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Z_2 structure and gas–solid effect in the stopping of slow ions

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Abstract

A recent claim by Paul of a systematic gas–solid difference in stopping cross sections for ions such as nitrogen and oxygen in the velocity range $v \simeq v_0$ is studied on the basis of existing experimental data. We find that all existing data support the commonly known Z_2 structure which, by and large, follows the valence structure of the target material. Existing experimental evidence is not found to support a specific gas–solid difference in the velocity range under consideration. The possibility of such an effect due to a gas–solid difference in charge state is rejected on theoretical grounds. Data for compound gases and solids are found to be well described by the Bragg additivity rule.

We have also studied nitrogen/helium and oxygen/helium stopping ratios which determine the so-called effective-charge ratio. Taking into account the scatter of experimental data, we do not find clear evidence against Northcliffe's assumption of a stopping ratio independent of Z_2 and common for gases and solids in the considered velocity range, although the absolute value appears too high.

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1. Introduction

Z_2 structure or Z_2 oscillations is the common term denoting the observation that the electronic stopping cross section of an elemental material for an energetic ion does not depend monotonically on its atomic number Z_2, as had been expected originally on the basis of Bloch's implementation [1] of the Bethe theory [2].

First indications of a nonmonotonic behavior were identified experimentally by Burkig and MacKenzie [3] with MeV protons and substantiated in subsequent work, especially in [4,5]. Similar results were found for He ions with energies around 1 MeV/u in a detailed comparison of available range data by Chu and Powers [6] who, at the same time, presented a theoretical estimate that predicted increasing amplitudes of the observed oscillatory structure with decreasing ion energy. Experimental and theoretical studies demonstrated that Z_2 structure persists into the keV regime for both H and He ions [7], and measurements with Li [8,9] and N [10] as well as several heavier ions [11] revealed similar findings.

The gas–solid effect in the stopping of heavy ions denotes a systematic difference in stopping cross section

(1) either between a group of gaseous and otherwise similar solid materials, especially materials of similar atomic number Z_2;
(2) or between a given material in the gaseous and the solid state.

An effect of the first kind had been searched for as a consequence of the well-known gas–solid difference in the

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equilibrium charge state of fission fragments [12] and was found experimentally for swift heavy ions over a quarter of a century later [13,14]. It was found small, of the order of 10–20%, stopping cross sections being higher in the solid phase, and consistent in sign with the difference in charge state which, however, may be significantly greater.

An effect of the second kind was searched for but not found by comparing stopping cross sections of α particles in solid and gaseous argon [15]. Only more recently was such an effect identified in a comparison of the stopping cross sections of Zinc in the vapor and solid phase [16], following up on a theoretical prediction [17]. For 25 keV protons, the stopping cross section in the gas phase was reported to be 60% higher than for the solid.

In a recent study, based on analysis of experimental data from the literature, Paul [18] concluded

(1) that there is a gas–solid difference in stopping cross sections also for ions much lighter than fission fragments, in particular nitrogen and oxygen;
(2) that it is the solid materials that have higher stopping cross sections;
(3) that this difference persists to low velocities around and below v_0, the Bohr speed;
(4) that there are systematic differences in equilibrium charge states of ions penetrating solid and gaseous targets in this velocity range, and
(5) that it is this difference that is primarily responsible for the gas–solid difference in stopping cross sections.

In short, 'the well-known density effect in the stopping power for fast ions, explained by the gas–solid difference in the mean equilibrium charge, persists down to low energies' [18].

Even without a detailed analysis, several objections may be made against this surprising conjecture:

- Separating a gas–solid effect from Z_2 structure is notoriously problematic, since inert gases tend to have low stopping cross sections due to their high binding energies.
- In the absence of a charge-state effect, atoms in the condensed state tend to have lower stopping cross sections than isolated atoms because of higher binding energies.
- The majority of stopping data for gas targets refers to inert gases. Therefore, in a statistical analysis, the average behavior of all gases will be dominated by the average behavior of the inert gases.
- Experimental as well as theoretical evidence on gas–solid differences in charge states for the variety of ions and velocities in question is scarce.
- Clear evidence on ion-solid differences in charge state has been established mostly for ions much heavier than nitrogen and oxygen [11].
- Theoretical experience indicates a very minor influence of the charge state of nearly neutral ions of comparatively low atomic numbers on the stopping cross section [19,20].

Moreover, inspection of the empirical basis of the arguments brought forward in [18] indicates that a number of features were overlooked in the analysis:

- The element with the highest stopping cross section for both nitrogen and oxygen ions at $v \simeq v_0$ is hydrogen. This feature, which is not necessarily consistent with the claim of gases having lower stopping cross sections, is not mentioned in [18].
- While the *inert gases* have low stopping cross sections indeed, other gases like N_2, O_2 and atmospheric air fall roughly on line with neighboring solid materials, cf. Fig. 2 in [18] and the corresponding figure for oxygen in [21].
- Only a small fraction of the data in Paul's large database [21] entered the argument.

Despite these cautionary remarks, the above conjecture is important and deserves attention. Since it is empirically based and unsupported by theoretical arguments, we find it appropriate to investigate its empirical basis in detail. Theoretical arguments will enter occasionally to clarify the issue.

2. Preliminaries

As the empirical basis of our analysis we use Paul's database [21] which is not only close to comprehensive but, as far as we can tell from experience, reliable and loyal to the original sources.

However, it is instructive to have a brief look at how the problem under consideration was handled in the tabulation by Northcliffe and Schilling [22], which, although out of date now, has set standards for all subsequent work on stopping of ions heavier than protons.

Fig. 1 shows mass stopping forces $\mathrm{d}E/\rho\mathrm{d}x$ ($\rho =$ mass density) on protons at $v = v_0$ as a function of the atomic number Z_2 of the stopping medium. Separate lines 'to guide the eye' have been drawn in the upper graph, connecting data for gaseous (filled circles) and solid (filled triangles) targets, respectively. It is seen that with the exception of the high-Z_2 end, the line for gases lies below the one for solids. However, inclusion of the data for zinc vapor (empty circle) and solid zinc (empty triangle) from [16] shows a gas–solid difference with the opposite sign, in agreement with the theoretical expectation [17]. Therefore, the two lines in this graph are in direct contradiction with the measured gas–solid difference in the stopping of low-velocity protons.

In the lower graph in Fig. 1, we have drawn connecting lines reflecting the valence structure of the stopping material without consideration of the state of aggregation. This has been accomplished by means of power-law fits according to

$$-\frac{\mathrm{d}E}{\rho\mathrm{d}x} = BZ_2^{-q} \tag{1}$$