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ion beam mixing (review)

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Abstract

Ion beam mixing (IBM) is a widely used method for modifying the structure and properties of bilayer and multilayer systems. The idea behind IBM is to create a surface alloy by homogenizing alternate layers deposited in a thickness ratio so that they result in a desired final composition after mixing. In this review some basic aspects of ion beam mixing reviewed. these include describe the basic mechanism of the phenomena of ion beam mixing; also a brief description of the phenomenological various models used for quantitative calculation of different mixing parameters.

Keywords: mechanism of mixing, thermal spike, ballistic binary.

1.Introduction

Ion beam mixing (IBM) is one of the most efficient and effective routes to synthesize stable or metastable alloy phases. IBM may be defined as an intermixing and/or alloying that can occur at the interface of two different materials during ion irradiation [1]. This technique offers several advantages over conventional techniques, particularly in terms of spatial selectivity, relatively low-temperature process, less preparation duration and capability of mixing immiscible systems. Among these advantages, spatial selectivity offers a way to synthesize the phases in a particular region of interest by irradiating the films in that specified area. Moreover, this method does not require large temperatures like the thermal annealing, which can be performed even at the room temperature. Furthermore, it consumes less amount of time for the synthesis compared to the other techniques, if the ion current is sufficiently high. The capability of mixing immiscible systems is the most interesting aspect of this technique, which is only possible through this method. It is well known that the ion beam mixing depends on several parameters such as energy of ion beam, ion fluence, ion species, target temperature, number of layers and their thicknesses.

2.Histricial of ion beam mixing

This IBM technique was first discovered by Van der Weg et al. in 1973 [2], where Pd thin films on Si substrates were lead to form palladium silicide after Ar ion irradiation. There are several studies on IBM of binary multilayer systems in the literature. For example, Ibrahim et. al., have reported on the alloy phase synthesis of Bi-Sb system using this method by varying different parameters such as ion fluence, ion energy and temperature [3]. Also, Manju et. al. has shown the mixing of Co-Sb and Pb-Te systems as well as engineering their thermo-electric properties upon ion ion irradiation [4]. Cheng et. al. has extensively studied the IBM and the formation of amorphous alloys of various binary systems such as Au-Ag, Pt-Pd, Hf-Zr, W-Mo, Ta-Nb, Ru-Zr, Ru-Ti and so on [5]. Apart from the IBM study of metal-metal systems, there are few more interesting studies on metal-semiconductor systems. S. Dhar et al. have studied the IBM of metal-semiconductor systems such as Cu-Ge, Ni-Ge and Co-Ge and reported their phase formation [6]. Similar work has been carried out on ion beam mixing of Metal-Si system such as Co-Si [7], Ni-Si, Pt-Si, Pd-Si, Mo-Si, W-Si, Cr-Si and Mg-Si systems [8], where they have achieved their stable and/or meta-stable phases. Since, ion beam mixing is one of the best alternatives for conventional techniques for the phase synthesis with added advantages, it has been the subject of research interest to many research groups from last few decades.

3. Theoretical models of ion-beam induced transport of matter

3.1. Low energy ion beam mixing

When the energetic heavy ion penetrates a top (impurity) layer A to reach a bulk material B, it loses energy due to collision with target atoms, which receive sufficient energy to get displaced from their original positions. These displaced atoms in turn make multiple collisions with the target atoms to produce a displacement cascade. The effects of these collisions can be divided into two categories based on the time scales:

- Prompt effects ~ a few ps, termed as Cascade mixing.
- Delayed effects, exceeding several ns consisting of Radiation Enhanced Diffusion (RED) at higher temperatures and thermal spike diffusion at lower temperatures.

3.1.1. Cascade mixing:

The secondary collisions along the path of the incident ion produce a collision cascade which involves many atoms with kinetic energies much smaller than incident ion energy and the multiple relocations of the atoms occur resulting in the mixing of dissimilar impurity A and bulk B atoms across the interface. Thus each incident ion gives rise to a small volume across the interface containing both atoms A and B. The overlap of these cascades with increasing ion fluence, result in a continuous mixed region.

3.1.2. Radiation Enhanced Diffusion (RED):

Thermal diffusion proceeds through defect interactions. Normal defects which are produced by thermodynamic lattice disorders show exponential decrease with temperature so that little or no diffusion occurs at room temperature. Under energetic ion bombardment diffusion can be greatly enhanced because a large number of point defects [9,10] that accelerate normal thermally activated diffusion are produced and mixing occurs due to this mechanism. This phenomenon is termed as radiation and defect enhanced diffusion (RED).

3.1.3. Thermal spike diffusion:

At low temperatures where RED does not take place, a thermal ion beam mixing in metallic layers can result from a combination of recoil mixing, collisional mixing and thermal spike diffusion [11,12]. Thermal spike is defined as a limited volume inside a solid with the majority of atoms temporarily in motion [13] and the respective material may thus be regarded as being in a liquid like state. The origin of these spikes is the dissipation of the energy deposited in the collision cascades through atomic motions in time ~10−13 s.

3.2. Swift heavy ion beam mixing

During passage of SHI through the materials, cylinders contain highly charged ions and electrons are formed causing modifications. After the passage of the SHI, the solid returns to its equilibrium state leaving behind bulk and surface modifications. The nature of modification depends on the electrical, thermal and structural properties of the target material, the mass of the projectile ion and irradiation parameters. To explain SHI induced mixing, mainly two models Coulomb Explosion and Thermal Spike are used. One can focus either on the response of the ionized atoms in the presence of conduction electrons or on the process by which the excited electrons dissipate their energy in the lattice. The Coulomb Explosion Model is related to the first approach whereas the Thermal Spike deals with the second one. One another model used for heavy ion mixing is Szenes model. However, it is perceptible that at such high energies the electronic excitation and electron–phonon and phonon–phonon coupling processes are expected to produce defects due to the atomic rearrangements and thereby enhance mixing at interfaces.

4. Phenomenological model:

For quantitative understanding of mixing is important to estimate some of the useful mixing parameters like mixing rate **(X^2/Φ)** : rate at which the squared thickness X^2 of the mixed region increases as a function of irradiation dose (Φ)at given temperature for a particular incident ion beam ; mixing efficiency **($X^2/\Phi\ F_D$) :** normalized mixing rate with respect to the deposited energy density F_D .

For theoretical prediction of these parameters. several phonological models which depend on different experimental conditions viz atomic number of ion incident ion and target atoms, energy deposited in collisions ect.

4.1. ballistic binary collision model

The ballistic mechanism in case of ion beam-induced mixing of the metal bilayer systems can be explained in terms of the average atomic number and it was observed that when the average atomic number lies below 20, linear binary collision cascade dominates [14], as can be illustrated by SRIM-simulations, and therefore, pure ballistic transport occurs. According to first model of ion beam mixing, proposed by Sigmund and Gras-Marti[15], the mixing rate (k) of a bilayer system is given by : $\mathbf{k = \Delta\sigma^2}$ / $\varphi = \frac{1}{3} r \xi \frac{F_D R_d^2}{N E_d}$

where, r is a dimensionless parameter (=0.608), n is a mass-sensitive kinematic factor given by $[4M_1M_2/(M_1+M_2)^2]^{1/2}$, F_D is the energy deposited by the incident ion at the interface, E_d is the displacement energy and R_d is the minimum distance (1 nm) required for a stable Frenkel pair. This theoretical formulation was based on the pure ballistic binary collision model, i.e., ion beam mixing only depends on kinematical properties of the materials, it always takes place and it is independent on the chemical properties of the system and, therefore, various thermodynamically driving forces were not taken into consideration.

4.1. Thermal spike model

The thermal spike model, proposed by Johnson et al. [16] assuming the formation of a global thermal spike along the ion path which gives rise to a Darken type enhancement of the mixing process along with parameters like effective cohesive energy (ΔH_{coh}), heat of mixing (ΔH_{mix}) etc. According to their model, the phenomenological expression for the ion beam mixing is given by : $\mathbf{k = \Delta\sigma^2}$ /

$$\frac{1+k_2\frac{\Delta H_{mix}}{\Delta H_{Coh}}}{\varphi \frac{k_1 F_D^2}{N_{avg}^{5/3}} \text{¿}})$$

where, N_{avg} is the average atomic density, K_1 and K_2 are fitting parameters with values of 0.0034 nm and 27.4, respectively.

As this model predicts a quadratic dependence of the mixing rate on the deposited energy, it raised soon doubts about the formation of a global spike. Instead of this, some authors proposed the formation of 'local spikes' which was initiated only when the recoil energies were below a critical value of $3.923\ x10^{-2}\ Z^{2.23}$ eV, where Z is the average atomic number of the target material [17]. At such low energies, the recoil cascades approximately exhibit spherical shapes. But Bolse predicted that overlapping of sub-cascades ultimately generated a cylindrical spike where the linear dependence on F_D was restored. The 'local cylindrical thermal spike' model by Bolse [18] predicts a mixing rate equal to: $\mathbf{k' = \Delta\sigma^2}$ /

$$\frac{1+k'_2\frac{\Delta H_{mix}}{\Delta H_{Coh}}}{\varphi = \frac{Z_{avg}^{1.77} F_D}{N_{avg}^{2/3}(\Delta Hcoh)^2} \text{¿}})$$

where, K_1 and K_2 are empirical constants and Zavg is the average atomic number.

5. Conclusion:

Ion beam mixing has been studied extensively since 1973. It is considered as an alternative way for surface composition modification and stable and metastable phases formation. In spite of the large body of ion beam mixing data, further effort is needed to distinguish between ballistic and thermal spike contributions to the observed. Our study in future intend to understand the experimentally observed mixing in bilayer and multilayer system in the basis of the Thermal Spike Model (TSM) at two different energies.

References:

[1] M. Nastasi and J. W. Mayer, Mater. Sci. Eng., R, 12, 1 (1994).

[2] W. F. Van der Weg, D. Sigurd, and J. W. Mayer, Applications of ion beams to metals, edited by S.T. Picraux, E. P. EerNiss and F. L. Vook (Plenum Press, New York, p. 209 (1973).

[3] A. Ibrahim, D. Thompson, Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms 7 (1985) 566.

[4] Manju bala, PhD Thesis, Jawaharlal Nehru University, New Delhi, India (2016).

[5] Y. T. Cheng, Mat. Sci. Rep., 5, 45 (1990).

[6] S. Dhar, PhD Thesis, Dept. of Phys., IIT Kanpur (1996).

[7] W. Xia, C. A. Hewett, M. Fernandes, S. S Lau and D. B Poker, J. Appl. Phys., 65, 2300 (1989).

[8] S.M. Mayers, Nucl. Instrum. Methods Phys. Res. B 168 (1980) 403.

[9] W. Wagner, L.E. Rehn, H. Wiedersich, V. Naunderf, Phys. Rev. B 28 (1983) 6780

[10] W.L. Johnson, Y.T. Cheng, M. Van possum, M.A. Nicollet, Nucl. Instrum. Methods Phys. Res. B 7–8 (1985) 657.

[11] W. Bolse, Nucl. Instrum. Methods Phys. Res. B 80–81 (1993) 137. 34. W. Bolse, Nucl. Instrum. Meth. Phys. Res. B 80/81, 137 (1993).

[13] P. Sigmund, Appl. Phys. Lett. 25 (1974) 169.

[14]. P. Sigmund, Rev. Roumaine Phys. 17, 823, 969, 1079 (1972)

[15] P. Sigmund, A. Gras-Marti, Nucl. Instrum. Meth. Phys. Res. 182/183, 25 (1981)

[16] W.L. Johnson, Y.-T. Cheng, M. Van Rossum, M.-A. Nicolet, Nucl. Instrum. Meth. Phys. Res. B 7/8, 657 (1985)

[17]. W. Bolse, Nucl. Instrum. Meth. Phys. Res. B 80/81, 137 (1993)

[18]. P. Borgesen, D.A. Lilienfeld, H.H. Johnson, Appl. Phys. Lett. 57,1407 (1990)