Growth processes of magnetic clusters studied by direct simulation Monte Carlo method

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Film formation with deposited magnetic clusters has attracted strong attention as a new manufacturing technique to realize high-density magnetic recording media and to create materials with unique magnetic properties. Such clusters are typically obtained by adiabatic expansion of a metal vapor. It is therefore important to clarify the growth mechanism since this has a profound effect on the cluster magnetic moment. In this article a new simulation method based on a combination of Direct Simulation Monte Carlo (DSMC) and a cluster collision model is introduced to examine the effect of experimental conditions in cluster beam growth. We simulate the behavior of clusters and inert gas atoms in the flight path under different experimental conditions. In particular, we find a bimodal size distribution curve and a decreasing average moment as a function of flight path length. © *2000 American Institute of Physics.* [S0021-8979(00)81008-7]

I. INTRODUCTION

Recently, cluster deposition methods have been intensely studied as a means to create systems with precisely tailored properties, leading to the suggestion to use such techniques as novel ways to manufacture high-density magnetic recording media. For the production of a three-dimensional artificial lattice this method is considered to be better than conventional sputtering and chemical vapor deposition. As a result, in recent years there have already been several reports on size controlled clusters[1,2] which aim to realize the magnetic layer system. Numerous attempts have been made by researchers to demonstrate that cluster formation in a plasma-gas-condensate[1–4] is a useful method for size-control. It is agreed that the magnetic properties depend strongly on the cluster size, which ranges from a few atoms to about 1000 atoms.[5] In a previous paper,[6,7] we proposed that size and anisotropy controlled clusters could be used as building units to create a three-dimensional artificial lattice with different magnetic properties in each embedded cluster. However, the relation between the cluster growth process during the flight path and the cluster size has not yet been clarified. Growth is affected by many factors, such as the flight path length, the ratio of the material gas to the inert gas, and the temperature of the outer wall of the equipment. Since the effect of these experimental conditions is unknown it is difficult to optimize the operating conditions to create the desired magnetic medium. In fact, the strong interdependence of reactor design, experimental conditions, and growth mechanism poses a great challenge in trying to predict the behavior of a cluster beam deposition based on intuition alone.

The effect of the outer wall temperature on cluster beam growth and deposition remains an unsettled question, with different groups reporting conflicting findings. The experimental results by Haberland *et al.*[1] and Tanemura *et al.*[8] reveal that the cooling along the flight path produces a large number of small size clusters because formation of the cluster embryo is promoted. In contrast, Brechignac *et al.*[9] show that the effect of cooling leads to large clusters because the flight time under cooling conditions becomes longer. This fundamental problem in the operation conditions to control cluster size is still in controversy.

In simulation schemes the gas flow is treated as either a continuum or a molecular flow. Theoretically, continuum (fluid) approximations of the transport equations begin to break down at low pressure. These criteria are decided by the Knudsen number $K_n = \lambda / L$ which is defined by the ratio of the mean free path of the gas λ to the characteristic length of the system L. In the large K_n region, continuum models, such as the compressible Navier–Stokes equation do not hold. In other words, when λ becomes comparable to L, the linear transport relationship for mass, diffusion, viscosity, and thermal conductivity is no longer valid. For this reason, discrete models are proposed to examine the behavior of the rarefied gas flow.

A method to treat the large K_n region is the so-called Direct Simulation Monte Carlo (DSMC) method which gives a solution to the Boltzmann equation without any assumptions on the form of the distribution function. The method is appropriate for predicting transitional and free molecular flows. It was first introduced by Bird[10] and has been extensively used to study model chemical reactions by Anderson *et al.*[11] In the DSMC method, the sampled particles are traced over the entire system to model the Boltzmann distribution by a large number of particles. The DSMC and modi-

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fied DSMC methods have been used to describe gas flow and related phenomena of rarefied gases, leading to descriptions of phenomena such as the step coverage in chemical vapor deposition,[12] chemical reactions in a plasma reactor,[13] and homogeneous nucleation in a rarefied gas.[14] These papers show that the DSMC method is a powerful technique to estimate gas flow, heat, and mass transfer under rarefied conditions. In this research we apply the DSMC to the cluster beam deposition process to examine the effects of the experimental conditions on the size distribution of cluster. The present results reveal that the cluster growth process and the size distribution are strongly dependent upon the experimental conditions.

II. MODEL AND NUMERICAL METHOD

The central assumption of the DSMC method is that collisions in the gas are completely random, therefore in the conventional DSMC simulation only the velocities of the particles are kept. However, since our purpose is to estimate the effects of the geometry and the temperature on the cluster growth, we have to retain both position and velocity information. For details on the setting up of the calculations we refer the reader to a recent publication devoted to a full description of the simulation conditions[15] and in the following restrict ourselves to a brief outline.

In this model, we only consider three types of collision reactions: (i) Simple collision processes, (ii) sublimation processes, and (iii) sticking processes. Thus fragmentation and rearrangement processes are not considered. We justify whether cluster growth occurs or not by determining the binding energy of each cluster. Namely after the collision in the raw material, the difference between the sublimation and the sticking processes is determined by the internal energy of the cluster. The internal energy, which consists of rotational and vibrational energies, is limited by the binding energy which depends on the cluster size. We assume that the binding energy in each cluster is given by volume and surface terms:[16]

$$E_b(k) = a_v k + a_s k^{2/3}, \tag{2.1}$$

where a_v and a_s are constants, depending on the material, and k is the number of monomers in the cluster. We divide the entire system into a $10\times10\times1$ mesh, each mesh including about 100 to 1000 particles, with clusters colliding against other particles that belong to the same mesh, in order to solve the Boltzmann equation.

Figure 1 shows a schematic illustration of this simulation model. The vapor of the raw material comes from the left side and the place of the growth process is located between the vapor source and the nozzle. This distance corresponds to the growth length L_g in the real equipment. The inert gas plays the role of a heat transfer mechanism from the cooling wall within the growth process by diffusive reflection. At the source of the vapor, the total gas is assumed consist solely of monomers and the velocity distribution is a Maxwell distribution.

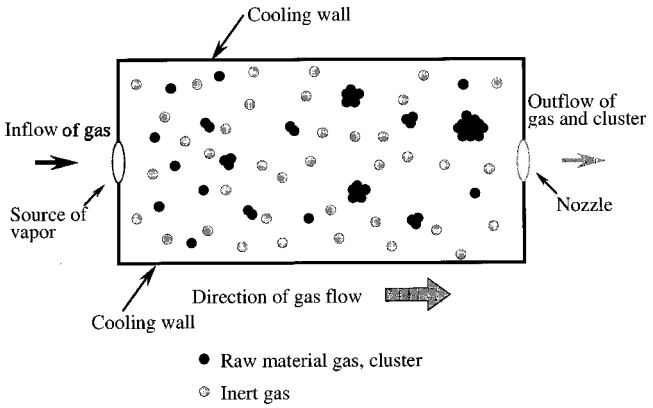

FIG. 1. Schematic illustration of the cluster growth apparatus. The large arrow indicates the rarefied gas flow of the raw material and inert gas. During the flow of raw material along the flight path cluster growth occurs.

III. RESULTS AND DISCUSSION

Figure 2 shows a typical size distribution of the clusters at the nozzle in the present model. In this figure, the horizontal axis indicates the cluster radius normalized by that of a monomer and the vertical axis shows the number of clusters. The number of monomers in the cluster is proportional to the cube of the radius of the cluster in Fig. 2. The main feature of these distribution curves is their bimodality, with a large peak for very small radius and a second maximum at larger sizes. In the small radius region, the figure indicates a large number of small clusters, such as monomers, dimers, trimers, and so on. However, in this region, the volume of raw material is negligibly small, because the size of the cluster is quite small. For the larger size region, the peak in the distribution shows that the size increases and the number

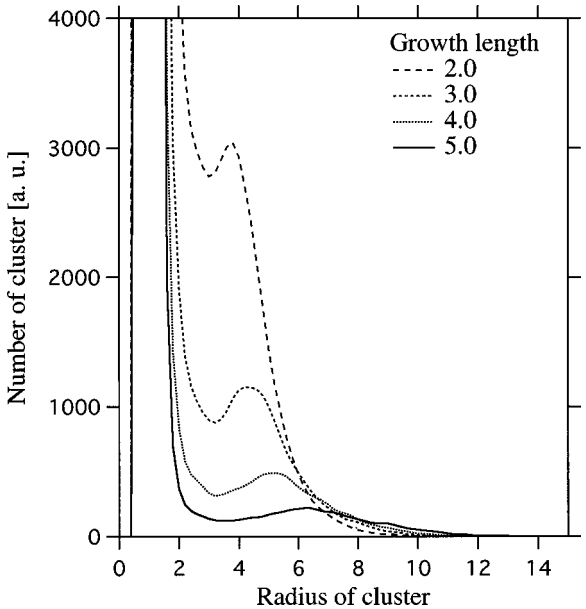

FIG. 2. Typical size distributions of the clusters at the nozzle. The concentration of the inert gas is 10% and the temperature of the wall is 1.0. The cluster radius is normalized by that of the monomer.

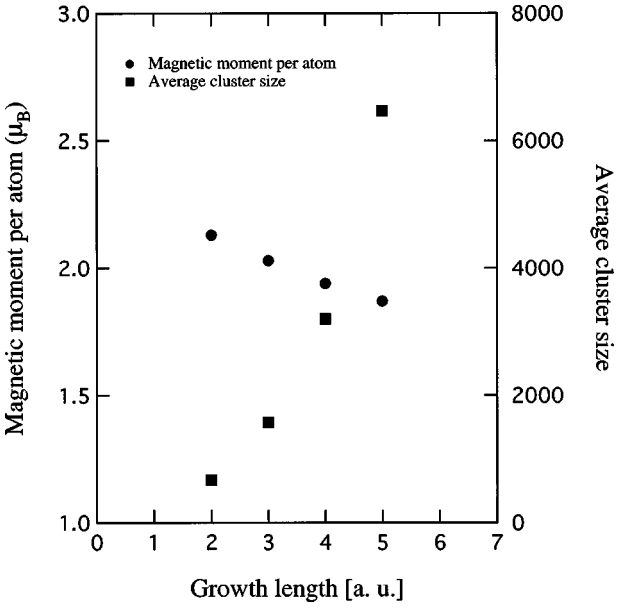

FIG. 3. Average magnetic moment per atom and average cluster size under various conditions corresponding to Fig. 2 as a function of the growth length.

decreases with an increase of the growth length. These results show corresponding previous experimental results of Fig. 7 in Ref. 3.

There is a fairly general agreement that the magnetism of a cluster depends on its size.[5] Here we use a simple model to calculate the cluster moment by relating it to the distribution of cluster sizes, which shows the feasibility of producing size-controlled magnetic clusters by means of controlling the experimental conditions. Figure 3 reveals the magnetic moment per atom under the various conditions corresponding to Fig. 2. In this estimation, we have assumed that the cluster material is Co and that the cluster size dependence of the magnetic moment may be obtained from Fig. 2(B) in Ref. 5. This figure shows that the magnetic moment decreases with increasing growth length, since the magnetic moment decreases with the increases of the cluster size.

In the present model, the time and length scales are in arbitrary units and we assume a binding energy expressed by surface and bulk terms. However, in the small cluster region it is clear that the energy of the cluster cannot be expressed by such a simple equation. Moreover, it is generally agreed that in the initial stages of cluster nucleation a three-body collision is necessary in the gas phase to release the latent heat of condensation, and that only after the cluster becomes larger two-body collisions are dominant for cluster growth. Namely, once the embryo of a cluster is formed subsequent growth occurs by cluster–cluster collisions.[17] However, this behavior is not completely understood and more detailed calculations are necessary to examine the internal energy and collision processes of the initial stages of embryo formation. Further work should also be done to elucidate the region of magic numbers, by means of a new calculation scheme based on a combination DSMC and first-principles method.[15]

IV. CONCLUSIONS

The DSMC was applied to the magnetic cluster growth process to examine the effect of the experimental condition on the size distribution and magnetic moments of clusters under various conditions. The results show clearly that the size distributions are strongly affected by the flight path length. Moreover, the average magnetic moments of the cluster also show a strong correlation with these variables. Although we have used very simple growth and moment formation models, the present results reveal that the desirable cluster sizes can be obtained by controlling the operation conditions opening the way to precisely fine-tuned magnetic nanostructures.

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