

Energy Loss by keV Ions in Silicon

H. O. Funsten, S. M. Ritzau,* R. W. Harper, and J. E. Borovsky
Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA

R. E. Johnson

University of Virginia, Charlottesville, Virginia 22904, USA
(Received 16 September 2003; published 24 May 2004)

Using silicon photodiodes with an ultrathin passivation layer, the average total energy lost to silicon target electrons (electronic stopping) by incident low energy ions and the recoil target atoms they generate is directly measured. We find that the total electronic energy deposition and the ratio of the total nuclear to electronic stopping powers for the incident ions and their recoils each follow a simple, universal representation, thus enabling systematic prediction of ion-induced effects in silicon. We also observe a velocity threshold at 0.05 a.u. for the onset of electronic stopping.

DOI: 10.1103/PhysRevLett.92.213201

PACS numbers: 34.50.Bw, 61.80.Jh, 61.82.Fk, 79.20.Rf

The processes by which an energetic ion loses energy as it traverses a solid have been the focus of considerable study for nearly a century [1–6] with the goal of systematically understanding and predicting the response of materials to ion irradiation. The energy loss is typically separated into electronic and nuclear processes [1,2]. That is, an incident ion transfers energy to target electrons, resulting in excitations and ionizations in the target material, and to target nuclei by repulsive (Rutherford-type) interactions. Because the latter process generates recoil atoms that also lose energy by electronic and nuclear stopping, the separation of total energy loss into electronic and nuclear components to predict the combined effects of ion irradiation of solids is quite complex.

Although nuclear and electronic energy loss govern important effects such as scattering [7], damage [8], sputtering [9], ion range [3], secondary electron emission [10], and fullerene fragmentation [11], direct measurements of the partitioning of the energy loss between nuclear and electronic processes at velocities less than the Bohr velocity ($v_B = 2.19 \times 10^8$ cm/s = 1 a.u.) have been exceedingly sparse. Further, theoretical and semi-empirical methods are inconsistent [5,12]. For example, while the Lindhard-Scharff-Schiott (LSS) theory [2–4,13] yields a reasonable approximation to the total energy lost to electronic and nuclear stopping processes by an ion (atomic number Z_1) incident on a target material (atomic number Z_2) when $Z_1 = Z_2$, the case for $Z_1 \neq Z_2$ is considerably more complicated and has quite limited applicability [2]. In this study, we directly and accurately measure the total energy lost to nuclear and electronic processes in the energy region for which nuclear stopping is significant.

Since an average of one electron-hole pair is generated in silicon for every $E_{eh} \approx 3.7$ eV lost to electronic energy loss processes by an incident ion or a recoil [14], measurement of the charge generated by an ion in a solid state detector (SSD) operated in a single particle detection

mode has been used to evaluate the total energy lost to electronic stopping processes at high incident energies. However, this technique is difficult to use for low ion energies for two reasons. First, SSDs typically have a thick dead layer (e.g., 400 Å [15]), which causes substantial energy loss or complete stopping of low energy ions before they reach the active region of the detector. Second, when the SSDs are operated in single particle detection mode, the energy measurement of a low energy ion is limited by the small magnitude of the pulse it generates in the device, which can approach the intrinsic noise level of the SSD and its electronics.

In this study we use silicon photodiodes having an SiO₂ passivation layer ranging in thickness from 40 to 60 Å [16] to accurately measure ion-induced charge generation and, therefore, the total electronic energy loss in silicon by low energy ions and their recoils. An additional benefit of these devices is a 100% quantum collection efficiency that eliminates any ambiguity introduced by unknown device recombination processes. Ion beams of H⁺, He⁺, C⁺, N⁺, O⁺, Ne⁺, Ar⁺, and Kr⁺ incident at energy E_0 were directed first into a Faraday cup for measurement of the beam current I_0 and then onto a photodiode [17,18]. The output photodiode current I_{PD} was subsequently measured. The photodiode was operated in current mode without external bias. Increasing I_0 likewise increases I_{PD} relative to the photodiode dark current I_D , enabling accurate measurement of I_{PD} even for low ion energies at which the SSD method fails.

The mean energy $\bar{\eta}$ lost to electronic processes by an incident ion and the recoils it generates over the entire ion range is

$$\bar{\eta} = \frac{(I_{PD} - I_D)E_{eh}}{I_0\tau}, \quad (1)$$

where τ is the fraction of incident ions transmitted through the SiO₂ passivation layer. The correction for ion transmission was small even for slow, heavy ions

(e.g., 14% for 50 keV Kr⁺) and was estimated using TRIM [6]. From Eq. (1), the average fraction of energy lost to electronic stopping processes is $f_E = \bar{\eta}/E_0$, and the average energy lost through nuclear stopping processes per incident ion is simply $\bar{\nu} = E_0 - \bar{\eta}$.

We have found a simple dependence of f_E on the ion's atomic number Z_1 and incident velocity v for those velocities at which the nuclear stopping power is a significant component of the total stopping power. Since f_E should increase with increasing velocity and decreasing mass of the incident ion, we tried to fit the data to numerous functional forms including the reduced parametrization of the LSS theory [3,4]. A good empirical fit of the experimental values of f_E follows the simple form

$$f_E = \frac{A}{Z_1^\zeta} \ln\left(\frac{v}{Bv_B}\right), \quad v < v_M. \quad (2)$$

The velocity v_M above which this simple dependence is no longer valid roughly corresponds to velocities in which the nuclear stopping power becomes small relative to the electronic stopping power and is derived below. The fit of the data to Eq. (2) (excluding H⁺ and He⁺ data for which $v > v_M$) resulted in $A = 0.432 \pm 0.014$, $B = 0.0493 \pm 0.0029$, and $\zeta = 0.202 \pm 0.009$. The correlation coefficient of the fit was $R^2 = 0.976$, implying a remarkably good fit, which is shown as the black line in Fig. 1. The error bars to the left of the data points represent the mean energy lost by an incident ion after transiting the passivation layer as computed using TRIM [6]. We note that charge generated by energy loss in the passivation layer can diffuse to the active region and contribute to I_{PD} [17], so the influence of energy lost in the passivation layer is less than the error bars imply. Over the entire velocity range, the different H and He isotopes have the same value of f_E at the same incident velocity, indicating the absence of an isotope effect.

Figure 2 shows data from this study as well as higher energy results from earlier studies: Ne [19], Si [20], Ar [19], Br [21], Ni [22], Ag [22], Au [22], and U [22]. Over a broad range of ion species and energies (1 keV H⁺ to 26 MeV U⁺) the experimental values of f_E fall close to the line representing Eq. (2). Therefore, Eq. (2) can be used to predict the average *total* energy lost to electronic and nuclear stopping processes by low energy ions in silicon. The red line in Fig. 2 is the estimate for incident Si using the least complex ($Z_1 = Z_2$) solution to the integral equations of the LSS theory governing total nuclear and electronic energy loss in a target [2]. This LSS result agrees well with the Si data when the value of k , which is the friction coefficient for velocity-proportional electronic stopping, is chosen to be 0.13. This value agrees well with the LSS theory prediction of $k = 0.146$ [2].

At high velocity the nuclear stopping power decreases rapidly and becomes small compared to the electronic

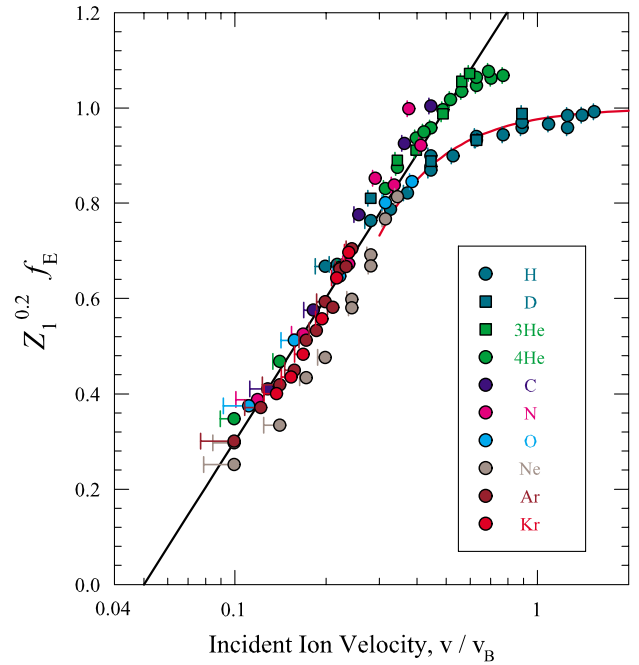


FIG. 1 (color). The quantity $Z_1^{0.20} f_E$, in which Z_1 is the incident ion atomic number and f_E is the fraction of the average total energy $\bar{\eta}$ lost to electronic stopping processes by an incident ion and its silicon recoils, is plotted as a function of incident ion velocity. The black line is the fit of Eq. (2) to the data for $v < v_M$, and the red line represents H⁺ in the high velocity case [Eq. (3)]. The extent of the error bars to the left of the data points corresponds to the calculated energy loss of the incident ion in the SiO₂ passivation layer.

stopping power, and the total energy lost to nuclear stopping processes approaches a constant value $\bar{\nu}_{\max}(Z_1)$ [3]. Therefore,

$$f_E \approx \frac{\bar{\eta}}{\bar{\eta} + \bar{\nu}_{\max}(Z_1)} = 1 - \frac{\bar{\nu}_{\max}(Z_1)}{E_0}, \quad v > v_M, \quad (3)$$

which approaches unity in the high energy limit. This behavior is clearly observed in Fig. 1 for incident H⁺ and D⁺, for which we obtain $\bar{\nu}_{\max} \approx 0.61$ keV for H⁺ (red line in Fig. 1) and $\bar{\nu}_{\max} \approx 1.1$ keV for D⁺. Furthermore, since isotopes with the same incident velocity have the same value of f_E , Eq. (3) yields $\bar{\nu}_{\max}(i)/\bar{\nu}_{\max}(j) = m_i/m_j$, where i and j refer to the different isotopes. For H⁺ and D⁺, the data yield $\bar{\nu}_{\max}(\text{H}^+)/\bar{\nu}_{\max}(\text{D}^+) = 0.55$, in good agreement with their mass ratio of 0.5.

As shown in Fig. 1, extrapolation of f_E to lower velocities using Eq. (2) indicates that $f_E = 0$ at $v = Bv_B = 0.05$ a.u. and, therefore, an ion loses all of its energy to nuclear stopping processes when $v < 0.05$ a.u. This suggests the presence of a velocity threshold for electronic stopping processes, as has been shown for gaseous targets [23,24], nearly free electron metals [25], and a high band gap insulator [26]. For semiconductors, this threshold corresponds to the minimum energy required to excite a valence electron into the conduction band. A related

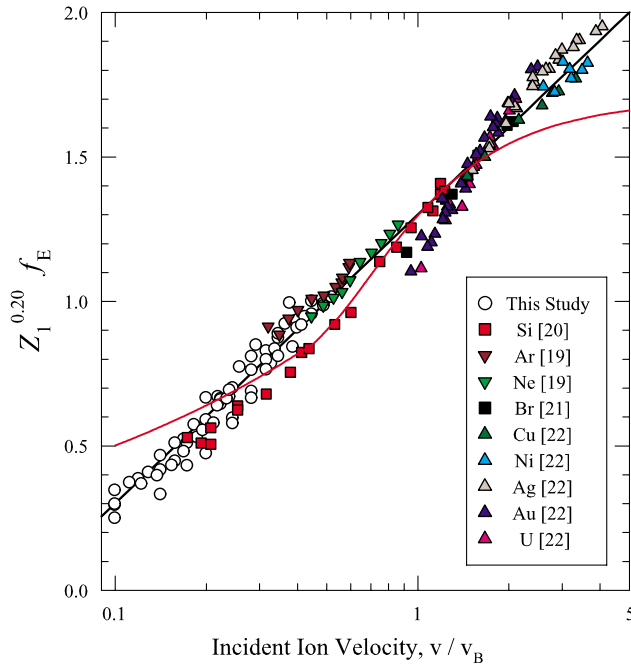


FIG. 2 (color). Similar to Fig. 1, this figure includes data (colored symbols) from earlier studies [19–22] for heavy ions at high energies derived using silicon solid state detectors operated in a single particle counting mode. The open circles represent the data of Fig. 1 for $v < v_M$. All data show surprisingly good agreement with Eq. (2) (black line), spanning 1 keV H to 26 MeV U. The red line is based on LSS theory [2] for incident Si ions (i.e., $Z_1 = Z_2$).

threshold exists for kinetic secondary electron emission from free-electron metals, for which a simple expression for the velocity threshold has been derived [10] based on the maximum energy transfer from an incident ion to a target electron for promotion of the electron across an energy gap. Applying this expression to silicon for excitation of a valence electron across a minimum band gap energy E_g of 1.1 eV (at 300 K) and using a maximum valence electron energy of 12.3 eV [27] yields a threshold velocity for the onset of electronic stopping of 0.02 a.u. This estimate is expected to be lower than the observed threshold of 0.05 a.u. due to the small number of valence electrons having an energy near 12 eV and the low probability of scattering a valence electron in momentum space directly into the conduction band minimum. If the average energy required to generate an electron-hole pair E_{eh} is used instead of E_g , this estimate yields a threshold velocity 0.07 a.u., in reasonably good agreement with the results presented here. Note that an explicit assumption of the LSS theory is that such a threshold does not exist, i.e., $f_E \rightarrow 0$ as $v \rightarrow 0$. Therefore, the LSS theory overestimates f_E for silicon for $v < 0.5v_B$.

The nuclear, S_N , and electronic, S_E , stopping powers describe the average energy *lost* per unit path length by an incident ion to the target material. However, Eq. (2) allows us to extract the relative amount of nuclear and electronic energy actually *deposited* per unit path length

by the incident ion and its recoils, which we write as the total stopping powers S'_N and S'_E . The prime on these quantities indicates the combined energy deposition by the incident ion and its silicon recoils: $S'_N(E) = \int d\sigma_n(T)\bar{\nu}(T)$ and $S'_E(E) = S_E + \int d\sigma_n(T)\bar{\eta}(T)$, where $d\sigma_n(T)$ is the nuclear elastic cross section for producing a recoil of energy T . Noting that $S'_N + S'_E = S_N + S_E$ and using the continuous slowing down approximation in which, on average, $T \ll E$, it is straightforward to show that the integral equations for $\bar{\nu}(E_0)$ and $\bar{\eta}(E_0)$ yield

$$\bar{\eta} = \int_0^{E_0} \frac{S'_E}{S'_E + S'_N} dE. \quad (4)$$

The derivative of Eq. (4) then gives the ratio of the total stopping powers

$$\frac{S'_N(E)}{S'_E(E)} = \frac{1}{d\bar{\eta}/dE} - 1. \quad (5)$$

Using $\bar{\eta} = f_E E$ and the general form in Eq. (2) for f_E yields

$$\frac{S'_N(E)}{S'_E(E)} = \frac{1}{f_E(E) + A/(2Z_1^\xi)} - 1, \quad v < v_M. \quad (6)$$

This ratio is evaluated using the parameters derived from the fit of Eq. (2) to the data.

Equation (6) can be used to study processes such as secondary electron emission and sputtering that arise from the combined effects of an incident ion and its recoils. Since this ratio contains the effects of the recoils, which convert a fraction of the nuclear stopping of the incident ion into electronic stopping, Eq. (6) represents a lower bound for the stopping power ratio S_N/S_E of the incident ion alone.

The velocity v_M below which Eq. (2) is valid can be estimated using Eq. (6). Since the nuclear stopping power rapidly decreases with increasing velocity near v_M , we define v_M as the velocity at which $S'_N/S'_E \rightarrow 0$. This is approximately the same velocity at which $S_N/S_E \rightarrow 0$ because only a very small fraction of the energy is lost to recoils at v_M . The results for Eq. (6) for $S'_N/S'_E \rightarrow 0$ are illustrated in Fig. 3, and we note that v_M can be roughly approximated by $v_M \approx 0.3Z_1^{2/3}v_B$. Equation (6) yields $v_M = 0.30$ a.u. for H^+ and $v_M = 0.43$ a.u. for $^4He^+$, which are consistent with velocities at which H^+ and He^+ are seen in Fig. 1 to begin deviating from the black line ($0.3v_B$ and $0.5v_B$, respectively). In Fig. 2, all of the data not from this study are for $v < v_M$ except for the Ni data, which lie in the vicinity of v_M .

Also from Eq. (6) we can derive the velocity v'_S at which the total nuclear and electronic stopping powers are equal, i.e., $S'_N = S'_E$. This is depicted in Fig. 3, which shows v'_S as a function of Z_1 . Also shown is the velocity $v_S \approx 0.1Z_1^{2/3}v_B$ for which $S_N = S_E$ as approximated by the LSS theory [4]. Our results for H^+ are close to those from the LSS approximation since few energetic recoils are produced by H^+ . At higher values of Z_1 , the

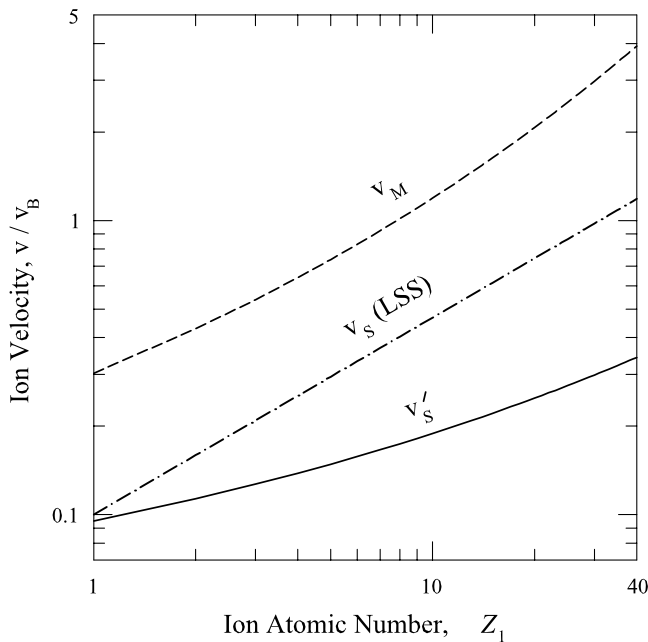


FIG. 3. The velocity v_M (upper dashed line) derived using Eq. (6) and $S'_N/S'_E \rightarrow 0$ is shown as a function of Z_1 . Also shown are the velocity v'_S at which $S'_N = S'_E$ derived using Eq. (6) and the velocity v_S at which $S_N = S_E$ using the LSS approximation $v_S \approx 0.1Z_1^{2/3}v_B$ [4].

difference between v'_S and v_S increases because of the corresponding increase in the production of fast recoils. Note that S'_N/S'_E , which includes the electronic energy lost by recoils, is a lower limit to S_N/S_E , so v'_S is therefore a lower limit to v_S .

Using new detectors having ultrathin dead layers and 100% quantum detection efficiency, we have measured for the first time the average total energy η deposited by electronic energy loss processes in silicon by low velocity ions and their recoils. In contrast to the complexity and limited applicability of the LSS theory for derivation of the total nuclear and electronic energy loss at low velocities, we find that these quantities and the ratio of the total nuclear and electronic stopping powers, which are the most relevant parameters governing ion-induced effects in materials, can be simply and usefully expressed as a function of incident ion velocity and atomic number. This dependence is valid above the apparent threshold velocity of ~ 0.05 a.u. for electronic stopping processes and below the velocity at which the nuclear stopping power becomes insignificant relative to the electronic stopping power.

This work was performed under the auspices of the United States Department of Energy and was supported by NASA Grant No. W-19 866. The authors gratefully thank H. O. Funsten Sr. (College of William and Mary), R. Korde (International Radiation Detectors, Inc.), and S. Datz (ORNL, deceased) for helpful discussions and insights.

*Present address: ON Semiconductor, East Greenwich, RI 02818, USA.

- [1] N. Bohr, *Philos. Mag.* **25**, 10 (1913); **30**, 581 (1915); *K. Dan. Vidensk. Selsk. Mat. Fys. Medd.* **18**, No. 8 (1948).
- [2] J. Lindhard, V. Nielsen, M. Scharff, and P.V. Thomsen, *K. Dan. Vidensk. Selsk. Mat. Fys. Medd.* **33**, No. 10 (1963).
- [3] J. Lindhard, M. Scharff, and H.E. Schiott, *K. Dan. Vidensk. Selsk. Mat. Fys. Medd.* **33**, No. 14 (1963).
- [4] J. Lindhard, V. Nielsen, and M. Scharff, *K. Dan. Vidensk. Selsk. Mat. Fys. Medd.* **36**, No. 10 (1968).
- [5] P. Sigmund, *Nucl. Instrum. Methods Phys. Res., Sect. B* **135**, 1 (1998).
- [6] J. F. Ziegler, J. P. Biersack, and U. Littmark, *Stopping and Range of Ions in Solids* (Pergamon Press, New York, 1985), Vol. 1.
- [7] W. Heiland and E. Taglauer, in *Methods of Experimental Physics*, edited by R. L. Park and M. G. Lagally (Academic, New York, 1985), Vol. 22, pp. 299–348.
- [8] T. Diaz de la Rubia and G. H. Gilmer, *Phys. Rev. Lett.* **74**, 2507 (1995).
- [9] H. H. Andersen and H. L. Bay, in *Sputtering by Particle Bombardment I*, edited by R. Behrisch, *Topics in Applied Physics* Vol. 47 (Springer-Verlag, Berlin, 1981), p. 145.
- [10] R. A. Baragiola, E. V. Alonso, and A. Oliva-Florio, *Phys. Rev. B* **19**, 121 (1979); E. V. Alonso, R. A. Baragiola, J. Ferrón, M. M. Jakas, and A. Oliva-Florio, *Phys. Rev. B* **22**, 80 (1980).
- [11] O. Hadjar *et al.*, *Phys. Rev. Lett.* **84**, 4076 (2000); J. Opitz *et al.*, *Phys. Rev. A* **62**, 022705 (2000); T. Kunert and R. Schmidt, *Phys. Rev. Lett.* **86**, 5258 (2001).
- [12] K. Arstila, J. Keinonen, and P. Tikkanen, *Nucl. Instrum. Methods Phys. Res., Sect. B* **101**, 321 (1995); P. Sigmund, *Nucl. Instrum. Methods Phys. Res., Sect. B* **135**, 1 (1998).
- [13] P. Sigmund, *Phys. Scr.* **28**, 257 (1983).
- [14] C. J. Wu and D. B. Wittry, *J. Appl. Phys.* **49**, 2827 (1978).
- [15] H. Grahmann and S. Kalbitzer, *Nucl. Instrum. Methods* **132**, 119 (1976).
- [16] R. Korde and J. Geist, *Appl. Opt.* **26**, 5284 (1987); E. M. Gullikson, R. Korde, L. R. Canfield, and R. E. Vest, *J. Electron Spectrosc. Relat. Phenom.* **80**, 313 (1996).
- [17] H. O. Funsten, D. M. Suszcynsky, S. M. Ritzau, and R. Korde, *IEEE Trans. Nucl. Sci.* **44**, 2561 (1997).
- [18] H. O. Funsten, S. M. Ritzau, R. W. Harper, and R. Korde, *IEEE Trans. Nucl. Sci.* **48**, 1785 (2001).
- [19] T. Karcher and N. Witherspoon, *Nucl. Instrum. Methods* **93**, 519 (1971).
- [20] A. R. Sattler, *Phys. Rev.* **138**, A1815 (1965).
- [21] L. Cliché, S. C. Gujrathi, and L. A. Hamel, *Nucl. Instrum. Methods Phys. Res., Sect. B* **45**, 270 (1990).
- [22] B. D. Wilkins *et al.*, *Nucl. Instrum. Methods* **92**, 381 (1971).
- [23] D. Semrad, *Phys. Rev. A* **33**, 1646 (1986).
- [24] R. Cabrera-Trujillo, J. R. Sabin, Y. Öhrn, and E. Deumens, *Phys. Rev. Lett.* **84**, 5300 (2000).
- [25] J. E. Valdés, J. C. Eckardt, G. H. Lantschner, and N. R. Arista, *Phys. Rev. A* **49**, 1083 (1994).
- [26] C. Auth, A. Mertens, H. Winter, and A. Borisov, *Phys. Rev. Lett.* **81**, 4831 (1998).
- [27] J. R. Chelikowsky and M. L. Cohen, *Phys. Rev. B* **10**, 5095 (1974).