Monte Carlo Simulation of Secondary Electron Emission from Dielectric Targets

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Abstract. In modern physics we are interested in systems with many degrees of freedom. The Monte Carlo (MC) method gives us a very accurate way to calculate definite integrals of high dimension: it evaluates the integrand at a random sampling of abscissa. MC is also used for evaluating the many physical quantities necessary to the study of the interactions of particle-beams with solid targets. Letting the particles carry out an artificial random walk and taking into account the effect of the single collisions, it is possible to accurately evaluate the diffusion process.

Secondary electron emission is a process where primary incident electrons impinging on a surface induce the emission of secondary electrons. The number of secondary electrons emitted divided by the number of the incident electrons is the so-called secondary electron emission yield. The secondary electron emission yield is conventionally measured as the integral of the secondary electron energy distribution in the emitted electron energy range from 0 to 50eV.

The problem of the determination of secondary electron emission from solids irradiated by a particle beam is of crucial importance, especially in connection with the analytical techniques that utilize secondary electrons to investigate chemical and compositional properties of solids in the near surface layers. Secondary electrons are used for imaging in scanning electron microscopes, with applications ranging from secondary electron doping contrast in p-n junctions, line-width measurement in critical-dimension scanning electron microscopy, to the study of biological samples.

In this work, the main mechanisms of scattering and energy loss of electrons scattered in dielectric materials are briefly treated. The present MC scheme takes into account all the single energy losses suffered by each electron in the secondary electron cascade, and is rather accurate for the calculation of the secondary electron yield and energy distribution as well.

1. Introduction

Many features appear in the secondary electron spectra, due to the different collision processes involved prior to low-energy secondary electron emission. When a stream of electrons impinges on a solid target, the incident particles (during their travel within the solid) lose energy and change direction at each collision with the atoms bound in the solid. Nuclear collisions deflect the electrons without relevant kinetic-energy transfer, owing to the large difference between the masses of the electron and the nucleus: the differential elastic scattering cross-section has to be used in order to describe these processes (Mott cross section). On the other hand, excitations and ejections of atomic electrons, and plasmon, phonon, and polaron excitations affect the energy dissipation but only slightly affect the direction of the electron in the solid. These can be described as inelastic processes, governed by the equations for the differential inelastic scattering cross-sections.

Transport MC is a very well-known numerical method which allows to predict the behavior of charged particles traveling through the matter. In particular, it has been widely utilized for the study of the electron-beams interacting with solid targets. This topic has many applications. Among them, on the side of the chemical analysis of the surfaces and the interfaces of materials, let’s mention Scanning Electron Microscopy, Transmission Electron Microscopy, Reflection Electron Energy Loss
Spectroscopy, Auger Electron Spectroscopy, X-Ray Photoelectron Spectroscopy. There are many other applications relative to the study of materials. If we limit ourselves to the secondary electron emission, MC simulation can be used for the study of image contrast in silicon p-n junctions, for line scan calculations of resist materials with given geometrical cross sections deposited on silicon substrates, for electric fields induced in insulating materials by electron beam irradiation, and so on.

2. Monte Carlo Modeling
While for electron kinetic energies higher than 10keV, MC simulations provide excellent results by the simple use of the Rutherford differential elastic scattering cross section and of the Bethe-Bloch stopping power formula (or semi-empirical stopping power formulas), when the electron energies of interest are much lower than 5keV – and this is the case of secondary electron emission – this approach can no longer be utilized\textsuperscript{1}. There are many reasons, and the most important ones are related to the three following facts: (i) The Rutherford formula is a high energy approximation, being the result of the first Born approximation. (ii) The Bethe-Bloch formula is valid only for quite high energies as well; in particular, the Bethe-Bloch stopping power does not work well when the electron energy $E$ becomes lower than the mean ionization energy $J$. It reaches a maximum for $E = J/1.166$. Below this energy value, the predicted stopping power becomes negative. Thus 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 use of the stopping power, and hence of the continuous-slowing-down approximation, for describing energy loss, 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 modeling of the electron trajectories should avoid the approximation of continuity in describing the electron energy losses.

A detailed approach able to accurately describe low energy elastic and inelastic scattering and to appropriately take into account the energy straggling (i.e. the different energy losses suffered by each electron of the penetrating beam) is required for the description of secondary electron cascade. The whole cascade must be followed: indeed any truncation, or cut off, underestimates the secondary electron emission yield. What is more, as we will discuss below, for insulating materials the main mechanisms of energy loss cannot be limited to the electron-electron interaction for, when the electron energy becomes very low (let’s say lower than 10-20eV), inelastic interactions with other particles or quasi-particles are responsible for electron energy losses. In particular, at very low electron energy, electron-phonon interactions and electron-polaron interactions (polaronic effects) become the main mechanisms for the electron energy loss.

3. Elastic and Inelastic Scattering
The results presented in this paper were obtained with the differential and total elastic scattering cross sections calculated using the Mott theory, i.e. obtained numerically solving the Dirac equation in a central field: this procedure is known as the “relativistic partial wave expansion method” and it has been demonstrated to provide excellent results when compared to experimental data\textsuperscript{2-9}.

The main mechanism which determines the inelastic scattering cross section and the relative energy losses, for energies higher than 20eV, is the interaction of the incident electrons with the collective excitations of the electron sea, known as plasmons. Such energy loss mechanisms can be described by calculating the so-called energy loss function, i.e. the reciprocal of the imaginary part of the dielectric function. The Ritchie theory\textsuperscript{10-20} was used--starting from the knowledge of the dependence of the dielectric function upon both the energy loss and the momentum transfer--to calculate the differential inverse electron inelastic mean free path and the electron inelastic mean free path. When the electron energy is higher than 50eV, both the electron inelastic mean free path and the electron stopping power calculated within the dielectric formalism are in very good agreement with the experiment (and with theoretical data obtained by other investigators). For details of the present calculation of the electron-electron inelastic mean free path $\lambda_{ee}$ in SiO$_2$, see Refs.\textsuperscript{16,17}.\textsuperscript{18}
When, on the other hand, the electron energy becomes lower than 50eV, the dielectric formalism alone is no longer able to accurately describe the energy loss phenomena. In fact, as the electron energy decreases, the electron inelastic mean free path calculated using only the electron-electron interaction increases indefinitely (while the stopping power goes quickly to zero). This means that if only electron-electron interactions were active for inelastic scattering, electrons with such a low energy would no longer interact inelastically (i.e. losing energy) with the solid (we are neglecting here, for the sake of simplicity, the little energy losses associated to the electron-atom elastic scattering collisions, which also occur for they actually are quasi-elastic). As a consequence they would travel without any change in their kinetic energy. For a semi-infinite target, this very long travel in the solid would continue forever or until the electron reaches the surface of the material and is able to emerge. Also note that the condition for emerging from the surface is not always satisfied. The interface with the vacuum presents, in fact, a potential barrier, and not all the electrons that reach the surface are able to go beyond it. When the electrons reaching the surface are not able to emerge, they are reflected back in the material.

In general, in order to be able to emerge from the surface and to be detected, an electron must have a residual energy $E$ satisfying the relationship

$$E \cos^2 \theta \geq \chi,$$  \hspace{1cm} \text{(1)}

where $\theta$ is the angle of the electron direction with respect to the normal to the surface and $\chi$ is the electron affinity, namely the potential barrier between the vacuum level and the minimum of the conduction band$^{21,22}$. Experimental and MC results relative to the slow electrons (secondary electron energy is lower than 50eV) depend on the value of the electron affinity: for SiO$_2$, $\chi = 0.9$eV.

Thus, if the only mechanism of energy loss was the electron-electron interaction, as the energy of the electrons become lower than approximately 10-20eV, the particles would no longer suffer energy losses, the inelastic mean free path would tend to infinite (and the stopping power would approach zero) so that many electrons could virtually walk in the solid without end. A MC simulation, in such a case, would require so long CPU times to converge as to be, practically, without any utility.

Luckily nature is much cleverer than MC code programmers. Indeed, when the electron energy is so low, electrons lose small and numerous amounts of energies interacting with lattice vibrations, or phonons. In fact, another important mechanism of electron energy loss for electrons traveling in solid targets, particularly effective when the electron energy is low (few eV), is represented by the interaction of the electrons with the optical modes of the lattice vibrations. Notice that this kind of interactions are responsible for electron energy gain as well. In his theory of the electron-phonon interaction, Fröhlich$^{23}$ considered, in particular, the interaction of free conduction electrons with the longitudinal optical mode lattice vibrations. The interaction was treated considering both phonon creation and phonon annihilation, corresponding to electron energy loss and to electrons energy gain, respectively. As the phonon generation probability is much higher than the phonon absorption probability, the last is often neglected in MC simulations$^{24}$. Furthermore, since, according to Ganachaud and Mokrani$^{25}$, the dispersion relation of the longitudinal phonons can be neglected in the optical branch, one can use a single phonon frequency $\omega$. For the present case we put $\hbar \omega = 0.1$eV. According to Fröhlich$^{23}$, Llacer and Garwin$^{24}$, and Ganachaud and Mokrani$^{25}$, the inverse inelastic mean free path for the electron-phonon interaction, $\lambda^{-1}_{\text{eph}}$, can be calculated by

$$\lambda^{-1}_{\text{eph}} = \frac{1}{a_0} \frac{n(T)+1}{2} \left[ \frac{\epsilon(0)-\epsilon(\omega)}{\epsilon(0)\epsilon(\omega)} \right] \frac{\hbar \omega}{E} \ln \frac{1}{\left( 1 - \frac{1}{E} \right)^{1/2}} \text{,}$$  \hspace{1cm} \text{(2)}

where

$$n(T) = \frac{1}{e^{\hbar \omega/KE} - 1} \text{.}$$  \hspace{1cm} \text{(3)}
Here $a_0$ is the Bohr radius, $k$ the Boltzmann constant, $\varepsilon(0)$ the static dielectric constant and $\varepsilon(\infty)$ the high frequency dielectric constant respectively. For SiO$_2$, $\varepsilon(\infty) = 2.13$ and $\varepsilon(0) = 3.9$.

Electron-phonon interaction is not the only mechanism of energy loss for very slow electrons interacting with insulating materials. Indeed a low energy electron moving in an insulating material induces around it a polarization field with a stabilizing (trapping) effect on the electron itself. This is known as the polaronic effect, as the quasi-particle constituted by the slow electron with the polarization field is known as a polaron. According to Ganachaud and Mokrani$^25$, the inverse inelastic mean free path for electron-polaron interaction, $\lambda_{ep}^{-1}$, is given by

$$\lambda_{ep}^{-1}(E) = C e^{-\gamma E},$$  \hspace{1cm} (4)

where, for the present calculations relative to SiO$_2$, we put $C = 1\text{nm}^{-1}$, $\gamma = 0.085\text{eV}^{-1}$.

In Fig. 1 the effects the inelastic electron mean free paths in SiO$_2$ corresponding to the various mechanisms of energy loss (electron-electron, $\lambda_{ee}$, electron-phonon, $\lambda_{eph}$, electron-polaron, $\lambda_{ep}$) are presented as long as the inelastic mean free path $\lambda$ which takes into account all these inelastic scattering mechanisms and is given by

$$\lambda^{-1} = \lambda_{ee}^{-1} + \lambda_{eph}^{-1} + \lambda_{ep}^{-1}.$$ \hspace{1cm} (5)

It is clear from the figure that, due to the inclusion of the electron-phonon and of the electron-polaron interactions in the calculation, the electron inelastic mean free path $\lambda$ approaches zero as the electron energy goes to zero.

4. Results and Discussion

The MC results we are going to present were obtained with a detailed modeling which takes into account all the described mechanisms of energy loss (electron-electron, electron-phonon, electron-polaron) and treats the elastic scattering events using the Mott cross section. Furthermore, the entire cascade of secondary electrons is followed. For a description of the present detailed MC approach, see Refs. 25-28.

The whole MC spectrum representing the energy distribution of electrons emerging from a SiO$_2$ sample due to a 250eV electron beam irradiation is presented in Fig. 2. Similar spectra have been observed for metals as well$^{29,30}$.

For the present calculation the target is considered as semi-infinite. The case of thin films requires special attention, in particular when the samples are thinner than the mean free path. The study of the secondary electron emission from thin films will be the subject of further investigations.

Many electrons of the primary electron beam can be backscattered, after having interacted with the atoms and electrons of the target. A fraction of them conserves the original kinetic energy, having suffered only elastic scattering collisions with the atoms of the target: these electrons constitute the so-called elastic peak, or zero loss peak, whose maximum is located at the energy of the primary beam. The plasmon peak in Fig. 2 represents the electrons of the primary electron beam that emerge from the surface after having suffered a single inelastic collision with a plasmon. Multiple collisions with plasmons are also present in the spectrum, but they are of very low intensity so that they are not visible on this scale. Electron-phonon energy losses are also not visible in Fig. 2, for (i) their intensity is much lower than that of the elastic peak and (ii) they are very close to the much more intense elastic peak whose width is of the order of 1eV.

On the other hand the secondary electrons produced by a cascade process are those electrons which have been extracted from the atoms by inelastic electron-electron collisions and are able to emerge from the target surface. Their energy distribution presents a pronounced peak in the very low energy region of the spectrum, typically below 50eV. The secondary electron emission yield is conventionally
measured integrating the area of the spectrum from 0 to 50eV (including, in such a way, also the tail of backscattered electron whose number, on the other hand, in this energy region is negligible – unless the primary energy be very low as well).

The comparison of the MC simulated secondary electron yield from SiO$_2$ with the Dionne experimental data$^{31}$ was shown in Fig. 3, to validate the code (in this case we used three values of $\gamma$: 0.080eV$^{-1}$, 0.085eV$^{-1}$, and 0.090eV$^{-1}$).

As the primary energy increases, the yield increases until a maximum is reached. Then the yield decreases by increasing the primary energy. Indeed, at very low primary energy, few secondary electrons are generated, and increasing the primary energy the number of secondary electrons emerging from the surface increases. Furthermore, at very low primary energy, the average depth at which the secondary electrons escaping from the surface are generated also increases as the primary energy increases. When the energy becomes higher than a threshold which depends on the target (the turning point for SiO$_2$ being approximately at 250eV, according to the mean value between experiment and simulation), the average depth of generation of the secondary electrons becomes so deep that just a small amount of the generated secondary electrons are able to reach the surface satisfying the condition, represented by Eq. (1), necessary to emerge from the sample and to be detected.

The agreement between MC results and experimental data is quite satisfactory. The MC model well predicts, in fact, the shape of the experimental secondary electron yield curve of SiO$_2$ as a function of the electron primary energy. In particular we observe that the dashed line looks similar in shape, but shifted in energy. In order to compare the shapes of the curves, the Monte Carlo secondary electron yields corresponding to $\gamma$=0.085eV$^{-1}$ were horizontally translated (shift in energy was -50 eV). With this shift in energy, the experimental and Monte Carlo data yield a very close match (see Fig. 4).

Adjusting other parameters does not yield a closer match. We conclude that the differences between experimental and Monte Carlo data corresponding the original calculation (i.e. without any energy translation) could be reasonably attributed to the poor approximation adopted to describe the polaronic effect.

Eq. (4), proposed by Ganachaud and Mokrani$^{25}$, is based on the principle that the lower the electron energy, the higher the probability for the electron to lose its energy in the generation of a polaron. It is assumed that, once the polaron has been generated, the residual kinetic energy of the electron could be neglected. Therefore, the electron is considered to stay trapped at the interaction site. Of course, this represents a quite rough approximation. In fact, as recognized by Ganachaud and Mokrani$^{25}$, trapped electrons can actually hop from one trapping site to the next, due to phonon-induced processes, which cannot be adequately taken into account with the present oversimplified model represented by Eq.(4). Further investigations are thus required to improve the code in order to better understand and describe the polaronic effect, which represents one of the more relevant phenomena in determining the yield of secondary electron emission from insulators.

5. Conclusion
The main mechanisms of scattering and energy loss of electrons scattered in dielectric materials have been briefly treated within the Ganachaud and Mokrani heuristic model$^{25}$. A MC scheme which takes into account all the energy losses suffered by each electron in the secondary electron cascade is used for calculating the secondary electron energy distribution and secondary electron emission yield from SiO$_2$. Satisfactory agreement has been found between the shapes of the curves corresponding to the MC simulation and the Dionne experimental data$^{31}$.

References
[1] Dapor M 1992 Phys. Rev. B 46 618

[2] Mott NF 1929 Proc. R. Soc. London Ser. 124 425
[3] Mott NF, Massey HSW 1965 *The Theory of Atomic Collisions*, Oxford University Press, Oxford

[4] Lin S-R, Sherman N, Percus JK 1963 *Nucl. Phys.* 45 492

[5] Bunyan PJ, Shonfelder JL 1965 *Proc. Phys. Soc.* 85 455

[6] Salvat F, Mayol R 1993 *Comput. Phys. Commun.* 74 358

[7] Dapor M 1996 *J. Appl. Phys.* 79 8406

[8] Dapor M 2003 *Electron-Beam Interactions with Solids: Applications of the Monte Carlo Method to Electron Scattering Problems*, Springer Tracts in Modern Physics 186, Springer, Berlin

[9] Jablonski A, Salvat F, Powell CJ 2004 *J. Phys. Chem. Ref. Data* 33 409

[10] Ritchie RH 1957 *Phys. Rev.* 106 874

[11] Ritchie RH, Howie A 1977 *Phil. Mag.* 36 463

[12] Penn DR 1987 *Phys. Rev. B* 35 482

[13] Ashley JC 1988 *J. Electrons Spectrosc. Relat. Phenom.* 46 199

[14] Yubero F, Tougaard S 1992 *Phys. Rev. B* 46 2486

[15] Cohen-Simonsen A, Yubero F, Tougaard S 1997 *Phys. Rev. B* 56 1612

[16] Dapor M 2006 *J. Electrons Spectrosc. Relat. Phenom.* 151 182

[17] Dapor M 2006 *Surf. Sci.* 600 4728

[18] Filippi M, Calliari L, Dapor M 2007 *Phys. Rev. B* 75 125406

[19] Taioli S, Simonucci S, Calliari L, Filippi M, Dapor M 2009 *Phys. Rev. B* 79 085432

[20] Taioli S, Simonucci S, Calliari L, Dapor M 2010 *Phys. Rep.* 493 237

[21] Dapor M, Inkson BJ, Rodenburg C, Rodenburg JM 2008 *EPL* 82 30006; 82 49901

[22] Dapor M 2009 *Nucl. Instr. Meth. B* 267 3055

[23] Fröhlich H 1954 *Adv. Phys.* 3 325

[24] Llacer J, Garwin EL 1969 *J. Appl. Phys.* 40 2766

[25] Ganachaud JP, Mokrani A 1995 *Surf. Sci.* 334 329

[26] Dapor M, Ciappa M, Fichtner W 2010 *J. Micro/Nanolith. MEMS MOEMS* 9 023001

[27] Koschik A, Ciappa M, Holzer S, Dapor M, Fichtner W 2010 *Proc. of SPIE* 7729 77290X
[28] Dapor M 2011 *Nucl. Instr. Meth. B* **269** 1668

[29] Cimino R, Collins IR, Furman MA, Pivi M, Ruggiero F, Rumolo G, Zimmermann F 2004 *Phys. Rev. Lett.* **93** 014801

[30] Furman MA, Chaplin VH 2006 *Phys. Rev. Spec. Top. – Accel. Beams* **9** 034403

[31] Dionne GF 1975 *J. Appl. Phys.* **46** 3347
Figure 1. Electron inelastic mean free paths in SiO$_2$ corresponding to the various mechanisms of energy loss. Electron-electron inelastic mean free path, $\lambda_{ee}$, is represented by the solid line. Electron-phonon inelastic mean free path, $\lambda_{eph}$, is represented by the dashed line. Electron-polaron inelastic mean free path, $\lambda_{ep}$, is represented by the dotted line. The electron inelastic mean free path $\lambda$ is given by $\lambda^{-1} = \lambda_{ee}^{-1} + \lambda_{eph}^{-1} + \lambda_{ep}^{-1}$ and is represented by the bold solid line. It approaches zero as the electron energy goes to zero. $\hbar\omega=0.1\text{eV}$, $C=1\text{nm}^{-1}$, $\gamma=0.085\text{eV}^{-1}$. 
Figure 2. MC spectrum representing the energy distribution of electrons emerging from a SiO$_2$ sample due to a 250eV electron beam irradiation. The elastic peak, or zero loss peak, whose maximum is located at the energy of the primary beam, represents the electrons which suffered only elastic scattering collisions. The plasmon peak represents the electrons of the primary electron beam that emerge from the surface after having suffered a single inelastic collision with a plasmon. Multiple collisions with plasmons are also present in the spectrum, but they are of very low intensity so that they are not visible on this scale. The secondary electrons energy distribution presents a pronounced peak in the very low energy region of the spectrum, typically below 50eV. $\chi=0.9\text{eV}$, $\hbar\omega=0.1\text{eV}$, $C=1\text{nm}^{-1}$, $\gamma=0.085\text{eV}^{-1}$. 

\[ \text{Intensity (a.u.)} \]

\[ \text{Energy (eV)} \]
Figure 3. Secondary electron yield of SiO$_2$. Comparison of the Dionne experimental data$^{31}$ (solid line) with the MC simulated data (dotted line: $\gamma=0.080\text{eV}^{-1}$; dashed line: $\gamma=0.085\text{eV}^{-1}$, dotted-dashed line: $\gamma=0.090\text{eV}^{-1}$). $\chi=0.9\text{eV}$, $h\omega=0.1\text{eV}$, $C=1\text{nm}^{-1}$.
**Figure 4.** Secondary electron yield of SiO$_2$. Comparison of the Dionne experimental data$^{31}$ (solid line) with the MC simulated data (bullets). $\gamma=0.085\text{eV}^{-1}$, $\chi=0.9\text{eV}$, $\hbar\omega=0.1\text{eV}$, $C=1\text{nm}^{-1}$. The Monte Carlo data were horizontally translated (shift in energy was -50 eV) in order to compare the shapes of the curves.