Nuclear Instruments and Methods in Physics Research B 372 (2016) 119–126

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Inelastic cross-sections and energy loss properties by non-relativistic heavy ions in zirconium dioxide

Jennifer Schofield [a,*], Simon M. Pimblott [a,b]

[a] School of Chemistry, The University of Manchester, Oxford Road, M13 9PL, UK
[b] The University of Manchester Dalton Cumbrian Facility, Westlakes Science and Technology Park, Moor Row, Cumbria CA24 3HA, UK

ARTICLE INFO

Article history:
Received 17 September 2015
Received in revised form 14 January 2016
Accepted 1 February 2016
Available online 21 February 2016

Keywords:
Heavy ion
Inelastic cross-section
Stopping power
ZrO_2
DOSD

ABSTRACT

A formalism for the inelastic cross-section for electronic collisions of protons and heavier ions in a material is developed based on a quadratic extrapolation of the experimentally based dipole oscillator strength distribution (DOSD) of the material into the energy momentum plane. The approach is tested by calculating various energy loss properties in zirconium dioxide. Mean free path, stopping power and continuous slowing down approximation (*csda*) range are predicted as a function of ion energy for various incident ions, with the stopping powers compared to experimental data to assess the effectiveness of the methodology. The DOSD is straightforwardly obtained from the experimentally measured energy loss function data below 80 eV and atomic photo-absorption cross-section data above 100 eV. Agreement between the results of the calculation for stopping power and the experimental data is within 10% for all ions when compared for energies greater than the Bragg peak. The discrepancy is larger below the peak due to limitations in the methodology, especially the failure to make corrections for the Barkas and higher order effects and the lack of charge cycling cross-section data.

1. Introduction

There have been numerous studies of the energy loss properties of charged particle radiation in matter since Bethe's pioneering work [1]. Over the last 20 years, extensive effort has been spent attempting to incorporate material and phase information, in particular Ashley has proposed an approach based on the use of the dipole oscillator strength distribution (DOSD) for a material [2]. This formalism has subsequently been deployed by a number of research groups [3–7] primarily to look at the energy loss of electrons, positrons and protons in water [3,4]; however there are limited applications to other materials including DNA and hydrocarbons [7,8]. The simulation and microscopic understanding of the structure of ion tracks in a material relies upon the availability of suitable cross-sections for the various collision processes, especially electronic ionisation and excitation and charge exchange between the irradiation particle and the medium.

This study describes the development formalism for the facile prediction of the inelastic cross-section for electronic ionisation and excitation of ions in a wide variety of materials which avoids the use of a model for the general oscillator strength (GOS) for each material. The formalism is based on the approximation developed by Ashley and the use of experimental DOSD from optical data thereby allowing significant flexibility and applicability. The formalism is applied to zirconium dioxide (ZrO_2) in order to assess its effectiveness. Elucidating radiation damage and the energy loss characteristics of ions in ZrO_2 is an important challenge in understanding the performance of light water reactor fuel cladding for both in-reactor operation and interim storage, as zirconium is the principal component of LWR fuel cladding.

In this paper, the methodology used for calculating both the inelastic cross-sections and energy loss properties of heavy ions is described, detailing the application of the 'Ashley' approximation to heavy ions and describing considerations of the effective charge of the incident ion on the stopping power. The DOSD of ZrO_2 is formulated from the available experimental data and the energy dependent inelastic cross-section, *Y* function and mean free path for electronic ionisation and excitation are evaluated as well as various energy loss properties, specifically the stopping power, and *csda* (continuous slowing down approximation) range. Mean energy loss characteristics for incident ions in ZrO_2 are compared, where possible, to experimental data.

2. Methodology

The electronic collision cross-section as well as stopping power and energy loss properties of an ion in a material can be related to

* Corresponding author.
E-mail address: Jennifer.schofield@postgrad.manchester.ac.uk (J. Schofield).

http://dx.doi.org/10.1016/j.nimb.2016.02.001
0168-583X/

the dielectric response function $\varepsilon(p,\gamma)$ for the medium in question. This complex function gives information on how a medium responds to a momentum, p, or energy, γ, transfer to it. When a fully stripped ion with incident energy $E = MV^2/2$ enters a material the probability of the ion undergoing an energy loss per unit distance is given by [3]

$$\tau(E,\gamma) = \frac{Z^2}{\pi E}\int Im[-1/\varepsilon(p,\gamma)]dp/p \tag{1}$$

where the reduced atomic units $e = m_e = \hbar = 1$ are used (and are used throughout this work) with e the electronic charge, m_e the electron mass, $\hbar$ Planck's constant and M the reduced mass of the ion. The Z^2 term in Eq. (1) distinguishes the expression for heavy ions from that for singly charged electrons and positrons, as noted in reference [9].

The probability of an energy loss event can be related to the inelastic collision cross-section by

$$\sigma(E) = \frac{1}{N}\int \tau(E,\gamma)d\gamma \tag{2}$$

with N being the number density of molecules in the medium. Furthermore,

$$\sigma(E) = (N\Lambda(E))^{-1} \tag{3}$$

where Λ is the mean free path of an ion in the medium. The stopping power, $S(E)$, and *csda* range, $R(E)$, are given by the two equations

$$S(E) = \int \gamma\tau(E,\gamma)d\gamma \tag{4}$$

$$R(E) = \int \frac{1}{S(E)}dE. \tag{5}$$

Crucially, the expressions for the energy loss properties in a medium for an ion given above rely on $Im[-1/\varepsilon(p,\gamma)]$, the imaginary part of the complex dielectric response function. This function is not available from experiment for most materials, however, the energy loss function (ELF) for optical data, where $p = 0$, is much easier to obtain. Ashley has suggested an approximation to allow the evaluation of the energy loss properties, namely, the quadratic extension of the DOSD into the energy–momentum plane [2]

$$Im[-1/\varepsilon(p,\gamma)] = \int \frac{\gamma'}{\gamma} Im\left[-\frac{1}{\varepsilon(0,\gamma')}\right]\delta\left(\gamma - \left(\gamma' + \frac{p^2}{2}\right)\right)d\gamma' \tag{6}$$

where γ' can be identified as a 'binding energy' associated with the molecules in the medium. This approximation allows us to use solely optical data in the previous Eqs. (1)–(5).

When evaluating the expressions (1)–(5) of an ion interacting with the electrons in a medium the kinematic constraints of the situation must be considered to determine the bounds of integration. The ion with mass M and incident energy $E = MV^2/2$ is assumed to collide with an electron at rest. The ion then leaves with energy $MV_1^2/2$ and the electron has energy $v_2^2/2$ with the contribution of γ' the 'binding energy' given to the system. According to energy conservation, we can say that $MV^2/2 = MV_1^2/2 + v_2^2/2 + \gamma'$. We can also define the 'energy loss' as $\gamma = (V^2 - V_1^2)M/2 > \gamma'$, which gives through simple rearrangement, $V_1 = \sqrt{V^2 - 2\gamma/M}$.

For all possible energy losses, the value of γ'_{min} is 0 while the maximum value of γ'_{max} always corresponds to the minimum possible momentum transfer for a given energy transfer. By applying conservation of momentum, the following inequality for the minimum momentum transfer is defined

$$p_{min} = M(V - V_1) < p. \tag{7}$$

When combined with the definition for V_1 above and the fact that $v_2 = \sqrt{2(\gamma - \gamma')}$, Eq. (7) becomes

$$\gamma' < \gamma - M^2\left(V^2 - \frac{\gamma}{M} - V\sqrt{V^2 - \frac{2\gamma}{M}}\right) = \gamma'_{max}. \tag{8}$$

We can now apply a Taylor expansion to the square root terms in Eq. (8) as $\gamma \ll MV^2/2$ for an ion colliding with an electron; giving

$$\gamma'_{min} = 0 < \gamma' < \gamma - \gamma^2/2V^2 = \gamma'_{max} \tag{9}$$

Taking the right hand side of Eq. (9) allows us to determine the limits on energy transfer as

$$\gamma_{min} = V^2 - V\sqrt{V^2 - 2\gamma'} < \gamma < V^2 + V\sqrt{V^2 - 2\gamma'} = \gamma_{max}. \tag{10}$$

These inequalities, Eqs. (9) and (10), define the bounds of integration to a symmetric curve in the energy-transfer/binding-energy plane with allowable values under the curve; see the hashed area in Fig. 1.

Applying these constraints to Eq. (1) gives the following probability of energy loss,

$$\tau(E,\gamma) = \frac{Z^2}{2\pi E}\int_{\gamma'_{min}}^{\gamma'_{max}} Im[-1/\varepsilon(0,\gamma)] - G(\gamma,\gamma')\gamma' d\gamma' \tag{11}$$

where $G(\gamma,\gamma') = \frac{1}{\gamma(\gamma-\gamma')}$. From this we are able to define the inelastic cross-section and stopping power for electronic collisions as:

$$\sigma(E) = \frac{Z^2}{2N\pi E}\int_0^{V^2/2} Im\left[\frac{1}{\varepsilon(0,\gamma')}\right]\ln\left[\frac{(1+s-a)(1-s)}{(1-s-a)(1+s)}\right]d\gamma' \tag{12}$$

$$S(E) = \frac{Z^2}{2\pi E}\int_0^{V^2/2} \gamma' Im[1/\varepsilon(0,\gamma')]\ln\left[\frac{1+s-a}{1-s-a}\right]d\gamma' \tag{13}$$

with $a = \gamma'/V^2$ and $s = \sqrt{1 - 2\gamma'/V^2}$. From Fig. 1 it is clear to see that when evaluating total cross-sections $\gamma'_{min} = 0$ and $\gamma'_{max} = V^2/2$.

When modelling ion track structures, the simulations follow the trajectory of an ion collision by collision until the energy of the ion is less than a defined cut-off energy. The nature of a collision is determined from relative cross-sections for each type of collision at the energy of the ion, i.e. ionisation, excitation, vibration or

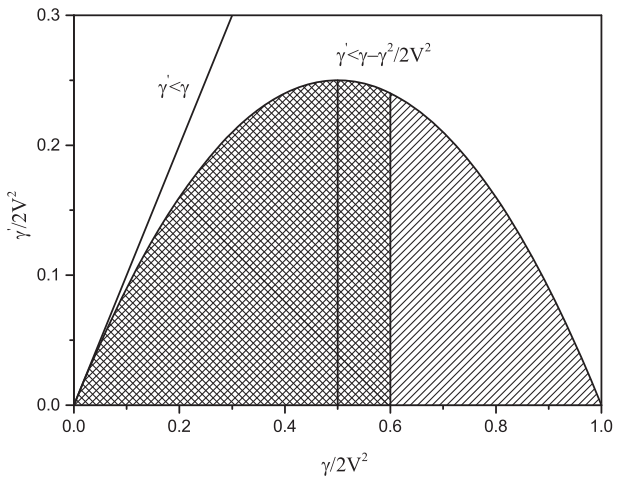

Fig. 1. The allowed integration region of the $\gamma - \gamma'$ energy plane for heavy ion–electron collisions. When evaluating cumulative inelastic cross-sections for energy transfers greater than $\gamma = V^2$ careful consideration of the bounds is necessary. For example, when integrating at 0.6 as shown in the graph (double hashed), one must calculate the area from 1 to 0.6 and subtract this from the whole cross-section.