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ARTICLE

Theoretical Study of Intermixing in Au/Si System under Swift Heavy Ion Irradiation

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Abstract: The behavior of the metallic multilayer Au100Å/Si100Å/Si under swift heavy ion irradiation has been studied within the inelastic thermal spike model. Gold has been chosen due to its insensitivity to irradiation. The heat transport in the electronic and lattice subsystem has been simulated using a three-dimensional numerical code that includes the energy transfer between the layers in a direction perpendicular to layer surfaces. The simulations have been done for four kinds of ions Pb, Xe, Kr, and Ar, at a specific energy of 3Mev/amu in order to study the influence of the electronic stopping power on the intermixing process. The results show that for Pb, Xe, and Kr, intermixing at the interface of Au/Si has been obtained. However, no mixing has been observed with Ar. Furthermore, the value of the interdiffusion coefficient for Au has been estimated to be approximately $11x10^{-4}$ cm^2 s^{-1}, which is characteristic of the liquid phase and, thus, supports the thermal spike model of mixing. For comparison, similar thermal spike simulations have been applied to Ti100Å/Si100Å/Si under the same irradiation conditions. We have found that Ti, which is a sensitive material, exhibits considerable intermixing with Si during Pb, Xe, and Kr irradiation. Whereas, a weak intermixing has been found after Ar irradiation; this is completely different from what has been observed in the Au/Si system.

Keywords: Irradiation, Swift heavy ions, Thermal spike model, Ion beam mixing.

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1. Introduction

The irradiation of metallic multilayers with swift heavy ions can induce considerable modifications. In this study, we are interested in the intermixing observed at the interface of the Au/Si system. Ion beam mixing (IBM) is the atomic intermixing and alloying that can occur at the interface separating two different materials during ion irradiation. It is applied as a process for adhering two multilayers, especially a substrate and deposited surface layer [1]. In the electronic energy regime, ion beam mixing can be explained using two different theoretical models, the Coulomb explosion [2] and thermal spike models [3, 4]. "According to the Coulomb explosion, as swift heavy ions pass through a material, they lose energy to the electrons in the material, thereby exciting the electrons and ionizing the atom. This leads to an ionized cylindrical zone along the path of the ions with a strong electronic deficiency in the centre. This deficiency creates electrostatic repulsion between the positively charged atoms in the centre, which results in an explosion-like atomic motion perpendicular to the path of the ions [5]. This can lead to mixing at the interface of bi-layer/multi-layer systems. This type of mixing is

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only seen in insulators" [6]. "In the case of metals and semiconductors, mixing is explained using the thermal spike model. Energy transferred by the swift heavy ion (SHI) to the lattice electrons will be imparted to the lattice via electron-phonon coupling. This results in the heating of the lattice and the generation of a Gaussian-like temperature profile around the path of the ions. The temperature can rise above the melting point of the material, and a cylindrical molten track of few nanometers can be formed. If the interface of the two materials in contact melted during irradiation, mixing between bilayer can occur by inter-diffusion through the molten track" [6].

In this work, we have used the thermal spike model to study the behavior of Au, which is insensitive to energetic heavy ions, in the metallic multilayer Au100Å/Si100Å/Si by describing the intermixing induced at its interface. For completeness and comparison, we have applied the same calculations to Ti100Å/Si100Å/Si.

2. The Thermal Spike Model

The concept of thermal spike was first introduced by Seitz[3], and analytically developed by Kaganov et al.[7].Then Toulemonde et al.[8, 9], Trautmann et al.[10], Meftah et al.[11, 12], Dufour et al.[13, 14],Wang et al.[15] and Kadid et al.[16] used this model to describe the track formation in different materials. For ion beam mixing using swift heavy ions, several researchers have demonstrated that the thermal spike model can explain the intermixing in the following multilayer systems: Ni /Ti [17], Fe/(Al or Au)[18], Fe/Si [19], Ni/Si [20], (Bi or Au)/Al_2O_3[21] and Cu/aGe [22]. Recently, by applying the thermal spike model, Patra et al. [23] showed that the swift heavy ion induced mixing across Pd/Si and Ni/Si interfaces is a consequence of the transient molten state. To the best of our knowledge, there has been no theoretical study of swift heavy ion induced intermixing in Au100Å/Si100Å/Si.

2.1. Mathematical Description

The thermal spike model is described mathematically by two coupled equations [3] governing the energy diffusion into the electron subsystem and into the lattice subsystem. A time-dependent transient thermal process coupling these two systems is expressed using a cylindrical geometry whose axis is the ion path:

$$C_e \frac{\partial T_e}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}(rK_e(T_e)\frac{\partial T_e}{\partial r}) - g(T_e - T_a) + A(r,t) \quad (1)$$

$$C_a \frac{\partial T_a}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}(rK_a(T_a)\frac{\partial T_a}{\partial r}) + g(T_e - T_a) \quad (2)$$

In these equations, T_e, T_a, C_e, C_a and K_e, K_a, are the temperature, the specific heat and the thermal conductivity of the electronic and atomic systems, respectively. The electron-phonon coupling constant [3] represented by g is linked to the electron-phonon mean free path λ [9] by the relation $\lambda^2 = D_e C_e/g$ and to the electron–phonon mean free time by the relation $\tau = C_e/g$, with $D_e = K_e/C_e$. In the present calculations, C_e and De are taken constant (C_e= 1J $cm^{-3}K^{-1}$, De = 2 cm^2 s^{-1}). When these values are applied for C_e and D_e, the thermal conductivity K_e is equal to 2J $cm^{-1}s^{-1}$ K^{-1}. A(r, t), is the energy density supplied by the incident ion to the electronic system at radius r and time t [7]. For A(r, t), the following equation was used [9]"

$$A(r, t) = b\, S_e \exp[-(t-t_o)^2/2t_0^2]\, F(r), \quad (3)$$

where S_e is the inelastic energy loss of ion calculated by the Bethe formula, t_0 is the time of electron gas thermalization ($t_{0\sim}$(1-5) 10^{-15} s), b is the normalizing constant and F(r) is the spatial distribution function of electron energy in the stopping media.

For multilayer systems, the two-dimensional code of the thermal spike model has been extended to have three dimensions by introducing the new term $\frac{\partial}{\partial z}\left(K_i \frac{\partial T_i}{\partial z}\right)$ (i = e for electrons and i = a for atoms), which describes the heat conduction between the layers in the direction perpendicular to the surface of the layers. In this case, the new coupled differential equations [24] are:

$$\frac{\partial T_e}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}(rK_e(T_e)\frac{\partial T_e}{\partial r}) + \frac{\partial}{\partial z}\left(K_e(T_e)\frac{\partial T_e}{\partial z}\right) - g(T_e - T_a) + A(r,t) \quad (4)$$

$$\frac{\partial T_a}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}(rK_a(T_a)\frac{\partial T_a}{\partial r}) + \frac{\partial}{\partial z}\left(K_a(T_a)\frac{\partial T_a}{\partial z}\right) + g(T_e - T_a) \quad (5)$$

The three-dimensional code was first applied to a Ni/Ti bilayer system by Wang et al. [17]. For bilayer- or multilayer systems, the calculation code solves coupled differential Eqs. (4) and (5), taking into account the evolution of all the parameters versus $T_{e,a}$ and the initial

energy distribution on the electrons, A(r,t). The main outputs of the code are the evolution of electronic and lattice temperatures versus time for different radial and depth distances.

3. Theoretical Results and Discussion

In order to get a theoretical understanding of intermixing, the Au/Si system has been studied. This system is composed of two different materials: gold and silicon. Gold is insensitive to the electronic excitations and its electron-phonon coupling constant, $g_{Au} = 3x10^{10}$ $W.cm^{-3}.K^{-1}$, is deduced from femto second laser experiments [25]. Silicon has an electron-phonon $g_{Si} = 1.8x10^{12}$ $W.cm^{-3}.K^{-1}$ [20].In a detailed study, Osmani et al. [26] have shown that Si is sensitive to swift heavy ions. The researchers used the thermal spike calculations to demonstrate that tracks can also appear at the Si surface at a threshold value equal to 8 keV/nm.

For the lattice subsystem, all the thermodynamic parameters in the solid and liquid phases were taken from the equilibrium measurements [15, 20]. Table1 shows the thermal properties of Au, Si, and Ti. We have noticed that the melting temperature of Ti is higher than that of Au and Si, respectively, which favors the appearance of a molten phase in Ti. The latent heat of fusion of Si is almost four times that of Ti. This may be the cause why Ti is more sensitive to swift heavy ions than Si. Table 1 also shows that Au exhibits higher ρ_s and ρ_l values than those of Ti and Si materials. This may be explained why Au is an insensitive material.

The electronic stopping power S_e values calculated with the TRIM code [27] are presented in Table 2.

TABLE1. Thermal properties of Au, Ti and Si.

	Au	Ti	Si
Latent heat of fusion (J/g)	63.7	420	1797
Latent heat of vaporization (J/g)	1698	8814	13722
Melting temperature (K)	1336	1933	1683
Vaporization temperature (K)	3243	3560	2953
Mass density solidρ_s (g / cm^3)	19.3	4.5	2.3
Mass density liquidρ_l (g /cm^3)	19.2	4.1	2.5

TABLE 2. Electronic stopping power Se (keV/nm) calculated with the TRIM code [27] for various ions.

	Pb	Xe	Kr	Ar
Au target	78	55	32	13
Si target	22	16	9	4
Ti target	36	26	15	6

TRIM (Transport of Ions in Matter)[28] is a Monte-Carlo algorithm that describes the trajectory of projectiles represented by a binary collision sequence [29].The most popular version of TRIM is contained in the SRIM package(The Stopping and Range of Ions in Matter), and was developed originally by Biersack and Haggmark[28]. However, more detailed studies on stopping powers and ranges have been done by Ziegler, Biersack and Littmark [27].

Equations (4) and (5) are solved numerically and the main calculation results are shown in Figs. 1 and 2. We can see that the electronic temperature in bulk Au increases to its maximum value (peak) of 243315 K within a very short time of ~ 10^{-15} s, and then it decreases. Further, the maximum electron temperature (Fig.1) attainable in bulk Au (243315 K) is higher than that in bulk Si (86539 K) because of the larger S_e value in Au. At the interface of the Au/Si system, the gradient of the electronic temperature drives energy flow from Au to Si until thermal equilibrium is reached in ~ 1,8 x 10^{-15} s (Fig.1) and, consequently, the maximum electron temperature (peak) of Au (198880 K) at the interface is lower than in bulk Au, whereas the maximum electron temperature of Si at the interface is higher than that in bulk Si (Fig.1).

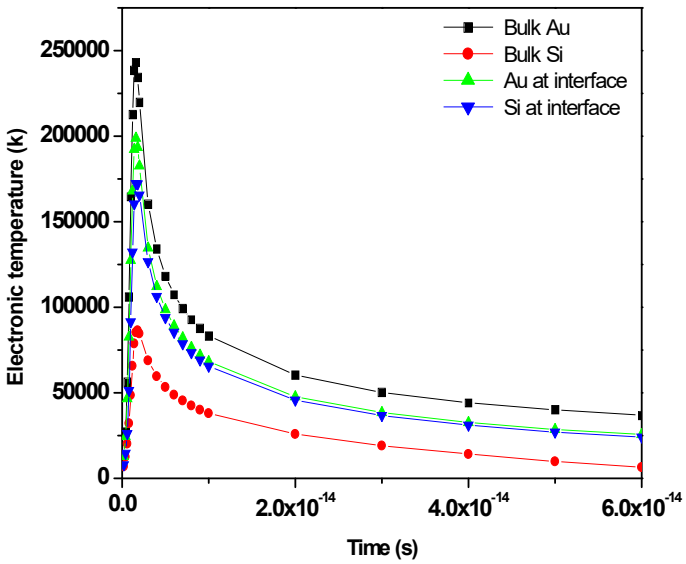

FIG. 1. Evolution of the electronic temperature versus time at a radius r = 1 nm: for bulk Au, bulk Si, and for Au, Si at the interface of Au/Si system, irradiated with 3 MeV/amu Pb ions at room temperature.

The evolution of the atomic temperature is plotted in Fig.2 versus time for various radial distances r. We can notice that for Pb ions of energy 3 MeV/amu, bulk Au does not reach its melting point equal to 1336 K [Fig.2(a)]. This agrees with the already established convention that Au is insensitive to electronic excitation. However, Au at the interface of Au/Si is molten [Fig.2(c)] due to the lattice temperature gradient at the interface, which leads to an energy flow from Si to Au. On the other hand, from Figs. 2(b) and 2(d), it can be noted that both bulk Si [Fig.2(b)] and Si at the interface of the Au/Si system [Fig.2(d)] reach the melting temperature equal to 1683 K. Because in these cases the electronic stopping power in Si equal to 22 keV/nm is higher than the calculated threshold value of 8 keV/nm [26]. The melting of Si near the interface will allow atomic transport through the interface, resulting in the observed intermixing. The calculated track radius R_m is defined as the maximum cylinder radius in which the molten phase is created. In this case, the Au layer melts with the track radius R_m = 10 nm, indicating that the whole Au layer is molten. The lifetime of the melt is about 1ps. The existence of a molten track can also be deduced from the estimated interdiffusion coefficients. To examine this aspect, the diffusion coefficient [30, 31] for Au is given as:

$$D = \frac{\Delta\Omega^2}{2t_n},$$

where $\Delta\Omega^2$ is the spatial width of Au at the interface due to irradiation and t_n is the diffusion time, which can obtained using the following equation:

$t_n = (\Phi/\Phi_c)t_s$.

In this equation, ts= duration of melt phase of Au, Φ= maximum fluence and $\Phi_c = 1/(2r)^2$is the fluence for complete overlap of the ion tracks.

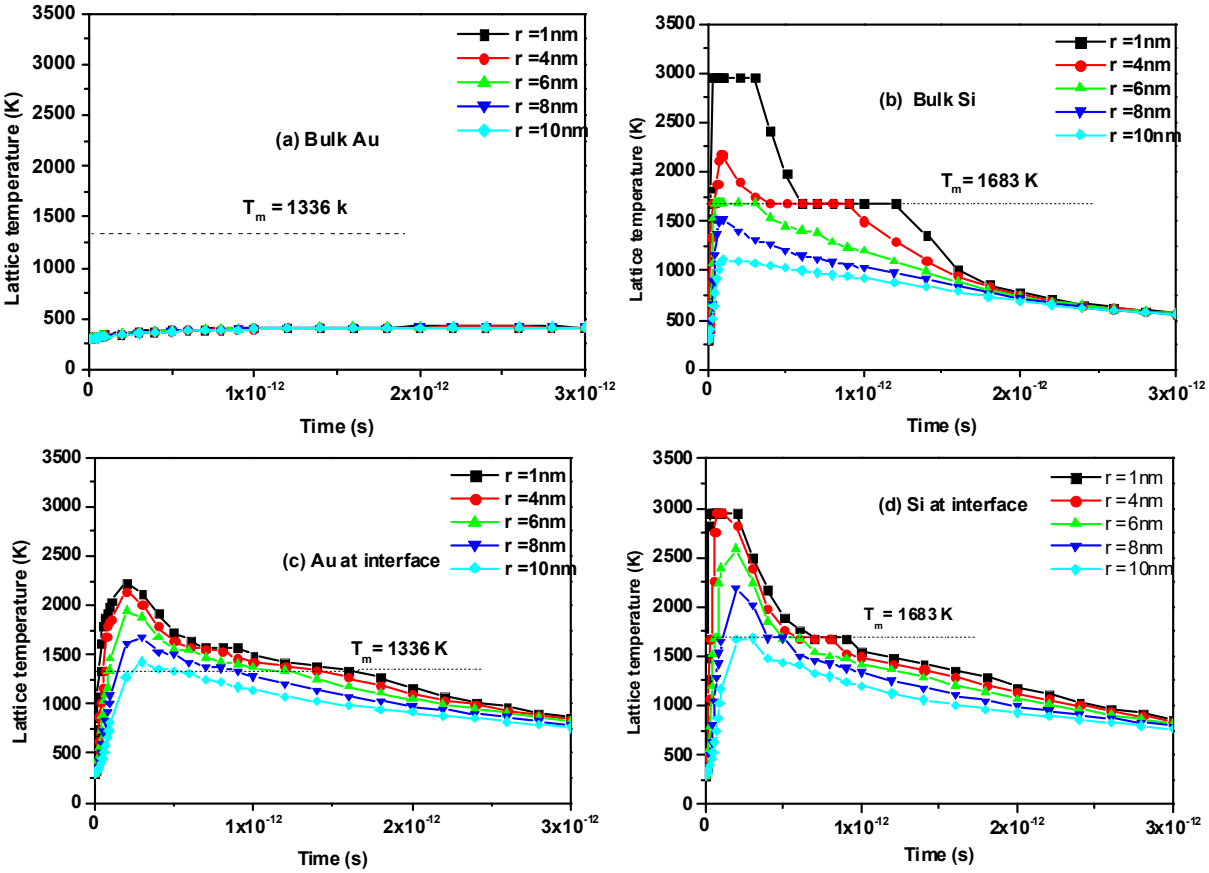

FIG. 2. Evolution of the lattice temperature versus time for various radial distances r: (a) bulk Au, (b) bulk Si, (c) and (d) are respectively Au and Si at the interface of the Au/Si system, irradiated with 3 MeV/amu Pb ions at room temperature.

Since the experimental values of $\Delta\Omega^2$, Φ_c and Φ are not available for Au, we use the values of another metal, tungsten (W), the data for which are available [32]. This substitution is possible because tungsten (W) and gold (Au) are similar in terms of both metals being insensitive materials, having the same density, and belonging to the transition metal category. This conclusion is tentative and requires further experimental investigation and validation. Taking the value of r = 10 nm and t_s = 1ps for Au, the value of interdiffusion coefficient D is estimated to be ~ 11×10^{-4} cm^2 s^{-1}.This value is in good agreement and in the range of the interdiffusion constants and thus supports the thermal spike model of intermixing. It may be noted that similar results were found for V/Si [33], Mo/Si [34], Mo/Si [35], Zr/Si [36], and Mn/Si [37].

Recently, by applying a unified thermal spike model [38], including the effect of the nuclear stopping power, Datta et al. [39] have performed a simulation of the 500 keV Xe ion irradiation in Au (6nm)/Si (6nm) bilayer on SiO_2 substrate. Their results show that both Au and SiO_2 are molten, and the radii of the molten zones are 4.5 nm and 2.5 nm for the Au and SiO_2 layers, respectively. It is also to be noted that the ion fluence applied in the experiment of Datta et al. [39] where nuclear energy loss dominates, is two orders of magnitude higher than that applied in swift heavy ion experiments. Thus, one of the advantages of ion beam mixing (IBM) using high-energy (≥ 1MeV/amu) heavy ions (SHI) over low-energy ion mixing is that the fluence required for mixing is about 10^{13} ions/cm^2 to 10^{14} ions/cm^2. This fluence is two to three orders of magnitude lower, compared to low-energy ion induced mixing [40].

3.1. Influence of the Electronic Stopping Power (S_e) on the Intermixing Process

Experimental studies on various multilayer systems suggest that the nature of swift heavy ion induced modifications in multilayers varies with the magnitude of electronic stopping power. In this work, to test theoretically its influence on the amount of intermixing in the Au/Si system, we have plotted in Figs.3(a) -3(d), the evolution

of the maximum lattice temperature T_{max}, normalized by the melting temperature T_m versus the depth of the multilayer system, for some radial distances, for four kinds of ions (Pb, Xe, Kr, Ar) at a beam energy of 3 MeV/amu, to see the influence of the different S_e values on the atomic intermixing. We can notice in Figs. 3(a)-3(d), that the ordinate was normalized by the melting temperature, i.e., the value of unity or higher shows the melting.

Different intermixing behavior is observed in the four irradiation cases. Irradiation with Pb ion, [Fig.3(a)] characterized by the electronic energy loss (S_e)values of 78 and 22 keV/nm in Au and Si layers, respectively, causes significant intermixing at the interface of the Au/Si system. The corresponding R_m value is the same for Au and Si and is found to be around 10 nm. In this case, the entire Au and Si layers are in the molten phase. This means that a complete mixing is obtained over the entire thickness of the two layers. On the other hand, the amount of intermixing is more with Xe irradiation [Fig.3(b)] than this with Kr irradiation [Fig.3(c)]. This is in conformation with the thermal spike model (TSM), according to which, the high electronic energy loss of Xe ion (as can be seen in Table 1) leads to a higher latent track radius and longer melt duration, leading to a higher amount of intermixing. These results are in good agreement with those of several researchers. In a systematic study, Avasthi et al. [41] have shown that atomic intermixing in the Fe/Si and the CuO/glass systems irradiated with 230 MeV Au ions and 210 MeV I ions, respectively increases with the increase in electronic stopping power. Similarly, Kumar et al.[42] have demonstrated that mixing in the Cu/Ge system due to the irradiation with 100 MeV Ag ions, 120 MeV and 140 MeV Au ions increases with fluence, electronic energy loss and irradiation temperature.

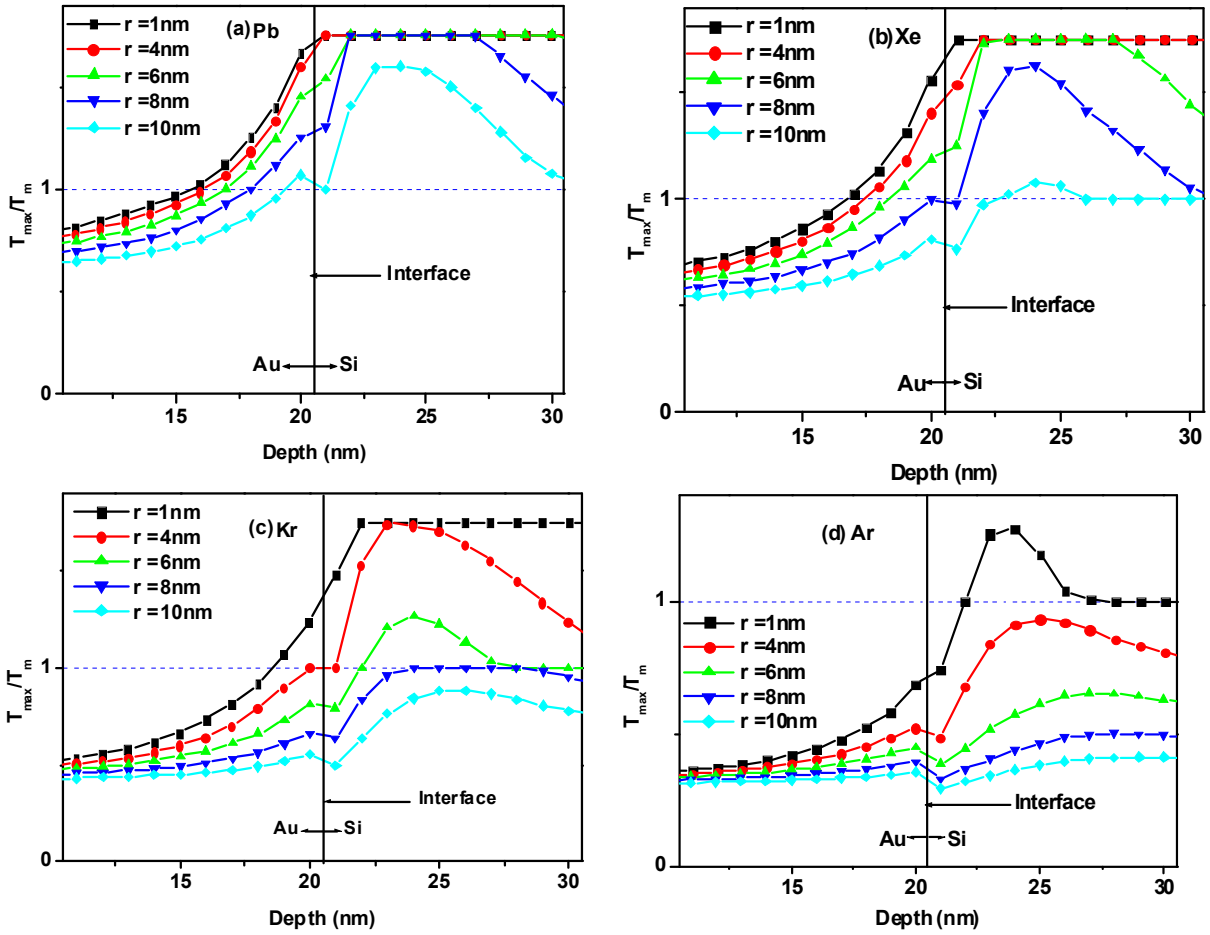

FIG. 3. Evolution of the maximum temperature T_{max} normalized by the melting temperature T_m versus depth for the Au/Si system irradiated with (a) Pb, (b) Xe, (c) Kr, and (d) Ar ions.

In contrast to Figs.3(a)-3(c), after Ar irradiation [Fig.3(d)] in the region of Au, the ratio T_{max}/T_m is lower than unity this means that no melting will be found in the Au layer at the interface, and consequently, the mixing does not exist. We have therefore concluded that intermixing occurs only if both sides of the interface must be molten to allow for atomic interdiffusion. This interpretation is in excellent agreement with the results of Bolse et al. [43] who have found that mixing occurs only when a melt is induced by the projectile in both components of the bilayer. According to theoretical calculations by Osmani et al.[26] based on the thermal spike model, the track formation threshold $(S_e)_{th}$ for crystalline Si is $(Se)_{th}$ (Si) = 8keV/nm. In the case of Ar irradiation, the electronic energy loss in Si layer is 4 keV/nm, which is below the theoretically calculated threshold. This may be another cause why no intermixing was observed in this case.

To further elucidate the mechanism of intermixing in the electronic stopping regime, some theoretical calculations were also performed for Ti100Å/Si100Å/Si multilayer system irradiated with Pb, Xe, Kr, and Ar at energy of 3Mev/amu [Figs.4 (a)-4(d)]. In this case, a higher mixing is observed in Ti/Si system as compared to the Au/Si system. A similar result has also been found by Kumar et al.[44], who have observed that the mixing in Ti/Fe system irradiated with 120 MeV Au at RT at a fluence of $1x10^{14}$ ions/cm^2 is higher than Ti/Au system at the same energy.

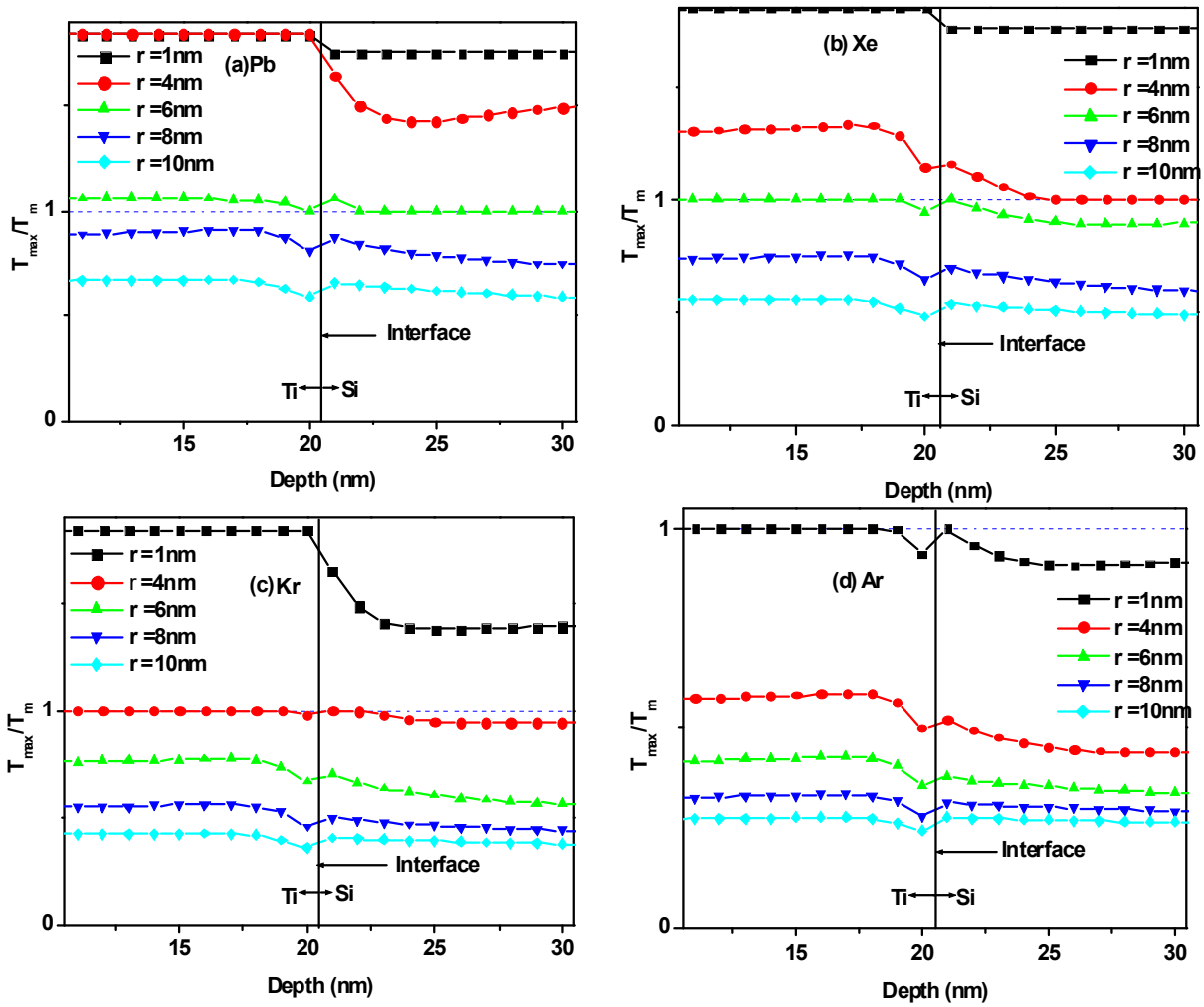

FIG. 4. Evolution of the maximum temperature T_{max} normalized by the melting temperature T_m versus depth for Ti/Si system irradiated with (a) Pb, (b) Xe, (c) Kr, and (d) Ar ions.

On the other hand, according to an empirical equation proposed by Bolse et al.[45] the mixing rate as a function of the electronic stopping power can be described by $k = \zeta^2 (S_e - S_{eth})$, ζ being the mixing efficiency. The authors showed that a strong interface mixing occurred if a threshold $(S_e)_{th}$ was exceeded. Considering that the S_e values for Pb, Xe, and Kr ions in Ti are 36,

26, and 15 keV/nm, respectively, and the threshold of intermixing is determined by the Si layer equal to ~8 keV/n m [26], this would nicely explain why a strong interface mixing has been observed in Ti/Si system.

4. Conclusion

The results of the calculations performed for different values of the electronic stopping power show that the inelastic thermal spike model can explain the intermixing at the interface of Au100Å/Si100Å/Si. The estimation of the interdiffusion constant of Au supports the appearance of a molten phase along the ion track. According to our theoretical results, we have concluded that for efficient mixing, both sides of the interface must be molten to allow for interdiffusion. We have also investigated ion beam mixing in Ti/Si. In this case, mixing is found to be higher than Au/Si at the same energy (3 MeV/uma) because Ti is sensitive to electronic stopping power S_e while Au is an insensitive material. We have also found that intermixing increases with the increase in electronic stopping power. Our results are in good agreement with previous studies on ion beam mixing due to electronic stopping power.

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