Scanning Microscopy

Volume 10 | Number 1 Article 1

12-11-1995

The Phase Effect in Electronic Stopping: A Survey of the Contributing Physical Processes

P. Bauer
Johannes Kepler University Linz, bauer@exphys.uni-linz.ac.at

E. Steinbauer
Johannes Kepler University Linz

F. Kastner
Johannes Kepler University Linz

Scanning Microscopy, Vol. 10, No. 1, 1996 (Pages 1-7)
Scanning Microscopy International, Chicago (AMF O'Hare), IL 60666 USA

0891-7035/96$5.00+.25

THE PHASE EFFECT IN ELECTRONIC STOPPING: A SURVEY OF THE CONTRIBUTING PHYSICAL PROCESSES

P. Bauer*, E. Steinbauer and F. Kastner

Institut für Experimentalphysik, Johannes-Kepler-Universität Linz, A-4040 Linz, Austria

(Received for publication May 7, 1995 and in revised form December 11, 1995)

Abstract

The phase effect in electronic stopping deals with the question whether the energy loss of an ion due to the interaction with electrons depends on the state of aggregation of the target. It is commonly accepted that charge changing collisions of the projectile and changes in the electronic states of the target contribute to the phase effect. In addition, the energy loss measurements might possibly be influenced by different impact parameter selection in the two experiments (solid and gas phase). Quantitative results of our calculations show that generally the impact parameter selection inherently present in a transmission experiment is quenched by the inevitable multiple scattering of the projectiles. Thus, electronic excitation and ionization in the projectile and the target are the only processes that contribute significantly to the phase effect.

Key Words: Ions, electronic energy loss, phase effect, charge changing collisions, excitation and ionisation, impact parameter selection, multiple scattering.

***Address for correspondence:**
Peter Bauer
Institut für Experimentalphysik
Abteilung für Atom- und Kernphysik
Johannes Kepler Universität
Altenbergerstraße 69
A-4040 Linz
Austria.

Telephone number: +43 7434 43792
FAX number: +43 732 2468 9677
E-Mail: bauer@exphys.uni-linz.ac.at

Introduction

The stopping of charged particles is of fundamental importance for any kind of ion beam application, such as target characterization, ion implantation, radiation therapy, etc. {see e.g., Børgesen (1991); IAEA-TECDOC-506 (1989)}. The energy range of interest (from a few keV to several MeV) and the interesting range of projectile atomic numbers, Z_1, are growing steadily. In addition, the desired level of accuracy is rising. While for experiments and applications the mass stopping power, $-dE/\rho dx$, expressed in eV per $\mu g/cm^2$, is a useful quantity, in theory the stopping cross section, ε, is the basic quantity. ε is defined by

$$\varepsilon = \int_0^\infty T\, d\sigma = \int_0^\infty T(b) P(T,b)\, 2\pi b\, db \qquad (1)$$

as sum over all possible energy losses T weighted by the corresponding cross section $d\sigma = 2\pi P(T,b) b.db$, b being the impact parameter and P(T,b) the probability of energy loss T at impact parameter b.

The relation between stopping power and stopping cross section is given by

$$-\frac{dE}{\rho dx} = \frac{\varepsilon}{M_2} \qquad (2)$$

where M_2 is the mass of the target atom (or molecule). At high energies, the projectiles are bare ions and target excitation and ionization is the only contribution to the stopping cross section. At lower energies, the ions can carry bound electrons while traversing a target material. Here, the capture and loss of electrons by the projectile will also contribute to the energy loss. This may be explicitly included in the definition of ε by using the charge state approach {Arnau (1994) and references therein}

$$\varepsilon = \sum_{i,j} \Phi_i (\varepsilon_i + \sigma_{ij} \cdot T_{ij}) \qquad (3)$$

where Φ_i and ε_i denote the probability to find the ion in charge state i (i = 0, ±1, ±2,...) and the partial stopping cross section for this fixed charge state i, resp., and σ_{ij} and T_{ij} denote the charge changing cross section (capture and loss) and the associated energy loss when the projectile charge state changes from i to j.

While there are numerous systematic studies of the electronic stopping of light ions in pure elemental targets (see e.g., Semrad *et al.* 1986), there are only very few systematic studies of how the stopping power of molecules is influenced by chemical bonds (*chemical effects*, see e.g., Bauer *et al.* 1992b) and of the influence of the state of aggregation (*phase effect*).

Comprehensive reviews of the literature on the phase effect in electronic stopping are given by Thwaites (1983), Thwaites (1985) which show that there is only a small number of investigations of this effect. Measurements have mainly been reported for light ions (H^+, He^+, Li^+) in inorganic and organic compounds (like H_2O, alkanes) and in gases (like O_2, Ar). One of the most frequently studied systems is H_2O as vapor and ice, because of its relevance for radiation therapy. Due to the fact that the changes in the valence electron states in these materials are minor, the observed phase effect is rather small. As demonstrated by Thwaites (1985), the general trend is that the energy loss in the solid phase is *smaller* than in the gas phase and the difference of the stopping cross sections increases with decreasing energy (up to ~10%). It is, however, difficult to deduce quantitative results from these studies because of the wide spread of the data from different labs.

For the frozen gases O_2, N_2, CO and Ar the findings are rather confusing: while Besenbacher did not find a phase effect for He^+ ions in Argon at the stopping power maximum (Besenbacher et al. 1981), Børgesen and co-workers found big effects in some and no effect at all in other gases for slow H^+ ions {see Børgesen (1985) and references therein}. This is a consequence of the fact that these experiments are difficult and it is not easy to obtain clear evidence how big these effects are. However, huge effects have been predicted by theory for point charge projectiles in metals and their vapors {Sabin *et al.* (1989); Meltzer *et al.* (1990)}. Independent of the detailed assumptions of the theory, a large phase effect should be expected for metals and vapors where the valence electron states are completely different in the different phases.

The Darmstadt group has measured the gas-solid difference for fast heavy projectiles and found significant effects {see e.g., Geissel *et al.* (1982); Geissel (1985)}, the stopping cross sections of the solids being higher than those of the gases, apparently due to projectile inelastic processes (see below).

In this review, we will describe the possible sources which contribute to the phase effect. As a model system we choose a monoatomic material which is a metal in the solid phase (Zn). This has the advantage that the physical processes involved are separated more easily from chemical effects additionally present in the case of compound targets. Furthermore, for the metal vapor system the changes of the valence electronic states and therefore also the phase effect are much more pronounced than for the frozen gas systems.

In principle, there are three different contributions to the phase effect:

1. Differences in the mean ionization potential of the target material due to the change of the state of aggregation (*target contribution*).

2. Differences in the charge changing processes of the ions in the different states of aggregation (*projectile contribution*).

3. Different impact parameter selection in the two measurements (solid and gas phase) due to different experimental geometries *(impact parameter selection)*.

These contributions are discussed in the following sections.

Target contribution

While wave functions and ionization energies (I_{nl} for the subshell n,l) of the inner shell electrons are nearly unaltered when the state of aggregation changes, the states of the valence electrons are altered significantly. From the atomic energy levels, Bethe's I-values (see eq. 4) are obtained as summation over all possible energy losses due to excitation and ionization, weighted by the corresponding dipole oscillator strengths. As a result, the I-values for solids I_{sol} are larger than the corresponding value for the gas phase I_{gas} (see e.g., ICRU Report 49, 1993). Consequently, the energy loss of bare ions is larger in the gas phase than in the solid or liquid phase.

As mentioned above, $I_{gas} < I_{sol}$ directly corresponds to $\varepsilon_{gas} > \varepsilon_{sol}$. This may be seen most easily from the Bethe equation which is valid for bare charges Z_1 at sufficiently high velocity, v. This theory relates ε to the number of target electrons, Z_2, and to Bethe's I-value, and is given in its simplest form by

$$\varepsilon = \frac{4\pi e^4 Z_1^2 Z_2}{mv^2} \cdot \ln \frac{2mv^2}{I} \qquad (4)$$

where m and -e are mass and charge of the electron. From this we get

$$\varepsilon_{gas} - \varepsilon_{sol} = \frac{4\pi e^4 Z_1^2 Z_2}{mv^2} \cdot \ln \frac{I_{sol}}{I_{gas}} > 0. \qquad (5)$$

This qualitative argument applies to all point charge projectiles for which capture and loss of electrons do not occur and therefore no projectile inelastic processes contribute to the energy loss.

For materials which are weakly bound in the solid phase like frozen gases or organic compounds, the phase effect is small due to the fact that the states of the valence electrons do not differ strongly. For example, the recommended mean ionization potentials of H_2O-water and H_2O-vapor are 75.0eV and 71.6 eV, resp., (ICRU Report 49, 1993) giving rise to a relative phase effect $\Delta\varepsilon/\varepsilon$ = 0.013 for 1 MeV protons which is so small that it can hardly be measured at these high energies. The effects are larger for lower energies (Bauer *et al.*, 1994) and for metals and their vapors due to completely different valence states and large lattice energies of the solid. Calculations of the phase effect in zinc gave $\Delta\varepsilon/\varepsilon \approx 0.1$ for 700 keV protons (Arnau *et al.*, 1994). At 700 keV, the neutral fraction is negligibly small so that the phase effect is entirely due to the target contribution.

Projectile contribution

The charge state of the projectiles fluctuates due to capture and loss of electrons bound to the projectile. The mean charge is defined only as the average of the instantaneous states. In the simplest case of a system with two charge states, the contribution to the stopping cross section due to capture and loss of electrons by the projectile is given by

$$\varepsilon_{cc} = \frac{\sigma_c \cdot \sigma_l}{\sigma_c + \sigma_l} \cdot (T_c + T_l) \qquad (6)$$

Here σ_c and σ_l denote the capture and the loss cross section, resp., and the sum $T_c + T_l$ describes the energy necessary to bring an electron from its initial state (bound to the target) via the intermediate state bound at the projectile to the final state (free electron state above the Fermi level). Allison and co-workers were the first to prove the usefulness of this concept {Allison *et al.* (1962); Huberman, (1962)}.

For protons, the contribution of charge changing collisions peaks at energies where the mean charge is 1/2, i.e. at typically 30 - 50 keV, and it amounts to a small fraction of the total stopping cross section (see e.g., Arnau 1994). Projectile inelastic processes contribute to the phase effect if the charge changing cross sections are different in different states of aggregation. There is, e.g., a density dependence of the electron loss probability postulated for heavy ions by Bohr and Lindhard (1954), who predicted a higher loss efficiency in the dense phase due to the higher collision frequency. Measurements with fast heavy ions by Geissel *et al.* (1982); see also Geissel (1985), yielded higher stopping cross sections in solids as compared to gases even in cases where the mean ionization potentials would give rise to the opposite expectation and thereby confirmed the model of Bohr and Lindhard qualitatively. These findings have been confirmed by the Orsay group {see e.g., Bimbot *et al.* (1989a); Bimbot *et al.* (1989b)}. Apparently the projectile contributions dominate the target contributions in the case of fast heavy projectiles.

Impact parameter selection

In addition to the contributions discussed above which are well documented in literature there is a further possible mechanism: a different impact parameter selection in the two experiments (solid and gas phase) could give rise to differences in ε {Martinez-Tamayo *et al.*, (1995)}. The argument could go as follows: a transmission experiment selects particles which are transmitted with little deflection and therefore have not suffered close collisions . This means that the particles reaching the detector were scattered only with impact parameters larger than a minimum impact parameter b_{min}. Thus, the possible energy losses T(b) are restricted (see eq. 1) and the resulting energy loss cross section ε^* is smaller than ε:

$$\varepsilon^* = \int_{b_{min}>0}^{\infty} T(b)P(T,b)2\pi b db \quad < \varepsilon \qquad (7)$$

would result. This argument does, however, not fully apply in reality, because the impact parameter selection due to the experimental geometry is often quenched by multiple scattering. Qualitatively speaking, all projectiles transmitted through the foil have been scattered through angles of up to about $\alpha_{1/2}$, where $\alpha_{1/2}$ is the half width angle of multiple scattering. Especially at low energies, $\alpha_{1/2}$ can exceed the experimental acceptance angle of a typical transmission experiment by a large amount. Systematic comparisons between transmission and backscattering experiments (Semrad *et al.*, 1986) have shown that the stopping power results from both measuring methods agree within the uncertainty of the experiments, i.e. within 3%, if the experiments are performed correctly. This shows that the influence of impact parameter selection in transmission geometry (see eq. 7) can be considerably reduced in practical cases.

This line of arguments need, however, not apply to transmission through a gas cell (Schiefermüller *et al.*, 1993). In this case, the exit aperture selects only those particles that leave the vapor cell on the axis and at angles close to 0°. In this case it is not at all clear to which extent the impact parameter selection is quenched by multiple scattering. We therefore have

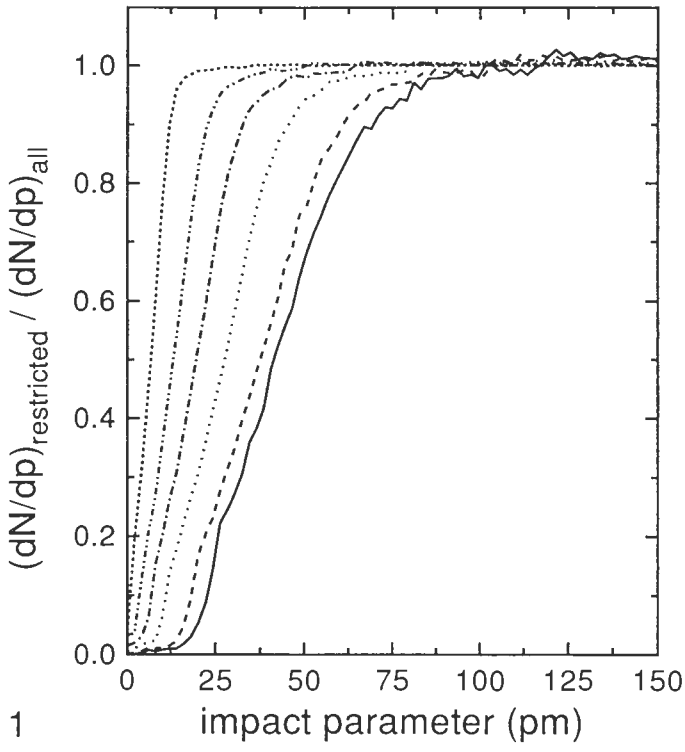

Figure 1: The ratio of calculated impact parameter distribution functions $(dN/dp)_{restricted}/(dN/dp)_{all}$ is shown as a function of impact parameter for protons transmitted through a zinc vapor of 2.7×10^{16} atoms/cm^2 areal density. The assumed length of the vapor cell is 30 cm. The calculations have been performed for the energies 40 keV (full line), 60 keV (dashed line), 130 keV (short dashed line), 240 keV (dot-dashed line), 430 keV (..-..-) and 700 keV (-----).

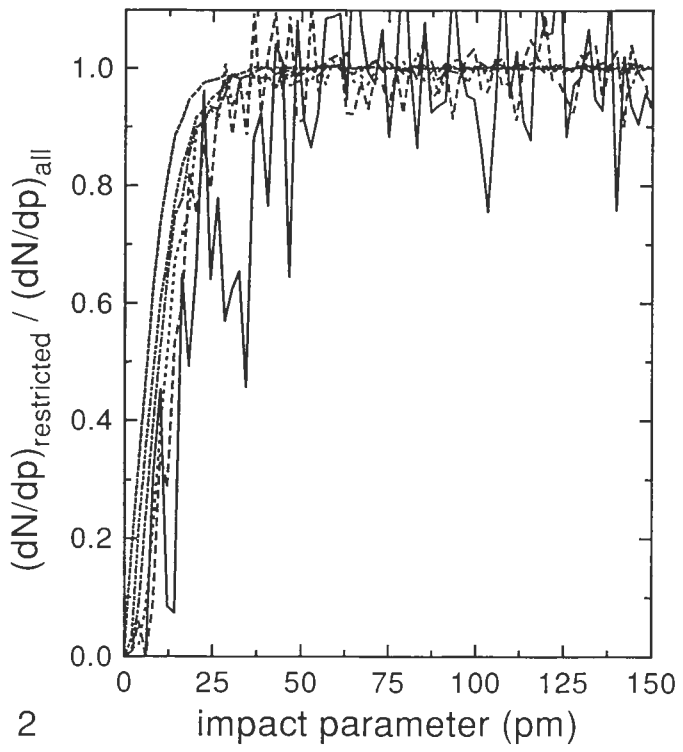

Figure 2: The ratio of calculated impact parameter distribution functions $(dN/dp)_{restricted}/(dN/dp)_{all}$ is shown as a function of impact parameter for protons transmitted through a zinc vapor of 2.7×10^{17} atoms/cm^2 areal density. The assumed length of the vapor cell is 30 cm. The calculations have been performed for the energies 40 keV (full line), 60 keV (dashed line), 130 keV (short dashed line), 240 keV (dot-dashed line), 430 keV (..-..-) and 700 keV (-----). The scatter of the data at 40 keV indicates that the intensity of the protons exiting the vapor cell at the axis (within 0.5 mm) at an angle <1° is extremely low.

---

performed Monte-Carlo simulations of the ion trajectories through a vapor cell of 30 cm length, with apertures of 1mm diameter and with an acceptance angle of the detector of 1°. The code is a derivation of the well known code TRIM TC (Biersack and Eckstein, 1984 and references therein) with the modification that the impact parameters are registered for all projectiles along their paths. A dilute Zn vapor was chosen as target and protons as projectiles. All scattering angles larger than 0.1° are treated as scattering events in a screened potential ('universal potential'), the electronic energy loss is subtracted after each collision. In all calculations, the total relative energy loss $\Delta E/E$ is smaller than 8%. The calculations have been performed in the energy range 40 - 1000 keV for two vapor densities, corresponding to the minimum and the maximum density used in our experiment (Bauer *et al.*, 1992a), i.e. 9.10^{14} Zn atoms/cm^3 and 9.10^{15} Zn atoms/cm^3 (Steinbauer *et al.*, 1996).

The results are shown in Fig. 1 and Fig. 2 where the ratios of the impact parameter distributions, $(dN/db)_{rest}/(dN/db)_{all}$, are given. $(dN/db)_{rest}$ is the statistical distribution of impact parameters experienced by those projectiles which leave the vapor cell through the exit aperture at angles smaller than 1°, while $(dN/db)_{all}$ is the impact parameter distribution of all calculated histories. The absence of impact parameter selection is equivalent to a horizontal line at $(dN/db)_{rest}/(dN/db)_{all} = 1$. As shown in Figs. 1 and 2, in both cases all impact parameters larger than 100 pm fully contribute to the transmitted particles.

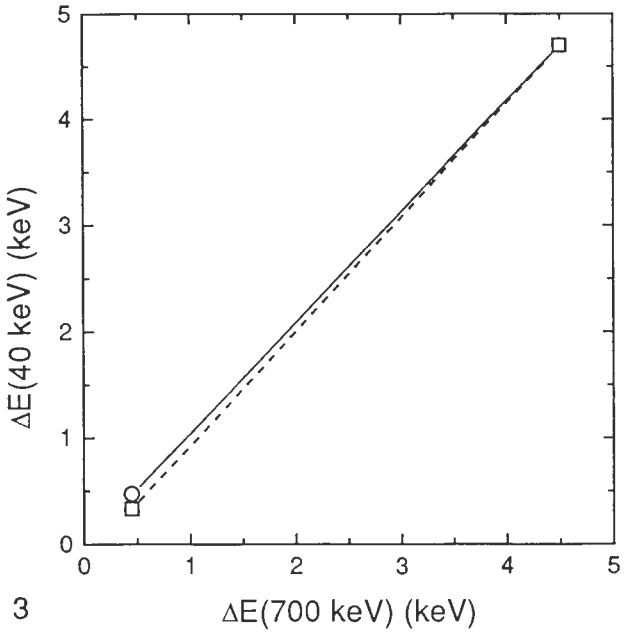

Figure 3. Visualization of the evaluation procedure to obtain the stopping ratio at 40 keV and at 700 keV, ε (40 keV)/ε (700 keV), from the measured energy loss values using Eq.(8). The influence of a hypothetical systematic error of 30% at the low vapor density (see text) on the evaluated stopping cross section is indicated by the dotted line.

For *low density* (Fig. 1), the cut-off values depend on the ion energy. At high energies, the cut-off occurs at impact parameters less than 10 pm which is small compared to the spatial extension of the Zn 4s-wave function. Therefore, the effect on ε is negligible at high energies. At 40 keV, the cut-off is at approximately 50 pm and may reduce the measured energy loss value ΔE_1(40 keV). This reduction is small, because the probability to miss the Zn 4s electrons at impact parameters ≥ 50 pm is < 5%. The observed energy dependence of the cut-off reflects the fact that for a given impact parameter the scattering angles become smaller with increasing energy. Therefore, smaller impact parameters are needed at high energies to scatter the projectiles off the beam direction.

At *high density* (Fig. 2), the cut-off appears at impact parameters less than 10 pm independent of the ion energy. This is due to the following reasons: first, multiple scattering leads to half width angles $\alpha_{1/2}$ which increase with decreasing energy, exceeding the assumed acceptance angle of the detector (1°) at low energies. Second, for a given impact parameter (10 pm) the scattering angle increases with decreasing energy in almost the same way as $\alpha_{1/2}$ does. For this density, the influence of the cut-off on the measured energy loss ΔE_2 is negligible at all energies.

The important question of the data evaluation is how to obtain stopping cross section data from the measured energy losses. Applying the standard procedure of evaluating ε as the ratio of ΔE over nΔx would for the low densities introduce systematic errors in ε of the same magnitude as the error in ΔE, due to the impact parameter selection. An alternative approach is to fix ε at high energies where systematic errors are negligible, e.g., at 700 keV, and calculate ε at lower energies E from the ratio ε (E)/ε (700 keV). This ratio is obtained as the slope in the plot of the energy losses ΔE_i(E) (i=1, 2), measured at the areal densities $n_i\Delta x$ (i=1, 2), versus the corresponding energy losses ΔE_i(700 keV):

$$\frac{\varepsilon(E)}{\varepsilon(700\text{keV})} = \frac{\Delta E_2(E) - \Delta E_1(E)}{\Delta E_2(700\text{keV}) - \Delta E_1(700\text{keV})} \qquad (8)$$

Fig. 3 shows how this procedure minimizes the influence of impact parameter selection on the stopping data. The larger the difference between the densities n_1 and n_2 is, the less is the slope influenced by a shift of the measured energy loss ΔE_1. In case $n_1/n_2 = 0.1$, the slope is influenced just by 1/10 of the reduction of ΔE_1(40 keV). Thus, an assumed reduction by 10% of ΔE_1(40 keV) would lead to a systematic error of 1% and therefore be negligible.

We conclude that for the densities necessary to measure energy losses in vapors (and gases), the impact parameter selection is in most cases quenched by multiple scattering and when a relative measurement is performed a further reduction of the systematic error is achieved leading to negligible errors. Thus, we finally end up with the fact that in phase effect measurements using light projectiles, e.g., for protons in Zn, only target and projectile contributions are responsible for the observed phase effect.

Acknowledgments

We would like to express our gratitude to A. Arnau, P. M. Echenique and A. Salin for many inspiring discussions, and express our thanks to J.P. Biersack for making available the TRIM TC program.

References

Allison SK, Cuevas J, Garcia-Munoz M (1962) Partial ionic stopping power and the energy expended in electron capture and loss collisions of protons in hydrogen gas. I, Phys Rev **127**: 792-798.

Arnau A (1994) Charge states and energy loss of ions in solids. Nucl Instr Meth **B93**: 195-202.

Arnau A, Bauer P, Kastner F, Salin A, Ponce VH, Fainstein PD, Echenique PM (1994) Phase effect in the energy loss of hydrogen projectiles in zinc targets. Phys Rev **B49**: 6470-6480.

Bauer P, Kastner F, Arnau A, Salin A, Fainstein PD, Ponce VH, Echenique PM (1992a) Phase effect in the energy loss of H projectiles in Zn targets: Experimental evidence and theoretical explanation. Phys Rev Lett **69**: 1137-1139.

Bauer P, Rössler W, Mertens P (1992b) Stopping of hydrogen ions in oxides: Influence of the chemical bond. Nucl Instr Meth **B69**: 46-52.

Bauer P, Käferböck W, Necas V (1994) Investigation of the electronic energy loss of hydrogen ions in H_2O: Influence of the state of aggregation. Nucl Instr Meth **B93**: 132-136.

Besenbacher F, Bøttiger J, Graversen O, Hansen JL, Sørensen H (1981) Stopping power of frozen argon for He ions. Nucl Instr Meth Phys Res **188**: 657-668.

Biersack JP, Eckstein WG (1984) Sputtering Studies with the Monte-Carlo program TRIM.SP. Appl Phys **A34**: 73-94.

Bimbot R, Cabot C, Gardes D, Gauvin H, Hingmann R, Orliange I, De Reilhac L, Hubert F (1989a) Stopping power of gases for heavy ions: gas-solid effect.I. 2-13 MeV/u Ne and Ar projectiles. Nucl Instr Meth **B44**: 1-18.

Bimbot R, Cabot C, Gardes D, Gauvin H, Orliange I, De Reilhac L, Subotik K, Hubert F (1989b) Stopping power of gases for heavy ions: gas-solid effect.I. 2-6 MeV/u Cu, Kr and Ag projectiles. Nucl Instr Meth **B44**: 19-34.

Børgesen P (1985) Measurements of the stopping power for keV light ions in condensed molecular gases. Nucl Instr Meth **B12**: 73-79.

Børgesen P (1991) Rutherford backscattering spectroscopy. In: Analysis of Microelectronic Materials and Devices. Grasserbauer M, Werner HW (eds.). John Wiley and Sons, Chichester, pp. 543-562.

Bohr N, Lindhard J (1954) Electron capture and loss by heavy ions penetrating through matter. Dan Mat Fys Medd **28** No. 7: pp. 1-31.

Geissel H (1985) Heavy-ion stopping. In: Semiclassical descriptions of atomic and nuclear collisions. Bang J, de Boer J, (eds.). North Holland, Amsterdam, pp. 431-462.

Geissel H, Laichter Y, Schneider WFW, Armbruster P (1982) Energy loss and energy straggling of fast heavy ions in matter. Nucl Instr Meth **194**: 21-29.

Huberman MN (1962) Partial ionic stopping power and the energy expended in electron capture and loss collisions of protons in hydrogen gas. II, Phys Rev **127**: 799-804.

IAEA-TEC-DOC-506 (1989) Atomic and Molecular Data for Radiotherapy. International Atomic Energy Agency, P.O.Box 100, A-1400 Vienna, Austria, pp. 51-72 and 136-160.

ICRU Report 49 (1993) Stopping powers and ranges for protons and alpha particles. International commission on radiation units and measurements (ICRU), 7910 Woodmont Avenue, Bethesda, MD 20814, USA. pp 6-33 and 48-106.

Martinez-Tamayo G, Eckart JC, Lantschner GH, Arista NR (1995) Energy loss of protons in Zn: Measurements between 2 and 200 keV. Phys Rev **A51**: 2285-2288.

Meltzer DE, Sabin JR, Trickey SB (1990) Calculation of mean excitation energy and stopping cross section in the orbital local plasma approximation. Phys Rev **A41**: 220-232.

Sabin JR, Oddershede J (1989) Stopping powers and mean excitation energies of the transition metals from iron to zinc. Phys Rev **A39**: 1033-1040.

Schiefermüller A, Golser R, Stohl R, Semrad D (1993) Energy loss of hydrogen projectiles in gases. Phys Rev **A48**:, 4467-4475.

Semrad D, Mertens P, Bauer P (1986) Reference proton stopping cross sections for five elements around the maximum. Nucl Instr Meth **B15**: 86-90.

Steinbauer E, Salin A, Arnau A, Diez-Muiño R, Kastner F, Bauer, P (1996). Nucl Instr Meth **B**, in print.

Thwaites DI (1983) Bragg's rule and stopping power additivity: A compilation and summary of results. Radiat Res **95**: 495-518.

Thwaites DI (1985) Current status of physical state effects on stopping power. Nucl Instr Meth **B12**: 84-89.

Discussion with Reviewers

J. Schou: It is known that the band gap of many frozen gases is much less than the ionization energy of the gas, (Ar-gas: 15.76 eV, Ar-solid: 14.16 eV, Kr-gas: 14.0 eV, Kr-solid: 11.01 eV, etc., from Zimmerer G (1987) in: Excited spectroscopy in solids. Grassano UM and Terzi N. (eds.) North Holland, Amsterdam, p. 37). This is valid for water ice as well: The band gap is about 8 eV (Baron B, Hoover D, Williams F (1978) J Chem Phys **68,** 1997), whereas the gas value is about 12.6 eV. Similarly, the ionization potential is about 1 eV lower for solid oxygen and carbon monoxide than for the gases. These values contrast the statement of the authors that the ionization value for gases always is larger than that of the solids. Which consequences does this gas-solid difference have for the target contribution?

Authors: You are right, the band gap of frozen gases is smaller than the ionization energy of the gases. However, the band gap of the frozen gases should not be compared with the ionization energy of the gases, but with the minimum excitation energy which is smaller than the corresponding band gap. E.g., the minimum excitation energy of a H_2O molecule in the gas phase is about 7.4 eV which is considerably less than the band gap of 8 eV of H_2O ice. Bethe's I-value includes excitations as well as ionization, thus it is not contradictive that the I-values of the gases are lower than for the corresponding solids, in accordance with the experimental findings that ε_{gas} is larger than ε_{solid} for bare ions.

F. Flores: Can you compare the relative importance of target and projectile contributions? How important are the charge changing processes in the phase effect?

Authors: Phase effect calculations for zinc by Arnau *et al.* (1994) show that excitation and ionization of the zinc valence electrons by projectiles of fixed charge state (0 or +1) are the dominating contribution to the phase effect. This means on the other hand that projectile excitation and charge changing collisions play a minor role in the case of hydrogen projectiles. One should, however, keep in mind that the charge states depend on both target and projectile properties.

For heavy ions the situation is completely different as demonstrated by the measurements by Geissel *et al.* (1982), {see also Geissel (1985)} where the phase effect in the projectile charge states dominates over the changes in target valence states.