NONLINEAR RESPONSE OF SI DETECTORS FOR LOW-Z IONS

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We report measurements of the relative mean pulse height produced by ¹H, ⁴He, ⁷Li and ¹⁶O ions in Si surface barrier detectors. The data are anomalous in that the pulse height for different ions of the same energy (after window and nonionizing losses are subtracted) increases with atomic number, contrary to observations for fission fragments. A simple model invoking a stopping power dependence of the energy required to create an electron-hole pair is consistent with all experimental data, suggesting that the response of Si detectors is nonlinear with particle energy.

1. Introduction

It has been reported [1-5] that the pulse-height response of Si semiconductor detectors is, for a given energy, larger for ⁴He particles than for protons. The magnitude of this effect appears to be independent of the projectile mass for ions of the same atomic number (Z) and not to depend on the detector type. Since the so-called pulse height defect, observed for e.g. fission fragments [6], increases with the mass and Z of the particle, the low-Z behaviour observed is anomalous. It is thus not clear whether this difference reflects a fundamental difference in the energy required to produce an electron-hole pair, ϵ , for various particles, or whether other factors related to the modes of energy loss in the sensitive volume of the detector may be responsible.

To further investigate this anomaly, we report here measurements of the variation in pulse height produced by incident ¹H, ⁴He, ⁷Li and ¹⁶O ions at low energies for two detector types. Such measurements require a precise determination of the energy lost in the window and dead layer regions, and a knowledge of the energy expended in nonionizing processes in the sensitive volume itself. By comparing the results for ⁴He and ⁷Li ions at the same energy, effects related to e.g. range differences should be smaller than for the ¹H-⁴He comparison.

2. Experiment

The experiment involved two measurements: (1) the ion velocity was measured by a time-of-flight system,

 Permanent address: GSI, Postfach 110541 D-6100 Darmstadt, FRG. and (2) the pulse height in the detector was determined relative to that produced by a convenient α -decaying radio-isotope, e.g. ²⁴⁴Cm, ²⁴¹Am or ²³⁹Pu. The time-of-flight technique to determine the projectile kinetic energy, $E_{\rm K}$, has been used previously in fission fragment measurements (see ref. [6]).

Beams of ¹H, ⁴He, ⁷Li and ¹⁶O ions produced by the CRNL 2 MV High Voltage Mass Separator first traversed a thin carbon foil, thereby liberating secondary electrons which impinged on a microchannel plate detector to generate a timing signal. A second timing signal was obtained from the Si detector under study, fitted with an Ortec VT100 fast preamplifier. These signals were fed to a time-to-amplitude convertor which was calibrated with an Ortec precision TAC calibrator. The Si detector was then translated collinear to the beam by an accurately known distance (16.25 cm) and the flight time again measured. The distance of travel was determined in situ to an accuracy of ±0.005 cm with precision dial indicators positioned at both extreme positions.

The preamplifier of the Si detector was changed to an Ortec 109A for the pulse height determinations. The corresponding pulse height was then measured relative to those produced by a mixed α source (239 Pu, 241 Am, 244 Cm with corresponding principal α energies of 5157, 5486, 5805 keV, respectively [7]) which the detector viewed simultaneously. A precision pulse generator, whose linearity was measured independently using γ -ray sources and a Ge detector, was employed for the relative pulse height measurements. Several measurements of the pulse height and the particle velocity were performed alternately. The energy resolution was 14 keV (fwhm) for 1 MeV protons.

Two Si detectors were used in these measurements:

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(1) a 50 mm² partially-depleted surface barrier detector of 100 μ m depletion depth (Serial No. Ortec 23-132A) having a Au window of 40 μ g cm⁻² nominal thickness; and (2) a totally depleted 100 μ m Si detector having a Au window on one side and an Al window on the other (Serial No. Ortec 22-704E), both of 40 μ g cm⁻² nominal thickness. The resistivity of both detectors was 1. 1 k Ω -cm. The detectors were collimated to expose a central area of ~5 mm².

The detectors were mounted in a rotatable housing controlled by a stepping motor which allowed a rotation of $\pm 45^{\circ}$ about an axis normal to the beam direction and lying in the plane of the detector surface as confirmed by a distant telescope observation. This facility allowed a determination of the rate of change of pulse height with angle, $dE/d(\sec\theta)$, which yields the window loss, $\Delta E_{\rm w}$, directly. $\Delta E_{\rm w}$ was measured for each ion at each energy. The detector was held at constant temperature (293 ± 1 K) by cooling the detector housing to offset the heat generated by the stepping motor.

Energy deposited in atomic motion, ΔE_n , is not available for creating electron-hole pairs and is therefore subtracted from the incident particle energy, E_K . ΔE_n was calculated by standard procedures [8] in four ways, with two values for the nuclear stopping and two for the electronic stopping. The nuclear stopping values were derived from fits to Lindhard's Thomas-Fermi value [9] and to Kalbitzer et al.'s [10] approximation to the Wilson-Haggmark-Biersack potential. The electronic stopping values were taken from: (1) Northcliffe and Schilling [11], interpolating linearly in Z, and (2) Andersen and Ziegler [12] and Ziegler [13] for $^1\mathrm{H}$ and

⁴He, respectively, where the effective-charge parameterization discussed in ref. [12] has been applied for higher-Z projectiles. The electronic stopping of the Si recoils was taken as the Lindhard velocity – proportional value throughout; use of the Northcliffe and Schilling values produced a negligible change in the ¹⁶O results, and thus would be even less significant for the lighter projectiles. Our results are, within experimental uncertainty, the same for all four calculations of ΔE_n .

The pulse height data were analyzed for their mean values; for ⁷Li and ¹⁶O ions, the most probable pulse height exceeds the mean pulse height by a measurable amount at the lower energies.

We have searched for effects resulting from instrument malfunction by using different amplifiers, including varying the time constants $(1-5 \ \mu s)$. Like other investigators [14,15], we found that all our results are internally consistent. The response of one of the detectors (23-132A) to a change in temperature was measured for 5.486 MeV ⁴He ions in the range 245K < T < 293 K. We found the relation $d(\ln E)/dT = 1.73(\pm 0.10) \times 10^{-4} K^{-1}$, consistent with the result of Pehl et al. [15], 1.80×10^{-4} K⁻¹.

3. Results

Fig. 1 shows typical time-of-flight spectra for ⁴He ions at ~ 2.5 MeV; the time resolution is 380 ns (fwhm). Using the known distance (16.25 cm) that the detector was moved between the two measurements shown, we obtain $E_{\rm K} = 2456.8(\pm 5.1)$ keV from these data.

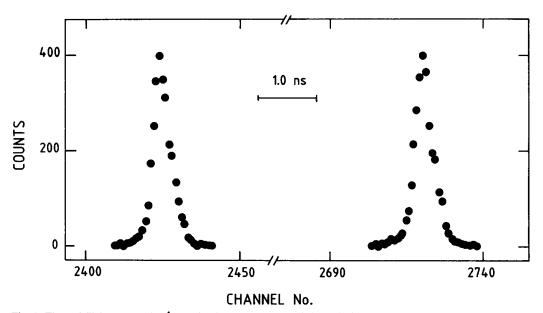


Fig. 1. Time-of-flight spectra for ⁴He. The detector was moved through $\Delta S = 16.25$ cm. The time resolution (fwhm) is 380 ps. The measured kinetic energy is $E_K = 2456.8 \ (\pm 5.1)$ keV derived from these data.

To determine $\Delta E_{\rm w}$, it was first necessary to find the normal to the detector surface. Fig. 2 shows data for ⁴He ions incident on detector 23-132A. To obtain the ADC dispersion (keV/channel) appropriate to the tilting data, we use the pulse height from ²⁴¹Am ($E_0 = 5486$ keV) which corresponds to a deposited energy of 5460 keV after correction for $\Delta E_n = 11$ keV, source thickness ~ 2 keV and $\Delta E_{\rm w} = 13$ keV.

The above procedure is repeated at several energies,

yielding results as shown in table 1. We then seek a linear relationship between relative mean pulse height and $E_{\rm K}' = E_{\rm K} - \Delta E_{\rm w} - \Delta E_{\rm n}$. In the fitting procedure, we have chosen $E_{\rm K}'$ as the dependent variable since the relative uncertainties in $E_{\rm K}'$ are larger than for the pulse height values. The values shown in the last column of table 1, $\delta E_{\rm K}'$, are the difference between the measured and fitted results. All data appear to yield linear relations with intercepts consistent with zero.

Table 1 Examples of pulse height data for 1 H, 4 He, 7 Li and 16 O ions incident on Si detectors. The nonionizing contribution, ΔE_n , shown in column 4 is obtained from a calculation using Northcliffe and Schilling electronic stopping (ref. [11]) and Lindhard's Thomas-Fermi nuclear stopping (ref. [9]), see text. The uncertainties are shown in parentheses. The values $\delta E_K'$ shown in the last column are the differences between the fitted and derived values of $E_K' = E_K - \Delta E_W - \Delta E_n$

Projectile	E _K	$\Delta E_{\rm w}$	ΔE_n	Relative	δΕ' _K
(detector)	(keV)	(keV)	(keV)	mean P.H.	(keV)
¹ H	547.3(1.3)	8.4(1.0)	1.5	98.24	-1.5
(22-704E, Al)	571.8(1.2)	8.2(1.0)	1.6	102.01	2.4
	776.1(2.2)	7.0(0.8)	1.7	139.19	3.1
	775.3(1.1)	7.0(0.8)	1.7	139.73	-0.6
	989.9(1.4)	6.0(0.7)	1.8	179.13	2.2
	998.9(2.9)	6.0(0.7)	1.9	180.32	0.3
	1138.8(2.7)	5.5(0.7)	1.9	206.69	-4.6
	1193.9(3.5)	5.4(0.6)	2.0	215.21	3.8
	1391.8(5.0)	4.9(0.6)	2.1	251.51	2.2
	1485.1(2.6)	4.7(0.6)	2.1	268.84	0.2
	1552.5(4.0)	4.5(0.5)	2.2	280.39	4.1
⁴ He	531.6(1.1)	19.7(1.2)	7.9	93.06	0.2
(22-704E, Au)	589.9(2.1)	20.4(1.2)	8.0	102.94	4.1
, , ,	781.4(1.1)	21.7(1.3)	8.2	138.34	1.7
	808.9(1.1)	21.8(1.3)	8.3	143.84	-0.7
	1075.6(2.4)	22.0(1.3)	8.6	192.77	-0.3
	1227.5(1.2)	21.8(1.3)	8.8	221.42	-3.7
	1268.0(1.8)	21.7(1.3)	8.8	228.10	0.5
	1570.2(1.6)	20.7(1.2)	9.1	284.04	-0.3
	1574.1(1.8)	20.7(1.2)	9.1	285.20	-2.7
	1584.9(1.1)	20.7(1.2)	9.1	287.01	-1.7
	2162.7(2.2)	18.8(1.1)	9.6	392.66	3.9
	2655.6(7.0)	17.4(1.0)	9.9	483.73	3.3
	2962.3(7.8)	16.7(1.0)	10.1	539.96	5.2
⁷ Li					
(22-704E, Au)	578.0(1.1)	24.4(1.5)	16.8	99.15	-0.5
, , ,	675.5(0.5)	26.3(1.6)	17.2	116.80	-0.3
	971.6(1.6)	30.5(1.8)	18.0	170.60	1.2
	972.5(0.5)	30.6(1.8)	18.0	170.87	0.6
	1268.7(0.9)	33.4(2.0)	18.6	225.22	0.8
	1278.1(0.7)	33.5(2.0)	18.6	227.21	-0.6
	1521.1(0.9)	35.4(2.1)	19.0	271.92	-0.6
	1567.1(1.7)	35.7(2.1)	19.1	280.35	-0.4
¹⁶ O					
(23-132A, Au)	730.0(1.0)	51.1(3.1)	59.9	113.21	-0.7
	1202.4(1.2)	68.4(4.1)	66.4	196.98	1.9
	1924.3(1.3)	90.0(5.4)	71.7	327.87	-0.3
	2522.9(1.8)	104.3(6.3)	74.5	437.18	-0.9

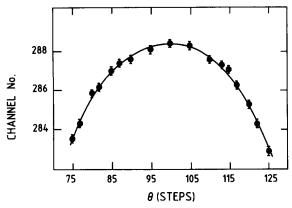


Fig. 2. Pulse height data as a function of detector tilt to find the normal to the detector surface. The stepping motor calibration is 1.800° /step. The smooth curve is a fit: $C = P_1 - P_2$ sec $(\theta - \theta_0)$, where C is the channel number. These data are for $\sim 500 \text{ keV}^4$ He ions incident on detector 23-132A.

In order to obtain reliable results for the energy per electron-hole pair, it is necessary that the applied bias voltage be sufficiently large to ensure complete charge collection, but small enough to avoid charge multiplication. We have operated the detectors with fields $< 10^4$ V cm⁻¹, to avoid the multiplication problem for surface barrier detectors [6]. However, the bias voltage was large enough to be in the so-called plateau region [1,2].

We have observed the effect of plasma recombination for both ⁷Li and ¹⁶O projectiles at low detector bias voltage. Table 2 shows the pulse heights observed for different ions relative to the ⁴He pulse height from ²⁴¹Am as a function of the bias voltage. The data were measured only for particles incident on the Au window side of both detectors.

Although the pulse height for 1.5 MeV ¹H projectiles

Table 2 Results for pulse height as a function of detector bias, relative to the 241 Am α source

Detector	Projectile	Approx. energy (keV)	Bias (Volts)	Relative pulse height
23-132A	Н	1250	50	226.64(0.03)
(partially depleted)	Н	1250	80	226.70(0.03)
	O	1150	25	195.98(0.2)
	0	1150	50	197.25(0.2)
	О	1150	75	197.34(0.2)
22-704E (Au)	Н	1500	25	271.35(0.04)
(totally depleted)	H	1500	50	271.37(0.04)
	Н	1500	80	271.41(0.04)
	Li	700	25	116.16(0.07)
	Li	700	80	116.55(0.07)

is almost the same at 25 and 80 V bias, the pulse height for ⁷Li projectiles is measurably less at 25 V than at 80 V bias. Similarly, a larger effect is seen in the ¹⁶O-¹ H comparison for 23-132A. At low bias voltage, the weaker electric field cannot collect all the charge before recombination occurs for the more highly ionizing radiation from heavier ions. We point out that the data in table 2 must be interpreted with care since the pulse heights are all measured relative to that produced by the 5.48 MeV alphas emitted by ²⁴¹Am for the same bias voltage (i.e. the measurements are made simultaneously).

4. Discussion

We describe our results in terms of the slopes, C, of the mean pulse height versus the deposited energy (E_{κ} $-\Delta E_{\rm w} - \Delta E_{\rm n}$) linear graphs, since all the data are consistent with a linear relationship between these two quantities. Thus, $C(^{1}H)$, $C(^{4}He)$,... refer to ^{1}H , ⁴He,... data. Our interest is in the slope ratios, since the experiment is not capable of yielding €-values on an absolute scale. Within the measurement uncertainties, we observe no differences among the detector type or configuration. We find $C(^{1}\text{H})/C(^{4}\text{He}) = 0.986 \pm 0.002$, $C(^{1}\text{H})/C(^{7}\text{Li}) = 0.975 \pm 0.004$ and $C(^{1}\text{H})/C(^{16}\text{O}) =$ 0.965 + 0.005, as shown in the last line of table 3. The ratio $C(^{1}H)/C(Z)$ may be identified with the ratio $\epsilon(Z)/\epsilon(^{1}H)$ [2], i.e. the ratio of the energies required to produce an electron-hole pair in Si by a projectile with atomic number Z, and by ¹H. Fig. 3 shows this ratio as a function of Z, determined in this experiment. It has been shown earlier [2] that $\epsilon(Z)$ does not depend on projectile mass for the light ion pairs ¹H, ²H and ³He, ⁴He. There is a marked decrease in apparent ϵ -values as Z increases, but the effect appears to saturate. For even heavier ions than ¹⁶O, it is well-known [15,16] that the pulse height decreases with increasing Z, which confirms the saturation effect observed here. For ¹⁶O ions, the effect exceeds 3%.

It thus appears that the energy to create an electron-hole pair in Si is a function of the charge of the ion. Such an effect has not been observed in Ge detectors, although the data are sparse [4]. Presumably this effect is present for all Z but may be masked by plasma recombination. Mitchell et al. [2] and Pehl et al. [15] have discussed and dismissed contributions due to electron-hole trapping, X-ray production and secondary electron escape. As our data have shown, the effect is not dependent on the detector type or window material but is dependent on Z and increases with increasing Z of the ion.

As an explanation for the Z-dependence of pulse height observed in this experiment, we suggest that ϵ is a function of the ionization density in the detector. Specifically, we propose that ϵ decreases as the elec-

tronic stopping power, dE/dx, increases, reflecting a smaller energy required to produce an electron-hole pair in the region where one has already been created. This effect appears plausible since ϵ_0 for Si is 3.67 eV/electron-hole pair (the subscript zero indicates the value derived from γ -ray measurements), whereas the stopping power of ⁴He in Si at 1 MeV is \sim 310 eV/nm. Thus, ⁴He ions lose \sim 80 eV of energy in each "atomic layer", assuming 0.25 nm per layer. This energy loss is shared among several atoms, and it seems reasonable to assume that ϵ is not independent of dE/dx.

We have modelled this effect by calculating the (relative) number of electron-hole pairs, $N_{\rm eh}$, produced by a projectile of incident energy E_0 , using

$$N_{\rm eh}(E_0) = \int_0^{E_0} \frac{\mathrm{d}E}{\epsilon_0 + k(\mathrm{d}E/\mathrm{d}x)}.$$
 (1)

Here, k was taken to be independent of Z, and dE/dx (electronic stopping, dE/dx < 0) values were taken from the literature [11–13,18]. For each ion (Z = 1, 2, 3, 8), $N_{\rm eh}$ was calculated in the energy region investigated here, and a linear fit was made to the $N(E_0)$ versus E_0 results.

Although the above supposition introduces a non-linearity into the energy response of the detector, we found that this nonlinearity was below the measurement errors of the experiment. Our results, using k = 0.0002 nm/electron-hole pair, are:

$$C(^{1}\text{H})/C(^{4}\text{He}) = 0.988(\pm 0.003)$$

 $C(^{1}\text{H})/C(^{7}\text{Li}) = 0.976(\pm 0.006)$
 $C(^{1}\text{H})/C(^{16}\text{O}) = 0.936(\pm 0.016).$

We show the uncertainties in the calculated results to illustrate the sensitivity to a 25% change (i.e. ± 0.00005) in the magnitude of k. If we assume that the competing effect of plasma recombination is nonnegligible for the 16 O case, then the agreement between the model calculations and data is satisfactory (see table 3).

Table 3
Summary of ratios of the slopes fitted to the mean relative pulse height versus deposited energy data. The uncertainties are shown in parentheses

$C(^{1}\mathrm{H})/C(^{4}\mathrm{He})$	$C(^{1}\mathrm{H})/C(^{7}\mathrm{Li})$	$C(^{1}\text{H})/C(^{16}\text{O})$
0.990(0.004)	0.978(0.006)	0.967(0.008)
0.982(0.003)	0.974(0.004)	0.964(0.013)
0.987(0.003)	-	_
0.986(0.002)	0.975(0.003)	0.966(0.007)
	0.990(0.004) 0.982(0.003) 0.987(0.003)	0.982(0.003)

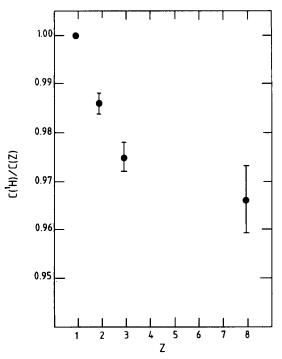


Fig. 3. Apparent electron-hole pair creation energy ratios for ⁴He, ⁷Li and ¹⁶O relative to ¹H, shown as a function of atomic number.

Using the k-value of 0.0002, we find that the magnitude of the product k(dE/dx) amounts to ~ 0.1 eV/electron-hole pair for 1 MeV ⁴He particles, a value much less than $\epsilon_0 = 3.67$ eV. This explains the apparent linear relation between pulse height and deposited energy that is obtained from measurements over any restricted energy region.

To test our model, we have simulated the measurements for relative electron-hole pair creation energies reported in refs. [1-4, 14]. Table 4 shows a summary of

Table 4 Comparison of ϵ -ratios measured to values predicted by our model, eq. (1) in text, with k=0.0002. Uncertainties are shown in parentheses

Ref.	Measured quantity	Experimental	Predicted
[4]	$\epsilon(^{4}\text{He})/\epsilon(^{1}\text{H})$	0.993(0.003)	0.996
[3]	$\epsilon(^{4}\text{He})/\epsilon(^{1}\text{H})$	0.991(0.0025)	0.991
įή	$\epsilon(^{4}\text{He})/\epsilon(^{1}\text{H})$	0.978(0.006)	0.986
[2]	$\epsilon(^4\text{He})/\epsilon(^1\text{H})$	0.989(0.003)	0.991
	$\epsilon(^2D)/\epsilon(^1H)$	1.000(0.001)	0.9990
	$\epsilon(^{3}\text{He})/\epsilon(^{4}\text{He})$	1.000(0.001)	1.0006
This work	$\epsilon(^{4}\text{He})/\epsilon(^{1}\text{H})$	0.986(0.002)	0.988
	$\epsilon (^7 \text{Li})/\epsilon (^1 \text{H})$	0.975(0.003)	0.976
[14]	$\epsilon(^{4}\text{He})/\epsilon(^{1}\text{H})$	0.986(0.006)	0.992

this comparison. The ¹⁶O data have not been included in this table because of the unknown influence of plasma recombination. We have corrected the original data for the effects of window loss and nonionizing processes in the following way:

- (1) Kemper and Fox [3] give $C(^{1}\text{H})/C(^{4}\text{He}) = 0.994 \pm 0.001$ with neither window nor nuclear loss corrections. If we assume a 50 μ g cm⁻² effective Au window, we obtain $C(^{1}\text{H})/C(^{4}\text{He}) = 0.991 \pm 0.0025$, where we have increased their error by adding one-half the correction to their error.
- (2) Martini et al. [4] obtained $C(^{1}\text{H})/C(^{4}\text{He}) = 0.9940 \pm 0.0015$, where their uncertainty just spans the window correction. Since the window/dead layer correction cannot be easily estimated for their detector, we take their value at its minimum value and double the probable error, yielding $C(^{1}\text{H})/C(^{4}\text{He}) = 0.9925 \pm 0.0030$.
- (3) We have used our calculations of ΔE_n to reanalyze the Ratkowski [14] data, and obtain $C(^1H)//C(^4He) = 0.986 \pm 0.006$.

We find excellent agreement between measured values and those predicted by the model proposed here. The observation that $\epsilon(^4\text{He})/\epsilon(^1\text{H})$ appears to depend on the energy region investigated (i.e. closer to unity for higher energy measurements) is resolved by this new description.

We have searched for further tests of our model using the ⁴He spectra from our mixed source. If we use the ion beam data for ⁴He on the three detector configurations, we can extrapolate the linear fits to the data to obtain the energy of the standard, i.e. ²⁴¹Am α energy. Table 5 shows the results, where the true ²⁴¹Am energy has been corrected for $\Delta E_{\rm w}$, $\Delta E_{\rm n}$ and source thickness. In all three cases, the energy so determined is too low, by 15 ± 12 , 31 ± 11 and 11 ± 10 keV. On average, $E_{\rm K}'$ is too small by 19 ± 7 keV. If we extrapolate, assuming a nonlinear response for the Si detectors and k=0.0002, we remove the discrepancy and obtain consistency between experiment and prediction, 5 ± 7 keV.

Finally, to further test the nonlinearity prediction, we have simulated our measurement of the energy difference between the principal 4 He groups from 239 Pu and 244 Cm. The energies are 5156.6 (\pm 0.41) and 5804.82(\pm 0.05) keV, respectively [7]. If we use the 241 Am peak to calibrate the system and assume a linear response, we will obtain a value for the energy difference that is 2.7 keV too small, using again k=0.0002. The Pu-Cm energy spread is largely insensitive to the values used for ΔE_n and ΔE_w since only differences are relevant. In fact, we measure an energy spread that is too small by 2.5 \pm 0.9 (N=16), 1.6 \pm 1.0 (N=15) and 2.4 \pm 0.8 (N=19) keV for the three configurations, or 2.2 \pm 0.5 keV averaged over the measurements (N=10 no. of individual measurements). This value is in excellent

Table 5 Results for ⁴He energy from α -decay of ²⁴¹Am. Col. 2 gives the values expected after source thickness, $\Delta E_{\rm w}$ and $\Delta E_{\rm n}$ corrections. Col. 3 is the value deduced from an extrapolation of the time-of-flight data. $\Delta = E_{\rm K} - E_{\rm K}'$. Uncertainties are given in

Detector	E _K (keV)	$E'_{\mathbf{K}}(\text{keV})$ $\Delta(\text{keV})$	
23-132A (Au)	5460 (4)	5445 (11)	15 (12)
22-704E (Au)	5459 (4)	5428 (10)	31 (11)
22-704E (Al)	5445 (4)	5436 (9)	11 (10)

a) $\langle \Delta \rangle = 19 \pm 7 \text{ keV}$

parentheses

agreement with the predicted discrepancy of 2.7 keV, thus providing a stringent test of our model.

In summary, we have measured the mean pulse height produced by 1 H, 4 He, 7 Li and 16 O ions in Si detectors at low energy. We find that the pulse height increases with increasing Z. We offer an explanation for this effect in terms of a nonconstant value for ϵ , the energy required to create an electron-hole pair. A simple model with ϵ being a weak function of the projectile stopping power is found to describe our data and all previous measurements. An important implication of this model is that the pulse height for a given ion is not a linear function of the deposited energy (after correcting for window and nonionizing losses). This conclusion is substantiated by experiments using α -decaying radionuclides.

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