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# Ordering and 'melting' of domain lattices in thin magnetic films

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**Abstract.** By virtue of minimization of a phenomenological expression for the free energy of domain structures of soft magnetic films the conditions under which the amorphization of domain structures takes place are found. It is shown that domain structure ordering or melting may be described by a thermal model in which the agitation by an oscillating magnetic field creates an effective temperature.

It is well known that domain structures (DSs) in thin films appearing spontaneously from a homogeneous magnetic state are disordered DSs. Furthermore, DS disorder in thin magnetic films may be the result of an order–disorder transition induced by a change in temperature or an external magnetic field [1–5].

This paper aims to describe ordering and melting of domain patterns in thin soft magnetic films. Considering a domain system as a set of interacting particles, we assume the existence of thermal fluctuations which is taken into account by the addition of entropy terms in a DS thermodynamic potential. We also suggest a definition of the entropy of melting which differs from the entropy defined as that of communal sharing of the volume [6].

If we assume that, with increasing temperature, $n < N$ ($N$ is the number of particles of the lattice and $n$ is the number of defects) particles leave lattice sites and occupy neighbouring voids, the configurational part of the lattice entropy is [7]

$$S = -kN[\xi \ln \xi + (1-\xi)\ln(1-\xi)] \tag{1}$$

where $\xi = n/N \ll 1$ and $k$ is Boltzmann's constant. The equilibrium value of $\xi$ can be calculated by minimizing the free energy

$$F = F_0 + N\xi w - TS(\xi) \tag{2}$$

where $w$ is the energy of one point defect, $F_0$ is the free energy of the perfect lattice and T is the temperature.

Minimizing (2) with respect to $\xi$, we obtain

$$\xi_0 = (1 + \exp(w/kT))^{-1} \simeq \exp(-w/kT). \tag{3}$$

Since the conditions for the existence of a free-energy minimum (2) are fulfilled at any $\xi < 1$, equations (3) determines the relative concentration of the equilibrium defects in the lattice. The average cluster size of a perfect lattice is $L_c = L/\sqrt{n}$ (where $L = a\sqrt{N}$ is

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the linear system size and $a$ is the lattice period), and taking into account (3) we can write $L_c = a\xi^{1/2} = a[1 + \exp(w/kT)]^{1/2}$.

We now consider the melting of a two-dimensional lattice as a continuous type of transition. Since the defect concentration (3) increases with increasing temperature and correspondingly the average cluster size decreases, at a certain temperature the lattice becomes amorphous. It is self-evident that a lattice can be considered as amorphous when $L_c \to a$. On the other hand, we can study the threshhold of percolation of bonds in a two-dimensional lattice assuming that the orientational order vanishes (orientational order is order in the system of bonds) if the fraction of undestroyed bonds $y \leqslant y_n$ (where $y_n$ is the percolation threshold). If we define a cluster as a set of bonded lattice nodes, an infinite cluster ceases to exist when $y \leqslant y_n$. The lattice in this state can be called a liquid-crystal phase. As is well known, the thresholds of percolation for problems of bonds for flat triangular and square lattices are $y_n^{(1)} = 0.3473$ and $y_n^{(2)} = 0.5$, respectively. The corresponding values of $\xi$ for these lattices are $\xi_n^{(1)} = 0.326$ and $\xi_n^{(2)} = 0.25$, respectively. Defining the lattice entropy of melting as $\Delta S = S(\xi_n) - S(0)$ we obtain $\Delta S^{(1)} = 0.631$ kN and $\Delta S^{(2)} = 0.562$ kN; there is good agreement with the results of numerical experiments on two-dimensional melting and the well known experimental rule $\Delta S = kN \ln 2$ (see [6] and references therein).

It is seen from (3) that the equilibrium defect number of a DS is determined by the relation between the energy of nucleation of a defect and the thermal energy. The defect nucleation energy is approximately equal to the domain interaction energy. Using a dipole approximation this energy can be written as

$$w = (4\pi M)^2 c h^3 x^4 z^{-3} \tag{4}$$

where $h$ is the film thickness, $M$ is the saturation magnetization, $x = d/h$, $z = a/h$, $d$ is the average domain size and $c$ is a numerical coefficient (for a hexagonal bubble lattice, $c = 0.086$ [8]). Usually $w \gg kT$ and $\xi_0 \to 0$ in accordance with (3).

Nevertheless, there are two cases when these energies can be of the same order and the entropy term contribution to the free energy (2) becomes significant.

The first case is the temperature range just below the Curie point $T_C$. Here the saturation magnetization decreases rapidly with increasing $T$ according to $M(T) = M_0(1 - T/T_C)^\beta$, where $\beta = 0.3$–$0.5$ and $M_0 = M(0)$. In this case the equilibrium defect concentration increases as

$$\xi_0 = \exp\left(-\frac{(4\pi M_0)^2}{kT_C^{2\beta+1}} c h^3 x^4 z^{-3}\, \delta T^{2\beta}\right) \tag{5}$$

where $\delta T = T_C - T$. The triangular lattice is amorphous at $\xi_0 = \xi_n^{(1)} = 0.326$. From (5) the temperature range $\delta T$ in which a DS can exist only in a disordered state (liquid-crystal phase) is

$$\frac{\delta T}{T_C} = \left(-\frac{kT_C \ln \xi_n^{(1)}}{(4\pi M_0)^2 c h^3 x^4 z^{-3}}\right)^{1/2\beta}. \tag{6}$$

Equation (6) gives $\delta T = 10^{-2}$–$10^{-1}$ K for $4\pi M_0 = 100$ G, $h \simeq 10^{-4}$ cm, $xz^{-1} = 0.1$ and $x = 1$. Amorphization of DSs near the Curie point has been observed experimentally [1].

The second case when the contribution of thermal fluctuations leads to amorphization of DSs may also be obtained from (6). Indeed, the structure is amorphous at all temperatures

in its range of existence ($\Delta T \sim T_C$) if the expression in large parentheses is (6) is about unity. We derive the film thickness from the last condition as

$$h \leqslant \left( -\frac{kT_C \ln \xi_n^{(1)}}{(4\pi M_0)^2 c x^4 z^{-3}} \right)^{1/3}. \tag{7}$$

Equation (7) gives $h \leqslant 10^3$ Å for the above values of the parameters. Thus we come to the conclusion that DSs in very thin films must be disordered at all temperatures in their range of existence.

DSs existing in an amorphous or cluster form in thicker films should be treated as frozen, since their relaxation to an equilibrium state (i.e. to a DS with a small defect concentration) takes an essentially infinite time, since $kT \ll w$. Indeed, let a system be in a non-equilibrium state with defect concentration $\xi \neq \xi_0$. Let us find the relaxation time to an equilibrium state. The entropy production rate is obtained from (1):

$$\dot{S} = -kN \ln\left(\frac{\xi}{1-\xi}\right)\dot{\xi}. \tag{8}$$

We choose the thermodynamic flow $J$ and the thermodynamic force $X$ satisfying the relation $\dot{S} = JX$ and equation (8):

$$J = N\dot{\xi} \tag{9}$$

$$X = -k[\ln \xi - \ln(1-\xi)]. \tag{10}$$

The physical meaning of the flow is the number of particles relaxing to an equilibrium state per second. We assume that the flow is proportional to the thermodynamic force, i.e.

$$\dot{\xi} = -\frac{kL_0}{N}[\ln \xi - \ln(1-\xi)] \tag{11}$$

where $L_0$ is the Onsager coefficient. Letting the deviation $\delta\xi = \xi - \xi_0$ be small and linearizing (11) with respect to $\delta\xi$, we can obtain the relaxation time

$$\tau = \frac{N\xi_0(1-\xi_0)}{kL_0}. \tag{12}$$

Assuming that the kinetic coefficient in (12) is proportional to the temperature (in analogy with the diffusion coefficient), we find that the relaxation time tends to infinity at low temperatures. That is why we can call such amorphous and cluster structures frozen.

On the other hand, it is well known that ordering of amophous DSs can be achieved by an oscillating external magnetic field with a small amplitude $H_m$. This field is necessary not only to overcome the static coercivity barrier of domain walls but also to act as an effective temperature. This effective temperature can be inntroduced as $\tilde{T} = \langle m_0 v^2/2 \rangle$ (here the effective temperature is measured in the units of an energy and $m_0$ is the domain wall mass) taking into account domain oscillations in the vicinity of lattice nodes with velocity $v = \mu H$ ($\mu$ is the mobility coefficient of a domain wall and $H = H_m \cos(\omega t)$, where $\omega$ is the frequency of the oscillating field). For bubble lattices the following can be obtained:

$$\tilde{T} = \frac{h^2 x \Delta_B H_m^2}{8\alpha^2} \tag{13}$$

where $\alpha$ is the relaxation coefficient and $\Delta_B$ is the width of the Bloch domain wall. The effective temperature is of the same order as the interaction energy of domains in a lattice if $H_m/4\pi M = 2\alpha(2hcx^3)^{1/2}(z^3\Delta)^{-1/2}$. For $\alpha = 10^{-1}$–$10^{-2}$ and $\Delta_B/h = 10^{-2}$ we obtain an estimate of the field amplitude $H_m/4\pi M \simeq (1\text{–}5) \times 10^{-2}$ which corresponds to the experimental values of field amplitude necessary to order DSs [9]. The dependence of the ordering time on the amplitude of the oscillating field, domain sizes and the film parameters follows from (12) and (13) ($\tau$ is determined up to a constant $B$):

$$\tau = \frac{BN}{8\tilde{T}}. \tag{14}$$

Using the effective temperature (13) it is possible to find the 'melting' temperature of a hexagonal bubble lattice. Assuming that the lattice turns into a liquid crystal phase at $\xi(\tilde{T}_{melt}) = 0.326$, we obtain from (3) and (13)

$$\tilde{T}_{melt} = 0.024(4\pi M)^2 d^4 h^2 \rho^{3/2} \tag{15}$$

where $\rho$ is the number of bubbles per unit area of the surface.

Finally, consideration of the entropy terms in the free energy of DSs of thin soft magnetic films leads to the conclusions that DSs must exist in a disordered state in one of two cases: Firstly, if the film temperature is close to the point where the saturation magnetization becomes zero and, secondly, if the film thickness does not exceed $10^2$–$10^3$ monolayers. DS disorder observed in defectless films is frozen in all other cases. The ordering of DSs by an oscillating external magnetic field is a relaxation from a DS with a large number of frozen defects to a DS with the equilibrium defect concentration. During this process the oscillating magnetic field acts as an effective temperature.

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