Formation mechanism of indium microcrystals on ion-bombarded InP surfaces

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Surface topography of argon-ion bombarded InP surfaces was investigated using *in situ* scanning electron microscopy. The density of indium microcrystals on the ion-bombarded surface was found to be proportional to the square root of the sputtering rate. The size of thses crystals decreased with increased sputtering rates. These results suggest that the density and size of indium microcrystals are determined by nucleation kinetics of free indium atoms created by preferential sputtering, just like the growth islands in deposition.

1. Introduction

Surface topography induced by ion-beam sputtering often causes degradation of depth resolution. For InP, the preferential sputtering of phosphorous results in indium-microball (microcrystalline island) formation [1,2]. In particular in depth profiling using Auger electron spectroscopy (AES), in which the ion energy and current density are lower than those used in dynamic secondary ion mass spectrometry (SIMS), depth resolution deteriorates drastically. Ogiwara et al found that the depth resolution in AES depth profiling depended on the sputtering rate of InP; the higher the sputtering rate, the higher the depth resolution [3]. To clarify the physical meaning of their observation, we investigated surface topography of argon-ion bombarded InP surfaces using *in situ* scanning electron microscopy (SEM). In this paper, we discuss the effect of sputtering rate on the surface morphology of InP.

2. Experimental

We performed argon-ion bombardment in an ultrahigh vacuum SEM instrument [4]. The sample chamber pressure during Ar^+ bombardment was 2×10^{-7} Torr using differential pumping. The ion energy was 4 keV. The Ar^+ beam was not rastered and was directed to an InP(001) surface at an impact angle of 30° to surface normal. The spot size was 0.5 mm in full width at half maximum. There were fine InP particles or other dust particles on the InP surface. These particles acted as masks during the ion bombardment and created shadowed regions, as schematically shown in Fig. 1. Thus, the sputtered depth could be evaluated at these particles from the center of the beam to the halo region, where the current density changed by two orders of magnitude. The surface morphology was observed by SEM near each particle. The maximum sputtered depth at the center of the beam spot was 3 μm.

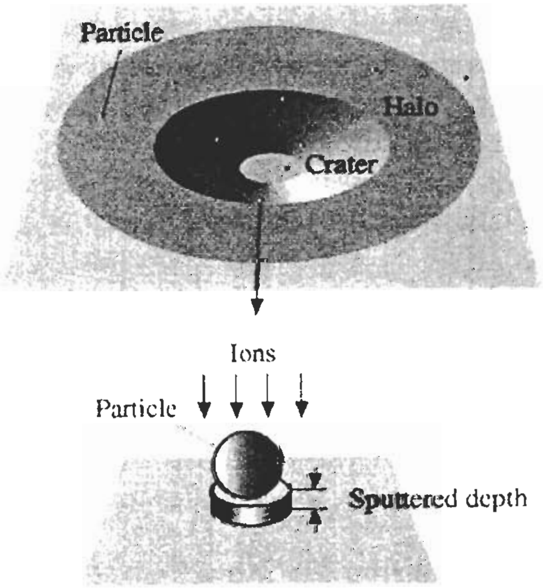

Fig. 1. Sputtering-rate dependence measurement.

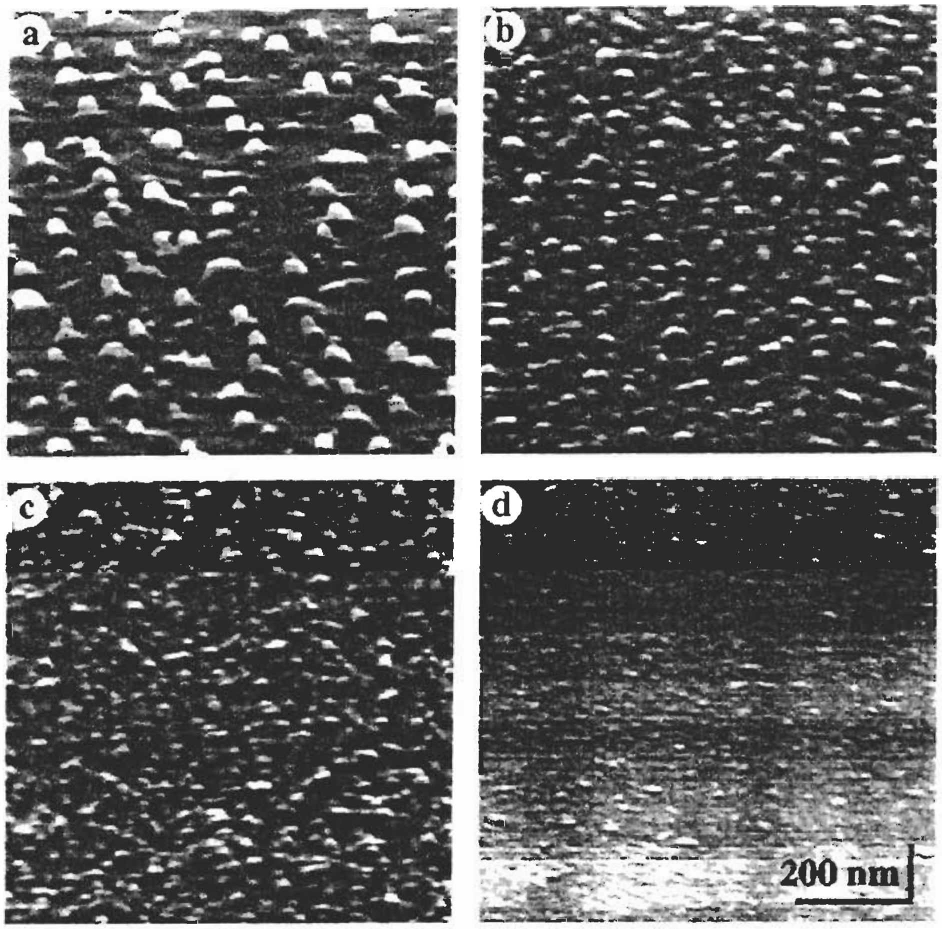

Fig. 2 SEM images of Ar$^+$ bombarded InP surfaces.
Sputtering rate and sputtered depth: (a) 4.6 nm/min, 0.1 μm, (b) 11.5 nm/min, 0.3 μm, (c) 26.5 nm/min, 0.69 μm, and (d) 115 nm/min, 2.9 μm.

3. Results and discussion

SEM images of the sputtered InP surfaces are shown in Fig. 2 for four different sputtering rates. The sputtering rate in Fig. 2(c) is close to that in dynamic SIMS. Surface topography is produced due to the formation of In islands. The size of the In islands is smaller for higher sputtering rates but the density of islands becomes higher.

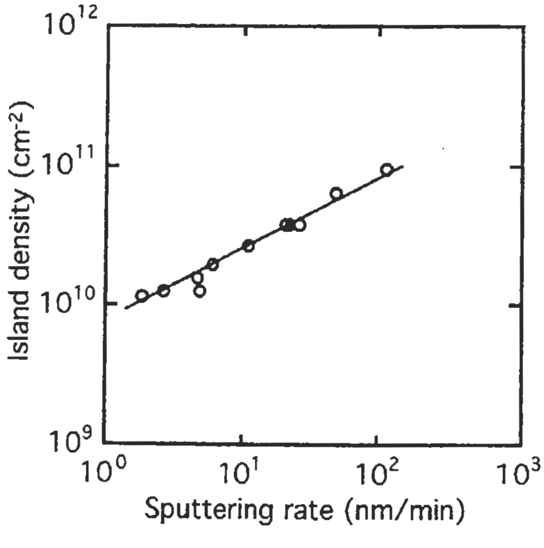

Fig. 3. Sputtering-rate dependence of In-island density

Although the sputtered depths are different for these surfaces, the morphology showed little dependence on sputtered depth for the same sputtering rate.

The sputtering rate dependences of the In island density and size are shown in Figs. 3 and 4, respectively. These were evaluated using SEM images. Note that the island height is affected by the sputtering rate difference between In and InP. The density is proportional to the square root of the sputtering rate. The average island area determined using the island diameter exhibits almost the reverse dependency as the density, thus the surface coverage of the In islands is almost constant. The deviation from the inverse-square-root dependency at higher sputtering rates might be due to three-dimensional growth of islands; island height might not be negligible for smaller islands. These sputtering rate dependences suggest that In-island formation is governed by the nucleation kinetics of islands, as is the growth of films.

The steady state concentration of In atoms created by preferential sputtering should be independent of the sputtering rate. However, the creation rate of In atoms J (atoms cm^{-2} s^{-1}) is proportional to the sputtering rate R. Therefore, the results shown in Fig. 3 mean that the In-island density N_s is proportional to $\sqrt{J}$. This corresponds to complete condensation of adsorbed atoms in vapor phase deposition [5],

$$N_s = \sqrt{J/D_s}, \tag{1}$$

where D_s is the surface diffusion coefficient of the adsorbed atom. This is the case where the lifetime of the adsorbed atoms before collision with other atoms λ_c is shorter than the lifetime of the adsorbed atoms before desorption λ_d,

$$\lambda_c < \lambda_d. \tag{2}$$

That is, adsorbed atoms collide with each other to form nuclei before desorption can occur. In the present case, the surface coverage of In islands is independent of the sputtering rate, indicating that all the In atoms created by preferential sputtering condense completely. Thus, inequality (2) should be satisfied.

The sputtering rate is smaller for In than for InP. Under a high sputtering rate, small In islands are completely sputtered away and a slight topography remains. However, the In island density is so high and the sputtered material is so ample that the topography is smoothed out. Under a low sputtering rate, on the other hand, larger In islands are more resistant to the sputtering and cause cone formation before being sputtered away. Therefore, the lighter the sputtering conditions (i.e., lower energies and lower current densities), the rougher the sputtered surface. The height of an In island was comparable to the sputtered depth for the sputtering rate of 1 nm/min.

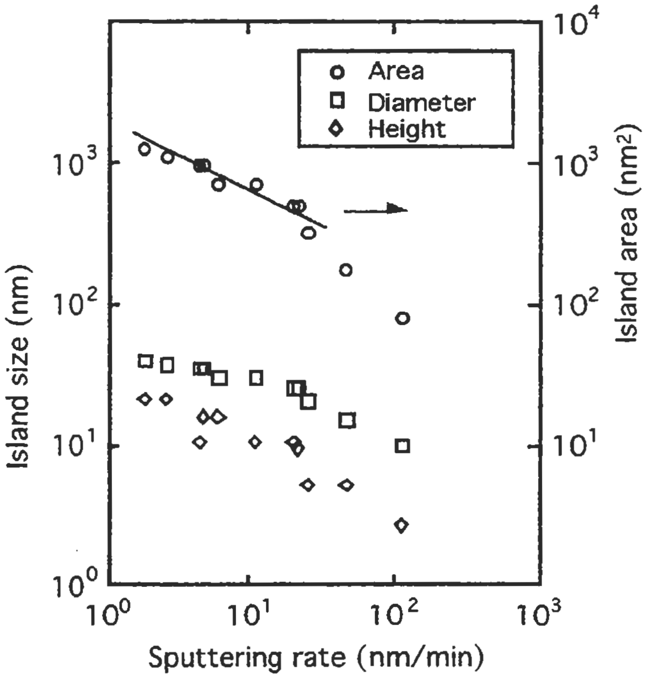

Fig. 4. Sputtering-rate dependences of In-island sizes and area

4. Conclusions

We have shown that indium islands on an argon-ion bombarded InP surface grow due to the nucleation kinetics of free indium atoms created by preferential sputtering. The island density was proportional to the square root of the sputtering rate. The island size decreased with increased sputtering rates. These results explain the previously reported results that a higher depth resolution was obtained by increasing the sputtering rate in AES depth profiling [3].

References

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Comments & Replies
Referees: Singo Ichimura (ETL) and Shigeo Tanuma (Japan Energy ARC)

Ichimura: The explanation of the experimental results is quite clear except one point described below.
The island density is proportional to the square root of the sputtering rate (R) at the region R<100 nm/min. However, it seems that the island area is proportional to the inverse of the sputtering rate at the region of 10 nm/min <R< 100 nm/min. (It is not proportional to the inverse square root of the sputtering rate.) Does it mean that the surface concentration of In depends on the sputtering rate at higher sputtering rate region? This point should be made clear in relation to the sentence "The steady state concentration of In atoms created by preferential sputtering should be independent of the sputtering rate"

Author: The deviation from the inverse-square-root dependence on sputtering rate might be due to the fact that the In islands are not two dimensional, but three dimensional. If the nuclei are two dimensional, the island area should be exactly proportional to the inverse square root of the sputtering rate. However, the area become smaller when 3D nucleation occurs. For larger islands, the height of the initial island might be negligible, but it is significant for smaller islands. This had been stated in the original text, but it was misleading. So I changed the expression (3rd page, upper left) to make it clear.

田沼：2. Experimental において" Ultrahigh vacuum SEM"を用いたとありますが，このときの真空度を示してはいかがでしょうか？また，このとき超高真空でなければならない理由はありますか？

著者：イオンスパッタ中の真空度に関する記述を本文中に追加しました．試料室の到達真空度は 3x10^{-10} Torr です．超高真空は本実験に不可欠ではありませんが，すくなくとも，スパッタ中に残留ガスによる汚染が生じない程度の真空度必要です．

田沼：sputtering rate を測定するために Fig.1 の様な測定をしていますが，この場合の誤差はどのくらいでしょうか？ これは SEM の倍率の補正も絡んでくると思いますが，いかがでしょうか？

著者：スパッタによる段差を測定するため，段差の大きさに応じて数千から 10 万倍の顕微鏡倍率を用いました．別の試料で, SEM で推定した段差と AFM で測定した段差がほぼ同じになることを確認しています．しかし，SEM の倍率の校正は行っていません．誤差は±15%程度と推定します．

田沼：Fig.3 の island density の決定は Fig.2 をみますと，(d)の様な場合はたいへん難しいような気がします．実際的にはどのように行ったかお教えいただけませんか？

著者：確かに，Fig. 2(d)のような場合は苦労します．測定方法は，各 SEM 像に対して決まった大きさの領域を 3 ヶ所設定し，その中の island density を数え，これを平均して求めました．異なる大きさの island が共存しますが，大きさには係わらず数えました．今回は目で見て数えましたが，このような処理が可能な画像処理ソフトもあります．

田沼：sputtering 後の InP 表面は sputter 速度に関係なく（元素的には）In でおおわれてるとこの論文からは考えれます．とくに速

い sputtering rate の場合は表面は平坦に近くなっています．このときのこの層の厚さはどのくらいでしょうか？

著者：スパッタ中にはスパッタ速度に応じた表面In原子のフラックスが存在するはずですが，スパッタを停止すると（フリーなIn原子の生成が停止されると）表面に残ったInは既に生成された核に凝集すると考えられます．したがって，In island以外の部分のInはかなり少ないのではないでしょうか．少なくとも，freeなInによる被覆率は１原子層に比べてかなり小さいと考えます．しかし，これらがArイオン照射によりミキシングされる効果も考えなければいけないので，表面付近のIn濃度の正確な推定はできてはいません．