PHYSICAL REVIEW B VOLUME 40, NUMBER 8 15 SEPTEMBER 1989-I

Fractal aggregation of magnetic particles in Ag-Co thin-film surfaces

J. R. Ding

Department of Materials Science and Engineering, Tsinghua University, Beijing 100084, China

B. X. Liu

Center of Condensed Matter and Radiation Physics, Chinese Center of Advanced Science and Technology (World Laboratory), P.O. Box 8730, Beijing, China

and Department of Materials Science and Engineering, Tsinghua University, Beijing 100084, China

(Received 21 March 1989; revised manuscript received 29 June 1989)

Fractal aggregations of Co particles were observed in $Ag_{30}Co_{70}$ thin films after the films were irradiated by 200-keV xenon ions to a dose greater than 8×10^{15} Xe atoms/cm^2. It was revealed that the observed fractals were the result of cluster diffusion-limited aggregation of magnetic particles. The fractal dimension D_f was determined to be 1.47 ± 0.02, which was in agreement with those reported in the literature. The generalized dimensions $D(q)$ and the f-α spectra were also calculated and compared to previously published results.

There has been much theoretical and experimental progress in the understanding of fractal phenomena. A variety of experiments on fractal aggregation and growth, such as electrochemical deposition,[1,2] dielectric breakdown,[3] viscous fingering,[4] irregular dendritic crystal growth,[5,6] and ion-induced fractals in thin solid films,[7,8] have been reported, and some models, such as the diffusion-limited-aggregation model[9,10] (DLA), the cluster diffusion-limited-aggregation model[11] (CDA), and the Niemeyer-Pietronero-Wiesmann model[3] (NPW), have been proposed and studied intensively. Recently, some authors reported their works on the particle aggregation with magnetic dipolar interactions,[12–14] and the corresponding computer simulations were performed by Mors, Botet, and Jullien.[15] In this Rapid Communication, we report a new experimental result on this issue, namely, the fractal aggregations of ferromagnetic particles (Co) in the surface layer of Ag-Co alloy films.

First, the multilayered samples were prepared by evaporating pure silver and cobalt alternatively onto the NaCl single-crystal substrates. Each multilayered film consisted of three Ag and three Co layers. The total thickness of the thin films was about 50 nm, and the relative thickness of the two components was adjusted to meet the designed composition. In this study, the composition was $Ag_{30}Co_{70}$. The evaporation was conducted in a system with a vacuum level better than 2×10^{-6} Torr. After evaporation, the multilayered samples were irradiated to a variety of doses ranging from 1×10^{15} to 2×10^{16} Xe atoms/cm^2 with 200-keV xenon ions at room temperature. The vacuum level was about 1×10^{-5} Torr during irradiation and the ion beam density was about 1 μA/cm^2. After dissolving the NaCl substrates by deionized water, the postirradiated films were collected on the Cu or Mo grids for transmission electron microscopy (TEM) examination. The details of the experimental process can be found in our previous publication.[16]

TEM examinations were carried out in a H-800 and/or JEM-200CX electron microscope. We observed the fractal aggregates of Co particles in the samples when the irradiation dose was greater than 8×10^{15} Xe atoms/cm^2. When the dose was less than 8×10^{15} Xe atoms/cm^2, no fractal aggregates were observed. Figure 1 shows a bright field image of a fractal aggregate observed in the Ag_{30}-Co_{70} sample after irradiating to a dose of 1×10^{16} Xe atoms/cm^2. TEM, energy-dispersive spectrum (EDS), and Rutherford backscattering spectrum (RBS) analyses revealed that the fractal aggregates existed in the surface layer of the alloy films. The fractal patterns observed in this study were very similar to those reported by other authors.[12–15] The average diameter of the magnetic particles in the fractal aggregates was about 80 nm. Figure 2 shows the distribution of the fractals in the same sample. In the film, there are many fractals and other small clusters (black) which are still in the aggregating stage. This experimental result indicated that the observed fractal aggregates were the result of cluster diffusion-limited aggregation. EDS analysis showed that both the fractal aggre-

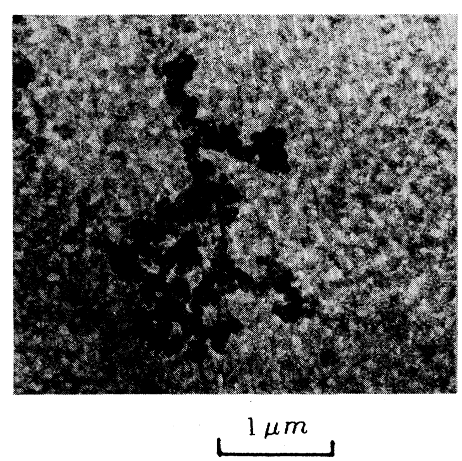

FIG. 1. A fractal aggregate observed in the surface layer of $Ag_{30}Co_{70}$ thin film. The irradiation dose was 1×10^{16} Xe$^+$/cm^2 with 200 KeV Xe$^+$. The fractal dimension is 1.47 ± 0.02.

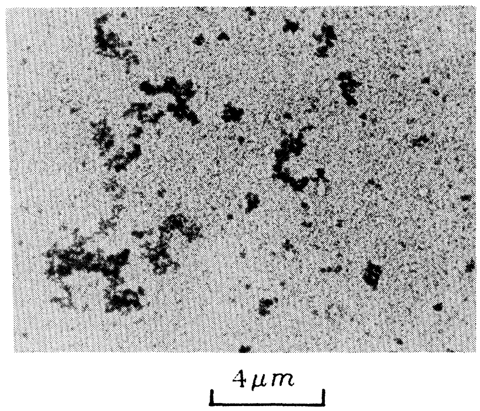

FIG. 2. The distribution of the fractal aggregates in the surface layer. The black area is cobalt, and the grey region is a mixture of Co and Ag.

gates and the clusters (black) were composed of Co particles. The matrix (grey) was a mixture of silver and cobalt, confirmed by electron diffraction analysis. The fractal pattern was analyzed by an image processor with a resolution of 512×512 pixels. The fractal dimension D_f was determined to be 1.47 ± 0.02, and was nearly the same for the fractals observed in different samples. The fractal dimension in our case, 1.47 ± 0.02, is smaller than the one expected by the CDA model, and is slightly larger than those reported earlier. The different matrices, on which the aggregation took place, may be a crucial point to understand the difference.

We now discuss the growth processes of the observed fractal aggregates. In the preparation of the multilayered samples, the outermost layer was ferromagnetic materials, i.e., Co. When 200-keV xenon ions irradiated the samples, mixing between two components occurred. The mixing efficiency was low, since Ag-Co binary alloy systems is immiscible and has a rather positive heat of formation (+26 kJ/mol).[17] When the dose went to a high level reaching 5×10^{15} Xe atoms/cm^2, the sample was uniformly mixed according to RBS analysis, which also determined the average composition of the alloy film being $Ag_{30}Co_{70}$. From the sputtering theory, we know that the sputtering yield of Ag (3.6) is larger than that of Co (1.5). Hence, the percentage of Co in the surface layer was larger than that of average after high dose irradiation. RBS analysis revealed that the surface composition was about 80 at.% of Co. Consequently, in those films that had been irradiated to a dose greater than 8×10^{15} Xe atoms/cm^2 there were many Co particles distributed uniformly and randomly in the surface. As mentioned above, Ag and Co are immiscible, diffusion of Co particles in the surface layer is relatively easy. Cobalt diffused and aggregated in the surface layer thus forming particles. When the radius of Co particles was larger than 4 nm, the particles became ferromagnetic.[18] The strong magnetic interactions among the Co particles then came into play and influenced the further aggregation, resulting in a smaller mass dimension (1.47 ± 0.02) of the final aggregates.

Similar experiments were also performed in the Fe-Cu binary-alloy system.[19] The composition of the samples was $Fe_{80}Cu_{20}$. The magnetic particles, which formed the fractal aggregates during the aggregation process in the surface layer, were Fe particles as confirmed by the EDS and the Auger-electron spectrum (AES). The diameter of the Fe particles in the aggregates was greater than 50 nm. The fractal dimension of the fractals was determined to be 1.42 ± 0.02. Obviously, D_f (Fe) is slightly smaller than D_f (Co), and this difference can probably be attributed to the difference in magnetic interactions involved in Fe and Co cases.[13-15] It should be pointed out that after storage the Fe films in air for several days, the Fe particles observed in the fractal aggregates were oxidized into Fe_2O_3, as detected by AES and observed by TEM. However, the shape of the preformed fractals remained unchanged, meaning the aggregation process was governed by Fe particles, but not Fe_2O_3 particles observed later.

In order to compare the fractal aggregates with magnetic dipolar interaction to other fractal aggregates, we calculated the generalized dimensions $D(q)$ with q ranging from -10 to $+10$, as well as the f-α spectra. The fractal patterns were put into a Laplacian field with the following conditions: The fractal pattern itself was equipotential (the potential was set to be 0), and the outer boundary was also equipotential (the potential was set to be 1), the diameter of the outer boundary was about three times larger than the fractal diameter. The Laplacian potentials were then calculated numerically. The Laplacian-potential gradient of each growth point was used for the growth probability at this point. The generalized dimensions $D(q)$ and the f-α spectra were calculated according to the following definitions:[20,21]

$$D(q) = \lim_{(\epsilon\to 0)} (q-1)^{-1} \ln\left[\sum_i p_i(\epsilon)^q\right] \Big/ \ln(\epsilon)\,, \quad (1)$$

$$\alpha(q) = \frac{d}{dq}(q-1)D(q)\,, \quad (2)$$

$$f[\alpha(q)] = q\alpha(q) - (q-1)D(q)\,. \quad (3)$$

Here $p_i(\epsilon)$ was the growth probability accumulated within the ith box of size ϵ. Figure 3 shows the general-

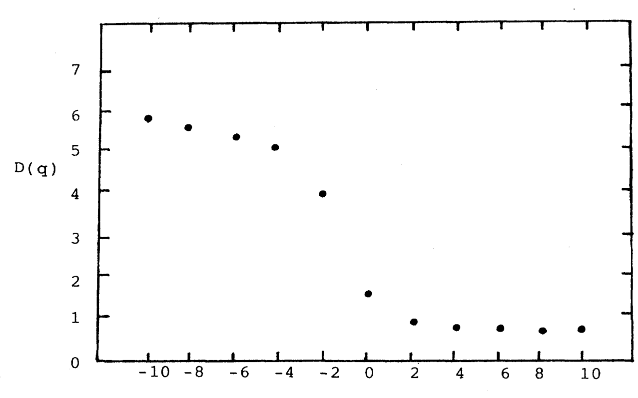

FIG. 3. The plot of the generalized dimensions $D(q)$ vs q of the fractal structure shown in Fig. 1. The Laplacian-potential-gradient measure was used.

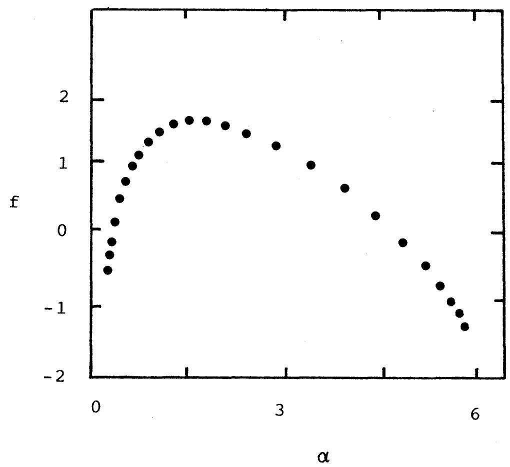

FIG. 4. The f-α spectrum of the observed fractal structure with the Laplacian-potential-gradient measure.

ized dimensions $D(q)$ vs q, and Fig. 4 shows the f-α spectrum.

The information dimension $D(1)=1.1$, and the correlation dimension $D(2)=0.79$. In the limit of $q\to\infty$ and $q\to-\infty$, $D(\infty)=0.62$ and $D(-\infty)=5.6$. Our results are slightly different from those reported by Ohta and Honjo.[21]

In conclusion, the fractal aggregation of magnetic particles (Co) was observed in the surface layer of Ag-Co alloy films after irradiations. The generalized dimension $D(q)$ and the f-α spectra were calculated, and showed that the observed fractals kept multifractal structures.

The authors are grateful to Dr. L. J. Huang and researchers of the Transmission Electron Microscopy Laboratory of Beijing University and the Analysis Center of Tsinghua University for their help. The financial aid from the International Atomic Energy Agency (Contract No. 4731/R1/RB) as well as by the National Natural Science Foundation of China is also acknowledged.

[1]M. Matsushita, M. Sano, Y. Hayakawa, H. Honjo, and Y. Sawada, Phys. Rev. Lett. **53**, 286 (1984).

[2]R. M. Brady and R. C. Ball, Nature (London) **309**, 225 (1984).

[3]L. Niemeyer, L. Pietronero, and H. J. Wiesmann, Phys. Rev. Lett. **52**, 1033 (1984).

[4]L. Paterson, Phys. Rev. Lett. **52**, 1621 (1984).

[5]H. Honjo, S. Ohta, and M. Matsushita, J. Phys. Soc. Jpn. **55**, 2487 (1986).

[6]L. J. Huang, H-D. Li, and B. X. Liu, Nuovo Cimento **10D**, 1117 (1988).

[7]B. X. Liu, L. J. Huang, K. Tao, C. H. Shang, and H-D. Li, Phys. Rev. Lett. **59**, 745 (1987).

[8]L. J. Huang, J. R. Ding, H-D. Li, and B. X. Liu, J. Appl. Phys. **63**, 2879 (1988).

[9]T. A. Witten and L. M. Sander, Phys. Rev. Lett. **47**, 1400 (1981).

[10]T. A. Witten and L. M. Sander, Phys. Rev. B **27**, 5686 (1983).

[11]R. F. Voss, J. Stat. Phys. **36**, 861 (1984).

[12]G. A. Niklasson, A. Torebring, C. Larsson, C. G. Granqvist, and T. Farestam, Phys. Rev. Lett. **60**, 1735 (1988).

[13]G. Helgessen, A. T. Skjeltorp, P. M. Mors, R. Botet, and R. Jullien, Phys. Rev. Lett. **61**, 1736 (1988).

[14]S. G. Kim and J. R. Block, J. Appl. Phys. **60**, 509 (1986); J. Colloid Interface Sci. **116**, 431 (1987).

[15]P. M. Mors, R. Botet, and R. Jullien, J. Phys. A **20**, L975 (1987).

[16]B. X. Liu, Phys. Status Solidi (a) **94**, 11 (1986).

[17]W. L. Johnson, Y. T. Cheng, M. Van Rossum, and M-A. Nicolet, Nucl. Instrum. Methods Phys. Res. Sect. B **7** and **8**, 657 (1985).

[18]B. D. Cullity, *Introduction to Magnetic Materials* (Addison-Wesley, Reading, MA, 1972), p. 410.

[19]J. R. Ding and B. X. Liu (unpublished).

[20]T. C. Halsey, M. H. Jensen, L. P. Kadanoff, I. Procaccia, and B. I. Shraiman, Phys. Rev. A **33**, 1141 (1986).

[21]S. Ohta and H. Honjo, Phys. Rev. Lett. **60**, 611 (1988).

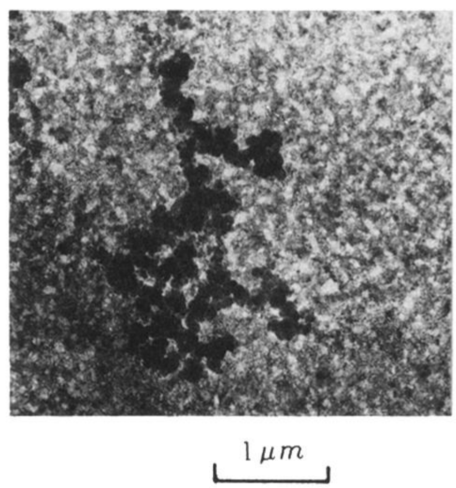

FIG. 1. A fractal aggregate observed in the surface layer of $Ag_{30}Co_{70}$ thin film. The irradiation dose was 1×10^{16} Xe^+/cm^2 with 200 KeV Xe^+. The fractal dimension is 1.47 ± 0.02.

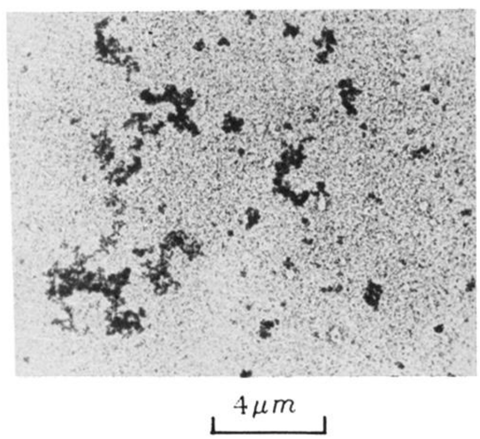

FIG. 2. The distribution of the fractal aggregates in the surface layer. The black area is cobalt, and the grey region is a mixture of Co and Ag.