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# ENERGY LOSS OF LIGHT IONS IN DIAMOND

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The energy loss of 2–12 MeV protons, 12–18 MeV alpha particles and 24 MeV lithium-7 ions transmitted through thin (4–18  $\mu\text{m}$ ) diamond crystals has been measured for random incidence and for incidence along the  $\langle 110 \rangle$ ,  $\langle 111 \rangle$  and  $\langle 100 \rangle$  axial channels. The ratio of channelled to random stopping is found to be the same for all three ions at the same velocity. The ratio of the stopping power at the leading edge of the channelled peak to random, taken as a measure of the ratio for best-channelled particles is found to be  $0.64 \pm 0.02$  for the  $\langle 110 \rangle$  axis for 12 MeV protons. The results are compared with several recent theories of channelled energy loss and are found to lie between the predictions of Dettmann and Esbensen and Golovchenko. A theoretical approach using a spatially periodic dielectric function is found to give results close to those of Esbensen and Golovchenko.

## 1. Introduction

The study of the reduced stopping power of channelled ions has received much attention in recent years, both experimentally<sup>1–7</sup>) and theoretically<sup>7–13</sup>). Extensive measurements have been made on silicon and germanium in the MeV energy range<sup>1–7</sup>) and most theoretical studies have made predictions for these crystals.

It is of interest to extend these measurements to diamond, partly because of the similarity in crystal structure and binding to silicon and germanium, but also because a large fraction of the electrons in diamond are valence electrons. In a recent theoretical treatment Dettmann<sup>12</sup>) has treated the valence electrons as a uniform electron gas, with the same contribution to random and channelled stopping, while other authors have taken the spatial periodicity of the valence electron gas into account<sup>8–11</sup>), and found a variation in stopping for channelled and random ions. It has been argued<sup>5</sup>) that this localisation of the valence electrons around the atoms is irrelevant for channelled ions, because the range of interaction of such ions with loosely bound valence electrons is much larger than channel dimensions, and thus that Dettmann's treatment of them as a uniform gas is to be preferred. In diamond, random stopping in the MeV energy range is predominantly due to the four valence electrons, the two core electrons accounting for about 10% of the stopping power, according to Dettmann's theory. In this theory, the reduction in stopping of channelled ions is due to the reduction in the core contribution, and thus the maximum reduction should be  $\sim 10\%$ , with a similar reduction in all axes. Other theories predict a much greater reduction, and a measure-

ment of the stopping power in diamond can provide a test of the validity of the various theories.

In this paper, measurements are reported of the stopping powers of 3–12 MeV protons, 12–18 MeV alpha-particles and 24 MeV Li ions in thin diamond crystals. The results are compared with the first-principle calculation of Dettmann<sup>12</sup>) and of Esbensen and Golovchenko<sup>13</sup>). The results indicate that the localisation of valence electrons should be taken into account in Dettmann's theory. The theory was modified by treating the valence electrons as a spatially periodic electron gas<sup>8</sup>) and an expression for the stopping power of this gas was derived. The modified theory gives predictions similar to those of the theory of Esbensen and Golovchenko; nevertheless both theories underestimate the channelled ion stopping power.

## 2. Experiment

Diamond crystals polished to a thickness of 4–18  $\mu\text{m}$  were obtained from D. Drukker & Zn, Amsterdam. The diamonds were natural type IIa<sup>14</sup>) crystals. It has been shown that polished diamond surfaces are free from damage on a scale that can be measured by channelling<sup>15</sup>). Other methods of preparation considered were ion milling and chemical etching: ion milling proceeds slowly and leads to amorphisation of the near surface region; chemical etching requires the use of oxidising agents at temperatures of  $\sim 1000^\circ\text{C}$  and cannot be easily controlled under these conditions. Thus polishing is the only realistic preparation method, as thin crystals cannot be grown. Nevertheless this is not without its difficulties. Diamond is polished on a cast iron scaife (polishing wheel) at linear velocities of

$\sim 30 \text{ ms}^{-1}$ , and with relatively large loads on the diamond. This cannot be compared with the gentle lapping of other crystals. The crystals obtained varied in thickness by  $\sim 10\%$  over the area of the beam spot, and this uncertainty in the thickness dominates the errors of the experiment. This variation is illustrated in fig. 1 for the two diamonds used for most of the measurements; the photographs were taken with sodium light ( $\lambda = 589 \text{ nm}$ ) and a change of one fringe indicates a variation in thickness of  $122 \text{ nm}$  (refractive index = 2.42). In view of the difficulties associated with the polishing of diamond, the uniformity obtained is a tribute to the diamond polisher's art.

Sample thickness was measured by using an infra-red spectrophotometer and determining the thickness from the oscillations in the observed transmittance<sup>3)</sup>. The value of the refractive index over the range of wavelengths used (3–20  $\mu\text{m}$ ) is

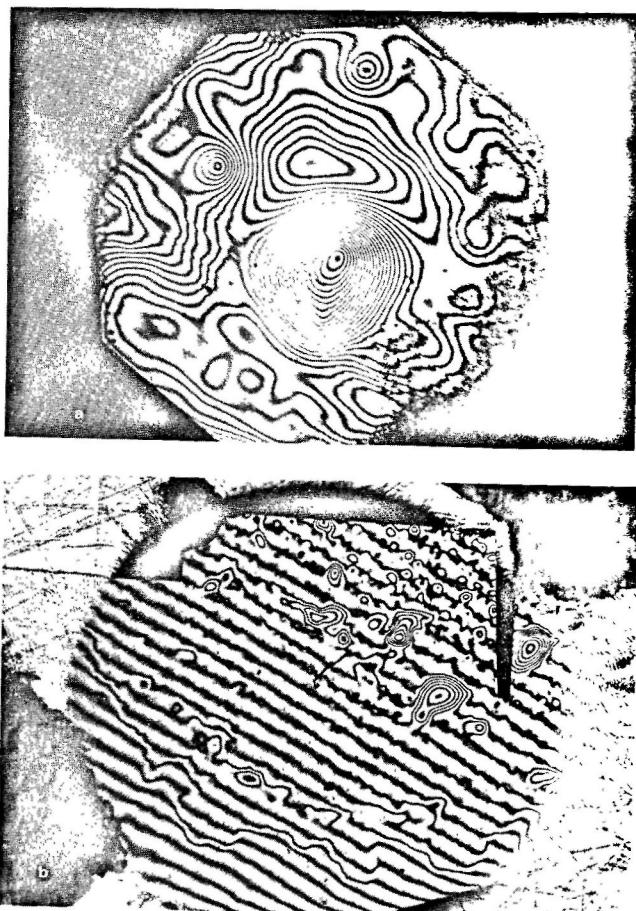


Fig. 1. Interference fringes in the two diamonds used. (a)  $18.7 \mu\text{m}$  diamond, (b)  $4.0 \mu\text{m}$  diamond.

$2.30^{16}$ ). Measurements at selected points on the diamonds were obtained by using monochromatic light to produce a fringe pattern (fig. 1) and then counting the number of fringes moving past a reference point while the wavelength of light from the monochromator was varied. The thickness can then be determined in a similar fashion to infrared determinations.

Incident beams were obtained from the tandem Van de Graaff accelerator of the NPL. The target was mounted on a two axis goniometry which allowed alignment of the target to  $0.01^\circ$ . The incident beam was collimated to a full-angle of  $0.01^\circ$  and passed through the target into a detector at  $0^\circ$  which subtended a solid angle of  $5 \times 10^{-3}$ . A monitoring annular detector, placed at a scattering angle (variable) of  $5^\circ$ – $10^\circ$  aided in aligning the target. Standard electronics were used to amplify and analyse pulses, with a resolution of 15 keV. Beam currents were kept to the order of  $10^{-16} \text{ A}$ .

### 3. Results and discussion

A typical transmitted energy spectrum for channelled protons is shown in fig. 2, together with the random and incident beam spectra. The mean crystal thickness was  $18.7 \mu\text{m}$  with a variation over the beam spot of 7%; the pathlength for  $\langle 110 \rangle$  channelled protons was  $23.4 \mu\text{m}$ . Random spectra were measured at the same angle of incidence as the channelled spectra. Skewed gaussian curves were fitted to the measured energy spectra, and the most probable energy loss determined at the peak of the fitted curves. In addition, for channelled spectra, the leading edge of the peak was determined by using the tangent to the higher energy side of the peak<sup>7)</sup>, as illustrated in fig. 2. It has been suggested<sup>3)</sup> that the leading edge is representative

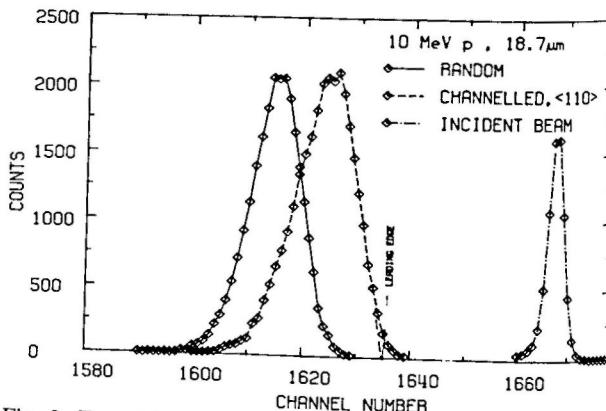


Fig. 2. Transmitted beam spectra.

of the energy of channelled ions with least energy loss, and that the value obtained may be compared with theoretical predictions for this quantity.

Random stopping powers were determined from the measured energy loss  $\Delta E = E_B - E_R$ , where  $E_B$  is the incident beam energy, and  $E_R$  the random peak energy, by

$$\frac{dE}{dx} \Big|_{E=E} = \frac{\Delta E}{\Delta x},$$

where  $\bar{E} = \frac{1}{2}(E_B + E_R)$  is the average energy of the ion in the target and  $\Delta x$  the target thickness. This is a good approximation for small energy loss  $\Delta E$ , and numerical calculations for a worst case show that the error introduced is, for all cases, much less than 1%. The ratio,  $\alpha$ , of the channelled ion stopping power to the random, was determined from

$$\alpha(\bar{E}) = \frac{E_C - E_R}{E_B - E_R},$$

where  $E_C$  is the energy of the channelled ions, either peak or leading edge value.

The resulting random stopping powers are shown in fig. 3 for 3–12 MeV protons, 12–18 MeV alpha-particles and 24 MeV  $^7\text{Li}$  ions. These are incorporated into the same set of axes by plotting  $Z_1^{-2} \cdot dE/dx$  against  $\bar{E}/A$ , where  $Z_1$  is the charge of the incident ion, and  $A$  the atomic mass. The agreement between results on the same diamond for different ions was found to be good. The observed scatter of ~10% is consistent with the uncertainty in target thickness, and errors from other sources amounted to 2%. Also plotted in figure 3 are values obtained from the recent compilation of energy loss data by Ziegler and Andersen<sup>17</sup>. The measured values agree with their recommended

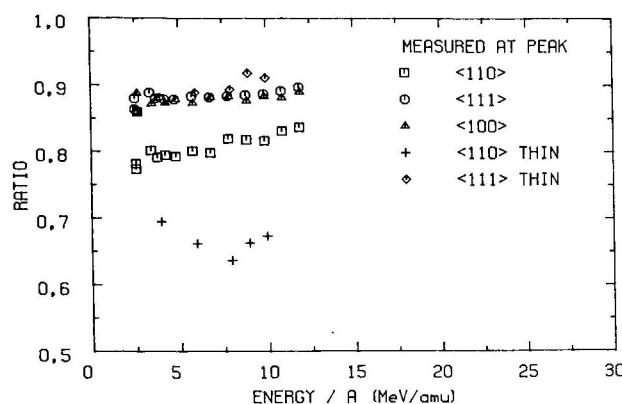


Fig. 4. Ratio of channelled to random stopping power as a function of energy for peak values.

values within the experimental uncertainty. The errors preclude the possibility of determining any deviation from their values due to the allotropic form of carbon used. Such allotropic effects have been observed in stopping in diamond and graphite<sup>18</sup> and graphite and vapour deposited carbon<sup>19</sup>.

The ratio of channelled to random stopping power is shown in fig. 4 for the peak values. The values obtained do not depend on a knowledge of target thickness, and thus a greater degree of precision is possible. Consideration of sources of error including analyser calibration, drift in amplifiers and accuracy in fitting the peaks allow a precision of 0.02 to be assigned to the thick ( $18.7 \mu\text{m}$ ) target values and 0.03 to the thin ( $4.0 \mu\text{m}$ ) target values. The  $\langle 100 \rangle$  and  $\langle 111 \rangle$  axis values for both diamonds are similar. There is an indication that  $\alpha$  is greater for the  $\langle 111 \rangle$  axis than for the  $\langle 100 \rangle$  axis. For the  $\langle 110 \rangle$  axis, the thin diamond values are considerably lower than the thick diamond values. There was no indication of any dependence of  $\alpha$  on  $Z_1$  and the points for the various ions in fig. 4 are not distinguished.

The relation of the energy of the leading edge of the channelled peak to the least energy loss of the channelled ions can only be meaningful if the width of the peak is much larger than the width expected from straggling and instrumental resolution. If this is true, the width can be ascribed to the effect of dechanneling and to variation of the stopping power with ion trajectory. The leading edge is then representative of the ions that have remained close to the channel centre throughout their passage through the crystal, and have experienced the least energy loss. If the width is dominated by straggling and instrumental effects, the least energy loss will

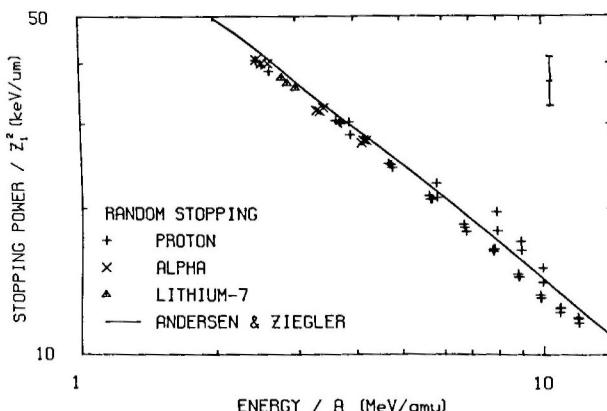


Fig. 3. Random stopping power for all energies, compared with values from the compilation of Andersen and Ziegler<sup>17</sup>.

be more truly represented by the peak of the spectrum, and the leading edge will give a misleadingly low value. In this experiment, the width of the peaks for ion channelled through the thinner diamond were similar to the incident beam widths, and the leading edge values were obtained only for the thicker diamond. These are shown in fig. 5. The values for  $\langle 110 \rangle$  are similar to the peak values obtained for the thin diamond, which is consistent with the argument given above. The results for the other two axes show a fairly pronounced decrease with energy. The peak values for the  $\langle 111 \rangle$  axis obtained with the thinner diamond were similar to the peak values of the thicker diamond, and this indicates that the peak is a better representation of the least energy loss for these ions. The energy dependence would then be an artifact of the analysis, due to the use of leading edge values. The values for  $\alpha$  obtained at lower energies, where dechannelling is more important, then provide a lower limit to the stopping power. Thus for the  $\langle 110 \rangle$  axis a value of  $0.64 \pm 0.02$  for the ratio  $\alpha$  for ions with least energy loss is obtained; for the other axes the position is less certain, but a lower limit of  $\alpha(\langle 111 \rangle) \approx 0.80$  and  $\alpha(\langle 100 \rangle) \approx 0.75$  is indicated.

The results were compared with values obtained for the first-principle theories of Dettmann<sup>12</sup>) and Esbensen and Golovchenko<sup>13</sup>). Both of these theories proceed from a similar generalized oscillator strength, but differ in the use of this to evaluate the stopping power. Dettmann treats the valence electrons as a uniform electron gas, and the core electrons independently in a Hartree-Fock approximation. Esbensen and Golovchenko treat the atom as a whole and obtain an expression that depends on the dipole oscillator strength. This they evaluated in

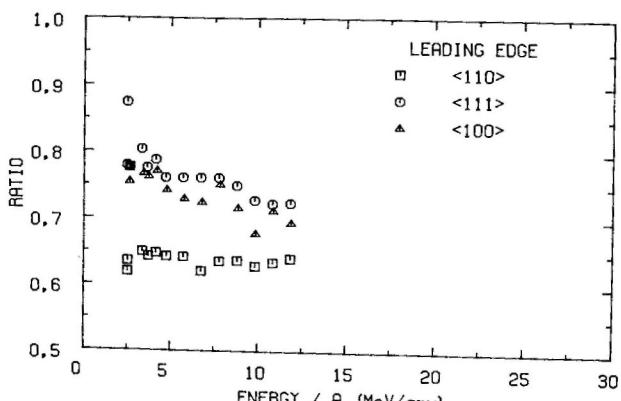


Fig. 5. Ratio of channelled to random stopping power as a function of energy for leading edge values.

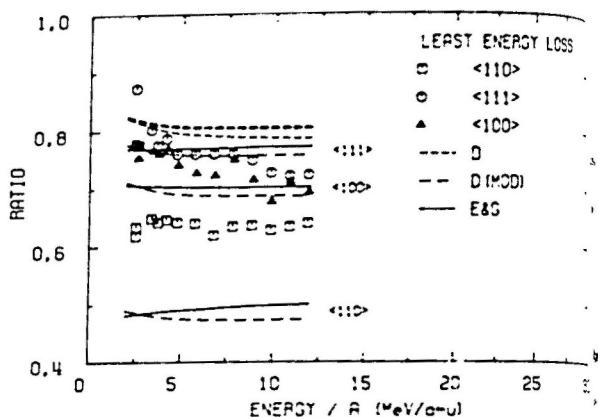


Fig. 6. Comparison with theory - E & G: Esbensen and Golovchenko<sup>13</sup>), D: Dettmann<sup>12</sup>); D(MOD): Dettmann core contribution with eq. (1). The lowest Dettmann curve is  $\langle 110 \rangle$ , and others in increasing order  $\langle 111 \rangle$  and  $\langle 100 \rangle$ .

a local density approximation. No attempt is made to distinguish valence electrons from core electrons and the theory is essentially based on a free atom model.

The stopping powers were evaluated in Dettmann's model using Clementi wave functions<sup>21</sup>) to evaluate the mean excitation energies. These were also used to determine the electron density of the carbon atom for the evaluation of the local density approximation to the expression of Esbensen and Golovchenko. The expression of these authors also requires X-ray structure factors; experimental values were used for these<sup>20</sup>). Both theories predict similar values for the random energy loss, in reasonable agreement with Andersen and Ziegler<sup>11</sup>. The ratios of channelled to random stopping power were determined for the best channelled ions and are shown in fig. 6, together with the experimental leading edge values. The theory of Dettmann predicts closely similar values for all three axes and is clearly not in agreement with experiment, even allowing for uncertainties in the definition of least energy loss. The theory of Esbensen and Golovchenko predicts a large axial dependence, but rather exaggerated when compared with the experimental results. Both theories predict a higher ratio in the  $\langle 111 \rangle$  axis than in the  $\langle 100 \rangle$  in accordance with experiment.

The results indicate that the reduced stopping of channelled ions cannot be explained solely in terms of the reduction of core-electron excitation. Thus localisation of valence electrons should be incorporated into Dettmann's theory. The stopping of ions in a spatially periodic electron gas in a covalent semiconductor has been discussed by Desalvo and

Rosa<sup>10</sup>) who evaluated the stopping power numerically. Using their model we have, with some approximation, evaluated the stopping power using a sum rule<sup>22</sup>) and obtain

$$\frac{dE}{dx} \Big|_{\text{valence}} (\mathbf{r}) = \frac{4\pi Z_1 Z_2 e^2}{m_e v^2} N \sum_{\mathbf{G}} e^{i\mathbf{G} \cdot \mathbf{r}} F_v(\mathbf{G}) \times \\ \times \ln \left\{ \frac{2m_e [v^2 + \sqrt{(v^4 + G^2 v^2 h^2/m_e^2)}]}{h(\omega_g + \sqrt{\omega_g^2 + 4G^2 v^2})} \right\}, \quad (1)$$

where  $v$  is the velocity,  $\mathbf{r}$  its position in the transverse plane,  $N$  the atomic density of the target,  $F_v(\mathbf{G})$  the valence electron structure factor for reciprocal lattice vector  $\mathbf{G}$  and  $h\omega_g = \sqrt{(h^2 \omega_p^2 + E_g^2)}$ , where  $\omega_p$  is the plasmon frequency and  $E_g$  the average energy gap<sup>24</sup>) between valence and conduction bands. The sum is over all vectors  $\mathbf{G}$  normal to the axis. For large  $v$  this reduces to

$$\frac{dE}{dx} \Big|_{\text{valence}} (\mathbf{r}) = \frac{4\pi Z_1 Z_2 e^2}{m_e v^2} N \left[ Z_2 \ln \frac{2m_e v^2}{h\omega_g} + \right. \\ \left. + \sum_{\mathbf{G} \neq 0} e^{i\mathbf{G} \cdot \mathbf{r}} F_v(\mathbf{G}) \ln \frac{2m_e v}{hG} \right], \quad (2)$$

which has also been obtained by Burenkov and Komarov<sup>23</sup>). Valence electron structure factors were obtained by subtracting the core structure factors, calculated from Clementi wave functions<sup>21</sup>), from the experimental structure factors<sup>20</sup>).

The expression (1) was then used for the valence contribution in Dettmann's theory and the results are also shown in fig. 6. The results are similar to those for the theory of Esbensen and Golovchenko. It may be noted that eq. (2) is similar to an expression by these authors [their eq. (23)] which may be regarded as the local density approximation for eq. (2) applied to the atom as a whole. The core electrons contribute only about 10% to the channelled energy loss, and thus a similarity between the two theories is to be expected.

Thus the ratio of channelled to random stopping powers in diamond indicates that the effect of a varying electron density of the valence electrons needs to be considered in Dettmann's theory. Closer examination of the agreement between theory and experiment is not possible without a better estimate of the least energy loss of channelled ions: it is probable that the leading edge of the channelled peak is not necessarily a good measure of this, especially in thin crystals. Nevertheless, the

results show that the theory overestimates the effect of channelling on the stopping power, while predicting the correct axial dependence. The accuracy of the results presented here is limited by the effect of target non-uniformity, and efforts are continuing in an attempt to improve this.

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