Resolution of the Frozen-Charge Paradox in Stopping of Channeled Heavy Ions

Peter Sigmund
Physics Department, Odense University (SDU), DK-5230 Odense M, Denmark

Andreas Schinner
Institut für Experimentalphysik, Johannes-Kepler-Universität, A-4040 Linz-Auhof, Austria
(Received 17 August 2000; revised manuscript received 27 November 2000)

The long-standing problem of the lacking signature of a Barkas effect in the stopping of swift heavy ions under channeling conditions has been analyzed theoretically. The stopping model provides explicit dependences on impact parameter and allows for projectile screening and higher-order Z_1 corrections. The analysis differentiates between principal target shells. A distinct Barkas correction is found in accordance with standard theory. It is less pronounced for channeled than for random stopping because of the dominance of outer target shells. Varying contributions from different target shells to the stopping force may give rise to an inversion of the commonly observed variation with ion energy and charge state of the Barkas correction.

DOI: 10.1103/PhysRevLett.86.1486 PACS numbers: 34.50.Bw, 29.30.Lw, 61.80.Lj, 61.85.+p

Swift heavy ions penetrating through crystals under channeling conditions can retain "frozen charge states," since the suppression of small-impact-parameter collisions may significantly reduce the frequency of electron capture and/or loss by the projectile [1]. This phenomenon has been utilized to study the electronic stopping of swift ions as a function of the ion charge $q_1 e$ in a series of by now classic experiments [2–4]. The main goal of those studies was to elucidate the Z_1^3 (or Barkas) effect, which refers to deviations from the Z_1^2 dependence of the standard stopping force on the atomic number Z_1 of the projectile [5]. Experiments were performed with bare and dressed ions of atomic number $Z_1 \leq 17$ at velocities $v/v_0 \simeq 9$–12 ($v_0 =$ Bohr velocity) penetrating along the (111) planar channel in Au [1–3] and the $\langle 110 \rangle$ axial channel in Si [4].

The Z_1^3 correction is caused by target polarization, it is positive for positively charged projectiles and decreases with increasing speed [6,7]. Such a behavior was not found in the experiments quoted. For bare ions with $5 \leq Z_1 \leq 17$, deviations from Z_1^2 scaling, when observed at all, were increasing with energy. For dressed ions, measured deviations from q_1^2 scaling were either insignificant or increasing with *decreasing* charge and hence were ascribed to incomplete screening.

The existence of the Z_1^3 effect is well established both experimentally and theoretically, in particular by experiments involving antiprotons [8]. As long as the experimental results quoted above can be taken as evidence against this effect, there is a fundamental problem that warrants resolving. While an effort in that direction was made on the basis of an electron-gas model [9], that analysis focused on bare ions and only qualitatively considered projectile screening.

Recent theoretical studies of heavy-ion stopping [10–12] based on the Bohr theory [13,14] incorporate screening and higher-order Z_1 effects. Good agreement has been obtained with experimental data on random slowing down over a wide velocity range [10,12,15,16]. Since these calculations invoke an impact parameter, they should be applicable to channeled ions as well. We note that ions and velocities involved in these experiments all lie well within the "classical" regime [17].

If $T(p)$ is the mean energy loss experienced in an individual collision event at impact parameter p, the stopping force is given by

$$-\frac{dE}{dx} = \sum_{\ell} z_{\ell} \frac{T(p_{\ell})}{d_{\ell}} \tag{1}$$

for axial channeling, where p_ℓ is the distance between the trajectory and the ℓth set of neighboring strings of atoms, z_ℓ the number of strings per set, and d_ℓ the interatomic distance in the string. For planar channeling and random trajectories, on the other hand, the stopping force may be found by appropriate integrations of $T(p)$ over the impact parameter.

For screened heavy ions the energy-loss function at large impact parameters has been found to be given by [10]

$$T(p) = \frac{2Z_1^2 Z_2 e^4}{mv^2 p^2} \sum_n f_n \zeta_n^2 \times \{[\beta K_1(\zeta_n) + (1-\beta)\alpha_n K_1(\alpha_n \zeta_n)]^2 + [\beta K_0(\zeta_n) + (1-\beta) K_0(\alpha_n \zeta_n)]^2\}, \tag{2}$$

where Z_2 is the atomic number of the target material, $\zeta_n = \omega_n p/v$, $\beta = q_1/Z_1$, $\alpha_n = \sqrt{1 + (v/a\omega_n)^2}$, $a = a_{\mathrm{TF}}(1-\beta)$, $a_{\mathrm{TF}} = 0.8853 a_0/Z_1^{1/3}$ ($a_0 =$ Bohr radius), and K_0, K_1 are modified Bessel functions in standard notation. ω_n and f_n are resonance frequencies and oscillator strengths characterizing the stopping medium ($\sum_n f_n = 1$). For a stripped ion Eq. (2) reduces to Bohr's result for distant collisions [13]. For $\beta < 1$ the relation reflects exponential screening by $Z_1 - q_1$ projectile electrons with a screening radius a.

 0031-9007/01/86(8)/1486(4)$15.00

TABLE I. Resonance frequencies and oscillator strengths for solid Si [8]. ξ^{-1} from Eq. (3) for 3.086 MeV/u Cl ions.

Shell n	$\hbar\omega_n$ (eV)	f_n	ξ^{-1}
1 (K)	3179	0.1222	1.448
2 (L)	249	0.5972	0.1134
3 (M)	20.3	0.2806	0.009 226

For small p, $T(p)$ needs to reflect the close-collision behavior. This is important for random stopping. Free-Coulomb scattering [13] is adequate as a first approximation [14]. Improved estimates have been found by the binary theory developed recently [12] which incorporates higher-order Z_1 terms nonperturbationally.

We have chosen measurements on Si [4] for our analysis because of the small number of target shells involved and because of the straight dependence of the stopping force on $T(p)$, Eq. (1), and chlorine projectiles because of the large number of charge states for which measurements were reported. However, no major qualitative differences have been pointed out between the experimental data on Si and Au and various projectile ions involved.

Table I shows resonance frequencies and oscillator strengths for the principal target shells, determined by bundling [8] tabulated optical spectra [18]. Also included is the factor

$$\xi^{-1} = Z_1 e^2 \omega / m v^3, \tag{3}$$

which is known to govern the magnitude of the Z_1^3 correction [6,7], for 3.086 MeV Cl ions in Si [4]. This factor varies rapidly from shell to shell. Channeled particles are stopped mainly by interaction with outer-shell target electrons. Clearly, the Z_1^3 effect must be less significant for channeled than for random stopping.

Figure 1 shows shellwise contributions to the calculated stopping force on fully stripped chlorine ions for random trajectories in silicon, calculated from Eq. (2) and from the binary theory, respectively. The contributions from the L and M shells cross over around 2 MeV/u, while the K shell appears insignificant in the depicted velocity range. In order to illustrate the effects of screening as clearly as possible we show the corresponding graph for neutral projectiles (Fig. 2) even though experimental data are available only for charge states down to 10. The contribution from the M shell is more heavily attenuated than that from the L shell, while that from the K shell is essentially unchanged. Clearly, screening affects distant interactions which are most pronounced for the most weakly bound target electrons. The total stopping force is reduced by less than a factor of 3 over most of the depicted velocity range; i.e., screening is far from complete. This reflects the importance of close collisions in random stopping.

The difference between full-drawn and dashed curves reflects third- and higher-order Z_1 terms. For the stripped ion, Fig. 1, the relative difference is seen to increase dramatically from the M shell to the K shell. For the neutral projectile (Fig. 2) the Z_1^3 effect appears enhanced on a relative scale. This, together with the attenuation of stopping by M electrons—where the Barkas correction was weakest—implies an enhanced Barkas effect with increasing screening. This is illustrated in Fig. 3, where the stopping number $L = (mv^2/4\pi Z_1^2 Z_2 e^4 N)(-dE/dx)$, divided by q_1^2, is plotted against the number of electrons on the ion. The curve labeled "Bohr"—which is based on Eq. (2)—illustrates the effect of incomplete screening. The curve labeled "binary" adds the Barkas correction which *enhances* the effect of incomplete screening, i.e., acts in the direction opposite to what would have been expected.

Figure 4 shows the calculated stopping force on chlorine ions well channeled along the ⟨110⟩ direction in silicon for both fully stripped and neutral ions, taking into account the interaction with the nearest-neighbor strings ($z_1 = 6$, $p_1 = 2.032$ Å) all calculated on the basis of Eq. (2). Figure 5 shows that second-neighbor rows do not influence the general picture and give rise to only a minor quantitative

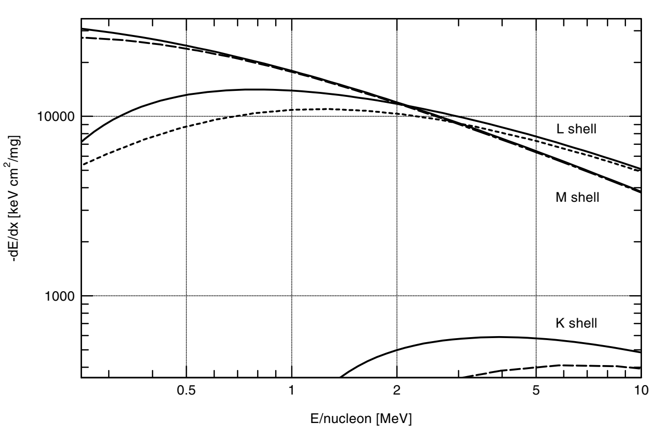

FIG. 1. Contribution of principal target shells to random-stopping force on fully stripped Cl ions in Si. Solid lines: nonperturbative [12]; dashed lines: Bohr allowing for screening [10].

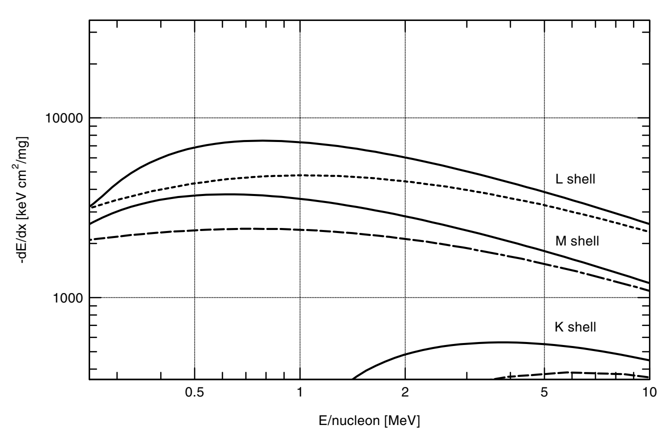

FIG. 2. Contribution of principal target shells to random-stopping force on neutral Cl ions in Si. Solid lines: nonperturbative [12]; dashed lines: Bohr allowing for screening [10].

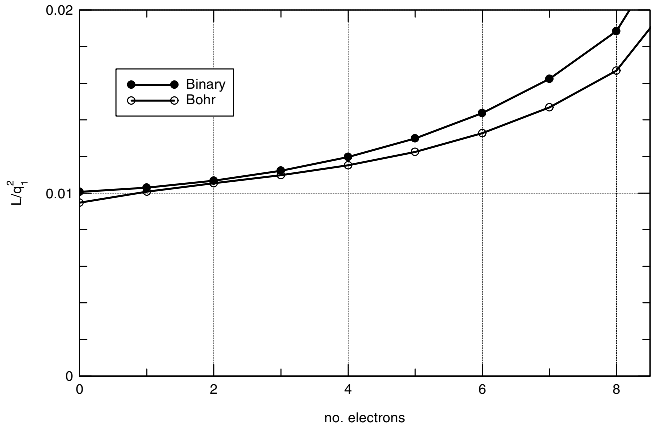

FIG. 3. Stopping number L for 3.086 MeV/u Cl^{q_1+} in Si divided by the square of the ion charge; random stopping.

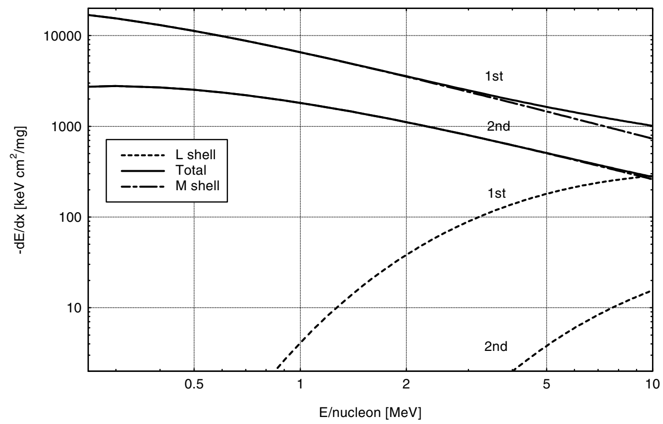

FIG. 5. Contributions from first and second neighboring rows to stopping. Parameters as in Fig. 4. Stripped ions only.

correction. For stripped ions stopping is heavily dominated by the M shell. According to Fig. 1 this must imply a very small Barkas correction. However, the contribution from the L shell increases rapidly over the depicted energy interval while that of the M shell decreases. This causes the Barkas term originating in the L shell to increase with *increasing* velocity. An effect like this is likely to cause a Barkas correction increasing with energy in gold [3].

For neutral projectiles we first notice a reduction in the total stopping force by 4 orders of magnitude. This means that screening is essentially complete. The fact that stopping due to L electrons is now competitive ought to produce a trend toward a Barkas effect increasing with decreasing ion charge and thus could explain experimental observations [3,4]. However, this trend is not dominant, as is seen in Fig. 6 which shows the calculated Barkas correction rising linearly with q_1 up to $\simeq 3\%$ for the fully stripped ion. This appears consistent with Fig. 4: In case of nearly complete screening a channeled ion with charge q_1 must be equivalent with a bare ion with the same charge.

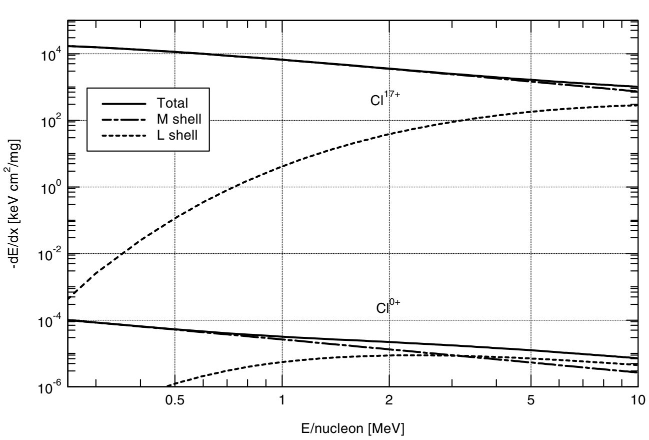

FIG. 4. Stopping of 3.086 MeV/u Cl ions in ⟨110⟩ axial channel in Si: Total stopping force and contributions from L and M shells for stripped and neutral particles; calculated from Eq. (2); first neighboring channels only; Barkas term not included.

While the Barkas effect hinges on inner electrons and is thus well characterized by the Bohr theory, a computation of the total stopping force is more problematic in view of the dominating role of valence electrons under channeling conditions. Since the Bohr theory assumes all target electrons to be located initially on lattice sites, close collisions are completely suppressed. This must underestimate stopping, in particular, due to valence electrons. For an error estimate we have considered two distinct effects:

(1) For a nonvanishing orbital radius r_n, the energy loss in the Bohr model is enhanced by an amount ΔT with $\Delta T/T \simeq 2r_n^2/3p^2$ at large impact parameters p. This causes a $\sim 4.5\%$ enhancement for $r_n = a_0$ which is more important than errors due to the neglect of thermal vibrations, Barkas effect, and the adopted excitation spectrum but has been ignored in the following.

(2) Valence electrons located near the channel center allow for close collisions. Their contribution to stopping is tentatively approximated by random stopping.

Table II shows results taking into account the second effect. Reference [9] quotes a calculated density of

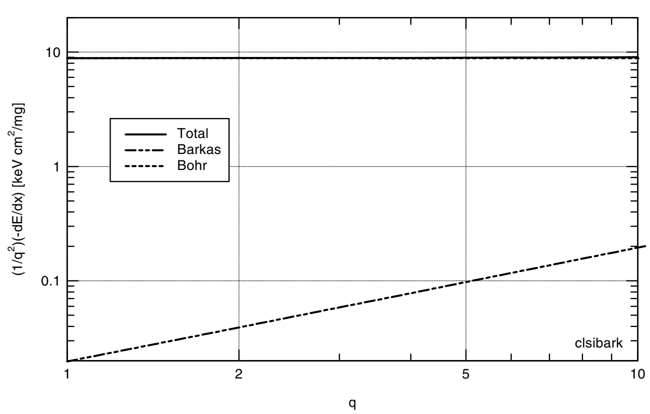

FIG. 6. Stopping force divided by q_1^2 for channeled 3.086 MeV Cl ions in Si; first-neighbor channels only.

TABLE II. Calculated contributions to stopping force (in MeV cm^2/mg) on 3.086 MeV Cl^{17+} ions in $\langle 110 \rangle$ axial channel in Si. See text.

	0% random	23% random	46% random
M shell, random	0.000	2.079	4.157
M shell, 1st neighboring string	2.337	1.797	1.257
M shell, 2nd neighboring string	0.796	0.612	0.428
L shell, 1st neighboring string	0.094	0.094	0.094
Total	3.227	4.582	5.936

$0.0495/\text{Å}^3$ electrons in the channel center, corresponding to 23% of all M electrons. The third column shows the contributions to the stopping force from individual shells taking this as a lower bound on random stopping. Doubling that contribution—which would reflect an average electron density being equal to the calculated electron density 0.7 Å away from the channel center—leads to the results quoted in the fourth column. The resulting stopping force of 5.936 MeV cm^2/mg compares well with the measured value of 6.105 MeV$/u$ Cl^{17+} [4], while a model assuming all M electrons on lattice sites (second column) would lead to an underestimate by almost a factor of 2.

It is the random component which is asserted to be responsible for the screening effect observed experimentally. The equivalent of Fig. 3 for just the M shell in Si predicts a 12.2% increase in L/q_1^2 from the stripped ion to four carried electrons and 17.8% for seven electrons. For a 23% random-stopping contribution from the M shell this leads to a 5.5% increase in the total stopping force for a Cl ion carrying four electrons and 17.8% for seven electrons. This may be compared with measured figures of 3.9% and 12%, respectively [4].

Our main conclusion is that the absence of a visible Barkas effect in stopping under channeling conditions—whether involving bare or screened ions—does not by any means contradict the presence of a pronounced effect in random stopping under otherwise identical conditions. We emphasize that under channeling conditions the Barkas effect does not only hinge on the parameter ξ, Eq. (3), but also on $\zeta_n = \omega_n p/v$ which causes further attenuation for the most widely open axial and planar channels. We find it interesting that incomplete screening and Barkas effect may act in the same direction instead of opposite, but note that this may be more important in random than in channeled stopping.

This work has been supported by the Danish Natural Science Research Council (SNF).

[1] S. Datz, F.W. Martin, C.D. Moak, B.R. Appleton, and L.B. Bridwell, Radiat. Eff. **12**, 163 (1972).

[2] C.D. Moak, S. Datz, B.R. Appleton, J.A. Biggerstaff, M.D. Brown, H.F. Krause, and T.S. Noggle, Phys. Rev. B **10**, 2681 (1974).

[3] S. Datz, J.Gomez del Campo, P.F. Dittner, P.D. Miller, and J.A. Biggerstaff, Phys. Rev. Lett. **38**, 1145 (1977).

[4] J.A. Golovchenko, A.N. Goland, J.S. Rosner, C.E. Thorn, H.E. Wegner, H. Knudsen, and C.D. Moak, Phys. Rev. B **23**, 957 (1981).

[5] H.H. Andersen, Phys. Scr. **28**, 268 (1983).

[6] J.C. Ashley, R.H. Ritchie, and W. Brandt, Phys. Rev. B **5**, 2393 (1972).

[7] J. Lindhard, Nucl. Instrum. Methods **132**, 1 (1976).

[8] S.P. Møller, E. Uggerhøj, H. Bluhme, H. Knudsen, U. Mikkelsen, K. Paludan, and E. Morenzoni, Phys. Rev. A **56**, 2930 (1997).

[9] J.A. Golovchenko, D.E. Cox, and A.N. Goland, Phys. Rev. B **26**, 2335 (1982).

[10] P. Sigmund, Phys. Rev. A **56**, 3781 (1997).

[11] A. Schinner and P. Sigmund, Nucl. Instrum. Methods Phys. Res., Sect. B **164–165**, 220 (2000).

[12] P. Sigmund and A. Schinner, Eur. Phys. J. D **12**, 425 (2000).

[13] N. Bohr, Philos. Mag. **25**, 10 (1913).

[14] P. Sigmund, Phys. Rev. A **54**, 3113 (1996).

[15] H. Paul, A. Schinner, and P. Sigmund, Nucl. Instrum. Methods Phys. Res., Sect. B **164–165**, 212 (2000).

[16] P. Sigmund and A. Schinner, Phys. Scr. (to be published).

[17] N. Bohr, Mat. Fys. Medd. Dan. Vidensk. Selsk. **18**, No. 8, 1 (1948).

[18] E.D. Palik, *Handbook of Optical Constants,* Academic Press Handbook Series Vol. 1 (Academic Press, Orlando, 1985).