PHYSICAL REVIEW B VOLUME 16, NUMBER 1 1 JULY 1977

Comparative study of uniaxial channeling-blocking and single-alignment channeling backscattering—reversibility and defect analysis*

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(Received 3 December 1976)

Scattering yields are examined for 200-keV proton channeling in silicon and gold under conditions of single and double alignment. Results are presented for 180° uniaxial channeling—blocking and 150° channeling—backscattering for nondamaged but thermally vibrating crystals (300 K). The analysis of the data is approached, however, from the point of view of the formalism required for decomposing an arbitrary mixture of interstitial and lattice-distortion-type crystal defects in order to assess the applicability of the reversibility rule of channeling and blocking to the defect analysis equations. In the limiting condition of thermal dechanneling it is observed that the equations are applicable only when the double-aligned spectra are corrected for the energy dependence of scattering into blocking configurations. The transformation is discussed in terms of scaling according to a set of energy-loss parameters that can be obtained from the scattered-energy spectrum. These procedures appear necessary since strict reversibility in channeling and blocking cannot be expected under conditions where electronic stopping introduces significant energy loss during penetration of the channeled beam.

INTRODUCTION

Analysis of defects in crystals by the channeling technique can be greatly facilitated through the use of comparative single-alignment channeling backscattering with double-alignment channeling blocking. An important capability of such an approach is the direct quantitative determination of the relative contribution to channeled particle scattering from atoms with large displacement from atomic rows ($x > a_F$) compared with scattering from atoms that have only slight misalignment from lattice rows ($x \sim a_F$, where x is displacement, and a_F is the Thomas-Fermi screening distance). The value of this approach is evident when one considers the parameters required to extract the number of direct scattering (interstitial-type) defect centers that are mixed with other defect types (e.g., strain fields or faults) which produce a predominance of dechanneling with little or no associated direct scattering.

In the present work, the formalism involved in such an analysis is examined, and a discussion is given of the necessary transformation required on the double-aligned data so that the rule of reversibility between channeling and blocking can be satisfied in the analysis of defects.

THEORETICAL CONSIDERATIONS

Assuming statistical equilibrium and a uniform distribution of channeled particles across the channel, the observed yield in single alignment is given as[1]

$$\chi_1(z) = \chi_R(z) + [1 - \chi_R(z)]N_D(z)/N, \quad (1)$$

where $\chi_R(z)$ is the randomized component of the beam, and $1 - \chi_R(z)$ is the remaining channeled component. To solve for the atomic fraction of direct scattering defects N_d/N (e.g., interstitials), it is necessary to know the value of $\chi_R(z)$ at all depths. Various approximation methods have been developed to extract this information from the theories of single, plural, and multiple scattering from interstitials[2]; however, as has been recently demonstrated,[3] these methods are generally inadequate for the analysis of complex defect structures that produce dechanneling independent of, or not in simple proportion to, the direct scattering process.

Double-alignment channeling-blocking measurements performed simultaneously with single alignment can provide a unique and unambiguous value of the random component of the beam through a second independent equation that gives solutions for all variables in Eq. (1) as a function of depth.

In the case of uniaxial double alignment, the expression for the observed yield is, under conditions of reversibility,[1,4,5]

$$\chi_2(z) = h\chi_R^2(z) + [1 - h\chi_R^2(z)]N_D(z)/N, \quad (2)$$

where h is a geometric factor unique to the angle of observation being employed (180° in the uniaxial case). According to theory, h is independent of depth.[5] The other terms in Eq. (2) have the same definition as in Eq. (1). Under the limiting condition for which no direct scattering centers are present, Eqs. (1) and (2) reduce to

$$\chi_1(z) = \chi_R(z), \quad \chi_2(z) = h\chi_R^2(z). \quad (3)$$

Thus, h becomes the parameter that connects the single-aligned dechanneling yield with that from double alignment when N_D approaches zero.

$$h=\chi_2(z)/\chi_1^2(z), \quad N_D(z)\to 0. \tag{4}$$

One can thus determine, under the conditions of Eq. (4), the experimental value of h near the surface, as well as its depth dependence and compare these results with theory.

A generalized solution for the random fraction of the probing beam can be obtained from Eqs. (1) and (2) and is applicable to arbitrary combinations of direct scattering and dechanneling. This is obtained by combining Eqs. (1) and (2) through division to eliminate the explicit appearance of the term N_D/N. The dechanneled component of the beam, at any depth, is then given directly in terms of the observable quantities χ_1, χ_2, and the parameter h. The resultant expression for χ_R is

$$\chi_R(z)=([1-\chi_2(z)] \pm \{[\chi_2(z)-1]^2-4h[1-\chi_1(z)] \times[\chi_1(z)-\chi_2(z)]\}^{1/2}) \times\{2h[1-\chi_1(z)]\}^{-1}. \tag{5}$$

When χ_R is real, it is necessary to use the negative branch of the solution since, under the limiting conditions $\chi_1=\chi_2=0$, χ_R must also be zero. When Eq. (5) is complex, it is necessary to restrict the solution so that the dechanneled fraction is given by the modulus of $\chi_R(z)$ which is

$$|\chi_R(z)|=\{[\chi_1(z)-\chi_2(z)]/h[1-\chi_1(z)]\}^{1/2}. \tag{6}$$

The value of Eq. (5) lies in its yielding a direct *in situ* experimental determination of the dechanneled fraction with no assumptions for the dechanneling process. However, it does require that the reversibility rule of channeling and blocking hold.

Once $\chi_R(z)$ is determined, then the direct scattering yield $\chi_D(z)$ is simply obtained by evaluating $\chi_D(z)=\chi_1(z)-\chi_R(z)$. The direct scattering yield is then related to the fractional concentration of direct scattering centers (i.e., interstitials) through the expression $\chi_D(z)=[1-\chi_R(z)]N_D(z)/N$, and so

$$N_D(z)/N=[\chi_D(z)][1-\chi_R(z)]^{-1}. \tag{7}$$

It has been pointed out[4] that under conditions of nonuniform transverse distribution of direct scattering centers, $N_D(z)/N$ is a transverse spatially averaged value.

EXPERIMENTAL

The channeling-backscattering experiments are performed with 200-keV protons in 150° single alignment and 180° uniaxial double alignment. An annular detector is employed in the uniaxial geometry so that the beam and scattering axes are the same. The acceptance angle of this detector is determined by its cover-plate aperture and is adjusted to 0.35°, which is $\frac{1}{5}$ the critical angle for channeling. The annular detector is also cooled and its temperature regulated with a thermocouple to about −50 °C so that its resolution is comparable to that of the 150° planar detector. Improvements in low-noise characteristics are also achieved in both detectors by using isolated ac power and grounds for the counting equipment. This requires that the detectors be electrically floating from the target chamber and accelerator beam line. Full width at half-maximum noise values, as measured with a pulser, are typically 7 to 9 keV. The annular detector must have the capability of being withdrawn from the beam axis into a protective shroud while under vacuum, in order that a large diameter heavy-ion beam can be directed onto the specimen for the purpose of producing radiation damage. This is accomplished by attaching the detector to a linear-motion bellows-type vacuum feedthrough. The detector is cooled by flowing liquid-nitrogen vapor through a cold head in the vacuum beam line and transferring heat from the detector mount to the cold head through a copper braid. This allows free motion of the cooled detector when it is moved in and out of the beam axis. Spectra are obtained in both detectors for random and aligned orientations with the dechanneled fractions versus depth obtained from a channel-by-channel ratio of the aligned-to-random yield. The energy-to-depth conversion is performed in the usual manner,[1] and constant average stopping powers[6] appropriate to the energy of the beam and the material under study are used. A further discussion of stopping power is presented below.

RESULTS FOR UNDAMAGED CRYSTALS

Figure 1 shows the single- and double-aligned channeling yields versus depth in silicon for 200-keV proton channeling along the ⟨111⟩ axial direction. The surface peak has been deleted so that the curves start at the minimum yield just behind the surface peak. The single-aligned yield starts at a minimum value of 3.5% dechanneled fraction, whereas the double-alignment yield starts at 0.5%. One of the advantages of double-alignment defect analysis is this extra sensitivity to interstitials that results from its lower-surface minimum yield (a factor of 7 lower than single alignment in the present case.) It will be noted, however, that as the particles penetrate to larger depths, the double-alignment yield rapidly approaches that of single alignment and, in fact, at ~5300 Å is seen to cross and then rapidly exceed the single-aligned yield. Physically, this is an anomalous result, since the double-aligned yield should reflect a lower dechanneled fraction across the entire depth of analysis. Figure 2 shows the single- and double-

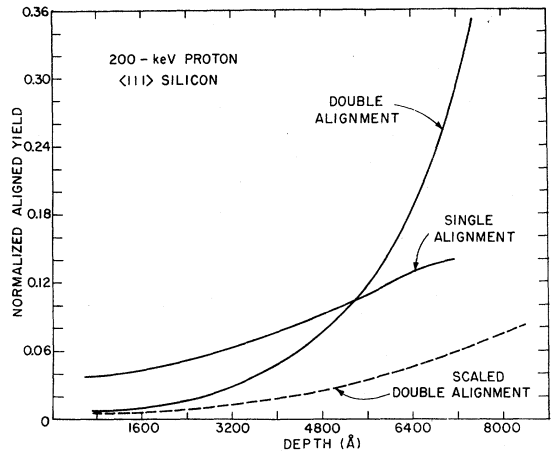

FIG. 1. Single- and double-alignment scattering yields as a function of depth for 200-keV proton axial channeling in ⟨111⟩ silicon. Also shown is the scaled version of the double-aligned scattering yield versus depth, as discussed in the text. For single alignment $dE/d\chi = 9.7\times10^5$ keV/cm and for double alignment $dE/d\chi = 8.7\times10^5$ keV/cm.

aligned yields for 200-keV-proton channeling in the ⟨100⟩ axial direction of gold. Again, the minimum yields near the surface differ from 5.8% for single alignment to 0.4% for double alignment. This is a factor of 14.5 lower for the double-alignment case. As the depth of analysis increases, the double-aligned yield again increases more rapidly than that of single alignment and becomes equivalent to the single-aligned yield at a depth of ~3800 Å. This rapidly increasing double-aligned yield as a function of depth is again anomolous inasmuch as the lower yields, compared with single alignment, should be preserved throughout

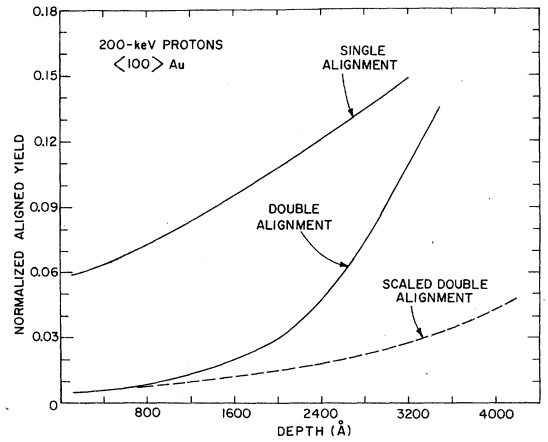

FIG. 2. Same as Fig. 1 except that the results are for the ⟨100⟩ axial direction in gold. For single alignment, $dE/d\chi = 2\times10^6$ keV/cm and in double alignment, $dE/d\chi = 1.8\times10^6$ keV/cm.

the entire depth of analysis. A possible explanation of the double alignment results could be that the stopping power in channels is not the same as for random directions resulting in different depth conversion for single and double alignment. In double alignment, the particles experience predominantly channeling, whereas in single alignment, they experience a combination of channeling and random stopping. In deciding how to handle the stopping power, it is necessary to recognize that most experiments dealing with differences in stopping power for a channeled versus random beam, are performed in transmission through single-crystal foils. In these cases, one compares energy-loss groups for random and aligned crystal orientations. It is generally concluded from these observations that the transmitted channeled particles experience substantially lower stopping power than do the particles in the random beam. However, Bøttiger and Eisen[7] have pointed out, as a result of their experiments, that in transmission it is the low transverse energy ($\epsilon_\perp$) particles that are observed in the channeling group and these will have experienced the lowest stopping power appropriate to the channel centers. Those particles of high $\epsilon_\perp$ will be penetrating near to the channel walls and thus experience stopping powers much more like the random value. It is these high $\epsilon_\perp$ particles that will dechannel and enter the random beam. They conclude from their observations that stopping powers similar to the random value should be used in dechanneling experiments. For the case of double alignment in undamaged thermally vibrating lattices, all particles enter the blocking configuration through collisions with string atoms. This may occur either from a particle in the random beam scattering into the exit blocking direction or a channeled atom reversing its direction (within the limits of the critical angle under uniaxial conditions) by scattering from a fluctuating string atom. Regardless of how the event occurs, the essential feature is that all the particles entering the blocking trajectory will have high transverse energies and therefore experience stopping powers similar to the random value. Thus in comparing dechanneling spectra with blocking spectra for moderate depths in undamaged crystals, both spectra should be analyzed with essentially the random value of the stopping power. Corrections for stopping-power differences may occur when particles are scattered into blocking configurations from interstitial atoms, since then they could originate with any value of transverse energy. Likewise, particles starting with low $\epsilon_\perp$ will increase their transverse energy, through multiple scattering, as a function of depth and eventually will undergo dechanneling or scattering

into the blocking direction being observed. This process will occur in the presence or absence of displaced atoms and could result in a difference in stopping power when comparing single- and double-alignment yields. This is because the outgoing beam will have a true random value in single alignment whereas in double alignment some averaged value will be in effect depending on the initial and final $\epsilon_\perp$ states. It is estimated from previous work[7] that this effect will amount to, at most, a 10% lower $dE/d\chi$ for double alignment, compared to single alignment in both the damaged and undamaged situation. Experimentally, we observe that in silicon crystals having a well-defined boundary between defective and nondefective regions, we obtain similar boundary depths in single or double alignment, using an approximate random value of the stopping power. Analysis of the present data is done with an energy-averaged value of stopping power for single alignment and a 10% lower value for double alignment. The numerical values involved are given in the captions of Figs. 1 and 2.

The origin of the observed excess double-aligned yield in Figs. 1 and 2 must therefore be sought in some other property of the system, rather than stopping power effects. In the following discussion the problem is examined in terms of the energy dependence of scattering into blocking directions. Analytical corrections to the anomalous double-aligned yields can be made and are discussed below in terms of theoretical predictions and the rule of reversibility.

DISCUSSION OF RESULTS

In comparing our results with theory, the first treatment is to calculate experimental values of h from Eq. (1), as a function of depth for the undamaged crystals. These values for silicon ⟨111⟩ and for gold ⟨100⟩ axial directions are presented in Figs. 3 and 4, respectively. The minimum value of h starts at the surface and increases as a regular function with depth for both specimens. The near-surface values of 4 for silicon and 1.4 for gold are in general agreement with theory.[5,8] The increase of h with depth is contrary to theory (which predicts a constant) and is a consequence of the anomalously rapid increase in χ_2 with depth. This result could be interpreted in terms of a breakdown of the reversibility rule since the observed yields as a function of depth must be considered in terms of the energy dependence of crystal-lattice dechanneling and scattering into blocking directions. This problem has recently been examined by Campisano *et al.*,[9] and they show that channeling and blocking, as a function of depth, may yield equivalent spectra if the energy at which a particle enters the channel is taken into account. They make use of this concept by applying z/E scaling to adjust the blocking data and thereby achieve reversibility. This procedure is invoked, since the ingoing channeling beam starts at a well-defined energy (i.e., the incident-beam energy) whereas the scattered particles, entering the channels for the blocking process, do so with a continuum of energies that correspond to the energy of the particles at the point where the scattering collision occurs. The excessively rapid rise of the double-aligned yield is thought to occur because the energy at which particles enter the blocking situation, at depth, is much lower than that of the incident channeling beam. Therefore, it is necessary to calculate, through the observed energy spectra, the energy at which particles enter the blocking configuration on their exit trajectory from the specimen. The depth at which a given yield is observed is then scaled from z to $zE_0/E(z)$, where E_0 is the initial energy and $E(z)$

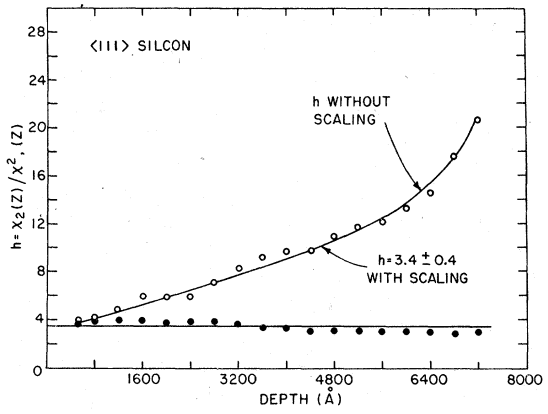

FIG. 3. Experimental values of h for ⟨111⟩ silicon determined from the spectra of Fig. 1 with and without scaling of the double-aligned spectra.

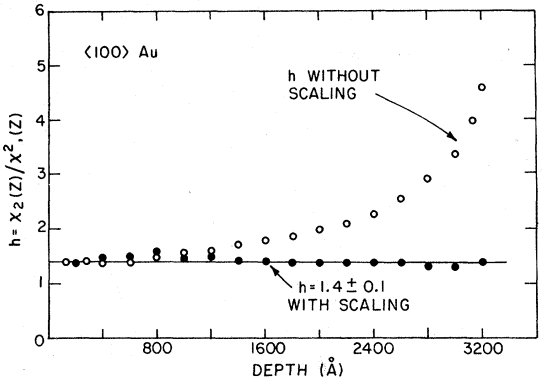

FIG. 4. Same as Fig. 3 except results are for ⟨100⟩ gold.

is the energy at the beginning of the blocking path. [$E(z) = E_i + z\,(dE/dz)$, where E_i is the observed energy and dE/dz the stopping power.] This produces an effective horizontal scaling of the double-aligned yield. The single-alignment yields do not require this correction since all particles enter channels at the same energy.

Application of the z/E scaling law to our data for silicon and gold yields the results shown in Figs. 1 and 2 for the scaled channeling yields and Figs. 3 and 4 for the computed values of h versus depth. It is seen that the double-aligned yields are always lower than those of single alignment and the h values of Figs. 3 and 4 now remain constant with depth as predicted by the rule of reversibility. Thus, one may conclude that the double-aligned yields, when appropriately scaled, are not anomalous and should be applicable to a calculation of χ_R as expressed in Eq. (5).

The adequacy of Eq. (5), under the scaling law, can be tested independent of the above h calculation since it is derived from a simultaneous solution of Eqs. (1) and (2) with no assumptions made for the relative contributions of dechanneling and direct scattering to the observed yields. Equation (5) can therefore be applied equally well to a damaged or nondamaged crystal. In the case of a nondamaged crystal, the calculated value of χ_R based on the combination of χ_1, the scaled values of χ_2, and a constant h should be the same as the experimentally observed value of χ_1, since all the yield, in this case, is from thermal dechanneling and none from direct interstitial-type scattering. The results of applying Eq. (5) to the nondamaged crystals at 300 K are shown by the points in Fig. 5. It is seen that the points, for both crystals, lie on or quite near the curves defining the single-aligned dechanneling yields. This demonstrates self-consistency of the equations with reversibility and the scaling procedure. The result of calculating χ_R through Eq. (5) will depend to some extent on the values of stopping power used for single and double alignment. The average value of h also depends on these parameters through the scaling operation. Thus, the points in Fig. 5 are subject to some uncertainties. The error bars represent effects introduced by the rms error in h as calculated in Figs. 3 and 4. It is seen that uncertainties in h introduce magnified errors at larger depths and that under the conditions of the present experiments, analysis should not be attempted much beyond the half-micron range.

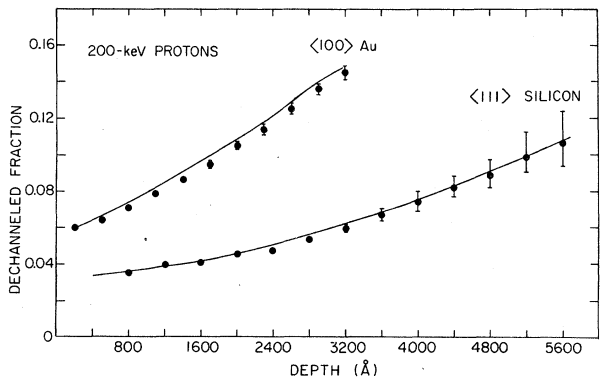

FIG. 5. Comparison of measured single-alignment thermal dechanneling for Si ⟨111⟩ and Au ⟨100⟩ with that predicted by Eq. (5) for the dechanneled fraction from a combination of single- and double-alignment data. The calculated results (solid circles) are obtained using the scaled values of χ_2 and their associated constant h parameters. The solid line is the measured single-alignment yields. Error bars demonstrate effect of rms deviation in average constant value of h.

The scaling which is outlined above is presented as an empirical process that results in achieving apparent reversibility in relation to the associated theoretical equations. However, little has been said thus far about the nature of the correction and why it is necessary. One approach is to consider the processes by which the observed yields are generated. Campisano *et al.*[9] have done this and point out that in single alignment the arrival of a particle at the detector occurs after two distinct events (i.e., dechanneling and backscattering) whereas in double alignment only a single wide-angle event is necessary. The process by which a spectrum is then constructed is very different for each configuration, since when particles exit from a channel with a given energy, they return to the single-aligned detector with a continuum of energies, whereas when particles scatter into channels, they all arrive at the double-aligned detector with a single energy determined by the energy at which they scattered. It is not clear that this explanation is in itself sufficient to account for the effects being observed. Beyond the details of the discrete events occurring, it must be recognized that an essential consideration is the reversibility rule as proposed by Lindhard[10] where he argues that the motion of a particle in a nondissipative crystal lattice can be reversed according to the principle of conservative mechanics. In such a case, point-scattering probabilities are equal for the incident path, compared to the reversed path when the particle finds itself at sites of equivalent potential. This leads to the statistical mechanical argument that a flux of particles moving from A to B and undergoing some process at B with a certain cross section would experience the same process at A with the same cross section if particle emission occurred from B. These precepts are meaningful only under

conservative motion along the penetration direction of the particle. Since in the present work substantial energy loss of the particle occurs (10–20 eV/Å), reversibility is seriously violated. Application of any theory requiring reversibility must therefore be corrected for energy loss effects on the particle's trajectory and scattering cross section at depth. As a channeled particle slows down, its oscillating wavelength becomes shorter and its cross section for multiple scattering, dechanneling, and wide-angle scattering increases. The equations governing the comparison of single and double alignment (or dechanneling and blocking) must therefore be appropriately modified or, alternatively, the observed spectra corrected in some systematic way in order to take this effect into account.

CONCLUSION

It is shown that simultaneous observation of single- and double-alignment backscattering can, in principle, be used for the unique separation of the dechanneled from the directly scattered component of a channeled beam. Proper depth analysis can only be accomplished, however, through appropriate energy scaling that achieves apparent reversibility in the scattering equations. This result makes it possible to apply the method to problems in radiation damage where it is necessary to determine the relative contribution to scattering from displaced atoms that are a distance greater than a_F (the Thomas-Fermi screening distance) from the lattice rows, compared with misaligned atoms that are displaced by distances comparable to the screening distance.

ACKNOWLEDGMENTS

Discussions with R. Behrisch, G. Foti and K. L. Merkle constituted helpful input to this work.

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*Work supported by the U. S. ERDA.

[1]E. Bøgh, Can. J. Phys. 46, 653 (1968).

[2]F. H. Eisen, *Channeling*, edited by D. V. Morgan (Wiley, London, 1973), Chap. 14, p. 415.

[3]P. P. Pronko, M. D. Rechtin, G. Foti, L. Csepregi, E. F. Kennedy, and J. W. Mayer, in Proceedings of the International Conference on Ion Implantation in Semiconductors and Other Materials, Boulder, Colorado (Plenum, New York, to be published).

[4]R. Behrisch, B. M. U. Scherzer, H. Schulze, Radiat. Eff. 13, 33 (1972); also R. Behrisch and J. Roth, *Proceedings of the International Conference on Ion Beam Surface Layer Analysis*, edited by O. Mayer, G. Linker, and F. Kaeppeler (Plenum, New York, 1975), Vol. 2, p. 539.

[5]B. R. Appelton and L. C. Feldman, *Proceedings of the International Conference on Atomic Collisions in Solids* (North-Holland, Amsterdam, 1970), p. 417.

[6]L. C. Northcliffe and R. F. Schilling, Nucl. Data Tables 7, 233 (1970).

[7]J. Bøttiger and F. H. Eisen, Thin Solid Films 19, 239 (1973).

[8]J. H. Barrett, Phys. Rev. B 3, 1527 (1971).

[9]S. U. Campisano, G. Foti, F. Grasso, and E. Rimini, *Atomic Collisions in Solids*, edited by S. Datz, B. R. Appleton, and C. D. Moak (Plenum, New York, 1975), p. 749.

[10]J. Lindhard, K. Dan. Vidensk. Selsk. Mat.-Fys. Medd. 34, No. 14 (1965).