Effective charge of low-velocity ions in matter: A comparison of theoretical predictions with data derived from energy-loss measurements

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Effective-charge fractions $\zeta_{\text{expt}}=Z_1^{-1}\,(S/S_p)^{1/2}$ of low-velocity ions with atomic number Z_1 are extracted from high-precision relative measurements of stopping powers S for He, N, Ne, and Ar ions in C, Al, and Au in the ion-velocity range $0.47\leq v_1/v_0\leq 1.28$. Proton stopping powers S_p were taken from literature data. The experimental effective-charge fractions ζ_{expt} are compared with calculated values ζ_{theor} as derived in linear-response theory on the basis of a statistical model of ions of given v-dependent degree of ionization q. At low ion velocities, ζ/q is predicted to be significantly larger than 1, and to approach 1 with increasing Z_1, v_1, or r_s. The experiments confirm these trends. The observed Z_1 dependence of ζ_{expt} is somewhat more pronounced than expected by the linear-response approximation.

I. INTRODUCTION

The calculation of the energy loss of low-velocity heavy ions in solids is complicated due to the fact that swift ions cannot be treated as point charges. One way of dealing with this problem is to factorize the stopping power S as

$$S\equiv S(Z_1^*,Z_2,v_1)=[Z_1^*(v_1,Z_1)]^2 S_p(v_1,Z_2)\ . \tag{1}$$

In Eq. (1), v_1 denotes the ion velocity, Z_1 and Z_2 the atomic numbers of the ions and the target atoms, respectively, and S_p the stopping power of the solid for bare protons at the same velocity v_1. By using the ansatz equation (1), stopping-power calculations are essentially reduced to the problem of evaluating the effective charge Z_1^*e of the swift ions. Previously, Z_1^*e has been assumed to be equal to the mean electric charge Q_1e of the partially stripped ion.[1] On the basis of standard criteria,[2] Q_1e may be evaluated in the Thomas-Fermi statistical atom approximation.[3] The identification of Q_1e as the relevant effective charge for stopping is justified if distant collisions dominate the energy-loss process. In close collisions, on the other hand, electrons of the medium penetrate into the screening cloud of bound electrons of the projectile where they experience an effective ion charge larger than Q_1e.

Apart from problems associated with the determination of the effective charge, the concept of a simple relation between the stopping power and the projectile velocity v_1, as exemplified by Eq. (1), may become questionable if v_1 approaches the Fermi velocity v_F of the valence electrons in the medium. Kreussler *et al.*[4] recently provided evidence that the relative velocity $v_r=v_r(v_F,v_1)$ is an appropriate quantity for low-velocity ion stripping; v_r may be approximated by

$$v_r=\begin{cases}\frac{3}{4}v_F[1+\frac{2}{3}(v_1/v_F)^2-\frac{1}{15}(v_1/v_F)^4] & \text{for } v_1\leq v_F\ ,\\ v_1[1+\frac{1}{5}(v_F/v_1)^2] & \text{for } v_1\geq v_F\ .\end{cases} \tag{2}$$

The Fermi velocity of the valence electrons can be expressed in terms of the one-electron radius r_s in the medium,

$$v_F/v_0=1.92a_0/r_s\ , \tag{3}$$

where

$$(4\pi/3)r_s^3 n_e=1\ , \tag{4}$$

and n_e is the density of the valence electrons ($a_0=\hbar/me^2=5.29\times10^{-11}$ m and $v_0=e^2/\hbar=2.18\times10^6$ m/s).

The effective stopping-power charge Z_1^*e for a given ion charge Q_1e was derived by Brandt and Kitagawa[5] in linear-response approximation. In this approach the electronic charge density in the ions is taken to be

$$\rho_e=[(Z_1-Q_1)e/4\pi R\Lambda^2]\exp(-R/\Lambda) . \quad (5)$$

The ion radius Λ is calculated variationally, and may be written in the form

$$\Lambda(y_r)=\frac{2a_0Z_1^{-1/3}c_1[1-q(y_r)]^{2/3}}{1-c_2[1-q(y_r)]} , \quad (6)$$

where $c_1=0.240$ and $c_2=0.143$ are variational constants. The degree of ionization q is defined as

$$q(y_r)=Q_1(Z_1,y_r)/Z_1 . \quad (7)$$

q can be calculated on the basis of a velocity stripping criterion.[3,6] The data tabulated in Refs. 3 and 6 may be used to calculate Λ in Eq. (6), provided the reduced variable

$$y_1=v_1/v_0Z_1^{2/3} \quad (8a)$$

used previously[3,6] is replaced by the more appropriate variable[4,5]

$$y_r=v_r/v_0Z_1^{2/3} . \quad (8b)$$

In order to determine the effective-charge fraction ζ, defined as

$$\zeta=Z_1^*/Z_1 , \quad (9)$$

Brandt and Kitagawa[5] calculated the stopping power

$$S_{Q_1}\equiv S(Q_1,\Lambda;y_r,r_s)$$

for an ion characterized by a charge distribution according to Eqs. (5) and (6) relative to the stopping power

$$S_{Z_1}\equiv S_{Q_1=Z_1}(Z_1;y_r,r_s)$$

one would observe for the bare nucleus. By using these results, the theoretical effective-charge fraction ζ_{theor} can be written in the form of a stopping-power ratio

$$\zeta_{\text{theor}}=(S_{Q_1}/S_{Z_1})^{1/2} . \quad (10)$$

To first order, the analytical result[5] is

$$\zeta_{\text{theor}}(y_r)\simeq q(y_r)\left[1+C(r_s)\frac{1-q(y_r)}{q(y_r)}\ln\{1+[4\Lambda(y_r)/r_s]^2\}\right] . \quad (11)$$

The constant $C(r_s)$ depends only weakly on r_s, viz., $C(r_s)=0.50\pm2\%$ for $1.2\le r_s/a_0\le4$. Equation (11) predicts that $\zeta/q>1$ and that the ratio approaches 1 when $q\to1$ or $\Lambda/r_s\to0$.

Alternatively, one may define an "experimental" effective-charge fraction according to Eqs. (1) and (9):

$$\zeta_{\text{expt}}=Z_1^{-1}(S/S_p)_{\text{expt}}^{1/2} . \quad (12)$$

The purpose of this study is a comparison of ζ_{theor} and ζ_{expt} as defined in Eqs. (10) and (12). Since available experimental stopping powers for ions at velocities $v_1\simeq v_F$ scatter by up to a factor of 2, we have performed an elaborate study of the energy losses of He, N, Ne, and Ar in C, Al, and Au.[7] Particular attention was devoted to elucidating detrimental effects introduced by the crystallinity of the target foils.

In contrast to heavy ions, stopping powers for protons are known with an uncertainty of about 20% or less.[8] In the evaluation of ζ_{expt}, to be presented below, we use our own heavy-ion stopping power data in combination with literature values of S_p.

II. EXPERIMENT

Energy-loss measurements were performed using the New York University 300-kV Dynatron accelerator. In order to attain proper vacuum conditions, as well as reproducible bombardment conditions, the beam transport system was redesigned. The base pressure in the new chamber was 2×10^{-5} Pa ($\sim10^{-7}$ Torr). The improved setup included a liquid-nitrogen-cooled pressure step. Carbon build-up on the targets was never observed.

Momentum analysis of the beam before and after passage through the target foils was achieved by two 90° magnetic spectrometers. The momentum resolution of the first spectrometer, which defines

the velocity of the incident ion, was $\Delta p/p = 2.4\times10^{-4}$ (corresponding to $\Delta E/E \simeq 5\times10^{-4}$). The resolution of the second spectrometer was about $\Delta E/E \sim 10^{-3}$. The angular divergence of the incident beam was less than 0.05°. Velocity spectra after passage of the ions through the target foil were measured with an angular resolution of less than 0.1° at angles of observation ϑ between 0° and 2° with respect to the incident-beam direction. As shown below, the ability to measure velocity spectra at scattering angles $\vartheta \neq 0°$ is essential for discriminating between random and (partially) channeled loss components of a spectrum. A detailed description of the experimental setup and method of data reduction will be given elsewhere.[9]

Carbon foils with a thickness of about 15 $\mu g/cm^2$ were provided by the Brookhaven National Laboratory. Foils of aluminium and gold were prepared at the Technological Laboratory of the Physics Department, University of Munich, with stated thicknesses[10] of 24 $\mu g/cm^2 \pm 10\%$ and 150 $\mu g/cm^2 \pm 10\%$, respectively. In an independent approach to determine the foil thicknesses we measured the energy loss of 300-keV He. The nonuniformity of the foils was found to be less than 2% (probed area $\sim 5\times1$ mm^2, beam size 0.3×0.2 mm^2). Assuming Ziegler's empirical He stopping-power functions[11] to be correct at 300 keV, the thicknesses turned out to be 13.6, 26.0, and 130 $\mu g/cm^2$ for C, Al, and Au, respectively. Judging from the scatter in the compilation of the literature data,[11] the foil thickness thus determined can be assumed to be uncertain to within $\pm 15\%$ or less.

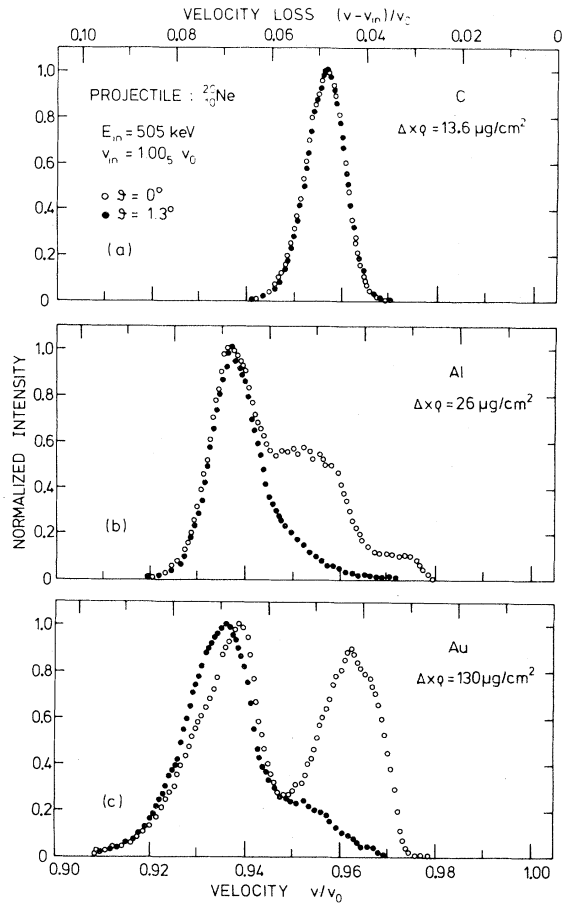

FIG. 1. Normalized velocity-loss spectra of 505-keV Ne in (a) C, (b) Al, and (c) Au. Parameter is the angle of observation ϑ, measured with respect to the direction of the incident beam.

III. EVALUATION OF THE MEAN ION-ENERGY LOSS AND STOPPING POWER

Figure 1 shows representative examples of velocity-loss spectra of 505-keV Ne in foils of C, Al, and Au at two angles of observation, $\vartheta = 0°$ and 1.3°. It is immediately evident that for Al and Au, a variation of the angle of observation results in pronounced changes of the shape of the spectra, whereas in our C foils the spectra remain essentially unaffected by ϑ variations. We attribute the low-loss (high-velocity) peaks in Figs. 1(b) and 1(c) to channeling effects. In a polycrystalline, textured foil there are apparently always crystallites with an orientation such that the beam strikes this "micro-target" within the critical acceptance angle for channeling. We have found that the effects seen in Figs. 1(b) and 1(c) cannot be removed by tilting the target with respect to the incident beam. The only practical way to circumvent these problems is to measure loss spectra at sufficiently large angles of observation. The same conclusions were reached by Mertens,[12] who observed loss spectra similar to those of Figs. 1(b) and 1(c), but less well resolved.

The energy-loss component of interest here is the low-velocity peak observed for $\vartheta = 1.3°$. This peak is taken to be the random energy loss. From the velocity spectra we find the mean energy $\bar{E}_{out}$ of ions transmitted through a foil of areal density $\Delta x\rho$. The corresponding stopping power S and the stopping cross section $\tilde{S}$ are then defined as

$$\tilde{S}(E_1) = S(E_1)/n = (A_2/N_A)(E_{in} - \bar{E}_{out})/\Delta x\rho \,, \quad (13)$$

where n (atoms/cm^3) and ρ (g/cm^3) are the number density and the density of the target, respectively. A_2 is the atomic weight of the target, and N_A is

Avogadro's number, Δx is the foil thickness, and $E_1 = 0.25(E_{\text{in}}^{1/2} + \bar{E}_{\text{out}}^{1/2})^2$, the energy corresponding to the mean ion velocity in the target foil.

The position of the random loss peak could be determined with an uncertainty of 1–2% for He and N, 2–3% for Ne, and 4–6% for Ar, the higher accuracy relating to the higher ion velocities. The total uncertainty in determining ΔE thus ranges from 3% to 8%. The uncertainty in determining E_1 is essentially given by the uncertainty in E_{in}, which is less than 2%.

In order to compare experimental stopping powers with theoretical data relating to electronic stopping, one must subtract the nuclear component from the measured total energy loss. We have calculated nuclear stopping contributions S_n^*, encountered in the experiment, on the basis of the procedure introduced by Fastrup *et al.*[13] By using the "average" potential from Wilson *et al.*[14] the fractional loss due to elastic interaction is found to vary between 0.2% for 600-keV N in C and 20% for 250-keV Ar in Au.

Figure 2 shows an example of uncorrected (+) and corrected (●) stopping cross sections for Ne in Au. The corrections exceed the experimental error only at velocities $v/v_0 < 0.6$. Also shown in Fig. 2 are stopping cross sections reported by other groups.[15–17] For all ion-target combination investigation, our results are in very good agreement with those of Ward *et al.*[17] For the purpose of this study, however, the velocity interval covered in Ref. 17 is too small (looking for Z_1 oscillations in stopping, Ward *et al.*[17] performed measurements only at $v/v_0 = 0.82$ and 1.01).

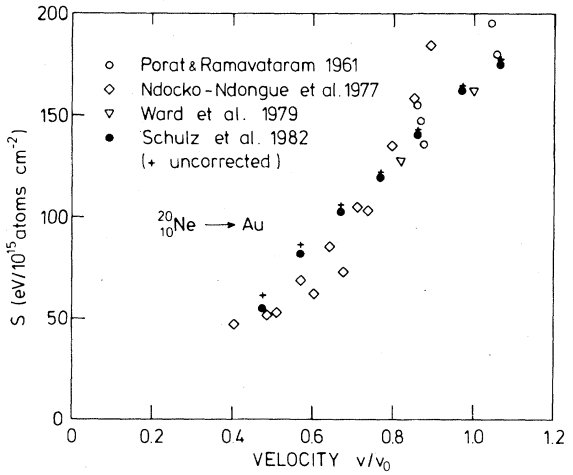

FIG. 2. Stopping cross section for Ne ions in Au vs the ion velocity.

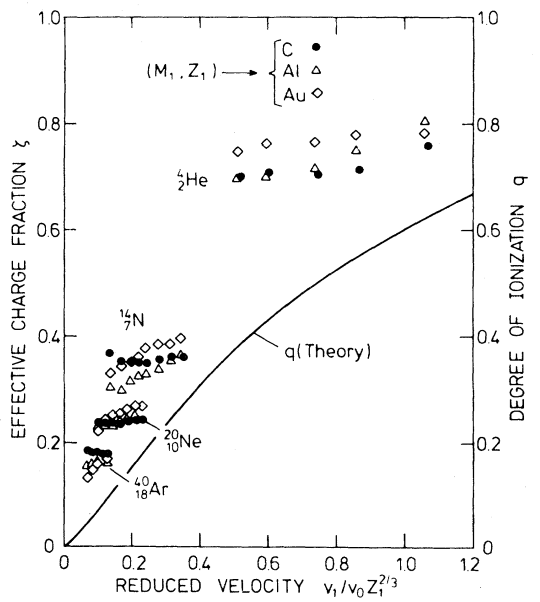

FIG. 3. Experimental effective-charge fractions ζ_{expt} of He, N, Ne, and Ar ions in C, Al, and Au, according to Eq. (12), vs the reduced ion velocity $y_1 = v_1/v_0 Z_1^{2/3}$. For comparison the degree of ionization q, taken from Ref. 3 (with $b = 1.33$), is also shown (solid curve).

A compilation of our stopping-power data is presented in Table I. The data are corrected for nuclear stopping and thus represent electronic stopping cross sections.

IV. EFFECTIVE CHARGE FRACTIONS

Using the data of Table I, we derive experimental effective-charge fractions as defined by Eq. (12). The result is presented in Fig. 3 as a function of the reduced ion velocity $v_1/v_0 Z_1^{2/3}$. Clearly the effective charges are up to a factor of 4 larger than the degree of ionization calculated by Brandt.[3] This effect is much larger than any conceivable experimental error. Inspection of proton stopping-power compilations,[8] for example, indicates that in the case of Au targets the possible error in the choice of S_p amounts to at most ±20% in the velocity range of interest here. For C and Al the accuracy appears to be much higher.[8] Taking into account another ±15% uncertainty in foil thickness determination we end up with a maximum uncertainty of S/S_p of ±35%. The corresponding uncertainty in ζ_{expt} is ±17%. Thus, the differences between ζ_{expt} and q_{theor} seen in Fig. 3 are real.

Another important finding is that the effective-charge fraction of an ion, at a fixed reduced veloci-

TABLE I. Electronic stopping cross sections $\tilde{S}$ for He, N, Ne, and Ar ions in C, Al, and Au. E_{in} is the energy of the incident ions, $E_1=0.25(E_{in}^{1/2}+E_{out}^{1/2})^2$ the "effective" projectile energy in the foil, v_1/v_0 the corresponding ion velocity in units of $v_0=e^2/\hbar=2.18\times10^6$ m/s, $v_1/v_0=0.20\,[E_1(\text{keV})/M_1(u)\,]^{1/2}$, where $M_1(u)$ is the ion mass in atomic mass units, and y_r the reduced relative velocity defined in Eq. (8b). Stopping cross sections $\tilde{S}$ are corrected for nuclear stopping (see text). E_{in} and E_1 are given in keV; $\tilde{S}$ is given in eV cm^2/10^{15} atom.

Target		Carbon				Aluminium				Gold			
$\Delta x\rho$		13.6 μg/cm^2				26 μg/cm^2				130 μg/cm^2			
r_s		$1.66a_0$				$2.12a_0$				$1.49a_0$			
Ion	E_{in}	E_1	v_1/v_0	y_r	$\tilde{S}$	E_1	v_1/v_0	y_r	$\tilde{S}$	E_1	v_1/v_0	y_r	$\tilde{S}$
$^4_2$He	75	68	0.82	0.722	20.2	66	0.81	0.638	30.9	66	0.81	0.764	43
	100	92	0.96	0.779	23.1	90	0.95	0.705	34.9	90	0.95	0.816	51
	150	141	1.19	0.889	26.2	138	1.17	0.827	41.1	137	1.17	0.917	62
	200	190	1.38	0.990	29.2	186	1.36	0.935	47.2	185	1.36	1.011	72
	300	288	1.70	1.168	34.3	284	1.68	1.123	54.6	283	1.68	1.184	85
$^{14}_7$N	100	85	0.49	0.265	42	86	0.49	0.221	45	86	0.50	0.290	63
	150	133	0.62	0.281	48	133	0.62	0.240	54	132	0.61	0.303	87
	200	181	0.72	0.296	54	179	0.71	0.258	70	179	0.71	0.316	103
	250	229	0.81	0.310	60	225	0.80	0.275	82	225	0.80	0.329	122
	300	278	0.89	0.325	64	273	0.88	0.292	90	270	0.88	0.342	144
	400	374	1.03	0.353	75	369	1.03	0.324	105	365	1.02	0.368	170
	500	471	1.16	0.380	83	464	1.15	0.353	122	462	1.15	0.393	188
	600	570	1.28	0.406	87	560	1.27	0.381	134	556	1.26	0.416	216
$^{20}_{10}$Ne	125	112	0.47	0.207	34	109	0.47	0.171	50	113	0.47	0.227	55
	180	165	0.58	0.217	40	161	0.57	0.183	62	162	0.57	0.235	82
	245	228	0.68	0.228	47	223	0.67	0.196	73	224	0.67	0.245	103
	320	301	0.78	0.240	54	294	0.77	0.211	86	295	0.77	0.256	119
	400	378	0.87	0.253	62	371	0.86	0.227	98	371	0.86	0.267	140
	505	481	0.98	0.270	68	472	0.97	0.246	112	472	0.97	0.283	162
	605	580	1.08	0.285	73					569	1.07	0.297	175
$^{40}_{18}$Ar	250	225	0.47	0.140	65	224	0.47	0.116	74	233	0.48	0.154	65
	360	332	0.58	0.147	76	329	0.57	0.124	98	337	0.58	0.159	100
	490	457	0.68	0.154	90	453	0.67	0.133	120	460	0.68	0.166	139
	640	605	0.78	0.163	97	599	0.77	0.144	136	606	0.78	0.174	163
	810	772	0.88	0.172	108	765	0.87	0.155	151	771	0.88	0.182	190

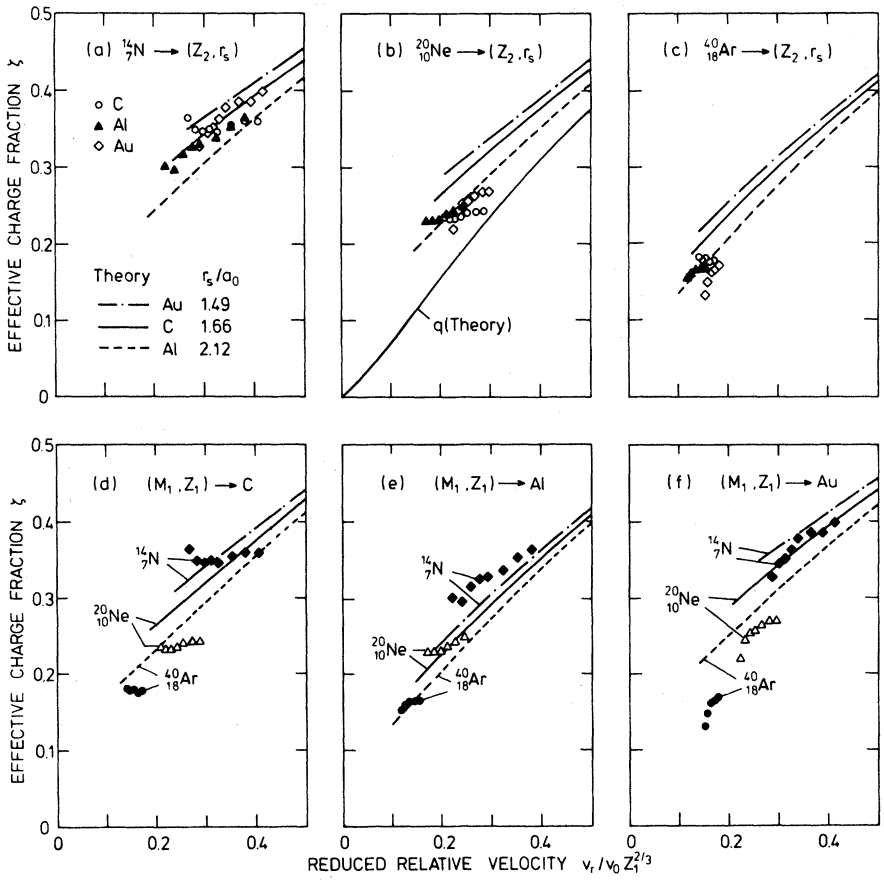

FIG. 4. Effective-charge fractions ζ of (a) N, (b) Ne, and (c) Ar ions in C, Al, and Au vs the reduced relative velocity $y_r = v_r/v_0 Z_1^{2/3}$. The theoretical curves are derived from Eqs. (11) and (6) using the r_s values given in the inset of (a). The curve $q(v_r)^5$ drawn in (b) for reference illustrates that $\zeta/q > 1$ and that it tends to 1 with increasing v_r. The same data as in (a)—(c) are reassembled for different ions in given targets in (d)—(f); ζ/q approaches 1 with increasing Z_1 more rapidly than predicted [Eq. (11)] in small-r_s materials.

ty, decreases strongly toward the q curve as the atomic number of the projectile increases (cf. Fig. 3). This effect is also evident in a compilation of literature data reported by Yarlagadda *et al.* (Fig. 2 of Ref. 6). It was attributed there to a velocity-dependent effective charge of the proton. As Eq. (11) demonstrates, the recent work of Brandt and Kitagawa[5] explains these trends without recourse to an effective proton charge less than 1. This supports the contention that protons do not carry a bound electron in a valence-electron gas.[3] According to Fig. 3 effective-charge fractions rise slowly with v_1, and, in the regime covered by the present study, they depend only weakly on the target material in the range $1.49 \leq r_s \leq 2.12$.

A comparison of experimental and theoretical effective-charge fractions is presented in Fig. 4. In order to illustrate the material or r_s dependence as well as the Z_1 effect, the data are plotted twice. Since the validity of a statistical atom model is doubtful for helium, the He data are not included in Fig. 4. The most important finding of Fig. 4 is that the ζ_{expt} values fall much closer on ζ_{theor} than on q_{theor}. The theoretical and empirical charge fractions differ from each other by at most 35% with a relative root-mean-square deviation of 16%, taking into account all experimental data points.

Detailed inspection of Fig. 4 reveals some trends not yet accounted for by theory.

(i) The influence of the target material on the effective charge is less pronounced than predicted [Figs. 4(a)—4(c)]; theory appears to overestimate the r_s dependence.

(ii) A v_r velocity dependence of ζ_{expt} is not evident in carbon [Fig. 4(d)]; theory predicts a monotonic increase in ζ_{theor} with v_r.

(iii) The Z_1 dependence of ζ_{expt} for $Z_1 \leq 18$ at small r_s is more pronounced than predicted [Figs. 4(d)–4(f)].

To ensure that the discrepancies seen in Fig. 4 are not a reflection of the choice of proton stopping powers taken from the literature, new experiments with protons on the same foil spots in the same apparatus for direct comparison with the present data are now in progress at the NYU accelerator facility. The large Z_1 dependence of ζ_{expt}/q and the small variations of ζ_{expt} in our ion velocity range suggest that screening in the theory may have to be reexamined, especially in light of new results of stopping-power calculations based on the phase-shift method.[18,19] It may also be necessary to account for the shell structure of the swift ion, which is known to cause the well-known Z_1 oscillations in electronic stopping.

V. CONCLUSION

Experimental effective-charge fractions ζ_{expt} of low-velocity heavy ions in solids, as extracted from the ratio of stopping cross sections for ions and protons of the same velocity, confirm the effect that $\zeta/q > 1$ predicted by the Brandt-Kitagawa theory. A firmer proton data base than employed here is required to improve the reliability of the empirical values. The theory may have to be refined to account in detail for the strong Z_1 dependence of the experimentally determined effective-charge fractions in materials with small r_s.

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