

A compound spike model for formation of nuclear tracks in solids

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Abstract Formation of nuclear tracks in solids has been described as a thermal spike as well as a Coulomb explosion spike. Here, formation of nuclear tracks is described as a compound spike including partial roles of both thermal and Coulomb explosion spikes in track formation. Fractional roles of both spikes depend on atomic and electronic structure of a track detector and deposited energy density in the track detector by the incident charged particle. Behavior of the cylindrical zone along the path of the incident particle is described mathematically in terms of bulk and individual atomic flow or movement. Defect structure of the latent nuclear tracks is described and conditions of continuity and discontinuity of latent tracks are evaluated and discussed. This paper includes mathematical description, analysis and evaluation of the nuclear track formation issue in the light of published experimental and theoretical results, which are useful for users of nuclear track detection technique and researchers involved in ion beam induced materials modification and ions implantation in semiconductors.

Key words Nuclear track detection technique, Track formation, Compound spike model, Defects, Nanostructure

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1 Introduction

Latent tracks are damaged zones created by moving charged particles in solids. These latent tracks can be stored indefinitely in insulating materials and can be amplified to get information about the track forming particles using a process called preferential chemical etching. Solid State Nuclear Track Detectors (SSNTDs) have some potential advantages such as low cost, less weight and the ability to discriminate against lightly ionizing particles. These detectors have provided very valuable results in various fields such as nuclear physics, astrophysics and geophysics. Due to numerous applications in different areas of applied science, many researchers have studied track formation mechanisms^[1-4]. Even after investigations of a few decades, a number of questions about the structure of charged particle tracks in solids are still un-answered. The use of this technique will be further improved if the track formation mechanism is understood thoroughly. A deeper understanding of interac-

tion of ions with solids is also useful for further improved ions implantation in semiconductors and sputtering.

2 A compound spike model for nuclear track formation

A charged particle traveling in a solid creates a superheated cylindrical zone with a modified structure containing defects of various types and sizes. A point $P(r, x)$ in a typical nuclear track is shown in Fig.1. In the inner dotted cylinder (Fig.1), bulk atomic flow and, in the outer shell, individual atomic flow happen during track formation. A fresh nuclear track in a solid is highly unsteady in time and after reaching thermodynamic equilibrium it becomes an inhomogeneous structure. The energy deposited by a track-forming particle in a cylindrical volume around the path is non uniform. It decreases exponentially along radial direc-

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tion whereas distribution along axis of the cylinder depends on energy of the particle and for MeV/u ions, it has a maximum at a depth into the target. Immediately ($\sim 10^{-17}$ s) after passage of an ion into the target material, a distribution of quasi-defects is formed. The density of these quasi defects is given by

$$N_d^q(r, x) = K \frac{N_o}{U} \varepsilon(r, x) \quad (1)$$

where N_o is the atomic density of the solid, U is the cohesive energy of the solid and $\varepsilon(r, x)$ is the deposited energy density at point $P(r, x)$ in the solid. These quasi-defects include ionized and excited atoms and atoms with broken bonds and displaced from their positions in the atomic structure of the solid. In this initial stage, energy deposited inside cylindrical track is continuous.

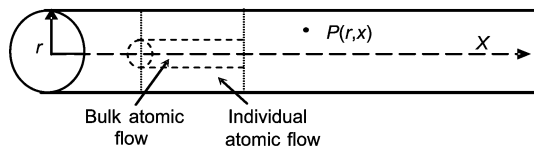


Fig.1 Latent nuclear track in a solid, showing cylindrical zones of bulk and individual atomic flow. Parameters are defined/shown in this figure for the purpose of mathematical description of the problem.

These quasi-defects produce separation of charges along the track and overlapping of electron clouds of excited atoms. Temperature of the cylindrical zone is raised through electron-phonon coupling. Coulomb repulsion and thermal diffusion (a compound spike) result into atomic movement. Atomic movement breaks the original atomic structure of the solid. This atomic movement is quenched by neutralization of ionized atoms and de-excitation of excited atoms. In good multi-atom dielectric materials, atomic movement can cause segregation of atomic species along radial direction due to difference in atomic masses. In a typical dielectric material, with three atomic species of significantly different masses, expected segregation of atomic species is shown in a cross-sectional view of a track in Fig.2. Atomic concentration of light, moderate and heavy elements will be highest in outer, middle and inner shells, respectively. The inner most solid circle represents the region of primary ionization produced by the track-forming particle and might contain extended defects, or very high density of point defects and it can be continuous or discontinuous. Its bigger size is shown only to differentiate it from atomic spe-

cies. The initial ionization produced by the incident charged particle can be considered as a line source. The density of these quasi-defects will satisfy

$$\frac{1}{r} \left[\frac{\partial}{\partial r} \left(r \frac{\partial N_d^q}{\partial r} \right) \right] = 0 \quad (2)$$

The solution that vanishes at $r=a$ is

$$N_d^q = N_d^0 \ln(a/r) \quad (3)$$

where a is radius of the damaged latent track ($0 < r < a$).

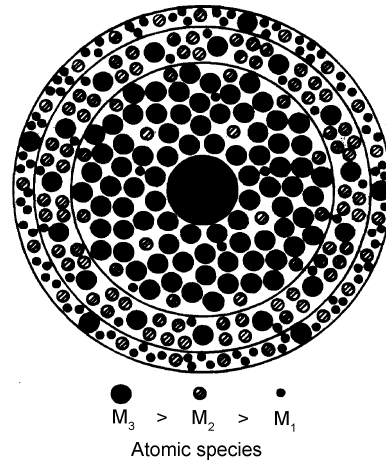


Fig.2 Atomic structure of a latent track in a compound solid containing atomic species of different masses, which is the case for most nuclear track detectors in use. The cross sectional view is shown.

The solution becomes infinite at $r=0$. It is not possible to determine density at axis without considering finite width of the source. A new defective structure is formed in a cylindrical track after its solidification.

The defect structure in a solidified latent track is loaded with a variety of stable defects formed aggregation and stabilization of quasi-defects. A fraction of quasi-defects are removed, a fraction becomes point defects and the remaining becomes extended defects. Point defects are broken and high energy bonds whereas extended defects are clusters of point defects. Mechanisms responsible for formation of extended defects are not understood completely. Nanometer-scale defect structure deduced from previously published^[2,3] and present investigation is shown in Fig.3. The density of final stable defects in a nuclear track is given by

$$N_d(r, x, v_d) = K_2 g(U, E_f) \int_0^{x_{\max}} \int_0^{r_{\max}} f(r, x) \varepsilon(r', x') dr' dx' \quad (4)$$

where $N_d(r, x, v_d)$ is density of defects with volume v_d at $P(r, x)$, $E_f = E_f(r, x, v_d)$ is energy of formation of this type of defects, $f(r, x)$ represents the energy contribution to the defect formation at $P(r, x)$ coming from an arbitrary point $P(r', x')$, r_{\max} is the radius of the damaged cylindrical zone and x_{\max} is the range of the charged particle in the solid, and K_2 is a constant of proportionality. The function $g(U, E_f)$ may be given by

$$g(U, E_f) = N_{\max} \exp(-E_f / kT) \quad (5)$$

where k is the Boltzman constant and T is the absolute temperature.

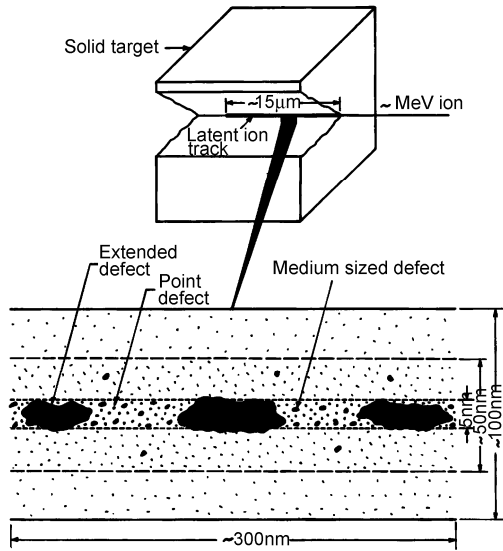


Fig.3 Nanometer-scale defect structure of a latent nuclear tracks deduced from previously published and present investigation.

Mechanisms of track formation of quasi-defect distribution into final fixed defect distribution (especially in case of discontinuous tracks) are not known. Observation of discontinuous^[2] and continuous^[4] tracks in different detector materials at the same beam energy 1–10 MeV/u suggests that most important factors in leading the track to be continuous or discontinuous are target (or detector) properties (composition and cohesive energy etc.). All tracks at high deposited energy density $\varepsilon(r, x)$ might be continuous due to overlap of extended defects in the track. At low deposited energy density, segregation of latent tracks into inten-

sively damaged and partly damaged sections is understandable in crystals if atomic composition of target material is not changed. Formation of continuous tracks in GaN^[4] might be the formation of Ga-rich material in the track due to evaporation and diffusion of nitrogen^[5] into surroundings of the track. The change in atomic composition is less probable in InP^[6] where discontinuity in latent tracks has been observed. Continuity/discontinuity of latent tracks in amorphous materials has not been tested yet, up to author's knowledge.

Fig.4 shows radial distribution of deposited energy density of 200 MeV ¹⁹⁷Au ions in CR-39 and GaN, calculated using formulation given by Tombrello^[7]

$$\varepsilon(r, x_0) = \frac{1}{7.4\pi} \left(\frac{dE}{dx} \Big|_{x_0} \right) \left(\frac{1}{R_{\perp}^{0.27} \bullet r^{1.73}} \right) \quad (6)$$

where R_{\perp} is a specific distance in Å and has a value of $840(e/\rho)$, where ρ is density in g/cm³ and e is energy of ion in MeV/u. Further details of equation (6) are given by Tombrello^[7]. Deposited energy density decreases very quickly with radius r . It is very important to know the radius of the cylindrical track. The experimentally observed value of 200 MeV ¹⁹⁷Au ion tracks is ~ 100 Å^[3]. The calculated deposited energy density at $r=50$ Å is 0.044 eV/Å³, which is 26% of density of cohesive energy (0.167 eV/Å³)^[8]. It may be deduced here that threshold energy deposition for formation of tracks is more than 25% of density of cohesive energy of the solid.

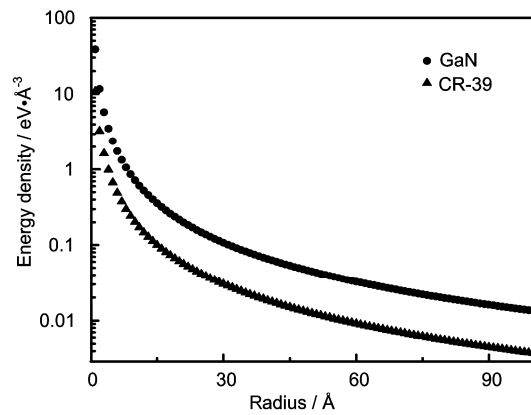


Fig.4 Radial distribution of deposited energy density of 200 MeV ¹⁹⁷Au ions in CR-39 and GaN.

3 Discussion

Table 1^[9-15] shows values of activation energy of annealing of nuclear tracks in different detecting solids along with detection threshold Z/β . We will refer elements lighter than carbon as light elements, the group of carbon, nitrogen, oxygen and fluorine as moderately heavy elements and the remaining as heavy elements. Activation energy of annealing is found to be higher in case of materials containing higher percentage of moderately heavy elements, suggesting that tracks inside these materials have relatively higher density of extended defects. This fact can be seen in Table 1, which contains the atomic percentage of moderately heavy elements in commonly used track detectors

along with the activation energy for tracks in these materials. We can also see in Table 1 that more sensitive detectors have a good fraction of light elements, which can produce a kind of Coulomb explosion as suggested by Schiwietz and co-workers^[16]. So, if detector material contains both light elements (suitable for Coulomb explosion) and moderately heavy elements (suitable for formation of extended defects), track formation can be expected provided free electron density is less than a certain limit to provide enough time for Coulomb explosion and formation of extended defects. The atomic composition of these materials (given in Table 1) can be seen in any standard material^[17,18].

Table 1 Mean values of activation energy of annealing of nuclear tracks in different commonly used track detectors. M and L stand for moderately heavy and light elements

Detector	Elements (M) /%	Elements (L) /%	Activation energy /eV	Z/β ^[15]
CR-39	51.4	49.6	0.197±0.005 ^[9] 0.210±0.060 ^[10]	10
Apatite	54	4	0.457±0.009 ^[11]	800
Mica	56	2	0.572 ± 0.064 ^[12]	
Garnet	60	0	1.209 ± 0.093 ^[13]	
Glass	60	0	0.160 ± 0.031 ^[14]	550

Coulomb explosion happens for lighter elements (e.g. hydrogen) even at lower deposited energy density along the path of charged particle as suggested by Schiwietz and co-workers^[16]. The formation of latent track also depends on some other detector material properties, especially the ability of a material to preserve the damage. Detector can preserve the damage in the form of latent tracks if it has free electron density lower than a certain limit as discussed previously in a bit detail. These electronic properties of the detector material can be represented by the concept of track potential, which can be written as suggested by Schiwietz et al.^[19]

$$V_T(\mathbf{R}) = \int d\mathbf{x} \frac{\rho_v(\mathbf{r}) - \rho_s(\mathbf{r})}{\mathbf{R} - \mathbf{r}} \quad (7)$$

where $\rho_v(\mathbf{r})$ and $\rho_s(\mathbf{r})$ are spatial charge densities of vacancies in all energy bands, and stopped electrons. Distribution, intensity and duration of nuclear track potential will determine whether or not the track formation will occur. It is quite difficult to determine distribution of nuclear track potential in the cylindrical

zone around the particle passage due to difficulties involved in measurement or calculation of density parameters $\rho_v(\mathbf{r})$ and $\rho_s(\mathbf{r})$.

4 Conclusion

A model for formation of nuclear tracks in solids is developed using well-known scientific facts or assumptions and mathematical modeling of the evolution of a spike induced by a charge particle in a solid. Previously proposed models of nuclear track formation are briefly reviewed in connection with the presently proposed model. The effects of thermal spike and Coulomb explosion spikes are incorporated in the description of the model. Partial roles of these spikes in nuclear track formation are discussed. Physical significance of mathematical equations in the developed model is also discussed in the light of related published experimental and theoretical results. Results and discussion presented in the paper are useful for other nuclear techniques like ion beam induced materials modification and ions implantation in semiconductors.

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References

- 1 Fleischer R L, Price P B, Walker R M. Nuclear tracks in solids – principles and applications. Berkeley: University of California Press, 1975.
- 2 Dartyge E, Daraud J P, Langevin Y, *et al.* Phys Rev B, 1981, **23**: 5213-5229.
- 3 Tombrello T A, Wie C R, Itoh N, *et al.* Phys Lett A, 1984, **100**: 42-44.
- 4 Kucheyev S O, Timmers H, Zou J, *et al.* J Appl Phys, 2004, **95**: 5360-5365.
- 5 Rana M A, Osipowicz T, Choi H W, *et al.* Chem Phys Lett, 2003, **380**: 105-110.
- 6 Herre O, Wesch W, Wendler E, *et al.* Phys Rev B, 1998, **58**: 4832-4837.
- 7 Tombrello T A. Nucl Instr Meth B, 1984, **2**: 555-563.
- 8 Northrup J E, Neugebauer J. Phys Rev B, 1996, **53**: R10477-R10480.
- 9 Rana M A, Qureshi I E, Manzoor S, *et al.* Nucl Instr Meth B, 2001, **179**: 249-254.
- 10 Virk H S, Modgil S K, Bhatia R K. Nucl Tracks and Radiat Meas, 1986, **11**: 323-325.
- 11 Green P F, Duddy I R, Gleadow A J W, *et al.* Chemical Geology, 1986, **59**: 237-253.
- 12 Roberts J H, Gold R, Ruddy F H. Proc. of 10th Inter. Conf. on Solid State Nuclear Track Detectors. Oxford: Pergamon Press, 1980: 177-188.
- 13 Singh S, Singh L, Singh J, *et al.* Nucl Tracks and Radiat Meas, 1991, **19**: 121-126.
- 14 Modgil S K, Virk H S. Nucl Instr Meth B, 1985, **12**: 212-218.
- 15 Khan H A, Lund T, Vater P, *et al.* Phys Rev C, 1983, **28**: 1630-1634.
- 16 Schiwietz G, Xiao G, Luderer E, *et al.* Nucl Instr Meth, 2000, **164-165**: 353-364.
- 17 Considine D M, Considine G D. Van Nostrand Reinhold Encyclopedia of Chemistry (Fourth edition). New York: Van Nostrand Reinhold Company, 1984.
- 18 Roberts W L, Campbell T J, Rapp G R. Encyclopedia of Minerals (Second edition). New York: Van Nostrand Reinhold Company, 1990.
- 19 Schiwietz G, Grande P, Skogvall B, *et al.* Phys Rev Lett, 1992, **69**: 628-631.