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Low energy electron transport in alkali halides

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Abstract

A model of electron transport in alkali halides, below 10 eV, is described. It is based on theoretically calculated microscopic cross sections of electron interactions with lattice phonons. Both acoustic and optical scatterings are taken into account, the former being also treated as a quasi-elastic process that randomizes the electron motion. Monte Carlo calculations based on the model simulate the UV-induced photoelectron emission from CsI. The calculated quantum efficiency and energy spectra are in good agreement with experimental data, in the photon energy range of 6.3 - 8.6 eV. The probability for an electron to escape from CsI, NaCl and KCl is provided as a function of its energy and creation depth. A comparison is made between our approach and other phenomenological models.

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1 Introduction

It is known that alkali halides are excellent secondary electron (SE) emitters. The SE yield induced by X-rays or electrons in these materials is by an order of magnitude larger than in metals [1]. In addition to the low work function, the main reason for the enhanced SE yield in alkali halides is that the low energy electron transport is dominated by electron-phonon interactions. These are characterized by exceptionally low energy losses when the electrons are slowed down, below the energy gap E_g . Furthermore, these energy losses, typically of a few tens of meV, can be partially compensated by energy gains of the same order.

Fröhlich [2, 3] developed a theoretical model for calculating the scattering rate of an electron interacting with the longitudinal optical modes of the lattice vibrations of an ionic crystal. His model is based on the time-dependent perturbation theory and has been widely used in other works. After the pioneering works of Dekker [4] and of Khokley et al. [5], Llacer and Garwin [6] used Fröhlich's theory to calculate the low energy electron transport in ionic solids by a direct Monte Carlo simulation. They assumed that the longitudinal optical phonon-electron (LOPE) interaction is predominant in the low energy electron transport and neglected the elastic scattering. They calculated the values of some parameters related to low energy electron transport and emission, particularly the escape length L_s , for several alkali halides. However, the predicted L_s values were found to be larger than experimental ones but no reason for this discrepancy was given.

Henke et al. [7] proposed a model based on Kane's "random walk" theory [8]. This model predicts SE emission (SEE) spectra, in good agreement with the experimental data of the same group. However, these authors did not consider the different inelastic interactions separately, but rather treated them as a single effective inelastic process.

Recently, the low energy electron transport in SiO_2 has been investigated, and both LOPE and acoustic phonon-electron (APE) interactions were taken into account [9, 10, 11]. In this material, it was found that the APE scattering is indeed the main inelastic process for electron energies higher than E_{BZ} (i.e. the energy of an electron having a wavenumber equal to the crystal first Brillouin zone edge). For low energy electron transport under an electric field, the inclusion of the acoustic process causes the mitigation of the electron velocity runaway. This phenomenon could be observed in experiments but

was not predicted by models which include only LOPE interactions, like that of Fitting et al. [12]. The APE scattering was considered so far as a quasi-elastic process, which randomizes the direction of motion of the electrons [13].

In recent years, Ausmees et al. [14, 15] used a Monte Carlo simulation for calculating the XUV-induced electron photoemission in NaCl and KBr. They also included in their model both LOPE and APE scatterings and adjusted unknown model parameters in order to fit experimental quantum yields for both materials. They concluded that for electron energies above 2-3 eV, the electron transport is mostly dominated by APE interactions.

Adamchuk et al. [16] used the diffusion equation and calculated the spectra of photoelectrons, with energies below E_g , induced by UV-photons in CsI and escaping into vacuum. They included the conduction band structure in their calculations and assumed that the LOPE interaction is the principal scattering mechanism in the electron slowing-down process.

In previous works, Akkerman et al. [17, 18] proposed a theoretical model and performed Monte Carlo simulations of SEE induced from CsI by X-rays and electrons in the energy range of 1-30 keV. The model was further extended up to 100 keV [19, 20]. The authors used, at low SE energies, elastic scattering in addition to the LOPE process. Their integral results, namely the total emission yields, agree fairly well with experimental data [19] but the SE spectra do not match existing experimental results. Hence some questions concerning the low energy electron transport in alkali halides remained open.

In the present work, we propose a microscopic model which includes both electron-phonon interaction processes. We present the results of Monte Carlo calculations of electron transport in CsI, NaCl and KCl, in the energy range below E_g . A comparison with experimental data for CsI shows that our present model permits precise calculation of both integral and differential characteristics of low energy electron transport and emission in alkali halides.

2 Model Description

Our present approach to electron transport in alkali halides is based on a model of low energy ($E < E_g$) electron interactions. It assumes that the electrons move along stochastic trajectories as classical particles. However, their interactions in the solid are described

by quantum mechanically calculated cross sections. As mentioned in the introduction, we assume that the transport of such electrons is governed by electron-phonon (both optical and acoustic) and elastic scatterings. Moreover, we consider the material as a perfect single crystal, although thin evaporated alkali halide films, generally constituting photoconvertors, are polycrystalline [21]. The problem of electron transport in covalent crystals, including the same processes and taking into account solid structure effects, has been discussed in more details by Jacoboni et al. [22].

2.1 The longitudinal optical phonon-electron interaction

The inverse mean-free-path (IMFP) for the LOPE interaction has been calculated following the method of Llacer et al. [6] and is given by:

$$\frac{1}{\lambda_{opt}^+} = \frac{e^2 m^*}{2\hbar^2} (n_q + 1) \frac{\hbar\omega_{opt}}{E} [1/\epsilon_\infty - 1/\epsilon] \ln\left(\frac{1+A}{1-A}\right), \quad (1)$$

if the electron loses energy to the lattice by phonon emission (i.e. phonon creation) or by:

$$\frac{1}{\lambda_{opt}^-} = \frac{e^2 m^*}{2\hbar^2} n_q \frac{\hbar\omega_{opt}}{E} [1/\epsilon_\infty - 1/\epsilon] \ln\left(\frac{B+1}{B-1}\right), \quad (2)$$

if the electron gains energy from the lattice by phonon absorption (i.e. phonon annihilation).

In formulae (1) and (2), the expressions defining A and B are:

$$A = \sqrt{1 - (\hbar\omega_{opt}/E)} \quad (3)$$

$$B = \sqrt{1 + (\hbar\omega_{opt}/E)} \quad (4)$$

and e is the electron charge, m^* is the electron effective mass in the conduction band, $\hbar\omega_{opt}$ and E are the energies of the optical phonon and of the interacting electron respectively. ϵ_∞ and ϵ are the optical and the static dielectric constants. n_q is the phonon occupation number:

$$n_q = n(q_{opt}) \quad (5)$$

where q_{opt} is the phonon momentum and $n(q)$ is the Bose-Einstein function:

$$n(q) = \frac{1}{\exp(\hbar\omega(q)/k_B T) - 1}, \quad (6)$$

$\omega(q)$ and k_B being the phonon frequency and the Boltzmann constant, respectively. The angular deflection distribution, $g(\theta)$, for the electron after a LOPE interaction is also given in [6]:

$$g(\theta) = D \frac{E'^{1/2} \sin \theta}{E + E' - 2(EE')^{1/2} \cos \theta} , \quad (7)$$

where θ is the scattering angle, D is a normalization constant and E' is the electron energy after the collision, i.e.:

$$E' = \begin{cases} E - \hbar\omega_{opt} & \text{for phonon emission} \\ E + \hbar\omega_{opt} & \text{for phonon absorption} \end{cases} \quad (8)$$

In this work, the dielectric constants, ϵ_∞ and ϵ , and the optical phonon energy, $\hbar\omega_{opt}$, are taken from [23] and [6] respectively.

2.2 The acoustic phonon-electron interaction

For this interaction, we have used the description of Ashley et al. [24]. They followed the treatment of Sparks et al. [9] as it has been applied to SiO₂ by Fischetti et al. [10, 25]. The IMFPs for this interaction are:

$$\frac{1}{\lambda_{ac}^+} = \frac{3S^2}{4\pi M_p N_c \hbar v^2} \int_0^{q_{max}^+} dq \frac{q^3}{\omega(q)} f(q) [n(q) + 1] (1 - \hat{k} \cdot \hat{k}')_+ \quad (9)$$

and:

$$\frac{1}{\lambda_{ac}^-} = \frac{3S^2}{4\pi M_p N_c \hbar v^2} \int_0^{q_{max}^-} dq \frac{q^3}{\omega(q)} f(q) n(q) (1 - \hat{k} \cdot \hat{k}')_- \quad (10)$$

where the subscripts refer to phonon creation (+) and annihilation (-).

v is the velocity of an electron of energy E , $n(q)$ is given by (6) and $(\hat{k} \cdot \hat{k}')_\pm$ is the cosine of the deflection angle of the electron in the collision:

$$(\hat{k} \cdot \hat{k}')_\pm = \frac{1 - q^2/2k^2 \mp \hbar\omega(q)/2E}{\sqrt{1 \mp \hbar\omega(q)/E}} . \quad (11)$$

q_{max}^+ and q_{max}^- can be easily obtained from the conservation laws of energy and momentum:

$$q_{max}^\pm = \begin{cases} 2k \mp 2m^* C_s / \hbar & q < k_{BZ} \\ k[1 + \sqrt{1 \mp \hbar C_s k_{BZ} / E}] & q \geq k_{BZ} \end{cases} \quad (12)$$

In this model, the three acoustic modes are treated as an equivalent branch with a sound velocity C_s . S is the interaction Hamiltonian matrix element, M_p is the primitive

cell mass, $f(q)$ is the primitive cell mass correction function [24] and N_c is the density of primitive cells in the crystal.

The phonon dispersion relation is approximated by:

$$\omega(q) = \begin{cases} C_s q & q < k_{BZ} \\ C_s k_{BZ} & q \geq k_{BZ} \end{cases} \quad (13)$$

where k_{BZ} is the sphere radius which has the same volume, in phase space, as the first Brillouin zone [25].

There is a high probability for large angular deflections to occur in the APE interaction [10]. This is different from the LOPE process where forward deflections are largely favored. In this work, we assume isotropic angular deflections after the acoustic interaction, as discussed below (2.3).

Some parameters like C_s , m^* and S are not well-known. We assume that the effective mass of the electron, m^* , is equal to the free electron mass like in Refs. [10, 24]. The value of S is obtained following the scaling method of Sparks et al. [9], where the cross section for electron scattering from the alkali halide molecule is approximated by the scattering cross section from the halogen ion. C_s is calculated according to [9, 10, 24] through:

$$\frac{3}{C_s} = \frac{2}{C_t} + \frac{1}{C_l} \quad (14)$$

where C_t and C_l are the transverse and longitudinal sound velocities of the corresponding alkali metals [26]. The acoustic phonon energy, $\hbar\omega_{ac}$ is obtained by averaging the phonon energy over electron energies.

The values of the parameters used in our calculations are summarized in table I. APE and LOPE calculated interaction mean-free-paths are presented in Fig. 1 versus the electron energy, for CsI, NaCl and KCl. As can be observed, the APE scattering mean-free-path continues to decrease in a non-physical way, even at electron energies above the gap. A similar discrepancy has been recently underlined by Bradford et al. in SiO₂ [13]. In order to overcome this difficulty, they introduced a pseudo-potential that takes into account the atom screening at energies higher than E_g , leading to a mitigation of the APE cross section, that approaches the elastic one at these energies.

2.3 The elastic scattering

The elastic scattering of electrons in solids, in the considered energy range, is far from being clearly understood. Evidently the "free atom approximation" of the interaction potential, which is widely used at higher energies, is not valid for the low energy electrons. Therefore, many-body interactions in the solid have to be taken into consideration. For example, in the case of Al, a self-consistent muffin-tin potential [27] has been used, showing a large difference between the elastic mean-free-paths calculated by both methods at $E = 10\text{eV}$. No such muffin-tin potential exists for other monoatomic solids. The situation becomes even more complicated in the case of alkali halides, which are ionic crystals and where the halogen atom can preferentially influence the scattering process. There are no direct experimental measurements of the elastic cross sections in solids. The only known experiments, done with solid Xe, provide a large scatter of elastic cross section data [28]. The estimated cross sections for solid Xe are summarized in table II. The experimental cross section for gaseous Xe [29] is also given for comparison. These data are relevant to this work since the atomic number of Xe is the mean value of that of CsI.

The cross sections calculated from the mean-free-paths for the acoustic phonon interaction in CsI (see Fig. 1a) are in the range of $1.8 \cdot 10^{-15} - 1.2 \cdot 10^{-14}\text{cm}^2$, i.e. larger than the experimental cross sections in solid Xe. Thus the dominant scattering process will be the APE interaction. The fact that the maximal energy transfer in such a process is small ($\hbar C_s k_{BZ} \ll E$, where E is the electron energy) and that the angular distribution of the scattered electrons is practically isotropic justifies the neglect of the elastic scattering from the simulation model, at least at energies $E > 2\text{eV}$. In addition, as it follows from [28], at $E < 1.5\text{eV}$, the mean-free-path for the elastic process drastically increases, which further justifies its neglect in this range. It should be noted that this simplification could be valid only if the distance d , along which the electron propagates in the solid, is sufficiently long to complete the randomization due to the isotropic scattering. d should be larger than $5\lambda_{ac}$. An even larger value is required, if we take into account the fact that the angular distribution for the acoustic process is not exactly isotropic. Nevertheless, the investigation of the elastic scattering will continue in order to get a more complete picture of all processes involved in the low energy domain.

3 Results

The presented results were obtained using our simulation code on a PC-486. The number of tracked electrons (or trajectories) used in the simulations has been selected in each case as to get fair statistical precision, as discussed below.

3.1 Transport and emission of UV-induced low energy electrons

As a first test of our model, we have considered the energy spectrum and the quantum efficiency of electrons photoproduced by monochromatic UV-photons, in the energy range of 6.3 to 8.6 eV, on a CsI layer. The lower energy limit is the sum of the energy gap and the affinity, ϕ (in CsI, E_g and ϕ are 6.2 eV and 0.1 eV respectively [30]). It is the theoretical threshold energy for electron emission. The upper limit was chosen to be less than $2E_g$ in order to avoid SEE induced by the photoelectrons. The value 8.6 eV also ensures, according to Distefano et al. [31], that exciton influence on the spectrum is negligible. We have assumed that the UV-photon is absorbed only through an interband transition.

In the first step of the simulation, the photon absorption depth is selected according to recent experimental mean absorption depths [32]. In order to save computation time, the "forced Monte Carlo" scheme [33] is used. In a second step, the initial electron energy and direction are chosen. The electron has the initial energy:

$$E_{init} = \hbar\omega - E_g - \chi, \quad (15)$$

where χ is randomly sampled from a normal distribution (see [17]) The initial direction of the electron is selected isotropically. The electron trajectory is followed step by step where, at every step, the collision process is selected randomly according to the relative IMFPs of the different elementary interactions, calculated by the model described above. The scattering angle of the electron is randomly chosen either from the optical angular distribution (7) or from the isotropic acoustic one, depending on the kind of interaction. The electron is followed until its energy becomes lower than ϕ (in this case it is considered as lost) or until it reaches the back or the front CsI surface. In the latter case, the electron

is counted as transmitted or backscattered, respectively, only if its angle of incidence to the surface satisfies the "escape cone" condition (see formula (3.21) in [34]).

Our first goal was to calculate the energy spectra of electrons induced by UV-photons in the energy range 6.8 - 8.4 eV and emitted from an 11 nm thick semitransparent CsI photocathode. The number of simulated trajectories was 10^5 in this case. Fig. 2 presents our results in comparison to experimental data of Distefano et al. [31]. Although the energy resolution of their spectrometer was 0.3 eV, there is a good agreement between the two sets of data. The systematic deviation of the experimental results from the simulated ones, at photon energies above 7.6 eV can be explained by the omission of the density of states of the conduction band from our calculations. The consideration of the density of states starts influencing the emission spectra at the mentioned energies, as was shown in [31, 35], shifting the peak position to higher energies and increasing the distribution width.

We have calculated the quantum efficiency of photoemission from both reflective and semitransparent CsI photocathodes. The number of trajectories was 10^4 , providing a statistical uncertainty of about 2 %. Fig. 3 shows the calculated quantum efficiency of a 500 nm thick reflective photocathode versus the photon wavelength, compared to experimental results of Breskin et al. [36]. The fluctuations, observed in the calculated quantum efficiency, could result from the oscillations in the experimental, wavelength-dependent, absorption length [32]. We would like to note that the reflection from the vacuum-CsI interface was not taken into account in our calculations. Using the approximate refraction index relation, provided in [37], would result in a reduction of our calculated quantum efficiency by the order of 15 %. This is within the systematic uncertainty of the experimental data. Fig. 4 presents the quantum efficiency of a semitransparent photocathode as a function of its thickness, irradiated at 170 nm and 180 nm, in comparison to the experimental results of Lu et al. [32]. In this case, the authors included most corrections in their experimental data, considering the reflection at the Cr-vacuum interface. An additional correction, due to the Cr-CsI interface, of the order of 3 % can be neglected. As can be seen from figures 3 and 4, in both cases the simulation results agree well with experimental data.

3.2 The escape probability

We have calculated the escape probabilities for electrons as a function of their initial energy and creation depth within the photocathode. The calculations were done for three alkali halides: CsI, NaCl and KCl. The procedure is as follows: an electron is created at a certain depth from the exit surface; its initial direction is isotropically chosen and its transport and emission are followed as described above. In these calculations, the number of tracked electrons was 5×10^3 for each calculation, providing a statistical uncertainty of about 3 %.

The calculated escape probabilities for electrons of 1, 2 and 6 eV, versus the electron creation depth, are summarized in figures 5-7, for the three photoconvertors. Also shown are the previously calculated values for CsI and KCl by Akkerman et al. [17, 18]. One can see that the agreement between both sets of calculations is rather good at 6 eV. At lower energies, the escape probability is higher compared to the one calculated in [17, 18]. The reason for this discrepancy is the following: in the previous works, a constant electron elastic cross section was assumed over the whole energy interval $E < E_g$; here, the cross section of the APE interaction, treated as a quasi-elastic process, decreases with the electron energy.

4 Discussion

From the results presented above, it appears that our model describes well the low energy electron transport in alkali halides. Despite of uncertainties in the values of some parameters used and the exclusion of both conduction band structure and exciton states, it should be noted that the calculated energy spectra and quantum efficiencies are in good agreement with experimental data of different sources. Therefore our model provides an important tool for calculating quite accurately integral and differential emission characteristics and for optimizing some experimental parameters, like for example the photoconvertor thickness.

The difference at lower electron energies between the escape probability calculated in the present and previous [17, 18] works also affects the shape of the calculated SEE spectrum, giving more weight to the low energy electrons (around 1 eV) as was indeed

observed experimentally. As in Ref. [17], it can be noted here that the escape probability does not have a pure exponential dependence on the electron creation depth, contrary to what has been supposed by several authors. Thus, the simple approach of a constant escape length, estimated to be of the order of 22 nm in CsI by Fraser [38], could be considered as a crude approximation. The presumed exponential behaviour of the escape probability is derived from the assumption that the low energy electron transport is similar to a diffusion process. However, the main condition required by the diffusion approach, as formulated in [39], is that the mean-free-path for the phonon-electron interaction is independent of the electron energy. This condition is obviously not valid as can be seen in Fig. 1.

Henke et al. [7, 40] developed a model for estimating SEE yields and energy spectra induced by soft X-rays. In the low energy range, they used Kane's "random walk" model. Their work was based on two principal assumptions:

- The electrons generated at a given depth from the exit surface lose, before reaching the surface, an average energy that is proportional to this depth.
- The variance of the electron arrival energy distribution can be neglected.

Using our model, we checked both assumptions in the case of CsI for several initial electron energies as follows: we estimated the mean electron energy loss and the electron energy variance within thick slices, across the convertor depth (the slice has to be thick enough in order to contain a large number of interactions). As can be seen in Fig. 8 for an electron with an initial energy of 3 eV, the mean differential energy loss is obviously not a constant function of the depth, thus the first assumption is not valid. Moreover, the calculated energy variance was found to be of the order of an eV and can indeed not be neglected, in contrary to the second assumption.

Therefore, models based on the diffusion approximation, including those derived from Boltzmann's kinetic equation, are less accurate than direct Monte Carlo calculations. The latter seem to be the most adequate approach to elucidate electron transport and emission even at energies below the energy gap.

5 Conclusions

We have described a model for the transport of electrons with energies lower than the energy gap in alkali halides, taking into account both optical and acoustic phonon-electron interactions.

Monte Carlo simulations based on this model have been performed for estimating some parameters of secondary electron emission induced in CsI by UV-photons. Both integral and differential calculated characteristics, like electron energy spectra and quantum efficiency, agree fairly well with existing experimental photoemission data. The acoustic phonon-electron interaction plays an important double role: randomizing the electron trajectories, similarly to an elastic scattering, and introducing additional energy losses.

Considering the dependence of the electron escape probability on its creation depth, we have shown that the diffusion approximation, often used for low energy electron transport calculations, is not well established and is therefore limited when performing quantitative estimations. Our low energy electron transport model is presently incorporated into the previous model of Akkerman et al. [17] for investigating the secondary electron emission induced from alkali halides by X-rays or energetic electrons.

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Figure captions

- Fig. 1** Acoustic and optical phonon-electron interaction mean-free-paths calculated with the present model, versus electron energy in: (a) CsI, (b) NaCl and (c) KCl.
- Fig. 2** The peak position (a) and the FWHM value (b) of UV-induced SEE energy spectra, versus photon energy. Shown are our calculations (full curves) and experimental data (Ref.31, dashed curves).
- Fig. 3** The quantum efficiency of a 500 nm thick reflective CsI photocathode versus the UV-photon wavelength. The hollow symbols and the dashed curve are the respective calculated and measured (Ref. 36) data.
- Fig. 4** The quantum efficiency of a semitransparent CsI photocathode versus its thickness, at photon wavelengths: (a) 170 nm and (b) 180 nm. The full curves are our simulation results and the dashed ones are the experimental data of Lu et al. (Ref. 32).
- Fig. 5** Calculated escape probability for electrons with initial energies of 1, 2 and 6 eV as function of their creation depth, in CsI. Full curves are the results of the present work and dashed or dotted ones are the results of previous calculations (Ref. 17).
- Fig. 6** Escape probability as calculated by our model in NaCl, versus electron creation depth at various initial electron energies.
- Fig. 7** Calculated escape probability in KCl as a function of creation depth. Full and dashed curves present the results of respective present and previous (Ref. 18) calculations.
- Fig. 8** Calculated mean differential energy loss versus the electron distance from the entrance surface, z , in a 100 nm thick CsI photocathode. The electron creation distance and energy are 40 nm and 3 eV. The differential energy loss was estimated within 5 nm thick slices. The mean energy loss at a given distance z refers to the energy lost in average by an electron between $(z-5)$ and z .

Table I: Parameter values used in the simulation

Material	ϵ_∞	ϵ	C_s [m/s]	S [eV]	$\hbar\omega_{opt}$ [eV]	$\hbar\omega_{ac}$ [eV]	ϕ [eV]
CsI	3.	5.65	696.	1.	0.010	0.003	0.1
NaCl	2.25	5.90	1952.	1.8	0.033	0.013	0.5
KCl	2.10	4.85	1492.	1.8	0.026	0.009	0.5

Table II: Elastic scattering cross sections in Xe

Energy [eV]	Solid Xe, experiment [cm ²]	Gaseous Xe, exp. [cm ²]	Estim. from the ionic radius [cm ²]
2 - 6	1.2 10 ⁻¹⁶ - 1.2 10 ⁻¹⁵		1.5 10 ⁻¹⁵
20		(2. - 2.3) 10 ⁻¹⁵

Fig. 1

Fig. 2

Fig. 3

Fig. 4

Fig. 5

Fig. 6

Fig. 7

Fig. 8