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QUANTUM THEORY OF SPIN WAVE RESONANCE IN THIN FERROMAGNETIC FILMS. PART I. SPIN WAVES IN THIN FILMS

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The essential facts and problems of spin wave resonance (SWR) are reviewed. Using the surface inhomogeneity (SI) model (an effective surface anisotropy field pinning the spins and identical on either surface of the film is introduced) as well as the Heisenberg model in the spin (exchange and Zeeman) Hamiltonian, an energy dispersion relation and spin-wave functions for the ferromagnetic film are derived. The theory is valid for Bravais lattices with arbitrary type of film surface orientation (provided it falls under the SI model) at arbitrary configuration of the static field and arbitrary value of the surface anisotropy. A surface parameter, having the physical meaning of a "measure" of surface spin pinning is introduced and is related to the surface anisotropy energy. The semi-classical picture of spin waves in thin films is elaborated and the influence of the surface parameter on the shape of standing spin waves is discussed. A detailed discussion of the properties and conditions of occurrence of surface spin waves in thin films is given as well as a method of their experimental identification.

1. *Introduction*

Spin wave resonance (SWR) consists in the absorption, by a ferromagnet immersed in a static magnetic film $\boldsymbol{H}$, of energy from an external electromagnetic field $\boldsymbol{h}$ oscillating perpendicularly to $\boldsymbol{H}$. There are commonly several and often more than 10 lines, in contradistinction to usual, simple ferromagnetic resonance. Each peak corresponds to excitation of a distinct spin wave. Kittel [1] was the first to have predicted the possibility of observing SWR in thin ferromagnetic films. An experimental confirmation was immediately provided by Seavey and Tannenwald [2] in permalloy samples. Soon after that, SWR was made apparent in thin samples of pure ferromagnetic metals (Fe, Ni, Co) and in ferrites ($NiFe_2O_4$, Suran [3]). Research by various authors permitted to establish and interprete the basic facts of SWR, though some problems are as yet unresolved.

Experimentally, SWR is carried out some well-defined configuration Φ of the static

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field $\boldsymbol{H}$ with regard to the surface of the film (we denote $\Phi \equiv \sphericalangle(\boldsymbol{H}, \boldsymbol{n}_z)$, with $\boldsymbol{n}_z$ the normal to the surface); most commonly, measurements are performed at perpendicular ($\Phi = 0$) or parallel $\left(\Phi = \frac{\pi}{2}\right)$ configuration. The alternating field $\boldsymbol{h}$ oscillates in the plane of the film perpendicularly to the static field $\boldsymbol{H}$. The film thickness ranges from 100 to 6000 Å. The AC field frequency is constant and in most cases lies within the 10–100 GHz range.

The basic fact of SWR consisted in the finding that the resonance spectrum of "good" samples (predominantly pure metals obtained in ultra-high vacuum) differed from those of "bad" ones (samples obtained in classical vacuum) (see, Kooi *et al.* [4]). In "good" samples, the separations $H_n - H_1$ between positions of the n-th ($n = 1, 2, 3, \ldots$) and first resonance peak grow according to an approximately n^2 law whereas the absorption of consecutive peaks diminishes in accordance with an approximately n^{-2} law. In "bad" samples, the separations are initially linear ($\sim n$) and, for higher n, quadratic in n; the absorption of the first several peaks is strikingly high and fails to follow the n^{-2} law.

The majority of existing SWR theories resort to the semi-classical approximation of the Landau-Lifshitz equations of magnetization motion. All the theories are unanimous in concluding that, in order that resonance shall occur, the film has to present inhomogeneities. The above stated, basic fact consisting in a relationship between the nature of the resonance spectrum and the technology of preparing the sample is accessible to explanation within the framework of two, fundamental models, namely the Surface Inhomogeneity (SI) and the Volume Inhomogeneity (VI) model. With regard to "good" samples, the approximation of the SI model is adequate; here, the internal spins are assumed to be acted on by an effective magnetic field which is homogeneous, whereas the surface spins are moreover subject to a "surface anisotropy field" affecting their pinning. The earlier SI theories took into consideration only the limiting cases of spin pinning: either "perfect freedom", or "perfect pinning" (Kittel [1] considered the latter case); subsequent theories included "partial pinning" (Soohoo [5], Wolf [6]). The SI model always leads to a quadratic k^2 law of resonance peak spacing (k — wave number); however, the experimental occurrence of deviations of the first several peaks from the k^2 law even in very "good" samples (Phillips and Rosenberg [7], Nisenoff and Terhune [8]) as yet lacks satisfactory explanation. The VI model, where the effective field within the sample is assumed as inhomogeneous, is efficacious in explaining the nature of the SWR spectrum of "bad" samples, but the results (linear spacing of the first several peaks, quadratic spacing of those with higher n) can hardly be said to depend on the type of volume inhomogeneity assumed there (effective field varying linearly with the distance from the central layer (Schlömann [9]), or varying quadratically (Portis [10], Hirota [11], Davies [12])).

Neither is there clarity as to the source and nature of the inhomogeneities. Surface inhomogeneity is predominantly considered to be due to the presence of a very thin foreign magnetic stratum on the surface proper of the sample. SWR measurements yield surface anisotropy energies of the order of 0.1 to 3 erg/cm^2 (Kooi *et al.* [13]) leading to the conclusion that the Néel surface energy is insufficient for giving rise to SWR. Wigen *et al.* [14, 15] consider the pinning of surface spins as due to a lower magnetization value at the film surface as compared with the magnetization within the film leading to a difference $4\pi\Delta M$ of demagnetizing fields ("dynamical pinning"). This explained certain effects observed in SWR (the critical effect, as well as the dependence of the SWR spectrum on frequency, see further on) but met with severe criticism on the part of Soohoo [5], who showed that a confrontation of the "dynamical pinning" theory with experiment required the assumption of quite unreasonable ΔM values of the order of the total magnetization of the sample. The source of volume inhomogeneities is seen chiefly in the magnetization varying along the normal through the sample. This inhomogeneity in magnetization can arise in the process of deposition by evaporation or, as shown by Ferchmin [16] as well as Sokolov and Tavger [17], can be "induced" by surface defect.

Various experiments have been performed with the aim of clarifying how a foreign stratum can influence the SWR spectrum of the sample. Kooi *et al.* [13] and Searle *et al.* [18] found SWR in 80% Ni permalloy to depend highly critically on the degree of surface oxidation: among other findings, an increase in oxidation entailed a very marked "damping" of peaks with even *n*. Searle *et al.* [18] ans Stankoff [19] deposited Fe or Ni strata several tens to several hundred Å thick on both surfaces of a (80% Ni) permalloy film and found this to modify the SWR spectrum essentially. These experiments are accessible to an explanation by the SI model and, consequently, argue in favour of the hypothesis that surface spin pinning depends on the presence of substances coating the surface of the sample; moreover, they permit the statement that, at perpendicular configuration, antiferromagnetic oxides cause an increase of spin pinning (particularly so Fe_2O_3; whose Néel temperature is higher than that of NiO), whereas a layer of pure ferromagnetic metal Fe or Ni causes a decrease in spin pinnig *i.e.* acts inversely.

There is no doubt as to the fact that SWR spectra depend on the configuration of the static field, but as yet no full explanation of this has been given. Results for different samples diverge. In most samples there exists a "critical" angle Φ_{crit}, for which the SWR spectrum reduces to a single absorption peak. As a rule, in such samples at angles $\Phi < \Phi_{\text{crit}}$ the spectrum consists of a large number of absorption peaks, whereas at $\Phi > \Phi_{\text{crit}}$ there are only several (2 to 3) peaks. This has been found to be the rule in numerous 80% Ni permalloy (both "good" and "bad") samples (Wigen *et al.* [14, 15], Rossing [20], Nisenoff and Terhune [8]) as well as in samples of cobalt [15] and $NiFe_2O_4$ ferrite (Suran [3]). On the other hand, in measurements carried out with a high degree of accuracy, Okochi and Nosé [21] failed to observe a critical effect in samples of 76% Ni permalloy. Searle *et al.* [18], in an 80% Ni permalloy sample "coated" with a 50 Å Fe stratum, obtained a single absorption peak at perpendicular configuration and a very well-defined multi-peak SWR spectrum at parallel configuration, which is a quite exceptional result. All this proves that the changes in SWR spectrum caused by a transition from one configuration of the static field to another depend very stronglyon the material of the film and on the chemical compound used for coating its surface. A quantum theory of the critical effect will be proposed in Part 2 of this paper; it will permit to explain the preceding, heterogeneous experimental results.

Comparisons by various authors between SWR from films prepared in identical conditions but differing in thickness had, essentially, the sole aim of ascertaining the presence (or lack) of an absorption peak attributable to the uniform spin wave ($k = 0$). This wave cannot be expected to be excited in massive metallic bodies (Kittel and Herring [22]), but cannot be ruled out in a thin film (thickness less than skin-effect depth). Theoretically, the position of the absorption peak of the uniform spin wave cannot depend on the film thickness. In experiments where the uniform peak was thought to appear, its position was said by the authors to be "virtually" independent of the thickness; in reality, this statement implied that, as the film thickness decreased, this peak shifted slightly towards *stronger* fields, whereas the other peaks shifted towards weaker fields. A number of authors have reported observing the "uniform" peak at various configurations (for the literature, *cf.* Ref. [21]). Thus, some report it to appear at perpendicular configuration only, whereas others restrict its occurrence to parallel configuration (see also Frait and Mitchell [23]); yet others admit both eventualities according to the material of the film (Searle *et al.* [18]), or decidedly exclude either (Okochi and Nosé [21]). Wigen *et al.* [14, 15], from an analysis of the critical effect, suggest that the absorption peak obtained at the configuration Φ_{crit} corresponds to the uniform mode, while Holzer *et al.* [24] uphold that the uniform peak occurs at configurations $\Phi_{\text{crit}} \leqslant \Phi \leqslant \frac{\pi}{2}$. The theory to be proposed here will reconcile these seemingly inconsistent results. It will be shown that the uniform spin wave can be excited in a film only in conditions of "natural" surface defect (absence of surface anisotropy) or if a "non-natural" surface defect is compensated in conditions of resonance. It is this author's belief that the former conditions cannot be achieved in practice; the latter will be shown to be fulfilled in the critical configuration; whereas the peak occurring at non-critical configurations and interpreted by various authors as the uniform peak turns out, in reality, to be one corresponding to a surface spin wave.

The problem of the occurrence of a surface peak in SWR has arisen but recently. Wolf [6] was the first to point to the theoretical possibility of its excitation. In later years, Sokolov *et al.* [25, 26] and, independently, Puszkarski [27, 28] worked out a method of identifying such a peak in the SWR spectrum. They showed theoretically that, in very thick films, its position has to be practically insensitive to thickness, whereas in thinner ones (10^2—10^3 Å) it should shift towards growing field strengths with decreasing thickness. Puszkarski [28] showed that a surface peak can appear in the SWR spectrum if the surface spins possess "freedom" in excess of the freedom arising by natural defect (*i.e.* if they are *unpinned*). From considerations to follow, it will be seen that such a situation can indeed occur in various samples at various configurations of the static field, leading to an explanation of the preceding divergences between experimental results (see, Part 2).

Experiments on SWR have revealed yet other facts: thus, strong roughness of the surface causes the spectrum to vanish, although in some cases its effect is restricted to a broadening of the lines (Searle *et al.* [18]). Inhomogeneity of oxidation in the plane of the film acts similarly (Wolff [6]). Also, the SWR spectrum shows a dependence on the AC field frequency, which affects the positions and intensities of the peaks strongly at parallel configuration but quite insignificantly at perpendicular configuration (Wolff [6], Nisenoff and Terhune [29]).

It is our aim to construct a quantum theory of SWR that will account for all the facts and provide a clarification of the as yet unresolved problems. Certain papers have dealt with SWR in a quantum approach, thus: Pincus [30] (linear chain), Ferchmin [31] and

Valenta and Wojtczak [32] (thin film of cubic structure), and Puszkarski [28] (hexagonal structure). The present theory is based on the SI model and comprises: (a) arbitrary Bravais structures, (b) arbitrary surface orientations (within the scope of the SI model), (c) arbitrary orientation of the static field, and (d) arbitrary surface anisotropy.

2. *Assumptions*

Let us consider a sample, consisting of non-conducting material, in the shape of a thin film of homogeneous crystallographical structure (arbitrary Bravais lattice) extending unboundedly in directions parallel to the surface (fulfilling Born-Kármán periodic boundary conditions in these directions). On the above assumptions, atoms lying in the same lattice layer parallel to the surface (to be termed in brief "a layer") are in identical physical conditions *i.e.* are mutually equivalent, forming a magnetic sublattice (Valenta [33, 34]). The approximation of the SI model consists in the distinction of only two sublattices in the film, namely a sublattice consisting of the two surface layers and one comprising all the internal layers. An atom is labelled by means of an index $\boldsymbol{lj}$, where l is a number denoting the layer, thus: $l = 0$ (substrate surface), $l = 1, 2, \ldots, L-2$ (internal layers), $l = L-1$ (free surface layer), whereas $\boldsymbol{j}$ is a two-dimensional vector lying in the plane of the film (there are N such vectors). The crystallographical XYZ axes are chosen as in Fig. 1, unit vectors being respectively $\boldsymbol{n}_x$, $\boldsymbol{n}_y$, $\boldsymbol{n}_z$.

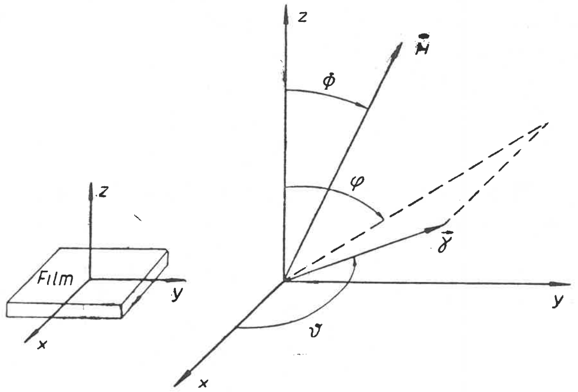

Fig. 1. Choice of XYZ coordinates. $\vec{\gamma}$ is the versor of the magnetization direction in the sample (in the text, the angle $\vartheta = \frac{\pi}{2}$ throughout); $\vec{H}$ is the static magnetic field

We perform the calculations in accordance with the Heisenberg localized spin model assuming an exchange (nearest neighbour interaction) and Zeeman Hamiltonian in standard form: $\hat{\mathscr{H}} = \hat{\mathscr{H}} + \hat{\mathscr{W}}$,

$$\hat{\mathscr{H}} = -2 \sum_{(\boldsymbol{lj};\, l+g,\, \boldsymbol{j}')} J_g \hat{\mathbf{S}}_{\boldsymbol{lj}} \cdot \hat{\mathbf{S}}_{l+g,\boldsymbol{j}'} - g\mu_B \sum_{\boldsymbol{lj}} \boldsymbol{H}_l^{eff} \cdot \hat{\mathbf{S}}_{\boldsymbol{lj}}, \tag{2.1}$$

$$\hat{\mathscr{W}} = -g\mu_B \sum_{\boldsymbol{lj}} \boldsymbol{h} \cdot \hat{\mathbf{S}}_{\boldsymbol{lj}}, \tag{2.2}$$

where summation extends over different pairs of neighbouring spins, J_g is the exchange integral between nearest neighbours situated respectively in layers l and $l \pm g$, whereas $\boldsymbol{H}_l^{eff}$ anf $\boldsymbol{h}$ are respectively the effective static magnetic field and the alternating magnetic field acting on a spin of layer l. We shall assume throughout $h \ll H_l^{eff}$, meaning that $\hat{\mathscr{W}}$ is a small perturbation with regard to $\hat{\mathscr{H}}$. On diagonalization of $\hat{\mathscr{H}}$ (we use the second quantization method), we calculate the peak intensities by methods of perturbation calculus in a first approximation.

Within the framework of the SI model, only such orientations of the film surface can be considered at which the nearest neighbours of a given spin belong to the same layer and the two closest lying layers: $g = 0, \pm 1$. Admitting only such orientations, a lack of active neighbours resulting from the thinness of the sample is "felt" only by spins lying in the surface. According to an analysis by Jelitto [35, 36], in cubic lattices orientations of this kind occur most commonly in practice (these are: *sc*(1, 0, 0), (1, 1, 0), (1, 1, 1); *fcc*(1, 1, 1), (1, 0, 0); *bcc* (1, 1, 0), (1, 0, 0)).

We shall be applying the following terminology: *Surface Defect* is a defect in the structure of the body due to: (1) the absence, with regard to atoms on the surface, of part of their neighbourhood and simultaneously (2) to the presence of an effective field $\boldsymbol{K}_{surf}$ on the surface; whereas *Natural Surface Defect* is the defect due solely to the finite thickness of the film.

Our calculations will be performed in the approximation of quasi-saturation. Consequently, the results will be valid only for temperatures low with respect to the Curie point of the material.

3. Diagonalization of the Hamiltonian

3.1 Equilibrium condition of the system

Let us assume, in a semi-classical approximation, a spin $\boldsymbol{S}_{lj} = S\vec{\gamma}$ (with S in $\hbar$ units) as present in each lattice site, $\vec{\gamma}$ being the quantization direction shared by all the spins. We determine the direction $\vec{\gamma}$ by minimalization of the expression $E_0(\vec{\gamma})$, derived from the Hamiltonian (2.1) on replacing therein the operators by classical spin vectors (Tyablikov [37]). We obtain:

$$\vec{\gamma} \| (\sum_l \boldsymbol{H}_l^{eff}) = L\boldsymbol{H}^{eff} + 2\boldsymbol{K}_{surf} \tag{3.1}$$

whence the dependence of the direction $\vec{\gamma}$ on the effective surface anisotropy field is seen to be significant in very thin films only (small L). For thicker films, we have $\vec{\gamma} \| \boldsymbol{H}^{eff}$, which leads to the following equation of equilibrium of the system:

$$\frac{H}{4\pi M} = \frac{\sin 2\varphi}{2 \sin(\varphi - \Phi)}\,. \tag{3.2}$$

We assume the externally applied static field $\boldsymbol{H}$ and the magnetization $\boldsymbol{M}$ as homogeneous throughout the sample and able to take any orientation with regard to its surface (see, Fig. 1) though remaining within the YZ-plane ($\vec{\gamma}$ is the versor of the $\boldsymbol{M}$ direction). The effective field $\boldsymbol{H}_l^{eff}$ within the sample is the sum of the external field and demagnetization field and, at the surfaces of the film, includes moreover the effective surface anisotropy field $\boldsymbol{K}_{surf}$:

$$\boldsymbol{H}_l^{eff} = \boldsymbol{H}^{eff} + (\delta_{0,l} + \delta_{l,L-1})\boldsymbol{K}_{surf}, \tag{3.2a}$$

$$\boldsymbol{H}^{eff} = \boldsymbol{H} - \boldsymbol{n}_z 4\pi M \cos\varphi. \tag{3.2b}$$

The alternating field $\boldsymbol{h}$ is linearly polarized in the X-direction, thus $\boldsymbol{h} = \boldsymbol{n}_x h_0 e^{i\omega T}$, and ω is the circular frequency.

The model of unidirectional surface anisotropy, represented on either surface by the effective field $\boldsymbol{K}_{surf}$, is implied (the case is assumed to be that of symmetrical boundary conditions). The field $\boldsymbol{K}_{surf}$ is considered as a phenomenological quantity accounting for the difference between the "pinning in $\vec{\gamma}$-direction" of the surface spins and internal spins, and containing contributions from all physical factors giving rise to differences between the effective fields active within the sample and on its surface (in particular, the field $\boldsymbol{K}_{surf}$ comprises the contribution from any foreign magnetic compounds that may be present on the surfaces of the sample). We assume $\boldsymbol{K}_{surf}$ as lying in the YZ-plane and subtending an angle α_0 with the Z-axis. Both α_0 and $\boldsymbol{K}_{surf}$ can take arbitrary values. Moreover, $\boldsymbol{K}_{surf}$ is assumed as independent of the configuration and strength of the field $\boldsymbol{H}$ and magnetization $\boldsymbol{M}$.

3.2. The diagonalizing transformations

With the aim of finding the eigenstates of the Hamiltonian (2.1), we perform four consecutive canonical transformations. The first transforms the spin operators from crystallographical X, Y, Z-axes to an orthogonal sytem of axes X′, Y′, Z′ with Z′-axis coinciding with the spin quantization axis $\vec{\gamma}$ and Y′-axis lying in the XOY-plane (Tyablikov [37]):

$$\hat{\boldsymbol{S}}_{lj} = \vec{\gamma}\hat{S}_{lj}^{z'} + \frac{1}{\sqrt{2}}(\boldsymbol{A}\hat{S}_{lj}^{\prime +} + \boldsymbol{A}^*\hat{S}_{lj}^{\prime -}); \tag{3.3}$$

$\boldsymbol{A}$ is a vector fulfilling the following relations:

$$(\vec{\gamma}\cdot\vec{\gamma}) = 1,\ (\vec{\gamma}\cdot\boldsymbol{A}) = 0,\ (\vec{\gamma}\times\boldsymbol{A}) = i\boldsymbol{A},$$

$$(\boldsymbol{A}\cdot\boldsymbol{A}^*) = 1,\ (\boldsymbol{A}\times\boldsymbol{A}^*) = i\vec{\gamma}. \tag{3.4}$$

Next, we go over from spin operators $\boldsymbol{S}'_{lj}$ to Bose operators by means of Holstein-Primakoff's transformation in the quasi-saturation approximation:

$$\hat{S}_{lj}^{\prime +} = \sqrt{2S}\,\hat{a}_{lj},\ \hat{S}_{lj}^{\prime -} = \sqrt{2S}\,\hat{a}_{lj}^{+},\ \hat{S}_{lj}^{z'} = S - \hat{a}_{lj}^{+}\hat{a}_{lj}. \tag{3.5}$$

$\hat{a}_{lj}^{+}$ and $\hat{a}_{lj}$ are respectively creation and annihilation operators of spin deviation, localized in the lattice site $l\boldsymbol{j}$, and satisfying the commutation rules:

$$[\hat{a}_{lj}, \hat{a}_{l'j'}^{+}] = \delta_{ll'}\delta_{jj'},\ [\hat{a}_{lj}, \hat{a}_{l'j'}] = 0. \tag{3.6}$$

In order that the Hamiltonian shall become diagonalized in the boson variables, we perform two more transformations (see, Corciovei [38], Ferchmin [31]): a Fourier transformation "in the plane" of the film,

$$\hat{a}_{lj}^{+} = \frac{1}{\sqrt{N}}\sum_{\vec{\varkappa}} e^{i(\vec{\varkappa}\cdot\boldsymbol{j})}\,\hat{b}_{\vec{\varkappa}l}^{+}, \tag{3.7}$$

with $\vec{\varkappa}$ denoting the reciprocal lattice vector, lying in the XOY-plane (the coordinates $\varkappa_x$ and $\varkappa_y$ are quantized in accordance with Born-Kármán conditions); and a transformation "in the direction" normal to the film,

$$\hat{b}^+_{\vec{\varkappa}l} = \sum_{\tau} u^*_l(\tau)\hat{\xi}^+_{\vec{\varkappa}\tau}. \tag{3.8}$$

On effecting the transformations (3.3), (3.5) and (3.7), the Hamiltonian (2.1) takes the following shape (constant terms are neglected, terms linear in the operators $\hat{b}^+_{\vec{\varkappa}l}$ and $\hat{b}_{\vec{\varkappa}l}$ vanish as a result of minimalizing $E_0(\vec{\gamma})$ (Tyablikov [37])):

$$\hat{\mathscr{H}} = \sum_{\vec{\varkappa}} \sum_{l} \sum_{g=0}^{\pm 1} R_{l,\,l+g}(\vec{\varkappa})\hat{b}^+_{\vec{\varkappa}l}\hat{b}_{\vec{\varkappa},\,l+g}, \tag{3.9a}$$

$$R_{l,\,l+g}(\vec{\varkappa}) = -2SJ_g\Gamma^{\vec{\varkappa}}_g + \delta_{0,g}[2S \sum_{d=0}^{l}{}' z_dJ_d + g\mu_B(H^{eff}_l \cdot \vec{\gamma})], \tag{3.9b}$$

$$\Gamma^{\pm\vec{\varkappa}}_g = \sum_{\boldsymbol{j}'}{}' e^{\pm i\vec{\varkappa}\cdot(\boldsymbol{j}-\boldsymbol{j}')},$$

$$(\boldsymbol{j} \in l,\ \boldsymbol{j}' \in l+g,\ \boldsymbol{j}\ \text{given}) \tag{3.9c}$$

$$\Gamma^{\vec{\varkappa}}_{-g} = \overset{*}{\Gamma}{}^{\vec{\varkappa}}_g = \Gamma^{-\vec{\varkappa}}_g,\ \Gamma^{\vec{\varkappa}}_0 = \overset{*}{\Gamma}{}^{\vec{\varkappa}}_0,\ \Gamma^0_g = z_g. \tag{3.9d}$$

$\sum_{\boldsymbol{j}}{}'$ stands for summation over the nearest neighbours of a lattice site; summation over nearest layers (up to layer l) is denoted by $\sum_g{}^l$ (but $\sum_{g=0}^{l}$ denotes summation including the layer l); and z_g symbolizes the number of nearest neighbours (of a lattice site of layer l) lying in the layer $l \pm g$.

The transformation (3.8) makes the Hamiltonian go over into the diagonal form:

$$\hat{\mathscr{H}} = \sum_{\vec{\varkappa}\tau} E(\vec{\varkappa}, \tau)\hat{\xi}^+_{\vec{\varkappa}\tau}\hat{\xi}_{\vec{\varkappa}\tau}, \tag{3.10}$$

where $\hat{\xi}^+_{\vec{\varkappa}\tau}$ is the creation operator of a spin wave of energy $E(\vec{\varkappa}, \tau)$, provided the functions $u_l(\tau)$ satisfy (1) the set of difference equations

$$u_l(\tau)E(\vec{\varkappa}, \tau) = \sum_{g=0}^{l} R_{l,\,l+g}(\vec{\varkappa})u_{l+g}(\tau),\ l = 0, 1, \ldots, L-1 \tag{3.11}$$

and (2) the orthonormality conditions

$$\sum_l u_l(\tau)u^*_l(\tau_0) = \delta_{\tau\tau_0}, \tag{3.12a}$$

$$\sum_\tau u_l(\tau)u^*_{l_0}(\tau) = \delta_{ll_0}. \tag{3.12b}$$

The set of Eqs (3.11) can be rewritten in the form:

$$[R(\vec{\varkappa}) - E(\vec{\varkappa}, \tau) - a_l]u_l(\tau) - 2S \sum_g{}^l J_g\Gamma^{\vec{\varkappa}}_g u_{l+g}(\tau) = 0, \qquad l = 0, 1, \ldots, L-1; \tag{3.13a}$$

$$R(\vec{\varkappa}) = R(-\vec{\varkappa}) = 2SJ_0(z_0 - \Gamma^{\vec{\varkappa}}_0) + 4Sz_1J_1 + g\mu_B(\boldsymbol{H}_{eff} \cdot \vec{\gamma}); \tag{3.13b}$$

$$a_l = 0 \quad \text{for} \quad l = 1, 2, \ldots, L-2,$$

$$a_0 = a_{L-1} \equiv a = 2Sz_1J_1 - g\mu_B(\boldsymbol{K}_{surf} \cdot \vec{\gamma}), \tag{3.13c}$$

which is conveniently solved by a method due to Jelitto [35]: one introduces two "fictitious" layers $l = -1$ and $l = L$, thus replacing the set of L equations (3.13a) by an equivalent set of $L+2$ equations consisting of two "subsets":

(1) the system of equations of the "bulk body" ($a_l \equiv 0$, $\sum\limits_g^{l} \equiv \sum\limits_{g=\pm 1}$):

$$-2SJ_1\overset{*}{\Gamma}{}_1^{\vec{\varkappa}}u_{l-1}(\tau)+[R(\vec{\varkappa})-E(\vec{\varkappa},\tau)]u_l(\tau)-2SJ_1\Gamma_1^{\vec{\varkappa}}u_{l+1}(\tau)=0,$$

$$l = 0, 1, \ldots, L-1 \tag{3.14a}$$

(2) the system of boundary equations:

$$Au_0(\tau) = \frac{1}{z_1}\Gamma_1^{*\vec{\varkappa}}u_{-1}(\tau),$$

$$Au_{L-1}(\tau) = \frac{1}{z_1}\Gamma_1^{\vec{\varkappa}}u_L(\tau), \tag{3.14b}$$

with the notation:

$$A \equiv \frac{a}{2Sz_1J_1} = 1-\frac{g\mu_B}{2Sz_1J_1}(\boldsymbol{K}_{surf}\cdot\vec{\gamma}). \tag{3.15}$$

The sets (3.13a) and (3.14a, b) are indeed equivalent, since on eliminating the fictitious layers from Eqs (3.14a) by means of Eqs (3.14b) one returns to the set (3.13a).

Eqs (3.14a) can be considered as boundary equations imposed on the general solution of the set (3.14b). We shall refer to the quantity A in the boundary equations as the *surface parameter* (not to be confounded with exchange constants, commonly denoted by an A in the literature). In Section 5, it will be shown that A measures the degrees (strength) of "pinning" of the surface spins. The next subsection will be devoted to solving the set of Eqs (3.14).

3.3. The dispersion relation, and the transformation functions $u_l(\tau)$

Introducing a particular solution of the form:

$$u_l(\tau) = e^{i\tau l} \tag{3.16}$$

into the set of equations of the "bulk body", we come to the dispersion formula:

$$E(\vec{\varkappa},\tau) \equiv E(\vec{\varkappa},\tau') = g\mu_B(\boldsymbol{H}^{eff}\cdot\vec{\gamma})+$$

$$+2SJ_0(z_0-\Gamma_0^{\vec{\varkappa}})+4SJ_1(z_1-|\Gamma_1^{\vec{\varkappa}}|\cos\tau'), \tag{3.17}$$

where τ' is a new quantum number, defined as follows

$$\tau' = \tau+\varphi, \tag{3.18}$$

$$\Gamma_1^{\vec{\varkappa}} = |\Gamma_1^{\vec{\varkappa}}|e^{i\varphi}, \quad \varphi = \varphi(\vec{\varkappa}). \tag{3.19}$$

The quantity φ is yet a new quantum number, the value of which is defined by the *structural factor* $\Gamma_1^{\vec{\varkappa}}$. It is worth noting that φ is non-zero only if the nearest neighbours acting from the layer $g = +1$ are distributed non-symmetrically with respect to the projection of the lattice site under consideration onto that layer. For a given $\vec{\varkappa}$ and well-defined structure and orientation of the surface, φ takes one and only one value[1].

The quantum number τ' can in general be complex; however, from the condition of real energy $E(\vec{\varkappa}, \tau')$ it results that only three "types" of τ' are permitted, to each of which there corresponds a different kind of spin wave:

(1) $\tau' = k$ — space spin waves,

(2) $\tau' = it$ — "acoustic" surface spin waves,

(3) $\tau' = \pi + it$ — "optical" surface spin waves. (3.20)

Above, k and t are real numbers. A justification of the preceding terminology will be given in the next Section. On inserting (3.20) into Eq. (3.17) one obtains formulas which show that the acoustical and optical surface spin waves have energies lying respectively below and above the energy band of the space spin waves.

The energy values $E(\vec{\varkappa}, \tau')$, as obvious from Eq. (3.17), are twice degenerate, being related to two particular solutions of the type (3.16) with quantum numbers $\tau_1 = -\varphi+\tau'$ and $\tau_2 = -\varphi-\tau'$. The general solution of the set (3.14a) is thus of the form:

$$u_l(\tau) = e^{-i\varphi l}u_l(\tau'), \qquad u_l(\tau') = c_1 e^{i\tau' l} + c_2 e^{-i\tau' l}. \tag{3.21}$$

The superposition constants c_1 and c_2 are to be had from the boundary conditions (3.14b) and normalization condition (3.12a). After some calculations, we obtain two kinds of normalized solutions:

$$u_l^+(\tau') = c_+(\tau')\cos\left(\frac{L-1}{2}-l\right)\tau' \qquad \text{(symmetric solutions)}, \tag{3.22a}$$

$$u_l^-(\tau') = c_-(\tau')\sin\left(\frac{L-1}{2}-l\right)\tau' \qquad \text{(antisymmetric solutions)}, \tag{3.22b}$$

the normalization factor being:

$$c_\pm(\tau') = \sqrt{2}\left(L \pm \frac{\sin L\tau'}{\sin\tau'}\right)^{-1/2}. \tag{3.23}$$

3.4. Characteristic equation

We still have to find what τ' — values satisfy the boundary conditions (3.14b). Inserting (3.22a, b) into Eqs (3.14b), we get the following *characteristic equations* determining τ':

$$f(\tau') \equiv \cos\frac{L+1}{2}\tau'/\cos\frac{L-1}{2}\tau' = A(\vec{\varkappa}) \qquad \text{for symmetric waves}, \tag{3.24a}$$

$$g(\tau') \equiv \sin\frac{L+1}{2}\tau'/\sin\frac{L-1}{2}\tau' = A(\vec{\varkappa}) \qquad \text{for antisymmetric waves}, \tag{3.24b}$$

where we have used the notation:

$$A(\vec{\varkappa}) = z_1 A|\Gamma_1\vec{\varkappa}|^{-1}, \qquad A(0) \equiv A. \tag{3.25}$$

---

[1] **A method of computing structural factors, and its application to cubic structures, has been proposed by Jelitto [36].**

The spectrum of allowed τ'-values is thus seen to depend on the surface parameter A and (by way of the structural factor) on the quantum number $\vec{\varkappa}$ as well as the structure and orientation of the surface. It is easily verified that all distinct solutions $u_l(\tau')$ are obtained on restriction to the interval $k \in (0, \pi)$ for space spin waves and $t \in (0, +\infty)$ for surface spin waves.

There is no general analytical method available for obtaining solutions of the equations (3.24) for arbitrary values of the quantity $A(\vec{\varkappa})$. In order to gain some essential information on the τ'-spectrum, we shall resort to a graphical method. In Fig. 2 are plotted the functions $f(\tau')$ and $g(\tau')$ (in the middle — for space waves; to the right and left, respectively, for optical and acoustical surface waves). On fixing some value of the parameter $A(\vec{\varkappa})$, the roots of the characteristic equations are found by searching for the points of intersection of the straight line $A(\vec{\varkappa}) = \text{const}$ (which is parallel to the axis of abscissae) with the curves. This graph shows that:

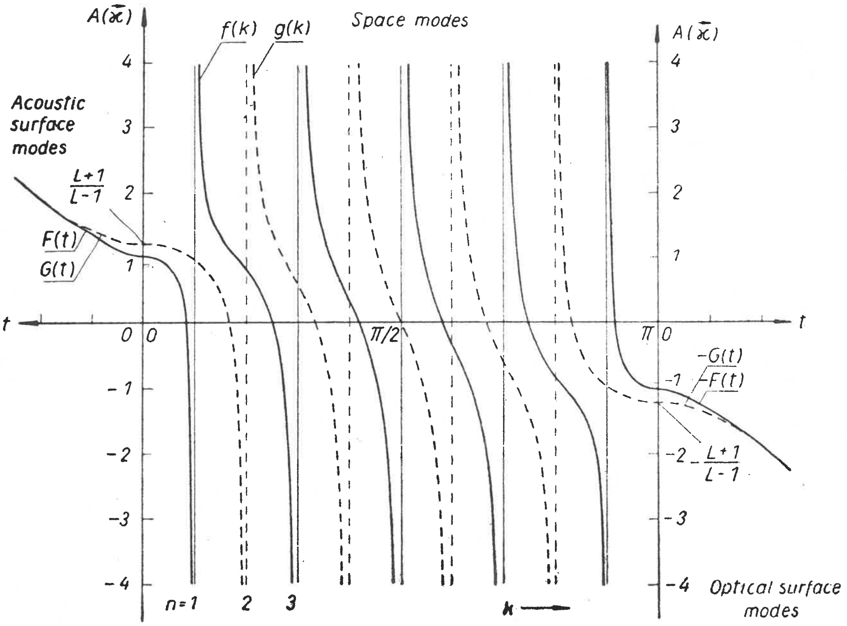

Fig. 2. Accessory graph for the discussion of the characteristic equations (3.24a, b) (here, we denot, $F(t) = f(it)$; $G(t) = g(it)$. Dashed lines — antisymmetric states; continuous lines — symmetric states. n — labels the states. $L = 11$ is assumed

1) We always obtain L distinct values of τ'; for $|A(\vec{\varkappa})| \leqslant 1$ the spectrum consists of space states only, whereas for $|A(\vec{\varkappa})| > 1$ also surface states appear (one such state or two states, see Table I);

2) As $A(\vec{\varkappa})$ increases, all values of the τ'-spectrum shift towards lower energies; the density of states varies insignificantly for the space waves but strongly for the surface waves (for the latter, at sufficiently large $A(\vec{\varkappa})$, we have practically twofold degeneracy);

3) The uniform wave ($k = t = 0$) — the limiting case between space waves and acoustical surface waves — occurs in the spectrum for two, particular values only: $A(\vec{\varkappa}) = 1$ (for symmetric waves) and $A(\vec{\varkappa}) = (L+1)(L-1)^{-1}$ (for antisymmetric waves).

It will now be our convention to label the spin-wave states corresponding to distinct τ'-values by numbers $n = 1, 2, \dots L$ running from the energetically lowest state. In this way, to a root τ' of the characteristic equations, a number is attributed according to the following principle: (1) The curves in Fig. 2 are labelled, beginning from the one furthest to the left, by numbers $n = 1, 2, \dots L$ (but note that the curves $n = 1, 2, L-1$ and L consist of two halves each, the one half "belonging" to space and the other half to surface states); (2) a root (t or k) is labelled with the number of the curve from which it was obtained by the graphical procedure.

TABLE I

	$\lvert A(\vec{\varkappa})\rvert > \dfrac{L+1}{L-1}$	$1 < A(\vec{\varkappa}) < \dfrac{L+1}{L-1}$	$A(\vec{\varkappa}) \leqslant 1$
number of space states	$L-2$	$L-1$	L
number of surface states	2	1	0

TABLE II

	k	n for symmetric waves	n for antisymmetric waves
$A(\vec{\varkappa}) = 0$	$n\dfrac{\pi}{L+1}$	$1, 3, 5, \dots, L$*	$2, 4, 6, \dots, L-1$
$A(\vec{\varkappa}) = 1$	$(n-1)\dfrac{\pi}{L}$	$1, 3, 5, \dots, L$	$2, 4, 6, \dots, L-1$
$A(\vec{\varkappa}) = -1$	$n\dfrac{\pi}{L}$	$1, 3, 5, \dots, L$	$2, 4, 6, \dots, L-1$
$\lvert A(\vec{\varkappa})\rvert = \infty$	$n\dfrac{\pi}{L-1}$	$1, 3, 5, \dots, L-2$	$2, 4, 6, \dots, L-3$

* L is assumed odd.

For certain particular values of $A(\vec{\varkappa})$, it is possible to obtain strict analytical formulas for roots k (see, Table II). The roots k for all other values of $A(\vec{\varkappa})$ and roots t for surface waves can be determined analytically to within some approximation only. Approximate formulas for roots t will be adduced in subsection 4. 3. For space waves, the following approximation is of particular practical interest:

$$k_n = (n-\delta)\,\frac{\pi}{L-1}\,, \tag{3.26}$$

with $L-1$ defining the film thickness (in lattice units). On inserting (3.26) into (3.24a, b) we obtain the following equation whence δ can be determined:

$$\sin\left[\delta\,\frac{L+1}{L-1}-\frac{2n}{L-1}\right]\frac{\pi}{2}=A(\vec{\varkappa})\sin\delta\,\frac{\pi}{2},$$

$$\begin{aligned}&n=1,3,5,\ldots\ \text{for symmetric waves,}\\&n=2,4,6,\ldots\ \text{for antisymmetric waves.}\end{aligned}\tag{3.26a}$$

It hence results that the "shift" δ is a function of the label of the wave, the parameter $A(\vec{\varkappa})$, and the thickness L; thus: $\delta=\delta(A(\vec{\varkappa}), L, n)$ with, always, $\delta\in(0, +2)$.

4. *Spin waves*

4.1. Semi-classical picture of spin waves in thin films

The transformations (3.7) and (3.8) can be rendered jointly by a single transformation, as follows:

$$\hat{a}^{+}_{l\boldsymbol{j}}=\sum_{\vec{\varkappa}}\sum_{\tau}u_{l\boldsymbol{j}}(\vec{\varkappa},\tau)\hat{\xi}^{+}_{\varkappa\tau}.\tag{4.1}$$

Acting with each side of this equality on the ground state $|0\rangle$, we obtain:

$$|l\boldsymbol{j}\rangle=\sum_{\vec{\varkappa}\tau}u_{l\boldsymbol{j}}(\vec{\varkappa},\tau)|\vec{\varkappa},\tau\rangle,\tag{4.2}$$

where $|l\boldsymbol{j}\rangle$ and $|\vec{\varkappa},\tau\rangle$ are, respectively, the state with one spin deviation localized in the lattice site $l\boldsymbol{j}$, and the state with one spin wave characterized by quantum numbers $\vec{\varkappa}, \tau$. By equalities (4.2), $u_{l\boldsymbol{j}}(\vec{\varkappa},\tau)$ is a function of the spin wave in Bloch's meaning. On adjoining the standard time factor, we obtain from Eq. (3.21), with T the time:

$$u_{l\boldsymbol{j}}(\vec{\varkappa},\tau',\varphi)e^{-i\frac{E(\vec{\varkappa},\tau')}{\hbar}T}=\frac{1}{\sqrt{N}}\exp i\,[\vec{\varkappa}\cdot\boldsymbol{j}+\varphi l-\hbar^{-1}E(\vec{\varkappa},\tau')\,T]\,u^{*}_{l}(\tau'),\tag{4.3}$$

whence the spin wave is found to be a plane wave characterized by the four quantum numbers $\varkappa_x$, $\varkappa_y$, φ and τ'.

To use the picture of precessioning classical spins (precession about $\vec{\gamma}$), one sees from (4.3) that the amplitude of the precession cone $N^{-\frac{1}{2}}u_l(\tau')$ varies from one layer to another in a manner depending on the quantum number τ' (in bulk bodies, the precession amplitudes of all spins are the same), the precession frequency is $\hbar^{-1}E(\vec{\varkappa},\tau')$, whereas the initial phase in the lattice point $l\boldsymbol{j}$ is $\vec{\varkappa}\cdot\boldsymbol{j}+\varphi l$. Propagation of a spin wave consists in displacement of the plane of constant phase in the direction $\vec{\varkappa}+\varphi\boldsymbol{n}_z$ (the quantum number $\vec{\varkappa}$ is the wave vector of propagation in the plane of the film and φ the wave number of propagation in that of the normal $\boldsymbol{n}_z$). Certain highly significant differences appear with regard to the picture of spin wave propagation in bulk bodies; namely, in thin films:

1) For any given vector $\vec{\varkappa}$, the number φ takes (see, subsection 3.3) only one value depending on the structure of the body and type of orientation of the surface. This means

that, given a thin film sample and a well-defined propagation direction $\vec{\varkappa}$ in the plane of the film, only one oblique direction of spin wave propagation is allowed;

2) Propagation in the direction of the normal is due to structural causes, since the shift in phase by the amount φ on going over from layer l to layer $l+1$ (see, Fig. 3) is due to the asymmetric distribution, in layer $l+1$, of nearest neighbours of the lattice site lj. As a consequence, the energy of the spin wave does not depend on the quantum number φ

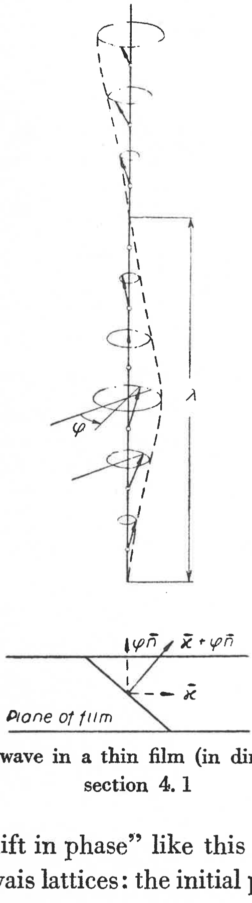

Fig. 3. Semi-classical picture of a spin wave in a thin film (in direction of the normal). Explained in subsection 4. 1

(*cf.* Eq. (3.17)). (A "structural shift in phase" like this occur sin bulk bodies only in the case of structures that are not Bravais lattices: the initial precession phases of spins belonging to different sublattices are different; *cf.* the case of hexagonal cobalt dealt with by Low [39] and Valenta [40].)

In the case $\vec{\varkappa} = 0$ (which involves $\varphi = 0$), the spin wave function is of the form:

$$u_{lj}(\vec{\varkappa}, \tau', \varphi) = \frac{1}{\sqrt{N}} u_l(\tau'), \tag{4.4}$$

stating that all spins perform a precession in phase with one another. For this reason, the function $u_l(\tau')$ is commonly referred to in the literature as that of the standing spin wave (*mode*). Since the quantum number τ' takes L values, L is the number of types of spin-wave modes able to appear in a thin film for a given value of the surface parameter.

4.2. Effect of the surface parameter on the spin-wave modes

The shape of the spin-wave mode $u_l(\tau')$ depends on the value of τ', which in turn depends on the surface parameter A by way of the characteristic equation. The following, general rule can be stated: for real τ', the spin-wave mode amplitudes vary sine-wise along the thickness of the film (oscillation modes or *space modes*; $n-1$ is the number of all nodes of the n-th mode), whereas for complex τ' the waves are of a "localized" nature, since their amplitudes vary monotonously from a largest value at the surface to a smallest value in the middle of the film (*surface modes*). It is readily verified that the amplitudes of two adjacent layers l and $l \pm 1$ have opposite signs for $\tau' = k \in \left(\frac{\pi}{2}, \pi\right)$ and $\tau' = \pi + it$; by analogy with the terms employed in the theory of vibrations of the dia-atomic chain, we shall refer to such spin-wave modes as "optical" ones. On the other hand, modes corresponding to $\tau' = k \in (0, \pi/2)$ and $\tau' = it$ will be referred to as "acoustical". The shapes of some low-energy (*i.e.* acoustical) spin-wave modes corresponding to different values of the surface parameter A are shown in Fig. 4; we now proceed to discuss certain conclusions from these graphs.

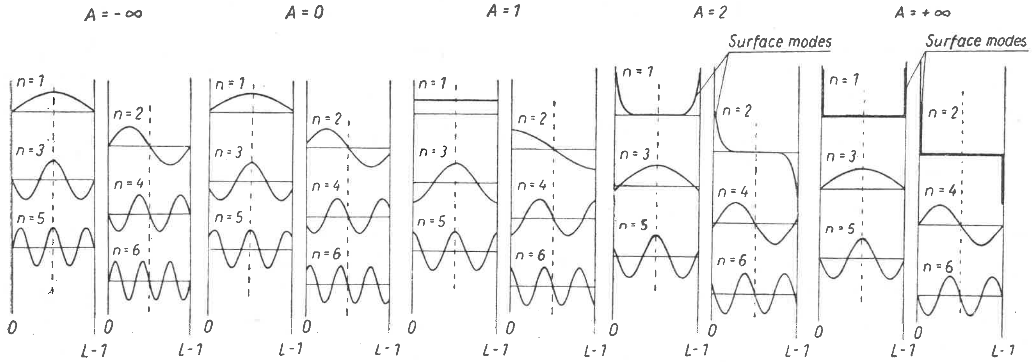

Fig. 4. Shapes of spin wave modes (with low n) for various values of the surface parameter A. Symmetric and antisymmetric modes correspond to odd and even n, respectively

As A increases (consider Fig. 4 in the horizontal direction), the oscillations of all of the space modes decrease (*i.e.* the effective number of wavelength "halves" per mode decreases) and the localization of the surface modes augments (see, Fig. 5). This last property is illustrated in a different way in Fig. 6, which shows the surface mode amplitude in a given lattice layer l in its dependence on the number t (it will be kept in mind that to an increase in A there corresponds an increase in t, see Fig. 2). It may be of significant interest to draw attention (Fig. 5) to the manner in which the shapes of the two lowest modes ($n = 1$ and $n = 2$) change with varying A. These modes are space modes for some values of A and surface modes upward of a well-defined, critical value of A (*cf.*, Fig. 2). The change in shape is found to be continuous, the mode for $\tau' = 0$ ($k = t = 0$) being of the nature of a limiting, space-surface mode. (Note the shape of the antisymmetric mode $k = 0$, Fig. 5, at $A = 1.083$.)

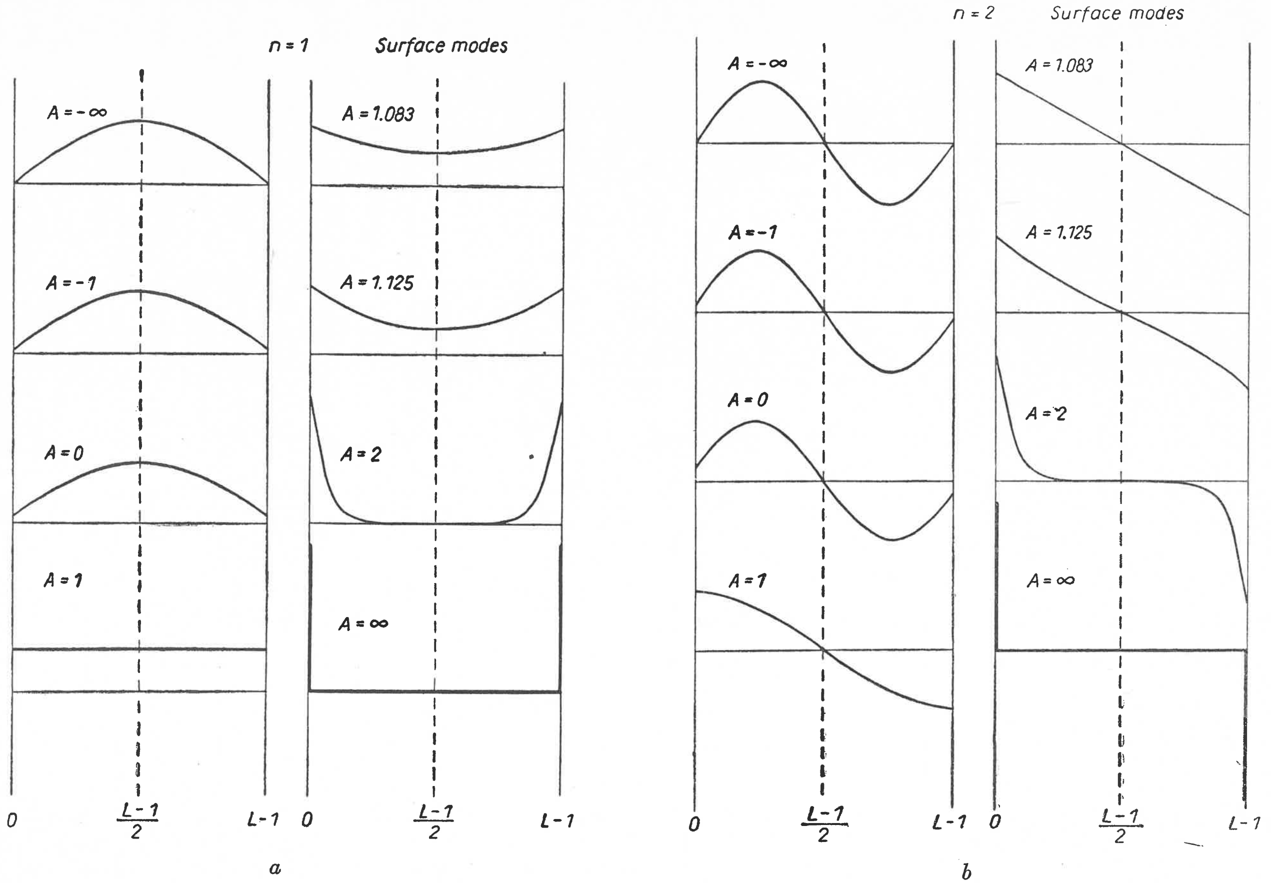

Fig. 5. Shape of (*a*) the symmetric spin wave mode $n = 1$ and (*b*) antisymmetric mode $n = 2$ in their dependence on the surface parameter A. At a well-defined limiting value of A the modes change their character from oscillational to localized

From the preceding, we easily find the physical meaning of the quantum numbers k and t: these are, respectively, the wave number of the space mode and the *localization increment* of the surface mode.

4.3. Energy of surface spin waves

Localized states are of considerable interest in solid state theory. The whole of this subsection will be devoted to a discussion of the general formula (3.17), which we shall specialize to render the energy of accoustical surface spin-wave states.

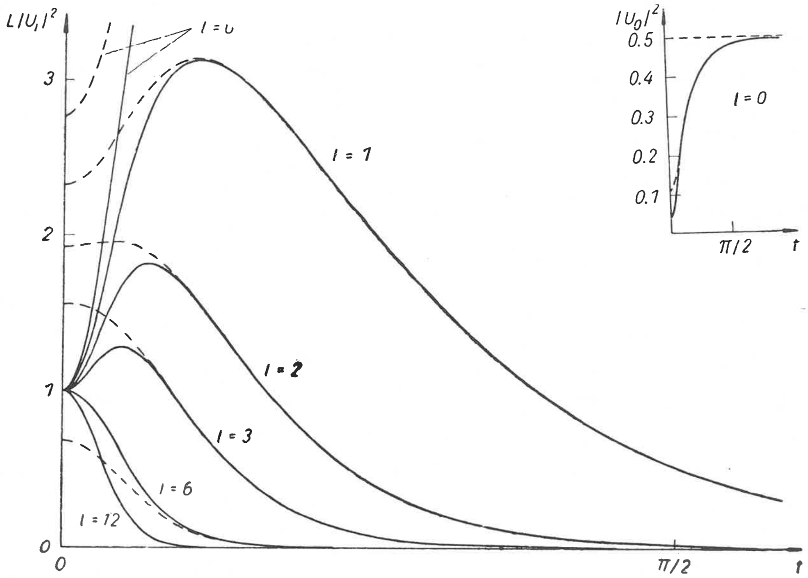

Fig. 6. Amplitudes of surface spin-wave modes (symmetric — continuous line, antisymmetric — dashed line) for various layers in their dependence in the localization increment t (l labels the layer; the film contains 25 layers). The graph to the right deals with the case $l = 0$ separately. As t increases, the modes are found to localize at the surface

Inserting $\tau' = it$ into Eqs (3.24) and introducing the notation $v = e^{-t}$, we obtain after some calculations:

$$\frac{1 \pm v^{L+1}}{1 \pm v^{L-1}} = A(\vec{\varkappa})\, v. \tag{4.5}$$

Throughout this subsection, the upper sign ("+" in Eq. (4.5)) corresponds to symmetric states, and the lower sign ("—" in Eq. (4.5)) to antisymmetric states. Eq. (4.5) will now be solved in the approximation

$$v^L = e^{-tL} \ll 1. \tag{4.6}$$

This inequality is satisfied at $L \gg 1$ or at $A(\vec{\varkappa}) \gg 1$. Resorting to (4.6), we can rewrite Eq. (4.5) as follows:

$$1 - A(\vec{\varkappa})v = \pm v^{L-1}(1 - v^2). \tag{4.7}$$

Since the right hand term is very small, we can assume in a first approximation $v = A(\vec{\varkappa})^{-1}$. In the next approximation, we obtain:

$$v = A(\vec{\varkappa})^{-1} \mp A(\vec{\varkappa})^{-L}[1-A(\vec{\varkappa})^{-2}] \tag{4.8}$$

which, with (3.17), finally leads to the following, approximate formula for the energy of surface states:

$$E(\vec{\varkappa}, A) = g\mu_B(\boldsymbol{H}^{eff}\cdot\vec{\gamma}) + 2SJ_0(z_0 - \Gamma_0^{\vec{\varkappa}}) + 4SJ_1z_1 - \\ -2SJ_1|\Gamma_1^{\vec{\varkappa}}|[A(\vec{\varkappa}) + A(\vec{\varkappa})^{-1}] \mp 2SJ_1|\Gamma_1^{\vec{\varkappa}}|A(\vec{\varkappa})^{-(L-2)}[1-A(\vec{\varkappa})^{-2}]^2. \tag{4.9}$$

As already mentioned (subsection 3.3), the acoustical surface states lie below the space state branch in the energy spectrum. In addition, Eq. (4.9) gives us the following information (for comparison, the respective properties of the two space states lying lowest in the energy spectrum are adduced in brackets):

1) the symmetric state has an energy lower than that of the antisymmetric state (the same is true of the space states);

2) splitting between the two states amounts to

$$\Delta E_{surf} = 4SJ_1|\Gamma_1^{\vec{\varkappa}}|\, A(\vec{\varkappa})^{-(L-2)}[1-A(\vec{\varkappa})^{-2}]^2, \tag{4.10}$$

and decreases with growing $A(\vec{\varkappa})$ and L (exponentially) (the energy gap between space states is much less sensitive to changes in $A(\vec{\varkappa})$ and L);

3) the position of the *centre of energy*, about which the two surface states are disposed symmetrically, does not depend on the thickness L (but does so in the case of space states);

4) with growing film thickness L, the symmetric state shifts towards higher energies and the antisymmetric state towards lower energies (whereas both space states shift towards lower energies). These properties can be resorted to as criteria for the experimental identification of surface states (*e. g.* in spin-wave resonance).

Eq. (4.9) and the conclusions to which it leads are, in principle, valid for the majority of samples used in experimental work, since their thickness ranges from 10^2 to 10^4 Å, corresponding to $L > 30$ and satisfying the condition (4.6) with sufficient accuracy. In the case of very thick films, one can neglect the splitting ΔE_{surf} in Eq. (4.9); in this approximation, Eq. (4.8) yields:

$$t = \ln A(\vec{\varkappa}) \tag{4.11}$$

and the energy of the two surface states (the symmetric and antisymmetric one) is practically the same and does not depend on L.

All formulas of this subsection extend to optical surface states on replacing $A(\vec{\varkappa})$ by $-A(\vec{\varkappa})$.

5. *The surface parameter as a measure of the pinning of surface spins*

5.1. Semi-classical interpretation

The notion of *pinning* was first introduced by Kittel [1] with regard to spins to account for their tendency to "keep to" the magnetization direction $\vec{\gamma}$. In the literature, various quantities serve as a measure of surface spin pinning. In this Section, we shall propose the surface parameter as a convenient measure.

Dealing with a spin as a semi-classical vector $\boldsymbol{S}_{lj}$ we can take, as an absolute measure of its pinning in the $\vec{\gamma}$-direction, the energy ε_{lj} possessed by it in the ground state of the system. For surface spins, we obtain (see (2.1)):

$$\varepsilon_{surf} = \varepsilon_{int} + 2S^2 z_1 J_1 A, \tag{5.1}$$

where $\varepsilon_{int} = -2S^2 z_0 J_0 - 4S^2 z_1 J_1 - g\mu_B S(\boldsymbol{H}^{eff} \cdot \vec{\gamma})$ is the energy of an internal spin in the ground state. By (5.1), the surface parameter A can be considered as a *relative measure* of the pinning of surface spins as compared with the pinning of internal spins. At $A = 0$, the two kinds of spins are equally „strongly" pinned; at $A > 0$ the surface spins are the freer ones; whereas at $A < 0$ they are more strongly pinned (less free) than the spins within the film. The difference in pinning between surface and internal spins is due to natural defect as well as to the action of the surface anisotropy field $\boldsymbol{K}_{surf}$. Natural defect alone, to which corresponds the value $A = 1$, obviously sets free the surface spins; let us agree to denote as *natural freedom* the state of surface spin pinning which corresponds to $A = 1$.

The field $\boldsymbol{K}_{surf}$ of surface anisotropy *contributes additionally* towards freeing or stronger pinning of the surface spins, thus changing their freedom to greater or lesser than natural. It is common usage in the literature to say that a surface spin is "unpinned" in the former case but "pinned" in the latter. As a measure of this "anisotropic surface spin pinning", we can take the energy of a surface spin when in the field $\boldsymbol{K}_{surf}$ or, equivalently, the energy of all the spins present per unit area of the surface (in ergs per cm²):

$$E_s = -g\mu_B S d^{-2}(\boldsymbol{K}_{surf} \cdot \vec{\gamma}) = 2S^2 z_1 J_1 d^{-2}(A-1), \tag{5.2}$$

with d — the lattice constant. In brief, we shall be using the following terminology[2]:

1) At $A = 1$ ($E_s = 0$), the surface spins have natural freedom;
2) At $A > 1$ ($E_s > 0$), they are unpinned;
3) At $A < 1$ ($E_s < 0$), they are pinned.

In the situation when $A = 0$, the natural freedom of surface spins is compensated, and their pinning becomes equal to that of internal spins. The cases $A = +\infty$ and $A = -\infty$ are, respectively, those of "perfect freedom" and "perfect pinning". The last case is the one considered by Kittel [1] (*cf.* Fig. 4).

5. 2. Quantum-theoretical interpretation

As a quantum-theoretical measure of the pinning of a spin $\boldsymbol{S}_{lj}$ we can assume the mean energy $\overline{E}_{lj}$ of the state $\hat{a}_{lj}^{+}|0\rangle$ *i. e.* the state with one spin deviation localized in the site lj. Eq. (4.2) now yields:

$$\overline{E}_{lj} = \sum_{\vec{\varkappa}\tau} |u_{lj}(\vec{\varkappa}, \tau)|^2 E(\vec{\varkappa}, \tau). \tag{5.3}$$

The larger is $\overline{E}_{lj}$, the more difficult it is to deviate the spin simultaneously creating a "packet"

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[2] In our earlier papers [27, 28, 41], the terms "pinned" and "unpinned" occur in a different meaning.

of spin waves. For thin films, it results that $\overline{E}_{lj}$ depends only on the number l labelling the layer (by Eq. (4.3)) and on the surface parameter A:

$$\overline{E}_{lj} \equiv \overline{E}_l(A) = \sum_{\vec{\varkappa}} \sum_{\tau'} N^{-1} |u_l(\tau')|^2 E(\vec{\varkappa}, \tau'). \tag{5.4}$$

An idea of $\overline{E}_l = \overline{E}_l(A)$ for surface spin ($l = 0$) is to be had from Fig. 7, showing "spectra" of $|u_l(\tau')|^2$ for various values of A (for simplicity, we have put $\vec{\varkappa} = 0$). With growing A, the position of the maximum of $|u_l(\tau')|^2$ shifts towards the bottom of the energy band, proving that $\overline{E}_0(A)$ is smaller for larger values of A *i. e.* that surface spin pinning is weaker. A similar analysis for internal layers (Kurczewski [42]) shows that their $|u_l(\tau')|^2$-spectra vary with A insignificantly.

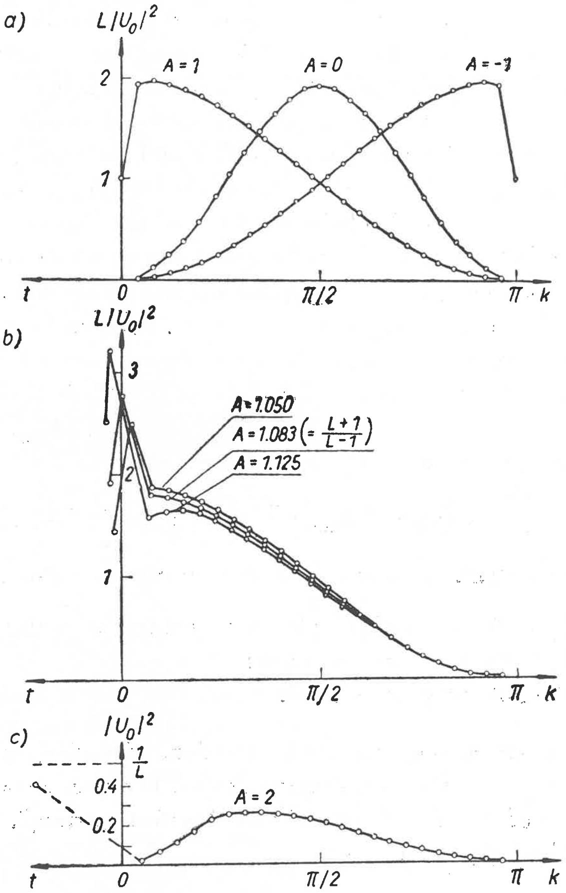

Fig. 7. Spectra of squared amplitudes of spin-wave modes in the surface layer ($l = 0$) corresponding to various values of the surface parameter A ($L = 25$). As A increases, the position of the maximum shifts towards the bottom of the energy band pointing to a weakening of surface spin pinning (*cf.* Eq. (5.4))

The preceding considerations, as well as those of subsection 1, permit the following conclusion: *The surface parameter A has the physical meaning (in both the semi-classical and quantum-theoretical approach) of a measure of the pinning of surface spins.*

Numerous authors take the surface anisotropy energy E_s as a measure of this pinning. By Eq. (5.2), a relation of proportionality exists between E_s and A. Others attribute this role to the value taken by the spin-wave mode amplitude at the film surface. An analysis of Fig. 4 (and 5) convinces us that the surface value of the amplitude can serve for measuring surface spin pinning only in the cases of modes $n = 1$ and $n = 2$, since only these modes present a monotonous dependence of the surface amplitude $|u_{0;L-1}(\tau')|$ on the surface parameter A.

In Part 2 of this paper, we shall calculate the spin wave resonance spectrum and shall show that the majority of experimental results are accessible to an interpretation within the framework of the Surface Inhomogeneity model taking into consideration the dependence of the surface parameter A on various physical factors.

REFERENCES

[1] C. Kittel, *Phys. Rev.*, **110,** 1295 (1958).
[2] M. H. Seavey, P. E. Tannenwald, *Phys. Rev. Letters*, **1,** 168 (1958).
[3] G. Suran, *C. R. Acad. Sci.* Paris, **266,** 998 (1968).
[4] C. F. Kooi, P. E. Wigen, M. R. Shanabarger, J. V. Kerrigan, *J. Appl. Phys.*, **35,** 791 (1964).
[5] R. F. Soohoo, *Magnetic Thin Films*, Harper and Row, N. Y. 1965.
[6] P. Wolf, *Proceedings of the International Symposium*, Göttingen 1965 (p. 329).
[7] T. G. Phillips, H. M. Rosenberg, *Phys. Letters*, **8,** 298 (1964); Proceedings of the Intern. Conf. on Magnetism, Nottingham 1964 (p. 306).
[8] N. Nisenoff, R. Terhune, *J. Appl. Phys.*, **36,** 732 (1965).
[9] E. Schlömann, *J. Appl. Phys.*, **36,** 1193 (1965).
[10] A. M. Portis, *Appl. Phys. Letters*, **2,** 69 (1963).
[11] E. Hirota, *J. Phys. Soc. Japan*, **19,** 1 (1964).
[12] J. T. Davies, *J. Appl. Phys.*, **35,** 804 (1964).
[13] C. F. Kooi, W. R. Holmquist, P. E. Wigen, J. T. Doherty, *J. Phys. Soc. Japan, Suppl.* B1, **17,** 599 (1962).
[14] P. E. Wigen, C. F. Kooi, M. R. Shanabarger, T. D. Rossing, *Phys. Rev. Letters*, **9,** 206 (1962).
[15] P. E. Wigen, C. F. Kooi, M. R. Shanabarger, V. K. Cummings, M. E. Baldwin, *J. Appl. Phys.*, **34,** 1137 (1963).
[16] A. R. Ferchmin, *Acta Phys. Polon.*, **32,** 289 (1967).
[17] V. M. Sokolov, B. A. Tavger, *Radiofizika*, **9,** 308 (1966).
[18] C. W. Searle, A. H. Morrish, R. J. Prosen, *Physica*, **29,** 1219 (1963).
[19] A. Stankoff, *Proceedings of the International Symposium*, Irkutsk 1968 (p. 422), (in Russian).
[20] T. D. Rossing, *J. Appl. Phys.*, **34,** 1133 (1963).
[21] Makoto Okochi, Hiroshi Nosé, *J. Phys. Soc. Japan*, **25,** 1017 (1968).
[22] C. Kittel, C. Herring, *Phys. Rev.*, **77,** 725 (1950).
[23] Z. Frait, E. N. Mitchell, *Proceedings of the Intern. Conf. on Magnetism*, Nottingham 1964 (p. 316).
[24] W. C. Holzer, B. W. Perry, A. M. Portis, *J. Appl. Phys.*, **37,** 1222 (1966).
[25] V. M. Sokolov, B. A. Tavger, *Fiz. Tverdogo Tela*, **10,** 1793 (1968).
[26] V. M. Sokolov, *Fiz. Tverdogo Tela*, **11,** 753 (1969).
[27] H. Puszkarski, *Phys. Status Solidi*, **22,** 355 (1967).
[28] H. Puszkarski, *Acta Phys. Polon.*, **33,** 769 (1968).
[29] M. Nisenoff, R. W. Terhune, *J. Appl. Phys.*, **35,** 806 (1964).
[30] P. Pincus, *Phys. Rev.*, **118,** 658 (1960).
[31] A. R. Ferchmin, *Phys. Letters*, **1,** 281 (1962).
[32] L. Valenta, L. Wojtczak, *Proceedings of the Intern. Conf. on Magnetism*, Nottingham 1964 (p. 830).

[33] L. Valenta, *Czech. J. Phys.*, **7**, 126 (1957).
[34] L. Valenta, *Phys. Status Solidi*, **2**, 112 (1962).
[35] R. J. Jelitto, *Z. Naturforsch.*, **19a**, 1567 (1964).
[36] R. J. Jelitto, *Z. Naturforsch.*, **19a**, 1580 (1964).
[37] S. V. Tyablikov, *Methods of the quantum theory of magnetism*, Moskva 1965 (in Russian).
[38] A. Corciovei, *Phys. Rev.*, **130**, 2223 (1963).
[39] G. G. Low, *Proc. Phys. Soc.*, **A79**, 473 (1962).
[40] L. Valenta, *Czech. J. Phys.*, **B14**, 278 (1964).
[41] H. Puszkarski, *Phys. Letters*, **30A**, 227 (1969).
[42] N. Kurczewski, *Paper for the degree of M. Sci.*, A. Mickiewicz University, 1969.