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Development of oxygen plasma source and ZnO sputtering experiment

新型 ZnO プロセス研究用酸素プラズマ源の開発と
ZnO スパッタリング実験

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We consider a new method of fabricating ZnO cluster by use of a trapping technique of non-neutral plasmas called Penning trap (H. Himura *et al.*, 25E08 in this conference). To estimate the growth time of clusters in the penning trap, we firstly calculate the time evolution of the particle number in a unit volume with the Becker-Döring model.

Introduction

Zinc oxide (ZnO) has attractive potential in applications such as ultraviolet or blue light emitting diode, laser, solar cell, and transparent thin films transistor. This is because ZnO has a wide direct band gap of 3.37 eV, large excitation binding energy of 60 meV at room temperature, and efficient radiation recombination.

One of most crucial issues on the ZnO applications is how the amount of impurities is decreased during processes. For developing a new process to solve the problem, We consider promoting particle growth in a non-neutral plasma trap where little impurities or residual gas exist because of ultra-low vacuum pressure. In fact, this may fabricate even ZnO clusters that become ZnO quantum dots if they grow up in the trap.

Regarding the cluster growth in a non-neutral plasma trap, the growth of hydrogenated silicon clusters in an ion trap was reported[1]. In their experiment, Si_nH_x (n = 2-10) clusters are confirmed by trapping the ionized hydrogenated silicon cluster ions ($Si_xH_x^+$). However, in this experiment, the cluster size was less than 10.

One of possible reasons may be due to heating of the cluster because of the AC electric field used in their trap. The Penning trap, on the other hand, employs no AC E-fields so that no heating is occurred. This is actually a favor aspect if we intend to grow up clusters larger than 10.

Density limit in penning trap

The Penning trap shows long confinement time, for instance, even for several days. On the other hand, there is an inherent limitation on its density that is called Brillouiun density limit *Brillouin density limit* expressed in $n_\mathrm{B} = \epsilon_0 B^2/2m$. For N-sized ZnO cluster ions, which is defined as $(\mathrm{ZnO})_N^+$ based on the ZnO unit, The value of n_B is shown in Fig. 1.

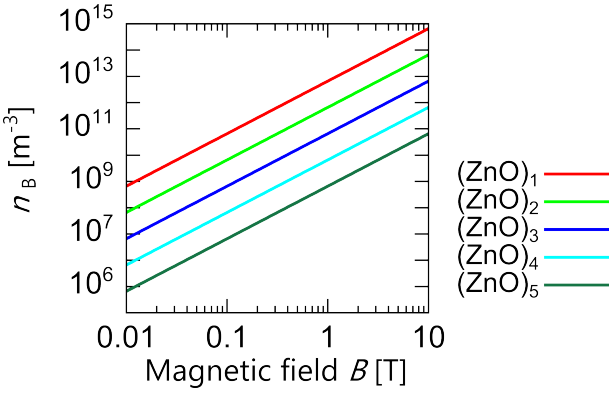

Fig. 1: Brillouin density limit n_B.

Calculating ZnO cluster growth time by Becker-Döring model

Let us estimate the growth time of clusters in the Penning trap. To calculate it, we firstly adopt the Becker-Döring model [2]. The particle number of a cluster containing N molecules is written as $n_N(t)$.

Assuming that most molecules are monomer with $N = 1$ and particle numbers increase or decrease only by either a binary collision or/and coalescences between a cluster and a monomer or the disaggregation of a monomer as shown in Fig. 2, the time evolutions of the particle numbers of N-sized cluster are expressed as

$$\begin{cases} \dfrac{\partial n_N(t)}{\partial t} = n_1(t)n_{N-1}(t)\sigma_{N-1} - n_1(t)n_N(t)\sigma_N \\ \qquad\qquad + n_{N+1}(t)\lambda_{N+1} - n_N(t)\lambda_N \quad (N \geq 2) \\ \dfrac{\partial n_1(t)}{\partial t} = -2n_1(t)^2\sigma_1 - n_1(t)\displaystyle\sum_{N=2}^{\infty} n_N(t)\sigma_N \\ \qquad\qquad + 2n_2(t)\lambda_2 + \displaystyle\sum_{N=3}^{\infty} n_N(t)\lambda_N \quad (N = 1) \end{cases}$$

, where σ_N is the product of the cross-section between N-sized cluster and monomer and collision velocity, and λ_N is the rate of disaggregation of a monomer. Thus, time evolutions of the particle number for different sized clusters are calculated by solving these equations.

Here one notes that in this calculation, the particle velocity is assumed to be the thermal velocity, the Zn-O bond distance $r_{\mathrm{Zn-O}}$ is 0.18 nm, and $\sigma_N = \pi\left(r_{\mathrm{Zn-O}}/2\left(1+N^{1/3}\right)\right)^2 v_{\mathrm{th}}$. The final assumption would be valid because the cross section becomes larger proportion to the superficial area of the cluster.

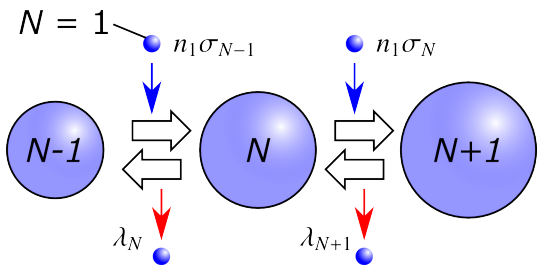

Fig. 2: Changes in cluster size.

Results

Figure 1 shows number densities against the confinement time of $(ZnO)_1$ monomer for the case of $n_1 \sim 10^{11}\,[\mathrm{m}^{-3}]$ at $B = 1\,[\mathrm{T}]$. Therefore, for the initial condition of the monomer particle with $n_1 = 10^{11}\,[\mathrm{m}^{-3}]$, $n_N(t)$ is calculated as Fig. 3

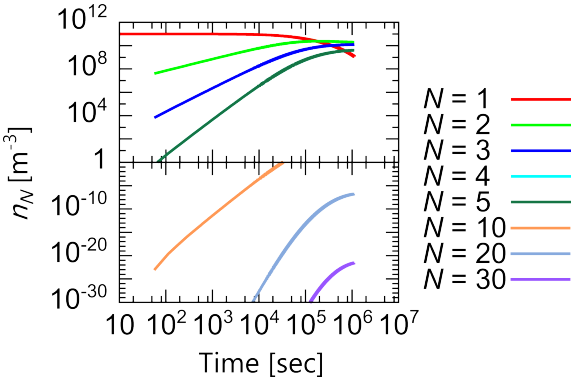

Fig. 3: Density distribution in the initial condition of $n_1 = 10^{11}\,[\mathrm{m}^{-3}]$.

As recognized from these results, no clusters bigger than ($N = 10$) are formed within the calculation time.

Fig. 4 shows the dependence of cluster numbers to the initial monomer particle number $n_1(t)$.

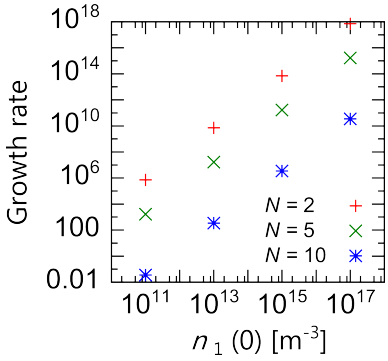

Fig. 4: The dependence of growth rate to initial monomer particle number.

As seen from the data, values of the growth rate extend to wide ranges of power. Thus, this suggests that the growth of clusters strongly depends on the number densities of the monomer particle.

More detailed analysis will be presented.

Reference

[1] H. Murakami , T. kanayama, Appl. Phys. Lett. **67** (16) (1995)

[2] R. Becker , W. Döring, Ann. Phys. **24** 719 (1935)