Drastic Enhancement of Energy-Loss Straggling of Relativistic Heavy Ions due to Charge-State Fluctuations

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Stopping power and energy-loss straggling of ^{197}Au, ^{208}Pb, and ^{209}Bi projectiles have been measured in different solids ($4 \leq Z_2 \leq 82$) in the energy range (100−1000) MeV/u. The experimental results clearly demonstrate the influence of the different charge states of the ions. Because of charge-state fluctuations the energy-loss straggling is up to 7 times larger than the pure collisional straggling. The selected energy domain in combination with the heavy projectiles allows for the first time an unambiguous interpretation of the long-standing problem of charge-changing collisions in energy-loss straggling.

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In previous publications [1–3] we have shown that the variance of the energy-loss distribution of bare Xe, Au, and U ions systematically deviates by a factor of up to 3 from the well known relativistic Bohr formula [4,5]. However, those experimental results are in good agreement with the predictions of the theory by Lindhard and Sørensen (LS) [6] which applies an exact phase-shift analysis based on the Dirac equation.

The energy-loss straggling of a swift bare ion in matter has its origin in the statistical nature of the collisions with target electrons, i.e., the fluctuation of the number of collisions and the fluctuation of the energy transfer in each collision. For partially ionized projectiles stochastic fluctuations of the ionic charge states due to electron-capture and electron-loss processes lead to the so-called charge-exchange straggling which contributes in addition.

The role of charge-state fluctuations in heavy ion energy straggling has motivated many theoretical [7–10] and experimental [11–17] activities. In general, most theoretical papers suggest a strong influence of charge changing on the energy distribution. However, the lack of knowledge of charge-changing cross sections limits their predictive power. In most previous experimental studies in the MeV/u region the interpretation for solids was severely influenced by artifacts like target thickness variations making an interpretation in terms of charge-exchange straggling impossible. Only few experimental data with reliable target qualities contribute in a more quantitative way for low-Z projectiles to the role of charge exchange [14,16]. Still, a clear separation of the different straggling contributions has not been experimentally possible so far [15].

In the energy range of the present experiments targets of at least several 100 mg/cm^2 were used which have negligible thickness inhomogeneities. In addition, we have the unique possibility to select the target material such that either the collisional straggling or the charge-exchange straggling is the dominant contribution.

The basic quantities determining the variance of the energy-loss distribution due to charge-state fluctuations (Ω^2_{chex}) are illustrated for the case of two charge states in thick targets [7,10]:

$$\Omega^2_{\text{chex}} = 2\,\Delta x\,\frac{F_0 F_1}{N(\sigma_{01} + \sigma_{10})}\,(S_0 - S_1)^2. \tag{1}$$

Besides the target properties like the thickness Δx and the number of atoms per unit volume N the charge-exchange straggling depends on the charge-exchange cross sections σ_{ij} for changing from a charge state with i electrons attached to a charge state with j electrons and the corresponding partial stopping powers S_i. The equilibrium charge-state fractions are denoted by F_i, which are also obtained from the charge-changing cross sections. Therefore, the knowledge of partial stopping powers and charge-changing cross sections is indispensable for a reliable prediction of charge-exchange straggling.

In our calculations the partial stopping powers were obtained from the LS theory including the following corrections [2]: the shell effect (up to 2% in lead at the lowest energy) [18], the Barkas term (up to 5% in lead at the lowest energy) [19,20], and the Fermi density effect (up to 2% in beryllium at the highest energy) [21]. The projectiles were treated as pointlike particles of the ionic charge. The stopping power for charge-state equilibrium can be calculated from the partial stopping powers and the charge-state fractions according to

$$S = \sum_i F_i S_i\,. \tag{2}$$

In Fig. 1 our measured stopping power values ($S_{\text{exp.}}$) for ^{197}Au, ^{208}Pb, and ^{209}Bi projectiles in different solid materials [22] are plotted in comparison with the theoretical predictions according to Eq. (2) inserting the measured F_i. The agreement between data and theory is within the experimental errors of about ±1%. Assuming only bare ions

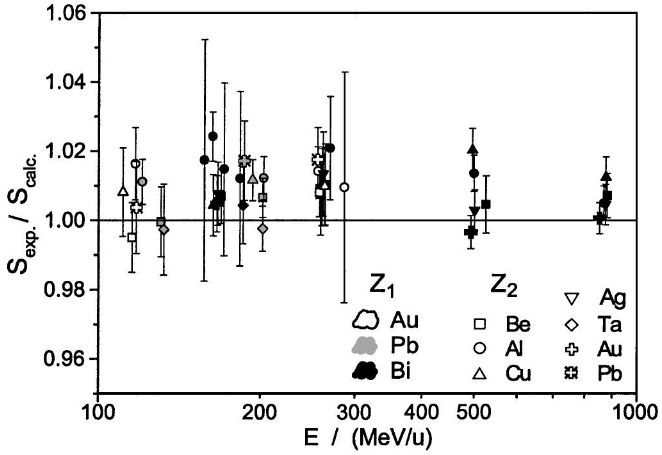

FIG. 1. Experimental stopping powers for ^{197}Au, ^{208}Pb, and ^{209}Bi projectiles in different solid targets (Z_2) normalized by the calculated values of the LS theory [6] with corrections. Equilibrium charge-state populations have been taken into account according to Eq. (2).

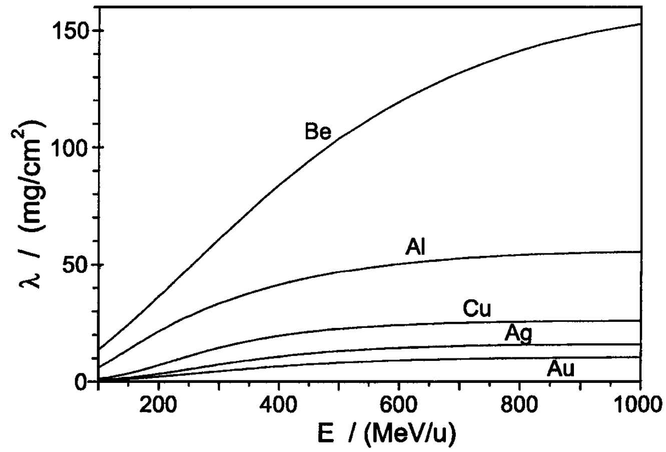

FIG. 2. Calculated mean free path lengths for charge exchange of H-like ^{209}Bi projectiles in different solids.

the calculated stopping powers would differ up to 9% from the experimental ones [22].

The other necessary input for a reliable prediction of the charge-exchange straggling are the cross sections for electron capture and ionization, which have been measured in the present and previous experiments [22–24]. For the low-Z targets, the most interesting ones in this work, the calculated values from the program GLOBAL [24] agree with the experimental data within the error bars of about ±20%. Thus we have detailed knowledge of all parameters for a reliable prediction of $\Omega^2_{\rm chex}$.

The cross sections are related to the mean free path lengths λ_i for a charge-changing event by

$$\lambda_i = 1 \Bigg/ \left(N \sum_j \sigma_{ij} \right). \qquad (3)$$

In case of H-like ^{209}Bi projectiles they are illustrated for different target materials in Fig. 2. It is clearly demonstrated that the mean free path lengths for charge exchange are crucially dependent on the projectile kinetic energy and on the atomic number of the target material; i.e., in beryllium they are more than a factor of 10 larger than in gold targets. This drastic Z_2 dependence provides a unique feature to select target materials with very different contributions from charge-state fluctuations.

The new experimental results on energy-loss straggling have been obtained with the magnetic spectrometer FRS at the GSI UNILAC/SIS heavy ion accelerator facilities [25] using the same procedure as in our earlier studies [1,22]. The measurements were performed in the energy range (100–1000) MeV/u in different solid target materials ranging from beryllium to gold. For each material and incident energy the energy-loss distribution was measured for three targets of different thickness. Depending on the projectile energy and the target material the equilibrium charge-state distributions are characterized by three significantly contributing charge states at 1000 MeV/u and up to seven at 160 MeV/u. The most prominent fractions are bare ions at the high energy and He-like for the low energy [22,24].

In Fig. 3 the new energy straggling data of ^{209}Bi projectiles in different solid materials at 600, 290, and 200 MeV/u incident energy are presented. They are compared with pure collisional straggling according to the LS theory and with the results of a Monte Carlo simulation. In the Monte Carlo code the theoretical probabilities for charge-changing collisions were implemented as statistically selected free path lengths and the slowing down calculated with the LS theory. It should be noted that the charge-exchange straggling sensitively depends on the difference of the partial stopping powers, cf. Eq. (1).

The charge-exchange straggling can also be treated analytically as it has been described by Sigmund [9]. We applied this method at higher energies for an equilibrium charge-state distribution of three charge states and it agrees with the results of our Monte Carlo calculation.

In our experiments with the high-resolution magnetic spectrometer we measured the energy distribution of the ions entering and leaving the target in a single, the most probable, charge state. Therefore, our simulations were also made for this case. The resulting straggling would be even larger for a complete equilibrium charge-state distribution throughout the target; e.g., for beryllium at 600 MeV/u the enlargement would be 10%.

The comparison of the results at 600 MeV/u clearly demonstrates that we obtain excellent agreement with the LS theory for collisional straggling in case of heavier target materials, whereas strong deviations are observed for light target materials like beryllium where charge-exchange straggling becomes important. The Monte Carlo simulation can reproduce the heavier-Z material very well and indicates already a strong transition to larger straggling values for the lighter target materials.

The energy-straggling results are 290 and 200 MeV/u incident energy are presented in the middle and lower parts

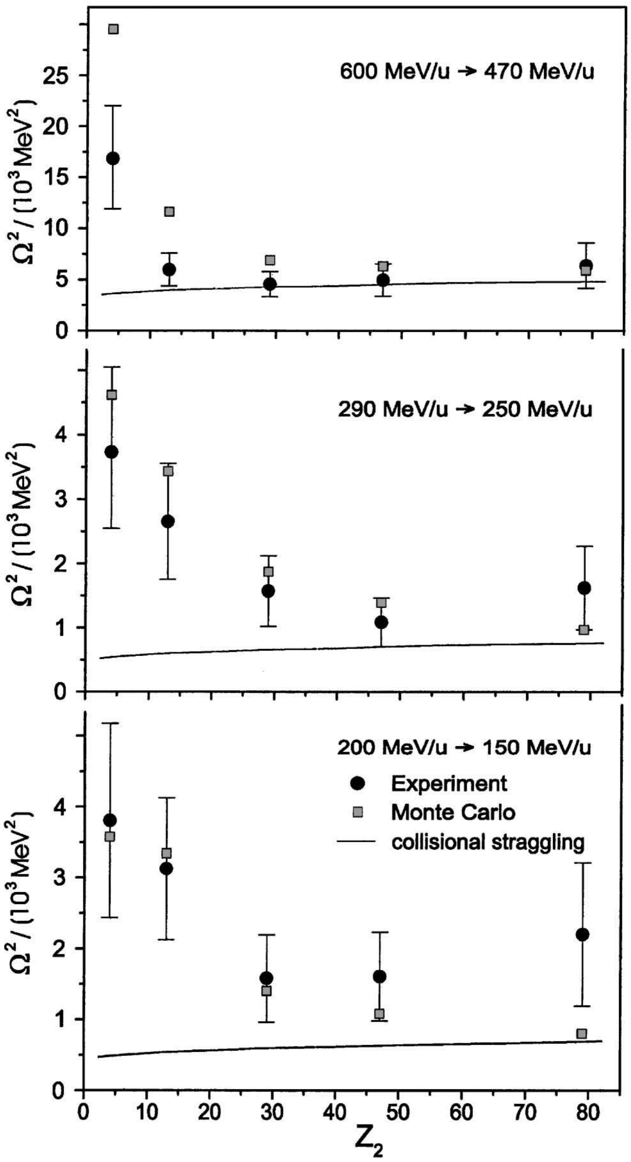

FIG. 3. Experimental energy-straggling values (circles) of ^{209}Bi projectiles in different solid target materials compared to a Monte Carlo simulation (squares) and the contribution from pure collisional straggling according to LS theory (line). The incident and exit energies are stated in the figure.

of Fig. 3. The experimental values show again systematic deviations from the LS theory. At these low energies the charge-exchange straggling yields an important contribution also for heavy materials and for the light media (Be, Al) the pure collisional prediction is too low by a factor of about 7.

We conclude that the different statistics of collision processes in low- and high-Z targets (see also Fig. 2) yield completely different contributions to the straggling and that the observed deviation is a direct consequence of the charge-changing collisions. This insight is in general affirmed by the Monte Carlo results, which are in good agreement with the experimental data over the entire Z_2 range, except for the overestimation in beryllium and aluminum at 600 MeV$/u$. In principle, the simulation at 600 MeV$/u$, characterized by the lowest number of charge states, should be the easiest.

The deviation can be caused by the uncertainties of the partial stopping powers and the charge-changing cross sections. A change of the cross sections by more than a factor of 2 would force agreement but is inconsistent with former cross-section measurements [23,24]. Therefore, we attribute the observed discrepancy to incomplete screening for close collisions.

Using the nonrelativistic screening corrections of Refs. [26,27] already reduces the difference between experiment and simulation by a factor of 2. At the lower velocities closer collisions contribute less to the partial stopping powers; therefore, such screening estimations change the simulated values only within the experimental error bars. Presently, there is no theoretical screening theory in the relativistic velocity regime. Therefore, one can use the comparison between model calculations and experimental energy-straggling data to deduce quantitative information on the incomplete screening for partial stopping powers. Note that the discussed screening corrections would shift the mean stopping powers in Fig. 1 only within the error bars towards even better agreement with theory, whereas the straggling calculations would be changed significantly. This statement is also confirmed by new straggling experiments in the range from 600 to 1000 MeV$/u$ [28].

In case the width of the collisional energy-loss distribution is much bigger than the maximum energy transfer in a single collision one expects Gaussian shaped collisional energy-loss distributions. However, as was shown by Närmann and Sigmund [10] there may also exist skewness due to projectile charge exchange. This skewness again becomes important for large mean free path lengths, and it reflects the asymmetry of the charge-state distribution. When the largest fraction of ions is fully stripped, the charge-state distribution is skewed and the corresponding energy-loss distribution has a tail towards less energy loss. When the K shell of the projectile is mostly filled and there are still very few electrons in the L shell, the observed asymmetry changes to the other side. At lower velocities the L shell is also filled, and again the skewness of the energy-loss distribution can change its sign. Hitherto such skewness effects have not been observed.

Figure 4 shows the experimental skewness for ^{209}Bi projectiles in beryllium and silver targets as a function of the projectile kinetic energy in comparison with the results of our Monte Carlo calculation. These two materials are representative for the different magnitude of charge-exchange straggling. For beryllium we observe an asymmetry, whereas for silver a Gaussian energy-loss distribution was obtained as expected from the dominance of the collisional straggling. The skewness and its dependence on the velocity are in good agreement with the

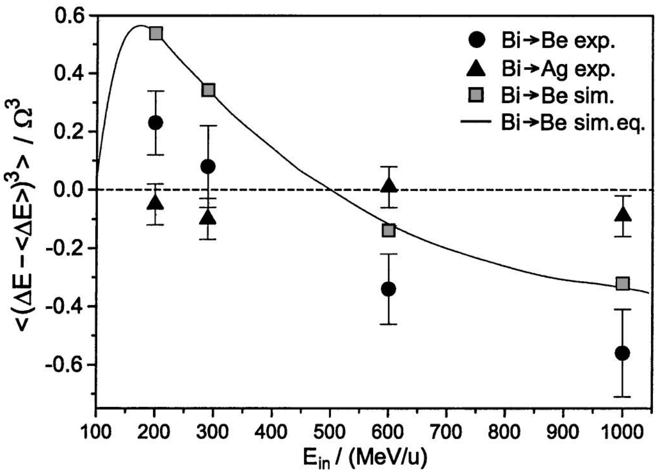

FIG. 4. Skewness, third moment of the energy-loss distribution $\langle(\Delta E - \langle\Delta E\rangle)^3\rangle$ normalized by the standard deviation Ω, measured (exp.) and simulated (sim.) as a function of the incident energy for 20% energy loss. The curve would be obtained for an equilibrium charge state distribution through the target (sim.eq.).

results of the simulation. Considering the full charge-state distribution, instead of only a single charge state at the entrance and exit of the target, does not change the illustrated skewness effects. We want to stress that the measured skewness is only due to the presence of charge-exchange straggling. Therefore, it persists even for thick targets in contrast to the well known Landau distribution caused by collisional straggling [29].

In conclusion, both the stopping powers and the charge-changing cross sections are well known in the relativistic energy regime of the present experiment. Moreover, the quite different charge-exchange cross sections of high- and low-Z targets both yield a unique possibility to disentangle the different contributions to the energy-loss straggling. This knowledge is the base of a clear interpretation and quantitative description of the energy-straggling results. The present experiment demonstrates for the first time unambiguously the dominant contribution of charge changing to heavy ion energy straggling characterized by the second and the third moments.

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