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Angular dispersion of protons passing through thin metallic films

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Abstract

The angular distributions of protons after traversing thin polycrystalline Al targets (~15 nm) with an incident energy of 10 keV have been measured and an analysis of the targets by means of transmission electron microscopy (TEM) techniques has been made. The separate influence of the different crystal characteristics and defects has been evaluated by numerical simulation considering the interaction of the ion with all the nearest neighboring atoms simultaneously. In the analysis we included the evaluation of the effects of lattice vibrations, oxide layers and foil roughness on the angular distributions. Previous experimental data in monocrystalline and polycrystalline Au targets has also been analyzed. For a consistency check a comparison with the results of the MARLOWE code for the simpler case of proton channeling in ⟨1 0 0⟩ Al has been performed. As in the case of Au, the present results indicate that the experiments can be explained in terms of a modified Moliere potential, and confirm the critical influence of crystal characteristics, in this case the amorphous oxide layer on the surface and the thermal vibration of the lattice atoms.

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1. Introduction

The penetration of swift ions in matter is determined mainly by two phenomena: the energy loss and the angular dispersion. In this work we investigate some aspects of the latter process.

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The angular dispersion of swift ions in solids, mainly determined by multiple scattering of the ions by the atomic cores of the target, has been investigated since the twenties of the past century [1–3]. The first approaches to describe the multiple scattering (MS) were based on the transport theory [1]. Scott published an extensive review of the advances obtained from the multiple scattering theory in the small angle approximation [4]. Later on, Meyer [5] presented a more general approach, also for small angles, which was further developed by Sigmund and Winterbon [6]. However, these approaches are not adequate to describe effects arising from the crystalline structure of the solids. One of these effects, channeling, manifests as an anomalously long range of the ions, and has been observed by Davies [7] and confirmed by computer simulations by Robinson and Oen [8]. The basic theory of this phenomenon has been presented by Lindhard [9] and a complete review has been published by Gemmell [10].

When describing the angular dispersion of ions in polycrystals, these are generally treated as random material and the MS formalism is used. However, important discrepancies have been found when doing so [11]. At low energies additional complications arise because of the limitations implicit in the MS model, such as the non-applicability of the small angle approximation. Therefore the availability of theoretical models is even more critical in this region.

The aim of this work is to improve the understanding of the passage of low energy protons through polycrystalline targets. For this purpose we made measurements of angular distributions of protons in thin polycrystalline Al foils (15 nm), and study crystalline effects such as channeling and different crystal defects and foil characteristics by computer simulations with an own code which does not rely on the binary collision approximation. This code contemplates different atomic crystalline structures of the target and also layers of random atoms. We also apply the simulations to previously investigated polycrystalline and monocrystalline Au targets [12].

In order to understand the phenomena on a realistic basis we made an analysis of the targets by means of transmission electron microscopy (TEM) techniques.

In the following sections we sketch the experimental method, describe the numerical simulations and present the results.

2. Experimental method

The proton beam was generated in a hot discharge ion-source followed by electrostatic acceleration, steering and focusing stages, and a Wien filter for mass selection. Neutral particles were eliminated by electrostatic bending of the beam. Some additional features has been described previously [13]. The angular analysis has been made rotating an electron multiplier detector with angular resolution of 1.9°. Self-supported polycrystalline targets were elaborated by evaporation under clean vacuum conditions on a very smooth plastic substrate [14] which was subsequently dissolved. The resulting foils are very smooth as was previously determined by an ion-beam technique [15]. The foil thickness (15.9 nm) was determined by measuring the proton energy-loss at 200 keV and using the stopping power values from the Andersen and Ziegler tables [16]. The reason of choosing 200 keV for this purpose is the higher precision of the tabulated stopping power at higher energies.

3. Simulations

The paths of the ions traversing the crystal are calculated considering the ion situated in a central cell of a cluster of $3 \times 3 \times 3$ fcc cells. The resultant force of the interaction of the ion and all the surrounding atoms inside this cluster, up to a distance equivalent to one lattice constant, is derived from the potentials. In this way the simultaneous interaction of the ion with up to 21 atoms is considered. The trajectory of the ion is obtained by the discretization of the standard kinematic equations into small time steps. Each time the ion exits the central cell of the cluster a new slab of 3×3 cells is generated in the corresponding direction. The backward slab of cells is eliminated and therefore the projec-

tile is always in the central cell of a $3 \times 3 \times 3$ cluster.

The different starting points of the projectile when it hits the foil are simulated randomizing the initial position inside the central cell of the first cluster. In the case of a polycrystal the different random orientations of the microcrystals are simulated by randomizing the initial direction of the projectile relative to a coordinate system coincident with the principal axes of the crystal. When the ion travels through two or more overlapped microcrystals the direction of the new microcrystal is again simulated randomly changing the ion direction.

The angular distributions are obtained recording the emergence angle of each ion after a number of time steps corresponding to the target thickness. To obtain a good statistics we calculate 5×10^5 trajectories per spectrum in the case of channeling and in the case of polycrystals, where the angular dispersion is larger, we increment this number to 10^6.

We checked our simulation method comparing results for a simple case (channeling) with those obtained with the MARLOWE code for protons of different energies traversing 10 nm aluminum and gold foils, both using the same standard Moliere potential. The results for Al are displayed in Fig. 1. Some differences can be observed, which arise from the different approaches. Our results show some structure and a stronger dependence with the projectile energy.

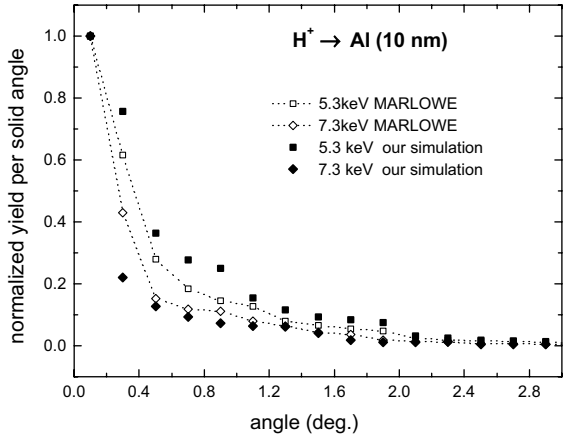

Fig. 1. Comparison of the results with the MARLOWE code and our simulations for the case of proton channeling in Al⟨100⟩ excluding thermal vibrations. For these calculations a standard Moliere potential has been used.

4. Results

4.1. Aluminum

Fig. 2 shows the experimental angular distribution for 10 keV protons on a polycrystalline Al target of 15.9 nm thickness together with the simulation results. To make a realistic simulation we analyze the incidence of different crystal characteristics present in our experiments, such as defects, surface impurities and lattice vibrations. The target has been analyzed by transmission electron microscopy, which showed that it was composed of randomly oriented microcrystals of irregular shapes and more or less homogenous dimensions in the plane of the foil (~25 nm), somewhat larger than the thickness of the target. That is to say a foil consisted of a single layer of microcrystals. This contrasts with our previously analyzed Au foils where grain sizes in the range of 5–40 nm lead to regions with crystallite superposition.

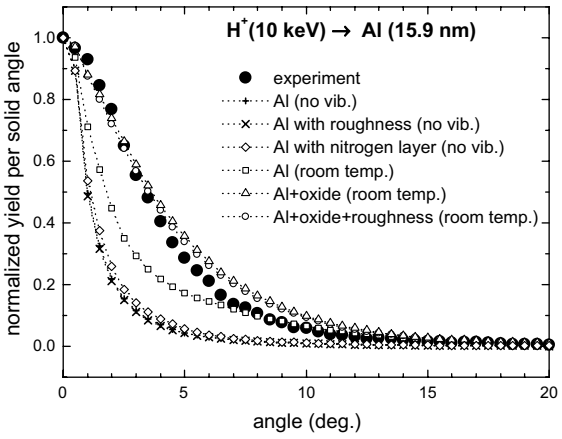

Fig. 2. Experimental angular distribution and simulation results including the analysis of the influence of thermal lattice vibration, N_2 and O_2Al_3 surface layers and effect of target roughness. The open circles show the results considering roughness in the polycrystalline Al layer and in the oxide layer. The simulations neglecting lattice vibrations are indicated as "no vib."

In this analysis of the influence of crystal characteristics we also include the basic phenomenon of channeling and the analysis of defects appearing in Au foils we have previously investigated [12].

A particular feature of the aluminum is the existence of thin oxide layers on the surfaces whose effect has also been studied.

The different effects illustrated in Fig. 2 can be successfully analyzed using a modified Moliere potential, as will be discussed in the following sections.

Calculations with other standard potentials like $1/r^n$, Lenz-Jensen, Thomas Fermi in the Sommerfeld approximation and ZBL have been performed, however when introducing the lattice structure these potentials do not yield good results.

4.2. Channeling

As a fraction of the microcrystals which constitute the target is aligned in channeling directions this phenomenon has been analyzed in more detail on the basis of our previous data [12]. Fig. 3 shows the simulation results of angular distributions for channeled 10 keV protons in a 10 nm monocrystalline gold foil oriented in the $\langle 100 \rangle$ direction and the corresponding experimental values [12]. For comparison with the experiment the simulation results have been convoluted with the detector acceptance. As in the present case of polycrystalline Al, a reasonable agreement was achieved after incrementing the screening distance of the Moliere potential by 80%. However, we consider this modification in the screening distance as tentative and subject for further investigations.

Fig. 3 also shows the simulation curves for a thin (10 nm) single layer of tiny crystals with random orientation. As expected they are significantly broader than the channeling curve. We additionally have included the multiple scattering function for an amorphous target of the same thickness. One can observe that the curve corresponding to a polycrystal curve is narrower than that corresponding to multiple scattering. We assume that this is caused by coherence effects produced by the regular lattice array of target atoms [11], an effect that is not included in the standard MS theory.

In order to prove the consistence of our simulation results we also simulated the experiments performed by Machlin et al. [17] and obtained a very good agreement with their measurements modifying Moliere's screening distance in the same percentage we had used to simulate our experiments. The results are shown in Fig. 4.

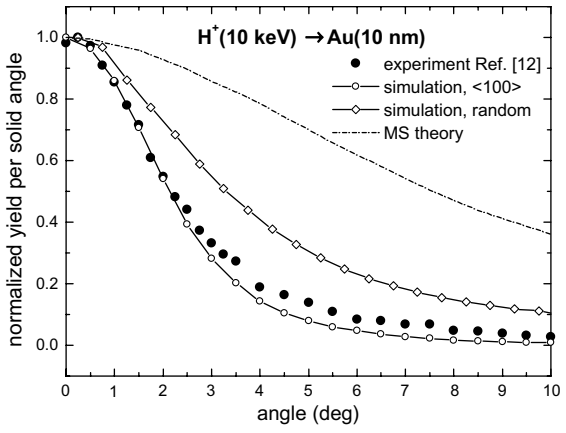

Fig. 3. Comparison of the simulated distributions resulting from a 10 nm thick $\langle 100 \rangle$ monocrystal layer, a 10 nm polycrystal layer with random orientated crystallites, including thermal vibrations, and an amorphous layer of the same thickness. In this case a multiple scattering calculation with the formalism of [6] and a standard Moliere potential is displayed. The solid circles represent the experimental data of [12].

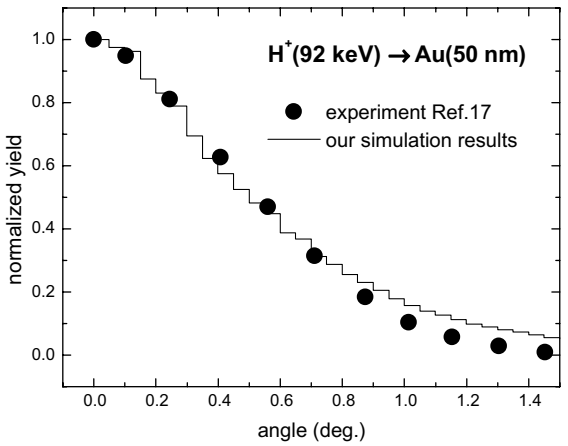

Fig. 4. Experimental angular distribution from [17] for 92 keV proton channelled through a $\langle 0,0,1 \rangle$ monocrystal gold film, together with our simulation results.

4.3. Effects of oxides, target roughness, thermal vibrations and crystal disorder

Aluminum exposed to atmosphere is covered by a thin oxide layer of approximately 2 nm thickness [18]. This oxide has been included in the simulations as an amorphous Al_2O_3 layer on each side of an 11.9 nm Al foil. The effect of these layers can be seen in Fig. 2 which also shows the results for pure polycrystalline Al of 15.9 nm. As can be seen the oxide leads to an appreciable broadening of the distribution.

We also evaluated the possible influence of nitrogen molecules adsorbed on the target surface by introducing an amorphous monomolecular layer at each side of the foil. The result can be observed in Fig. 2, which shows a small, but not completely negligible effect.

The effect of a typical foil roughness was also analyzed considering a Gaussian target thickness distribution with a standard deviation of 13% respect to the mean foil thickness. The net effect is a slight reduction of the angular dispersion when the roughness is considered as distributed on the polycrystal and the oxide layer. However, a negligible effect results when the roughness is assumed to be on the polycrystal only. A more complete analysis of these effects is shown in Figs. 2 and 5.

Another crystal characteristic which has to be taken into account is the thermal vibration of the lattice atoms. They were included in the simulations adjusting the amplitude to make our results compatible with MARLOWE code results for channeling. The thermal vibration amplitude resulting this way is 1.06×10^{-1} a.u. for Al. Figs. 2 and 5 show the influence of thermal vibrations in the case of polycrystals composed by 1 or 2 overlapped crystal layers with different orientation.

Here we also include the evaluation of the effect of twins, the most frequent crystal defect present in the gold foil used in our previous measurement. Twins are interfaces characterized by a high angle tilt (70° in this case) of the atomic rows, generating two rotated crystal regions with specular symmetry. Fig. 6 shows the influence of twins on angular dispersion when the ions traverse sequences of 1–4 twin interfaces inside of a single Au microcrystal of random orientation. The TEM analysis of Al foils reveals a negligible amount of twin layers, so that they are not relevant in this case.

The impurity concentration both in Au and Al crystals are less than 10^{-4} so that the effect on the proton scattering can be neglected.

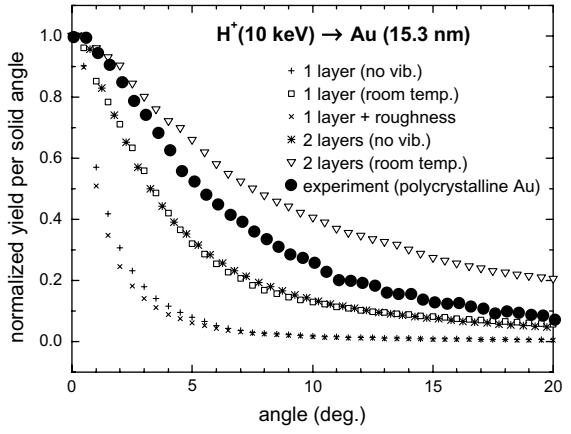

Fig. 5. Simulation results of the effects of thermal vibrations and foil roughness in a polycrystal composed by 1 or 2 overlapped random oriented crystallite layers. Roughness has been evaluated in the case of 1 random oriented crystal layer at room temperature considering a value of 12%. The solid circles represent the experimental data of [12].

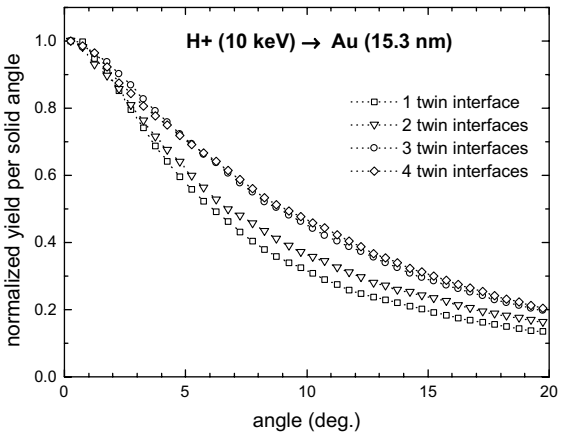

Fig. 6. Simulation results of the effects of twin interfaces in a single layer polycrystal of random oriented crystallites at room temperature. The different curves correspond to sequences of 1, 2, 3 and 4 twin interfaces.

Including the discussed features in the simulations of the Al target we achieve a fairly good accordance with the experiments as shown in Fig. 2. In conclusion, these simulations show the importance of thermal vibrations of the lattice atoms and of the oxide layers on the surfaces, while the influence of the foil roughness is found to be very small.

5. Concluding remarks

We measured angular distributions of low energy protons in thin polycrystalline Al foils and studied the effects of different crystal characteristics found in the foils of the present measurements and in the Au foil previously studied, and observed the following:

- The simulations of the angular distributions in ideal polycrystalline Al and Au foils using a standard potential are narrower than the experimental values.
- From an extensive simulations study we find that the oxide layers present on the surfaces of aluminum, as well as the thermal lattice vibrations lead to a significant broadening of the distributions, while the influence of foil roughness is found to be very small.
- In the case of Al targets the widths of the distributions arising from numerical simulation fit the experiment if the screening length of the employed Moliere potential is modified as previously done for Au foils, and observed crystal characteristics such as oxide layers and thermal lattice vibrations are considered.

Acknowledgements

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References

[1] W. Bothe, Z. Phys. 5 (1921) 63.
[2] G. Wentzel, Ann. Phys. 69 (1922) 335.
[3] J. Lindhard, V. Nielsen, M. Scharff, Mat. Fys. Medd. Dan. Vid. Selsk. 36 (10) (1968).
[4] W.T. Scott, Rev. Mod. Phys. 35 (1963) 231.
[5] L. Meyer, Phys. Stat. Sol. B 44 (1971) 253.
[6] P. Sigmund, K. Winterbon, Nucl. Instr. and Meth. 119 (1974) 541.
[7] J.A. Davies, J. Friesen, J.D. McIntyre, Can. J. Chem. 38 (1960) 1526.
[8] M.T. Robinson, O.S. Oen, Appl. Phys. Lett. 2 (1963) 30.
[9] J.L. Lindhard, Mat. Fys. Medd. 34 (14) (1965).
[10] D.S. Gemmell, Rev. Mod. Phys. 46 (1974) 129.
[11] H.H. Andersen, J. Bottiger, H. Knudsen, P. Moller-Petersen, Phys. Rev. A 10 (1974) 1568.
[12] C. Archubi, C. Denton, J.C. Eckardt, G.H. Lantschner, F. Lovey, J. Valdés, C. Parra, F. Zappa, N.R. Arista, Phys. Stat. Solidi (b) 241 (2004) 2389.
[13] M. Famá, G.H. Lantschner, J.C. Eckardt, C.D. Denton, N.R. Arista, Nucl. Instr. and Meth. B 164–165 (2000) 241.
[14] A. Valenzuela, J.C. Eckardt, Rev. Sci. Instr. 42 (1971) 127.
[15] J.C. Eckardt, G.H. Lantschner, Thin Solid Films 249 (1994) 11.
[16] H.H. Andersen, J.F. Ziegler, in: J.F. Ziegler (Ed.), Stopping and Ranges of Ions in Matter, Vol. 3, Pergamon, New York, 1977.
[17] E.S. Machlin, S. Petraglia, Nuovo Cimento LVB (1) (1968) 263;
E.S. Machlin, Phys. Lett. A 24 (1967) 173;
E.S. Machlin, S. Petralia, A. Desalvo, R. Rosa, Fl. Zignani, Philos. Mag. 22 (1970) 101.
[18] N. Cabrera, N.F. Mott, Rep. Prog. Phys. 12 (1949) 163.