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G. Heber, F. Goedsche, E. Heiner, J. Monecke, H. Wenzel. SURFACE-EXCHANGE MODES IN FERROMAGNETIC PARTICLES AND THIN FILMS. Journal de Physique Colloques, 1971, 32 (C1), pp.C1-1165-C1-1167. 10.1051/jphyscol:19711417 . jpa-00214459

HAL Id: jpa-00214459

<https://hal.science/jpa-00214459>

Submitted on 1 Jan 1971

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SURFACE-EXCHANGE MODES IN FERROMAGNETIC PARTICLES AND THIN FILMS

G. HEBER, F. GOEDSCHE, E. HEINER, J. MONECKE, H. WENZEL

TU Dresden, DDR, Sektion Physik, Sektion Mathematik

Résumé. — A l'aide de la théorie phénoménologique des ondes de spin, on calcule de telles ondes à l'intérieur d'une particule ferromagnétique entourée d'une matière paramagnétique, en se limitant à des particules sphériques et rectangulaires (y compris des couches minces). L'anisotropie de surface est prise en considération tandis que les effets magnétostatiques sont négligés. On donne les conditions pour l'existence des ondes de surface. Pour une particule rectangulaire ces ondes de surface peuvent être aussi trouvées de façon rigoureuse à partir d'un calcul quantique des spins localisés. Quelques conséquences thermodynamiques de telles ondes de surface sont énoncées.

Abstract. — Spin-wave-modes inside a ferromagnetic particle (imbedded in a paramagnetic matrix) are deduced by means of phenomenological spin-wave-theory for a spherical and a square-shaped particle (including a thin film). Surface-anisotropy is included, magnetostatic effects are excluded. The conditions for the occurrence of surface-exchange-modes are given. It is shown that these surface modes also follow from an exact quantummechanical treatment of localized spins in a square-shaped particle. Some thermodynamic consequences of such surface modes are pointed out.

I. Surface modes by phenomenological spin-wave method. — Let us consider one small ferromagnetic particle imbedded in a paramagnetic matrix. All other ferromagnetic particles should be so far away that magnetic dipole-dipole interaction may be omitted. We discuss collective magnetic excitation, with wavelengths λ , which are big (relative to lattice constant a), but not so big that magnetostatic interactions are more important than exchange interactions. That means, we assume λ to fulfill :

$$200 a > \lambda > 20 a. \quad (1)$$

Under such conditions, magnon modes may be simply derived from a phenomenological approach [1, 2], the fundamental equation being

$$(\Delta + x(\omega)) m(\mathbf{r}) = 0, \quad (2)$$

with

$$x(\omega) = \frac{1}{MB} \left(\frac{\omega}{\gamma} - H_i \right)$$

and M = saturation magnetization, B = exchange constant, ω = frequency of spin wave, γ = gyromagnetic factor, H_i = effective magnetic field inside the particle, $m = m_x + im_y$, $\mathbf{m}$ = deviation from the saturation magnetization M .

For simplicity, we consider only cases here, in which H/H_i is constant exactly or in a good approximation.

We wish to show that surface modes appear as solutions of (2), if the boundary condition at the surface of the particle has an appropriate structure. In fact this boundary condition is very essential for the surface modes. We use a boundary condition, derived by Rado and Weertman [3], to be written as

$$B \frac{\partial m}{\partial n} \pm K_s m = 0, \quad (3)$$

with B , m as in (2),

$\partial/\partial n$ = normal-derivative, K_s = surface-anisotropy constant. $+$ — sign holds, if z -axis is magnetically easy axis at the surface, $-$ — sign holds, if z -axis is magnetically hard axis at the surface.

If the geometry of particle is simple, one is able to solve (2) and (3) explicitly. The most simple case is a square-shaped particle (including a plane film). In this case the solution may be separated in cartesian coordinates; we have in each component solutions of the form

$$f(x) = C_1 e^{ik_x x} + C_2 e^{-ik_x x}, \quad (4)$$

which have to fulfill (due to (3))

$$e^{i\tilde{k}} = \pm \frac{\tilde{k} + i\tilde{\lambda}}{\tilde{k} - i\tilde{\lambda}} \quad (5)$$

with

$$\tilde{k} = lk_x,$$

l = length of particle in x -direction,

$$\tilde{\lambda} = l(\pm K_s)/B.$$

Note : For $\tilde{\lambda} = 0$ we have free surface-spins, for $\tilde{\lambda} \rightarrow \infty$ however completely pinned surface-spins. (5) determines the spectrum of the allowed $\tilde{k}$ -values. We have examined (5) in detail [4, 5]. Of course, a discrete manifold of real $\tilde{k}$ -solutions is always present, which belong to volume modes. Interesting for us are solutions with imaginary $\tilde{k}$ (surface mode). Such solutions appear only, if the negative sign in (3) holds, i. e., if z -axis is magnetically hard at the surface. In this case there are only one or two solutions, depending on the value of $\tilde{\lambda}$, see figure 1. For the positive sign in (3) no imaginary $\tilde{k}$ is a solution of (5). Complex $\tilde{k}$ -solutions do not exist in both of the cases. The strong dependence of these imaginary $\tilde{k}$ -solutions on $\tilde{\lambda}$ and the sign in (3), (5) may give the possibility to measure K_s by measuring these surface modes by ferromagnetic resonance, by low-energy-electron scattering, or by cold-neutron-small angle scattering from surfaces of ferromagnetic films.

We have also considered a spherical ferromagnetic particle with a similar result. Surely one can treat also other geometrical cases similarly.

FIG. 1. — Wavenumber $\tau = |\tilde{k}|$ for the surface modes as function of $|\tilde{\lambda}| = lK_s/B$.

II. Surface modes in a Heisenberg model with surface anisotropy. — The method [4, 5] is limited to long wavelengths λ (see (1)). From physical arguments however such surface modes should exist also for smaller λ . This was shown by Monecke [6] considering the Hamiltonian

$$\begin{aligned} \mathcal{H} = & -2J \sum_{(l,m)} \mathbf{S}_l \cdot \mathbf{S}_m - g\mu_B H_0 \sum_l S_l^z + \\ & + g\mu_B H_A \sum_{\text{sides}} |S_l^z| + 2g\mu_B H_A \sum_{\text{edges}} |S_l^z| \\ & + 3g\mu_B H_A \sum_{\text{corners}} |S_l^z| \end{aligned} \quad (6)$$

for a square-shaped particle, with

H_0 = external field, H_A = surface-anisotropy field, the three last summations are including only surface spins of the particle.

$H_0 > H_A$ gives a simple ground state (all spins in z-direction). This Hamiltonian gives as eigenvalue-equation for the x-component k_x of the wave vector $\mathbf{k}$

$$\left. \begin{aligned} \operatorname{tg} \frac{x_1}{2} \\ -\operatorname{cotg} \frac{x_1}{2} \end{aligned} \right\} = - \frac{g\mu_B H_A + 2SJ \left(1 - \cos \frac{x_1}{l_1 - 1}\right)}{2SJ \sin \frac{x_1}{l_1 - 1}} \quad (7)$$

with

$x_1 = ak_x(l_1 - 1)$, al_1 = length of particle in x-direction, a = lattice constant.

For small k_x (7) reduces to (5), i. e. surface modes appear also in the Heisenberg model with surface anisotropy.

Remark that in a paper by Wallis et al. [7] the existence of such modes was shown for a Heisenberg-model, in which the exchange at the surface is different from that in the bulk material.

We may conclude that $\lambda > 20a$ is not essential for the occurrence of surface modes. In the case $\lambda > 200a$ magnetic dipole-dipole interactions have to be included [8], [9].

III. Macroscopic consequences of exchange-surface modes. — Let us consider the case, in which one or two surface-exchange modes do exist. It follows

FIG. 2. — Magnetic part c of specific heat of a cube-shaped particle as function of temperature T for different values of $\tilde{\lambda}$: (units of c : k_B ; external field: 84 kOe; cubes with edge-length 60 Å).

FIG. 3. — Magnetic part c of specific heat of a cube-shaped particle as function of $\tilde{\lambda}$ for fixed temperature ($k_B T = 0.05$ meV; unit of c : k_B ; external field: 20 kOe; cubes with edge-length 300 Å).

from (2) that the dispersion relation for all modes has the form

$$\omega = \gamma MB(k_x^2 + k_y^2 + k_z^2) + \gamma H_i. \quad (8)$$

Therefore surface modes (with one imaginary k) lie below the volume modes (real $\mathbf{k}$) in the frequency spectrum of the particle. If H_i is high enough, the frequency of these modes is located in the gap between $\omega = 0$ and $\omega = \gamma H_i$. In this case, the surface modes should have a strong influence on thermodynamic properties of the particle (or film) for temperatures T with $k_B T < \hbar \gamma H_i$.

For H_i of order 10^4 G the interesting temperatures are of order 1 °K.

We have calculated in this region the magnetization M and the magnetic part of the specific heat c as function of temperature from our mode spectrum [10], [11]. The most interesting features show $c(T, \tilde{\lambda})$. For strong surface anisotropy and high magnetic fields typical anomalies appear in c as function of T (see Fig. 2), but we have still more anomalous behaviour, considering c as function of $\tilde{\lambda}$ for fixed T (see Fig. 3). These anomalies are connected with the appearance (or disappearance) of the surface modes by changing $\tilde{\lambda}$ (see Fig. 1).

It seems that such features not yet have been observed experimentally.

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