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* In Kazan University the Electron Paramagnetic Resonance (EPR) was discovered by Zavoisky E.K. in 1944.

Computer simulation of growth and magnetic properties of quasi 2D magnetic cluster†

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The computer simulation of the formation of 2D magnetic cluster is performed. It is shown that the magnetic interaction between growing cluster and randomly walking atoms results in the cluster elongation in the cluster magnetization direction. The features of microwave field absorption (FMR or SPR signals) in thin granular film consisting of elongated clusters are discussed. Magnetic resonance measurements can provide the detailed information on shape anisotropy of the particles and give an evidence for considered model of magnetic cluster growth.

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1. Introduction

The method of ion-beam synthesis is a very effective way to obtain thin films of new chemical phases within the surface layer of any target. In particular, high-dose implantation of iron ions into the silicon results in the formation of thin films of different silicides FeSi, Fe_2Si, Fe_3Si, depending on the implantation dose [1]. It is assumed that the synthesized films are not continuous and consist of clusters of the silicide phase. Of particular interest are thin films of magnetic silicide Fe_3Si as a promising material for microelectronics and spintronics. Moreover, when ion-beam synthesis is carried out in presence of external magnetic field parallel to the target surface, the synthesized films reveal noticeable magnetic in-plane anisotropy [2]. It was suggested that shape anisotropy of created ferromagnetic Fe_3Si clusters constituting the film may be considered as one of the reasons for such effect. In this paper the process of magnetic cluster growth in external magnetic field and the features of magnetic resonance in the film, consisting of the clusters, are discussed.

The formation of magnetic Fe_3Si silicide clusters begins with a creation of stable grain of this new phase. During ion bombardment along with the creation of the grains the process of their destruction due to atomic collisions takes place. Hence, an initial randomly created grain of the Fe_3Si phase should reach a certain critical size to become stable and serve as a nucleus of the Fe_3Si cluster growth. It can be assumed that the process has the noticeable probability only within thin layer, where the concentration of implanted Fe atoms is maximal. Therefor the computer simulation of cluster growth in 2D system can be used to study geometrical properties of created clusters. For this purpose the Witten-Sander model of the particle-cluster diffusion-limited aggregation (DLA) [3] is widely used. There are numerous modifications "sticking" of moving particle to the growing cluster (the different probabilities of the process).

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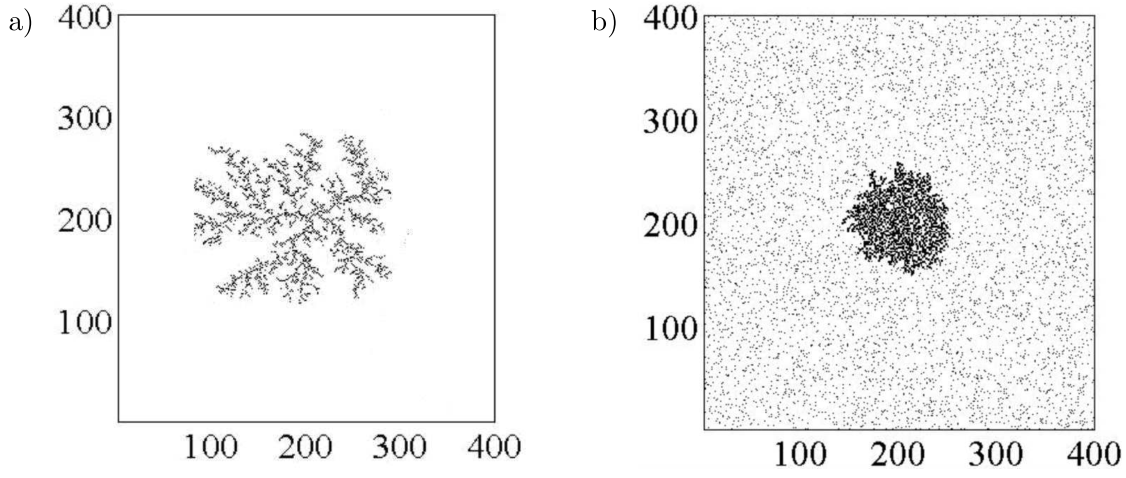

Figure 1. Clusters generated according to classical DLA model (a) and in frame of the model considered in present paper (b). The number of Fe atoms included in cluster in both cases is the same.

In the case of the formation of magnetic (ferromagnetic) clusters from diffusing magnetic particles it is necessary to take into account the influence of the magnetic field created by the cluster on the motion of particles near the cluster surface. Such type of the anisotropic diffusion of magnetic particles was considered in the description of the growth of electrodeposited ferromagnetic aggregates in the applied magnetic field [4] and the shape transformation of iron precipitates in alloys under the strong magnetic field [5]. The modified multi-center DLA model, taking into account the effect of the dipole-dipole magnetic interaction between the cluster and randomly walking atoms was developed in [6]. It was shown that the interaction results in cluster elongation in the direction of the cluster magnetization. Thus, if growing in the external magnetic field cluster elongates along the field.

In all mentioned modifications of DLA model the basis of the original DLA approach remains: the cluster grows by taking the diffusing particles up one by one and each of the particles becomes a new unit of the cluster. However, the atoms embedded into the solid react with the host atoms and form a new chemical compound. Such a situation occurs during the ion-beam synthesis of Fe_3Si silicide when not one but several Fe atoms must come near the cluster boundary at a time to add a new unit (with Fe_3Si structure) to the cluster. To take into account this process a modified model was developed [7]. In the model a new unit is added to the cluster in the site of the cluster perimeter near which the local concentration of randomly walking Fe atoms becomes sufficient to provide the Fe_3Si phase composition. The examples of clusters generated in frame of original DLA approach and according to the suggested model are presented in Fig. 1. The figure shows that in case B) generated cluster has a much denser structure.

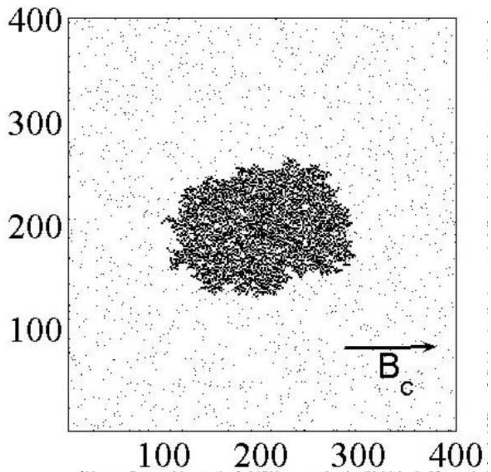

Figure 2. Cluster simulated in the presence of the external magnetic field directed along the OX axis.

In the considered model the influence of the magnetic field created by the cluster of magnetic silicide Fe_3Si on the motion of iron atoms near the cluster also take place. Accounting of the magnetic interaction results in cluster elongation (see Fig. 2). The simulation of the cluster growth was performed on square lattice (400×400). The model critical size of the stable "grain" is taken to be $3a$ (a is the lattice constant). Each of 10000 free iron atoms makes 10 steps during each stage of the simulation. It was assumed that the saturation magnetization M_S of the growing ferromagnetic cluster is equal to that for silicide Fe_3Si ($M_S = 900\,\mathrm{emu/cm^3}$). All calculations were performed using the C++ code compiled under Linux OS.

Of course, in a real situation each cluster within the film is not two-dimensional but has a certain size in the direction perpendicular to the plane of the implanted layer. However, it is conceivable that found above effect of cluster elongation along the applied magnetic field will remain. Thus, ion-beam synthesized magnetic film can be considered as a planar system of elongated ellipsoids, the long axes of which lie in the implanted layer plane.

2. Magnetic resonance

Magnetic resonance method (ferromagnetic resonance (FMR), superparamagnetic resonance (SPR)) is usually used to obtain the information about the shapes of particles constituting the granular film, their size distributions and inter-particle interactions. In the scientific literature there are a great number of theoretical and experimental works devoted to the study of the FMR in granular systems (see the detailed overview [8] and references in it). The theoretical description of microwave absorption in real granular magnetic film is very complex, multi-parameter problem, but if the particle density in the film is sufficiently low the inter-particle interactions can be neglected and one can easily obtain the microwave absorption for a single particle. Then, the expression for absorption power must be averaged on the particle parameter distributions over the film to obtain the experimentally observed FMR signal. Below the simple calculation of FMR for planar system of elongated and non-interacting ellipsoids is made and several features of calculated signals connecting with the shape anisotropy of particles are discussed.

The microwave (at the frequency ω) absorption $P(\omega)$ in the film can be represented as the sum of absorption signals $P_i(\omega)$ for i-th ellipsoid:

$$P(\omega) = \sum_i P_i(\omega). \tag{1}$$

According to Kittel approach [9] the magnetization $\vec{M}_i$ of i-th ellipsoid in external magnetic fields is obeyed the equation:

$$\frac{\partial \vec{M}_i}{\partial t} = \gamma \left(\vec{M}_i \times \vec{H}_i \right) + R_i, \tag{2}$$

where $\vec{H}_i = \vec{H}_0 - 4\pi \hat{N}_i \vec{M}_i + \vec{h}(t)$, $\vec{H}_0$ is the static magnetic field, $\vec{h}(t)$ is the microwave fields, $\hat{N}_i$ is the demagnetization tensor and R_i is the term describing magnetization relaxation. In the principle axes of ellipsoid demagnetization tensor is written as

$$\hat{N}_i = \begin{pmatrix} N_{ix} & 0 & 0 \\ 0 & N_{iy} & 0 \\ 0 & 0 & N_{iz} \end{pmatrix}, \tag{3}$$

where $N_{ix} < N_{iy} < N_{iz}$ (the elongation is supposed along x-axis). Regarding the relaxation term R_i in the equations (2) it should be noted the following. The differences in the demagnetization factors (the presence of the easy magnetization axis of the ellipsoid) cause a predetermined orientation of the equilibrium magnetization $\vec{M}_{i0}$ in the absence of a microwave field. The energy dissipation processes will seek to bring the magnetization to this direction. It is convenient to write the relaxation terms and to solve the equations (2) in the coordinate system in which one of the axes coincides with the equilibrium direction for the magnetization.

The FMR absorption lines are usually observed in two static field $\vec{H}_0$ orientations: perpendicular one, when $\vec{H}_0$ is normal to the film plane and $\vec{h}(t)$ lies in the plane (case A) and parallel orientation, when $\vec{H}_0$ lies in the film plane and $\vec{h}(t)$ is normal to the plane (case B). Below both of these cases are briefly considered.

Case A. When $\frac{H_0}{M_S(N_{iz}-N_{ix})} > 1$ the magnetization relaxes toward direction of static field $\vec{H}_0$ (along z-axis) and equations (2) in usual stationary resonance approximation can be written as:

$$\begin{aligned} \frac{\partial M_{ix}}{\partial t} &= \omega_{11} M_{iy} - \gamma h(t) \sin\alpha M_s - \frac{M_{ix}}{T}, \\ \frac{\partial M_{iy}}{\partial t} &= \omega_{12} M_{ix} - \gamma h(t) \cos\alpha M_s - \frac{M_{iy}}{T}, \\ M_{iz} &\approx M_S, \end{aligned} \tag{4}$$

where α is the angle betwenn $\vec{h}(t)$ and longest axis of ellipsoid. In (4) the Bloch-type relaxation terms $R_{ix,y} = -\frac{M_{ix,y}}{T}$ are introduced. The frequencies ω_{11} and ω_{12} are determined as follow:

$$\begin{aligned} \omega_{11} &= \gamma H_0 - \gamma\left(N_{iz} - N_{iy}\right) M_S, \\ \omega_{12} &= \gamma H_0 - \gamma\left(N_{iz} - N_{ix}\right) M_S. \end{aligned} \tag{5}$$

Equations (4) lead to the FMR absorption for i-th ellipsoid:

$$P_i(\omega) = \frac{2\pi(\gamma h)^2 M_S[\omega_{11}\cos^2\alpha + \omega_{12}\sin^2\alpha]}{T[(\omega^2 - \omega_{11}\omega_{12} + \frac{1}{T^2})^2 + \frac{4\omega_{11}\omega_{12}}{T^2}]}. \tag{6}$$

Case B. In this case static magnetic field $\vec{H}_0$ lays in the (xy) plane, where x-axis coincides with longest axis of ellipsoid, microwave field $\vec{h}(t)$ being directed along z-axis. In coordinate system (x', y', z') in which x'-axis is directed along equilibrium magnetization $\vec{M}_{i0}$ the equations (2) can be written as:

$$\begin{aligned} M_{ix'} &\approx M_S, \\ \frac{\partial M_{iy'}}{\partial t} &= \omega_{21} M_{iz'} - \gamma h(t) M_S - \frac{M_{iy'}}{T}, \\ \frac{\partial M_{iz'}}{\partial t} &= \omega_{22} M_{iy'} - \frac{M_{iz'}}{T}, \end{aligned} \tag{7}$$

where

$$\begin{aligned} \omega_{21} &= \gamma[H_0(\sin\phi\sin\theta + \cos\phi\cos\theta) + (N_{iz} - N_{iy}\sin^2\theta - N_{ix}\cos^2\theta)M_S], \\ \omega_{22} &= \gamma[H_0(\sin\phi\sin\theta + \cos\phi\cos\theta) + (N_{iy} - N_{ix}\cos^2\theta - N_{ix}\sin^2\theta)M_S]. \end{aligned} \tag{8}$$

In (8) ϕ is the angle between static magnetic field $\vec{H}_0$ and longest axis of ellipsoid and the angle θ is determined from the condition

$$H_0(\cos\phi\sin\theta - \sin\phi\cos\theta) = (N_{ix} - N_{iy})M_S\cos\theta\sin\theta. \tag{9}$$

The solving of (7) leads to the microwave absorption:

$$P_i(\omega) = \frac{2\pi(\gamma h)^2 M_S \omega_{22}}{T[(\omega^2 - \omega_{21}\omega_{22} + \frac{1}{T^2})^2 + \frac{4\omega_{21}\omega_{22}}{T^2}]}. \tag{10}$$

The expressions (6) and (10) need to be averaged on parameters of ellipsoids to obtain the experimentally observed FMR signals. To make some numerical estimations of the FMR lines (6) and (10) it can be supposed that all ellipsoids in the film have the same shape, but the orientations of ellipsoid axes in film plane may be different. Let the dimensionless axes of the ellipsoid a, b and c are supposed to be equal to: $a = 200$, $b = 180$ and $c = 20$. These values correspond to a thin cluster with in-plane elongation in x-axis direction. These parameters of the cluster lead to the following values for demagnetization coefficient [10]: $N_x = 0.095$, $N_y = 0.105$, $N_z = 0.8$. The value of the magnetization M_S is taken equal to 1000G and the frequency ω of microwave field is set equal to 9.5 GHz (X-band).

Microwave absorption in case A (Eq. (6)) is described by single resonance line with the maximum at $H^{\perp}_{\text{res}} = \frac{1}{\gamma}\sqrt{\omega_{11}\omega_{21}}$ and the width, determined by relaxation parameter $1/T$. Note, that the resonance frequency $\omega \approx \sqrt{\omega_{11}\omega_{21}}$ is not dependent on the angle α. Thus, the averaging on orientation of the ellipsoid in the film plane only leads to a change in amplitude of FMR signal, but not to its broadening. Consequently, in case A the FMR signal is the same in ordered or disordered arrangements of ellipsoids and at above introduced numerical parameters the signal is the resonance line with maximum at $H^{\perp}_{\text{res}} = 12900$ Oe.

Contrary to the case A, in case B the resonance frequency $\omega \approx \sqrt{\omega_{21}\omega_{22}}$ depends on the angle ϕ. If the arrangement of ellipsoid is ordered i.e. the longest axes of all ellipsoids have the same direction in the plane (taken as x-axis of coordinate system), the absorption maximum will be observed at $H^{\parallel}_{\text{res1}} = 880$ Oe when $\vec{H}_0$ lies along x-axis and at $H^{\parallel}_{\text{res2}} = 1320$ Oe, when $\vec{H}_0$ lies in plane and perpendicularly to x-axis. This case was considered in [11]. The numerical values were obtained using introduced above ellipsoid parameters, the width of each signal being determined by relaxation time T. If the orientation of the longest axes in the film plane is random the FMR absorption is described by inhomogeneously broadened line, practically absorption "band", extended from 800 Oe to 1200 Oe.

3. Conclusion

Finally, a simple model of the formation of Fe_3Si clusters during high-dose Fe ion implantation into silicon is considered. The magnetic interaction between growing cluster and randomly walking Fe atom results in the cluster elongation in the cluster magnetization direction. This can explain the origin of the uniaxial in-plane magnetic anisotropy in films ion-beam synthesized in the presence of an external magnetic field. The features of microwave field absorption (FMR, SPR signals) in thin granular film consisting of elongated clusters are discussed. The observation of discussed above differences between resonance lines in perpendicular ($\vec{H}_0 \perp$ film plane) and parallel ($\vec{H}_0 \parallel$ film plane) orientations of static magnetic field provides the useful information on shape anisotropy of particles constituting the magnetic granular film and can give evidence for the considered model of cluster growth.

References

1. Petukhov V., Khaibullin I., Zaripov M., Manapov R., *Fizika Tverdogo Tela* **25**, 1392 (1984) [in Russian].
2. Gumarov G., Petukhov V., Zhikharev V., Valeev V., Khaibullin R., *Nucl. Instrum. Methods Phys. Res. B* **267**, 1600 (2009).
3. Witten T., Sander L., *Phys. Rev. Lett.* **47**, 1400 (1981).
4. Cronemberger C., Sampiao L., Guimaraes A., Molho P., *Phys. Rev. B* **81**, 021403 (2010).

5. Beaugnon E., Arras E., *J. Phys.: Conf. Ser.* **51**, 439 (2006).
6. Balakirev N., Gumarov G., Zhikharev V., Petukhov V., *Comput. Mater. Sci.* **50**, 2925 (2011).
7. Balakirev N., Zhikharev V., Gumarov G., *Nucl. Instrum. Methods Phys. Res. B* **326**, 61 (2014).
8. Kliava J., *in book Magnetic Nanoparticles*, edited by S.Gubin (WILEY-VCH Verlag, GmbH@Co,KGaA, Weinheim, 2009) pp. 255–302.
9. Kittel C., *Phys. Rev.* **73**, 155 (1948).
10. Aharoni A., *J. Appl. Phys* **83**, 3432 (1998).
11. Balakirev N., Zhikharev V., *Phys. Status Solidi C* **12**, 39 (2015).