single scattering angle), and topological factors like the foil thickness and density inhomogeneities. The elastic scattering of the protons with the atomic nuclei determine the resulting angular distributions. This is described by theories based on the transport equation and collision theory [1–4]. In most cases, the electronic energy loss is described as an angle-independent magnitude formulated either as the stopping power $dE/dx$ or the stopping cross-section per atom $S$ [5,6]. Only in the last two decades some measurements performed at higher energies showed the need for considering impact parameter-dependent electronic energy losses $Q_{\text{inelastic}}$ per single collision to describe this process [7–14], because neither the geometrical path enlargement nor the elastic energy loss could explain the experimental results. These effects have been also found to be relevant in similar studies with heavy ions [15–19].
Foil thickness inhomogeneities also generate an observation angle dependence of the energy loss because the thinner sectors transmit the particles into a narrower cone than the thicker ones, so that when observing projectiles in the forward direction one collects preferentially those which came out from thinner sectors and therefore lost less energy [20,21].

The standard multiple scattering calculations are valid as long as the small angle approximation holds, the energy loss is negligible compared to the projectile energy, and the path length is equivalent to the penetrated depth. At the present energies not all of these conditions hold. Essentially, there is a difference between the travelled path length and the penetrated depth. This affects the tails of the distribution. The energy loss effect in the multiple scattering distribution is adequately taken into account, for low-energy ions, by using an average energy corresponding to the mean velocity of the projectiles in the target [22]. But to our knowledge there is not a simple way to take into account the breakdown of the other approximation in calculating angular distributions.

Angular dispersions of protons in the low-energy range have been previously measured for other materials such as carbon, for energies between 1 and 20 keV [23], and between 3 and 54 keV [24]. These papers show angular halfwidth as a function of energy and foil thickness. In [25] angular distributions of hydrogen, helium and argon ions in VYNS films are compared at energies ranging from 4 to 30 keV. However, a detailed study of the applicability of the multiple scattering theory in the range of very low energies has not yet been accomplished.

Regarding the angular dependence of proton energy loss at low energies, a simple approximation proposed by previous authors [23,26] considers a geometrical increase of the energy loss, proportional to the secant of the observation angle. Högberg [27] proposed a somewhat different path length enlargement for the energy loss of protons in C, considering a single deflection in the middle of the foil. All these authors claim an independence of the energy loss with the observation angle and they ascribe the observed differences to path length effects. On the other hand, Blume et al. [28] studying protons in gold observed an increase of the energy loss with observation angle beyond the simple length enlargement proposed by Högberg [27].

The purpose of this work is to obtain a better knowledge of the energy and angular distributions of low-energy hydrogen ions, after traversing thin solid foils and to test the applicability at these low energies of theoretical models used at higher energies.

In this work, we present measurements of energy spectra and angular distributions of 9 keV protons after traversing polycrystalline aluminum and gold targets.

The angular distributions are compared with tabulated values using the multiple scattering theory [4] for the Thomas–Fermi and Lenz–Jensen potentials, and with distributions corresponding to $r^{-n}$ power potentials calculated following the multiple scattering formalism of the same reference.

Monte Carlo simulations of the angular dependence of the energy loss have been made assuming a random atom distribution in the solid, $r^{-n}$ scattering potentials, and a Gaussian-like foil thickness distribution. As will be shown below, a very good reproduction of the experimental data can be achieved with these simulations.

We also present a simplified model containing the main physical features of the process, which reproduces the experimental data with a surprisingly good degree of accuracy. These calculations take into account the variations with observation angle of the energy loss, due to: variations of the electronic energy loss through changes in the path lengths, the effect of foil roughnesses, and the variations of the nuclear energy loss with the scattering angle.

2. Experimental procedure

The proton beam was generated by a small accelerator with a hot discharge ion source followed by focusing and accelerating stages, a Wien filter for mass selection, electrostatic beam steerers, and an 18° deflector to get rid of neutral particles. The beam-energy spread is as small as
some eVs, and the intensity is variable between $10^{-13}$ and $10^{-9}$ A/mm² and it is very stable. The target holder was preceded by two diaphragms of 4 and 1 mm diameter respectively separated by 58 cm, defining the angular divergence to less than 0.4° and the beam spot size on the target to 1 mm diameter. The multiple target holder allowing foil removal during the experiment, and a rotatable 127° cylindrical electrostatic energy analyzer with the detection system, were located in the same chamber. The energy and angular resolutions of the analyzer were respectively 1% and ±0.58°. A combination of a sorption pump for the preliminary vacuum and a diffusion pump with a UHV-suitable LN₂ trap maintained an oil-free high vacuum.

The self-supported targets were made by evaporation under clean vacuum conditions on a very smooth plastic substrate [29] which is subsequently dissolved. The foil thicknesses were determined by matching the proton energy-loss measurements at 9 keV to previous stopping cross-section $S$ determinations [30], which were based on comparisons with absolute empirical values at higher energies [31] 1. The values obtained were 20.0 nm for Al and 14.3 nm for Au. The roughness has been determined through a simple in situ procedure based on measurements of the energy straggling $\Omega$ of protons transmitted through the foil. For Gaussian foil thickness distributions this straggling can be expressed as [32]

$$\Omega = \sqrt{\Omega_0^2 + \left(\frac{\Delta E}{x}\right)^2} \approx \sqrt{\Omega_0^2 + \rho^2 \Delta E^2}, \quad (1)$$

where $\Omega_0$ is the energy straggling of an ideal foil of thickness $\langle x \rangle$, $\sigma_x$ the standard thickness deviation, $\Delta E$ the energy loss and $\rho$ is a parameter characterizing the foil roughness, defined as $\rho = \sigma_x/\langle x \rangle$. Taking the experimental energy loss values $\Delta E(\theta)$ at $\theta = 0$, and using in Eq. (1) the theoretical $\Omega_0$-values, $\Omega_0$(Al) = 140 eV and $\Omega_0$(Au) = 164 eV, resulting from [6], we extracted the following approximate values of the roughness parameter: $\rho$(Al) = 15% and $\rho$(Au) = 11%. These values have been checked by analyzing a foil of the same production batch with an atomic force microscope.

A TEM analysis previously made on similar targets [33] revealed a polycrystalline structure of random orientation with grain size of ~20 nm.

The foil thickening due to ion beam bombardment was held within insignificant limits by using a low current density of ~$10^{-10}$ A/mm², and short irradiation times. This was checked by comparison of the energy loss at the beginning and the end of the measurement series.

The energy loss at large observation angles $\theta$ is very sensitive to departures of the foil orientation from orthogonality to the incoming beam direction. This was carefully adjusted and checked by comparison of a spectrum corresponding to a large $\theta$ with its similar corresponding to $-\theta$.

The energy spectra for the different observation angles $\theta$ have been measured maintaining a constant incident number of projectiles for each value of the angle. So these measurements allowed the determination of the angular distributions by integration of the individual energy spectra.

The energies of the distributions have been determined by fitting them with Gaussians and taking its central value. Given the nearly Gaussian shape of body of the measured distributions, the resulting values are very close (within ~10 eV) to the most probable value. The differences between these values and the mean values are also small (~10–25 eV depending on the spectra, over a total energy loss of about 1400 eV), which is less than the experimental uncertainties. The energy losses $\Delta E(\theta)$ are obtained by subtracting the energies of the corresponding distributions determined in the preceding way, from the incident beam energy.

### 3. Models and computer simulations

#### 3.1. Angular distributions

Using the Sigmund and Winterbon formalism [4] we calculated the angular distributions for $r^{-n}$ power potentials varying the $n$ values and search-
ing the best fit to our experimental data. In this fitting procedure the large angle tails of the distributions were not considered because of the breakdown of an assumption required for the validity of the formalism, namely the equivalence of travelled path length and penetrated depth.

The scaled angular distribution of a particle beam after traversing a thin layer is given by

\[ f_1(\tau, \tilde{x}) = \int_0^\infty z \, J_0(\tilde{x}z) \exp[-\tau \Delta(z)] \]

in terms of the reduced thickness and angle units

\[ \tau = \frac{Ea}{2Z_1Z_2e^2}, \quad \tilde{x} = \frac{Ea}{2Z_1Z_2e^2} x, \]

where \( a = 0.8853 a_0/(Z_1^{2/3} + Z_2^{2/3})^{1/2} \) is the screening radius, \( N \) the atomic density, \( x \) the penetrated depth, \( E \) the ion energy, \( Z_1 \) and \( Z_2 \) the atomic numbers of the ion and target, respectively, and \( e \) the elementary charge. In Eq. (2) \( J_0 \) is the zero-order Bessel function of the first kind, \( z \) is an integration variable and

\[ \Delta(z) = \int_0^\infty d\tilde{\phi} \frac{f(\tilde{\phi})}{\tilde{\phi}^2} \left[ 1 - J_0(z\tilde{\phi}) \right]. \]

For a power potential \( V(r) \propto r^{-n} \) the scattering function is analytically given by \( f(\tilde{\phi}) = \tilde{\phi}^{1-2/n} \) [34], where \( \lambda \) is a numerical constant evaluated following [34,35]. For this kind of potential one obtains

\[ \Delta(z) = cz^{2/n}, \quad c = -\frac{\lambda \Gamma(-1/n)}{2^{2/n+1} \Gamma(1+1/n)}. \]

This formalism was also used to calculate the derivatives of the multiple scattering function with respect to the foil thickness, necessary for the evaluation of the foil roughness effect as it will be described below.

### 3.2. Energy loss as a function of observation angle

#### 3.2.1. Monte Carlo simulations

Monte Carlo simulations of the energy loss as a function of the observation angle have been performed following the scheme of [36,37] using the binary collision model. A random distribution of atoms in the solid is assumed. In this case, one has a random probability \( k_a \) of occurrence of a collision, related to the total cross-section \( \sigma_{tot} \) and to the travelled distance between collisions \( D \) by the expression

\[ k_a = 1 - \exp(-N\sigma_{tot}D), \]

\( \sigma_{tot} \) being the total scattering cross-section and \( N \) the atomic density. In reduced units [34] of path length, energy and mass, this expression is

\[ k_a = 1 - \exp[-(M_1 + M_2)J_{tot}\xi/4\mu], \]

where \( \xi \) is the reduced travelled distance between collisions and \( J_{tot} = (r_0/a)^2 \) is the reduced total scattering cross-section, with \( r_0 = 0.562N^{-1/3} \) being half the distance between neighboring atoms in an fcc lattice. With Eq. (6), \( \xi \) can be written in terms of the random number \( k_a \) as

\[ \xi = -\frac{4\mu}{M_1 + M_2} \ln(1 - k_a)/J_{tot}. \]

An electronic energy loss proportional to this travelled path length was included, taking the \( \Delta E/\Delta x \) values corresponding to a zero observation angle previously measured in this laboratory [30].

Using a second random number \( k_b \), the reduced scattering angle \( \tilde{\phi} \) in each collision is obtained through the expression [36,37]

\[ \tilde{\phi} = J^{-1}[J(\xi) + (k_b - 1)J_{tot}], \]

where \( J \) is the reduced scattering cross-section, \( \xi \) the reduced energy, and \( J^{-1} \) the inverse function of \( J \).

The reduced cross-section was derived following the Lindhard formalism [34] using an \( r^{-n} \) potential extracted from a fit of the measured angular distribution.

In terms of the center of mass scattering angle \( \phi \), the elastic energy loss \( Q_{nucl}(\phi) \) is

\[ Q_{nucl}(\phi) = 4 \frac{M_1M_2}{(M_1 + M_2)^2} E \sin^2(\phi/2). \]

The effect of foil roughness was included in this simulation by adding a random thickness generator, reproducing the Gaussian-like target thickness distribution with a roughness coefficient \( \rho \) similar...
to the one determined experimentally, as previously described.

3.2.2. Simple model calculation

Guided by the results of the Monte Carlo simulations we present here a simple model which includes the main physical features relevant for the angular dependence of the energy loss $\Delta E$. Although a rigorous treatment requires a more elaborated theory [10,11], this model leads to a remarkably good reproduction of the experimental $\Delta E(\theta) - \Delta E(0)$ data. These calculations include the following three contributions to the $\Delta E$-variation with the observation angle $\theta$:
1. Changes in the electronic energy loss due to path length increase.
2. The angular dependence of the nuclear scattering.
3. The effect of foil roughness.

The path length increase with the observation angle (originated in the multiple scattering process) is estimated by the simple expression
$$\frac{1}{2} \left( \frac{1}{\cos(\theta)} - 1 \right)$$
which, although it is not derived from multiple scattering theory, behaves closely like the multiple scattering path length enlargement, as can be observed from our own Monte Carlo simulations, as well as the simulations of [38].

Under these assumptions the variation of the electronic energy loss can then be written as
$$\Delta E_{\text{elec}}(\theta) - \Delta E_{\text{elec}}(0) = \frac{1}{2} \left( \frac{1}{\cos(\theta)} - 1 \right) \Delta E_{\text{elec}}(0),$$
where $\Delta E_{\text{elec}}(0)$ is the electronic energy loss measured in the forward direction.

The nuclear (elastic) contribution for $M_1 \ll M_2$, is approximated by
$$\Delta E_{\text{nucl}}(\theta) - \Delta E_{\text{nucl}}(0) \approx \Delta E_{\text{nucl}}(\theta) \approx 4 \frac{M_1 M_2}{(M_1 + M_2)^2} E \sin^2(\theta/2).$$

As described in the introduction, the roughness effect arises from the preferential scattering onto small angles of the projectiles traversing thinner sectors, and therefore losing less energy than projectiles collected at larger angles. Following [21] the foil roughness effect is given by
$$\Delta E(\theta)_{\text{rough}} - \Delta E(0)_{\text{rough}} = \Delta E \rho^2 \left( \frac{\partial \ln | f_1(\tau, \theta)|}{\partial \ln \tau} - \frac{\partial \ln | f_1(\tau, 0)|}{\partial \ln \tau} \right),$$
which requires a previous knowledge of the variation of the multiple scattering function with the reduced foil thickness $\tau$. $\Delta E$ is the angular average of the energy loss, which we evaluated with our angular distributions and $\Delta E(\theta)$ data.

The total variation of the energy loss $\Delta E(\theta) - \Delta E(0)$ with observation angle $\theta$ is given by the sum of Eqs. (9)–(11).

4. Results and discussion

Figs. 1 and 2 show the measured energy spectra for different observation angles for Al and Au targets of 20 and 14.3 nm thickness respectively together with the fitted Gaussians. In both cases...
one can clearly observe a shift of the distributions towards lower energy with increasing observation angle. The angular distributions resulting from the integration of these spectra are depicted in Figs. 3 and 4. In these figures we also show the well-known tabulated values of multiple scattering distributions from [4] corresponding to Lenz-Jensen and Thomas–Fermi potentials. We observe that the experimental distributions are narrower than those calculated with the Thomas–Fermi and Lenz-Jensen potentials especially for Au. A similar discrepancy was observed by other authors at higher energy experiments [39,40].

The solid lines in Figs. 3 and 4 show the calculations of the angular distributions resulting from the best fitting power potential \( r^{-n} \). The most suitable powers \( n \) were determined to be 2 for Al and 2.8 for Au. This adjusted potential was used in the previously discussed numerical simulations and to evaluate the foil roughness effect through Eq. (11).

Figs. 5 and 6 show the energy losses as a function of the angle of emergence \( \Delta E(\theta) - \Delta E(0) \) referred to the energy loss at \( \theta = 0 \), corresponding to the same data of the previous figures. A common feature of the depicted angular dependences is the important increase for small angles followed by a change in slope and a further increase at large angles.

In these figures, one can also observe the results of the previously mentioned model calculation, showing separately the contributions of Eqs. (9)–(11). One can observe a remarkably good agreement with the experimental data using the previously specified \( \rho \) values. In spite of its simplicity, this model calculation clearly shows that the first
increase at small angles is mainly due to foil roughness, whereas the stronger increase at large angles results as the composed effect of path length enlargement and growing elastic energy loss.

The Monte Carlo simulations depicted in these figures show also a remarkably good agreement with the experimental data. We have performed a set of simulations with different roughness coefficients in order to test its influence, and have confirmed that the initial increase of $\Delta E(\theta) - \Delta E(0)$ at angles $\lesssim 10^\circ$ is due to the effect of foil roughness. Figs. 7 and 8 show path length enlargements and elastic energy losses, resulting from the Monte Carlo simulations compared to our simplified model. A very good agreement can be observed.

We point out that for hydrogen ions, neither these simulations nor the simple model include an impact parameter dependent single-scattering inelastic energy loss as required for higher energies [7–14]. So the good agreement with the experimental data indicates that if there were an impact parameter dependence of such a single-scattering inelastic energy loss in the case of protons, its effect would be too small to be observable in the present low-energy experiment.
5. Concluding remarks

Energy distributions of 9 keV protons after traversing thin Al and Au foil have been determined as a function of exit angle between 0° and 40° and associated model calculations and Monte Carlo simulations have been performed. From the preceding analysis one concludes that:

1. The measured angular distributions can be well reproduced with multiple scattering functions corresponding to 1/r m potentials, within the range of validity of the small-angle formalism given in [3,4] (without excluding the possibility of using other potential forms to fit the experiments).

2. Within the precision of our measurements there is no evidence of an impact parameter dependence of the electronic energy loss at the low energy of these experiments.

3. The angular dependence of ΔE at small angles is dominated by foil roughness effects.

4. The angular dependence of ΔE at larger angles is essentially due to path length enlargements and to the increase of the elastic contribution.

5. Our Monte Carlo simulations reproduce very closely the angular dependence of the energy loss.

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