Electronic Stopping Cross Sections for ^{1}H and ^{4}He Particles in Cr, Mn, Co, Ni, and Cu at Energies near 100 keV

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(Received 22 May 1969)

Electronic stopping cross sections have been measured for 30- to 140-keV ^{1}H and 50- to 120-keV ^{4}He particles in Cr, Mn, Fe, Co, Ni, and Cu ($24 \leq Z_2 \leq 29$). In this Z_2 region the cross sections at a given energy show a steady *decrease* with increasing Z_2, thus confirming and extending earlier observations by others. The combined data show a dependence on Z_2 which is markedly similar to the periodic dependence of electronic stopping cross sections on the Z of the projectile. The measurements were made utilizing high-resolution solid-state-detector spectra of the particles backscattered from thin films of the target materials evaporated on thick substrates. The probable error of the stopping-cross-section measurements is 3%.

INTRODUCTION

THE penetration in solids of heavy charged particles of a few hundred keV energy or less has recently become a subject of renewed interest. The possibility of producing solid-state electronic devices by ion implantation has added a new impetus to both experimental and theoretical studies of penetration phenomena. References 1–3 provide extensive bibliographies pertinent to this and to related fields.

Experimental data concerning energy loss[4,5] are still relatively sparse because of the difficulties of preparing and weighing the thin uniform stopping layers, and of measuring the energy losses which, though small, are appreciable fractions of incident beam energies. However, in recent years there have been general improvements in thin-film technology, and in detection methods for low-energy particles, so that the region is now more accessible to measurement.

Stopping theory has also improved in the last 10 years and an extensive literature[6–12] pertinent to the low-energy region is now available. That the theory is not yet a reliable quantitative guide is perhaps best borne out by the recent, and as yet incompletely understood, discovery of the oscillation of the electronic stopping cross section S_ϵ with the atomic number of the projectile (Z_1 oscillation[13–22]). The subject matter of the present investigation bears on a closely related phenomenon, namely, the Z_2 oscillation[23–26] of S_ϵ. In this case S_ϵ for ^{1}H or ^{4}He particles of a given energy, *decreases* as Z_2 is varied from 24 to 29, in contrast with the increase predicted by theories which do not include the effect of the periodic variations in radial electron density.

METHOD

The basic details of the method have been described in previous publications.[27,28] The method utilizes spectral details of particles reflected from thin films deposited on thick substrates and thus avoids the use of thin self-supporting films. The targets were prepared by thermal evaporation, onto glass substrates, of a sandwich structure[29] comprised of the stopping material between two thin gold marker layers. One such target is shown in Fig. 1 together with the spectrum of backscattered ^{4}He particles as obtained by a low-noise silicon surface-barrier detector system. The thin gold layers do not contribute appreciably to energy loss,

[1] J. F. Gibbons, Proc. IEEE **56**, 295 (1968).
[2] S. Datz, C. Erginsoy, G. Leibfried, and H. O. Lutz, Ann. Rev. Nucl. Sci. **17**, 129 (1967).
[3] W. M. Gibson, in Proceedings of the Conference on the Use of Small Accelerators for Teaching and Research, 1968, Report No. CONF 680411 USAEC, p. 306 (unpublished).
[4] W. Whaling, in *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1958), Vol. 34, p. 193.
[5] H. Bichsel, in *AIP Handbook*, edited by D. E. Gray (McGraw-Hill Book Co., New York, 1963), pp. 8–20.
[6] O. B. Firsov, Zh. Eksperim. i Teor. Fiz. **36**, 1517 (1959) [English transl.: Soviet Phys.—JETP **9**, 1076 (1959)].
[7] J. Lindhard and V. Nielsen, Phys. Letters **2**, 209 (1962).
[8] J. Lindhard, M. Scharff, and H. E. Schiøtt, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. **33**, 14 (1963).
[9] H. E. Schiøtt, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. **35**, 9 (1966).
[10] E. Bonderup, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. **35**, 17 (1967).
[11] B. Fastrup, P. Hvelplund, and C. A. Sautter, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. **35**, 10 (1966).
[12] J. Lindhard, V. Nielsen, and M. Scharff, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. **36**, 10 (1968).
[13] Ya. A. Teplova, V. S. Nikolaev, I. S. Dimitriev, and N. L. Fateeva, Zh. Eksperim. i Teor. Fiz. **42**, 44 (1962) [English transl.: Soviet Phys.—JETP **15**, 31 (1962)].
[14] J. H. Ormrod and H. E. Duckworth, Can. J. Phys. **41**, 1424 (1963).
[15] J. H. Ormrod, J. R. Macdonald, and H. E. Duckworth, Can. J. Phys. **43**, 275 (1965).
[16] F. H. Eisen, Can. J. Phys. **46**, 561 (1968).
[17] P. Hvelplund and B. Fastrup, Phys. Rev. **165**, 408 (1967).
[18] L. Erickson, J. A. Davies, and P. Jespersgaard, Phys. Rev. **161**, 219 (1967).
[19] A. H. El-Hoshy and J. F. Gibbons, Phys. Rev. **173**, 454 (1968).
[20] D. E. Harrison, Appl. Phys. Letters **13**, 277 (1968).
[21] K. B. Winterbon, Can. J. Phys. **46**, 2429 (1968).
[22] R. S. Nelson, Phys. Letters **28A**, 676 (1969).
[23] D. W. Green, J. W. Cooper, and J. C. Harris, Phys. Rev. **98**, 466 (1955).
[24] M. Bader, R. E. Pixley, F. S. Mozer, and W. Whaling, Phys. Rev. **103**, 32 (1956).
[25] J. L. Whitton, Can. J. Phys. **46**, 581 (1968).
[26] H. H. Andersen, H. Sørensen, and P. Vadja, Phys. Rev. **180**, 373 (1969).
[27] W. White and R. M. Mueller, J. Appl. Phys. **38**, 3660 (1967).
[28] R. M. Mueller and W. White, Rev. Sci. Instr. **39**, 291 (1968).
[29] The above method of target fabrication is a variation of previous sandwich methods designed to avoid the use of thin self-supporting films; cf., Refs. 24 and 36.

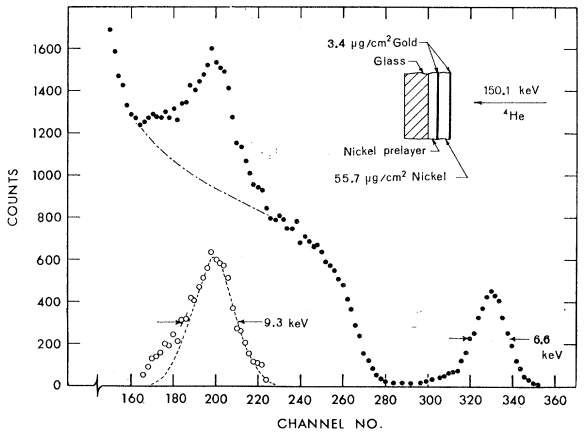

FIG. 1. Silicon surface-barrier-detector spectrum of ^{4}He particles backscattered from a sandwich-structure target. The asymmetric tail of the low-energy peak (arising from violent nuclear collisions) does not affect the most probable energy loss as measured by the peak-to-peak difference.

but, because of the large Rutherford scattering cross section of gold relative to the stopping material, they produce pronounced peaks in the spectrum. The energy difference of the peaks is then closely proportional to the energy loss ΔE in the stopping layer. For normal incidence the observed stopping cross section S_0 and the average energy $\bar{E}$ of the particle in the stopping layer are closely given by

$$S_0=(1/N)(\Delta E/\Delta m)$$
$$=[k(n_1-n_2)-\tau]/Nm[1-(1-\rho)^{-1}\sec\theta], \quad (1)$$

$$\bar{E}=E_i(1-\tfrac{1}{2}\rho)-\tfrac{1}{2}k(n_1-n_2)(1-\rho)-\tfrac{1}{4}\tau, \quad (2)$$

where N is the number of atoms per gram of the stopping material, m is the areal density of the stopping layer, Δm is the effective areal density traversed by the particle, i.e., including the path length after reflection from the second gold layer, $\Delta m=m(1-\sec\theta)$, where θ is the (large) scattering angle, and n_1-n_2 is the peak-to-peak energy difference, in channels, of the gold peaks. The calibration constant k (eV/channel) is taken from a plot of channel number of the higher-energy gold peak versus the incident-particle energy E_i. A slight variation of k with energy might be expected because of the change of stopping cross section of the gold (and other dead layers) on the surface of the detector. Such a variation is within the errors of the calibration and was not observed. The constants ρ and τ are required to correct for reflection and transmission energy losses, respectively, of the gold layers. At $\theta=165.5°$, the scattering angle used throughout most of this work, the fractional energy loss ρ upon reflection from a gold nucleus is 0.0199 for ^{1}H and 0.0784 for ^{4}He. The quantity $\frac{1}{2}\tau$ is the energy loss experienced by an ion in a one-way passage through one of the two (equally thick) gold layers. Thicknesses of 1–7 μg/cm^2, but typically 3–4 μg/cm^2, were used with equivalent results. For gold layers of 2 μg/cm^2 each, the energy loss τ was experimentally determined to be 0.4 keV for ^{1}H and 0.6 keV for ^{4}He.

The use of a high-resolution solid-state detector system permits rapid and precise energy measurement, and extension of the measurements to energies as low as 25 keV in this study. Commercial preamplifier-amplifier systems with zero capacitance noise widths of 1–2 keV full width at half-maximum (FWHM) are now common, and in all, three such systems were used in conjunction with a 7-mm^2 silicon surface-barrier detector cooled to about −120°C. Total resolution of the accelerator-detector system as measured by the width of the upper gold peak (1–3 μg/cm^2) was in the range 2.4–3.0 keV FWHM for incident protons energies under 150 keV. The systems were not optimized for resolution and better performance can be achieved[30] thus permitting extension of the method to somewhat lower energies and stopping materials of higher Z.

The detector system was calibrated by means of a plot of machine energy setting versus channel number of the higher-energy gold peak (using a multichannel analyzer), and exhibited an integral linearity of better than 1% in the range 20–150 keV for both ^{1}H and ^{4}He. The energy loss ΔE thus relates directly to the machine energy determination, i.e., a measurement of the terminal voltage of the Cockcroft-Walton accelerator. For this measurement, a precision high-voltage divider was used, with the division ratio known to about 0.1% accuracy. This unit, patterned and tested according to a design by Park,[31] has been in use for a period of approximately three years. Immediately upon completion of the present work the unit was again tested

TABLE I. Experimental details.

Material[a]	Evaporation source	No. of runs	Range of target thicknesses m (μg/cm^2)	Experimental spread σ% ($\bar{E}$=100 keV) ^{1}H	^{4}He[b]
Cr	Electroplated W filament, W basket	8	20.0– 92.5	±2.5	±2.5
Mn	W basket, Al_2O_3 crucible in shielded heater	4	40.9– 90.7	±2.2	±1.8
Fe	BeO crucible in shielded heater	6	34.8–108.0	±1.6	±2.8
Co	BeO crucible in shielded heater	5	45.6– 85.4	±4.0	±2.5
Ni	BeO crucible in shielded heater	4	43.9–116.1	±3.4	±3.2
Cu	Ta dimple, Al_2O_3 crucible in shielded heater	8	44.2–223.3	±3.0	±1.8

[a] Purity >99.9%.
[b] In general, ^{4}He results were calculated from a smaller amount of data, since ΔE was too large for measurement in the case of very thick samples.

[30] J. A. Ray and C. F. Barnett, IEEE Trans. Nucl. Sci. **NS-16**, 82 (1968).
[31] J. H. Park, J. Res. Natl. Bur. Std. (U. S.) **66C**, 1 (1962).

by an independent method.[32] The division ratio matched that of the original determination to within 0.5%. Only negligible error is introduced by assuming particle energy at the target to be that determined from terminal voltage measurements.

Two simultaneous measurements of the areal densities of the stopping layers were made by means of a Cahn model RG electrobalance and a Westinghouse model 701A quartz-crystal microbalance with the pan and crystal head, respectively, located a few centimeters on either side of the glass target substrate, and approximately 48 cm from the thermal source. The use of balances in positions adjacent to the substrate used for the S_0 measurement leaves the method open to inaccuracies due to vapor density variations. However such variations were within the assigned errors as was shown by comparing (via scattering) the relative thicknesses of thick deposits of Cr, Mn, Fe, Ni, and Cu on the electrobalance pan, the target substrate, and small glass plates so positioned as to check over-all geometry variations. Geometry corrections of the order of 1% were determined for the balances and a probable error of 2.0% assigned to include both geometry factor error and the vapor density variation. A precision of ± 0.6 $\mu g/cm^2$ pe was obtained with the electrobalance, and since this was appreciably lower than that of the quartz-crystal balance (± 0.9 $\mu g/cm^2$), the electrobalance values were used in the determination of S_0. The calibrations of the quartz-crystal microbalance and the electrobalance agreed within 2% of the theoretical value for the quartz-crystal balance, so that in effect two independent determinations of m were obtained. Further details of the weighing method are given elsewhere.[28,33]

TABLE II. Electronic stopping cross sections for ^1H and ^4He in Cr, Mn, Fe, Co, Ni, and Cu.

	S_ϵ 10^{-15} eV cm^2/atom					
E (keV)	Cr	Mn	Fe	Co	Ni	Cu
			^1H			
25	18.5			14.6		
30	20.0	17.4	17.2	15.8	15.4	14.9
35	21.5	18.8	18.7	17.1	16.9	15.7
40	22.6	19.9	19.6	18.1	18.1	16.5
50	24.6	21.8	21.5	19.9	19.9	17.9
60	26.1	23.1	22.8	21.4	20.9	19.2
80	27.9	24.9	24.2	23.3	22.2	21.0
100	28.6	25.8	24.7	24.0	22.9	21.7
120	28.8	26.1	25.0	24.3	23.1	22.1
140	28.8	26.1	25.0	24.3	23.3	22.1
			^4He			
40	31.5				24.3	
50	37.5	31.5	30.3	28.7	27.2	
60	41.9	35.2	33.7	31.9	29.9	27.4
80	49.0	41.1	39.8	37.8	34.9	31.7
100	54.3	46.1	45.0	42.4	39.3	35.3
120	58.9	50.1	49.5	46.3	43.5	38.6

[32] R. N. Lewis, Rev. Sci. Instr. 36, 532 (1965).
[33] R. M. Mueller and W. White, Rev. Sci. Instr. (to be published).

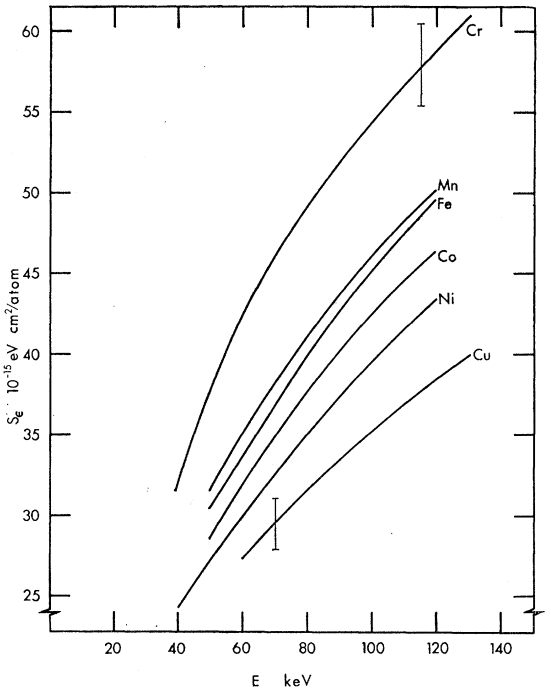

FIG. 2. Electronic stopping cross sections for ^4He in Cr, Mn, Fe, Co, Ni, and Cu. The data have been smoothed and averaged. Error bars represent standard deviations.

The vacuum evaporator was connected directly to the scattering chamber of the Cockcroft-Walton accelerator so that the target could be transferred under conditions of high vacuum. With typical beam currents of about 10 nA and vacuums of 10^{-6} Torr or better, repeated bombardments (as much as 25 μC) on a given 1-mm-diam spot, gave reproducible results over periods of many hours, and produced no visible marking. Thus contamination was considered to be negligible. However, general practice was to change the target bombardment position for each data point. The vacuum pumping system (ion pumps with titanium sublimator) permitted deposition rates of 0.1–5 ($\mu g/cm^2$)/sec (but typically ~ 0.8 ($\mu g/cm^2$)/sec) at vacuums in the low 10^{-7} Torr range. On occasion, severe outgassing resulted in pressures as high as 10^{-5} Torr. However, within the errors of this study, no effects attributable to pressure or deposition rate were noted.

The existence of possible effects due to channeling in long-range structure was checked by transferring targets of both copper and chromium to a goniometer and observing the spectrum of scattered protons when the target was rotated in increments of about 1° about each of two mutually perpendicular directions at right angles to the beam. To an accuracy of about 1% in ΔE no effect was observed. This observation does not rule out possible channeling in microcrystals of appropriate alignment. However, we would expect only negligible error from this effect.

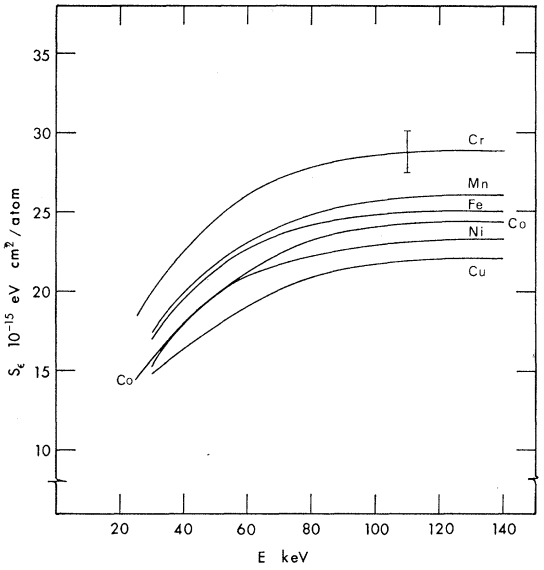

FIG. 3. Electronic stopping cross sections for ^{1}H in Cr, Mn, Fe, Co, Ni, and Cu. The data have been smoothed and averaged. Error bars represent standard deviations.

In Fig. 1, the low-energy peak shows an asymmetry attributable to violent nuclear collisions. Since ΔE is determined by a peak-to-peak measurement, i.e., as a most probable energy loss, the low-energy "tailing" does not affect the determination of S_0. "Soft" nuclear collisions, on the other hand, may be expected to add to the Gaussian spread of the peaks and to contribute to ΔE. For the conditions of this study the effect is entirely negligible for protons. For ^{4}He, a small correction, less than 0.8% in the worst case, was computed in the manner of Fastrup *et al.*[11] and applied to the data. Such calculations assume ΔE small with respect to the incident energy, a condition usually violated in our case. However, because of the small nuclear-stopping contributions, the assumption gives rise to only negligible error in this work.

In general, because the stopping cross section is energy-dependent, the average value given by Eq. (1) does not apply strictly at the average energy given by Eq. (2). Accordingly, the results include corrections [to Eq. (1)] based on the method of Chilton *et al.*[34] These corrections, largest for the ^{4}He data, in no case exceeded 3%.

In a number of cases the outer gold layer was not deposited on the stopping medium (a separate deposition being used for calibration instead) in order to check for possible effects due to passage of the ions through the gold. Within experimental error no effects were noted. In general practice, the use of two gold layers on the measurement substrate is preferrable,

TABLE III. Probable-error assessment expressed as the percent error contributed to the electronic stopping cross section. Errors quoted are representative of films in the 60–80 μg/cm^2 range and tend to decrease slightly with increasing mass.

Energy measurements	
Energy difference of the peaks	±1.6%
Calibration of detector	±0.7%
Energy loss in gold	±0.3%
Nuclear stopping (^{4}He only)	±0.5%
Areal density measurements	
Weight of deposit	±1.0%
Area of deposit	±0.1%
Geometry (including vapor density variation)	±2.0%

TABLE IV. Comparison with other measurements.

Cross section	A This method pe%	B Comparison work Ref.	B Comparison work pe%	Range of intercomparison (keV)	Difference[a] $[S_\epsilon(B)-S_\epsilon(A)/S_\epsilon(A)]\times 100\%$
		15	±4.6	20– 50	+ 1 to −1
^{1}H → Al	±4[b]	36	±5[c]	50–140	−11 to −6
		d	···	≈20	+ 6
		35	···	≈30	− 5
^{4}He → Al	±4[b]	15	±5	30– 60	− 2 to −5
^{1}H → Ni	±3	e	±5[c]	30– 90	+ 7
^{4}He → Ni	±3	e	±5[c]	40– 90	+ 2
		24	±3	50–140	+20 to +9
^{1}H → Cu	±3	36	±5[c]	50–140	+ 9 to +7
		35	···	≈30	+45

[a] Where the difference varies continuously over the range the first value refers to the lowest energy.
[b] Ref. 27.
[c] Error stated only as a limit in these cases.
[d] J. R. Young, J. Appl. Phys. **27**, 1 (1956).
[e] G. F. Bogdanov, V. P. Kabaev, F. V. Lebedev, and G. M. Novikov, Atominaia Energya **22**, 126 (1967).

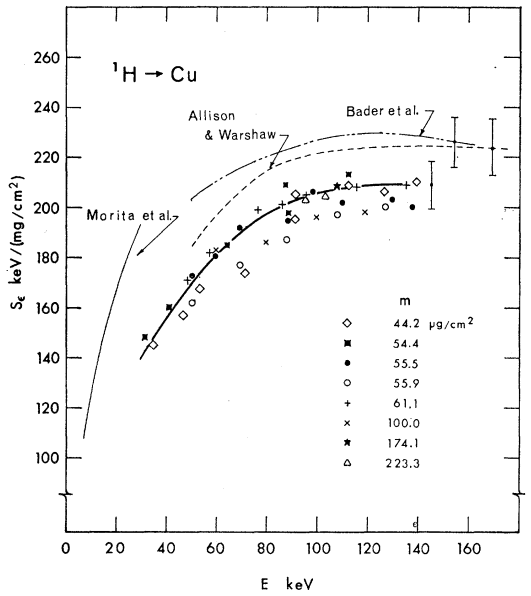

FIG. 4. Comparative data for the electronic stopping cross sections of ^{1}H in Cu. The curve has been fitted to the experimental points shown by weighting the data according to the errors assigned for individual runs. Error bars represent standard deviations except for the Allison and Warshaw case, where the error was stated as ±5%.

[34] A. B. Chilton, J. N. Cooper, and J. C. Harris, Phys. Rev. **93** 413 (1954).

since ΔE is then not dependent on small electronic drifts, a matter which can give rise to appreciable error for small ΔE.

Further details pertinent to the method are given in Table I together with the measured spread of results expressed as the fractional standard deviation σ in S_0 for $\bar{E}=100$ keV. The spread at other energies is roughly the same but becomes slightly larger at the lowest energies.

RESULTS AND ACCURACY

The smoothed and averaged electronic stopping cross sections are given in Table II and Figs. 2 and 3. An absolute probable error of $\pm 3\%$ has been assigned, based on the estimated individual error contributions given in Table III. The relative accuracy of the results may be expected to be somewhat better than the absolute accuracy and can be assessed from the individual experimental spreads listed in Table I.

The results obtained by the above method are, for the most part, in good agreement with the few measurements available for comparison (see Table IV). The results of Morita *et al.*,[35] while in agreement for aluminum, appear high ($\approx 45\%$) in the case of copper and also as much as 25–75% high with respect to other investigations[4,24,36,37] for beryllium and gold. Our complete data for copper are shown in Fig. 4 in comparison with the results of other groups. The discrepancy between our work and that of Bader *et al.*,[24] though not large except at the lower limit of the comparison, appears significant and is difficult to understand in view of the care and techniques employed in both works.

DISCUSSION

The observed decrease of S_ϵ with Z_2 confirms and extends earlier investigations,[23–26] of a Z_2 oscillation. Current theories,[6–12] predict the general trend with Z_2 but do not account for the superimposed oscillations shown in Fig. 5. While the data are fragmentary, the Z_2 oscillation bears a marked similarity with the Z_1 oscillation[17] in that the maxima and minima appear at about the same positions.

In the region $24 \leq Z_2 \leq 29$, there appears to be fine structure superimposed on the general trend of S_ϵ versus Z_2. In general, the variations are smaller than the precision which can be claimed for these results. However, we note that there is at least a suggestion of a correlation with the structure in the curve of excitation potential versus Z_2 as presented by Andersen *et al.*[26]

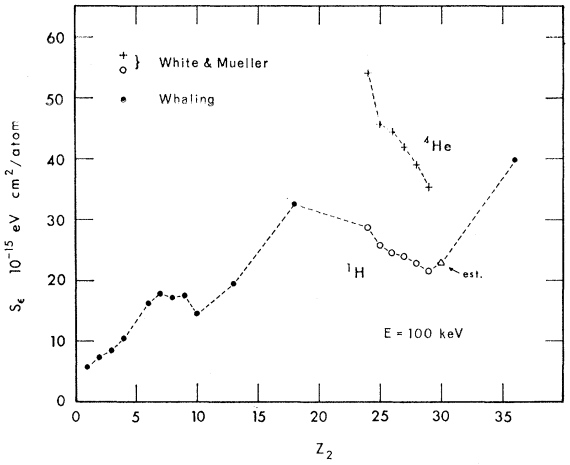

FIG. 5. Z_2 oscillation of the electronic stopping cross section at $E=100$ keV. The Whaling compilation is that of Ref. 4. S_ϵ for fluorine ($Z_2=9$) has been computed from that of Li and LiF assuming additivity of atomic stopping cross sections. For zinc ($Z_2=30$), S_ϵ has been estimated from 200 keV data (Ref. 24) and the trends of this work and Ref. 24. The data clearly indicate a minimum at $Z_2=29$.

The Z_1 oscillation has been attributed to the variation of radial charge distribution of the projectile incident on a given target.[19–21] The data indicate that the positions of the maxima and minima of the Z_1 oscillation are approximately independent of Z_2 and the physical state of the target. In view of the data of Fig. 5, we submit that the periodic dependence of S_ϵ on Z_2 is also necessary for a complete description of electronic stopping, and arises from the Z_2 radial charge distribution. While the Z_2 periodicity is likely to have only a small effect on the positions of the maxima and minima of the Z_1 oscillation, the effect on the magnitudes of the Z_1 oscillation may be expected to be more pronounced.

ACKNOWLEDGMENTS

We wish to express our appreciation for the expert technical assistance of G. O. Engelmohr and J. A. Brooks throughout the course of this work. We also wish to thank J. A. Davies for a critical reading of the manuscript.

[35] K. Morita, H. Akimune, and T. Suita, J. Phys. Soc. Japan **25**, 1525 (1968).
[36] S. K. Allison and D. Warshaw, Rev. Mod. Phys. **25**, 779 (1953).
[37] A. H. Morton, D. A. Aldcroft, and M. F. Payne, Phys. Rev. **165**, 415 (1968).