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On the validity of trajectory methods for calculating the transport of very low energy (<1 keV) electrons in liquids and amorphous media

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HIGHLIGHTS

- Low energy electrons do not follow well-defined trajectories.
- However, trajectory simulation is applicable, i.e. valid.
- Limits of validity are reviewed for a point scatterer system.
- An extrapolation to liquids and amorphous solids is proposed.

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ABSTRACT

It is easily demonstrated that a trajectory picture of low energy electron transport in condensed matter is not compatible with the Heisenberg uncertainty principle. The uncertainty in the position of a low energy electron is large and may in fact be larger than an entire simulated trajectory. This might be interpreted to mean that trajectory methods are not applicable. However, this conclusion is not correct. In the present paper, the evidence for the validity of low energy electron trajectory simulation is discussed, as well as the wave aspects and quantum nature of low energy electron transport in liquids and amorphous solids.

It is pointed out that the validity of a trajectory approach to low energy electron transport in a liquid or amorphous solid partly is due to its ability to reproduce the average results of coherent elastic multiple wave scattering in a randomlike medium, and moreover that this ability may be further enhanced by the presence of inelastic scattering. The resulting validity of the trajectory method may be referred to as circumstancial validity, which is of a nature different from the explicit validity of trajectory methods which are compatible with the uncertainty principle.

A previous systematic analysis of the limits of circumstancial validity is revisited and discussed for the basic case of multiple elastic scattering of a particle in a random medium of point scatterers. The detailed limits of circumstancial validity are graphically demonstrated in terms of particle wavelength, average distance between scatterers and elastic mean free path. Their immediate applicability to neutron transport is noted. The approximate nature of the point scatterer model as regards electron transport is adressed. In order to obtain an extrapolation of the result of the point scatterer model, it is observed that an increasing error of the trajectory method appears together with an increased amplitude of the multiple wave scattering taking place within the medium. On the basis of this observation, an extrapolation is proposed which provides a rough estimate of the relative error of the trajectory method when applied to multiple elastic scattering of low energy electrons in real liquids or amorphous solids.

1. Introduction

The scattering and energy loss in condensed matter of electrons with energies below 1 keV is of interest in for example photoelectron and Auger electron spectroscopy, secondary electron emission theory and, not least, in microscopic approaches to the theory of biological effects of radiation. As regards the last subject it is now recognized that electrons with energies about and below a few hundred eV are of major importance in radiation damage to the DNA molecule in the cell nucleus (Nikjoo and Girard, 2012; Nikjoo et al., 2012; Nikjoo and Lindborg, 2010). In view of the fact that the living cell predominantly consists of water, the transport of such electrons in liquid water is of particular interest. In what follows, we will use this case as an illustrative and important example.

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Electron plural and multiple scattering in condensed matter is often calculated by means of trajectory methods, usually Monte Carlo trajectory simulation. We recall the standard assumptions made in a detailed electron trajectory simulation. The medium is regarded as a continuum in which the electron moves like a free particle, except when subject to discrete elastic or inelastic scattering events. The probability for the electron to move a distance s without any scattering event is $\exp(-s/\lambda_t)$, where λ_t is the total mean free path, given by $\lambda_t=(n\sigma_t)^{-1}$. Here, n is the density of scatterers and σ_t is the total cross section per scatterer. In the calculation of scattering cross sections, the electron is assumed to be scattered elastically by single atoms or possibly by small molecules. Inelastic interaction is also assumed to be with single atoms or molecules, except for collective interaction modes such as plasmon excitation and phonon creation or annihilation. Basically, quantum methods or empirical data have to be used to calculate all these individual cross sections. This standard detailed trajectory simulation is equivalent to analytic trajectory methods such as solving the classical Boltzmann particle transport equation, assuming that the same mean free paths are used.

As an example, to be further discussed below, a simulated trajectory of a 100 eV electron in liquid water is shown in Fig. 1 with a cut-off at 10 eV. It should be remarked that the cross sections for such low energy electrons in liquid water are uncertain, in particular as regards the elastic scattering (Emfietzoglou et al., 2005, 2013; Nikjoo et al., 2006; Liljequist et al., 2012). The simulation in Fig. 1 was made with the track structure code KURBUC, details of which are described e.g. by Nikjoo et al. (2006). The inelastic interaction is based on a recent dielectric response model for liquid water (Emfietzoglou et al., 2005), while the elastic interaction at low energies is based on empirical data for scattering from free water molecules (Nikjoo et al., 2006). Of particular interest in the present context are the elastic and the inelastic mean free paths λ_e and λ_i used in the simulation (Table 1).

The trajectory method is an approximation, and clearly not valid in the presence of manifest electron diffraction phenomena. As will be discussed in some detail below, it may from the uncertainty principle also be suspected that trajectory methods are generally invalid if the electron wavelength λ is not much smaller than the average distance d_{nn} between nearest neighbour scatterers in the medium; this distance is conveniently estimated as

$$d_{nn} = n^{-1/3} \tag{1}$$

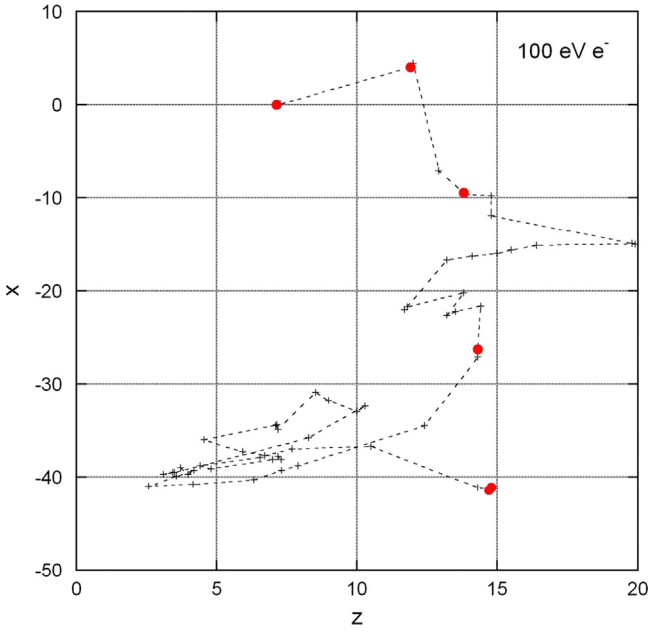

Fig. 1. One simulated primary electron trajectory in liquid water for an initial electron energy of 100 eV and a cut-off energy 10 eV, obtained with the Monte Carlo track structure code KURBUC. Elastic events are shown as crosses and inelastic events as dots. For clarity no secondary electrons are shown in the picture. The scale is in Å (10^{-10} m). Elastic and inelastic mean free path values are exemplified in Table 1.

Table 1
Examples of the elastic mean free path λ_e and the inelastic mean free path λ_i at different electron energies E, used in the KURBUC simulation of the electron trajectories in liquid water shown in Fig. 1.

E (eV)	λ_e (Å)	λ_i (Å)
100	7.0	15.2
50	4.1	17.5
20	2.3	37.5
10	1.7	78.6

If trajectory methods are invalid, full quantum methods for calculating the single, plural or multiple electron scattering have to be used (Pendry, 1974). An example from a biological context is given by calculations of the coherent multiple elastic scattering of electrons within the DNA molecule (Caron and Sanche, 2003; Caron et al., 2009).

However, quantum calculations of plural or multiple scattering tend to be very complex as well as computationally very time-consuming (Ankudinov et al., 2002), whereas Monte Carlo trajectory simulation is, by comparison, very simple. An estimate of the limits of validity of trajectory simulation is therefore definitely of practical interest in all applied calculations where low energy electron transport is involved.

The remainder of the present article is organized as follows. In Section 2, we discuss the application of the uncertainty principle to a low energy electron trajectory in condensed matter. We point out evidence showing that trajectory methods can be used also at very low electron energies, although the physical content of the trajectory concept is then quite different from the classical one. In Section 3, we discuss a previously found expression for the relative error (RE) of the trajectory method when applied to multiple elastic scattering of a particle in clusters of point scatterers (Liljequist, 2009). On basis of this expression, we present a useful graphical display of the detailed limits of validity of the trajectory method. In Section 4 we address the fact that while the point scatterer is a good model e.g. for the elastic scattering of low energy neutrons by nuclei, it is at most a lowest-order approximation for the elastic scattering of low energy electrons by atoms. In Section 5, we propose an extrapolation of the point scatterer expression for RE to low energy electron multiple elastic scattering in media with more realistic atomic or molecular elastic scattering potentials.

2. The uncertainty principle versus wave propagation in random media. Circumstancial validity of the trajectory method

It is readily shown that a low energy electron in condensed matter cannot be said to follow a well-defined trajectory such as illustrated in Fig. 1. For the argument, assume first that the electron is initially localized to the vicinity of an atom or an molecule, i.e. with an uncertainty $\Delta x \approx d_{nn}$ in the particle position, measured along an arbitrary x-axis. The uncertainty principle,