

Table 8.2

RANDOM STOPPING POWERS IN DIAMOND

\bar{E} (MeV)	$\frac{dE}{dx}$ (keV/ μ m)	\bar{E} (MeV)	$\frac{dE}{dx}$ (keV/ μ m)
<u>Protons (thick crystal)</u>		<u>Protons (thick crystal)</u>	
2.469	40.68	7.786	16.47
2.537	39.68	7.810	16.31
2.633	38.58	7.843	16.50
3.649	30.50	8.809	14.66
3.721	30.22	8.831	14.47
3.749	30.07	8.863	14.45
4.715	24.84	9.827	13.28
4.771	24.66	9.847	13.10
4.797	24.34	9.876	13.09
5.722	21.28	10.837	12.48
5.756	20.93	10.857	12.24
5.801	20.92	10.881	12.51
6.758	18.56	11.844	11.94
6.787	18.28	11.862	11.86
6.830	17.92	11.890	11.62
<u>Protons (thin crystal)</u>		<u>Protons (thin crystal)</u>	
3.897	28.3	7.950	17.6
3.922	27.8	8.942	16.0
5.923	21.1	8.955	16.0
5.942	20.6	9.948	14.2
7.933	18.4	9.961	13.8
\bar{E} (MeV)	$\frac{\bar{E}}{A}$ (MeV/amu)	$\frac{dE}{dx}$ (keV/ μ m)	$\frac{1}{Z_1^2} \frac{dE}{dx}$ (keV/ μ m)
<u>Alphas (thick crystal)</u>			
9.867	2.467	162.9	40.72
10.132	2.533	159.9	39.98
10.478	2.620	160.0	40.00
13.326	3.332	127.8	31.96
13.511	3.378	127.5	31.88
13.768	3.442	129.5	32.38
16.565	4.141	109.6	27.40
16.704	4.176	111.0	27.75
16.947	4.237	110.7	27.68
<u>${}^7\text{Li}$ (thick crystal)</u>			
19.606	2.801	336.3	37.37
20.175	2.882	327.5	36.39
20.955	2.994	320.2	35.57

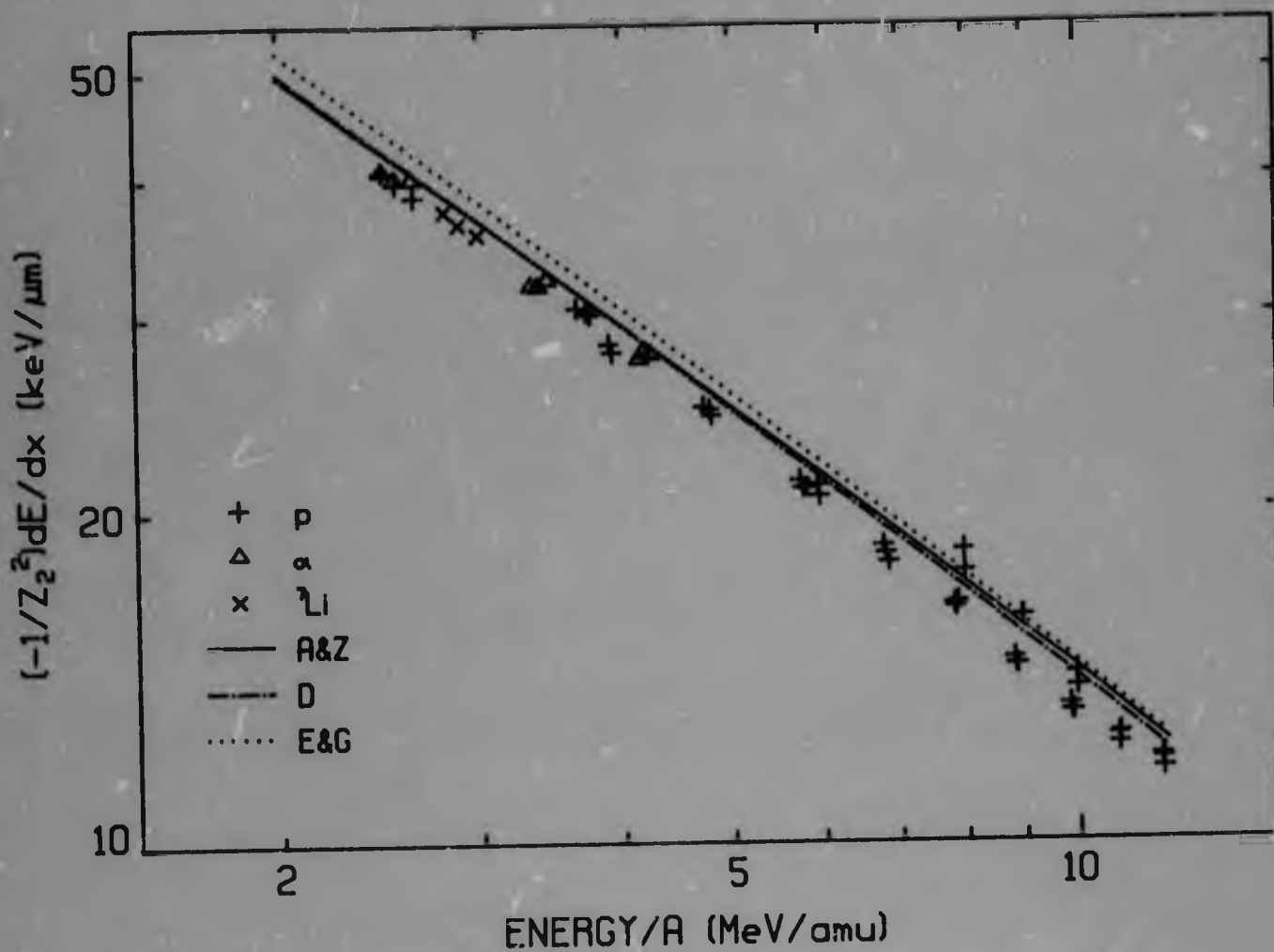


Figure 8.3: Random stopping power of light ions in diamond. A & Z: Andersen and Ziegler, D: Dettmann, E & G: Esbensen and Golovchenko.

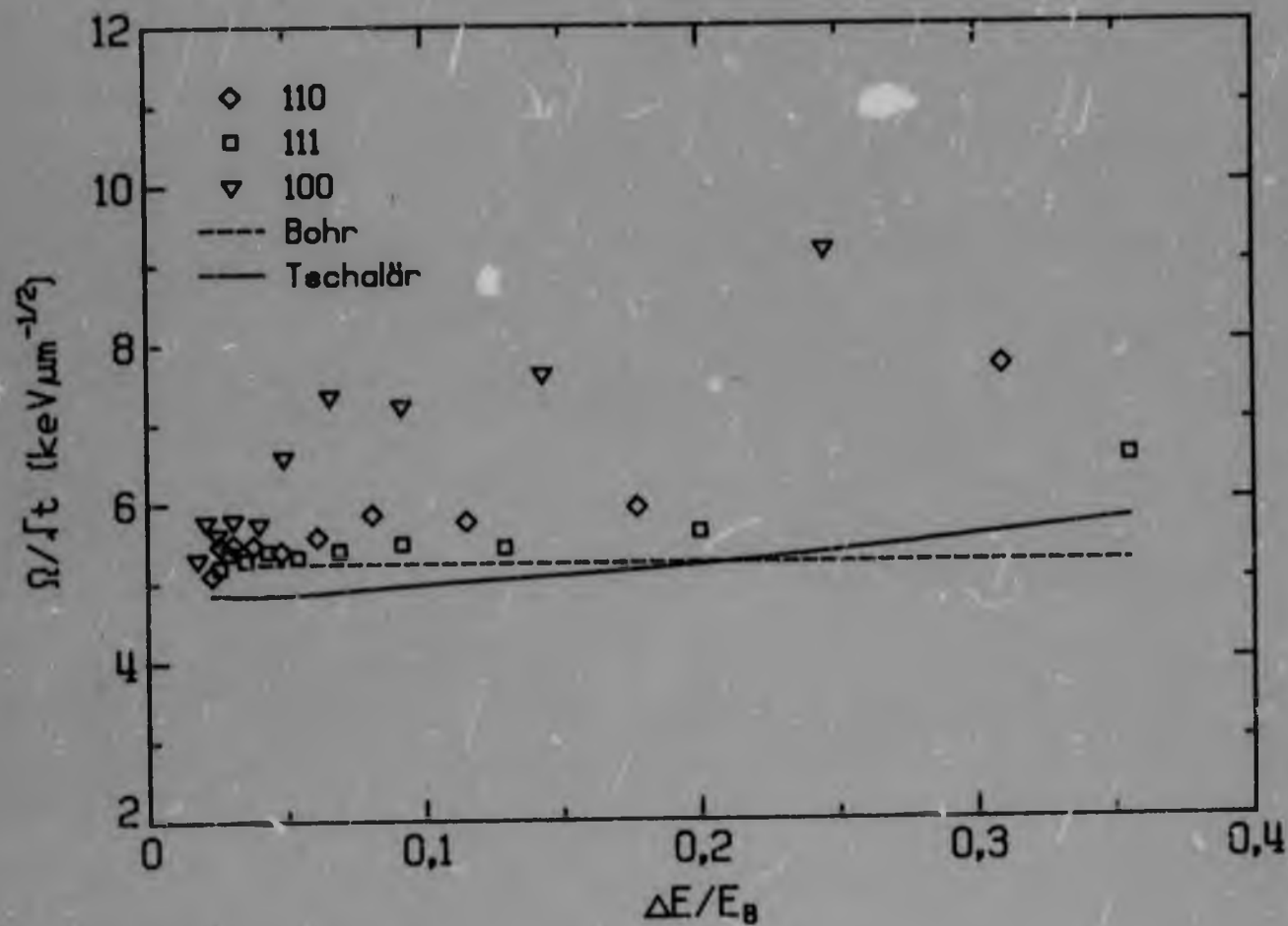


Figure 8.4: Straggling of protons transmitted through thin diamond in random orientations.

and the rate of increase. It is possible that this is due to approximations in the theory, but it is more probable that the experimental points are perturbed by the crystal structure of the target. It was observed that the random peak tended to be skewed towards higher energies, especially for larger relative energy losses in the target, and more so in $\langle 100 \rangle$ where the points lie above the other axes in Figure 8.4. It is probable that some feeding into channels is occurring; this would be expected to be more probable in targets where more multiple scattering can occur, that is, at larger energy loss. This then leads to a lower energy loss for some ions and a skewing of the peaks to higher energies, with a consequent apparent increase in straggling. This is likely to occur in any crystal-line target: it can be concluded that crystals are not the ideal target in which to study straggling at larger relative energy losses. Nevertheless it can be concluded that the Bohr approximation is valid at smaller energy losses, in the Gaussian energy loss distribution regime. Straggling in carbon has also been examined by Langley and Brice [Lal 78] who determined it from the broadening of the resonances at 459 keV and 1.7 MeV in elastic proton scattering. A value close to that predicted by the Bohr theory was found at 1.7 MeV.

8.4.3 Channelled Ion Stopping Power

The ratio of channelled to random stopping is given in Table 8.3 and is plotted in Figure 8.5 for the three axes $\langle 110 \rangle$, $\langle 111 \rangle$ and $\langle 100 \rangle$. There was no evidence for any ion dependence of these values, and the points in Figure 8.5 are not distinguished according to the ion. It is interesting that the peak values for the $\langle 110 \rangle$ axis in the thin diamond are close to the leading $\langle 110 \rangle$ values for the thick diamond. The width

Table 8.3

RATIO OF CHANNELLED TO RANDOM STOPPING POWER

E/A (MeV)	$\Delta E_C/\Delta E_R$ <110>		E/A (MeV)	$\Delta E_C/\Delta E_R$ <111>		E/A (MeV)	$\Delta E_C/\Delta E_R$ <100>	
	Peak	Leading edge		Peak	Leading edge		Peak	Leading edge
Protons (thick crystal)								
2.537	0.774	0.619	2.469	0.880	0.775	2.633	0.888	0.754
3.721	0.791	0.642	3.649	0.880	0.775	3.749	0.880	0.761
4.771	0.793	0.642	4.715	0.879	0.760	4.797	0.879	0.741
5.756	0.801	0.641	5.722	0.883	0.760	5.801	0.873	0.728
6.787	0.799	0.620	6.758	0.882	0.757	6.830	0.882	0.723
7.810	0.820	0.635	7.786	0.884	0.760	7.843	0.883	0.729
8.831	0.818	0.636	8.809	0.885	0.749	8.863	0.876	0.715
9.847	0.817	0.628	9.827	0.887	0.727	9.876	0.883	0.718
10.857	0.831	0.634	10.837	0.891	0.722	10.881	0.881	0.711
11.862	0.837	0.640	11.844	0.897	0.723	11.890	0.890	0.693
Protons (thin crystal)								
3.922	0.695		3.897	0.881		3.939	0.77	
5.942	0.662		5.923	0.889		5.955	0.75	
7.950	0.637		7.935	0.894		7.961	0.71	
8.955	0.663		8.942	0.918		8.965	0.79	
9.961	0.674		9.948	0.911		9.970	0.90	
Alpha (thick crystal)								
2.533	0.782	0.635	2.467	0.864	0.779	2.620	0.860	0.776
3.378	0.802	0.649	3.332	0.889	0.803	3.442	0.872	0.766
4.176	0.795	0.647	4.141	0.879	0.787	4.237	0.873	0.770
⁷Li (thick crystal)								
2.882	0.775	0.610	2.801	0.856	0.772	2.994	0.860	0.725

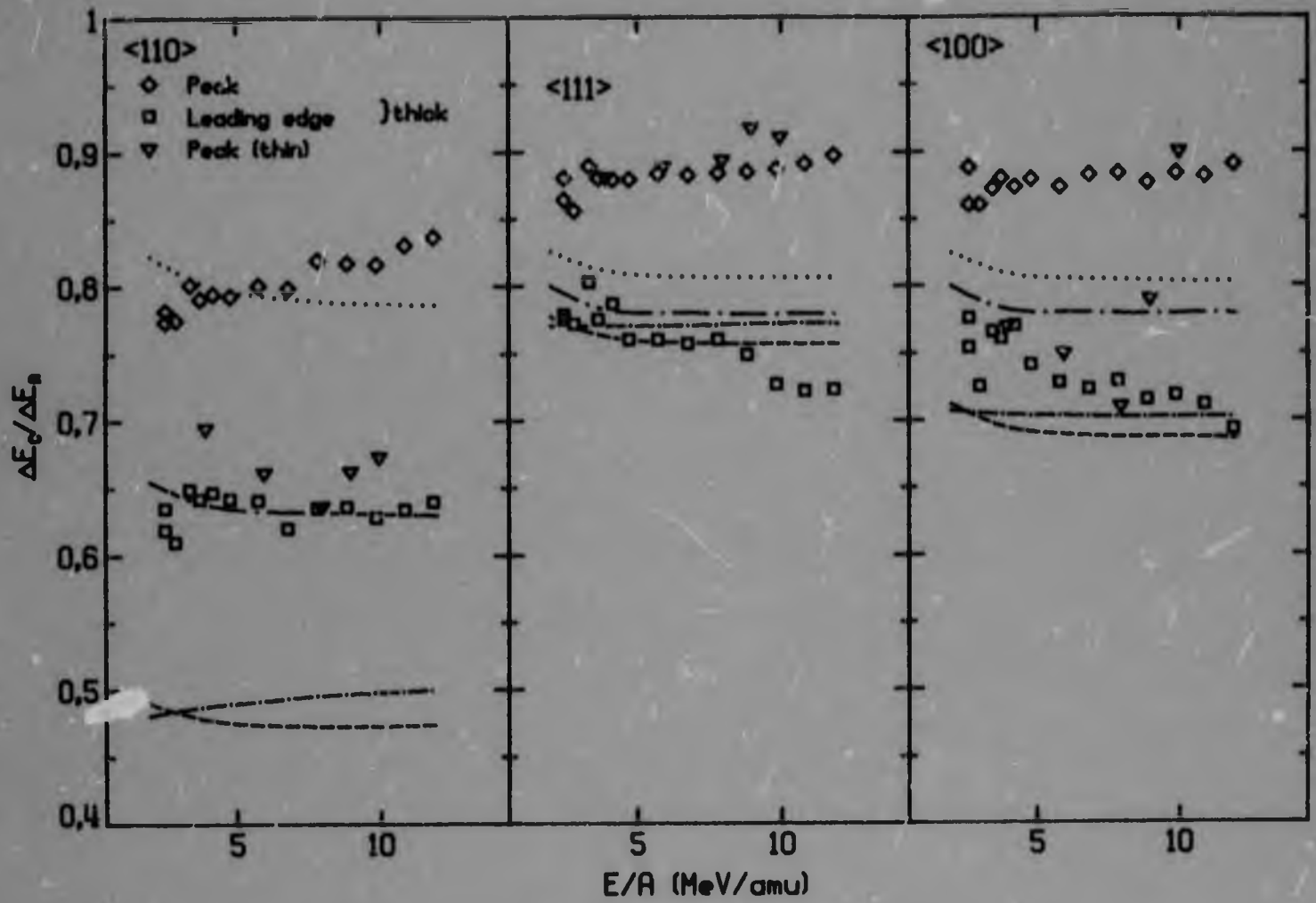


Figure 8.5: Ratio of channelled to random stopping powers. Dettmann; ----- modified Dettmann; -·-·- Esbensen and Golovchenko; - - - - planar.

of the thin diamond $\langle 110 \rangle$ peak was consistent with that expected to be produced by resolution and straggling if the channelled ions see a somewhat reduced electron density. The $\langle 100 \rangle$ values are also close to the $\langle 100 \rangle$ thick target leading-edge values. The $\langle 111 \rangle$ values are close to the $\langle 111 \rangle$ thick target peak values. It is possible that this is due to mis-alignment of the crystal, as there was some difficulty in aligning the thin crystal. It is possible that some mosaic spread and strain was present in the diamond, because it is a Type IIa crystal.

The leading-edge values for $\langle 111 \rangle$ and $\langle 100 \rangle$ measured on the thicker target are apparently energy dependent, increasing to lower energies. In these channels the local electron density at the channel centre is higher, and thus straggling will be greater. Thus it is possible that the lower value at higher energies is due to the influence of straggling in the peaks leading to lower leading-edge values. The influence of straggling would be expected to be less at lower energies, where the effect of dechanneling would be greater. Thus the leading edge for these axes would give a better reflection of the least energy loss, at lower energies. This is consistent with the $\langle 100 \rangle$ peak values obtained for the thin diamond, which are comparable with the lower energy $\langle 100 \rangle$ leading-edge values of the thick diamond.

Thus, the least energy loss can be represented by the $\langle 110 \rangle$ thick target leading-edge values, and the $\langle 111 \rangle$ and $\langle 100 \rangle$ values at lower energy. Combining these with the thin target peak values leads to estimates of the ratio, m , of channelled to random ions of 0.64 ± 0.02 for $\langle 110 \rangle$ and $m_{100} = 0.77$, $m_{111} = 0.80$. These represent average values for all energies: from the theoretical results the energy variation is expected to be small.

The results can now be compared with the theoretical results also shown in Figure 8.5. It is immediately clear that there is a much more pronounced axial variation than that predicted by the Dettmann theory. The theory must, therefore, be modified to include the effect of channelling on the valence electron stopping, and this has been done by treating the valence electrons as a spatially periodic gas as described in Chapter 2. It can be seen that the results from this modified theory are very close to those of the theory of Esbensen and Golovchenko, and these may be considered together, as the approximations involved in the theories render this difference insignificant. This agreement is perhaps to be expected for the reasons given in Section 2.4.5: the equation (2.4.29) from the theory of Esbensen and Golovchenko can be regarded as a local density approximation obtained from equation (2.4.35) for the periodic electron gas. The theoretical values then follow the trend of the experimental values, but the reduction in stopping power and axial dependence are more pronounced than that observed in experiment.

It has been pointed out that the true least energy loss will only be experienced by hyperchannelled ions, that is, those trapped in one channel throughout their motion in the crystal [Ap 72]. It can be expected that these will be observed only in experiments using the thinnest crystals [Ap 72]; the least energy loss in thicker crystals will then be that of ions that have wandered from channel to channel along the most widely spaced plane intersecting the channel [E1 72]. This widest planar channel is $\{111\}$ (wide spacing) for the $\langle 110 \rangle$ axis and $\{110\}$ for both $\langle 111 \rangle$ and $\langle 100 \rangle$ axes. Accordingly, this minimum planar energy loss was evaluated and is also shown in Figure 8.5, for the modified Dettmann theory. These values are more in accord with the experimental least energy loss values.

The slightly higher $\langle 111 \rangle$ experimental value is perhaps due to greater multiple scattering, or due to the possibilities in feeding into various planes. Thus the next widest plane intersecting the $\langle 111 \rangle$ axis is $\{112\}$ and the $\langle 100 \rangle$ is $\{100\}$. The latter is wider than $\{112\}$ and hence will have a lower energy loss.

Finally, some observations can be made about the theoretical approaches. Both the modified Dettmann theory and the Esbensen and Golovchenko theory give similar results. The former is, however, computationally more simple, that is, no numerical integration over electron densities is required, and fewer terms need to be summed (~ 10 compared with ~ 150) for convergence. The modified Dettmann theory also allows the solid-state structure of the target to be included both in the random and channelled energy loss calculations. This could be important in the examination of allotropic effects, if more accurate measurements become available. It should be noted that the use of realistic, that is, experimental X-ray scattering factors can be important in both theories. Thus, in the theory of Esbensen and Golovchenko, use of theoretical scattering factors calculated from spherically symmetric charge distributions (for example, those of Doyle and Turner [Do 68] as used by Esbensen and Golovchenko) lead, in the case of diamond, to energy losses 15 % higher in $\langle 100 \rangle$, and lesser variation in the other axes. Such theoretical scattering factors are probably adequate for large Q ; for small Q the scattering factors are sensitive to the valence electron distribution and more realistic values should be used.

8.5 CONCLUSION

The energy loss of channelled and random ions has been measured for

3 - 12 MeV protons, 12 - 18 MeV alpha particles and 24 MeV lithium ions for the $\langle 110 \rangle$, $\langle 111 \rangle$ and $\langle 100 \rangle$ axes in thin diamond crystals. The dependence of the random stopping power on velocity and atomic number of the ions is in agreement with that expected from the Bethe-Bloch theory. The actual values measured agree within errors with the values recommended by Andersen and Ziegler [Ans 77]. The straggling of the random beam has also been investigated. At small energy losses it is in agreement with the Bohr value, and increases at larger values in qualitative agreement with the TschalMr theory. It is probable, however, that the results at larger energy losses are influenced by the crystal structure: an amorphous target would be more suitable for exploring this regime.

The leading edge of the channelled spectra for larger energy losses in the thicker diamond used has been interpreted as being characteristic of the least energy losses for the channelled ions. The values obtained indicate that the Dettmann theory is inadequate and should be modified to include the effect of channelling on the valence electron energy loss. The modified theory is then in agreement with the theory of Esbensen and Golovchenko. Both theories predict lower values than observed. It is probable that the observed values are those characteristic of non-hyperchannelled ions, and values for the widest planes intersecting the axes are more in accordance with the measured values.

Both the modified Dettmann theory and the theory of Esbensen and Golovchenko give similar results. However, the Dettmann theory is computationally simpler and is more amenable to the incorporation of solid-state bonding (that is, allotropic) effects in the random case. The use

of realistic X-ray scattering factors is important in both theories, if detailed comparison with experiment is to be made.

The results reported here are somewhat limited by the variation in target thickness and subsequent limitation on obtainable accuracy. It would be worthwhile to improve the uniformity of thin diamond crystals. In this regard, the crystals should be of Type Ia, with a low A-nitrogen concentration, and with little (or preferably no) B-nitrogen or platelets. These can be expected to be the best channelling diamonds. A more uniform crystal would enable more accurate measurements to be made, and also allow the investigation of allotropic effects.

CONCLUSION

Channelling measurements on natural diamond have been extended to include dechannelling, the effect of amorphous and impurity layers on the surface, and energy loss measurements. The increase in dechannelling in a series of Type Ia diamonds (which comprise the majority of natural diamonds) compared with the best crystals has been quantified and shown to be due to the presence of platelets on {100} planes.

The yield, as a function of depth, has been measured for the major axial directions at a variety of energies and temperatures. The variation of yield with temperature is rather small on account of the high Debye temperature of diamond and the scaling of the yield with $u_2^2 z$, that has been observed in silicon and germanium, has not been found in diamond. However, the yield as a function of depth has been found to scale with an energy dependent characteristic length, z_e , determined by electron multiple scattering. This has been interpreted as being the dominant dechannelling mechanism in good crystals. The scaled yield is very similar for the three major axes, indicating that the same mechanism is acting in all cases, and that small differences between the axes in the initial transverse energy distributions, and in the distribution of electrons in the channel are of relatively minor importance. Such differences that do exist are rapidly 'washed out' in the diffusion process in transverse phase space.

Theoretical calculations using the steady increase model fail to reproduce the experimental behaviour, but the diffusion model gives good results, if realistic electron scattering terms are used. The model employed used multi-string transverse energy distributions and included damping and absorption terms, although the effect of these is small. An analytical solution of the diffusion equation has been given for the Lindhard single-string initial distribution, and a constant $\frac{dE_L}{dz}$. This can give reasonable results for small depths, but fails at larger depths where the detailed variation of the diffusion function with transverse energy becomes important.

The numerical solutions have enabled the effect of various terms of the diffusion model to be examined and it was noted that the damping term can lead to an increase in the yield at large depths (while the yield is decreased at smaller depths) in contrast to a decrease at all depths noted with the steady increase model.

The effect of amorphous surface layers on the minimum yield at the surface has been studied in $\langle 110 \rangle$. Layers of carbon, aluminium and gold were evaporated onto a polished diamond surface, and a wide range of thicknesses was obtained by removing the layers sequentially using argon ion milling. The effect could then be studied down to relatively small increases in the yield. The use of gold, aluminium and carbon layers allowed a wide range of reduced thickness, τ , to be covered, from $\tau = 0.2$ to 140.

The results were, in general, in good agreement with calculations based on Thomas-Fermi multiple-scattering theory, but small deviations

could be due to deviations of the true ion-atom interaction potential from the statistical Thomas-Fermi potential. It is probable that high precision measurements could be used to distinguish between different potentials. The increase in yield is due predominantly to the tail of the multiple-scattering distribution and thus this type of measurements is complementary to direct measurement of the distribution which gives greatest weight to the maximum of the distribution at small angles.

A scaling law for the yield as a function of τ , derived for power law scattering, has been demonstrated to hold approximately for the range of τ used in the experiment and has been used to correct the aluminium thickness for a small oxygen contribution.

The use of small layer thicknesses has enabled comparisons to be made with Lindhard's estimates of the effect on the yield of contamination layers. While his expression, derived from Rutherford scattering, gives reasonable results for low Z layers, deviations occur at high Z. Using an approximation to the Thomas Fermi single scattering cross-section, an alternative expression has been derived, which gives good results up to $\Delta x = 0.1$.

Calculations of the yield as a function of depth in a crystal overlaid with an amorphous layer have been made using the diffusion model. Results are in good agreement with experiment, lending further weight to the validity of the diffusion model.

The dechannelling produced by defects has been studied in a group

of strong Type Ia diamonds. It has been shown that the dechannelling probability is proportional to the infrared absorption at 1370 cm^{-1} , which is in turn proportional to the area per unit volume of platelets on {100} planes. The dechannelling probability is also proportional to the concentration of B-aggregates of nitrogen. The dechannelling in a diamond with only B-features is found to be similar to, but smaller than, that in diamonds with similar B concentration, but with platelets, and is probably due to dislocation loops found in these diamonds.

A model for the dechannelling cross-section of a platelet has been given, combining the effect of a stacking fault and a dislocation loop. The dechannelling cross-section of a stacking fault has been evaluated taking into account the distribution of transverse energies, and has been found to be about 4 to 5 times larger than simple approaches based on the minimum yield. An expression for the dechannelling cross-section of a dislocation loop has been obtained by combining the approaches of Kudo and Quéré. This expression approximates the radial dependence of Quéré and the energy dependence of Kudo. Estimates of the area per unit volume of platelets obtained with this model are in accord with estimates obtained from electron micrograph counts. If it is assumed that the B-aggregates and platelets are associated, the loop contribution can be estimated from the results for the B-only diamond, and a similar value for the platelet area per unit volume can be obtained. The concentration of B-nitrogen is such that it is consistent with platelets having the Lang structure. Some consequences of this model and the suggested sequence of growth and decay of platelets are pointed out.

The energy loss of protons, alpha particles and lithium ions in

thin diamond crystals has been measured. The random energy losses are in agreement with those tabulated by Andersen and Ziegler, but the errors are such that it cannot be determined if any differences are due to the allotropic form of carbon used. The straggling in the random beam is in accord with the theory of Bohr for small energy losses in the crystal, and increases in qualitative agreement with the theory of Tschalär for larger energy losses. It is possible that the width of the spectrum at larger energy losses is affected to some extent by feeding into channels, and close comparison has not been made.

The channelled ion energy loss has also been measured, and values for the ratio, m , of channelled to random stopping for 'best channelled' ions are $m = 0.64 \pm 0.02$ for $\langle 110 \rangle$, $m \leq 0.3$ for $\langle 111 \rangle$ and $m \leq 0.75$ for $\langle 100 \rangle$. No dependence of m on the atomic number of the incident ion was observed.

Comparison has been made with the theories of Dettmann and of Esbensen and Golovchenko. It has been shown that the effect of channelling on the valence electron contribution to stopping should be taken into account in the theory of Dettmann. An expression for the energy loss in a periodic electron gas has been derived by using a sum rule. When combined with the core contribution of Dettmann, results close to those of Esbensen and Golovchenko are obtained. The theoretical results calculated at the channel centre are rather lower than the experimental results, but calculations for the principal plane passing through the axis are in closer accord with the experimental results. This indicates that effects of hyperchannelling are not seen.

The use of diamond as a target in channelling experiments is not without difficulties, but these can be overcome with care in the selection, characterisation and preparation of crystals. Selection from a large suite of crystals allows perfect crystals, at least by channelling standards, to be obtained and differences between the channelling in these and in less perfect crystals may be explained quantitatively in terms of known defects. Channelling experiments may be performed on diamond with the same confidence as on other laboratory-grown, crystals.

APPENDIX 1

CHANSPEC

A FORTRAN computer program, CHANSPEC, was written for the off-line analysis of channelling spectra on the Nuclear Physics Research Unit's Interdata (Perkin-Elmer) 7/32 computer. This programme accepted input spectra from cards or magnetic tape and performed operations on the data defined by a sequence of command cards ('card' is used here loosely as a term for any suitable sequence of records and thus input or commands could be on cards, disc file or read from a terminal):

i) READ

Spectra can be read in, and a number of random spectra averaged.

ii) FIT

The surface edge of the spectra could be 'deconvoluted' and the surface peak extracted from the channelling spectra. The yield could then be extrapolated to the surface. This was performed by fitting a suitable function to the edge, using least-square methods. The surface peak was fitted with a gaussian,

$$h \exp \left[- \frac{(x - x_0)^2}{2\sigma^2} \right],$$

and the underlying edge with a quadratic step (that is,
 $n(x) = a + b(x - x_0) + c(x - x_0)^2$, where n is the number of

counts and x the analyser channel or energy and a, b, c are fitting parameters, with $n = 0$ for $x > x_0$, where x_0 corresponds with the surface) convoluted with a gaussian. This yields for the fitting function

$$i(u) = E_1 + E_2 + E_3 + P; \quad u = x - x_0$$

where $E_1 = (a + bu + cu^2) \operatorname{cerf} \left(\frac{u}{\sigma} \right)$

$$E_2 = \frac{(b + 2cu)\sigma}{\sqrt{2\pi}} e^{-\frac{u^2}{2\sigma^2}}$$

$$E_3 = c \int_u^{\infty} \frac{1}{\sqrt{2\pi}\sigma} e^{-\frac{v^2}{2\sigma^2}} dv$$

$$P = \frac{h}{\sqrt{2\pi}\sigma} e^{-\frac{u^2}{2\sigma^2}}$$

Here, $\operatorname{cerf}(x)$ is the complementary error function. The coefficient c is usually small and thus E_3 may be neglected. E_2 may also often be neglected.

The gaussian peak term, P , was not used in fits to random spectra. After fitting, the unconvoluted edge was used to replace the convoluted experimental points.

iii) DEPTH

A conversion from an energy scale to a depth scale could be performed using the method of Zaidins [Za 74] and his parameterisation of the stopping-power and range, with values suitable for diamond. The program allowed a lower channelled energy loss (a constant fraction of random energy loss) to be used if required. This is discussed in Chapter 5.

iv) NORM

Spectra could be normalised to random.

v) SMOOTH

Statistical fluctuations in the data could be smoothed using a least squares cubic spline smoothing technique [Rei 67]. A parameter to define the amount of smoothing was calculated by the program, but could be over-ridden. The former was generally adequate.

vi) DAMAGE

This command allowed extraction of radiation damage profiles [Der 78] or the analysis of dechannelling probability using the single scattering model (Chapter 7), depending on the version of the program.

Processed data could be printed (PRINT) or plotted on the line printer (PLOT) at any stage, or analysis could be re-started at the as-read-in stage without a re-read (RESTART). Additional commands allowed a

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