Energy-Loss-Straggling Experiments with Relativistic Heavy Ions in Solids

C. Scheidenberger,[1] H. Geissel,[1] H. H. Mikkelsen,[2] F. Nickel,[1] S. Czajkowski,[1] H. Folger,[1] H. Irnich,[1] G. Münzenberg,[1] W. Schwab,[1] Th. Stöhlker,[1] T. Suzuki,[1] and B. Voss[3]
[1]*GSI Darmstadt, Planckstr. 1, D-64291 Darmstadt, Germany*
[2]*Odense Universitet, Campusvej 55, DK-5230 Odense M, Denmark*
[3]*TH Darmstadt, Schlossgartenstr. 9, D-64289 Darmstadt, Germany*
(Received 8 July 1996)

Energy-loss straggling measurements performed at the high-momentum-resolution magnetic spectrometer FRS with bare and highly charged $_{8}$O, $_{54}$Xe, $_{79}$Au, and $_{92}$U ions with specific kinetic energies (700–1000) MeV/u are reported. The results are in good agreement with rigorous calculations recently reported by Lindhard and Sørensen and reveal systematic deviations from the well-known relativistic Bohr formula, which was obtained within the framework of the first-order Born approximation. [S0031-9007(96)01598-0]

PACS numbers: 34.50.Bw, 25.75.–q

The energy-loss straggling of charged particles is of interest not only because of its fundamental aspects but also because of its impact on many applications in physics and other sciences. For instance, the energy-deposition and the related straggling put basic restrictions on the resolution of so-called ΔE detectors used for a large variety of purposes [1]. The statistical nature of energy loss and charge-changing collisions is the reason for the range straggling of charged particles. A precise knowledge of range distributions is essential to well-controlled implantation of charged particles as required, e.g., in material science, nuclear-spectroscopy experiments, and cancer therapy.

To our knowledge, no energy-loss straggling measurements with relativistic heavy ions have been reported to date. This lack of data was the motivation for the measurements performed at the GSI UNILAC/SIS heavy ion accelerator facilities in combination with the magnetic spectrometer FRS, which provides high momentum resolution. The energy-loss straggling was measured for $_{8}$O, $_{54}$Xe, $_{79}$Au, and $_{92}$U ions impinging fully stripped with specific kinetic energies ranging from 700 to 1000 MeV/u on various targets made of Be, Ti, Au, and Pb. In this energy range even the heaviest projectiles like Au and U can capture at most two electrons when penetrating through matter [2–4]. Thus theoretical predictions are greatly simplified, and in contrast to measurements at low projectile velocities, where many different charge states occur, the strongly reduced complexity allows for an unequivocal interpretation of the measured data.

Theoretical descriptions found in the literature [5] refer to cases where either a classical treatment holds, i.e., where the Sommerfeld parameter $Z_1\alpha/\beta \gg 1$, Z_1 being the projectile atomic number, α the fine-structure constant, and β the projectile velocity relative to the speed of light, or where a first-order perturbation treatment can be applied, i.e., where $Z_1\alpha/\beta \ll 1$. Thus the intermediate region $Z_1\alpha/\beta \sim 1$ requires some attention and in this Letter the regime where β approaches unity will be treated.

Only the atomic interaction of the penetrating ions with the target electrons will be considered, nuclear effects can be neglected as well as radiative processes whose cross sections are too small in the considered velocity range to affect the results. Furthermore, the case of thin absorbers, where only few collisions occur and thus the resulting energy-loss distributions have to be described by Landau-type approximations, is beyond the scope of this Letter. More specifically, the theoretical description concentrates on Gaussian energy-loss distributions, which occur in the limit of many collisions corresponding to targets which are thick enough to ensure that the energy-loss distribution can be completely described by its mean value and its variance Ω^2. According to Bohr [6] the accumulated contributions of successive collisions yield

$$\Omega^2 = NZ_2 \int_0^{\Delta x} \sum_n E_n^2 \sigma_n \, dx \,, \tag{1}$$

where NZ_2 denotes the number of target electrons per unit volume, Δx is the penetrated thickness of the absorber, and E_n and σ_n are the energy transfer to an electron of a target atom initially in the ground state and the corresponding cross section, respectively. The summation must be understood as a summation over all discrete excitation states of the target electrons and as an integration over all possible continuum states. When the slowing down of the ions does not affect the energy dependent cross sections, the integration becomes trivial and $\Omega^2 \propto \Delta x$. In the following it will be assumed that the projectile charge equals its atomic number.

As shown by Bethe [7] the cross section depends on the momentum transfer Q to the target electrons, and in analogy to the determination of the mean energy loss of a heavy charged particle the sum in Eq. (1) can be split into two parts referring to small and large momentum transfers, respectively. Fano [8] has carried out the calculation for small momentum transfers neglecting shell corrections and relativistic corrections, which both are presumed to be of minor importance. Using the first moment $S(1)$ of the oscillator-strength distribution [see Ref. [9]] we can rewrite his result

$$\Omega^2_{\text{low-}Q} = 4\pi Z_1^2 e^4 N Z_2 \Delta x \left(\frac{\text{Ry}\ S(1)}{Z_2 mc^2 \beta^2} \ln \frac{2mc^2\beta^2}{I_1} \right). \quad (2)$$

Here Ry $\simeq$ 13.6 eV is the Rydberg, mc^2 and e are electron rest energy and charge, respectively, and I_1 denotes the mean excitation energy for straggling [8]. A definition of the quantity $S(1)$ as well as simple functions for $S(1)$ and I_1 can be found for target materials with atomic number $Z_2 \leq 38$ in Ref. [9]. These functions can be applied also for target materials up to the highest atomic numbers with reasonable accuracy [10].

Bohr [11] considered the contribution of large momentum transfers to the energy-loss straggling, which is much larger than the contribution of small momentum transfers. The target electrons can be treated as being unbound, and thus the cross section for the scattering of free electrons off nuclei can be used. With the result for the relativistic first-order Born cross section the calculation yields

$$\Omega^2_{\text{high-}Q} = 4\pi Z_1^2 e^4 N Z_2 \Delta x \, \frac{1 - \beta^2/2}{1 - \beta^2}. \quad (3)$$

This equation is commonly known as the "relativistic Bohr formula." Since the applicability of the first-order Born approximation is restricted to projectile-charge and velocity ranges where $Z_1 \alpha / \beta \ll 1$ is fulfilled, Bohr's perturbation result even at relativistic velocities applies only for projectiles with $Z_1 \lesssim 10$, whereas for higher projectile atomic numbers a higher-order Born result [12–14] or a treatment using the exact Mott cross section [15,16] is required. A rigorous treatment of the close-collision contribution of relativistic heavy-ion slowing-down, which applies for all ions up to the highest kinetic energies, has been derived recently by Lindhard and Sørensen [17]. For moderate relativistic energies, where the scattered bare nuclei can be regarded as point charges, their close-collision result for the energy-loss straggling can be retrieved with the exact Mott cross section $d\sigma_{\text{Mott}}$ [15,16]

$$\Omega^2_{\text{high-}Q} = 2\pi N Z_2 \Delta x \int_{\Theta_0}^{\pi} E^2(\Theta) \frac{d\sigma_{\text{Mott}}(\Theta)}{d\Theta} \sin(\Theta)\, d\Theta. \quad (4)$$

Here $E(\Theta)$ is the energy transfer to a target electron in a collision leading to a center-of-mass scattering angle Θ, and Θ_0 corresponds to the smallest energy transfer for which the assumption of scattering off free electrons is valid. In practice the integral has to be computed numerically. For small values of Θ_0 there is only a very weak dependence on the choice of Θ_0 and to a very good approximation one can set $\Theta_0 = 0$. The results for different projectile atomic numbers are shown in Fig. 1 in comparison with the relativistic Bohr result. The Z_1^2 dependence found by Bohr, which is characteristic for a first-order-perturbation treatment, is no longer valid when the exact phase shifts are used.

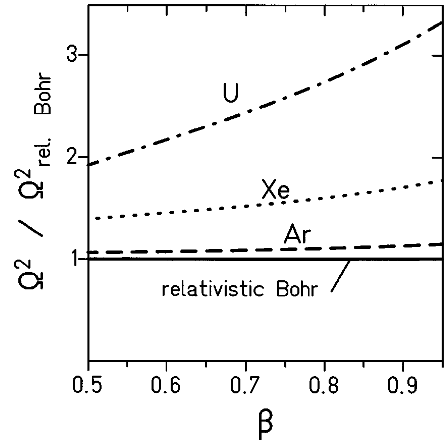

FIG. 1. Close-collision energy-loss straggling of heavy charged particles according to Eq. (4) relative to the relativistic Bohr straggling, Eq. (3), depending on the projectile velocity for different projectiles: $_{18}$Ar (dashed line), $_{54}$Xe (dotted line), $_{92}$U (dash-dotted line). The results which take into account Mott scattering vary drastically with increasing Z_1, and an increase of up to a factor of 3 is expected.

An additional contribution to the collisional straggling arises from charge-changing events of the projectile inside the target. Sigmund [18] has derived a general analytical expression for the charge-exchange straggling. For the case of three contributing charge states, which is appropriate for the present considerations, the equilibrium-charge-exchange straggling reads

$$\Omega^2_{\text{ch.ex.}} = \frac{\Delta x}{N\beta^*} \cdot \sum_{i=0}^{2} \sum_{j=0}^{2} \left[\left(\frac{dE}{dx} \right)_i - \left(\frac{dE}{dx} \right)_j \right]^2 \times F_i^{(\infty)} (\alpha^* F_j^{(\infty)} - \sigma_{ij}), \quad (5)$$

where α^* and β^* (see Ref. [18]) depend on the charge-exchange cross sections σ_{ij}, i and j denoting the number of electrons attached to the projectile before and after a charge-changing collision, respectively, and $F^{(\infty)}$ and dE/dx are equilibrium fraction and stopping power of ions in a certain charge state, respectively. Since these quantities are known from our measurements [19–21], the contribution of the charge-exchange straggling can be calculated reliably. Agreement was found between the results of Eq. (5) and the results of a Monte Carlo calculation; in particular, to a very good approximation the impinging bare ions can be treated as if they would enter the target already in charge-state equilibrium, since the targets used in the experiments were much thicker than the required equilibrium thicknesses.

The experiments were performed at the heavy-ion synchrotron SIS accelerator facilities [22] and the fragment separator FRS [23] of GSI. For the present experiments $_8$O^{8+} was accelerated to a specific kinetic energy of 702 MeV/u, $_{54}$Xe^{48+} to 800 MeV/u, $_{79}$Au^{64+} to

1000 MeV/u, and $_{92}U^{73+}$ to 950 MeV/u, respectively. The ion beams were extracted from the SIS and transported to the magnetic spectrometer FRS. It consists of two stages which are arranged mirror-symmetrically with respect to the middle focal plane. At the entrance of the FRS the Xe, Au, and U ions passed through a stripper causing an energy loss of less than 1.2%. Only those ions which emerged from the stripper completely stripped were transported to the middle focal plane, where the targets for the energy-loss straggling measurements were placed. In the energy-loss mode of the FRS [23] the momentum-loss distribution of the ions is analyzed with the second stage, which is characterized by a dispersion of 8.4 cm/%. The measured momentum distribution of the ions is to first order independent of possible momentum fluctuations of the beam in front of the target. That is why the energy-loss mode is well suited for the present investigations.

The energy-loss straggling in the target increases the momentum width of the ion beam. This leads because of the spectrometer dispersion to the broadening of the position spectra measured with two multiwire proportional counters at the final focus of the FRS. Since in all measurements the fully stripped ions were well centered on the ion-optical axis, the mean momenta of the beams can be determined from the measured field strengths and bending radii of the dipole magnets. The achieved relative accuracy was better than 2×10^{-4}. By means of the mean momenta and of the dispersion, the measured position spectra were transformed into energy-loss spectra, which proved to be Gaussian in all cases. The variances were determined by applying Gaussian fits, and the straggling was obtained from the difference of the variances measured with and without target. The measured variances of the angular and energy-loss distributions of the ions emerging from the targets were in all cases at least five times smaller than the corresponding acceptance of the FRS. Consequently, for each target the complete energy-loss distribution could be recorded with one single magnetic setting. Mainly the precision of dispersion and position-distribution measurement determines the experimental error which is of the order of 25%.

Special efforts were made to prepare suitable targets, since target inhomogeneities can dominate the effect of energy-loss straggling. Thus it is important to minimize the thickness inhomogeneity. These efforts have been described in detail in Ref. [24], but the main features are briefly summarized: The targets were prepared by the GSI target laboratory, and their surfaces were polished in a four-step process in order to achieve a mirrorlike surface. The thickness was determined by means of an optomechanical method [25,26] with a relative accuracy of better than 1%. With a laser interferometric technique [27] the microscopic surface inhomogeneities of the targets were determined, and only those targets which had root mean square thickness variations of less than 0.1 mg/cm^2 were selected for the experiments. In addition, a Ta collimator with 0.3 mm diameter was used in order to eliminate the influence of a possible macroscopic wedged shape of the targets. The targets used for the energy-loss straggling measurements consist of Be, Ti, Au, and Pb and cover a thickness range 1000–8000 mg/cm^2.

In Table I the projectile-target combinations and the specific kinetic energies of the incident ions used in our study are shown together with the theoretical and experimental results. Since the energy loss partly amounted to a significant fraction of the projectile kinetic energy (e.g., 943 MeV/u U ions loose about 25% of their kinetic energy when penetrating through a 4.7 g/cm^2 Au target) the velocity dependence of all parameters contributing to the energy-loss straggling was taken into account by carrying out the calculations according to [8]

$$\Omega_m^2 = \left(\frac{dE(E_1)}{dx}\right)^2 \int_{E_0}^{E_1} \frac{d\Omega_m^2(E')/d(\Delta x)}{[dE(E')/dx]^3}\, dE', \qquad (6)$$

in agreement with the results of Tschalär [28]. Here E_0 and E_1 denote the incident and exit kinetic energy of the projectile, respectively, and the index m denotes the contribution under consideration, i.e., either collisional or charge-exchange straggling. It can be seen from the table that the small momentum transfers as well as the fluctuations of the ionic charge states inside the target cause only small contributions to the straggling, whereas by far the largest contribution arises from close collisions leading to large energy transfers. Within the error bars the experimental values agree well with the sum of charge-exchange, low-Q, and Mott straggling, whereas the sum of charge-exchange, low-Q, and relativistic Bohr result applies only for the lightest projectile.

In order to point out clearly the large systematic deviations from the relativistic Bohr theory the contributions of charge exchange and small momentum transfers have been subtracted from the measured values. This procedure is acceptable since it induces only minor corrections. What remains is the contribution of large energy transfers and the results are shown in Fig. 2 normalized to the relativistic Bohr results. For all data points the projectile velocity in the middle of the targets differs only slightly, i.e., β varies between $\beta = 0.81$ and $\beta = 0.87$. The theoretical results for Mott scattering are shown in the figure for both these velocities. The relevance of Mott scattering for the energy-loss straggling of relativistic heavy ions is clearly confirmed by our measurements, whereas the energy-loss straggling of light ions can be approximated well by the relativistic Bohr formula based on the first-order Born approximation. These results are in satisfactory agreement with results of stopping-power measurements [19]. Of course, for the energy-loss straggling of heavy ions the difference between the perturbation and the exact result is much more pronounced since the second moment of the energy-loss distribution is strongly determined by violent collisions, where the difference between the first-order Born and the Mott cross section is significant.

TABLE I. Theoretical and experimental results for the energy-loss straggling of the investigated projectile-target combinations. The theoretical values are the results according to Eqs. (2)–(5).

Projectile	Incident energy (MeV/u)	Target, thickness (mg/cm^2)	Rel. Bohr Eq. (3) (MeV^2)	Mott Eq. (4) (MeV^2)	Low Q Eq. (2) (MeV^2)	Ch. ex. Eq. (5) (MeV^2)	Experiment (MeV^2)
$^{18}_{8}$O	698	Pb 7931	63.4	67.8	1.46	0	52.9 ± 22.3
$^{136}_{54}$Xe	799	Be 1006	454	732	0.61	47	773 ± 211
		Be 2011	907	1464	1.26	100	1680 ± 409
		Be 4010	1829	2952	2.70	223	3160 ± 701
$^{197}_{79}$Au	988	Ti 1004	1169	2839	6.62	233	2980 ± 647
$^{238}_{92}$U	943	Au 1897	2508	7404	42.8	763	7379 ± 1740
		Au 4672	6147	17 868.8	113.1	2315	18 934 ± 3502

In conclusion, the first measurements of the energy-loss straggling of relativistic heavy ions up to and including uranium have been reported, and the experimental data show quite good agreement with new theoretical results. With respect to the energy-loss straggling the importance of the exact Mott cross section for the scattering of heavy nuclei off electrons has been demonstrated, and the well-known relativistic Bohr result is exceeded by up to a factor of 3.

We would like to thank the FRS technicians and the GSI accelerator staff. The excellent work of the members of the GSI target laboratory is gratefully acknowledged. The support of L. O. T.-Oriel GmbH [27], which provided the laser interferometer was an essential prerequisite for the success of the measurements. Fruitful discussions with M. Inokuti and P. Sigmund are gratefully acknowledged. It is a pleasure to thank A. H. Sørensen and J. Lindhard for helpful discussions and for providing us their latest results concerning the energy-loss straggling.

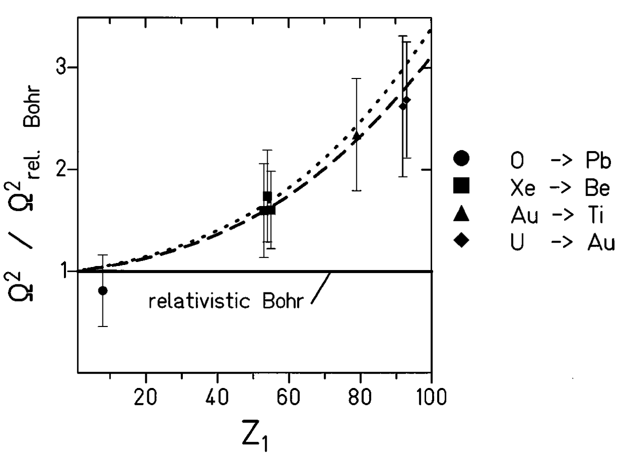

FIG. 2. Ratio of the close-collision energy-loss straggling and the predictions of the relativistic Bohr formula, Eq. (3), as a function of the projectile atomic number Z_1: The theoretical results for Mott straggling are shown for $\beta = 0.81$ (dashed line) and for $\beta = 0.87$ (dotted line).

[1] H. Pfützner *et al.*, Nucl. Instrum. Methods Phys. Res., Sect. B **86**, 213 (1994).
[2] R. Anholt *et al.*, Phys. Rev. A **36**, 1586 (1987), and references therein.
[3] H. Gould *et al.*, Phys. Rev. Lett. **52**, 180 (1984).
[4] Th. Stöhlker *et al.*, Nucl. Instrum. Methods Phys. Res., Sect. B **61**, 408 (1991).
[5] S. P. Ahlen, Rev. Mod. Phys. **52**, 121 (1980), and references therein.
[6] N. Bohr, Philos. Mag. **30**, 581 (1915).
[7] H. Bethe, Ann. Phys. (Leipzig) **5**, 325 (1930).
[8] U. Fano, Ann. Rev. Nucl. Sci. **13**, 1 (1963).
[9] M. Inokuti *et al.*, Phys. Rev. A **23**, 95 (1981).
[10] M. Inokuti (private communication).
[11] N. Bohr, Mat. Fys. Medd. K. Dan. Vidensk. Selsk. **18**, No. 8 (1948).
[12] William McKinley, Jr. and Herman Feshbach, Phys. Rev. **74**, 1759 (1948).
[13] W. R. Johnson, T. A. Weber, and C. J. Mullin, Phys. Rev. **121**, 933 (1961).
[14] B. Rosen, J. Math. Phys. **4**, 392 (1963).
[15] N. F. Mott, Proc. Roy. Soc. (London) A **124**, 425 (1929).
[16] N. F. Mott, Proc. Roy. Soc. (London) A **135**, 429 (1932).
[17] J. Lindhard and A. H. Sørensen, Phys. Rev. A **53**, 2443 (1996).
[18] P. Sigmund, Nucl. Instr. Methods Phys. Res., Sect. B **69**, 113 (1992).
[19] C. Scheidenberger *et al.*, Phys. Rev. Lett. **73**, 50 (1994).
[20] C. Scheidenberger *et al.*, Nucl. Instrum. Methods Phys. Res., Sect. B **90**, 36 (1994).
[21] C. Scheidenberger *et al.* (to be published).
[22] K. Blasche *et al.*, Part. Accel. **32**, 83 (1990).
[23] H. Geissel *et al.*, Nucl. Instrum. Methods Phys. Res., Sect. B **70**, 286 (1992).
[24] Th. Schwab *et al.*, Nucl. Instrum. Methods Phys. Res., Sect. B **48**, 69 (1990).
[25] H. Folger and J. Klemm, in *Preparation of Nuclear Targets for Particle Accelerators*, edited by Jozef Jaklovsky (Plenum Publishing Corporation, New York, 1981), p. 171.
[26] H. Folger *et al.*, Nucl. Instrum. Methods Phys. Res., Sect. A **303**, 24 (1991).
[27] The laser interferometer was provided by L. O. T.-Oriel GmbH, Im Tiefen. See 58, D-64293 Darmstadt.
[28] C. Tschalär, Nucl. Instrum. Methods Phys. Res. **61**, 141 (1968).