

# An extensive theoretical quantification of secondary electron emission from silicon

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## ABSTRACT

Though intensive experimental studies have been carried out on electron emission properties in past decades, the reliable data from accurate experimental measurements for clean and smooth surfaces are still quite limited. In this work, we have performed a comprehensive Monte Carlo simulation of electron emission yields, the secondary electron yield (SEY,  $\delta$ ), backscattered electron coefficient (BSC,  $\eta$ ) and total electron yield (TEY,  $\sigma = \delta + \eta$ ), from silicon and the uncertainty quantification of the theoretical calculation results for the first time. The simulations were made on total 17280 scattering models. The considered uncertainty factors in the physical modelling that can influence the calculated yields include work function data, optical energy loss function data, dielectric function models for electron inelastic scattering and the scattering potentials for electron elastic scattering. The calculation results show that  $\delta$  is significantly influenced by the work function while the dielectric function model has a slight effect. Elastic scattering potential (or elastic scattering cross section) and energy loss function affects to a certain extent both  $\delta$  and  $\eta$ . Our simulated  $\eta$  data agree with most of the experimental measurements while the simulated mean  $\delta$  data are well below most of the experimental data. At a 75% level of confidence, the experimental  $\sigma$  data measured by Goto et al. lie within the uncertainty range of our simulation results. This work has provided sufficient ground for the necessity of building a theoretical database of electron emission yield considering the fact that most of the previous experimental data are not reliable due to the presence of surface contamination during the measurements.

## 1. Introduction

The study of electron transport in condensed matter is of great interest to researchers in many applied fields because different kinds of signals produced in the interaction process are employed for the characterization of the material with electron beam techniques [1–6], including electron probe microanalysis, scanning electron microscopy, transmission electron microscopy, reflection high energy electron diffraction, scanning transmission electron microscopy, Auger electron spectroscopy, x-ray photoelectron spectroscopy, electron energy loss spectroscopy and reflection electron energy loss spectroscopy, etc. The

generated electron or photon signals carry rich information, like the elemental composition, structure, and electronic properties, about the material. Therefore, a sound knowledge of electron interaction in materials is crucial for both fundamental research and practical application, and the modelling of electron interaction with matter remains a topic of active development.

One generally distinguishes an electron emitted from the surface as a secondary electron or backscattered electron by an energy threshold of 50 eV. The low energy electrons (<50 eV) are considered as secondary electrons that are excited from the valence- or core-shells of the material by the incident electrons. While high energy electrons (>50 eV) are

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defined as backscattered electrons that are due to those incident electrons having lost certain energies during inelastic scattering collisions and backscattered from the surface. The study of secondary electron emission from matter under the bombardment of an electron beam has a long history from the 1950s [7–13]. The electron emission yields of solids are essential parameters for application devices like photo-multipliers, plasma thrusters [14], Hall effect engines [14], satellite communication devices [15], etc. and in various fundamental research fields of physics, chemistry, and engineering, including catalysis [16, 17], surface science, chemical analysis [18]. It is thus important to carry out both theoretical and experimental studies of electron emission yields, i.e. the secondary electron yield and backscattering coefficient. But the serious problem is that the available experimental databases [19, 20] for the secondary electron yield (SEY,  $\delta$ ), backscattered electron coefficient (BSC,  $\eta$ ) and total electron yield (TEY,  $\sigma = \delta + \eta$ ) reveal quite large inconsistency, frequently over 100% for SEY, among the historical data measured by different researchers, probably mainly due to factors regarding to sample preparation method and vacuum condition [13,20,21]. The surface state of the sample particularly plays a significant role in the measured SEY due to the low energy secondary electron emission being easily modulated through the change of work function. Meanwhile, the surface contaminants may also affect the low energy electron backscattering from the surface. The old measurements done in a poor vacuum condition were actually representing the property of contaminated surfaces with contaminants like hydrocarbon molecules. The modern ion sputtering technique cannot completely remove oxidation for some materials but often induces surface roughness which may significantly alter the SEY. Therefore, experimental databases cannot provide reliable and accurate SEY and BSC values so far, and there are no experimental benchmark data established.

On the other hand, tremendous theoretical efforts have also been put into the modelling of electron emission and derivation of electron yields. Particularly, Monte Carlo simulation methods and physical models have been developed and extensively applied to the simulation of secondary electrons and backscattered electrons [22–35]. Early semi-empirical Monte Carlo physical models of electron scattering [25] often need some parameters which are to be determined from the fitting of the calculation results to the experimental SEY data. It is clear that such kind of theoretical modelling can only suggest a general shape of the yield curve but cannot provide the absolute calculation yield values from the first-principles. Since the introduction of the full momentum- and frequency-dependent dielectric function,  $\epsilon(q, \omega)$ , which is practically derived from the experimental measured optical dielectric function data  $\epsilon(q=0, \omega)$  by the theoretical approach of Penn [36], the dielectric functional approach has enabled the Monte Carlo physical model [37] to be the first-principles like, i.e. the inelastic scattering cross section is calculated based on a full physical background and is free of the artificial fitting parameters. The up-to-date Monte Carlo simulations [37–65] are almost based on such a dielectric functional approach to electron inelastic scattering, especially for metals and other elemental solids. This would shed light on the reliable modelling of electron emission yields and for establishing a theoretical database of SEY and BSC in absolute values for ideal clean surfaces. However, as stated above, the available experimental database of electron yields hardly provides a solid foundation for validation of a particular theoretical model due to large scatter involved in the experimental data. Even though a study has indicated such a possibility for validation by carefully selecting possibly more reliable experimental datasets [46], while the tendency to choose a particular dataset in comparison without reasonable justification is certainly inadvisable. In addition, there is still a problem with the determination of the scattering cross section model, which is particularly useful for low energy electrons. People would wonder how much we can trust the present theoretical modelling and Monte Carlo simulation data.

To answer this question, we have to consider uncertainty quantification (UQ) for the theoretical simulation results. UQ is a process of

identifying, characterizing, and quantifying those factors that can affect simulation output. The present different modelling areas, e.g. political sciences [66], climate sciences [67], financial processes [68], civil engineering [69], and computational biology [70], are already at various levels of implementation of UQ standard. Recently the estimation of the uncertainty in theoretical modelling is becoming an important issue [71]. Like experimental data, simulation results also need to be judged to establish the simulator as a reliable productive instrument, and in future, the modelling data would be considered as incomplete without assessing the associated uncertainties. However, as far as to our knowledge, in the field of Monte Carlo simulation of electron interaction with solids, only limited work has been done on the sensitivity analysis but none on the UQ. They are sensitivity analysis of the effects of, for example, inelastic scattering cross section and surface potential on linewidth measurements [72], the elastic and inelastic scattering cross sections on line edge roughness simulations [73], and the scattering cross sections on SEY [74]. The reason for the limited attention from the researchers to the UQ of Monte Carlo simulation of electron emission is quite obvious: the high computation cost prevents from the simulations associated with the scattered input data and varied theoretical models for the UQ. In a previous work, a simple uncertainty analysis was done on BSC where the uncertainty solely due to the elastic cross section was considered [62]. Therefore, in this work we aim to build a theoretical foundation for calculating simulation uncertainties related to SEY and BSC by performing a comprehensive Monte Carlo simulation, leading to a detailed UQ. The analysis could thus be helpful for predicting reliability of Monte Carlo simulations of electron emission.

In the present status of Monte Carlo modelling and simulation, one faces a few questions: “which is the most reasonable scattering cross section?”, “are we using the correct scattering physical model?”, “what are the correct optical properties for input?”. These factors are considered as the main sources of the present theoretical uncertainties. For electron scattering calculations, the full uncertainty  $u_{\text{simulation}}$  may be given as [62,71],

$$u_{\text{simulation}}^2 = u_{\text{modelling}}^2 + u_{\text{numerical}}^2 + u_{\text{specimen}}^2 \quad (1)$$

where the three uncertainty components are due respectively to the physical modelling, numerical calculation and the specimen parameters of the sample geometrical structure. As we explained elsewhere [62] that the uncertainty term due to numerical calculation,  $u_{\text{numerical}}$ , is negligible in our present calculation with the CTMC-SEM code. The term,  $u_{\text{specimen}}$ , due to specimen geometrical structure is vanishing here because we deal with the structureless sample, or more specifically the plane surface of a semi-infinite bulk. The uncertainty term due to the physical modelling,  $u_{\text{modelling}}$ , is hence the only important term which is contributed by the uncertainties involved in the physical modelling of electron elastic scattering and inelastic scattering. The Mott’s cross section and dielectric functional theory are generally accepted as the most accurate physical models for the description of electron elastic scattering and inelastic scattering, respectively. However, the inputs to the calculations still contain certain uncertainties: for elastic scattering there are a variety of forms for electron-atom interaction potential, and for electron inelastic scattering the dielectric function has different models and expressions and the input data of experimental optical constants [75–77] may also differ in the database. In addition, the uncertainties of the parameters used for describing the physical properties of the specimen, such as mass density, work function etc., also affect the calculated scattering cross sections.

In this study, we have considered an extensive range of theoretical uncertainties related to the physical modelling and the required experimental input data. Our analysis encompasses factors such as nuclear density, electron distribution, exchange potential, correlation-polarization effects, atomic vs. muffin-tin approximation, inelastic scattering model, work function, and energy loss function data. We acknowledge the inherent challenges in accounting for potential

systematic errors that are suspected but unmeasurable. We have made reasonable efforts to address the known uncertainties within the scope of our study while acknowledging the limitations of theoretical modelling.

The material considered here for our current UQ of electron emission is crystalline Si for the following reasons: Si has a wide range of practical applications; Si has different work functions and optical property data. In addition, the experimentally measured SEY data vary as large as 100% and the theoretical evaluation is thus very important. Furthermore, for a semiconductor its modelling of electronic excitation would be different from that of a metal. We have considered a range of work function with three different datasets of energy loss functions (ELFs) obtained from various sources for pure and doped crystal Si. As regards to the uncertainties related to the modelling for the determination of particle interactions, we have considered totally 384 Mott's elastic cross section datasets calculated from the respective different scattering potential models and three inelastic cross section datasets derived from the three dielectric function expressions, where a new model of dielectric function [78] proposed for semiconductor/insulator has been included.

## 2. Theoretical modelling

An up-to-date Monte Carlo simulation code (Classical Trajectory Monte Carlo simulation of Scanning Electron Microscopy, CTMC-SEM) [62] is adopted in this calculation by tracing trajectories of primary electrons and excited secondary electrons inside the bulk target with a random sampling of electron scattering events, including elastic and inelastic scattering. Below we will present in detail, the involved physical models and the related modelling uncertainties.

### 2.1. Elastic scattering models

Mott's differential cross section [79] derived from the solution of Dirac's equation is considered to be the most accurate model to describe the elastic scattering of electrons with atoms:

$$\frac{d\sigma_e}{d\Omega} = |f(\theta)|^2 + |g(\theta)|^2 \quad (2)$$

where  $f(\theta)$  and  $g(\theta)$  are the scattering amplitudes and can be calculated with the partial wave expansion method,

$$f(\theta) = \frac{1}{2iK} \sum_{l=0}^{\infty} \{ (\ell+1)(e^{2i\delta_l^+} - 1) + \ell(e^{2i\delta_l^-} - 1) \} P_\ell(\cos \theta) \quad (3)$$

$$g(\theta) = \frac{1}{2iK} \sum_{l=1}^{\infty} \{ -e^{2i\delta_l^+} + e^{2i\delta_l^-} \} P_\ell^1(\cos \theta) \quad (4)$$

where  $\hbar K$  is the momentum of the electron,  $\delta_l^+$  and  $\delta_l^-$  are spin up and spin down phase shifts of the  $\ell$  th partial wave, respectively;  $P_\ell(\cos \theta)$  and  $P_\ell^1(\cos \theta)$  are the Legendre and the first order associated Legendre functions, respectively. The phase shifts are calculated a radial equation of electron motion in an atomic potential field  $V(r)$ . The effective interaction potential for a projectile electron at the distance  $r$  from the atomic nucleus is described by the means of an optical potential model,

$$V(r) = V_{st}(r) + V_{ex}(r) + V_{cp}(r) - iW_{abs}(r) \quad (5)$$

where  $V_{st}(r)$  is the electrostatic potential,  $V_{ex}(r)$  is the exchange potential,  $V_{cp}(r)$  is the correlation-polarization potential (necessary only for projectiles at low energies below 10 keV), and  $W_{abs}(r)$  is the magnitude of imaginary absorption potential, which is taken as zero in this work. There are various different potential models and the accurate one is hardly known. Thus, the alternative forms of potential play as the source of uncertainty in the calculation of Mott's cross section. For the numerical calculation of differential and total cross sections, we have employed Salvat's latest version of ELSEPA [80], which is an extension of the previous version [81] with additional functionality while

maintaining a detailed description of the underlying calculations. Totally 384 Mott's cross sections datasets were obtained by combinations of various alternative models.

The electrostatic interaction potential  $V_{st}(r)$  is expressed as,

$$V_{st}(r) = -e\varphi(r) = -e[\varphi_n(r) + \varphi_e(r)] \quad (6)$$

where  $\varphi_n(r)$  and  $\varphi_e(r)$  are respectively, the electrostatic potentials contributed by the protons in the nucleus and orbital electrons of the target atom. They can be calculated by

$$\varphi_n(r) = e \left( \frac{1}{r} \int_0^r \rho_n(r') 4\pi r'^2 dr' + \int_r^\infty \rho_n(r') 4\pi r' dr' \right) \quad (7)$$

$$\varphi_e(r) = e \left( \frac{1}{r} \int_0^r \rho_e(r') 4\pi r'^2 dr' + \int_r^\infty \rho_e(r') 4\pi r' dr' \right) \quad (8)$$

where  $\rho_n(r)$  and  $\rho_e(r)$  represent the nuclear and electron charge distributions, respectively. The ELSEPA code [80] provides four nuclear charge distribution models (point nuclear, uniform distribution, Fermi distribution, and Helm's uniform-uniform distribution) and four electron charge distribution models (Thomas-Fermi-Molière density, Thomas-Fermi-Dirac density, Dirac-Hartree-Fock-Slater density, and Dirac-Fock density). It also includes three electron exchange potential models (Furness-McCarthy potential, Thomas-Fermi potential, and Riley-Truhlar potential). In the presence of slow projectiles, charge polarization occurs in the target atom, resulting in an electric field from the induced dipole moment that attracts the projectile. ELSEPA incorporates the Buckingham polarization potential and local density approximation models to accurately describe this polarization effect. Additionally, to account for the solid-state effect, the muffin-tin potential [82] is used as a representation of the potential well in a close-packed crystal lattice. In the muffin-tin approach, it is assumed that  $Z$  electrons of the neutral atom are confined within a sphere of a certain radius  $R_{mt}$  centred at the nucleus. This assumption implies that at distances  $r > R_{mt}$  the electrostatic interaction potential of a bound atom is smaller (in magnitude) than that of a free atom. Consequently, at small scattering angles, the differential cross sections obtained from these solid-state potentials are smaller than those of atomic potentials.

The electrostatic interaction potential for an atom in the solid is assumed to be modified as,

$$V_{st,mt}(r) = V_{st}(r) + V_{st}(2R_{mt} - r), r \leq R_{mt} \quad (9)$$

thus, the electron density of a bound atom will be

$$\rho_{e,mt}(r) = \begin{cases} \rho_e(r) + \rho_e(2R_{mt} - r) + \rho_u, & \text{if } r < R_{mt}; \\ 0, & \text{if } r > R_{mt}. \end{cases} \quad (10)$$

where  $\rho_u$  is a constant ensuring proper normalization,

$$\int_0^{R_{mt}} \rho_{e,mt}(r) 4\pi r^2 dr = Z \quad (11)$$

thus, Eq. (5), for scattering by atoms in solids becomes

$$V_{mt}(r) = \begin{cases} V_{st,mt}(r) + V_{ex}(r) + V_{cp}(r) - iW_{abs}(r), & \text{if } r \leq R_{mt}; \\ V_{st,mt}(R_{mt}) + V_{ex}(R_{mt}) + V_{cp}(r), & \text{if } r \geq R_{mt}. \end{cases} \quad (12)$$

Whereas, default model in ELSEPA is for free atoms, while in this work both free atomic potential and the muffin-tin potential are considered. In all these Mott's cross sections, we have considered four nuclear charge distribution models, four electron distribution models, four electron exchange potential models and three correlation-polarization potential models, resulting in totally 384 ( $= 4 \times 4 \times 4 \times 3 \times 2$ ) different scattering potentials (see Table A1 of Ref. [62]) and the corresponding 384 cross section datasets whereas in previous works [62,

63] only 48 ( $=4 \times 4 \times 3$ ) potentials were considered. Fig. 1 shows the obtained elastic mean free paths from the calculated 384 total cross section datasets. The variation among the cross section datasets is obvious in the whole energy range and becomes quite serious in the low-intermediate energy range, which should affect particularly the description of transport of low energy secondary electrons.

## 2.2. Inelastic scattering models

An electron moving inside a sample interacts with other constituting charged particles through Coulomb's interaction. The excitation of the core-shell and valence electrons in an inelastic scattering event consumes the kinetic energy of the energetic moving electron. The differential inelastic scattering cross section for electrons moving in a solid is expressed in terms of the differential inverse inelastic mean free path [83],

$$\frac{d^2\lambda_{in}^{-1}}{d(\hbar\omega)dq} = \frac{2\gamma^2}{1+\gamma} \frac{1}{\pi a_0(E-E_g)} \text{Im} \left[ \frac{-1}{\epsilon(q,\omega)} \right] \frac{1}{q} \quad (13)$$

where  $\lambda_{in}$  is electron inelastic mean free path (IMFP), the average distance that an electron can travel in the material before losing energy;  $\hbar q$  and  $\hbar\omega$  are the momentum transfer and the energy loss (or the photon energy), respectively.  $\epsilon(q,\omega)$  is the dielectric function of the medium. The relativity correction factor  $\gamma$  is given by  $\gamma = 1 + (E - E_g)/m_e c^2$ , where  $E_g$  is the band gap energy of a semiconductor/insulator ( $E_g = 0$  for a metal),  $m_e$  is electron rest mass,  $c$  is the speed of light, and  $a_0$  is the Bohr radius. The kinetic energy  $E$  of an electron in the above equation is referenced to the bottom of the valence band.

The energy loss function (ELF), defined as  $\text{Im}\{-1/\epsilon\}$ , thus determines completely the behaviour of electron inelastic scattering in a specific material. There are two modes excitation of conduction electrons, i.e. single particle excitation or collective excitation [84], which can be explicitly described by the Lindhard dielectric function [85,86] for a free electron metal. For non-free-electron materials, Ritchie and Howie [87] suggested to derive the  $q$ -dependent ELF from the optical dielectric function,  $\epsilon(\omega)$ , which are available from the experimental measured optical constants. Penn has further extended the experimental optical ELF,  $\text{Im}\{-1/\epsilon(\omega)\}$ , to the full momentum-dependent ELF in a simple integration formalism [36]. This opens the way to more accurately describe electronic excitations in a realistic material from the valence electron excitation at the low energy loss ( $\hbar\omega$ ) region, to the interband transition at the intermediate energy region and the

inner-shell ionization in the high energy region for a Monte Carlo simulation. Further modifications to the Penn's modelling are considered in this work.

After obtaining the ELF, the inverse of the IMFP at an electron kinetic energy  $E$  can be calculated by the integration,

$$\lambda_{in}^{-1} = \iint_D \frac{d^2\lambda_{in}^{-1}}{d(\hbar\omega)dq} d(\hbar\omega)dq \quad (14)$$

Regarding to the integration limit of  $\hbar\omega$ , the smallest energy loss allowed for electronic transition is the energy difference between the bottom of unoccupied states (the bottom of the conduction band) and the top of occupied states (the top of the valence band) or the band gap  $E_g$ , which could be defined as the lower integration limit. According to the Pauli exclusion principle, the electron of energy  $E$  should not fall into the occupied states after an energy loss  $\hbar\omega$ ; then the largest energy loss would be  $E - E_g - E_v$ , where  $E_v$  is the valence bandwidth of a solid, which defines the upper integration limit. From the conservation of energy and momentum, the integration limits of  $\hbar q$  are obtained as:

$$\hbar q_{\pm} = \sqrt{m_e(E - E_g) \left( 2 + \frac{E - E_g}{m_e c^2} \right)} \pm \sqrt{m_e(E - E_g - \hbar\omega) \left( 2 + \frac{E - E_g - \hbar\omega}{m_e c^2} \right)} \quad (15)$$

Therefore, the integral region of the energy loss and momentum transfer is defined by Ref. [83]:  $D = \{(q, \omega) | E_g \leq \hbar\omega \leq E - E_g - E_v, q_- \leq q \leq q_+\}$ . However, in the present numerical evaluation for Si we have set the lower integration limit of  $\hbar\omega$  as 0 for the FPA and the SMA dielectric functions described later by considering the fact that below  $E_g$  there is an additional contribution to ELF from electron-phonon scattering channel. In this case, it is easy to find that the energy requirement for non-zero values of  $\lambda_{in}^{-1}$  is  $E \geq E_g + E_v$  and below which the IMFP tends to be infinite. While for the LLM dielectric function, due to the variable change, the electron-phonon scattering is in fact omitted and, hence, the actual lower integration limit of  $\hbar\omega$  is still  $E_g$ ; the condition for non-zero values of  $\lambda_{in}^{-1}$  is  $E \geq 2E_g + E_v$ . For Si,  $E_g \simeq 1.1$  eV and  $E_v \simeq 12.5$  eV.

### i. Full Penn algorithm

In the full Penn algorithm (FPA) [36], the ELF is obtained by extrapolating the optical ELF,  $\text{Im}\{-1/\epsilon(\omega)\}$ , onto the  $(q, \omega)$ -plane by an expansion according to the Lindhard ELF,

$$\text{Im} \left\{ \frac{-1}{\epsilon(q, \omega)} \right\} = \int_0^\infty d\omega_p g(\omega_p) \text{Im} \left\{ \frac{-1}{\epsilon_L(q, \omega; \omega_p)} \right\} \quad (16)$$

where the expansion coefficient  $g(\omega)$  is found to be

$$g(\omega) = \frac{2}{\pi\omega} \text{Im} \left\{ \frac{-1}{\epsilon(\omega)} \right\} \quad (17)$$

where  $\epsilon_L(q, \omega; \omega_p)$  is the Lindhard dielectric function of the free electron gas with plasmon energy  $\hbar\omega_p$  in the long wave limit  $q \rightarrow 0$ . The real and imaginary parts of the Lindhard dielectric function are respectively represented by,

$$\epsilon_L^r = 1 + \frac{2}{\pi a_0 q} \frac{1}{Z} \left[ \frac{1}{2} + \frac{1}{8Z} F\left(Z - \frac{X}{4Z}\right) + \frac{1}{8Z} F\left(Z + \frac{X}{4Z}\right) \right] \quad (18)$$

and

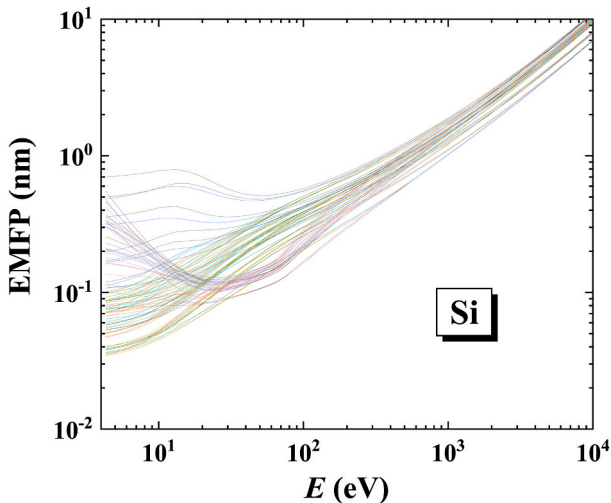


Fig. 1. Calculated elastic mean free paths from the 384 different elastic scattering cross section datasets by the ELASP code for crystalline Si. The kinetic energy  $E$  of an electron is referenced to the vacuum level.



$$\varepsilon_L^i = \begin{cases} \frac{1}{8a_0k_F} \frac{X}{Z^3}, 0 \leq X \leq 4Z(1-Z); \\ \frac{1}{8a_0k_F} \frac{1}{Z^3} \left[ 1 - \left( Z - \frac{X}{4Z} \right)^2 \right], |4Z(1-Z)| \leq X \leq 4Z(1+Z); \\ 0, \text{ otherwise.} \end{cases} \quad (19)$$

where  $F(x) = (1 - x^2)\ln|(x+1)/(x-1)|$ ,  $X = \hbar\omega/E_F$ ,  $Z = q/2k_F$  and  $E_F = \hbar^2k_F^2/2m$  is the Fermi energy and  $k_F$  is the Fermi wave vector.

The Lindhard ELF is the  $\delta$ -function along the plasmon dispersion line, where  $\varepsilon_L^e = 0$  and  $\varepsilon_L^i = 0$ . The calculation is thus implemented individually for the single electron excitation part and the plasmon excitation part,

$$\text{Im} \left\{ \frac{-1}{\varepsilon(q, \omega)} \right\}_{\text{FPA}} = \text{Im} \left\{ \frac{-1}{\varepsilon(q, \omega)} \right\}_e + \text{Im} \left\{ \frac{-1}{\varepsilon(q, \omega)} \right\}_{\text{pl}} \quad (20)$$

The single excitation part can be calculated straightforwardly as,

$$\text{Im} \left\{ \frac{-1}{\varepsilon(q, \omega)} \right\}_e = \int_0^\infty d\omega_p g(\omega_p) \text{Im} \left\{ \frac{-1}{\varepsilon_L(q, \omega; \omega_p)} \right\} \quad (21)$$

$$\Theta[q^+( \omega; \omega_p) - q] \Theta[q - q^-( \omega; \omega_p)] ,$$

where

$$\begin{cases} q^-( \omega; \omega_p) = -k_F(\omega_p) + \sqrt{k_F^2(\omega_p) + 2m\omega/\hbar} ; \\ q^+( \omega; \omega_p) = k_F(\omega_p) + \sqrt{k_F^2(\omega_p) + 2m\omega/\hbar}, \end{cases} \quad (22)$$

are the left and right boundaries of the area for  $\varepsilon_L^i \neq 0$ , respectively.

Due to the character of the  $\delta$ -function, the plasmon excitation part can be calculated by

$$\text{Im} \left\{ \frac{-1}{\varepsilon(q, \omega)} \right\}_{\text{pl}} = g(\omega_p) \frac{\pi}{|d\varepsilon_L^e(q, \omega; \omega_p)/d\omega_p|} \Big|_{\varepsilon_L^e(q, \omega; \omega_p)=0} \Theta[q^-( \omega; \omega_p) - q] \quad (23)$$

where the slope of the real part of the Lindhard dielectric function at the plasmon dispersion line is given as,

$$\begin{aligned} d\varepsilon_L^e(q, \omega; \omega_p)/d\omega_p|_{\varepsilon_L^e(q, \omega; \omega_p)=0} = & -\frac{2}{3\omega_p} \left\{ 2 + \frac{1}{\pi a_0 q} \frac{1}{Z} + \frac{1}{2\pi a_0 q} \frac{1}{Z} \times \left[ 2 \right. \right. \\ & - C_1 \ln \left| \frac{C_1 + 1}{C_1 - 1} \right| - C_2 \ln \left| \frac{C_2 + 1}{C_2 - 1} \right| + \frac{X}{4Z^2} \left( C_1 \ln \left| \frac{C_1 + 1}{C_1 - 1} \right| \right. \\ & \left. \left. - C_2 \ln \left| \frac{C_2 + 1}{C_2 - 1} \right| \right) \right] \Big\}, \end{aligned} \quad (24)$$

where  $C_1 = Z - X/4Z$  and  $C_2 = Z + X/4Z$ .

## ii Super-extended Mermin algorithm

There are also some other dielectric function models. To account for the plasmon damping, Mermin introduced a relaxation-time approximation in the Lindhard dielectric function by turning the frequency  $\omega$  into a complex frequency  $\omega + i\gamma$ , where  $\gamma$  represents the damping rate of the plasmon and derived the following expression by preserving the local number of electrons [88],

$$\varepsilon_M(q, \omega; \omega_p, \gamma) = 1 + \frac{(1 + i\gamma/\omega)[\varepsilon_L(q, \omega + i\gamma; \omega_p) - 1]}{1 + (i\gamma/\omega)[\varepsilon_L(q, \omega + i\gamma; \omega_p) - 1]/[\varepsilon_L(q, 0; \omega_p) - 1]} \quad (25)$$

where  $\varepsilon_M(q, \omega; \omega_p, \gamma)$  is the Mermin dielectric function for a single oscillator at plasmon energy  $\omega_p$ . The Mermin-type ELF can be expressed

by the Drude-Lorentz function at  $q = 0$ . By fitting the experimentally obtained optical ELF as a linear combination of Drude-type ELFs,

$$\text{Im} \left\{ \frac{-1}{\varepsilon(0, \omega)} \right\} = \sum_{i=1}^N a_i \text{Im} \left\{ \frac{-1}{\varepsilon_M(0, \omega; \omega_{pi}, \gamma_i)} \right\} = \sum_{i=1}^N a_i \frac{\gamma_i \omega \omega_{pi}^2}{(\omega^2 - \omega_{pi}^2)^2 + \gamma_i^2 \omega^2} \quad (26)$$

where the fitting parameters,  $\gamma_i$ ,  $\omega_{pi}$  and  $a_i$ , are the width, energy, and oscillator strength of the  $i$ th oscillator, respectively. In the above expression  $a_i$  is usually taken as positive, which should satisfy the requirement of  $f$ -sum rule [89]. However, at an ionization edge this fitting may lose accuracy. Da et al. [90,91] have proposed the super-extended Mermin algorithm (SMA), with which some negative oscillator strengths  $a_i$  among a large number of oscillators ( $N \sim 50 - 150$ ) are employed to describe accurately the experimental optical ELF spectrum in the entire energy loss range including not only the valence electron excitation but also the phonon and the inner-shell excitations. With the help of these negative oscillators, which do not hold any physical meaning but are simply useful for fast convergence, even the sharpest features of optical ELF around the band gap energy or the ionization edge for the inner-shell electron excitation can be accurately described. The SMA dielectric function is thus,

$$\text{Im} \left\{ \frac{-1}{\varepsilon(q, \omega)} \right\}_{\text{SMA}} = \sum_{i=1}^N a_i \text{Im} \left\{ \frac{-1}{\varepsilon_M(q, \omega; \omega_{pi}, \gamma_i)} \right\} \quad (27)$$

The FPA is a special case of the SMA for  $\gamma_i \rightarrow 0$  and  $N \rightarrow \infty$  so that negative oscillator strengths are no more useful. The previous calculations [90,91] have shown that the SMA improves the calculated IMFPs at low kinetic energies by comparing with the experimental data. In Table 2 of Supplementary Materials, we have listed all the Drude-type oscillator terms for fitting the three experimental optical datasets by Eq. (27).

## iii Levine-Louie model

In FPA and SMA the material is treated as metallic without a bandgap. A modification to the Lindhard dielectric function has been proposed by Levine and Louie (LL) [78] for semiconductors and insulators having a band gap. The LL model (LLM) dielectric function is represented by the formula of the Lindhard dielectric function as,

$$\varepsilon_{\text{LLM}}(q, \omega; \omega_p) = \varepsilon_L(q, \omega'; \omega_p'), \hbar\omega > E_g \quad (28)$$

by introducing a variable transformation  $(\hbar\omega')^2 = (\hbar\omega)^2 - E_g^2$ . It is easy to show that the  $f$ -sum rule is maintained for the variable change. It should be noted that the position of the plasmon pole is changed simultaneously, i.e.  $\hbar\omega_p' = \sqrt{(\hbar\omega_p)^2 - E_g^2}$ . Thus, the expansion of the dielectric function in Eq. (16) is performed according to the LLM dielectric function as follows,

$$\begin{aligned} \text{Im} \left\{ \frac{-1}{\varepsilon(q, \omega)} \right\}_{\text{LLM}} &= \int_{E_g}^\infty d\omega_p g(\omega_p) \frac{\omega_p^2}{\omega_p'^2} \text{Im} \left\{ \frac{-1}{\varepsilon_{\text{LLM}}(q, \omega; \omega_p')} \right\} \\ &= \int_{E_g}^\infty d\omega_p g(\omega_p) \frac{\omega_p^2}{\omega_p'^2} \text{Im} \left\{ \frac{-1}{\varepsilon_L(q, \omega'; \omega_p')} \right\} \end{aligned} \quad (29)$$

where  $\hbar\omega_p' = \sqrt{(\hbar\omega_p)^2 - E_g^2}$ . Note that the factor  $\omega_p^2/\omega_p'^2$  is introduced to satisfy the  $f$ -sum rule and the ELF formulas for the single electron and the plasmon excitation parts are, respectively,

$$\text{Im}\left\{\frac{-1}{\varepsilon(q, \omega)}\right\}_{\text{pl}} = g(\omega_p) \frac{\omega_p^2}{\omega_p^2} \left| \frac{\pi}{\frac{d\varepsilon_L(q, \omega; \omega_p(\omega_p))}{d\omega_p} \frac{d\omega_p}{d\omega_p}} \right|_{\varepsilon_L(q, \omega; \omega_p')=0} \quad (30)$$

$$\Theta\left[q^-(\omega(\omega); \omega_p(\omega_p)) - q\right].$$

and

$$\text{Im}\left\{\frac{-1}{\varepsilon(q, \omega)}\right\}_{\text{e}} = \int_0^\infty d\omega_p g(\omega_p) \frac{\omega_p^2}{\omega_p^2} \text{Im}\left\{\frac{-1}{\varepsilon_L(q, \omega(\omega); \omega_p(\omega_p))}\right\} \quad (31)$$

$$\Theta\left[q^+(\omega(\omega); \omega_p(\omega_p)) - q\right] \Theta\left[q - q^-(\omega(\omega); \omega_p(\omega_p))\right].$$

The ELF can then be obtained by

$$\text{Im}\left\{\frac{-1}{\varepsilon(q, \omega)}\right\}_{\text{LLM}} = \begin{cases} \text{Im}\left\{\frac{-1}{\varepsilon(q, \omega)}\right\}_{\text{e}} + \text{Im}\left\{\frac{-1}{\varepsilon(q, \omega)}\right\}_{\text{pl}}, & \hbar\omega > E_g; \\ 0, & \text{otherwise.} \end{cases} \quad (32)$$

#### iv Comparison between the models

To see the difference between the FPA, SMA and LLM, the surface and contour plots of the three  $q$ -dependent ELFs as functions of momentum transfer and energy loss are shown in Fig. 2. The optical constants were taken from the Palik's database [75]. Fig. 2 illustrates that

the difference between these three models is not very significant in the whole energy loss and momentum transfer range. The optical ELF demonstrates a damped plasmon peak at  $\sim 16.7$  eV whose intensity decays quickly along the dispersion entering into the single particle excitation region. Compared with the FPA the plasmon peak in the LLM is a little bit enhanced and the dispersion is shifted to higher energies. In other approximations, like single-pole approximation [36] and the simple Ritchie-Howie algorithm [87], the single particle excitation is ignored. In all the three dielectric function models the dispersion follows that of the Lindhard dielectric function for free electron gas, and the exchange-correlation effect is omitted in the present dielectric function formalism, Eq. (13), which may contribute to the plausible systematic error on the estimated IMFP and the SEY.

Fig. 3(a) shows the comparison on the IMFPs calculated by the three dielectric function models. It is seen that the difference between the FPA and LLM is quite small because of the small bandgap  $\sim 1.1$  eV for Si. Nguyen-Truong has used the LL formalism, but with the Mermin dielectric function in replace of Lindhard dielectric function in Eq. (29), in the calculation of IMFP for water having a wide gap  $\sim 6$  eV [92]. His LL result demonstrates a significant reduction of IMFP at very low energies when compared with the FPA result for water, which is perhaps mainly due to a different choice of the integration limit.

#### 2.3. Optical ELF

During the transportation of electrons in a solid, the inelastic interaction is the vital process related to the secondary electron production. Therefore, the uncertainty of optical ELF data as an essential input to our

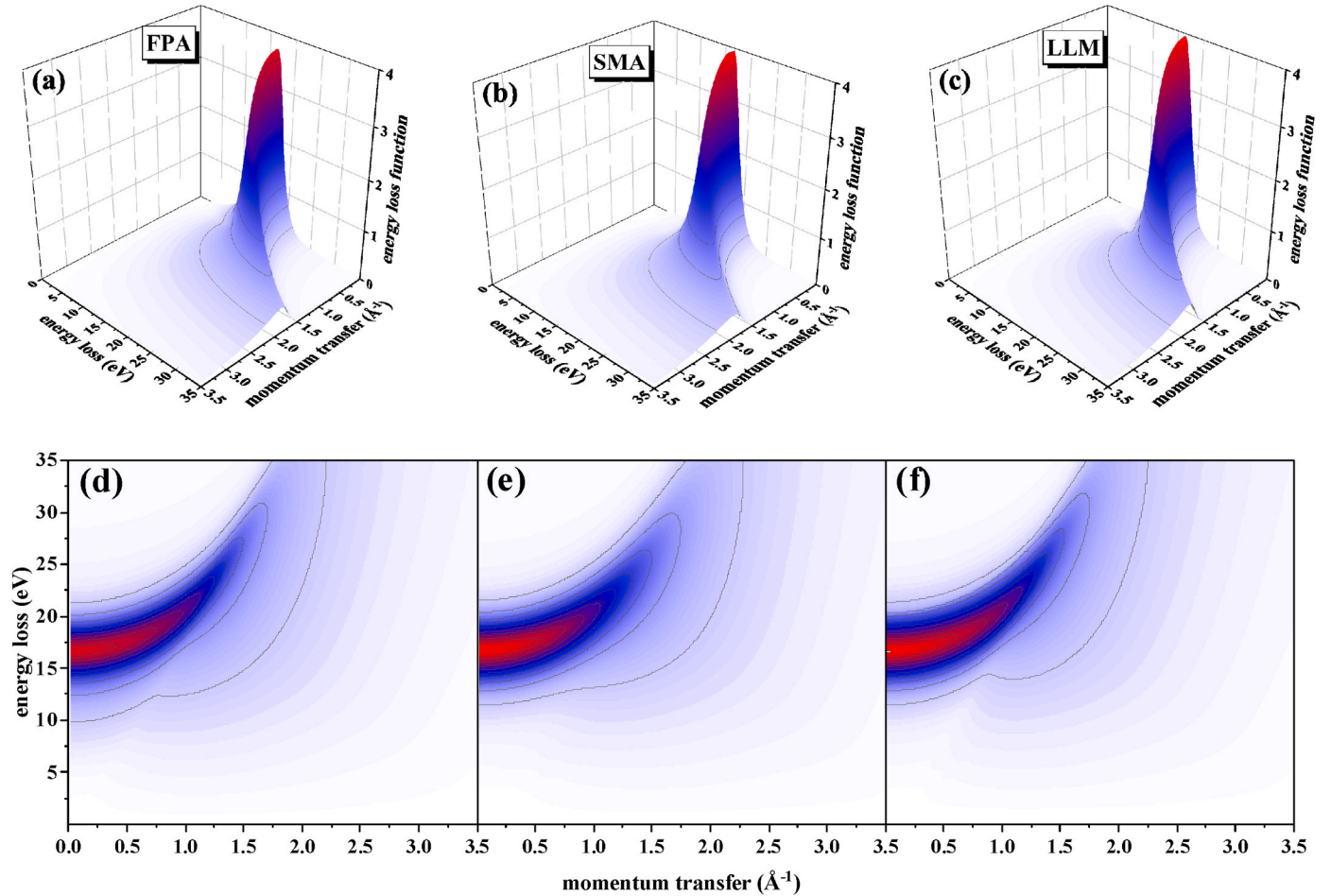
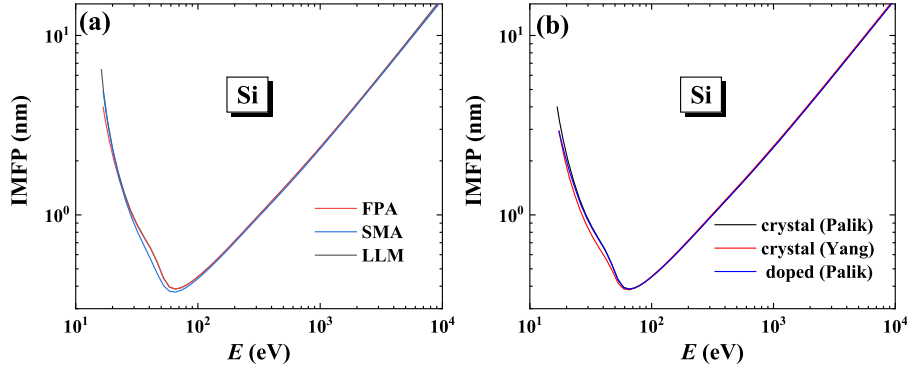


Fig. 2. The surface plots (top panel) and contour plots (bottom panel) of the  $q$ -dependent ELFs for crystalline Si, calculated by the FPA (left), SMA (middle) and LLM (right).



**Fig. 3.** (a) Calculated IMFPs by the FPA, SMA and LLM with the “crystal (Palik)” ELF; (b) calculated IMFPs by the FPA with three different ELFs, i.e. “crystal (Palik)”, “crystal (Yang)” and “doped (Palik)”. The kinetic energy  $E$  of an electron is referenced to the bottom of the valence band.

Monte Carlo simulation may have an impact on the simulated yield, particularly to SEY. Palik has compiled optical constants, usually in the low to the medium photon energy  $\hbar\omega$  range from below 1 eV to above 1 keV, for some elements and compounds. These compiled data from various sources measured with optical methods form a database [75]. In recent years there have also been other data sources of measured optical constants by reflection electron energy loss spectroscopy technique for some materials in the low  $\hbar\omega$  range from 1 eV to  $\sim 100$  eV [93–101]. Other databases [102,103] provide complementary data at higher photon energies. Here we have selected three ELF datasets for Si, as shown in Fig. 4, named as “crystal (Palik)”, “crystal (Yang)” and “doped (Palik)” by combining various sources from Palik [75], Henke et al. [102] and Yang et al. [100,101]. The details of these combined ELF datasets are listed in Table 1. For the Sb doped (at concentration of  $\sim 0.002$ ) n-type Si, the ELF is much enhanced below the bandgap as compared with that of pure Si. Limited by the energy resolution of electron beam technique, the ELF in the “crystal (Yang)” dataset below 2 eV was approximated by a straight line such that the sum rules are satisfied. Therefore, all the ELFs for crystalline silicon (c-Si) are very close in value above the bandgap, and their main difference among the ELFs is at the phonon excitation part below the bandgap.

The accuracy of the optical constants can be verified by several sum rules [111,112]; the most frequently used ones are the oscillator strength sum rule ( $f$ -sum rule) and the perfect screening sum rule ( $ps$ -sum rule). The  $f$ -sum rule  $Z_{\text{eff}}$  is given by,

$$Z_{\text{eff}} = \frac{2}{\pi \Omega_p^2} \int_0^{\omega_{\text{max}}} \omega \text{Im} \left\{ \frac{-1}{\varepsilon(\omega)} \right\} d\omega \quad (33)$$

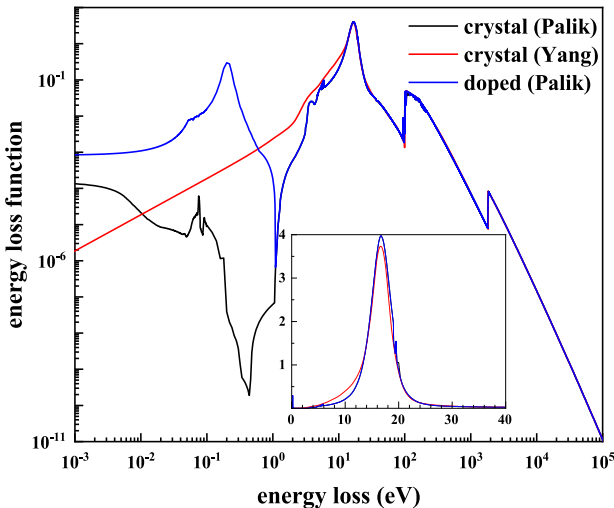
where  $\hbar\Omega_p = \sqrt{4\pi n_a e^2 / m}$ . The expectation value of  $Z_{\text{eff}}$  equals to the atomic number  $Z$ , or the total number of electrons per atom or molecule, when  $\omega_{\text{max}} \rightarrow \infty$ . The  $ps$ -sum rule  $P_{\text{eff}}$  can be obtained from the Kramers-Kronig relation [113,114] as:

$$P_{\text{eff}} = \frac{2}{\pi} \int_0^{\omega_{\text{max}}} \frac{1}{\omega} \text{Im} \left\{ \frac{-1}{\varepsilon(\omega)} \right\} d\omega + \text{Re} \left\{ \frac{-1}{\varepsilon(0)} \right\} \quad (34)$$

where  $\text{Re} \{ -1/\varepsilon(0) \} = 0$  for conductors. For non-conductors, the refractive index  $n$  is much larger than the extinction coefficient  $k$  at low frequencies. Therefore,  $\text{Re} \{ -1/\varepsilon(0) \} \simeq 1/n^2(0)$ . For Si,  $n(0) = 3.4155$  is taken for the calculation of  $P_{\text{eff}}$ . The expectation value of  $P_{\text{eff}}$  should be unity when  $\omega_{\text{max}} \rightarrow \infty$ . The accuracy of the  $f$ -sum rule is dominated by the accuracy of the high-energy part of the ELF according to the  $\omega$ -factor in the integration of Eq. (33). In contrast, the accuracy of the  $ps$ -sum rule is dominated by the low energy part of the ELF, as indicated by the  $\omega^{-1}$ -factor in the integration of Eq. (34), and this is the most important energy loss region in electron inelastic scattering responsible for valence electron excitation in addition to phonon scattering. It is thus concluded that each sum rule emphasizes the respective energy regions of importance. However, this is anyway a simple overall estimation, and the accuracy of an ELF in a certain photon energy region cannot be definitively stated.

Table 2 presents the  $f$ - and  $ps$ -sum rules and the relative errors for the considered three ELFs. Fig. 5 shows the plot of sum rules as functions of integration limit. These sum rule values vary among the three different ELF datasets. Since all the  $f$ -sums are rather satisfactory, being below 1%, while the  $ps$ -sums are quite different, we may suggest that the “crystal (Yang)” dataset would be better.

Fig. 3(b) compares the energy dependent IMFPs derived from FPA with the three different ELF datasets. It can be seen that the three IMFP curves are quite close in the logarithmic scale plot. The difference among them is under  $\sim 10\%$ , which is expected to cause a slight variance of the simulated yield data. Above 60 eV, all the curves agree; below 60 eV, the “crystal (Yang)”-IMFP curve is lower than “crystal (Palik)”-IMFP curve while the “doped (Palik)”-IMFP is in between them. This behavior is consistent with the sum rule results shown in Table 2, where the negative sum rule errors for “crystal (Palik)”-ELF indicate that this ELF is underestimated in the whole range of photon energy  $\hbar\omega$  and, hence, the resultant IMFP is overestimated in the whole range of kinetic energy  $E$  and particularly at low energies. The previous calculation on BSCs for some metals by using different ELF datasets, for which very different errors of sum rules are observed, have already indicated that, sum rules



**Fig. 4.** Comparison of three selected ELFs from different sources, i.e. “crystal (Palik)”, “crystal (Yang)” and “doped (Palik)”.

**Table 1**

The optical ELF of Si data sources used.

| Si form         | photon energy range (eV) and the corresponding dataset in literature |                 |                 |                 |               |                |
|-----------------|----------------------------------------------------------------------|-----------------|-----------------|-----------------|---------------|----------------|
| crystal (Palik) | 0.01–1.10 [104,105]                                                  | 1.10–3.10 [106] | 3.10–6.00 [107] | 6.00–20.0 [108] | 20.0–100 [75] | 100–2000 [109] |
| crystal (Yang)  | 0.01–200 [101]                                                       |                 |                 |                 |               | 200–2000 [109] |
| doped (Palik)   | 0.01–1.10 [75,110]                                                   | 1.10–3.10 [106] | 3.10–60 [107]   | 6.00–20.0 [108] | 20.0–100 [75] | 100–2000 [109] |

**Table 2**The  $f$ - and  $ps$ -sum rules and their relative errors for ELFs of Si.

| ELF             | $f$ -sum | relative error | $ps$ -sum | relative error |
|-----------------|----------|----------------|-----------|----------------|
| crystal (Palik) | 13.950   | −0.36%         | 0.962     | −3.84%         |
| crystal (Yang)  | 14.134   | 0.96%          | 1.000     | 0.02%          |
| doped (Palik)   | 14.015   | −0.10%         | 1.051     | 5.09%          |

can explain well the discrepancy of the calculated BSCs with different ELFs [115]. Therefore, the quality of ELF in terms of sum rules can indeed impose certain impact to the simulation results. Because the IMFPs are different at energies below 60 eV by different ELFs, it can be predicated that ELF input data can affect the simulated SEY. But, in view of the small difference among the sum rules, the difference between the secondary yields obtained with different ELFs is expected to be several percent. It is worth noting that the three optical ELFs investigated in this study represent bulk optical property and, hence, the electron inelastic scattering modelled by Eq. (13), while the surface plasmon excitation effect omitted in the modelling could introduce a systematic error. However, the magnitude of the error is expected to be small and potentially negligible.

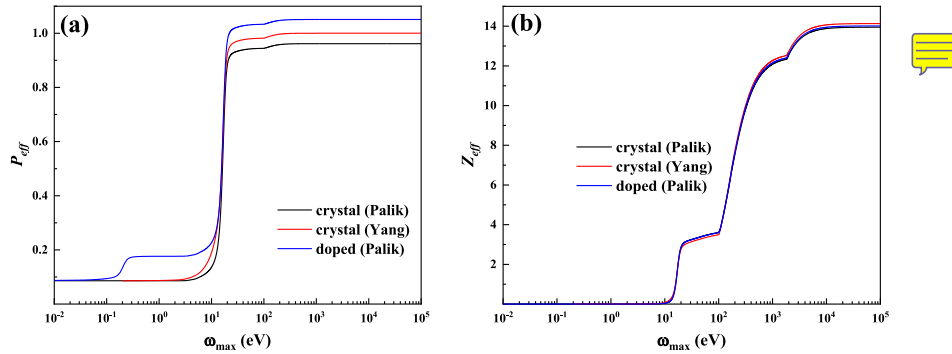
#### 2.4. Secondary electron excitation and emission

##### i Work function data

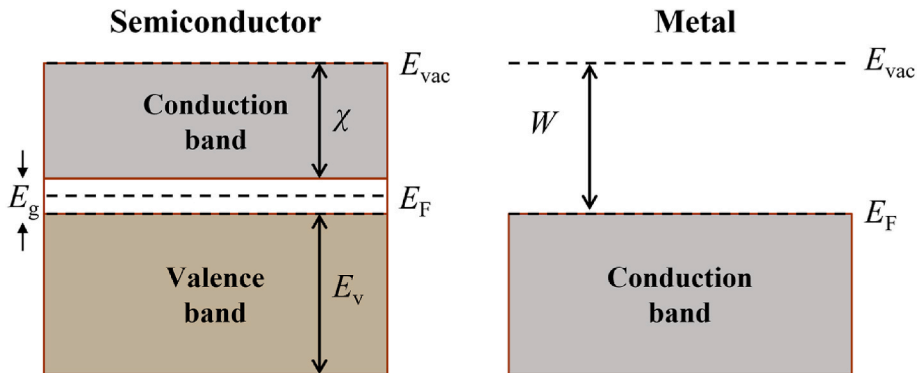
After certain elastic and inelastic collisions inside the sample, an electron may arrive at the surface for emission, and the decision about refraction or reflection of the electron from the surface will be judged by a quantum mechanical transmission coefficient [116] as,

$$T(E, \beta) = \begin{cases} \frac{4\sqrt{1 - U_0/E \cos^2 \beta}}{[1 + \sqrt{1 - U_0/E \cos^2 \beta}]^2}, & \text{if } E \cos^2 \beta > U_0; \\ 0, & \text{otherwise.} \end{cases} \quad (35)$$

where  $\beta$  is the angle between the direction of moving electron and surface normal and  $U_0$  is the surface potential barrier, which is related to the energy reference point. In this work, the electron energy in a Monte Carlo simulation is referenced to the bottom of the conduction band of a semiconductor or an insulator. Therefore,  $U_0$  is electron affinity  $\chi$  (Fig. 6). An electron gains or losses its kinetic energy by the potential barrier when penetrating the surface from the vacuum side or from the bulk side. Hence, the surface barrier plays a significant role in the secondary electron emission. There are several methods for the experimental determination of the work function, i.e. the thermionic method, the photoelectric method, the field-emission method, the effusion method, the contact potential difference method and the calorimetric method. In Table 1 of Supplementary Materials, we list the available data of the work function and electron affinity  $\chi$  for Si measured by different researchers under different experimental conditions, and Fig. 1



**Fig. 5.** Comparison on (a)  $f$ -sum rules and (b)  $ps$ -sum rules, as functions of integration limit, for three different ELFs.



**Fig. 6.** Energy diagram, where  $E_{vac}$  is the vacuum level,  $E_g$  the bandgap,  $E_v$  the valence bandwidth,  $E_F$  the Fermi level,  $W$  the work function and  $\chi$  the electron affinity.



of Supplementary Materials illustrates the statistical distribution of reported work function values. The range of measured  $W$  is from 3.59 eV to 5.4 eV; the uncertainty of the work function value is considered as partly due to the surface contamination effect in experiments. We used this range as our uncertainty range of work function in the simulation.

In the simulation, the energy reference level of an electron in a vacuum is set as a vacuum level  $E_{vac}$ , and inside the material the lowest unoccupied state. For metals, the energy reference level inside the material is the Fermi level  $E_F$ ; for semiconductors, it is the bottom of the conduction band (Fig. 6). Since the Fermi level of a semiconductor is at the mid of energy gap at room temperature, the electron affinity measured from the bottom of the conduction band to the vacuum level is  $\chi = W - E_g/2$  and  $U_0 = \chi$ .

## ii Cascade electron generation

When inelastic scattering occurs, the energy loss  $\hbar\omega$  and momentum transfer  $\hbar q$  are determined by random sampling. The cascade electron is assumed to be excited from the valance band. The excitation probability is proportional to a joint density of states, i.e.  $p(E', \hbar\omega) \propto \sqrt{E'(E' + \hbar\omega)}$ , where  $E'$  is the energy of the electron in the valance band.  $E'$  is sampled by a uniform random number  $\xi \in [0, 1]$  by

$$\xi = \frac{\int_{E_0}^{E'} \sqrt{E(E + \hbar\omega)} dE}{\int_{E_0}^{E_v} \sqrt{E(E + \hbar\omega)} dE} \quad (36)$$

where the lower limit of the integral is

$$E_0 = \begin{cases} E_g + E_v - \hbar\omega, & \hbar\omega < E_g + E_v; \\ 0, & \text{otherwise.} \end{cases} \quad (37)$$

After  $E'$  is obtained, the energy of the excited secondary electron can then be determined as

$$E_s = E' + \hbar\omega - (E_g + E_v) \quad (38)$$

The direction of the cascade electron is along the direction of momentum transfer  $\hbar q$ .

## 2.5. Monte Carlo uncertainty quantification

The uncertainty propagation from the uncertainty of variables can be derived analytically once a functional expression of these variables is given. For a function  $y = f(x_1, x_2, \dots, x_N)$ , the combined standard uncertainty for the measurand  $y$  is determined as  $u_y^2 = \sum_{i=1}^N (\partial f / \partial x_i)^2 u_{x_i}^2$ , where  $u_{x_i}$  is the uncertainty for the  $i$ th input quantity [117]. However, in our present case the functional dependence of electron emission yields on the variables (elastic scattering cross section, inelastic scattering model, ELF and work function) cannot be explicitly given. Therefore, we have adopted a Monte Carlo uncertainty quantification (MCUQ) approach to evaluate the propagation of uncertainties [118,119]. The chief procedure of the approach is to set the probability density functions for the input quantities and then to run the Monte Carlo simulation. The simulation will provide the distribution of the output and, hence, the uncertainty of the output. The Monte Carlo procedure acts as a computational experiment for the output statistics; in this work the

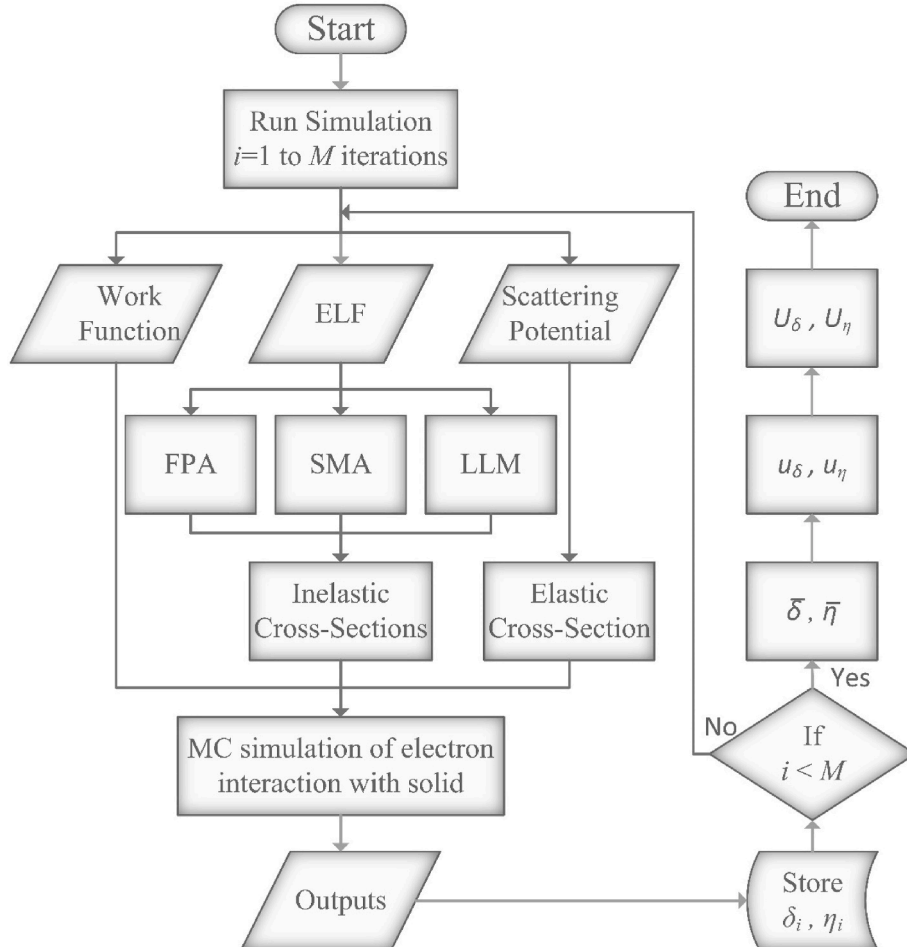


Fig. 7. Flowchart of MCUQ used for estimation of theoretical uncertainty of Monte Carlo simulated electron emission yields.

procedure is just the Monte Carlo calculation process of electron emission yields through the tracing of electron trajectories.

Fig. 7 demonstrates the flowchart for the present theoretical calculation of electron emission yields and the related UQ. Uncertain inputs of experimental data involve work function and ELF while uncertainty for the theoretical modelling includes elastic scattering potential and dielectric function model. Here, we consider that all the uncertain inputs are equally important and uniformly distributed in a certain range. Then, a number of Monte Carlo trials,  $M = 17280$  (384 scattering potentials, 3 dielectric function models, 5 work function values, 3 ELF datasets) for each incident energy is chosen priori. We have employed a parallel computer to perform the Monte Carlo calculation, which is very suitable for our present purpose. At each simulation we have traced  $1 \times 10^5$  incident electron trajectories and several dozens of secondary electron cascade trajectories.

### 3. . Results and discussion

Fig. 8 compares the simulated energy spectra of secondary electrons obtained by the FPA, SMA and LLM with the theoretically calculated and experimentally measured distributions by Amilio et al. [120], Dapor et al. [121] and Goto et al. [122]. The inset figure presents normalized energy spectra calculated with the FPA, showcasing the variation within the range of work function values from 3.59 to 5.4 eV. This inset provides further insight into the behaviour of the energy spectra with respect to different work function values, specifically focusing on the FPA. All the three models yield the similar curve shape with a peak around  $\sim 2$  eV due to secondary cascade process. Note that the experimental spectrum was originally measured as  $EN(E)$  curve by a cylindrical mirror analyser; the peak position at very low energies may thus not be very accurate when the spectrum is converted into  $N(E)$  curve. A weak shoulder found experimentally around 10 eV is due to secondary electrons generated via plasmon damping as in the case of aluminium [123,124], while in the simulation this feature is more obvious by the FPA than by the LLM.

Fig. 9 compares our calculated values of SEY, BSC and TEY along with their corresponding standard deviations for Si for the varied work function with experimentally measured data. Bronstein and Fraiman

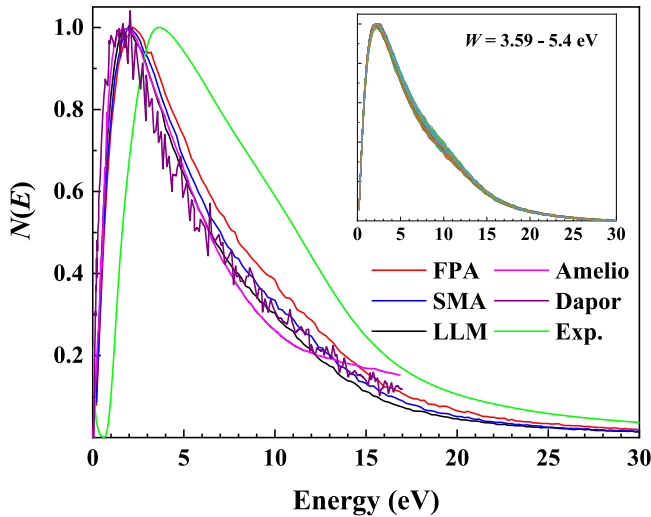
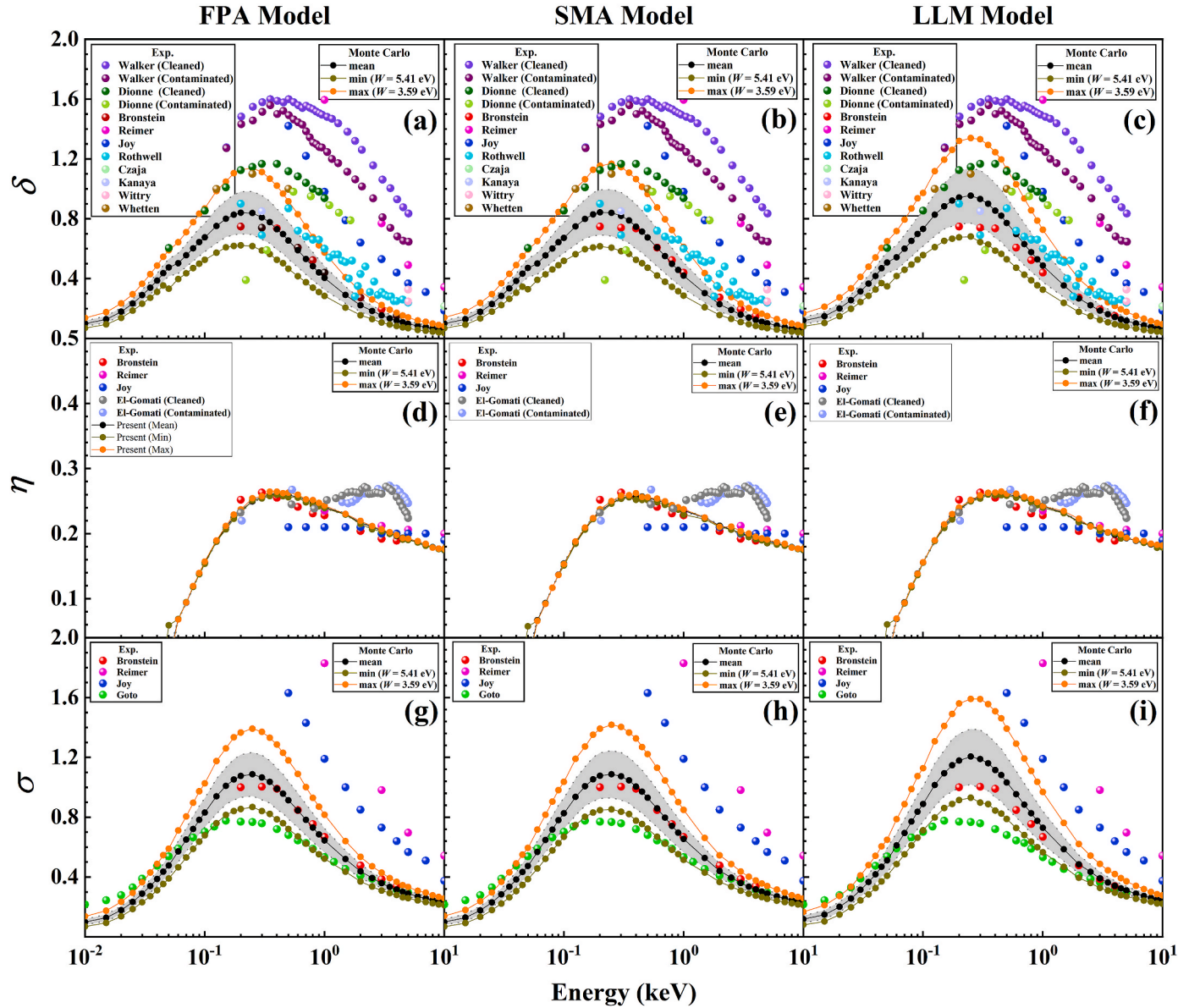


Fig. 8. Comparison of energy spectra of secondary electrons calculated with the FPA (red), SMA (blue) and LLM (black) dielectric functions with theoretically calculated curves by Amilio (pink) [120] and Dapor (purple) [121] and experimentally measured curve by Goto (green) [122]. The present simulations were performed for:  $E_p = 1$  keV, number of incident trajectories = 100000, ELF = “crystal (Palik)”, elastic potential No. = “42001”, and work function  $W = 4.61$  eV.

have performed SEY and BSC measurements in relatively good vacuum conditions for incident beam energies of 50 eV-4 keV. They have prepared the sample using the in-situ evaporation technique to achieve a cleaner surface [125]. Reimer and Tollkamp have measured SEY and BSC for incident beam energies of 1–30 keV in a scanning electron microscope [126]. Walker et al. have measured SEY of cleaned and contaminated Si in an ultra-high vacuum at incident beam energies of 250 eV-5 keV [13]. In addition, there are five other data sources [127–131]. It can be seen from our simulation results that the work function affects SEY and hence TEY greatly, but hardly influences BSC. The calculated BSC curve agrees with experimental data very well, where the three datasets measured by Bronstein [125], Reimer [126] and Joy [132] have a good consistency. Additionally, El-Gomati et al. [133] have conducted BSC measurements on cleaned and contaminated samples, revealing a noteworthy discrepancy with other data; the authors suspected the possibility of carbon or oxygen residual contamination even though the surface was subjected to ion bombardment for periods. However, our previous studies [62,63] have indicated that the lower atomic number of element contamination will reduce the BSC value other than elevate it.

For SEY and TEY, the situation becomes very different; the reported experimental data are very scattered among the different measurements and the data dispersion is greater than other materials. Bronstein’s SEY data are the smallest, while the Walker’s data are the highest. In between of them Rothwell’s data [134] are closer to Bronstein’s data, while those of Dionne [21], Joy [132] and Reimer [126] are more close to Walker’s data. Among the three dielectric function models, the LLM yields the highest SEY values, while the FPA and SMA present the similar SEY values. Both the mean values of SEY and TEY by the FPA and SMA are found to be in a better agreement with Bronstein’s data than other experiments. Goto has conducted TEY measurements for the incident electron energies ranging from 10 eV to 5 keV using a state-of-the-art cylindrical mirror analyzer in an ultrahigh vacuum environment; the sample surfaces were cleaned by ion sputtering. It is believed that the Goto’s database [122] is more reliable and provides estimates that are closer to the true values than the data in the Joy’s database [135]. Interestingly, Goto’s TEY values are quite close to the SEY values of Bronstein, the lowest data in the Joy’s SEY database, and, hence, are significantly smaller than the TEY data obtained by Bronstein. In other words, this suggests that all existing experimental data on SEY in the Joy’s SEY database are overestimated. The primary cause of the large dispersion in the available experimental data stems from the surface conditions during measurements, such as surface contamination and roughness. However, the available information in literature about the effect of contamination on SEY are even controversial. On the one hand, some experiments indicate that compared with the clean surface, the SEY is reduced for the carbon covered surface [136] or for the as-inserted sample with the presence of carbon or oxygen on the surface [13]. On the other hand, some other experiments have shown the opposite tendency that the cleaned surface will present lower SEY values compared to the as-received sample [137–139]. A very recent theoretical work has explored the likely origin for the increase of SEY for adsorbed surfaces, although the work function may also be increased: both carbon and oxygen atoms play a role by the chemical adsorption to modulate the density of states and IMFP [140]. This observation supports the Goto’s data of very low TEY values for a clean surface. Therefore, even very small amount of contaminates on the surface could cause a systematic error for the measurement of SEY data through the combined effects of the change of work function and change of the density of states.

In the present approach, we have considered the uncertainty factor of work function in the range of 3.59–5.41 eV, that has been reported by different researchers [141–154]. In Fig. 9, we have shown minimum, maximum, and mean SEY, BSC and TEY where all other input parameters were kept constants. The elastic scattering cross section was calculated with a specific scattering potential (No. “42001” whereas 4



**Fig. 9.** Comparison between the simulated SEY (top), BSC (middle) and TEY (bottom) and experimental data of Si. The simulation results are derived for varied work function (3.59–5.41 eV) and calculated by the FPA (left), SMA (middle) and LLM (right). ELF = “crystal (Palik)”, elastic potential = “No. 42001”. The simulated results are presented for the mean (black) values, the maximum (orange) and minimum (green) values (corresponding to minimum and maximum work functions, respectively) and the grey area represents the standard deviation range.

stands for Helm’s uniform-uniform distribution of nuclear charge distribution model, 2 stands for Thomas-Fermi-Dirac distribution model of electron distribution model, 0 stands for no exchange potential, 0 stands for no correlation-polarization potential, and 1 stands for bound atom muffin-tin model) while many theoretical uncertainties are not yet included. To obtain the mean values, we have considered two approaches: the simple average and the weighted average. The weighted average is performed with the statistical distribution of reported work function values as given in Fig. 1 in Supplementary Materials. Both approaches give the nearly same averaging result.

Fig. 10 presents the calculation of the standard deviation in SEY, BSC and TEY due to uncertain scattering potential models with the 384 elastic cross sections, where all other input parameters were kept constant. Because the variation tendency of the total elastic cross section (or the elastic mean free path in Fig. 1) on kinetic energy is different for different scattering potential, there is no a definite scattering potential corresponding to the minimum or the maximum SEY/TEY in Fig. 10. In

addition, one can observe an evident sudden change of the SEY curve at  $\sim 50$  eV due to the inclusion of backscattered electrons as secondary electrons by definition for  $E_p < 50$  eV. The calculated mean SEY is seen obviously lower than the Bronstein’s data, while the maximum SEY is very close. The small standard deviation of BSC shows that the elastic cross sections have negligible effect to the BSC calculation of Si for beam energy above 100 eV.

To observe the impact of ELF on the standard deviation in the calculation of SEY, BSC, and TEY by the FPA, SMA and LLM, we have performed the simulations using the three different ELF datasets, whereas all other input parameters were kept constant. Therefore, there are totally  $3 \times 3$  different IMFPs involved. Fig. 11 shows that the calculation of electron emission yields is insensitive to the ELF by the LLM, while it is mostly obvious by the SMA. The ELF-“crystal (Yang)” produces the lower SEY/TEY data than other two ELFs. Notice the fact that both the ELF-“crystal (Palik)” and the ELF-“doped (Palik)”, whose values are very different below the band gap, have resulted in almost the



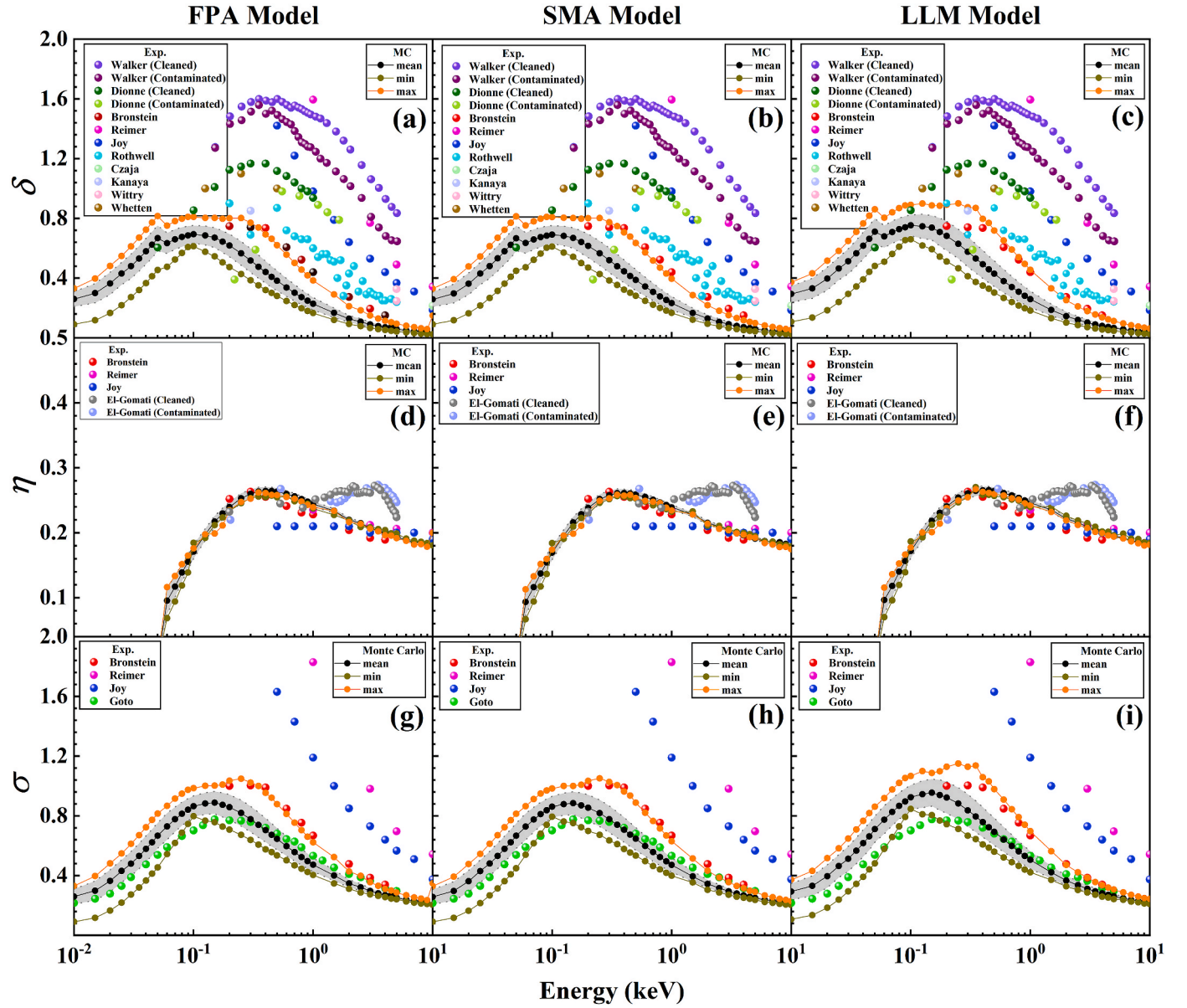


Fig. 10. Comparison between the simulated SEY (top), BSC (middle) and TEY (bottom) and experimental data of Si. The simulation results are derived for 384 elastic scattering potentials and calculated by the FPA (left), SMA (middle) and LLM (right). ELF = “crystal (Palik)” and the work function is taken as 4.61 eV. The simulated results are presented for the mean (black) values, the maximum (orange) and minimum (green) values and the grey area represents the standard deviation range.

identical electron emission yields. Therefore, the lower SEY/TEY values by the ELF-“crystal (Yang)” should be mainly attributed to the small intensity difference within the energy loss range of 2–20 eV with other two ELFs.

Fig. 12 shows the final calculated mean values of SEY, BSC and TEY together with their combined standard uncertainty obtained from the standard deviation,  $u_y = \sqrt{\sum_{i=1}^M (y_i - \bar{y})^2 / (M - 1)}$ , where  $y$  is either  $\delta$ ,  $\eta$  or  $\sigma$ , and  $\bar{y}$  represents the mean value. At each primary energy the calculations are performed over total number of  $M = 17280$  Monte Carlo simulations by changing input parameters or scattering models, and for each simulation  $10^5$  primary electron trajectories and about several tens of times of secondary electron trajectories were traced. The bottom panel shows the statistics of the simulated data range, where the BSC data distribute in a very narrow range of values and obey a Gaussian function. But the SEY, and hence the TEY, have a rather wide standard uncertainty range and the distribution is not Gaussian any more. It is seen that the simulated mean SEY data together with the combined

uncertainty is lower than the Bronstein’s SEY data except below 300 eV, and thus far below than all other experimental data. However, the simulated mean TEY data together with the combined uncertainty are in very close agreement with the Goto’s TEY data in the whole energy range simulated. More specifically, below 200 eV the lower limit of uncertainty range agrees with Goto’s data; in the range between 200 eV and 2 keV the Goto’s data fall within the present uncertainty range of TEY data, and above 2 keV the upper limit of uncertainty range agrees with Goto’s data. Although the Goto’s data are the lowest among all the experimental data, but they could be better in quality for a clean surface than other experimental data. Our present simulation results by taking important theoretical uncertainties into account strongly supports this consideration.

According to the ISO-JCGM 100 [117], the following parameters should be reported as measurement data: i) an estimation of the output quantity, taken as the mean values generated; ii) the standard uncertainty, taken as the standard deviation of these generated values; iii) the chosen coverage probability or the level of confidence (usually 95%),



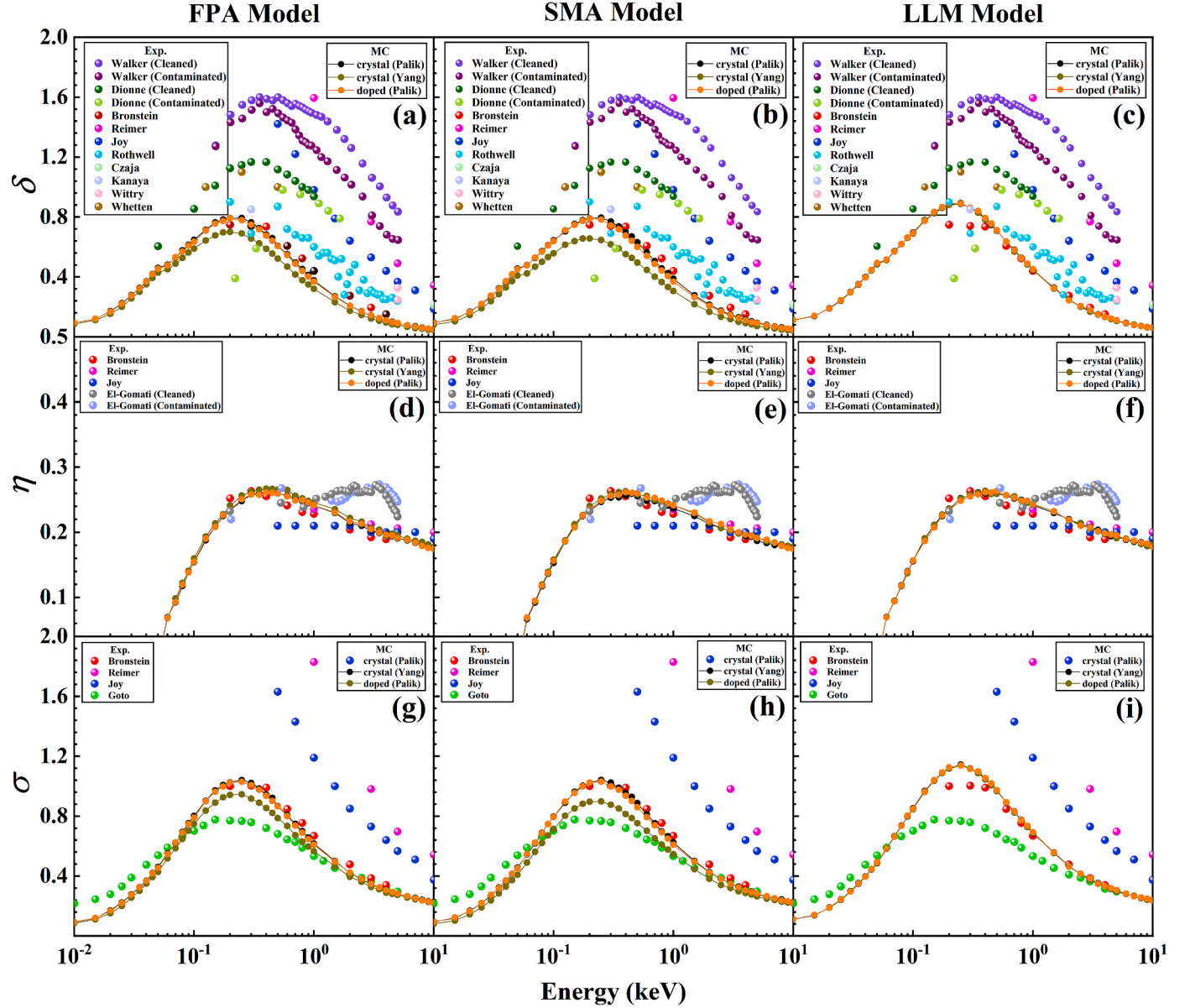


Fig. 11. Comparison between the simulated SEY (top), BSC (middle) and TEY (bottom) and experimental data of Si. The simulation results are derived for three different ELFs, “crystal (Palik)”, “crystal (Yang)” and “doped (Palik)”, and calculated by the FPA (left), SMA (middle) and LLM (right). The elastic potential = “No. 42001” and the work function is taken as 4.61 eV.

and iv) the endpoints corresponding to the selected coverage interval. The last step of UQ is about the expanded uncertainty at the  $k\%$  level of confidence, i.e.

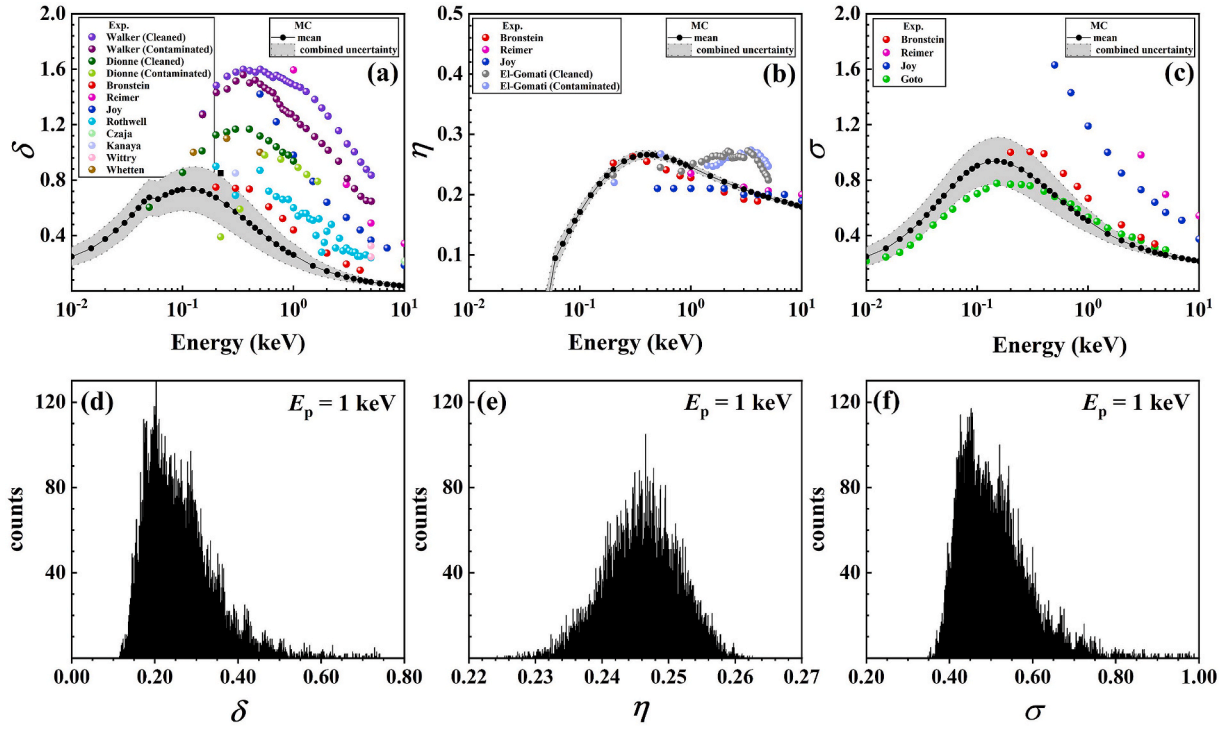
$$U_{\%} = k_{\%} u_{(\delta, \eta, \sigma)} \quad (39)$$

where the value of  $k\%$  at 95% and 75% confidence levels is  $k_{95} = 1.96$  and  $k_{75} = 1.16$ , respectively. Fig. 13 shows the expended uncertainty plots for the calculated SEY, BSC and TEY data at 95% (light grey) and 75% (dark grey) levels of confidence. One can easily observe that the calculated TEY data agree with Goto’s data at 75% confidence level, and agree with Bronstein’s data at 95% the confidence level.

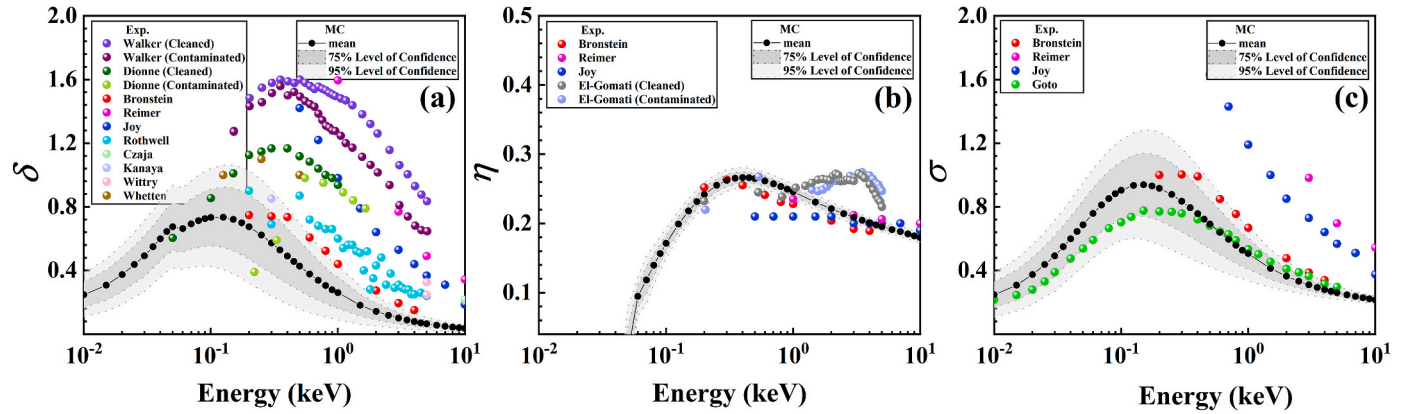
#### 4. Conclusion

Existing experimental database of the measured SEY, BSC and TEY values show large scatter particularly for SEY. The data dispersion is mostly related to the surface status under the measurement via sample

preparation and cleaning procedures. Therefore, experimental databases cannot provide reliable and accurate electron emission yields and render a service for validation of a theoretical modelling of electron-material interaction. In this study, we have conducted a thorough theoretical uncertainty quantification of electron emission from Si by considering several theoretical modelling uncertainty factors, i.e. the elastic potential models for electron elastic scattering cross section calculation, the dielectric function models for electron inelastic scattering cross section calculation, the optical ELF dataset and work function value as input to a Monte Carlo simulation. From the calculation results, we have found that the combined standard uncertainty for BSC is quite small. However, the combined standard uncertainty for SEY and TEY is no more negligible and the dominant uncertainty factors are the work function and the elastic potential. The most remarkable finding of the present work is that our theoretical calculation results of SEY are quite smaller than all the published experimental SEY data in the Joy’s database. But at the 75% confidence level, the Goto’s experimental TEY data lie within the expanded uncertainty range of our simulation results



**Fig. 12.** Top panel: Comparison between the simulated SEY (left), BSC (middle) and TEY (right) and experimental data of Si. At each energy, the simulation is performed for total samples of uncertainty input,  $M = 17280$ . The grey area shows the final combined standard uncertainty range. Bottom panel: the statistics of the simulated 17280 data of SEY (left), BSC (middle) and TEY (right) at 1 keV.



**Fig. 13.** Comparison between the simulated SEY (left), BSC (middle) and TEY (right) and experimental data of Si. At each energy, the simulation is performed for total samples of uncertainty input,  $M = 17280$ . The light and dark grey areas show respectively the expanded uncertainty range at the level of confidence 75% and 95%.

of TEY, which provides a validation of our approach. The present work implies that the previously measured SEY data may contain serious systematic errors and are overestimated. This work has thus laid the foundation for establishing a theoretical database of electron emission in the future.

In summary, our study on electron emission from Si has provided a valuable insight into the uncertainties of the calculated SEY associated with theoretical modelling, meanwhile we also acknowledge the existence of the unconsidered systematic uncertainties, including the surface effects in theoretical modelling and the limitations of the dielectric function theory formalism. Despite the limitations, our finding highlights the importance of considering these theoretical uncertainty factors in future theoretical modelling efforts. Moving forward, it is crucial to address the remaining uncertainties and explore additional sources of error. This will contribute to a more accurate understanding of electron-

material interactions and enable the development of more reliable theoretical models, ultimately leading to enhanced accuracy in predicting and understanding electron emission phenomena. Our present study hence paves the way for future investigations and motivates further research to improve the comprehensive uncertainty quantification of electron emission processes."

#### CRediT authorship contribution statement

**M.S.S. Khan:** Writing – review & editing, Writing – original draft, Visualization, Validation, Methodology, Investigation. **S.F. Mao:** Software, Methodology. **Y.B. Zou:** Methodology, Funding acquisition, Conceptualization. **D.B. Lu:** Software, Investigation. **B. Da:** Software, Methodology. **Y.G. Li:** Software, Methodology, Formal analysis, Conceptualization. **Z.J. Ding:** Writing – review & editing, Supervision,

Project administration, Methodology, Funding acquisition, Formal analysis, Data curation.

### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

### Data availability

Data will be made available on request.

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### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.vacuum.2023.112257>.

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