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MODELLING TRANSIENT PROCESSES IN THE COMPETITIVE PHENOMENA INDUCED BY HEAVY PROJECTILES IN MATERIALS FOR DETECTORS*

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Abstract. One of the clues in detection in modern physics is the development of detector materials able to produce simultaneously two or more different processes, as well as the measurement of the physical quantities associated to these processes in order to discriminate and to identify different particles. In the same time, the absorption of radiation in matter is not a continuous process and due to the interaction mechanism it is localized. As a result, for short time, transient processes are produced, some of them used in detection, other producing disorder that affects the properties of the detector material. In the present paper the thermal spike model is extended and used for semiconductors and liquefied noble gases used as target media for detectors. Both classes of materials are very promising for the detection of heavy particles in nuclear and astroparticle physics.

Key words: semiconductors, noble gases, transient processes, detection, degradation.

1. INTRODUCTION

The passage of heavy projectiles produces disturbances in the targets, which could be either transient or permanent. The energy released by the incoming particle as a result of the interaction with the electrons and/or the atoms of the medium is eventually found in “stable” lattice defects in crystalline materials and generally in defects in solids [1], is collected as electron – hole (ion) pairs in an applied electric field in all cases, is collected as light in the case of scintillators and of Cherenkov effect, etc. In all types of materials, part of this energy is transformed into heat. On the other side, part of the energy transferred to the target is used for detection, *e.g.* recording ionization, scintillation or even heat, and part of it produces damage.

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The study of the partition of the deposited energy into energy transferred to electrons and to atomic recoils in all target materials, together with the study of specific phenomena produced by electrons and /or atomic recoils would permit to reveal what are the common features and what are the peculiarities of the phenomena in different classes of materials. In particular, we are interested in this paper in crystalline semiconductors and in liquid noble gases as detector media, and we shall study comparatively the transient processes induced by the passage of heavy projectiles. In the next section, the energy lost by the incoming particle is divided into energy transferred to electrons and to atomic recoils. In each case the energy is transported from the interaction place. The mathematical description is given by a diffusion-type equation, and the interaction of the two subsystems could lead to a system of coupled equations. In section three, the peculiarities of the phenomena specific to semiconductors and to liquid noble gases will be presented.

2. ENERGY PARTITION

Following the illuminating article by Lindhard [2], we consider the division of the energy dissipated by the incoming particle, E, into energy ultimately given to electrons and energy ultimately left for atomic motion. The energy transferred to electrons is, for ejected electrons, the kinetic energy plus the original binding, while for excited electrons it is the excitation energy. Correspondingly, the energy transferred to atoms excludes the internal excitation of atoms.

The partition of the energy loss into these two channels depends on the electronic and nuclear stopping cross sections (stopping powers).

In fluids, particularly in liquid noble gases, the energy transferred to atomic recoils is found as thermal energy, and does not contribute to ionization or excitation. On the other side, in the case of neutral, massive projectiles, the primary collision process produces recoils, and it is this energy which is subsequently partitioned. So, in these cases, the Lindhard curves for the self recoils are of interest.

In crystalline solids the lattice and the electronic system are coupled by electron-phonon interaction, which is of particular interest in the case of high electronic energy loss regime, where part of the energy transferred to electrons is ultimately found as lattice defects.

3. RESULTS

3.1. CRYSTALLINE SEMICONDUCTORS

At low energies of incoming particles, where the nuclear energy loss is much greater than the electronic energy loss, the transient phenomenon characterizing the damage is the formation of displacement spikes. Following Seitz, this is a limited volume with the *majority* of atoms temporarily in motion [3]. An important quantity in determining the spatial distribution of radiation damage is the average distance, or mean free path travelled by an energetic particle between primary recoils of energy greater than a given energy.

As examples, we present in Fig. 1 the mean free path between recoils for He, Si and Pb ions in a Si target. The results of the calculation show that at high energies all ions produce primary recoils that are well separated. As the ion energy decreases, the distance between recoils decreases and for Si and Pb ions it approaches the interatomic spacing of Si atoms in the target. So, as the ion slows down in the target, the mean free path decreases until a PKA is generated at every lattice site along the path. In the limit where the mean free path approaches the interatomic distance, the displacement cascade cannot be thought as a collection of isolated vacancies and interstitials, since a large number of defects are created in a very short time, in a limited region. It was suggested [4] that as the mean free path approaches the interatomic distance in the target, a highly damaged region is formed:

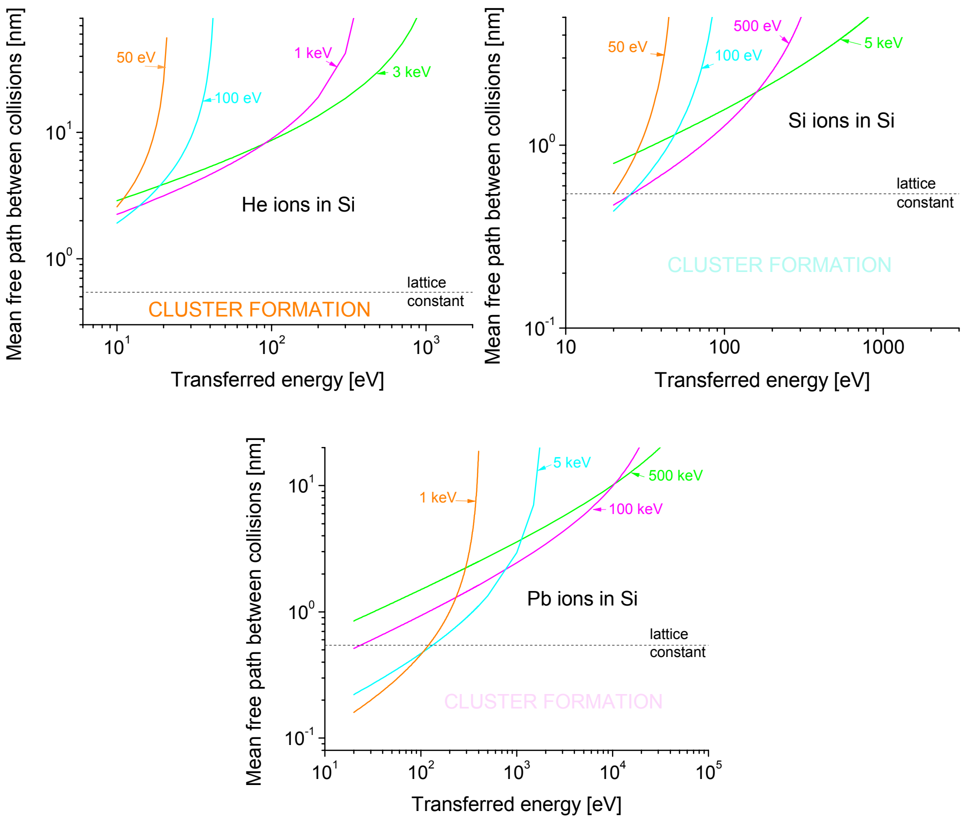

Fig. 1 – Mean free path between recoils of He, Si and Pb ions of different energies in a silicon target.

every displayed atom is forced away from the ion (or PKA) path, producing a volume of material composed of a core of vacancies surrounded by a shell of interstitials, referred to as a displacement spike.

The results of the calculations for He, Ge and Pb ions, of same energies, in a Ge target are presented in Fig. 2. From the examination of the figure, it could be seen that He ions produce well separated recoils at all energies, while for Ge and Pb ones al low enough energies the separation between recoils approaches the interatomic spacing in the target.

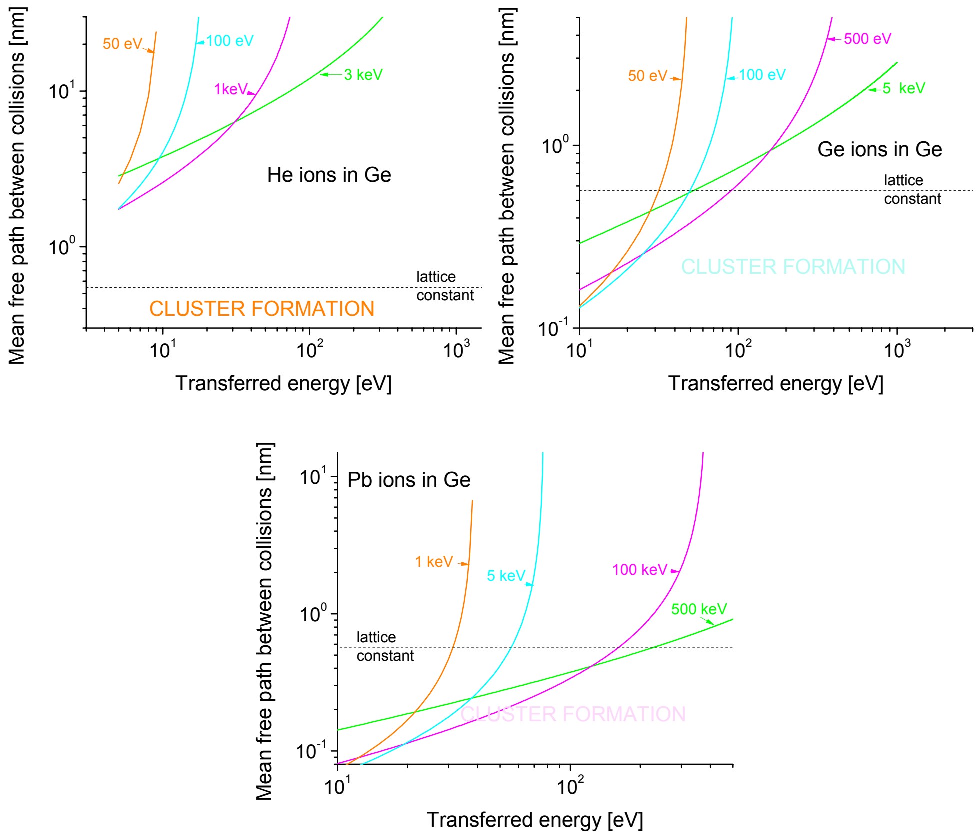

Fig. 2– Mean free path between recoils of He, Ge and Pb ions of different energies in a germanium target.

To gain an idea of the spatial distribution of the cascade in the region where the mean free path between recoils is less than the interatomic distance, we calculated the longitudinal and transversal moments of the spheroid containing the transport cascade [5]. A Si ion of 1 keV kinetic energy in Si has a range of around 35 Å, and the momenta of the spheroid are 13 and 12 Å respectively. In this region, the energy per atom is ~ 2.5 eV. The slowing-down time of this projectile is of the order of $8\cdot10^{-14}$ s. Subsequent energy dissipation among recoil atoms is characterized by a time constant of the order of $1.2\cdot10^{-13}$ s.

At the other extremum, of high kinetic energies of incoming particles, the main mechanism of energy loss is ionization.

The high electronic energy loss regime in metals has been treated in the frame of the thermal spike model from the beginning of the nineties [6], in the aim to explain the track of the ion as due to the modifications of the structural characteristics of the material. The model has been successively extended to insulators [7] and eventually to semiconductors [8].

At intermediate energies, where electronic and nuclear energy losses are comparable, both of them must be considered, as well as the coupling between the electronic system and the lattice. The behaviour of Si under ion irradiation has been quantitatively analysed in Ref. [9], in the frame of a thermal spike model, which takes for the first time into consideration both ionization and nuclear energy losses. The results evidence also the influence of the coupling between the subsystems, on the case of heavy ions, where a peak or shoulder is seen in the time distribution of the electronic temperature, in correspondence to the maximum atomic excitation.

In Fig. 3, the electronic and atomic temperature profiles produced by 600 keV Xe, 5GeV Fe and 5 GeV Ar ions in Si are represented, as calculated in the present work. We would like to underline that in the case of Xe ions of 600 KeV, the nuclear stopping power surpasses the electronic stopping power, while in all other cases studied here we are in the case of high electronic energy loss regime.

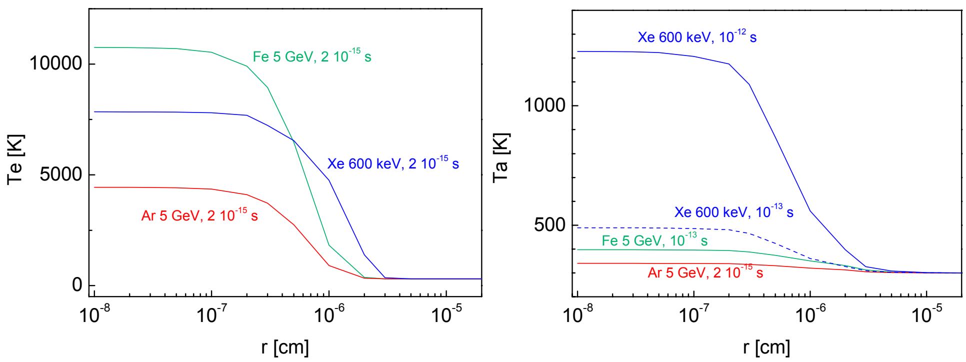

Fig. 3 – Electronic (left) and atomic (right) temperature dependence in silicon on the distance to the ion trajectory after the passage of the ion, at a time interval indicated in the legend.

It is interesting to note that in the case of 600 keV Xe irradiation, an important rise in the atomic temperature was obtained; in this case the components of the energy loss are: 1.89 keV/nm nuclear and 0.7 keV/nm electronic stopping powers respectively. It is much more efficiently communicated directly to the lattice system, and in fact the time development of the thermal spike differs in respect to Fe and Ar irradiation: the highest temperature is attained after 1ps after the passage of the ion, while in the case of Fe and Ar ions this is attained after 0.1 ps. In this last situation, the nuclear energy loss is negligible, and the electronic energy loss is 0.97 and 0.384 keV/nm respectively.

3.2. LIQUID NOBLE GASES

As explained in section 2, in fluids only the energy transferred by an incoming particle to the electronic system (or part of it) could be at the origin of radiation damage.

As stressed by Seitz [10], this energy could reappear in different forms, as scintillation or/and Cherenkov radiation – see for example Ref. [11]. Another part, used also in detection, is related to the generation of electron – ion pairs, and to their collection: ionization [12]. But in all cases, in the balance of the energy transmitted by the nuclear recoils (appearing as a result of elastic scattering of the incoming particle in the fluid) to the electronic system, a part of energy could be found as heat. Detectors having liquefied noble gases as working materials operate in the vicinity of the liquid – gas transition on the phase diagram, and the remaining energy could be used in bubble production, *i.e.* in the phase transition.

In the case where scintillation could appear, it is produced / used for detection. The detection process includes the absorption of radiation and conversion of absorbed energy into a number of photons, the collection of scintillation photons at the photodetector and the conversion of the photons into photoelectrons. If the incoming particle is a neutral one, the nuclear recoils produced in the primary interaction lose energy by interactions with electrons and nuclei of the fluid. The energy transmitted by the recoils towards the electronic system of the fluid is used to produce excitons and electron-ion pairs along the track. Free excitons collide with ground state atoms and form excited molecules (excimers) through the quasichemical reaction:

$$X^* + X \rightarrow X_2^*,$$

where X stands for any type of noble liquid (Ar, Xe, Kr, Ne, He). Free ions undergo collision, recombination and deexcitation processes:

$$\begin{aligned} &X^+ + X \rightarrow X_2^+ \\ &X_2^+ + e^- \rightarrow X^{**} + X \\ &X^{**} \rightarrow X^* + \text{heat} \\ &X^* + X \rightarrow X_2^+ \end{aligned}$$

to form excimers. The excimers then decay radiatively from the lowest-excited molecular states $^1\Sigma_u^+$ and $^3\Sigma_u^+$ to the repulsive ground state $^1\Sigma_g^+$, with different decay time constants.

The equations satisfied by the concentrations of excimers are also of diffusion type, this phenomenon being also modelled in the frame of the theory of diffusion limited chemical reactions.

The calculation of the relative scintillation response of scintillators to an ionizing particle (which could be a self-recoil) must consider: the fraction of the energy lost which is transferred to the electronic system, the quenching produced by biexcitonic collisions [13, 14], and also the *escaping electrons* [15]. These are electrons produced by ionization that thermalize outside the Onsager radius and become free from recombination even in the absence of an electric field. In Reference [15], the model has been applied to Xe, and has been found as reasonably agreeing with experimental data.

In Fig. 4, the initial radial distribution of excitons in the cylindrical track core produced by various particles in LXe, as estimated in Ref. [14], are represented. These are the same ions and of the same energies for which the calculations presented in Figure 3 were performed in Si. The distribution is modeled with a Gaussian with a size parameter the same as the core radius, given by the Bohr-criterion: $r_0 = \hbar v / 2E_1$, where $\hbar$ is Plank's constant divided by 2π, v is the velocity of incident ion and E_1 is the energy of lowest electronic excited state of the medium, and the pre-exponential factor was calculated using the averaged electronic energy loss.

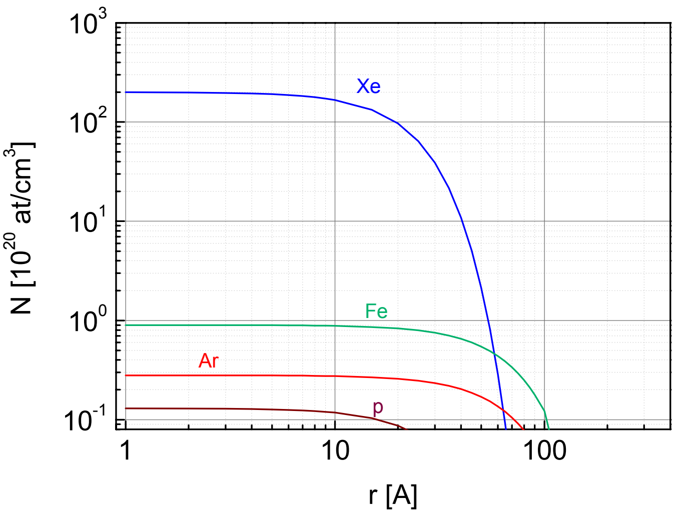

Fig 4 – Calculated distribution of excited atoms in cylindrical track created by Xe, p, Fe and Ar ion irradiation of LXe.

Consequently, in the case of neutral massive particles incoming on liquid noble gases, all previously mentioned phenomena are produced by the atomic recoils, more specifically by the fraction of their energy communicated to the electronic sub-system. The division of the recoil energy between transfers to electrons and to atomic recoils is the solution of the Lindhard integro-differential equation [2], for particles identical with the medium, for uni-component media. Different parameterisations have been used in the literature for their calculation (see Refs. [16] for an example). We have calculated these curves using the simplifying hypotheses presented in Ref. [17], and the results are presented in Fig. 5, for Xe selfrecoils in LXe, as the dependence of the energy deposited into the electronic system on the kinetic energy of the recoil.

The energy deposited into excitation has been calculated from scintillation measurements, and is represented on the same figure as calculations based on the model in Ref. [15].

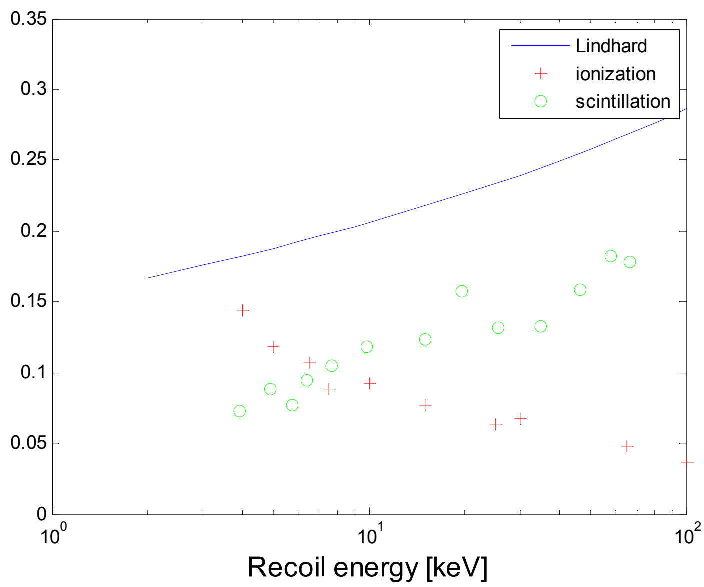

Fig. 5 – Fraction of the energy transferred to the electronic system by self-recoils in Xe (continuous line); fraction of the energy used for scintillation: (exp. data); fraction of the energy used for ionization (exp. data).

The energy used for the formation of electron-ion pairs has been estimated from measurements of ionization yield (number of electrons escaping recombination per unit recoil energy), and is also represented in the figure. The measurement of the energy deposited in the formation of electron-ion pairs supposes the measurement of the charge associated with an ionizing event, and is related to charge collection. Charge collection is affected by electron-ion recombination, which is high for heavily ionizing particles, and is also dependent on the applied electric field. On the other side, an electric field applied to LXe suppresses the electron-ion recombination and, thus reduces the scintillation yield. So, the data on scintillation and ionization are correlated and only data sets from simultaneous measurements might be considered.

However, the dispersion of data measured at different values of the electric field (0.10, 0.27, 0.73, 1.00, 2.00, and 2.30 kV/cm is not high, as seen from Fig. 14 of Ref. [15]. The analysis in Fig. 3 is performed using data collected independently on ionization and scintillation in LXe.

The energy in the electronic system which isn't used for ionization or scintillation remains available as heat, and could be used in bubble formation.

It was shown that liquid rare gases have a band structure [18]. Due to the large band gap, in the order of ten eV, they are excellent insulators.

For the formation of a bubble, which is based on the thermal spike model of Seitz [10] for fluids, there are two steps: nucleation of the bubble as a result of the evaporation of the fluid, and growth of the nucleus to a macroscopic bubble. First, an intense energy deposition along a particle's path can provide enough localized heating to create bubbles of a critical size or larger; than, if a vapor bubble grows larger than a critical radius (which is in the order of tens of nm), it becomes thermodynamically unstable and continues to expand evaporating the liquid. The main assumption is that the process is so rapid, that the energy is furnished from the interaction process, not from the ambient energy of the fluid.

The conditions necessary for radiation induced nucleation [19] are consequently related to the following steps:

– The total energy deposited must be larger than a critical energy for bubble formation, $E_c(T)$, computed from the sum of the thermodynamically reversible processes of vaporization, formation of bubble surface and bubble expansion against the liquid.

– The stopping power of the particle in the target material ($\mathrm{d}E/\mathrm{d}x$) must be such that $E_c(T)$ is deposited within a small distance $L(T)$ of order r_c :

The minimum energy required for bubble formation is:

$$E_c = 4\pi r_c^2 \left(\sigma - T \frac{\partial \sigma}{\partial T} \right) + \frac{4}{3} \pi r_c^3 \rho_v \frac{\lambda}{\mu} + \frac{4}{3} \pi r_c^3 P,$$

where σ – surface tension, P – operating pressure, T – operating temperature, ρ_v – saturated vapour density, λ – heat of vaporization per mole, μ – molecular mass, r_c – critical radius. This phenomenon will not be considered in detail in this paper. Different aspects were investigated by different authors; see for example Fladerera and Strey [20].

4. CONCLUSION

In the present paper, the problem of transient processes induced by the passage of heavy projectiles in materials for detectors, particularly in crystalline semiconductors and noble liquid gases, has been treated. In some cases, the competition between different mechanisms and the coupling between the electronic system and that corresponding to atomic recoils must be taken into consideration. In semiconductors, high electronic excitation could lead to defect production.

A better understanding of the microscopic processes of interaction is necessary as well as further clarifications on the mechanisms of degradation. Thus new ideas for detection would be possible.

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