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Kinetics of diamagnetic phase transitions

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The kinetics of first-order diamagnetic phase transitions into a Condon domain phase, which may occur in normal metals under a strong magnetic field, is considered. Expressions for profile, velocity, and width of the interface between the homogeneous and diamagnetic domain phase are obtained. Critical sizes of clusters of the diamagnetic domain phase are calculated as a function of temperature. The experimental feasibility of observation of the kinetics of first-order diamagnetic phase transitions is discussed.

In a strong magnetic field the transverse motion of electrons is quantized into cyclotron orbits. The electron energy spectrum is split, therefore, into Landau levels. Many properties of the electron gas are periodic functions of magnetic fields as successive Landau levels sweep through the Fermi level due to an increase of the external magnetic field.[1] Thus the oscillations of magnetization occur (de Haas–van Alphen effect). It has been shown[2] that the field acting on charges in a metal coincides with the magnetic induction B. This means that each charge is acted upon by a magnetic moment produced by all the remaining charges. As a result, a self-consistent nonlocal interaction is produced between the charges, and leads[3] to a stratification into diamagnetic or Condon domains,[4] when the magnetic, susceptibility χ ($\chi = dM/dB$, where M is the magnetization) is larger than $1/4\pi$. Macroscopically the formation of Condon domains is understandable since the magnetic induction B can be a multivalued function of the field H when strong enough oscillations in $M(B)$ occur.[2]

In the nuclear magnetic resonance (NMR) experiment on the direct evidence for Condon domains[5] it has been noticed that the existence of the NMR splitting due to domains depended on whether the sample has been heated or cooled indicating the thermal hysteresis in the phase-transition temperature. Therefore, diamagnetic phase transitions may be of *first-order* ones. The creation of the diamagnetic domain phase may be thus connected with nucleation processes and appearance of metastable states. In the case of first-order phase transitions the homogeneous and diamagnetic domain phases must coexist in the temperature interval between limits of stability of two phases. Therefore, by studying a diamagnetic phase transition in a constant magnetic field, as a function of temperature, one gets new insight in the appearance and development of the diamagnetic domain structure. We present here a *theory of kinetics of first-order diamagnetic phase transitions* and discuss possibilities of its measurement.

When the diamagnetic domain structure appears as a result of a *first-order phase transition,* the Landau expression for the thermodynamic potential Φ for a spatially inhomogeneous case has a following form:[6,7]

$$\Phi = \Phi_0 + \int \phi \, dv \ , \tag{1}$$

where ϕ is the density of the thermodynamic potential presenting an expansion in degrees of magnetization:

$$\phi = \tfrac{1}{2} a M^2 - \tfrac{1}{4} b M^4 + \tfrac{1}{6} c M^6 + D(\nabla M)^2 \ , \tag{2}$$

where

$$M = 4\pi M = B - H \tag{3}$$

and D is the positive coefficient of the nonuniform term. The analogous expansion has been used for diamagnetic periodic structures in[8,9] where the amplitude of spatial oscillations of the magnetization was an order parameter. The expression (2), without the term of a sixth order (i.e., leading to a second-order phase transition) has been applied for the two-dimensional electron gas in Ref. 10.

One supposes here that the external magnetic field is fixed and the temperature is varied. If coefficients b and c are positive, a first-order phase transition occurs for the temperature-dependent $a(T)$ which is given by

$$a(T) = a'(T - T_0) \ , \tag{4}$$

where a' is a positive constant and T_0 is *the stability limit of the homogeneous phase.* On the other hand, as is shown in Ref. 8 [see also (Ref. 10)],

$$a = 1 - 4\pi\chi \ . \tag{5}$$

The homogeneous and diamagnetic domain phases coexist on the two sides of the phase transition temperature in the range restricted by points of absolute instability of phases:

$$T_0 < T < T^* \ , \tag{6}$$

where T^* is *the stability limit of the diamagnetic domain phase.* Temperatures T_0 and T^* are given by

$$T_0 = T_c - \frac{3b^2}{16a'c} \tag{7}$$

and

$$T^* = T_0 + \frac{b^2}{4a'c} \ , \tag{8}$$

where T_c is the phase-transition temperature in which the densities of thermodynamic potentials of the two phases are equal. The range (6) gives the thermal hysteresis on supercooling and superheating, where T_0 and T^* are supercooling and superheating limits of the thermal hysteresis range (8).

For temperatures $T_0 < T < T^*$ the thermodynamic potential density exhibits three minima: two, corresponding to two Condon domains ($M \neq 0$), and the third one corresponding to the homogeneous phase $M=0$. Since the thermodynamic potential is an even function of magnetization, we have 180° domains. For temperatures $T_0 < T < T_c$ the homogeneous phase is metastable, whereas the diamagnetic domain phase is stable. At the phase transition temperature T_c the thermodynamic potential densities of the two phases are equal. In the range $T_c < T < T^*$ the homogeneous phase is stable, whereas the diamagnetic domain phase is metastable. At $T < T_0$ and $T > T^*$ one of the phases exists only. Temperature changes in the range $T_0 < T < T^*$ give rise to a motion of the interface between the two phases.

The evolution of the order parameter M is determined by the thermodynamic force, i.e., the derivative of the thermodynamic potential density ϕ with respect to the order parameter M. The larger is this thermodynamic force, the faster is the regression $\partial M/\partial t$ of a given fluctuation in M towards equilibrium. In order to study the evolution of M we use the time-dependent Ginzburg-Landau equation:[11,12]

$$\frac{\partial M}{\partial t} = -\Gamma \frac{\delta \phi}{\delta M} , \tag{9}$$

where Γ is the Landau-Khalatnikov kinetic coefficient which is assumed to depend noncritically on temperature,[12] $\delta\phi/\delta M$ is the variational derivative of ϕ with respect to M (Ref. 13) that drives the system towards equilibrium. The kinetic equation (9) reflects the fact that the order parameter M is not a conserved quantity.[14,15]

In the one-dimensional case[4,10] after substitution of Eq. (2) into (9) we obtain:

$$\frac{\partial M}{\partial t} + \Gamma(aM - bM^3 + cM^5) - 2\Gamma D \frac{\partial^2 M}{\partial x^2} = 0 . \tag{10}$$

The original partial differential equation in the independent variables of coordinate x and time t, Eq. (10), can be reduced to an ordinary differential equation in the variable s by rewriting $s = x - vt$, where v is a velocity in the direction x. After this transformation in (10) we obtain

$$2\Gamma D \frac{d^2 M}{ds^2} + v \frac{dM}{ds} - \Gamma(aM - bM^3 + cM^5) = 0 . \tag{11}$$

We seek for a stationary solution in the moving coordinate system, of a wave-front type, in order to describe the interface (the front) between two phases. We assume therefore the following boundary conditions: $dM/ds \to 0$ where $s \to \pm\infty$ and $M \to M_d$ (the index d is referred to the diamagnetic domain phase) for $s \to +\infty$, $M \to 0$ for $s \to -\infty$, where M_d and O are values of the order parameter in the diamagnetic and homogeneous phases correspondingly. M_d is given by

$$M_d = \frac{b}{2c}[1 + (1 - 4ac/b^2)^{1/2}] , \tag{12}$$

The required solution of Eq. (11) is known:[16,17]

$$M = \frac{M_d}{\sqrt{1 + \exp(-s/\Delta)}} , \tag{13}$$

where

$$\Delta = \pm \left[\frac{3D}{2|bM_d^2 - a|} \right]^{1/2} . \tag{14}$$

The expression (13) presents a kink (soliton) solution of the time-dependent. Ginzburg-Landau equation. In Ref. 17 it has been proved that this solution is stable under small perturbations and it has therefore a physical meaning. The kink solution (13) is shown in Fig. 1. It describes the interface between the diamagnetic domain phase $M \neq 0$ and the homogeneous phase ($M=0$) with the interface width Δ. The negative sign in the exponent of Eq. (13) corresponds to a situation in which the homogeneous phase is located to the left, the positive one puts the homogeneous phase to the right. The antikink solution of Eq. (11)

$$M = \frac{M_d}{\sqrt{1 + \exp(s/\Delta)}} \tag{15}$$

exists, if we suppose that $M \to M_d$ for $s \to -\infty$ and $M \to 0$ for $s \to +\infty$. This change leads to a mirror transformation of Fig. 1. Equations (13) and (15) presents particular solutions provided v is given by

$$v = \Gamma \frac{4a - bM_d^2}{(|bM_d^2 - a|)^{1/2}} \sqrt{2D/3} , \tag{16}$$

where v is the velocity of the interface between the diamagnetic domain and homogeneous phases. The sign of the interface velocity in (16) depends on the direction in which the interface propagates thus leading to formation

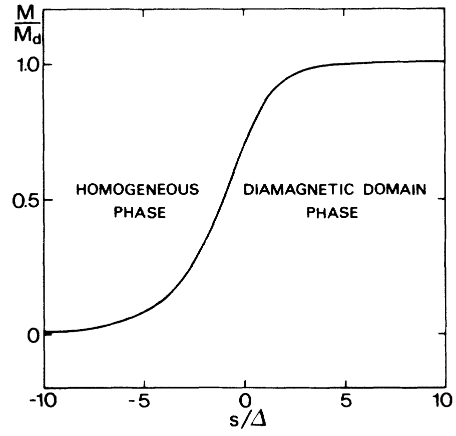

FIG. 1. The interface separating the homogeneous and diamagnetic domain phases: M/M_d as a function of s/Δ.

of the homogeneous or diamagnetic domain phase in the volume of a sample under the influence of temperature (cooling or heating).

Substituting (7) into (14) and (16) we obtain

$$\Delta=\tfrac{3}{4}\left[\frac{D}{2a'(T_c-T_0)[1-\frac{3}{8}\delta+(1-\frac{3}{4}\delta)^{1/2}]}\right]^{1/2} \tag{17}$$

and

$$v=2\Gamma[Da'(T_c-T_0)]^{1/2}f(T)\ , \tag{18}$$

where

$$f(T)\equiv\frac{\delta-\frac{2}{3}[1+(1-\frac{3}{4}\delta)^{1/2}]}{[1-\frac{3}{8}\delta+(1-\frac{3}{4}\delta)^{1/2}]^{1/2}}$$

and

$$\delta=\frac{T-T_0}{T_c-T_0} \tag{19}$$

is a dimensionless temperature.

The interface exists between $\delta=0(T=T_0)$ and $\delta=\frac{4}{3}(T=T^*)$; $\delta=1$ is the phase transition temperature $(T=T_c)$. In Figs. 2 and 3 the interface width Δ and velocity v are presented as functions of the dimensionless temperature δ according to equations (17) and (18). It is seen from Fig. 2 that the interface width is finite for all the temperatures in the range $T_0<T<T^*$. In other words the sharp interface between the two phases is well determined in the above-mentioned temperature range, including the phase transition temperature. On approaching the stability limit of the diamagnetic domain phase T^* the width of the interface increases. We see from Fig. 3 that at the phase-transition temperature $T_c(\delta=1)$ the interface does not move $(v=0)$. It is caused by the equality of thermodynamic potential densities of coexisting phases. The velocity of the interface is actually one of the growth of the diamagnetic domain phase.

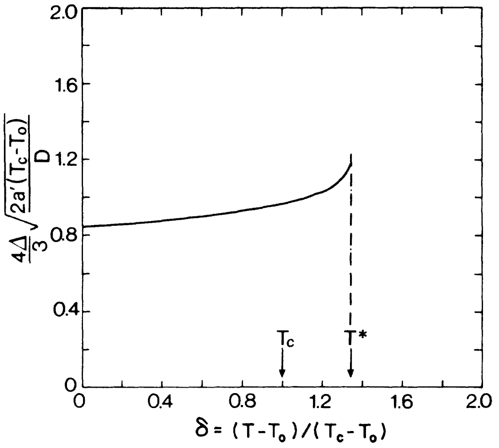

FIG. 2. The temperature dependence of the interface width $\Delta(4\Delta/3)[2a'(T_c-T_0)/D]^{1/2}$ as a function of the dimensionless temperature $\delta=(T-T_c)/(T_0-T_c)$. Here T_0 is the stability limit of the homogeneous phase, and T_c is the phase transition temperature.

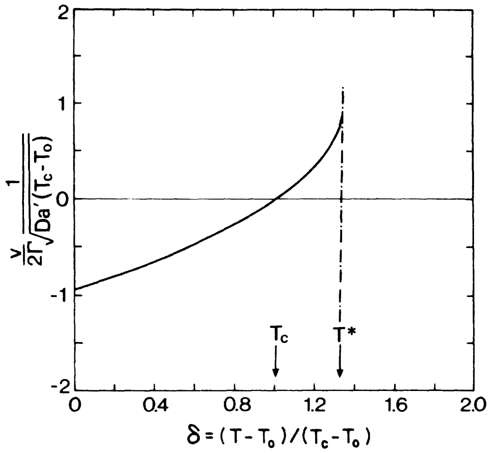

FIG. 3. The temperature dependence of the interface velocity v $((v/2\Gamma)1/[Da'(T_c-T_0)]^{1/2}$ as a function of δ).

At first-order phase transitions a supercooled homogeneous phase in the temperature range, given by Eq. (6), is stable against infinitesimal fluctuations and unstable against finite heterophase fluctuations[18] which are critical nuclei of the diamagnetic domain phase. Let us calculate the critical size of the nucleus (cluster) R_c of the diamagnetic domain phase. According to the classical theory of nucleation,[18] the new phase cluster whose radius corresponds to the maximum of the thermodynamic potential change, required for formation of this phase, is the critical nucleus. Consequently, to calculate the critical size of the diamagnetic domain phase cluster one maximizes the thermodynamic potential by varying R.[18] For simplicity we assume, according to,[15,19] that the order parameter as a function of the coordinate $M(r)$ is approximately given by

$$M=M_d,\quad |r|<R\ , \tag{20a}$$

$$M=M_d\left[1-\left[\frac{r-R}{\Delta}\right]\right],\quad R\le r\le R+\Delta\ , \tag{20b}$$

$$M=0,\quad |r|\ge R+\Delta\ , \tag{20c}$$

where R is the radius of the cluster. Integrating the thermodynamic potential density $\phi(R)$ over the volume V, in Eq. (1), we obtain:[19]

$$\Phi=\phi(M_d)V+\frac{4\pi}{3}R^3[\phi(M_h)-\phi(M_d)]$$
$$+4\pi R^3 D\frac{M_d^2}{\Delta^2}\left[\frac{\Delta}{R}+\frac{\Delta^2}{R^2}+\frac{\Delta^3}{3R^3}\right]\ , \tag{21}$$

where the index h denotes the homogeneous phase. For large clusters $(\Delta/R\ll1)$, when this macroscopic consideration is applicable, we have, after maximization of $\phi(R)$,

$$R_c = 24\frac{D}{\Delta|4a - bM_d^2|} . \tag{22}$$

Substituting Eqs. (4) and (7) into Eq. (22) we obtain

$$R_c = \frac{8}{|f(T)|}\left[\frac{2D}{a'(T_c - T_0)}\right]^{1/2} . \tag{23}$$

In Fig. 4 the temperature dependence of R_c is demonstrated. It is seen that at $T = T_c$ ($\delta = 1$), $R_c \rightarrow \infty$. It is caused by the equality of thermodynamic potentials at the phase transition in which the minimum work required for formation of nuclei of the diamagnetic domain phase is equal to zero.

Since the formation of the diamagnetic domain phase is connected with increase of thermodynamic potential it is possible only through thermal fluctuations. The probability of such fluctuations is

$$W \propto e^{-\Delta\Phi/k_B T} , \tag{24}$$

where $\Delta\Phi$, the work needed for formation of a critical cluster of the diamagnetic domain phase, is given by

$$\Delta\Phi = 3840\pi\frac{D^3 M_d^2}{\Delta^3|4a - bM_d^2|^2} . \tag{25}$$

This result is in accord with the nucleation theory in fluids[18] as $\Delta\Phi \propto D^3$.

Let us discuss, now, the conditions for possible experiments. The diamagnetic domain phase has been firstly observed in NMR experiments.[15] Therefore we shall naturally discuss the possibility of measurements of the kinetics of first-order diamagnetic phase transition by this method.[20] Since the phase transition must be a first-order one, three NMR lines accompanied by a thermal hysteresis should be observed in the temperature range $T_0 < T < T^*$: one line is originated from the homogeneous phase and two lines are of the diamagnetic domain phase resulting from each domain.

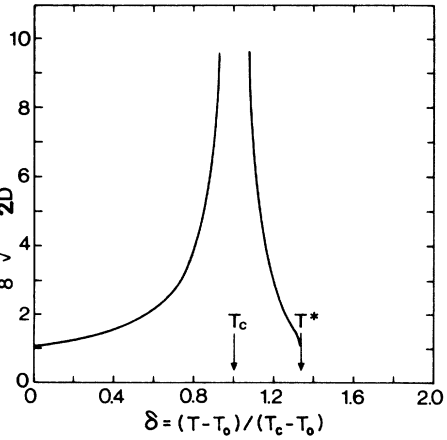

FIG. 4. The temperature dependence of the critical size of the cluster of the diamagnetic domain phase R_c $((R_c/8)[a'(T_c - T_0)/2D]^{1/2}$ as a function of δ).

The observation of the three states on a time scale of NMR is possible, if

$$\omega_N \tau_{\rm cl} \gg 1 , \tag{26}$$

where ω_N is the NMR frequency (for example, in the experiment[5] for Ag $\omega_N \simeq 18$ MHz and $\tau_{\rm cl}$ is the time during which the cluster of the diamagnetic domain phase grows as a result of cooling. The time τ_c is determined by

$$\tau_{\rm cl} = \frac{\Delta R_c}{v} , \tag{27}$$

where ΔR_c is an increase of the cluster on a cooling (see Fig. 4) and v is the interface velocity in the given temperature range (it is seen from Fig. 3 that the temperature dependence of v is relatively weak). We can present Eq. (18) in the form

$$v = \Gamma R_L[(a'/2)(T_c - T_0)]^{1/2} f(T) , \tag{28}$$

where R_L is the cyclotron radius given by

$$R_L = \frac{v_f}{\omega_c} . \tag{29}$$

Here v_f is the Fermi velocity and $\omega_c = eH/mc$ is the cyclotron frequency. We use here $D \simeq \chi R_L^2$.[4,10]

Using Eqs. (26), (27), (28), and (29) we obtain

$$\omega_N \tau_{\rm cl} = \frac{\omega_N}{\omega_c}\frac{v_f}{v}\frac{\Delta R_c}{R_L} . \tag{30}$$

To check the criterion (26), we take characteristic values of applied parameters. For estimates we use

$$R_L = 10^{-4}\text{–}10^{-5}\ \text{cm} .$$

The coefficient a' may be estimated from the susceptibility χ [see (4) and (5)] as $\chi \propto 1/4\pi$ near T_c. The difference $T_c - T_0$ is taken as 0.5 K. Taking the definite $T_c - T_0$, we do not need to use the coefficients b and c. However, they may be calculated from measurements of the thermal hysteresis $T^* - T_0$ according to Eq. (8) and the latent heat of the phase transition

$$\Delta Q = \tfrac{3}{8}\frac{a'b}{c}T_c . \tag{31}$$

Near T^* we obtain that $\Delta R_c/R_L \simeq 10\text{–}10^2$ [Fig. 4 and Eq. (23)]. It is reasonable to apply $\omega_N/\omega_c \simeq 10^{-5}$. The measured values of the interface velocities for ferroelectric materials are

$$v = (0.6\text{–}70)\times 10^{-3}\ \text{cm/sec}$$

(Refs. 21–23). Consequently $v_f/v \simeq 10^{10}$. Therefore it is clear that $\omega_N \tau_{\rm cl} \gg 1$ and the criterion, Eq. (26), is fulfilled. Thus the simultaneous observation of three phases by NMR is possible.

The temperature dependence of the NMR spectrum intensity near the first-order diamagnetic phase transition temperature may provide an interesting information on the phase separation at diamagnetic phase transition. At $T > T^*$ the single line corresponding to the homogeneous phase is presented. For $T_c < T < T^*$ three lines appear:

the line of the largest intensity corresponds to the homogeneous phase, whereas the two other lines are of Condon domains. At $T=T_c$ all the three lines have an equal intensity. In the temperature range $T_0<T<T_c$ the intensities of the two NMR lines resulting from Condon domains are larger than that of the homogeneous phase. At $T<T_0$ two NMR lines of the diamagnetic domains emerge. It is clear that the interface velocity v is equal to the rate of the temperature change of the intensity of NMR lines in the temperature range $T_0<T<T^*$.

To observe the pure effect of the interface motion it is desirable that the rate of establishing of temperature in a sample is sufficiently large in comparison with the interface velocity: $v_T \gg v$. Let us check whether this condition is fulfilled for silver samples used in (Ref. 5) for the direct evidence of presence of the diamagnetic domains.

A process of a temperature establishing in a sample may be approximately described by the Fourier equation[24] (for simplicity we consider a one-dimensional case):

$$\frac{1}{\alpha}\frac{\partial T}{\partial t}=\frac{\partial^2 T}{\partial x^2}\,, \tag{32}$$

where α is the thermal diffusivity given by

$$\alpha=\frac{K}{C\rho}\,, \tag{33}$$

where K is the conductivity coefficient, C is the specific heat per unit volume and ρ is the density. Since conductivity expresses the rate of heat flow into a sample and specific heat denotes its ability to store this received energy, it follows that the ratio (33) is a measure of the rate of change of temperature of the material.[25] Therefore we can approximately estimate the rate of propagation of temperature changes in a sample v_T as

$$v_T=\frac{\alpha}{d}\,, \tag{34}$$

where d is the thickness of the sample. We use $\alpha=1.71$ cm^2/sec (Ref. 21) for silver. The specimen used in (Ref. 5) was a plate—8 mm$^2\times 0.8$ mm thick along a [100] direction. Calculations of v_T according to Eq. (34) for two directions in the silver sample give $v_T=6$ cm/sec (along the width of the sample) and $v_T=21$ cm/sec (along the thickness of the sample). Thus the criterion $v_T \gg v$ is fulfilled providing the possibility of the measurement of the interface velocity. The related condition required to observe the growth of clusters of the diamagnetic domain phase $\tau_{\rm cl} \gg \tau_T$, where τ_T is the time of the temperature establishing, is also fulfilled. Indeed, if we roughly estimate τ_T as $\tau_T \simeq d/v_T$, then using the latter expression and Eq. (27) we obtain

$$\tau_{\rm cl}/\tau_T=(\Delta R_c/d)v_T/v \gg 1$$

for the values used of these parameters.

To summarize, the interface between the homogeneous and diamagnetic domain phases is obtained as a particular kink (soliton) solution of the time-dependent Ginzburg-Landau equation. Expressions for the profile, width, and velocity of the interface are derived. Temperature dependences of the interface width and velocity and that for the critical size of the cluster of the diamagnetic domain phase are presented. It is shown that the kinetics of formation of the diamagnetic domain phase can be investigated in NMR experiments.

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