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LETTER TO THE EDITOR

The demagnetization heat capacity of uniaxial ferromagnets near the critical point

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Abstract. The effect of a demagnetizing field on the heat capacity of a ferromagnet is discussed. Mean field calculations of the contributions of demagnetization energies to the heat capacity of a uniaxial ferromagnet are presented and illustrated by application to heavy rare earth metals.

In the renormalization group theory of magnetic systems with dipolar interactions the contribution to the free energy of the system from the demagnetization effects can be neglected by considering a needle-shaped sample having only one magnetic domain and uniform bulk magnetization (Aharony and Bruce 1974). If an external magnetic field is present it has to be parallel to the long axis of the sample. However, for samples of other shapes the free energy and hence the heat capacity will have a contribution from the energies associated with demagnetization effects. Knowledge of the magnitude of such a correction term is particularly important close to T_c because of the possible effect of this correction on the critical behaviour of the heat capacity.

The heat capacity of a ferromagnetic sample in an external magnetic field depends on its shape (Levy and Landau 1968). This shape dependence of the heat capacity is caused by the shape-dependent demagnetizing field. Griffiths (1969) argued that the heat capacity of a ferromagnetic sample in an external magnetic field has a singularity (a non-analytical behaviour, not a discontinuity) at a temperature close to and below T_c where the spontaneous magnetization of the sample equals the external magnetic field divided by the demagnetization factor.

In this letter we report calculations of the demagnetization free energy and the demagnetization heat capacity for an uniaxial ferromagnet in a zero or vanishingly small external magnetic field in the mean field approximation. These calculations were motivated by our investigation of the critical heat capacity of Gd near its Curie point (Bednarz 1992, Bednarz *et al* 1992). Therefore our results will be illustrated primarily by using the example of Gd for a typical sample geometry used in our experimental studies.

The magnetic energy, F_{dom}, in the low temperature regime, for a ferromagnetic plate magnetized along the normal was calculated by Kittel (1949) for stripe domains with narrow walls, in a thick slab (figure 1)

$$F_{dom} = VC_1 M_s^2 a_w / L_z \tag{1}$$

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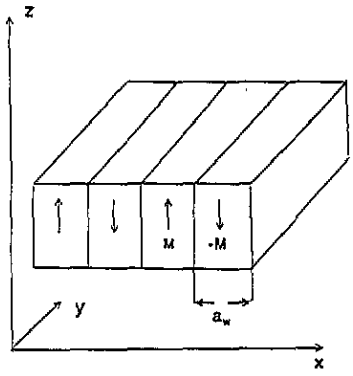

Figure 1. Model for calculation of the magnetic field energy of coplanar strip domains of alternate sign.

where V is the slab volume, M_s is the spontaneous magnetization, a_w is the domain width, L_z is the slab thickness and C_1 is a numerical factor equal approximately to 0.85 in this case and around 0.5 in some other cases (Barker and Gehring 1983).

It is possible to devise a domain arrangement for the rectangular slab from figure 1 which will have no demagnetizing field (Kittel 1949, Vonsovskii 1974). The magnetic flux circuit is completed within the crystal by means of the domains of closure and no magnetic 'poles' are formed on the crystal surface. The magnetic energy associated with the demagnetization field is zero, but the anisotropy energy is not zero.

In a uniaxial crystal the magnetization within the domains of closure is oriented in a direction of hard magnetization; this involves the anisotropy energy (Kittel 1949, Vonsovskii 1974)

$$F_{\text{anis}} = V a_w \sum_i K_i / 2L_z \tag{2}$$

where V is the sample volume, K_i are the anisotropy constants, a_w is the domain width and L_z is the sample thickness. The ratio of the energy of a domain structure with domains of closure to the energy of a slab-like structure is determined primarily by the ratio of the sum of the anisotropy constants to the square of the spontaneous magnetization, M_s^2. If the ratio $4\pi(\sum K_i)/\mu_0 M_s^2 \ll 1$ where μ_0 is the permeability of free space, then the flux closure configuration gives the lower energy but with the increasing value of the ratio the domains of closure will gradually open. The uniaxial ferromagnets usually possess a slab-like domain structure (Kittel 1949, Kooy and Enz 1960).

In order to estimate the total energy associated with the domain structure one also has to estimate the domain wall contribution to that energy. Usually, the walls are described by rotation of the magnetization vector, M, at constant magnitude, M, within the wall. This magnetization profile is known as a rotational solution (Kittel 1949, Bulaevskii and Ginzburg 1964). However, another possible wall structure (linear solution) is given by a varying magnitude of M with the magnetization direction always either parallel or antiparallel to the easy axis of the magnetization (Bulaevskii and Ginzburg 1964, Bar'yakhtar and Klepikov 1972, Lawrie and Lowe 1980, Winternitz *et al* 1988). The latter magnetization profile is expected to occur near T_c (Bulaevskii and Ginzburg 1964, Lawrie and Lowe 1980).

Bulaevskii and Ginzburg (1964) analysed a single wall structure near T_c using a mean field approximation to the free energy expansion in powers of magnetization under the assumption that the domain width is much larger than the wall thickness. They showed that close to T_c the linear solution for the magnetization profile was given by

$M_s(x) = M_s \tanh(x/\lambda)$ where x is the distance perpendicular to the wall and λ is a measure of the wall thickness and is of the order of the correlation length ξ.

The solution of Bulaevskii and Ginzburg (1964) for one domain wall can be used to estimate the domain wall energy of N walls close to T_c in the mean field approximation provided the walls do not interact ($a_w \gg \xi$). The free energy of the domain walls of the slab-like structure (figure 1) can be obtained by integrating the free energy functional derived from the exchange Hamiltonian of the LGW form

$$\frac{H_{ex}}{2J_L(0)} = \int d^3x \left(\frac{1}{2} \frac{T - T_{c0}}{T_{c0}} (\sigma(x))^2 + \frac{c}{2} (\nabla \sigma(x))^2 + u(\sigma(x))^4 \right) \tag{3}$$

where c and u are constants defined later, $\sigma(x)$ is the spin density with dimensions of $1/(\text{volume})^{1/2}$ and the integration is over the sample volume (Wegner 1976, Winternitz *et al* 1988). The zeroth-order term, $J_L(0)$, of a series expansion of the exchange energy, $J_L(k)$, in powers of wavenumber k is related to the mean field critical temperature, T_{c0} by $k_B T_{c0} = \frac{2}{3}(g_J - 1)^2 J(J+1) J_L(0)$, where J is the total angular momentum quantum number and g_J is the Landé factor (Stanley 1971, Coqblin 1977).

The spin density can be related to the magnetization density by

$$\sigma(x) = M(x) \Omega_0^{1/2} / \gamma \mu_B \tag{4}$$

where μ_B is the Bohr magneton, $\gamma = g_J(g_J - 1)$ and $\Omega_0 = V_{mol}/N_A$, where V_{mol} is the molar volume and N_A is Avogadro's number. The total free energy of a single wall, F_{tot}, is

$$F_{tot} = \tau \int d^3x \left(-\frac{1}{2} \frac{\kappa^2}{\kappa_0^2} (M(x))^2 + \frac{a}{2\kappa_0^2} (\nabla M(x))^2 + u'(M(x))^4 \right) \tag{5}$$

where the new symbols are defined as follows: $\kappa = \xi^{-1} = \xi_0^{-1}(-t)^\nu = \kappa_0(-t)^\nu$ where ν is the correlation length critical exponent equal to $\frac{1}{2}$ in the mean field theory, and $c = a/\kappa_0^2$ (a is a dimensionless constant), and $u' = u\Omega_0/(\gamma\mu_B)^2$. The constant τ is defined as

$$\tau = 2J_L(0) / \left[(\gamma \mu_B)^2 / \Omega_0 \right]. \tag{6}$$

Taking the domain wall to lie in the y–z plane and the origin of the coordinate system in the centre of the wall, the Bulaevskii and Ginzburg (1964) solution $M_s(x) = M_s \tanh(x/\lambda)$ minimizes the functional given by (5) with $\lambda = (2a)^{1/2}\xi$ under the constraint that $M_s^2 = [1/(4u')](\kappa^2/\kappa_0^2)$. This condition implies uniform magnetization in the bulk of the domain far from the wall.

For one wall, F_{tot} can be expressed as a sum of several terms resulting from the integral given by (5),

$$F_{tot} = -\tfrac{1}{4}\tau(\kappa^2/\kappa_0^2) M_s^2 V_1 + F_{inh} \tag{7}$$

where V_1 is the volume of the domain including one wall, the first term on the right hand side is the homogeneous background magnetization energy and F_{inh} contains the quadratic and quartic contributions to the free energy from the non-uniform magnetization. The various integrals can be evaluated under the approximation $a_w \gg \xi$ (integration limits can be extended to $\pm\infty$ where $\tanh(x) \to \pm 1$). The inhomogeneous contribution to the free energy from $N = L_x/a_w$ independent walls, F_{wall}, is given by N times F_{inh}:

$$F_{wall} = (2\sqrt{2a}/3) V \tau M_s^2 / \xi \kappa_0^2 a_w \tag{8}$$

where V is the sample volume. The total free energy associated with the demagnetization effects is obtained by adding to F_{wall} either the magnetic energy of the N volume domains or the anisotropy energy of the N domains of closure, depending on the domain structure, i.e. on the ratio $4\pi K_2(T)/\mu_0 M_s^2$ close to T_c. The total free energy associated with the demagnetization effects, F_{dem}, for the slab-like domain structure is obtained by combining (1) and (8):

$$F_{dem} = V M_s^2 \left[2\sqrt{2a}\tau/(3\xi\kappa_0^2 a_w) + C_1 a_w/L_z \right]. \tag{9}$$

F_{dem} is a minimum with respect to the domain width, a_w, when

$$a_w = \left[2\sqrt{2a}\tau L_z/\left(3\xi_0\kappa_0^2 C_1\right) \right]^{1/2} (-t)^{\nu/2} = a_{w0}(-t)^{\nu/2} \tag{10}$$

where the reduced temperature dependence of a_w is written explicitly and is given by the exponent $\nu/2$. The minimized demagnetization free energy (in SI units) is then given by

$$F_{dem} = \left(\mu_0/2\pi\right) V M_{s0}^2 \left[2\sqrt{2a} C_1 \tau/\left(3\bar{L}_z \xi_0 \kappa_0^2\right) \right]^{1/2} (-t)^{2\beta+\nu/2} = F_{dem}^0 (-t)^{\psi}. \tag{11}$$

The reduced temperature dependence of the minimized demagnetization energy, F_{dem}, is determined by the exponent $\psi = 2\beta + \nu/2$. There is agreement between the exponents obtained here for the reduced temperature dependence of the domain width and for the demagnetization free energy with the mean field limit of the corresponding exponents derived by Stauffer (1972) who analysed a scaling form of the free energy of the domain wall. Stauffer obtained, for the domain width, the exponent $\nu(1-\eta)/2$, which becomes $\nu/2$ putting $\eta = 0$ as the mean field result; the calculated exponent for the free energy also reduces to the mean field exponent obtained here for $\eta = 0$. The contribution from the demagnetization energy to the total sample heat capacity can be estimated by

$$\Delta C_{dem} = -T\left(\partial^2 F_{dem}/\partial T^2\right) \simeq \left(-1/T_c\right) F_{dem}^0 \psi(\psi - 1)(-t)^{\psi-2}. \tag{12}$$

The above results will now be used to estimate the contribution from the demagnetization processes to the critical heat capacity of Gd. The various parameters required will be taken directly from numerical fits to experimental data.

For Gd the saturation magnetization at $T = 0$ K is $M_{s0} = g_J \mu_B N_{Gd}[J(J+1)]^{1/2} \simeq 223 \times 10^4$ A m^{-1}, where N_{Gd} is the number of Gd atoms per m^3. The temperature dependence of the spontaneous magnetization is given for small t by the power law $M_s(t) = M_{s0}(-t)^{\beta}$ where $\beta \simeq 0.39$ for Gd (Aliev *et al* 1988). The anisotropy constants K_2 and K_4 were measured by Graham (1963) as a function of temperature. The anisotropy constant K_2 reaches a maximum at around 285 K ($K_2 \simeq 2 \times 10^4$ J m^{-3}) and decreases to zero at around 350 K. In the region of the Curie temperature, $K_2 \simeq 1.5 \times 10^4$ J m^{-3}. The anisotropy constants K_4 and K_6 vanish above approximately 240 K (Graham 1963, Rhyne 1972). Close to T_c, $M_s^2(T)$ is much less than $K_2(T)$ and the domain structure is approximated by that in figure 1.

One also obtains $\tau \simeq 12 \times 10^2$ for Gd, taking $T_c \simeq 300$ K and estimating $J_L(\mathbf{0})$ using the relation given in (3) with $T_{c0} \simeq 1.5\, T_c$ to incorporate approximately the suppression of the Curie temperature by fluctuations (Fisher 1967, Stanley 1971).

The coefficient a was obtained using the data of Mackintosh and Møller (1972) for the exchange energy function $J_L(k)$ for Gd as a function of the reduced wave vector k/k_{max} in the c-axis direction:

$$\frac{J_L(k) - J_L(0)}{J_L(0)} \simeq \frac{m}{J_L(0)}\left(\frac{k^c}{k^c_{max}}\right) = -\frac{a(k^c_{max})^2}{\kappa_0^2}\left(\frac{k^c}{k^c_{max}}\right)^2 \tag{13}$$

where the coefficient $m \simeq -12$ meV can be estimated from a fit to the data in figure 5.8 of Mackintosh and Møller (1972). Taking $k^c_{max} \simeq 2\pi/c$ where $c \simeq 5.8$ Å and with $\kappa_0 = \xi_0^{-1} \simeq 0.5$ Å^{-1} for an estimate of the inverse of the correlation length at $T = 0$ K, one obtains $a \simeq 0.59$.

At this point a number of important estimates can be made. One of them concerns the range of the validity of the model. It was assumed in the above calculations that $a_w \gg \lambda \simeq \xi$. This is an important assumption because the sum of free energies of N non-interacting walls can give a reasonable approximation to the actual free energy of the domain walls only if the separation between the walls is sufficiently large. Otherwise, by not taking into account the interactions between the walls, the model developed here does not correctly address the entropy of the whole system. From (10), $a_w \simeq 6.7 \times 10^4(-t)^{0.35}$ Å for $L_z \simeq 0.22$ mm (this is the thickness of one of Gd samples investigated in our experimental study, Bednarz 1992) and for $\nu = 0.7$ (Guillou and Zinn-Justin 1985). The domain width is of the order of the correlation length ($6.7 \times 10^4(-t)^{0.35} \simeq 2(-t)^{-0.7}$) when $t \simeq 5 \times 10^{-5}$ so the model breaks down at $T \simeq T_c - 0.015$ K.

From (11), the free energy is: $F_{dem} \simeq 0.5(-t)^{1.13}$ J mol^{-1} putting for V the molar volume of Gd. From (12), at the reduced temperature $t = -5 \times 10^{-5}$ ($T_c - T \simeq 0.015$ K), the demagnetization contribution to the heat capacity is around -1.4 J mol^{-1} K^{-1} which is approximately 2% of the total heat capacity. The demagnetization contribution becomes negligibly small further away from T_c (it is around 0.3% at $t = -5 \times 10^{-4}$; $T \simeq T_c - 0.15$ K).

The magnetic structure of the rare earth metals terbium (Tb) and dysprosium (Dy) in their ferromagnetic state is more complex than that of Gd. However an estimate of the magnitude of the demagnetizing contribution to the heat capacity of Tb and Dy near their Curie points can be obtained under the simplifying assumption of strip domain structure. For this purpose, we used the data of Mackintosh and Møller (1972) on their exchange function $J_L(k)$ for Tb and Dy in order to estimate the coefficient a in (11) and also literature data on the heat capacity for these metals (Jayasuriya *et al* 1984, 1985). The sample geometry and dimensions were taken to be the same as those for Gd.

The relative demagnetization contribution to the heat capacity of Tb is practically the same as that for Gd, i.e. about 2.7% at $t = -5 \times 10^{-5}$ and 0.4% at $t = -5 \times 10^{-4}$. This contribution is larger for Dy: about 8% at $t = -5 \times 10^{-5}$ and 1.3% at $t = -5 \times 10^{-4}$. It should be emphasized that these estimates for Tb and Dy are intended only to indicate the magnitude of the contributions.

The above results suggest that the contribution from demagnetization effects to the heat capacity of Gd, Tb and Dy are negligible over most of the temperature range which is typically experimentally accessed even by a high resolution heat capacity measurement for high quality samples. The analysis also indicates that for $(-t) < 10^{-4}$ this contribution may need to be taken into account as one of the factors determining the shape of the heat capacity curve very close to T_c.

We emphasize that the primary objective of this work has been to give a reasonable estimate of the magnitude of the demagnetization heat capacity contribution to the total heat capacity over a range of reduced temperature below T_c. The model presented here is

not intended to give a complete description of the temperature dependence of the domain width, and hence the demagnetization free energy, in the *immediate proximity* of T_c. The self-consistent range of validity of this model requires $(T - T_c)/T_c < -5 \times 10^{-5}$ in the case of Gd, for example. At smaller values of $|t|$, the walls can no longer be considered independent. It has been suggested that a periodic magnetization profile replaces the linear domain structure (Garel and Doniach 1982, Barker and Gehring 1983). The period of the profile is finite at T_c.

In conclusion, the results presented in this letter show that demagnetization processes in thin plates of uniaxial ferromagnets may have a sizable effect on the heat capacity data in the critical region and should then be taken into account in the data analysis. The reduced temperature range where the demagnetization effects may become important in an analysis of the critical behaviour of the heat capacity will depend on the ferromagnet studied and on the sample geometry.

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