Chapter 1
Electron Transport in Solids

The Monte Carlo (MC) method is used for evaluating the many physical quantities necessary to the study of the interactions of particle-beams with solid targets. Studies of backscattered and secondary electrons are of great interest for many analytical techniques. A better comprehension of the processes which occur before the emission of backscattered and secondary electrons would allow a more comprehensive understanding of surface physics.

1.1 Motivation: Why Are Electrons Important

Electrons continuously interact with the matter around us. Plasma processing of materials, electron lithography, electron microscopy and spectroscopies, plasma-wall in fusion reactors, interaction of charged particles with the surfaces of space-crafts, and hadron therapy represent only a few technological examples where electrons are involved and play a role.

In fact, we use electron beams for our purposes, either on the front of production of materials or on that of their characterization. Let us think of the many applications such as processing of materials with plasma and of the local melting of materials for joining large components. We use electron beams also in electron lithography, an important technique utilized for the production of microelectronics devices. Let us consider the importance of the beams of electrons in material characterization, performed using techniques such as electron microscopy and all electron spectroscopies. Electrons interact with the surfaces of space-crafts. Plasma-wall interaction in fusion reactors also involves electron-matter interaction. Electrons play a role also in cancer proton therapy, where cascades of secondary electrons are produced. These very low energy electrons are toxic for human body cells, since they produce damage to the biomolecules due to ionizations/excitations and the resulting break of chemical bonds. Also secondary electrons which have ultra-low energy – and which, for a long

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time, were thought to be relatively harmless – are dangerous for biomolecules due to the so-called *dissociative electron attachment*. And, of course, we want to minimize the effects of irradiation on the healthy tissues near to the diseased cells.

In all the cases above, modeling the interaction of electrons with matter is very important, as it can be used to provide a solid theoretical interpretation of experimental evidence.

These are the reasons for which an accurate and detailed study of the interaction mechanisms of electrons with matter is of paramount importance.

1.2 The Monte Carlo Method

The world is ruled by quantum mechanics. The investigation of the processes of electron-matter interaction requires the use of quantum mechanics-based techniques. And since, typically, the number of particles involved in these processes is huge, it is crucial to use statistical approaches, such as those represented by the Monte Carlo method. This method provides a very accurate description of many of the phenomena that we can observe in nature when electrons interact with materials.

The Monte Carlo method is a numerical procedure which uses random numbers, theory of probability, and statistics to evaluate multiple integrals.

Suppose we need to calculate the area of a closed surface. In order to do so, we can surround the curve with a square of known side. Then we generate a large number of random points inside the square. Whenever a random point falls within the surface, we update a counter. When the number of points is very large, the ratio of those fallen within the surface and the total number of generated points will approach the ratio between the (unknown) area of the surface and the (known) area of the square.

It is very important to emphasize that, when the number of dimensions is high (as it is when dealing with all statistical problems) the Monte Carlo method is the best numerical procedure for the calculation of multiple integrals. Complicated problems of physics involving very large numbers of particles can be addressed with the Monte Carlo method: it can realize real numerical simulations of physical processes such as the interaction of an electron beam with a solid.

The Monte Carlo method is used, in particular, for evaluating the many physical quantities necessary to the study of the interactions of particle-beams with solid targets. By letting the particles carry out an artificial, random walk – taking into account the effect of the single collisions – it is possible to accurately evaluate the diffusion process [1–4].

1.3 The Monte Carlo Ingredients

To work properly, the Monte Carlo method needs a set of input data describing the interaction of the particle beam with the target. Such data specify the kind of materials and, of course, the kind of incident particles. The interactions of the particles

impinging on the specimen with the target atoms can be described by the cross-sections that describe different physical phenomena. In fact various kinds of interaction occur during the passage of the electrons through the material.

In particular we will investigate the elastic electron-atom processes, described by the so-called elastic scattering cross-section. The elastic scattering cross-section can be described by an equation known as the *screened Rutherford cross-section* formula. This is an analytical expression valid when the energy of the incident electrons is relatively high and the atomic numbers of the target atoms are relatively low, as it was deduced within the so-called *first Born approximation*. But, since the case of low energy electrons and high atomic numbers is not well described by such a simple formula, in order to simulate the elastic scattering cross-section with a more general formulation – valid for all energies and atomic numbers – a more complex approach is necessary, which is known as the *relativistic partial wave expansion method* (Mott cross-section [5]).

Concerning the inelastic scattering cross-section, we will use semi-empirical analytical formulas, when possible, and the so-called dielectric Ritchie's theory [6] for dealing with the more general cases.

When electron energy becomes relatively small (let us say, smaller than 20–30 eV, depending on the investigated material) another very important mechanism of energy loss (and energy gain) is related to the creation (and annihilation, respectively) of phonons. For describing such a phenomenon, we will introduce the electron-phonon cross-section utilizing the Fröhlich's theory [7].

In the end, in many cases also trapping phenomena are important and need to be considered in the simulations. They can be due to the polarization of materials (insulators) induced by the passage of very slow electrons through them [8] and/or also to defects in the materials. When dealing with insulating materials, trapping phenomena are mainly due to the so called polaronic effect, i.e., to the creation of quasi-particles constituted by slow electrons with the polarization field around them [8]. In the case of metals and semiconductors, traps are mainly due to defects in the materials (i.e., impurities, structural defects, grain boundaries etc.).

1.4 Electron-Beam Interactions with Solids

During their travel in the solid, the incident electrons lose energy and change direction at each collision with the atoms bound in the solid. Because of the large difference between the masses of the electron and the atomic nucleus, nuclear collisions deflect electrons with very small kinetic energy transfers. This process is described by the differential elastic scattering cross-section (which can be calculated by the so-called relativistic partial wave expansion method, corresponding to the Mott cross-section [5]). The Mott cross-section can be approximated with the screened Rutherford formula: this is possible when the conditions corresponding to the first Born approximation are satisified, i.e., for high energy and for low atomic number of the target atom. Additionally, excitation and ejection of atomic electrons, and excitation of

plasmons, affect the energy dissipation. These processes only slightly affect the direction of the incident electron in the solid, so that they can be described as inelastic events. Plasmon excitations are ruled by the equations for the differential inverse inelastic mean free path, calculated by the use of Ritchie's dielectric theory [6]. The Fröhlich theory [7] can be used for describing the quasi-elastic electron-phonon interactions in insulating materials. Electron-phonon interactions are considered quasi-elastic for the corresponding energy losses and gains are very small when compared to the plasmon energy losses. When, in insulating materials, electron kinetic energies considerably decreases, trapping phenomena due to the polaronic effect have to be taken into account as well [8].

While for electron kinetic energies higher than 10 keV, MC simulations provide excellent results by just using the Rutherford differential elastic scattering cross-section (elastic scattering) and the Bethe-Bloch stopping power formula or semi-empirical stopping power[1] formulas (inelastic scattering), when the electron energies become much smaller than 5 keV – and this is the case of secondary electron emission – this approach fails [10]. There are many reasons, and the most important ones are related to the three following facts:

(i) As the Rutherford formula is a result of the first Born approximation, it is a high energy approximation.
(ii) Also the Bethe-Bloch formula is valid only for quite high energies; in particular, the Bethe-Bloch stopping power does not provide the correct predictions when the electron energy E becomes smaller than the mean ionization energy I. It reaches a maximum and then approaches zero as E approaches $I / 1.166$. Below $I / 1.166$, the predicted stopping power becomes negative. The use of semi-empiric approaches can sometimes mitigate the problem. Actually, numerical approaches based on the calculation of the dielectric function - as a function of the energy loss and of the momentum transfer - are necessary to calculate low energy inelastic processes.
(iii) The inclusion of the stopping power in the MC code corresponds to the use of the so-called *continuous-slowing-down approximation* (CSDA). Such a way of describing energy losses completely neglects that actually electrons lose their energy in several inelastic collisions. Sometimes an electron can even lose all its energy in a single collision. In other words, any realistic model of the electron trajectories should avoid the approximation of continuity in describing the electron energy losses. CSDA can be used (and will be used, when possible, in the present work as well) but only in cases where the details of the energy loss mechanisms are not crucial for the accurate description of the process under investigation. CSDA can be used, for example, for the calculation of the backscattering coefficient. We will see that, in some specific cases, even the calculation of the secondary electron yield can be performed using CSDA. On

[1] In this book we will use the expression *stopping power* instead of *stopping force* to indicate the energy loss per unit distance of the electron in the solid. Even if consistent with the units, and hence more accurate, the use in the literature of the expression *stopping force*, as observed by Peter Sigmund [9], is only slowly appearing, after a hundred years of use of the term *stopping power*.

the contrary, the description of the energy distributions of the emitted electrons (both backscattered and secondary) have to be performed avoiding the approximation of continuity in the energy loss processes and including *energy straggling* (ES) – i.e., the statistical fluctuations of the energy loss due to the different energy losses suffered by each electron travelling in the solid – in the calculations.

A detailed approach able to accurately describe low energy elastic and inelastic scattering and to appropriately take into account the energy straggling is required for the description of secondary electron cascade. The whole cascade of secondary electrons must be followed: indeed any truncation, or cut off, underestimates the secondary electron emission yield. Also, as already discussed, for insulating materials the main mechanisms of energy loss cannot be limited to the electron-electron interaction for, when the electron energy becomes very small (lower than 10–20 eV, say), inelastic interactions with other particles or quasi-particles are responsible for electron energy losses. In particular, at very low electron energy, trapping phenomena due to electron-polaron interactions (polaronic effects) and electron-phonon interactions are the main mechanisms of electron energy loss. For electron-phonon interaction, even phonon annihilations and the corresponding energy gains should be taken into account. Actually the energy gains are often neglected, for their probability of occurence is very small: much smaller, in any case, than the probability of phonon creation.

Summarizing, incident electrons are scattered and lose energy, due to the interactions with the atoms of the specimen, so that the incident electrons direction and kinetic energy are changed. It is usual to describe the collision events assuming that they belong to three distinct kinds: elastic (scattering with atomic nuclei), quasi-elastic (scattering with phonons) and inelastic (scattering with the atomic electrons and trapping due to the polaronic effect).

1.5 Electron Energy-Loss Peaks

Electron energy-loss spectroscopy treats the primary process in which the incident electron loses amounts of energy which characterize the target material (see, for example, Refs. [6, 11–30]). An electron spectrum represents the number of electrons as a function of the energy they have after interaction with a target. The spectrum can be represented as a function of either the electron energy or of the electron energy-loss. In this second case, the first peak on the left of the spectrum, centered at zero energy-loss, is known as the *zero-loss peak*. Also known as the *elastic peak*, it collects all the electrons which were transmitted – in transmission electron energy loss spectroscopy (TEELS) – or backscattered – in reflection electron energy loss spectroscopy (REELS) – without any measurable energy loss: it includes both the electrons which did not suffer any energy loss and those which were transmitted (TEELS) or backscattered (REELS) after one or more quasi-elastic collisions with

phonons (for which the energy transferred is so small that, with conventional spectrometers, it cannot be experimentally resolved). In TEELS, elastic peak includes also all the electrons which were not scattered at all, namely which were not deflected during their travel inside the target and did not lose energy.

Actually, the energy of electrons of the elastic peak is slightly reduced. This is due to the recoil energy transferred to the atoms of the specimen. Elastic peak electron spectroscopy (EPES) is the technique devoted to the analysis of the line-shape of the elastic peak [31, 32]. Since lighter elements show larger energy shifts, EPES can be used to detect hydrogen in polymers and hydrogenated carbon-based materials [33–40] measuring the energy difference between the position of the carbon elastic peak and that of the hydrogen elastic peak: this difference between the energy positions of the elastic peaks – for incident electron energy in the range 1000–2000 eV – is in the neighborhood of 2–4 eV.

In the first 30–40 eV from the elastic peak a generally quite broad peak collects all the electrons which suffered inelastic interaction with the outer-shell atomic electrons. Typically it includes electrons which suffered energy loss due to inelastic interaction with plasmons (*plasmon-losses*) and corresponding to inter-band and intra-band transitions. If the sample is sufficiently thick (in TEELS) and in the case of bulk targets (in REELS), the probability that an electron, before emerging from the specimen, has suffered more than one inelastic collision with plasmons is not negligible: such multiple electron-plasmon inelastic collisions are represented in the spectrum by the presence of a set of equidistant peaks (the distance from each other being given by the plasma energy). The relative intensities of these multiple inelastic scattering peaks decrease as the energy loss increases, demonstrating that the probability of suffering one inelastic collision is greater than the probability of suffering two inelastic collisions, which is in turn higher than the probability of suffering three inelastic collisions, and so on. Of course, in transmission EELS, the number of measurable plural scattering peaks is also a function of the thickness of the sample. Plural scattering peaks at multiples of the plasma energy are clearly observable – in the energy-loss region between the elastic peak and approximately 100–200 eV (i.e., in the energy spectrum, between 100–200 eV and the elastic peak) – when the film thickness is greater than the electron inelastic mean free path. On the other hand, when the film thickness is much smaller that the electron inelastic mean free path, a strong elastic peak and only the first plasmon-loss peak can be observed in the energy-loss region below 100–200 eV (i.e., above 100–200 eV from the elastic peak, in the energy spectrum).

For higher energy-losses, edges (of relatively low intensity with respect to the plasmon-losses), corresponding to inner-shell atomic electron excitations, can be observed in the spectrum. These edges are followed by slow falls, as the energy-loss increases. The energy position of these steps or, better, sharp rises, corresponds to the ionization threshold. The energy-loss of each edge is an approximate measure of the binding energy of the inner-shell energy level involved in the inelastic scattering process.

With an energy resolution better than 2 eV, it is possible to observe, in the low-loss peaks and in the ionization threshold edges, detailed features related to the band structure of the target and its crystalline characteristics. For example, in carbon, plasmon peaks can be found at different energies in the spectrum, according to the carbon structure. This is due to the different valence-electron densities of the different allotropic forms of carbon, such as diamond, graphite, C60-fullerite, glassy carbon, and amorphous carbon [26, 41].

For an excellent review about electron energy-loss spectra, see the Egerton book [26].

1.6 Auger Electron peaks

Also Auger electron peaks can be observed in the spectrum: they are due to the presence of doubly ionized atoms. Auger [42] and Meitner [43] noted the presence of pairs of tracks – originating from the same point – in X-ray irradiated cloud chambers filled with an inert gas. One of them had a variable length which depended on the energy of the incident radiation. The other track had a fixed length. Auger suggested the presence of doubly ionized atoms in the gas. Two years later, Wentzel made the hypothesis of a two-step process. A primary ionization, in the Wentzel interpretation, was followed by a decay process [44]. The incident radiation ionizes the system in the inner shell S. The system can then decay according to two alternative mechanisms. One is radiative: one electron drops out of an outer shell R into the inner shell S and a photon is emitted. The other one is non-radiative: one electron drops out of an outer shell R into the inner shell S, and the excess of energy is used to eject out of the shell R' another electron (the Auger electron). The two processes are competitive. In the electron spectrum, Auger electron peaks – due to the non-radiative process – can be recognized.

1.7 Secondary Electron Peak

Secondary electrons – produced by a cascade process – are those electrons extracted from the atoms by inelastic electron-electron collisions. Actually not all the secondary electrons generated in the solid emerge form the target. In order to emerge from the surface, the secondary electrons generated in the solid must reach the surface and satisfy given angular and energetic conditions. Of course, only the secondary electrons which are able to emerge from the target are included in the spectrum. Their energy distribution presents a pronounced peak in the region of the spectrum below 50 eV. The secondary electron emission yield is conventionally measured by integrating the area of the spectrum from 0 to 50 eV (including, in such a way, also the tail of backscattered electron whose number, in this energy region is actually negligible – unless the primary energy be very low as well).

1.8 Characterization of Materials

Simulation of transport of electrons in materials has been demonstrated to be very important for many applications. The determination of electron emission from solids irradiated by a particle beam is of crucial importance, especially in connection with the analytical techniques that utilize electrons to investigate chemical and compositional properties of solids in the near surface layers.

Electron spectroscopies and microscopies, examining how electrons interact with matter, represent fundamental tools to investigate electronic and optical properties of matter. Electron spectroscopies and microscopies allow to study the chemical composition, the electronic properties, and the crystalline structure of materials. According to the energy of the incident electrons, a broad range of spectroscopic techniques can be utilized: for example, low energy electron diffraction (LEED) allows to investigate the crystalline structure of surfaces, Auger electron spectroscopy (AES) permits to analyze the chemical composition of the surfaces of solids, electron energy loss spectroscopy – both in transmission, when the spectrometer is combined with transmission electron microscope, and in reflection – can be used to characterize materials by comparing the shape of the plasmon-loss peaks and the fine-structure features due to interband and intraband transitions with those of suitable standards, elastic peak electron spectroscopy is an useful tool to detect the presence of hydrogen in carbon-based materials.

The study of the properties of a material using electron probes requires the knowledge of the physical processes corresponding to the interaction of the electrons with the particular material under investigation. A typical AES peak of an atomic spectrum, for example, has a width in the range from 0.1 to 1.0 eV. In a solid, many energy levels are involved which are very close in energy, so that broad peaks are typically observed in AES spectra of solids. Their features also depend on the instrumental resolution. Another important characteristic of the spectra is related to the shift in energy of the peaks due to chemical environment: indeed the core energy levels of an atom are shifted when it is a part of a solid. This property is used to characterize materials, as the shift can be determined theoretically or by comparison with suitable standards. Even the changes in spectral intensities and the appearence of secondary peaks can be used for analyzing unknown materials. Electron spectra are used for self-supported thin film local thickness measurements, multilayer surface thin film thickness evaluation, doping dose determination in semiconductors, radiation damage investigations, and so on.

The backscattering electron coefficient can be used for non-destructive evaluation of over-layer film thickness [45, 46], while the study of the energy distribution of the backscattered electrons may be utilized for materials characterization through the study of the shape of plasmon-loss peaks [47, 48].

Secondary electron investigation allows extraction of critical dimensions by modeling the physics of secondary electron image formation [49–51]. It permits to investigate doping contrast in *p-n* junctions and to evaluate accurate nanometrology for the most advanced CMOS processes [52, 53].

1.9 Summary

Transport Monte Carlo simulation is a very useful mathematical tool for describing many important processes relative to the interaction of electron beams with solid targets. In particular, the backscattered and secondary electron emission from solid materials can be investigated with the use of the Monte Carlo method.

Many applications of the Monte Carlo study of backscattered and secondary electrons concern materials analysis and characterization. Among the many applications of MC simulations to analysis and characterization, we can mention non-destructive evaluation of over-layer film thickness [45, 46], materials characterization through the study of the main features of the electron spectra and the shape of plasmon-loss peaks [47], extraction of critical dimensions by modeling the physics of secondary electron image formation [49–51], and doping contrast in *p-n* junctions for the evaluation of accurate nanometrology for the most advanced CMOS processes [52, 53].

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