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〈연구논문〉

Analytical Formula for the Collisional Atomic Mixing

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충돌에 의한 원자혼합현상의 해석적 모형

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Abstract—An analytical formula for the cascade atomic mixing has developed in this study. It has more realistic parameters involved in the ion-atom collision than Sigmund and Gras-Marti formula. The mixing efficiency calculated using present formula shows exactly the same result with that from Sigmund formula for the light element. It shows, however, slightly different result from Sigmund formula for the heavy element. And it shows good agreement with dynamic Monte-Carlo simulation result for the light element, while it shows slight discrepancy with the Monte-Carlo simulation result for the heavy element presumably due to sputtering and matrix relocation during ion irradiation.

요 약—충돌에 의한 원자혼합현상에 대한 해석적모형을 개발하였다. 이 식은 Sigmund와 Gras-Marti의 식보다 더 현실적인 이온-원자 충돌변수들을 포함한다. 이 식에 의해서 계산된 경량원소에 대한 혼합효율은 Sigmund 식에 의해서 계산된 것과 동일한 결과를 나타낸다. 반면, 중량원소에 대한 계산은 약간의 차이를 나타낸다. 또 이 결과는 경량원소에 대한 dynamic Monte-Carlo 모의실험결과와 잘일치하고 있는 반면, 중량원소에 대해서는 약간의 차이를 나타내는데, 이는 이온 조사시의 sputtering과 기지원자 재배치에 기인한 것으로 추측된다.

1. Introduction

Irradiation of layered thin film structures with energetic particles results in atomic mixing at the interfaces. At low temperature, the atomic mixing was proposed to arise mainly from collisional effects [1]. Recent investigations [2] suggest that thermal spike effect might influence the low temperature mixing in some cases. Nevertheless, it is necessary to provide a reliable theoretical description of the mixing due to collision only, since this is prerequisite for the understanding of other mixing phenomena such as thermal spike or radiation enhanced diffusion.

Serval theorical studies [3, 4, 5] for the collisional mixing have been published. Among them, the most frequently cited analytical treatment is that by Sigmund and Gras-Marti [4] who employed

transport theory in order to derive analytical formula for the collisional mixing. In this study, we present an analytical description of cascade collisional mixing formulated by way of simple diffusion model and physical parameters involved in ion-atom collision. The present formula is applied to bilayered thin film strucures, and mixing efficiences calculated by using the present formula are compared with those obtained using that of Sigmund and Gras-Marti and the results obtained using a dynamic Monte-Carlo simulation.

2. Description of model

Consider an ion beam incident normally with a dose rate of ions/cm^{2}sec onto the surface of a target whose composition varies with depth. The atomic concentration of atoms of type A at a depth x is $C_A(x)$. When atoms move in random directions with average velocity v, the net current density of atomic species A in the positive direction can be described approximately by [6]

$$J_+^A = \frac{1}{4} v\, N\, C_A(x-\frac{2}{3}l) - \frac{1}{4} v\, N\, C_A(x+\frac{2}{3}l) \qquad (1)$$

where l is the mean free path between atomic collisions, and N is the atomic number density. In Eq.(1), $2l/3$ is the average distance that an atom makes its last collision before crossing the interface. The first term on the right-hand side of Eq.(1) is the current density of type A atoms in the positive x direction which cross an interface at x without collision, and the second term is the corresponding quantity for the opposite direction.

During the cascade collision, however, only a fraction of atoms in a cascade region is displaced from their original lattice sites. This fraction (f) depends upon the local energy deposition or the nuclear stopping power (S_n). Thus Eq.(1) should be multiplied by the fraction of f,

$$J_+^A = \frac{1}{4} v\, N\, f[C_A(x-\frac{2}{3}l) - C_A(x+\frac{2}{3}l)] \qquad (2)$$

$$= -\frac{1}{3} v\, l\, N\, f\, \frac{\partial}{\partial x} C_A(x)$$

When we assume that the number of type A atom is conserved (neglecting the sputtering due to ion bombardment), the continuity equation can be applied,

$$N\frac{\partial}{\partial t} C_A(x,\ t) = -\nabla \cdot J_+^A \qquad (3)$$

or

$$\frac{\partial}{\partial t} C_A(x,\ t) = \frac{1}{3} v, l, f \frac{\partial^2}{\partial x^2} C_A(x,\ t) \qquad (4)$$

This is the well known diffusion equation or Fick's second law [7]. Thus the effective diffusion constant (D) becomes

$$D = \frac{1}{3} v\, l\, f \qquad (5)$$

In order to correlate the physical parameters involved in collisional process to the diffusion constant, firstly we interpret the average speed v. If the traveled distance of an atom participating in a cascade is taken to be the mean lateral cascade distance (r), the frequency of cascade can be described approximately by πr^2, thus the following expression

$$v \cong \pi r^3 \qquad (6)$$

gives a rough estimate of the average atomic drift velocity.

Secondly the fraction of the displaced atoms (f) can be determined by calculating displaced atoms and dividing this value by the total number of atoms in the average cascade volume calculated from the expression [8]

$$V_c = \frac{4}{3}\pi(R_p + \Delta R_p)\ y^2\ \alpha^3 \qquad (7)$$

where R_p is the mean projected range of ion in medium, ΔR_p and y are the longitudinal and transverse straggling of ion trajectories, respectively, and α is the correction factor for an individual cascade, whose value is assumed to be ~1 in this study. If the number of the displaced atoms is assumed to be roughly [9] $\Delta E/2E_d$, where E_d is the threshold displacement energy of the type A atom, and deposited energy (ΔE) is $S_n R_p$, then the fraction of atoms displaced in the cascade region be-

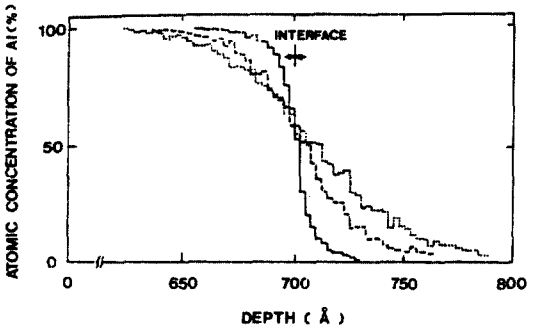

Fig. 1. Dynamic simulation results for the atomic concentration variation in Al layer at the interface of Al (70 nm)/Pd structure bombarded with 80 keV Ar^+ ions at doses of 1×10^{15} Ar^+cm^2 (——), 5×10^{15} Ar^+cm^2 (— — —), and 1×10^{16} Ar^+cm^2 (· · · ·).

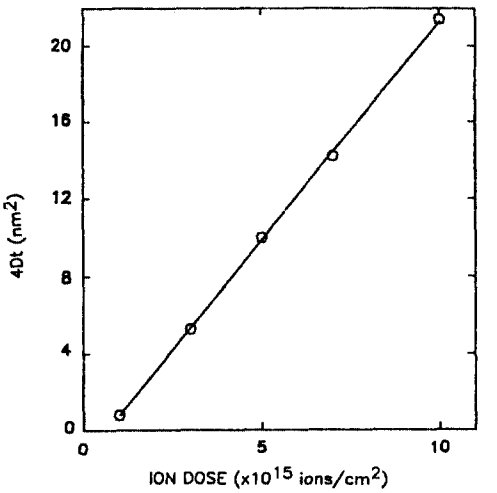

Fig. 2. The mixing variances of Al in Al (70 nm)/Pd structure as a function of ion dose. The incident energy of Ar^+ is 80 keV.

comes

$$f = \frac{S_n R_p/2E_d}{\frac{4}{3}\pi (R_p+\Delta R_p)\ y^2\ N} \tag{8}$$

Using Eq, (5), (6), and (8), the effective diffusion constant becomes

$$D = \frac{1}{8}\ \frac{lr^3 R_p S_n}{E_d N (R_p+\Delta R_p) y^2} \tag{9}$$

When we define the normalized quantity $\eta = 4Dt/\phi S_n$ (ϕ is the total ion dose) as the mixing efficiency, then η becomes

$$\eta = \frac{1}{2}\ \frac{lr^3 R_p}{E_{d(Rp}+\Delta R_p) y^2 N} \tag{10}$$

In Eq.(10), the unknown parameters are l and r, since the values of R_p, ΔR_p, y, and S_n can be determined using TRIM simulation code [10].

We assume that l is the interatomic spacing of target atom. It corresponds to hard sphere cross-section which is appropriate in low energy region [3]. And r is assumed to be approximately half of y, since the ratio of individual cascade to the average cascade distribution is about 0.6 [8].

3. Comparison with Sigmund's formula and Dynamic Monte-Carlo Simulation

Sigmund and Gras-Marti derived an analytical formula for the isotropic cascade mixing by using transport equation. The mixing efficiency is given as [4],

$$\eta = \frac{2}{3}\Gamma_o \frac{\xi_{21} R_{iv}^2}{N\ E_d} \tag{11}$$

where $\Gamma_o = 0.608$, $\xi_{21} = [4m_1 m_2/(m_1+m_2)^2]^{1/2}$, and R_{iv} is a typical seperation distance for vacancies and interstitials, whose value is usually taken as 1 nm [11]. One can see that the Eq.(10) has more realistic ion-atom collisional parameters than Eq.(11).

In order to compare our analytical formula (Eq. (10)) with Eq.(11), we calculated the mixing efficiency for the case that 80 keV Ar^+ ion is implanted into Al (70 nm) layer deposited on Pd substrate. Using E_d = 16 eV [12], l=0.4 nm, $N=6\times10^{-3}$ nm^{-3}, R_p=69.4 nm, ΔR_p=21.2 nm, and y=20.3 nm, we obtained $\eta = 41.1\times10^{-5}$ nm^5 eV from Eq.(10) and η=41.0 nm^5/eV from Eq.(11). Thus we found that the Eq.(10) reveals exactly the same result with that obtained from Eq.(11) for the Al (70 nm)/Pd system.

The analytical treatments have certain limitations such as the assumption of Gaussian profiles or the neglect of matrix recoil and the sputtering effect [13]. These limitations can be overcome by employing a dynamic Monte-Carlo computer simulation

Table 1. Mixing efficiencies obtained using analytical formulas and dynamic Monte-Carlo simulation for various system.

System [Top layer(thickness) /Substrate]	S_n (eV/nm)[a]	R_p (nm)[a]	ΔR_p (nm)[a]	y (nm)[a]	E_d (eV)[b]	η ($\times 10^{-5}$ nm^5/eV)		
						from Eq.11	from Eq.10	from MCS
Pd(40 nm)/Cu	930	34.0	18.0	18.0	26	20.2	16.2	13.4 ± 0.27
Ag(40 nm)/Fe	870	39.6	20.3	20.2	23	26.6	24.0	19.35± 0.84
Ag(40 nm)/Cu	770	37.9	19.6	21.2	23	26.6	25.9	21.2 ± 0.85
Al(70 nm)/Cr	650	70.0	21.2	20.7	16	41.0	40.8	42.54± 1.69
Al(70 nm)/Pd	500	69.4	21.2	20.3	16	41.0	41.1	43.8 ± 1.44

[a] These values are obtained using TRIM code. The values of S_n are those of top layer atom at the interface.
[b] in Ref. 12.

which simulate the entire ballistic process during the slowing down of an incident ion. Thus we have developed a dynamic Monte-Carlo simulation (MCS) code, and then compared the calculated mixing efficiencies using Eq.(10) and Eq.(11) with the result obtained using MCS. The dynamic MCS will be described in detail elsewhere [14], so that only the main features and the choice of the most important input parameters are described below.

The Ziegler-Biersack-Littmark potential [10] is adopted for elastic scattering between colliding species. The inelastic energy loss between or during the collision is described by the Lindhard [15] formula. The incident ion loses its kinetic energy through both inelastic and elastic collisions.

When the target atom acquires an energy T which is larger than the threshold displacement energy (E_d), the target atom becomes displaced and moves with a kinetic energy of T-E_b, where E_b is the bulk binding energy of the solid target atoms. Whenever the energy of the moving particle decreases to a certain cut-off energy (E_c), the atom stops moving and rest in the target. And when the recoil atoms obtain enough energy to generate other recoil process, this causes collision cascade. Those recoil atoms having kinetic energy greater than the surface barrier energy, U_s, leave the surface, which are called sputtered atoms. The following parameters are used for the Al/Pd system, $E_c = E_b$, $E_b = 2u_s$, $u_s = 3.36$ eV and 3.91 eV [16] for Al and Pd, respectively, and $E_d = 16$ eV and 26 eV [12] for Al and Pd, respectively.

In the dynamic mode, the target is subdivided initially into slabs of 2 nm thick. After each passage of a pseudoprojectile (representing a certain interval of incident dose), the atomic composition in each slab is rearranged according to the number of atoms being removed or deposited there. Fig. 1 shows the dynamic simulation results for the atomic concentration variation of Al at the interface of Al (70 nm)/Pd system bombarded with 80 keV Ar^+ with varing ion dose. It reveals that the calculated profiles are complementary error function distribution and tend to become broader across the interface with increasing ion dose.

The mixing variance (4Dt) at each ion dose is estimated by fitting the curve with a complementary error function $\mathrm{erfc}[x/\sqrt{4Dt}]$. Fig. 2 shows the dynamic simulation results for the mixing variance at various doses. The mixing variance increases linearly with ion dose. The mixing efficiency, η, can be estimated from the slope of this line in Fig. 2 and the nuclear stopping power for Al at the interface. The mixing efficiency for the dynamic simulation result is found to be $(43.8 \pm 1.44) \times 10^{-5}$ nm^5/eV. This value is very close to the calculated values using Eq.(10) and Eq.(11).

In order to compare the values of η from analytical formulas and that from MCS in detail, we calculated the mixing efficiencies for various systems using Eq.(10), Eq.(11) and MCS. These results are shown in Table 1. The mixing efficiencies obtained using Eq.(11) show same values when the atomic species of top layer are same (e.g. Al/Pd

and Al/Cr systems), since Eq.(11) has not the physical parameters related to the substrate material. In contrast, mixing efficiences obtained using Eq. (10) show different values, since Eq.(10) has some parameters related to the system structure such as R_p, ΔR_p, and y. When the atom of top layer is light element, both of analytically calculated η are exactly same. They show, however, slight difference for the heavy element of top layer. Neverthless, Eq.(10) is more appropriate for the experimental interpretation because it has more realistic parameters involved in ion-atom collision. The MCS results are in good agreement with those obtained using Eq.(10) within 7% error when the top layer is light element, while they show relatively large discrepancy (~20% error) with those from Eq.(10) for the top layer of heavy element. We suggest that the descrepancy between the calculated η using Eq.(10) and MCS result might be arised from sputtering effect and relocation of atoms during ion irradiation, since the sputtering yields of Al, Pd, and Ag are found to be 2.2, 5.3, and 7.2 atoms/ion, respectively [16].

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