Stopping power for fast channeled α particles in silicon

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The stopping power for 160-MeV α particles in silicon has been measured in the $\langle 110 \rangle$ and $\langle 111 \rangle$ channeling directions and also for random impact. The respective values 0.54 ± 0.02 , 0.67 ± 0.02 , and 1.10 ± 0.01 eV/Å are discussed in relation to conflicting theories of the channeled stopping power.

I. INTRODUCTION

Possibly the most contentious feature of the channeling phenomenon is the reduction in the stopping power $S = -\frac{dE}{dx}$ that occurs when ions are incident along low-index directions of a single crystal. Even if we confine the discussion to ions of high velocity ($\gg v_{Bohr}$), so that the stopping is dominated by energy loss to the electrons of the target crystal, the literature is extensive and often conflicting.¹ This is perhaps surprising when one recalls that for random incidence the problem was essentially solved by Bethe² and by Bloch.³

Two distinct approaches to the problem of calculating S for channeled particles of high, but nonrelativistic, velocities may be identified. Lindhard's⁴ approach separates the energy lost into two components: that lost in distant collisions with the target electrons, regarded as free, and that lost in close collisions. For fast ions there is equipartition between the former (plasmon excitation) and the latter (single-particle excitation) so that in channeling, where close collisions are suppressed, the stopping power $S_{\langle hkl \rangle}$ is just half the random value S_r . In its simplest form this prediction is independent of the target material and channeling direction in contrast to experimental results for silicon⁵ and, particularly, germanium.⁶ Golovchenko and Esbensen⁷ have given a more detailed study of stopping in silicon, based on Lindhard's ideas, and recognize that there is a channel dependence for $S_{(hkl)}$. Their model gives $R_{(hkl)} = S_{(hkl)}/S_r \le \frac{1}{2}$ and for 160-MeV α particles in silicon predicts $R_{(110)} = 0.34, R_{(111)} = 0.51.$

An alternative approach has its origins in attempts⁸ to explain the observed differences in $R_{(hel)}$ between silicon and germanium in terms of the different relative numbers of valence and core electrons in these two materials. The valence electrons are regarded as free but their contribution to stopping is the *same* for both channeled and random impact. The central problem now is to calculate, for a particular ion velocity and target, what fraction of the energy is lost to the core electrons and then to apportion this correctly into loss from distant and from close collisions. This has been attemped by Dettmann⁹ using Hartree-Fock functions for the silicon-target core electrons and paying particular attention to the calculation of the average excitation energy of these electrons. This results in $R_{\langle hkl \rangle} \ge \frac{1}{2}$ at high velocities and, specifically, Dettmann⁹ predicts $R_{\langle hkl \rangle} = 0.64$ for 160-MeV protons in silicon along any channel. The present experiment was performed in an attempt to test these conflicting theories using 160-MeV α particles, rather than protons, because of the availability of good energy resolution in both beam and detector for the former. The ratio $R_{(hkl)}$ is not expected to be sensitive to ion type provided the ion remains fully stripped and the high-velocity limit has been reached.

II. EXPERIMENT AND RESULTS

The α -particle beam from the Harwell synchrocyclotron was collimated by apertures of 2.5 and 1.0 mm diameter placed 7.5 m apart in an evacuated beam pipe with thin Mylar end windows. After passing through a silicon target, which was mounted in air in accordance with customary practice for experiments in this energy region, the beam traversed a second evacuated pipe and was detected by a Ge(Li) detector placed a further 1.7 m downstream; the overall energy resolution obtainable was at best 0.8 MeV full width at half maximum. Very good energy resolution is needed for accurate energy loss measurements in what necessarily must be a thin crystal because of dechanneling.¹¹ The Ge(Li) detector was placed on the beam axis and behind a 2.5-mm collimator so that only those particles substantially undeflected by their passage through the target would be recorded. From a knowledge of the synchrocyclotron design and the stopping powers of the air and Mylar we

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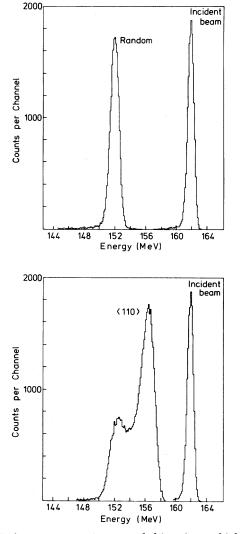


FIG. 1. Energy spectra recorded in a 1-cm-thick Ge(Li) detector with an α -particle beam of energy 166 MeV at the target position. The target was a 0.91-mm-thick silicon crystal, cut with the $\langle 110 \rangle$ axis normal to its surface. The upper spectrum was obtained with the beam incident at a small angle to the crystal normal and gives the random energy loss. The lower spectrum was obtained with the crystal inclination adjusted accurately to obtain the optimum channeling fraction along the $\langle 110 \rangle$ axis.

calculate the incident beam energy at the detector, with no target, to be 160.4 ± 1.0 MeV. Additionally, calibrating the detector with ²²Na and ²⁴¹Am γ -ray sources we find the beam energy to be 163 ± 0.8 MeV. We adopt an average of these two results, 162 ± 1 MeV, which implies an incident energy at the position of the specimen of 166 ± 1 MeV.

Two single-crystal silicon-disc specimens were used: 0.91 ± 0.01 mm thick, cut, polished, and etched perpendicular to the $\langle 110 \rangle$ direction and 2.06

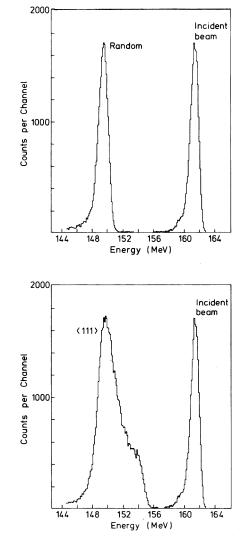


FIG. 2. Energy spectra obtained under similar circumstances to Fig. 1 except that the silicon crystal was set with its normal making an angle of $35^{\circ}16'$ to the beam direction and was adjusted by rotating in a plane parallel to its surface until the channeling fraction along the $\langle 111 \rangle$ axis was optimized.

±0.01 mm thick similarly prepared with respect to $\langle 111 \rangle$. They were mounted on a three-axis goniometer controlled by stepping motors of minimum movement corresponding to 0.01°. Energy spectra recorded in the Ge(Li) detector are shown in Figs. 1 and 2. We notice here a slight variation in the incident beam energy related to changes in running conditions of the synchrocyclotron on different days. Movement between $\langle 110 \rangle$ and $\langle 111 \rangle$ was obtained by turning the crystal through 35° 16' along a $\{110\}$ plane whilst "random beam" spectra were obtained from a deliberately misaligned target which nevertheless presents the same thickness to the beam. For the 0.91-mm crystal placed normal to the beam, the (110) direction (Fig. 1), we see a clearly distinguished channeled component together with a smaller "random" peak on the low-energy side. With progressively thicker samples, as seen by the beam, this channeled component changes to a shoulder and then to a weak tail on the high-energy side of the spectrum. For the (111) direction (Fig. 2) the channeled fraction is much smaller than for the $\langle 110 \rangle$, a difference we found characteristic of channeling direction and not attributeable solely to the extra thickness of the target due to its inclination to the beam. The relative number of α particles falling in the well-channeled portion of the spectrum (e.g. Fig. 1) is observably influenced by the presence of the 0.025-mm Mylar window and 10 mm of air between our final 1.0-mm collimator and the specimen but the position of the well-channeled portion and, therefore, the determination of the least energy loss is in no way affected. However, the multiple scattering so introduced prevented us from obtaining more than rough estimates of Lindhard's ψ_{crit} and the detector collimator prevented a measurement of the dechanneling length.

Dechanneling is evidently very strong in silicon. Owing to the collimation placed before the Ge(Li) detector those particles which failed to become channeled and most of those which became dechanneled during their passage through the crystal will not be recorded. Thus a significant fraction of particles contributing to the random portion of the bottom spectrum of Fig. 1, and most of the random portion of the bottom of Fig. 2, will represent particles dechanneled late in their passage through the crystal. This feature means that the random portion of a channeling spectrum will be wider than for a misoriented crystal and any apparent displacement of this pseudorandom peak upwards in energy is of no present concern. Acerbi et al.¹¹ have studied the dechanneling effect along the (111) axis of silicon and found an exponential law to be appropriate with a half length $L_{1/2}$ for 38.9-MeV protons of 0.12 mm. Since $L_{1/2}$ may be expected to scale¹ as E/Z, where E is the energy of the incident ion of charge Z, then for 160-MeV α particles we would expect $L_{1/2} \simeq 0.24$ mm. Using this estimate it is apparent that those particles which are channeled for a depth into the crystal sufficient to ensure that they contribute to the leading edge will have but a small further chance of becoming dechanneled before leaving the crystal since the width of the leading edge is only about 10% of the effective target thickness. Consequently, we can assume that most of the high-energy portion of the leading edge will be essentially uncontaminated by dechanneled particles.

The random stopping powers are determined from

the spectra shown in Figs. 1 and 2 by fitting Gaussian distributions. The Gaussian form is clearly adequate for the incident beam and is a good approximation to the Landau-Vavilov¹² energy loss distributions for targets of the thickness used here.

The correct procedure for computing the channeled stopping power is not so obvious. The theories of Dettmann and of Golovchenko and Esbensen both refer to the energy loss of the "best-channeled" particles so that one would wish to identify these with the high energy portion of the leading edge of a channeled spectrum. However, the leading edge arises not only because it contains the best channeled particles but also because these particles were initially amongst the most energetic in the beam. We can easily identify in our spectra those particles which have remained fully channeled during their passage through the crystal but the term "best channeled" implies a condition which is not unequivocally exhibited in our results, and which may not be susceptible to experimental demonstration. This difficulty is discussed by Fich et al.¹³ in connection with an observation of channeling by 1.35-GeV/c protons and π^+ in a germanium crystal.

Not only does the width of the channeled peak reflect the importance of dechanneling but there will also be a contribution (perhaps visible in Fig. 1) due to the variation in stopping power compatible with the spread in transverse energies of particles which remain fully channeled. The channeling theories do not make allowance for nonzero transverse energies which is why they must specify "bestchanneled" particles, which will correspond in some way to the leading edges. We have therefore fitted a series of Gaussian distributions to the leading edges starting with all the data points and gradually reducing their number so as to concentrate on the high-energy region. As the number of data points is reduced so the Gaussian distribution diminishes in height and width, and its centroid moves towards higher energies. Because of the residual dechanneling complication and since the width of the Gaussian is largely determined by the beam energy spread we have not considered it profitable to attempt to compute a theoretical width appropriate for each channeled spectrum but demand only that the fitted width should lie between the widths associated with the incident beam and random loss spectra. We then select as optimum that Gaussian distribution with centroid lying in the middle of this width range and associate with it an uncertainty of just half the observed spread of possible values. This uncertainty, whilst not large, dominates all others so that the errors quoted in Table I for $R_{\langle hkl \rangle}$ are to be understood as a measure of the possible systematic, rather than statistical,

TABLE I. Channeled $(S_{(hkl)})$ and random (S_r) stopping powers for α particles in silicon (eV/Å); also shown are the stopping-power ratios $R_{(hkl)}=S_{(hkl)}/S_r$. The incident beam energy is 166 ± 1 MeV so the average energy lies between 154 and 164 MeV, depending on sample, axis, and orientation. All experimental results quoted in this paper have been corrected (Ref. 10) to a mean energy of 160 MeV to facilitate comparison with theory.

	Experiment		Theory (160-MeV α particles)		
	0.91-mm sample	2.06-mm sample	Dettmann ^a	Golovchenko and Esbensen ^b	Bichsel and Tschaler ^c
S(110)	0.54 ± 0.03	0.54 ± 0.05	> 0.58	0.37	
$S_{(111)}$	0.66 ± 0.02	0.69 ± 0.04	0.64	0.55	
S _r	1.073 ± 0.014	1.114 ± 0.012	1.12	(1.082)	1.082
R (110)	0.50 ± 0.02	0.49 ± 0.04	>0.52	0.34	
$R_{\langle 111 \rangle}$	0.61 ± 0.02	0.62 ± 0.03	0.57	0.51	

^a Reference 9.

^b Reference 7.

^c Reference 10.

errors. This procedure was applied only to the 0.9-mm crystal, appears to be entirely self-consistent, and gives no evidence for the existence of a few "best-channeled" particles suffering an energy loss significantly lower than the rest. It was not practicable to fit Gaussian distributions to the 2.06-mm data since the channeled spectrum was relatively shapeless, possessing only a well-defined end point. This end point was identified as the high-energy intercept of the straight line drawn tangential to the leading edge of a Gaussian distribution, and the position of the centroid of this hypothetical Gaussian could then be inferred. The uncertainties involved in this procedure are quite large but are compensated to some extent by the fact that for the 2.06-mm crystal the energy differences are also large. The main reason for including these results in Table I is to demonstrate that our measurements of $R_{\langle hkl \rangle}$ are not dependent upon target thickness.

III. DISCUSSION

A. Experiment

The values of S_r deduced from the 0.91- and 2.06-mm samples do not agree very well, although the discrepancy is not statistically unacceptable. Combining the two values and their errors gives $S_r = 1.097 \pm 0.009 \text{ eV/Å}$ for 160-MeV α particles in silicon. Of the two theoretical values quoted in Table I only that of Dettmann is calculated absolutely and the prediction $S_r = 1.12 \text{ eV/Å}$ must be accorded as being in excellent agreement with the experiment. The stopping power of Bichsel and Tschalar,¹⁰ $S_r = 1.082 \text{ eV/Å}$, is based on an extrapolation of a formula fitted to stopping power data measured with rather low particle energies; it is not clear what accuracy should be attributed to this formula at 160 MeV but we might reasonably expect an uncertainty of about 1%.

We believe our best values for $R_{(hkl)}$ for 160-MeV α particles in silicon are 0.499 ± 0.021 and 0.615 ± 0.013 for $\langle 110 \rangle$ and $\langle 111 \rangle$, respectively. It will be appreciated that this experiment consists of measuring a small energy loss as the difference between two large energies observed in successive target-in and target-out measurements which necessarily involve changes to the synchrocyclotron beam intensity. Great care was taken to avoid concomitant changes to the primary beam energy. The experimental stopping powers $S_{(hkl)}$ quoted in Table I are secondary quantities deduced from the $R_{(hkl)}$ and S_r results.

Most other experiments with which the present results might be compared have been performed with very low energies (< 10 MeV/amu) where the high-velocity limit is far from being applicable; see Gemmell¹ for a summary of these measurements. At higher but still nonrelativistic energies there is only the work of Acerbi et al.¹¹ which concentrated on measurements of the stopping power ratios for 22-39-MeV protons channeled along the (100) plane in W, but in which a measurement for the (111) axis in silicon was obtained for 20.4-MeV protons yielding the rather imprecise result $R_{(111)}$ $= 0.64 \pm 0.10$. More interesting, perhaps, are the very-high-energy measurements of Fich et al. using 1.35-GeV/c protons and π^* , from which those authors tenatively deduced $R_{(110)} \approx 0.33$. However this result is not to be compared with the present measurements since it refers not to silicon, but to germanium for which the strong inner-shell binding energy is such that a high-velocity limit cannot be reached before the onset of relativistic effects, a point clearly realized by Dettmann, who did not claim such a limit for germanium. Fich et al. made no allowance for the statistical fluctuation contribution to energy loss straggling for the channeled particles and, moreover, they actually measured the ratio of the number of electron-hole pairs produced in channeled-random orientations and not the energy loss ratio, although careful consideration leads us to expect that any correction required for this last effect would be likely to be small.

B. Theory

Our work was motivated by a desire to provide an experimental test of the conflicting theoretical predictions exemplified by the treatments of Golovchenko and Esbensen⁷ on the one hand and of Dett $mann^9$ on the other. (We regard the earlier papers of Dettmann and of Dettmann and Robinson, see also Ref. 9, as "first tries" which, although they give valuable insight to the approach, are less exact.) We give a brief description of the two models since the former⁷ has only appeared as an abstract. Both theories are nonrelativistic and use guantummechanical perturbation methods involving the first Born approximation to describe the excitation and ionization of the target atoms; they should therefore be most accurate for projectile velocities which are high compared with those of the target electrons. Both theories accommodate the channeling process by working in terms of the impact parameter b of collisions between projectile and a collection of static noninteracting atoms arranged on the lattice sites of the crystal. The principal difference seems to us to lie in the approximation used to overcome the divergence at b = 0 characteristic of the dipole approximation and clearly seen in Bloch's artificial cutoff at small b = b', so arranged that the impact parameter theory gave the same result as the Bethe theory, for a randomly arranged target. Golovchenko and Esbensen⁷ use the dipole approximation for the collective excitations, roughly corresponding to large b, but utilize a sum rule for the single particle collisions, small b region. This sum rule is only exact for a target electron bound in the harmonic oscillator potential so that it will be a poor description of the electron states of the solid. The channeling stopping powers predicted by Golovchencko and Esbensen are significantly lower than the experimental values given in Table I but they do correctly predict a channel dependence, although somewhat exaggerated. They give no calculation of the random stopping and the tabulation of Bichsel and Tschalar¹⁰ is used to obtain the ratios $R_{\langle hkl \rangle}$.

The early models of Dettmann and of Dettmann and Robinson took hydrogenic ground states for the target electrons so that each electron was identified with a particular atom. These electron states were scaled, through the binding energy ϵ_{nl} , for the various shells in silicon and germanium to give Bohr radii $a_{nl} = \hbar^2 / 2m \epsilon_{nl}$. The avoidance of the dipole approximation forced the authors into a new way of calculating the average excitation energy $\overline{\omega}$, given to an electron in a collision, which was just twice Bloch's value, i.e., $\overline{\omega} = 2\epsilon_{nl}$. Their calculation of the b dependence of the projectile energy loss $\Delta E(b)$ showed that: (a) the divergence at b = 0 had been removed and provided $\overline{\omega} = 2\epsilon_{nl}$ the integral over all b supplied the Bethe formula; (b) the contribution to ΔE from the $b \ll a_{nl}$ region (what has been conventionally regarded as the "close" collisions) was small; (c) the major contribution to ΔE came from the region $a_{nl} \ll b \leq b_c$ where $b_c/v_{\text{projectile}} = a_{nl}/v_{\text{electron}}$ and hence for loosely bound electrons and fast projectiles b_c is many times any channel radius: thus the projectile will cause both excitation and ionization at atoms far removed from the channel wall and so the projectile cannot know it is channeled.

It was originally thought that sufficiently fast projectiles incident on light targets would interact roughly equally with *all* bound electrons at atoms far removed from the channel so that the claim was made $R \rightarrow 1$ for the high-velocity limit. Later, doubts arose over the approximation leading to the choice of $\overline{\omega}$ so that Dettmann and Robinson only claimed $R \ge \frac{1}{2}$.

The final paper of Dettmann⁹ is significantly different from those earlier in that there are quite separate treatments for the valence and for the core electrons. The four valence electrons are taken as a uniformly distributed free-electron gas responding as a plasma, in the manner of Lindhard. We see this as recognizing that the range of interaction for such loosely bound electrons is much larger than the channel radii so that the b integration will have the effect of averaging the electron density fluctuations which in fact occur. Again the local electron density is irrelevant in this approximation and these four electrons will therefore contribute equally to random and to channeled stopping. The paper then concentrates on the behavior of the core electrons treating them in a tight binding approximation with Clementi wave functions instead of the hydrogenic type. The $\overline{\omega}$ approximation is now handled differently so that it is only needed for those collisions causing excitation of the target core electrons (Dettmann's "low excitations") and is consequently of less weight in the theory. (The "high excitations" are treated in a different way which avoids any approximation for $\overline{\omega}$. Since they are only dominant at $b \leq a_{nl}$ and in channeling our minimum impact parameter is about three times the largest bound-electron orbit radius these high velocities will contribute marginally to stopping only in the narrower (111) channel. From Dettmann's Fig.

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5 we estimate the channeled stopping $S_{(111)}$ in Table I might be increased by 3% due to these high excitations. The contribution of the high excitations is of course included in the calculated random stopping.) Dettmann's work can be utilized directly to calculate $S_{(hkl)}$ for a given channeling experiment through his Eqs. (3) and (21). Using (21) one must sum the contribution from successive rings of atoms around the channel center out to b > b, (we use his notation⁹ in this part of the discussion and b_{c} , corresponds physically to the earlier b_{c} ; beyond b_{j} these contributions soon become small. Now the largest b_{j} is $b_{2s} = 5.086$ Å for our velocity particles (39.06 a.u.) so the contribution to ΔE only becomes negligible at $b \sim 6$ Å [Eq. (21a), see our reference list for a corrected version]. However, by b = 5.124Å (the fourth ring of atoms around the $\langle 110 \rangle$ channel) we already calculate $S_{(110)} = 0.58 \text{ eV/Å}$. We therefore enter the value >0.58 in Table I and recognize that Dettmann predicts too high a value in this channel. He has offered the argument that this is because the free-electron gas model for the valence electrons gives too high a value for the energy loss since the directions of the bonding orbitals are such as to localize the valence electrons along these channel walls and away from the channel center. For the $\langle 111 \rangle$ channel we have included contributions to ΔE , out to b = 5.757 Å beyond which they become negligible. We obtain a value for $S_{(111)}$ in fair agreement with experiment; no localization difficulty is expected with the valence electrons in this channel. Now while it is certainly true that the valence electron density near the center of the $\langle 110 \rangle$ channel is low whereas that in the (111) is not, see for example Desalvo and Rosa,14 this argument seems to us wrong in view of the earlier claim that localization is irrelevant. We have considered trying to put the suggestion on a quantitative basis but always come back to the problem of localization which we view as not in the spirit of Dettmann's papers. For the moment we recognize that Dettmann's predictions are a better fit to our data than those of Golovchenko and Esbensen but there is still room for improvement. We would particularly welcome a calculation of the straggling carried out in accordance with Dettmann's ideas since this is sensitive only to the high excitations. There is much confusion in the literature as to how the stopping power for best-channeled particles is to be extracted from the data which contains both straggling and dechanneling contributions. Our procedure of fitting a Gaussian to the high-energy side of the spectrum is undoubtedly correct but the error involved in our ad hoc specification for its width is still dominant and must remain so until the straggling can be separated out accurately.

Finally it is instructive to apply the simple arguments at the end of Dettmann's paper to our 160-MeV α particles. We think it clearer to write his last equation as

$$R(160 \text{ MeV}) \cong \frac{S_{v} + S_{r,1}}{S_{v} + S_{r,1} + S_{r,2}} \cong \frac{N_{v} + \frac{1}{2}N_{\text{core}}}{N_{v} + N_{\text{core}}} \quad (1)$$

since $S_{r,1} = S_{r,2}$. Then we recognize that our critical $b_{1s} = 0.225$ Å so that even for our smallest impact parameter, b = 1.108 Å (two of the wall atoms of the $\langle 111 \rangle$ channel) the $1s^2$ electrons can never contribute to channeled stopping, even through $S_{r,1}$, so that (1) becomes

$$R = \frac{N_v + \frac{1}{2} (N_{\text{core}} - 2)}{N_v + N_{\text{core}}} = \frac{4 + \frac{1}{2} \times 8}{4 + 10} = 0.57$$

close to the observed $R_{(111)}$. Since for even 160-MeV protons $b_{1s} = 0.411$ Å we note that Dettmann was wrong in his prediction of $R \simeq 0.64$, the correct prediction being $R \simeq 0.57$ as for α particles. It is clear that the high-velocity limit in which all core electrons contribute to the distant collision energy loss is not reached even for $\beta = v/c = 0.52$ and that it cannot be reached for silicon without moving into the relativistic energy region for which current theories are inapplicable. We conclude, therefore, that we have indeed reached a high velocity limit for silicon but with the meaning of the phrase restricted in a manner not originally envisaged, namely that the $1s^2$ electrons should not contribute. It will be interesting to test this high velocity limit for diamond where the smaller $1s^2$ binding energy may well allow these electrons to contribute and also for germanium where the stronger core electron binding will certainly prevent several subshells of electrons contributing to the low excitations so reducing R below the value predicted in Eq. 1.

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$$\frac{2\pi}{v^2}\sum_j \frac{1}{bb_j} e^{-2b/b_j}, \ b \gg b_j.$$

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