Early History of Ion Beam Physics

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Abstract

In order to understand better the physics of radiation damage due to fast-neutron recoil atoms in nuclear reactors, Dr. W.B. Lewis (the research director at Chalk River Nuclear Laboratories) suggested to me in 1956 that we should investigate the range of 0–100 keV ions in solids. Soon afterwards, a Harwell report on Ar and Kr trapping and release during implantation (later published by Carter et al., 1962) caught Dr. Lewis's attantion and re-enforced his belief in the need for 0–100 keV ion ranges in solids. Hence, for almost 50 years, I have wandered through the field of atomic collisions in solids and have collaborated with many of the Ion06 participants – especially with Ingmar Bergström, Peter Sigmund (our host), Hans Henrik Andersen, Len Feldman, Jens Ulrik Andersen, Preben Hvelplund, Jim Williams, Bruce Winterbon. Over the years, we have established strong bonds of friendship and have had a lot of fun together. Tonight, I shall enjoy sharing with you some personal reminiscences on this early history. Since time permits, I shall also outline two of the many unsolved problems we have encountered.

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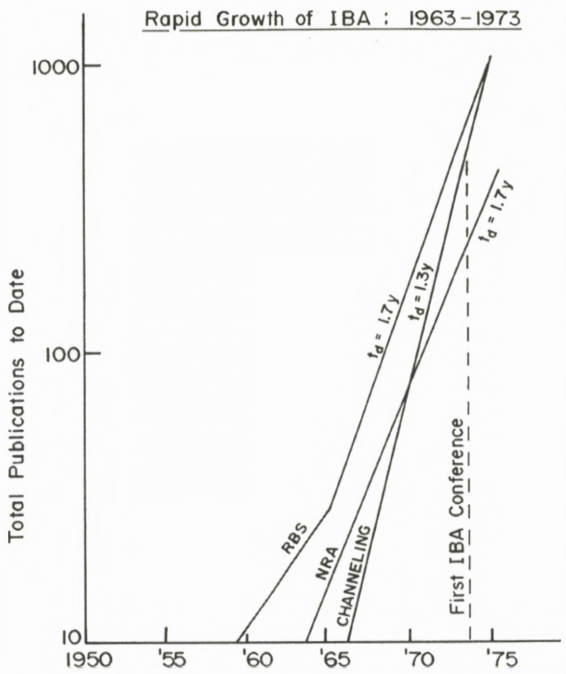

Figure 1. Publications growth rate in nuclear reaction analysis (NRA), Rutherford backscattering (RBS) and channeling. Adapted from Bujdoso et al. (1982).

1. Introduction

Figure 1 (Bujdoso et al., 1982) depicts the extremely rapid growth that occurred during the 1960s in three important sections of the field: namely, in Rutherford scattering, channeling, and nuclear reaction techniques. In each section, the total number of publications in the literature grew from less than 10 papers in the early 60s to more than 1000 by 1970: i.e., with doubling times under two years! By the end of the 60s, ion implantation of Si had become the major driving force in the field. However, prior to 1965 there was very little interest from the semiconductor field, apart from a couple of unsuccessful doping attempts by Ohl (1952) and by Kingsbury and Ohl (1952) at Bell Laboratories.

Indeed, the major interest in ion-beam physics came initially from two quite different sources: (i) Nuclear reactor sites such as Chalk River (F. Brown, J.A. Davies, G. Sims, J. Whitton), Oak Ridge (M. Robinson, O.S. Oen, S. Datz), Brookhaven (C. Erginsoy), Harwell (M.W. Thompson, R.S. Nelson), Aachen (G. Leibfried, C. Lehmann, P. Sigmund), ISPRA (Hj. Matzke) and Garching (R.

Behrisch, R. Sizmann, H. Lutz), where the primary motive was to study the physics of fast-neutron recoils; and (ii) European nuclear spectroscopy groups such as the Bohr Institute (N. Bohr, G. Sidenius, Skilbreid), and Oersted Institute (J. Koch) in Copenhagen, Aarhus University (J. Lindhard, K.O. Nielsen), the Nobel Institute of Physics in Stockholm (I. Bergström, B. Domeij, L. Eriksson), Chalmers Institute in Goteborg, (O. Almen, G. Bruce), the FOM Institute in Amsterdam (J. Kistemaker, P. Rol, J. Fluit) and Orsay in Paris (R. Bernas), who collaborated (after World War II) in developing low-energy (~50 keV) heavy-ion accelerators with high mass resolution, known as electromagnetic isotope separators. Their main objective was to prepare radioactive targets for nuclear spectroscopy. This latter group also had a keen interest in many related problems of ion-beam physics, such as sputtering, target stability, ion ranges and ion-source development. They even initiated their own conference series, with meetings in Harwell (1955), Amsterdam (1957), Vienna (1960), Orsay (1962) and Aarhus (1965).

Until the mid 1960s, there was almost no overlap between these two scientific communities.

2. Pre-1962 History

Theoretical work in ion-beam physics goes back to Bohr's (1948) comprehensive monograph on atomic particle penetration through matter, and to Lindhard's "Notes on Atomic Collisions" series (Lindhard and Scharf, 1961; Lindhard et al., 1963a, 1963b). In the nuclear reactor community, theoretical work was carried out mainly at Aachen and Jülich by Leibfried and his students, Lehmann and Sigmund, and at Oak Ridge by Robinson and Oen. Peter Sigmund moved from Aachen to Denmark at an early stage of his career, firstly in 1962 at the Danish Nuclear Reactor centre Riso where (in collaboration with H.H. Andersen) he initiated his lifelong interest in radiation effects, and then in 1964 at the Institute of Physics in Aarhus. His subsequent theoretical contributions to sputtering, scattering and energy-loss processes are well known to all of us.

Experimental work on ion ranges goes back to the 1957 publications[1] of Baulch and Duncan (1957) who studied the range of ~100-keV α-recoil atoms in gases, and of Bredov and Okuneva (1957) who used chemical etching to obtain range profiles of radioactive ^{137}Cs atoms in Ge (Figure 2). Bredov compared his experimental distribution with that predicted using Bohr's exponentially screened potential – and obtained rough agreement. However, at such low energies (4 keV),

[1] The 1956 Rutherford Backscattering study by K.O. Nielsen was never published – except in his doctoral thesis – and did not come to our attention until my first visit to Scandinavia in 1962.

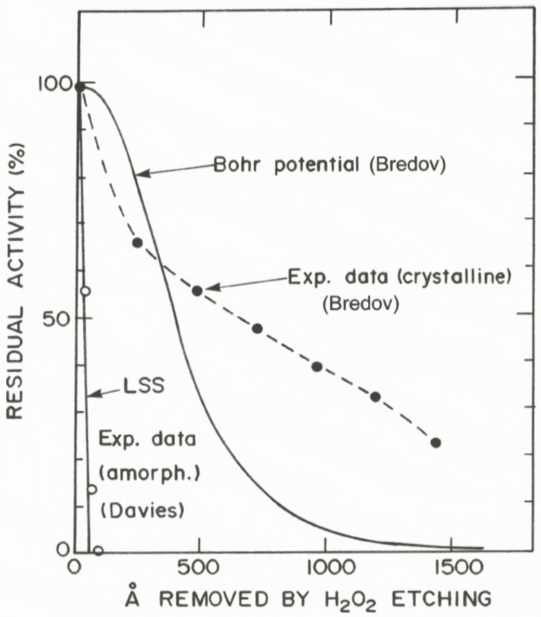

Figure 2. Depth profile of 4-keV ^{137}Cs in Ge. Adapted from Davies et al. (1962). Experimental data: • Bredov; ○ Davies. Theoretical curves based on the Bohr potential and on Lindhard et al.'s (1963b) Thomas–Fermi treatment.

Lindhard et al. (1963b) had shown[2] that Thomas–Fermi screening was more appropriate – and would predict much shorter ranges. At that time, we suspected that Bredov's chemical etching technique might be at fault. Hence, one of my early range measurements (Davies et al., 1962) was to repeat his ^{137}Cs in Ge experiment using our own special two-step transmission technique: namely, (i) deposit thin Ge films of various thicknesses on thick Al targets and implant 4-keV ^{137}Cs ions into each film; (ii) dissolve the Ge film completely in aqueous H_2O_2 (which does not attack the underlying Al) and measure the residual radioactivity.

Our data agreed well with Lindhard et al. (1963b) and hence we blamed Bredov's deep penetration on poor experimental technique. However, Bredov had used single-crystal Ge, whereas our evaporated films were probably amorphous. In hindsight, Bredov's (1957) result may even have been evidence of channeling, which he had failed to recognize because his theoretical estimate used a much too strongly screened potential.

[2] Although Lindhard did not publish this work until 1963, some of my nuclear spectroscopy colleagues at Chalk River were already familiar with his work as early as 1958.

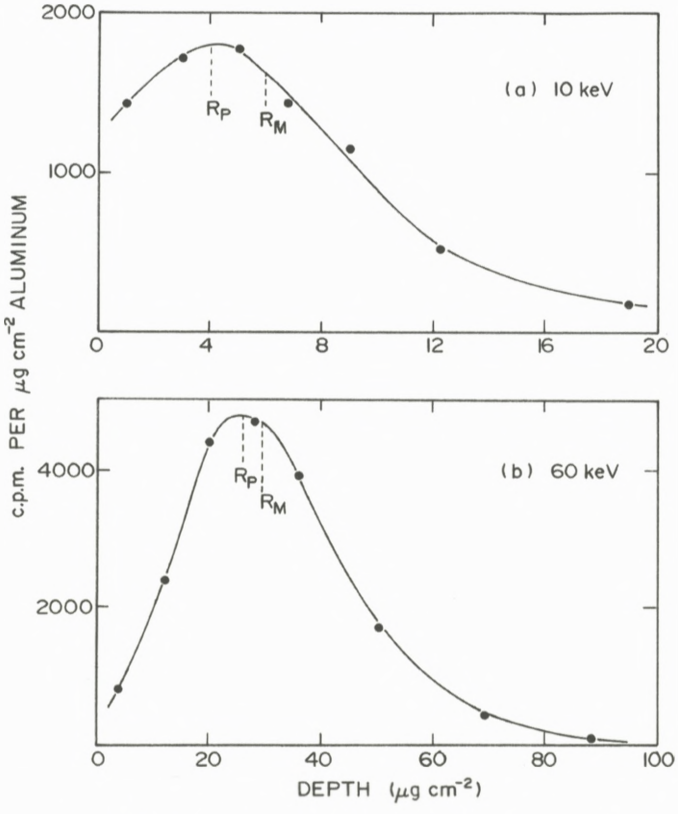

Figure 3. Depth profiles of ^{24}Na in polycrystalline Al (Davies and Sims, 1961). The peak position R_P represents the "most probable" value of the range, whereas R_M is the "median" value at which 50% of the implanted beam has been stopped. The prominent "tail", especially in the 10-keV case, encouraged Robinson and Oen (1963) to make Monte Carlo simulations of channeling.

Our early range studies (Davies and Sims, 1961; Brown and Davies, 1961; Davies et al., 1963; McCargo et al., 1963) consisted in developing a two-step anodic oxidation/stripping technique for obtaining detailed depth profiles in Al, W and (later) Si. In each of these targets, very uniform oxide films are prepared by anodic oxidation, with thicknesses varying linearly with applied voltage from a few atomic layers up to several hundred. In each case, a suitable solvent exists which rapidly dissolves the oxide layer, without attacking the underlying metal. Hence, by repeating this two-step process, a detailed depth profile is obtained. Two typical range profiles are shown in Figure 3.

In each case, the mean range agreed well with LSS theory, but a small fraction ($\sim$10%) penetrated much deeper than predicted. These small "tails" eventually led Robinson and Oen (1963) to postulate a channeling mechanism in the occasionally aligned polycrystalline grains. But at Chalk River, our thinking was influenced strongly by the neutron-diffraction concepts of Brockhouse and Iyengar (1958). Since the wavelength of a 60-keV Na beam is only $\sim 10^{-6}$ nm, the corresponding Bragg angle (θ_{Bragg}) is very small ($\sim 10^{-6}$ radians) and *diffraction* effects are therefore negligible.

In 1960, a mini-conference on keV ion ranges was held at Chalk River, in which the Powers and Schmitt groups both participated. Powers and Whaling (1962) reported on the use of proton backscattering to measure range profiles in high-dose implants of $\sim$100-keV nitrogen and oxygen ions. Schmitt and Sharp (1958) and VanLint et al. (1961) presented mean-range estimates derived by measuring the escape fraction of radioactive keV and sub-keV recoils produced in thin foils by (γ, n) nuclear reactions.

Sputtering is another field that was investigated extensively in these early years, especially in Amsterdam and Goteborg. In the early 1960s, Rol et al. (1960) and Fluit and Rol (1964) reported that sputtering yields in monocrystalline copper exhibited a strong orientation dependence. Initially, this was attributed to a geometrical "transparency" effect, because sputtering is largely a near-surface phenomenon. Almen and Bruce (1961a, 1961b) published two papers, documenting an exceptionally detailed study of sputtering behaviour of some 25 different metals bombarded by 45-keV Kr ions (Figure 4). Particularly noteworthy was their observation of a periodic dependence of sputtering yield on the atomic number Z_2 of the target. Large peaks in sputtering yield occur around Z_2 values of 30 (Zn), 48 (Cd) and 80 (Hg) which correspond nicely with minima in their binding energy U_s. Two metals, Mg and Al, exhibit anomalously low sputtering yields. However, years later, the Amsterdam group found that this could be attributed to the presence of protective surface oxides with much higher binding energies; under ultra-high vacuum conditions, the sputtering yield increased suddenly by almost a factor of ten when the surface oxide was sputtered away.

The year 1961 was a memorable one for two reasons: (i) the Chalk River nuclear spectroscopy group purchased a 70-keV isotope separator with excellent mass resolution (1:4000) from a Swedish company (this instrument was an ideal accelerator for our radioactive implantations); and (ii) Ingmar Bergström arranged to visit us for 2 months, since the new accelerator was an exact copy of his own isotope separator in Stockholm. His visit established the first major bridge between the two scientific communities – our nuclear reactor group and the European nuclear spectroscopists – and a remarkably fruitful interaction was

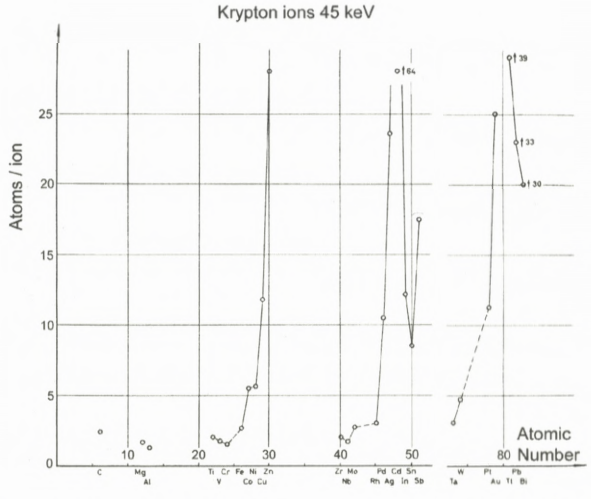

Figure 4. Sputtering yield Y_S as a function of target atomic number Z_2 for 45-keV Kr bombardment (Almen and Bruce, 1961a).

the result. Ingmar helped us to broaden the Chalk River program to include other aspects of ion-beam physics such as dose effects, sputtering and thermal stability of implanted targets (Bergström et al., 1963). He also invited me to Stockholm for 3 months to collaborate with his student Bo Domeij, en route to the 1962 isotope separator meeting in Orsay – he even persuaded my research director to finance my whole trip! During this Stockholm visit, he introduced me to many of his Scandinavian colleagues: O. Almen in Goteborg, G. Sidenius at the Bohr Institute, and especially K.O. Nielsen and J. Lindhard in Aarhus. Karl Ove Nielsen had only just arrived in Aarhus in 1962 as the newly appointed professor of experimental physics; Jens Lindhard on the other hand had been there already for 5–6 years. This resulted in my spending the year 1964–1965 in Aarhus, collaborating closely with several Danish graduate students, two of whom (J.U. Andersen and P. Hvelplund) are participants at Ion06 (J.U. Andersen and P. Hvelplund), and also with another well known "foreign guest" Peter Sigmund. But, by 1964 channeling had already been "discovered" – so let us first go back and complete our history of the pre-channeling era.

Other early studies of ion-beam physics include Sidenius' nuclear stopping cross-section data in gas targets (Bohr Institute) and Loftager's later work in Aarhus in which large peaks in the nuclear stopping cross-section were observed whenever the collision distance corresponded to an inner-shell overlap.

Electronic stopping of heavy ions was investigated by several experimental groups. Ormrod and Duckworth (1963) measured energy loss in transmission through thin carbon foils and observed an unexpected periodic dependence on the atomic number Z_1 of the ion beam. This Z_1 oscillation effect was later extended to higher energies and heavier ion beams by Fastrup et al. (1965) and by Hvelplund and Fastrup (1968). Using gas targets and single-collision coincidence techniques, Afrosimov et al. (1963) in Leningrad and Kessel et al. (1965) at the University of Connecticut reported sharp steps in the inelastic energy-loss Q, as successive inner shells overlap during the collision – i.e., firstly, L-L overlap, then K-L, and finally K-K. Eventually, this work led to the discovery of high-energy molecular X-ray bands (Saris et al., 1972).

A particularly memorable milestone in the field of electronic stopping was the discovery of the surface-barrier detector by Mayer and Gossick (Mayer and Gossick, 1956; Mayer, 1959). With this new energy-dispersive spectrometer, an entire energy spectrum could be obtained simultaneously in a single measurement (i.e., within minutes), whereas the old cumbersome magnetic spectrometers had required hours or even days of step-by-step data collection. Initially, the physics community failed to show much interest in this new detector. However, in September 1960, this was rectified when the Asveville workshop on Semiconductor Detectors brought together almost all the future pioneers of ion-beam analysis – Georges Amsel, Walter Brown, Geoff. Dearnaley, Walt Gibson, Jim Mayer, Laurie Miller.

For many of us, one puzzling aspect of the Si solid-state detector was the unexpectedly large value of 3.67 eV per electron/hole pair, i.e., more than three times the Si band gap energy E_i of 1.1 eV. In Ge, this discrepancy between ε and E_i was even larger, i.e., $\varepsilon_{\mathrm{Ge}} = 3.7$ eV whereas E_i is only 0.66 eV. However, a few years ago, Len Feldman came across a 1961 paper by Shockley (1961) which contains a surprisingly simple and accurate explanation for these large ε values, namely:

$$\varepsilon = E_i + 2E_{\mathrm{f}} + rE_{\mathrm{R}}, \tag{1}$$

where E_{f} ($\sim 0.6E_i$) is the mean final energy of an electron or hole when it can no longer create additional electron/hole pairs. The Raman phonon energies E_{R} (0.063 eV in Si, and 0.037 eV in Ge) were obtained from neutron scattering data (Brockhouse and Iyengar, 1958; Palevsky et al., 1959), and the mean-free-path ratio $r = L_{\mathrm{ionization}}/L_{\mathrm{phonon}}$ was obtained from the quantum yield measurements of Vavilov (1959). Substituting $r_{\mathrm{Si}} = 17.5$ and $r_{\mathrm{Ge}} = 57$ in Equation (1), we obtain:

$$\varepsilon_{\mathrm{Si}} = 2.2E_i + 1.10 = 3.5 \text{ eV}, \tag{2a}$$

$$\varepsilon_{\mathrm{Ge}} = 2.2E_i + 2.11 = 3.6 \text{ eV}. \tag{2b}$$

Note the good agreement with the experimental ε values of 3.67 eV in Si and 3.7 eV in Ge. Note too the similarity in magnitude between the first term $2.2E_i$ in Equation (2) and the modified Kinchin–Pease relationship (Sigmund, 1969) between the energy $(-\Delta E)$ to create a Frenkel pair and the displacement energy E_{d} in a collision cascade; namely, $(-\Delta E) = 2.38E_{\mathrm{d}}$.

We shall return later to a still unresolved aspect of solid-state detectors, namely why do high-energy heavy ions exhibit almost the same ε value as protons and electrons?

3. 1962–1965: Channeling

A detailed account of the "channeling story" has already been published (Davies et al., 1992) in the proceedings of the 10th Ion Beam Analysis conference in Eindhoven. A few personal reminiscences are recalled here.

In March 1962, just before my departure for Stockholm, Robinson and Oen (whom I had not yet even met) phoned from Oak Ridge to inform us that their computer program could successfully simulate the "tails" in our polycrystalline range profiles – and attributed them to a coulombic steering process, occurring in those crystalline grains which accidentally had a low-index direction aligned with the incident beam direction. At first, they named their steering process "tunneling" but, to avoid confusion with quantum mechanical tunneling, they soon changed the name to "channeling". Hence, by the time I arrived in Stockholm in 1962, we were already growing single crystals of Al and W in order to verify the existence of channeling.

During my Stockholm visit, Bent Elbek (Bohr Institute) gave a nuclear physics seminar at the Nobel Institute of Physics. While I no longer recall even the title of his seminar, I still vividly remember our midnight discussions, while sharing the Institute's top-floor guest apartment. We discussed the possible origin of the exponential "tails" in all the Chalk River range profiles – and suddenly Elbek recalled that K.O. Nielsen (1956) at the Bohr Institute had used helium backscattering plus a magnetic spectrometer to measure *in situ* the range distribution of 40-keV Gd in polycrystalline Al (Nielsen, 1956). His resulting depth profile (Figure 5) was an excellent Gaussian peak, with absolutely *no* evidence of a deeply penetrating "tail", even down at the 10^{-3} level.

This posed quite a dilemma! Various experimental artifacts might be able to *create* a spurious tail, but it is hard to imagine an artifact that could cause a real tail to disappear. Fortunately, the next morning Bo Domeij, who was collaborating in some range-profile measurements during my Stockholm visit, proposed

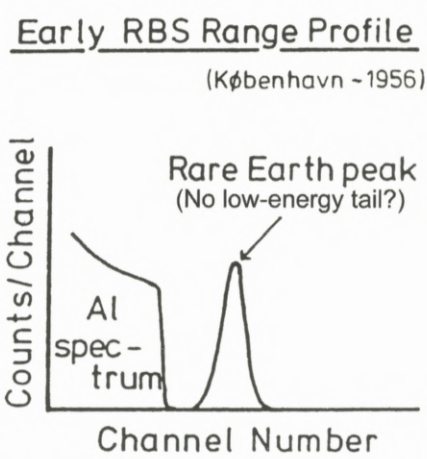

Figure 5. K.O. Nielsen's RBS spectrum of 40-keV Gd in polycrystalline Al, using a magnetic spectrometer (Nielsen, 1956).

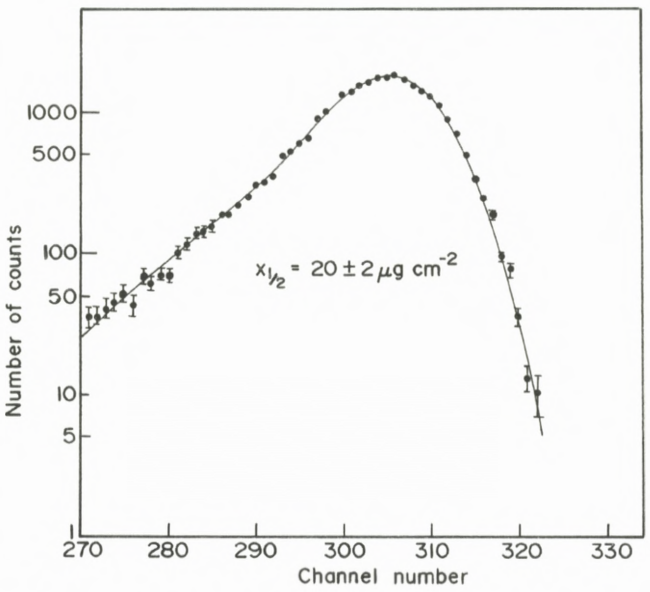

Figure 6. α-particle spectrum from 210-keV ^{222}Rn, implanted into polycrystalline Al. The resulting $x_{1/2}$ for the exponential "tail" agrees well with the value of 17 $\pm$ 2 μg/cm^{-2} obtained subsequently by the anodic oxidation/stripping technique (Domeij et al., 1963).

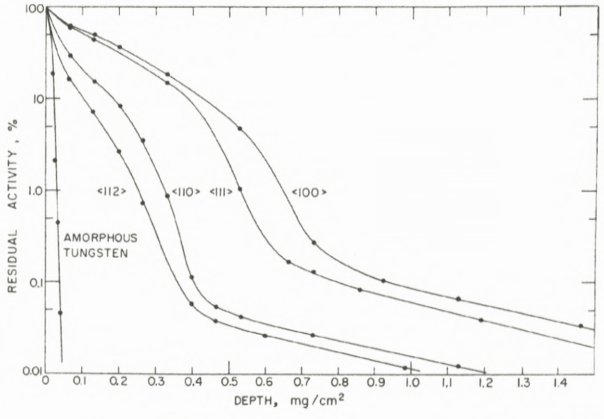

Figure 7. Integral penetration profiles of 40-keV ^{125}Xe, implanted into single-crystal W along various crystallographic directions (Domeij et al., 1964a).

an ingenious experiment to resolve the problem. We would implant ^{222}Rn into polycrystalline Al and determine the resulting depth profile by two independent methods: (i) measure the energy spectrum of the emitted α-particles, using one of the newly developed solid-state detectors; and (ii) use the anodizing/stripping technique on the same implanted target. The α-spectrum (Figure 6) verified the existence of a small penetrating "tail". Furthermore, both techniques exhibited the same magnitude and $x_{1/2}$ values for the exponential tail of the profile (Domeij et al., 1963).

A few months after my return to Canada, Robinson and Oen invited a small group of us to Oak Ridge to discuss their computer range simulations. By then, single crystals had finally become available – and range profiles in aligned monocrystalline tungsten (Figure 7), aluminum (Piercy et al., 1964), silicon (Davies et al., 1964) and copper (Lutz and Sizmann, 1963) soon confirmed the existence of channeling. Also, Domeij et al. (1964b) measured profiles in amorphous Al_2O_3 and WO_3 and, as would be expected, found no tail.

In September 1963, a small conference was held at Chalk River to discuss the significance of these single crystal results. Despite its small size (less than 25 participants), all major groups were represented, which indicates how small the ion-beam physics community was 43 years ago. By the end of 1963, channeling had been observed, not only in heavy-ion range profiles, but also in MeV proton-transmission experiments through Si crystals (Figure 8) by Dearnaley (1964) and through very thin Au crystals Nelson and Thompson (1963).

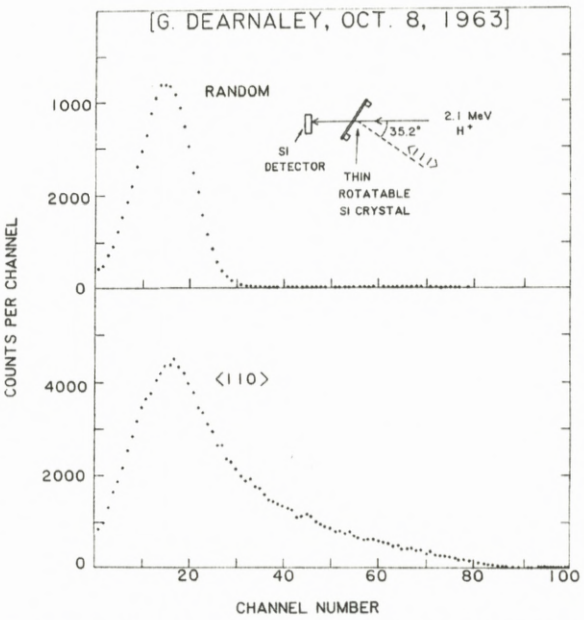

Figure 8. Energy spectrum of 2.1 MeV protons, after transmission through a 30 μm Si (111) crystal at 35.2° tilt from the ⟨111⟩ axis: (a) random incidence; (b) incidence along an ⟨110⟩ axis (Dearnaley, 1964).

In 1964–1965, I was a guest scientist with K.O. Nielsen's accelerator group in Aarhus. During my visit, Lindhard (1965) developed and published his definitive theoretical treatise on channeling, indicating the unique role of "close-impact" processes in measuring quantitatively the non-channeled component of the beam. Experimental verification of Lindhard's concepts soon followed, using nuclear reactions (Andersen et al., 1965), Rutherford scattering (Bogh and Uggerhoj, 1965) and inner-shell X-rays (Khan et al., 1966, 1967), Lindhard's theoretical paper contained also an elegant proof of reversibility between channeling of an external beam and "blocking" of energetic positive particles emitted (or backscattered) from lattice sites within the crystal. Since Stockholm at that time lacked a suitable MeV accelerator, Domeij (1965) again devised an ingenious ^{222}Rn implantation experiment. He injected ^{222}Rn into tungsten single crystals and used the blocking pattern of the emitted α-particles (Figure 9) to verify the concept of reversibility. His measurements also established that ~80% of the Rn atoms must reside on lattice sites. This was the first use of channeling to study lattice location of embedded foreign atoms (Domeij, 1965).

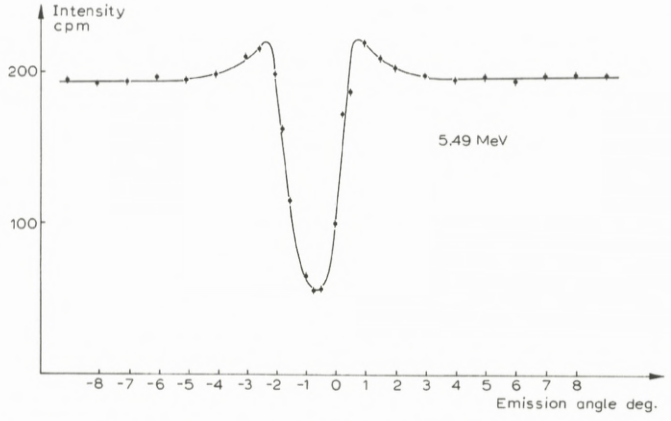

Figure 9. Angular dependence of the 5.49 MeV α-particles emitted from ^{222}Rn, implanted at 60-keV into (111) tungsten (Domeij, 1965).

Other pioneering studiers of MeV ion channeling include early blocking measurements by Tulinov et al. (1965) in the Soviet Union and by Gemmell and Holland (1965) at the Argonne, and also the single-crystal transmission studies of Gibson et al. (1965).

Many of these channeling studies were presented at the Electromagnetic Isotope Separators conference in Aarhus in mid-June 1965 and published in Nucl. Instr. Meth. Vol. 38. More than 50% of the papers at this meeting involved solid-state applications and hence a new biannual conference series (ICACS) was split off (Table 1), with the initial one being held in Chalk River in 1967. At the 1965 Aarhus meeting, less than 10% of the papers involved semiconductor applications. Yet, within two years, a fully dedicated Implantation of Semiconductors conference was held in Grenoble (1967) – and by 1970 this had expanded into another regular conference series. In 1968, the first Gordon conference on Particle-Solid Interactions was held in New Hampshire and this too became a biannual event. In 1973, another ion-beam conference series – Ion Beam Analysis – was initiated at IBM (Yorktown Heights) and the following year the inaugural conference of the Ion Implantation of Metals series was held at Sandia (Albuquerque). With the exception of the Gordon conference, all the biannual conference series in Table 1 are still flourishing.

Table 1. Ion Beam Physics Conferences

1965	ICACS – split off from the Electromagnetic Isotope Separators conference series – biennial.
1967	Grenoble – Ion Implantation of Semiconductors.
1968	Gordon Conference – Particle-Solid Interactions – biennial series, terminated in 1996.
1970	Implantation of Semiconductors – biennial.
1973	Ion Beam Analysis (IBA) – biennial.
1974	Implantation of Metals – biennial.
1978	Ion Beam Modification of Materials (IBMM) – formed by combining the above Implantation conference series (Semiconductors and Metals) – biennial.
1979	Radiation Effects in Insulators – biennial.
1980	Surface Modification of Metals by Ion Beams (SM^2IB) - biennial.

4. Unsolved Problems

This concludes my account of the early history of ion-beam physics. However, since "Unsolved Problems" is one of the main themes of Ion06, let me close by briefly recalling two unsolved problems from these early years: (i) the so-called "supertail" in well-channeled range profiles in tungsten; and (ii) the response of solid-state detectors to high-E heavy ions.

(i) Tungsten "supertails"

In almost all channeled range profiles in W, a small fraction (typically ~0.1%) of the radioactive ions penetrate to extremely large depths (Figure 7), independent of the incident energy (Domeij et al., 1964a, figure 3). Cavid Erginsoy (1964) postulated that perhaps some sort of quantum restriction was preventing the best-channeled ions from undergoing any energy loss. However, Carl Wandel (Aarhus, 1964) made an alternative suggestion that avoided having to invoke any exotic new physics. He pointed out that best-channeled ions probably create *no* displacements near the end of their tracks and hence could diffuse as free interstitial atoms. Perhaps, such interstitial diffusion in tungsten is rapid at room temperature and, if so, then the enhanced motion should occur equally in all three dimensions. Erginsoy's "super-channeling" model on the other hand would enhance the penetration only along the incident beam direction. One of the Aarhus graduate students (P. Jespersgaard) therefore injected ^{133}Xe into a W (111) crystal firstly

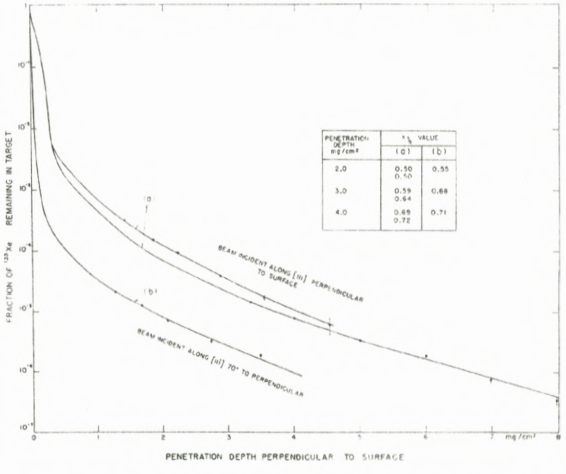

Figure 10. Integral penetration profiles for 20-keV ^{133}Xe in (111) W: (a) beam injected along [111] direction perpendicular to the surface; (b) beam injected along [11$\bar{1}$] direction at 70.7° from the perpendicular (Davies and Jespersgaard, 1966).

along the ⟨111⟩ axis perpendicular to the surface, and secondly along the ⟨111⟩ at 70.7° from the perpendicular (Davies and Jespersgaard, 1966). Note that the channeled depth profile (Figure 10) is reduced ~3-fold for the 70.7° axis, whereas the "supertail" depth scale is unaffected. Hence, a 3-dimensional diffusion-type process, as proposed by Wandel, must be involved.

At 30 K, the supertail is almost two orders of magnitude larger (i.e., about 10% of the beam), indicating that the fraction ending up as interstitial atoms is strongly temperature dependent (see figure 3 in Davies et al., 1968). Furthermore, the supertail can be completely suppressed by using a high-dose Ne bombardment to introduce trapping centres into the crystal before allowing it to warm up from 30 K. This shows that the interstitial diffusion process must be negligible at 30 K, but extremely rapid at room temperature. A similar experiment at 78 K, by Hermann et al. (1966) shows that the interstitial process is still negligible at liq. nitrogen temperature.

One other point: Andersen and Sigmund (1965) predicted that, when $Z_{ion} > Z_{target}$, even the best-channeled ions create displacements near the end of their track – and therefore, would not exhibit a "supertail". Unfortunately, for a high-Z target such as tungsten, there are not many heavier radiotracers available. Nevertheless, Domeij and Eriksson (1965) were able to implant ^{222}Rn and, as predicted, they found no "supertail".

So far, tungsten is the only crystal exhibiting such clear evidence of an interstitial diffusion process. Thus, one unsolved problem is whether channeling can be used to induce similar interstitial diffusion effects in other crystal lattices. The most likely candidate is the chemically similar bcc crystal, Mo, but perhaps other bcc crystals such as Ta and Nb should also be considered. Furthermore, in Si and other elementary semiconductors, group-III dopants (B, Al, etc.) and also Au exhibit a fast-diffusion behaviour that is probably interstitial.

(ii) ε (eV per electron/hole pair) for high-E heavy ions

Contrary to widespread belief, the so-called "pulse-height defect" (PHD) for heavy ions is surprisingly small, or even *negative*, provided the ion energy is higher than the Bragg peak. During the 1960s, electrons, protons, and even He ions were found to exhibit approximately the same ε value, namely 3.67 eV. Furthermore, ε seemed to be independent of ion energy and unaffected by channeling. Heavier ions at MeV energies exhibited somewhat larger ε values, but this was attributed to significant E-loss contributions from nuclear stopping and from the finite window thickness of the detector.

However, by the 1980s, the picture had changed considerably. Careful measurements in several laboratories (Kemper and Fox, 1972; Langley, 1973; Mitchell et al., 1975) showed that, at $E > 2$ MeV, helium ions produce a slightly *larger* pulse height than protons, i.e., the PHD for helium is actually *negative*. After applying small corrections for nuclear stopping and window loss, Lennard and Winterbon (1987) observed a linear dependence of ε on the stopping power, $\mathrm{d}E/\mathrm{d}x$ (MeV/micron), namely,

$$\varepsilon = 3.67 - 0.2\frac{\mathrm{d}E}{\mathrm{d}x}. \tag{3}$$

However, the cause of this $\mathrm{d}E/\mathrm{d}x$ dependence has not yet been established.

Note that Equation (3) would predict a very large negative PHD for very heavy ions. Recently, ε has been measured by Comedi and Davies (1992) and by Weijers et al. (2002), using heavy ions at energies above the Bragg peak value, so that nuclear stopping and window corrections become extremely small (<0.5%). Their observed ε values for a wide range of heavy ions (Table 2) are within a few % of the proton value of 3.67 eV, despite the much denser plasma along the heavy-ion track. A satisfactory explanation for this almost constant ε value has not yet been found. Also, why does the linear $\mathrm{d}E/\mathrm{d}x$ dependence of Equation (3) break down for ions heavier than helium?

Table 2. ε (eV per electron/hole pair) at $E > E_{\text{Bragg}}$.

Ion	E (MeV)	$\mathrm{d}E/\mathrm{d}x$ (MeV/μm)	$\varepsilon_{\text{predicted}}$	$\varepsilon_{\text{observed}}$	Reference
e, γ			3.67	3.67	Lennard and Winterbon (1987)
H	1.0	0.09	3.67	3.67	Lennard and Winterbon (1987)
He	5.0	0.30	3.63	3.64	Lennard and Winterbon (1987)
^{12}C	25	1.20	3.47	3.58	Comedi and Davies (1992)
^{35}Cl	30	4.50	2.80	3.55	Comedi and Davies (1992)
^{32}S	60	3.70	2.90	3.52	Weijers et al. (2002)
^{81}Br	140	9.30	1.80	3.71	Weijers et al. (2002)
^{107}Ag	2000	8.00	2.10	3.70	Weijers et al. (2002)

This seems an appropriate place to end our historical review, having reminded the reader that some unsolved (and hopefully interesting) ion-beam problems still exist.

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