CALCULATION OF DISPLACEMENTS PER ATOM DISTRIBUTIONS IN SOLID MATERIALS

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Abstract

The results of the calculations of the displacements per atom distribution induced by the gamma irradiation on $YBa_2Cu_3O_{(7-x)}$ superconductor and Iron slabs up to 15 MeV are presented. Firstly, a calculation procedure for the displacements cross sections and the displacements per atom distributions was introduced, relaying on the application of the energy flux distribution values of secondary electrons, formerly calculated using the Monte Carlo methods based code system MCNPX. Finally, calculation results of displacements per atom distribution were compared with the corresponding energy deposition profiles, obtaining a nearly linear correlation among them at different depth positions for a wide range of Gamma Radiation incident energy.

Key words: Monte Carlo method, atomic displacements, gamma radiation, simulation, superconductors, iron, cross sections

CÁLCULO DE DISTRIBUCIONES DE DESPLAZAMIENTOS ATÓMICOS EN MATERIALES SÓLIDOS

Resumen

Se presentan los resultados de los cálculos de la distribución de desplazamientos atómicos en el superconductor $YBa_2Cu_3O_{(7-x)}$ y en láminas de hierro inducida por radiación gamma hasta 15 MeV. Se introdujo un procedimiento de cálculo para las secciones eficaces de desplazamiento y para las distribuciones de desplazamientos atómicos, basado en el uso de los valores de la distribución del flujo energético de los electrones secundarios, obtenidos con ayuda del código de cálculo MCNPX basado en el método de Monte Carlo. Se compararon los resultados de los cálculos de la distribución de desplazamientos atómicos con los perfiles de energía depositada, obteniéndose una correlación casi lineal entre ambos a diferentes profundidades en las muestras para un amplio intervalo de energías de la radiación gamma incidente.

INTRODUCTION

Nowadays, the damage induced by the different types of radiations in advanced materials is broadly studied. Mainly, those materials that get involved in experiments and development of new technologies, like they are the high temperature superconducting materials (HTSC), semiconductors, metals, on the base of which are essentially constituted the radiation detectors, particle accelerators, nuclear reactors, etc. In this sense, present investigation tries a study of two kinds of materials used with these ends, the ceramic superconducting material $YBa_2Cu_3O_{7-x}$ and the metallic iron ones, through the mathematical simulation of the physical processes that take place in these solid materials when being irradiated, in a wide energy range that goes from some keV to 15 MeV. For it, the program MCNPX [1] is used, with the help of which the physical necessary magnitudes are determined to calculate the distribution of the Atom Displacements (AD) inside these materials, by means of the implemented algorithm.

Previous works involving AD determination on HTSC materials reported in [2-5] for electron fluxes and in [6,7] for the Gamma Radiation, led to first attempts for the overall estimation of its magnitude as a measurement of the potential microscopic structural modifications induced by the Gamma Radiation Damage on these materials.

However, in spite of the foregoing statement, the AD calculation approach presented by Belevtsev et al. [7] has a restricted character and meaning, since it follows the classical AD calculation procedure introduced by Oen-Holmes [8] (see also [9]), which does not take into account the shower and cross linked nature of gamma quanta and secondary electron interactions arising at Gamma Radiation transport in solids. This Gamma Radiation complex stochastic behavior can be nowadays, theoretically, very well simulated and described by means of calculation codes based on the Monte Carlo method, as for instance the MCNPX, EGSnrc [10], GEANT4 [11], modeling the transport of different types of radiations in the substance. An attempt introducing a direct Monte Carlo methods based simulation of AD

distribution induced by the Gamma Radiation on Iron slabs has being reported by Sato *et al.* [12].

Then, it seems to be justified the application of the standards outputs (tallies) of the available Monte Carlo methods based simulation codes, cited before, as an alternative way providing a posteriori realistic approach to the subject of Gamma Radiation Damage calculations. Following this last idea, the present work tries a first attempt to calculate AD rate distributions in solid materials involving Gamma Radiation transport properties, an approach to Gamma Radiation Damage determination, closer than previous reported works [7,13,14].

Thus, a calculation procedure was introduced, starting from the calculation code outputs of the energy flux distribution values of secondary electrons, which are then processed by the application of standard expressions describing the electron Primary Knock-on interactions leading to AD events. This procedure was thus finally applied for the calculation of AD distribution at different incident Gamma Radiation energies, regarding the plane crystalline sites on the YBCO superconducting material and the metallic iron ones.

Atom Displacements Cross Sections

The incident Gamma Radiation produces secondary electrons in the material. These electrons can remove an atom from the lattice through elastic scattering processes. The removed atom is the so called Primary Knock-on Atom (PKA) and the corresponding cross section, σ_{PKA}, was obtained starting from the McKinley-Feshbach approximation [15] (see Eq.(2)). If any of these recoil atoms has a kinetic energy above the displacement threshold energy T_d, secondary atoms can be knocked-on by PKA and removed from the lattice too. The number of secondary displaced atoms can be calculated introducing the damage function $\nu(T)$. Then, the total number of displaced atoms per target atom can be obtained writing the displacement per atom cross section as follow

$$\sigma_{dpa} = \sigma_{PKA} \cdot \nu(T) \tag{1}$$

with
$$\sigma_{PKA}(E) = \frac{\pi Z_A^2 r_0^2}{\beta^4 \gamma^2} \left\{ \left(\frac{T_m}{T_d} - 1 \right) - \beta^2 \ln\left(\frac{T_m}{T_d} \right) + \pi\alpha\beta \left[2\left(\sqrt{\frac{T_m}{T_d}} - 1 \right) - \ln\left(\frac{T_m}{T_d} \right) \right] \right\} \tag{2}$$

and for the damage function we use the Kinchin-Pease model

$$\nu(T) = \begin{cases} 0, & T < T_d; \\ 1, & T_d \leq T \leq 2T_d; \\ T/2T_d & T > 2T_d; \end{cases} \tag{3}$$

where

Z_A is the atomic number of target atom,

r_0 is the electron classic radius $(= e^2/mc^2)$,

$\alpha = Z_A / 137$,

β is the ratio of electron velocity to light velocity,

$\gamma^2 = 1/(1 - \beta^2)$,

$T_m = 2E(E + 2mc^2)/Mc^2$ (maximum kinetic energy of recoil atom),

E is the electron kinetic energy,

mc^2 is the electron rest energy,

M is the atom mass.

To evaluate the damage function, the obtained average atom recoil kinetic energy, T_{av}, was used, given by the expression:

$$T_{av} = \frac{1}{\sigma_{PKA}} \frac{\pi Z_A^2 r_0^2}{\beta^4 \gamma^2} \cdot T_m \left[\ln\left(\frac{T_m}{T_d} \right) - \beta^2 \left(1 - \frac{T_d}{T_m} \right) + \pi\alpha\beta \left(\sqrt{\frac{T_d}{T_m}} - 1 \right)^2 \right]. \tag{4}$$

The number of atoms displaced per electron was calculated following the expression proposed by Oen-Holmes [8].

$$N_{dpa}^e(E) = \int_{Ec}^{E} N_a \sigma_{dpa}(E') \frac{1}{(-dE'/dx)} dE',$$

(5)

where

$$E_c = \sqrt{(mc^2)^2 + \frac{Mc^2 T_d}{2}} - mc^2$$

(6)

is the cutoff kinetic energy of electrons in order to displace an atom from its crystalline site, N_a is the number of atoms in the unit of volume in the sample and $(-dE/dx)$ is the electron stopping power, calculated following the expression given by Bethe-Ashkin [16].

$$-\frac{dE}{dx} = 2\pi N_a Z_m^2 r_0^2 \left(\frac{mc^2}{\beta^2}\right) \left\{ \ln\left(\frac{mc^2 \beta^2 \gamma^2 E}{2I^2}\right) - \frac{1}{\gamma^2}\left[1 + (2\gamma - 1)\ln 2 + \frac{1}{8}(\gamma - 1)^2\right]\right\}$$

(7)

where Z_m^2 is the mean square atomic number of the sample material and is the mean excitation potential of the atom. The classical macroscopic number of displaced atoms per electron, $N_{dpa}^e(E)$, involves the calculation of AD events in a ¨*thick layer*¨ of thickness about the range of electrons in the material.

The dependencies of σ_{PKA} and σ_{dpa} on the electron kinetic energy are shown in figure 1, for the two studied materials. As can be observed in this figure, cross sections vanishes down certain value of electron kinetic energy, that in the case of oxygen atoms in YBCO, with $T_d = 20$ eV, takes a value above 130 keV. This means that electrons with energy lesser to this value can not produce AD in the Cu-O_2 planar sites. For the copper atoms in YBCO that energy is about 745 keV and for the iron atoms is near 626 keV.

Simulation with MCNPX code

The sample geometries and the irradiation conditions for calculations were similar as those used in a previously reported work by Piñera [17,18], as follows: the ceramic YBCO parallelepiped target was shaped with dimensions 2.8 mm x 16.0 mm x 1.5 mm (subdivided in voxells with 0.4 mm x 0.4 mm x 0.1 mm), on which impact 2 x 10[7] photons at different selected incident energies (in the wide range from 662 keV to
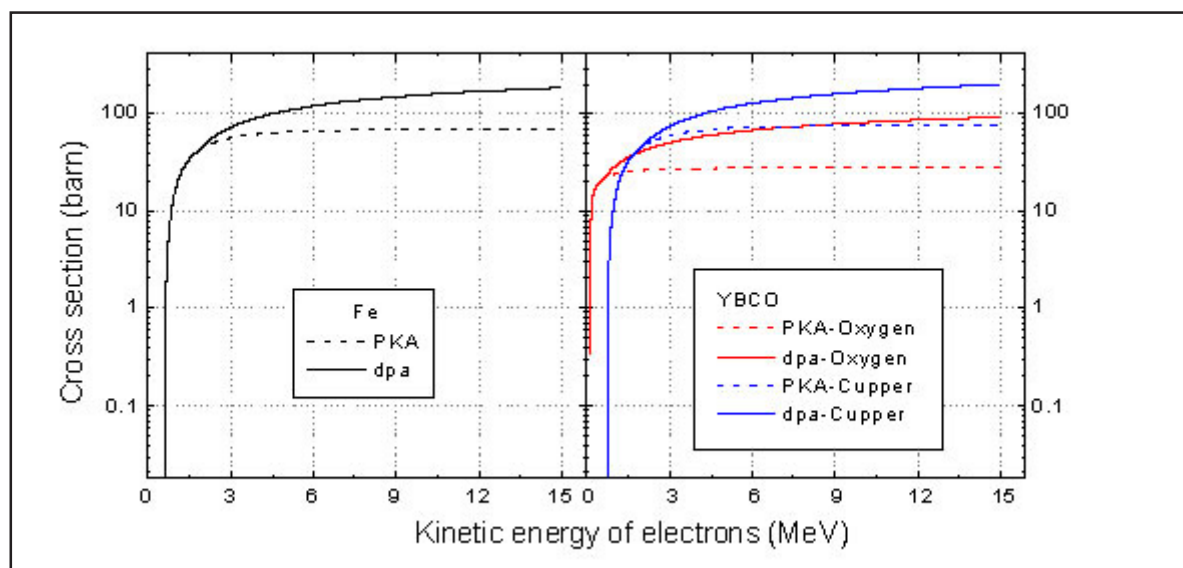

Figure 1. AD cross sections as function of electron kinetic energy. Continuous lines correspond to *dpa* cross section and dashed lines to PKA one's.

15.0 MeV). The simulations were carried out under the same number of independent histories for calculation purposes. In all the cases, the results shown are normalized to the total number of histories (incident photons and its secondary particles).

Determination of AD distribution

Starting from the integral expressions used to calculate AD rate for a continuous secondary electron energy distribution, we propose the following expression to calculate the number of displacement per atom (*dpa*) in the voxel volume, N_{dpa}, as a sum extended over the calculated discrete electron flux values at a given depth Z, generalized for any *k*-atom in the material

$$N_{dpa} = \sum_k \left(n_k \sum_i N^e_{dpa,k}(E_i) \Phi(E_i, z) \Delta E_i \right).$$

(8)

where n_k is the relative compositional weight of the *k*-atom in the material and $N^e_{dpa,k}$ arises from

the evaluation of Eq.(5) for each one of these atoms.

The previous expression was applied to the studied materials. In the case of Fe, was applied to iron atoms with T_d = 25 eV (= 1). In the case of YBCO, was applied to oxygen atoms with T_d = 20 eV (n_k= 4/7) and to copper atoms with T_d = 25 eV (n_k= 2/3), both belonging to the Cu-O$_2$ planar cites.

The calculation results of total *dpa* values for both materials are shown in figure 2 for different values of incident gamma energy E_γ. As can be observed, the number of *dpa* increases with the energy of incident radiation. Note also that the difference between *dpa* values for each E_γ increases more and more with depth, being strong this augment in the sample thickness. This abrupt increase of *dpa* can be related with the production of displacements in cascade, that takes place when increasing the electrons energy, since atoms with more energy are taken out from its sites and these atoms in turn, produce higher number of secondary atoms and consequently the cascade of displaced atoms is bigger.
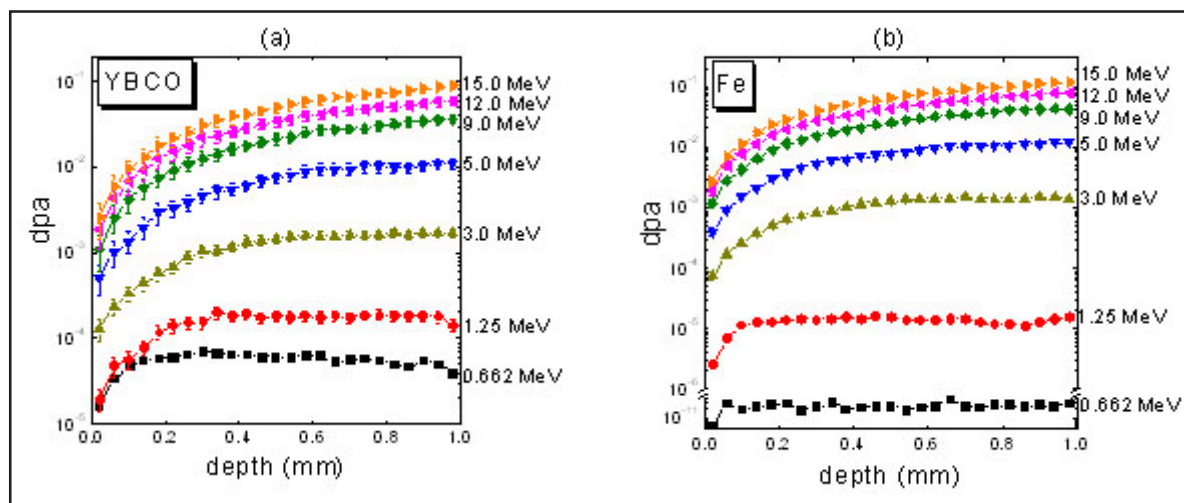

Figure 2. In depth distribution of *dpa* for different incident energy values in YBCO (a) and Fe (b).

For energies $E_\gamma \leq 1.25$ MeV, the *dpa* distribution reaches its maximum inside the material (unimodal distribution), as it has been observed in previous works [14,17,19]. Besides, these *dpa* distribution shapes are similar to the observed in the obtained energy deposition profiles at the same energies [18].

The maximum *dpa* values for each E_γ are shown in table 1, for both materials, taken into account the contribution of each atomic species.

From table 1 it seem that in the case of YBCO the oxygen atoms have a bigger weight in the total *dpa* values at low energies and, in contrast, the copper atoms at high energies. This result is consistent with the evidences observed previously [2-4]. Note

also that the Fe material is more resistant to radiation damage at low energies ($E_\gamma < 5.0$ MeV) and YBCO at high irradiation energies.

Comparison between AD and energy deposition

Starting from the similarity between the *dpa* distributions and energy deposition profiles, a comparison among them was performed, as it is showed below in figure 3. Due to the good correspondence that was observed in this figure involving both magnitudes, a linear fitting of this relationships was carried out for all incident energy. The results of this fitting are given away in table 2, showing the slope parameter, $\eta(E_\gamma)$, that represent the direct relation between *dpa* and energy deposition. The linear regression coefficient R is shown too.

Table 1. Maximum *dpa* values for each incident energy for both materials

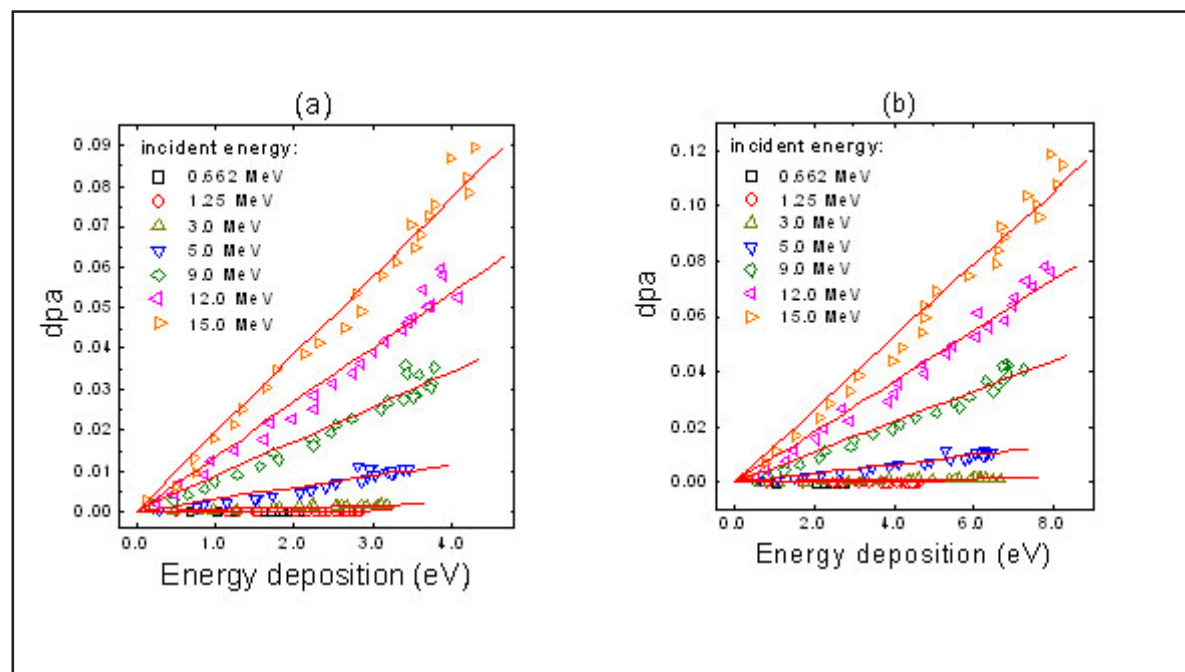|	YBCO			Fe
Eγ (MeV)	oxygen atoms	copper atoms	total	Total
0.662	6.91×10^{-5}	-	6.91×10^{-5}	3.89×10^{-11}
1.25	1.93×10^{-4}	5.56×10^{-6}	1.98×10^{-4}	1.58×10^{-5}
3.00	1.13×10^{-3}	5.84×10^{-4}	1.71×10^{-3}	1.51×10^{-3}
5.00	5.95×10^{-3}	5.25×10^{-3}	1.12×10^{-2}	1.19×10^{-2}
9.00	1.60×10^{-2}	2.01×10^{-2}	3.61×10^{-2}	4.28×10^{-2}
12.00	2.48×10^{-2}	3.49×10^{-2}	5.96×10^{-2}	7.80×10^{-2}
15.00	3.57×10^{-2}	5.37×10^{-2}	8.94×10^{-2}	1.19×10^{-1}

Figure 3. Relation between *dpa* and energy deposition values for every studied incident energy in YBCO (a) and Fe (b). Continuous lines represent the linear fitting of the data.

Table 2. Results of the linear fitting of the data shown in previous figure 3

	YBCO		Fe	
Eγ (MeV)	($\eta \pm \Delta\eta$) (dpa/eV)	R	($\eta \pm \Delta\eta$) (dpa/eV)	R
0.662	$(3.03 \pm 0.04) \times 10^1$	0.955	$(9.45 \pm 0.44) \times 10^6$	0.447
1.25	$(6.76 \pm 0.17) \times 10^1$	0.927	$(3.45 \pm 0.11) \times 10^0$	0.792
3.00	$(5.17 \pm 0.15) \times 10^2$	0.945	$(2.29 \pm 0.05) \times 10^2$	0.972
5.00	$(2.91 \pm 0.09) \times 10^3$	0.960	$(1.65 \pm 0.04) \times 10^3$	0.971
9.00	$(8.55 \pm 0.18) \times 10^3$	0.982	$(5.51 \pm 0.11) \times 10^3$	0.988
12.00	$(1.34 \pm 0.02) \times 10^4$	0.990	$(9.18 \pm 0.14) \times 10^3$	0.993
15.00	$(1.93 \pm 0.03) \times 10^4$	0.992	$(1.32 \pm 0.02) \times 10^4$	0.992

It can be clearly observed from figure 3 that AD depend linearly on the radiation energy deposition in the material, and that this dependence is stronger when increasing E_γ. This can be better distinguish in table 2, which shows how the values of $\eta(E_\gamma)$ increase abruptly when increasing E_γ, from approximately 30 dpa/eV for $E_\gamma = 0.662$ MeV up to about 2×10^4 dpa/eV for $E_\gamma = 15.0$ MeV. It can be also verify that the linear fitting is better (> R) at bigger E_γ.

These particular behaviors can be expected, since secondary electrons plays an important and decisive role on the general energy deposition mechanism and particularly on the displacement of atoms from their crystalline sites arising at the gamma quanta transport, which is extended over the whole target volume.

These findings seem to confirm partially the results reported by A. Leyva *et al.* [14], in which the calculated energy depositions were taken as a measurement of the Gamma Radiation Damage effects on the YBCO superconducting material.

CONCLUSIONS

The calculation of AD rates induced by the Gamma Radiation to different energies in the YBCO superconductor was carried out on the basis of a procedure relying on the use of the standards tallies of the Monte Carlo method based simulation code MCNPX, which are then applied to perform calculations through standard expressions describing the electron Primary Knock-on interactions leading to Atom Displacements events. This provides a realistic approach to the subject of Gamma Radiation Damage calculations, where the imported secondary electrons energy profiles emphasize the shower and cross linked nature of gamma quanta and secondary electron interactions arising at Gamma Radiation transport in solids.

A nearly position independent linear correlations among AD rate and energy deposition were found, which allows the application of an energy deposition scale for Gamma Radiation Damage determination.

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