Surface anisotropy energy in terms of magnetocrystalline anisotropy fields in ferromagnetic semiconductor (Ga,Mn)As thin films

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Surface anisotropy energy density is a key quantity in the description of the dynamics of surface spins in ferromagnetic thin films and is known to result in a ferromagnetic resonance fine structure of a thin film: A multipeak spectrum is observed in an effect known as spin-wave resonance (SWR). The so-called surface pinning parameter, directly correlated with the surface anisotropy energy, plays a fundamental role in determining the character of an SWR spectrum in the theory of SWR based on the surface inhomogeneity model. Here we extend the theory of SWR by introducing a new formula representing the surface pinning parameter as a series of contributions from different anisotropies existing in (Ga,Mn)As thin films. We show how to determine the coefficients in this series by comparing the SWR theory with data obtained in a resonance experiment. The proposed procedure allows one to determine the proportion with which each type of the anisotropy contributes to the total surface anisotropy of (Ga,Mn)As, and, consequently, to assess to what extent the proportions on the surface differ from those in the bulk. The presented considerations can be used for the study of any other thin-film system, since we provide also a general rule that allows one to express the surface pinning parameter of a thin film, as well as its surface anisotropy energy, by the free energy of the considered thin-film system. This enables the interpretation of experimental SWR spectra in terms of the free-energy density of the thin film.

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I. INTRODUCTION

Ferromagnetic semiconductors are in the class of materials that show promise of application in new spin-electronic—or, in short, spintronic—devices using both the charge and spin of the electron. Gallium manganese arsenide, (Ga,Mn)As, is a promising material in this class, hence the recent intensification of studies of its properties. Its magnetic anisotropy is of particular interest for its prospective technological applications. However, the origins of this anisotropy in (Ga,Mn)As have not been entirely elucidated yet, which can be expected to affect the control of its use in spintronic devices. Particularly, the magnetic anisotropy of thin films of gallium manganese arsenide, (Ga,Mn)As, is one of their most interesting properties, since it determines the direction of the sample magnetization, the manipulation of which is of key importance for prospective application of this material in memory devices. For this reason the magnetic anisotropy of (Ga,Mn)As thin films is being intensively investigated by many experimental techniques. These include spin-wave resonance (SWR), a method for studying the surface of (Ga,Mn)As thin films in this respect [1–16]. It is worthy of notice that the main objective of the SWR studies conducted so far in (Ga,Mn)As has been to obtain information on certain *volume* characteristics, such as the value of uniaxial anisotropy [7] or exchange constant [14] in the studied material. This is understandable, as the complex nature of ferromagnetism in dilute semiconductors prompts the search of methods that could provide new information explaining this new (volume) material property. Paradoxically, this leaves the main potential of SWR unexploited, since the chief message of SWR studies provides information on magnetic characteristics of the *surface* (see, e.g., [17]). We propose here to use SWR first of all for probing the surface magnetic anisotropy in (Ga,Mn)As thin films.

The structure of the multipeak SWR spectra observed in thin-film samples strongly depends on the type of inhomogeneity existing in the sample. Two extremely idealized models, the volume inhomogeneity (VI) model and the surface inhomogeneity (SI) model, have been used so far for the description of this dependence. The objective of this paper is to contribute to a present theory of spin-wave resonance based on the SI model adjusted in its touchiest point, namely, the boundary conditions. The model uses the concept of the surface pinning parameter, which describes the freedom of precessing surface spins in relation to that of precessing bulk spins.

The paper is organized as follows. In Sec. II we determine the contribution of the magnetocrystalline anisotropy to the free-energy density of a (Ga,Mn)As thin film; the contribution includes first-, second- and third-order cubic anisotropy terms, first- and second-order perpendicular-to-plane uniaxial anisotropy terms, and two in-plane uniaxial anisotropy energy terms. Next, in Sec. III we demonstrate how specifically (by what formula) each of the above-mentioned

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magnetocrystalline anisotropies contributes to the *surface anisotropy* and the *surface pinning*. In Secs. IV and V we consider, with reference to the experimental data, specific angular configurations of the static magnetic field with respect to the surface of the thin film typically used in SWR studies; of these configurations we indicate those that together provide an optimal basis for experimental determination of the contribution of each anisotropy present in (Ga,Mn)As to the surface anisotropy. Finally, in Sec. VI we formulate some observations based on the comparison of our results with those of the SWR studies of (Ga,Mn)As thin films reported to date.

II. BULK MAGNETOCRYSTALLINE ANISOTROPIES IN (Ga,Mn)As

The interpretation of most experimental studies of ferromagnetic resonance (FMR) in (Ga,Mn)As is based on a phenomenological formula for the free energy of the investigated sample. The main characteristic of the studied system contained in this formula is the angular dependence of the magnetocrystalline anisotropy energies. The free energy of a (Ga,Mn)As sample is expressed as a series of terms related to different symmetries; usually the series is limited to low-order terms related to the cubic and uniaxial symmetries. In the present study we are going to rely on a formula for the free-energy density F in (Ga,Mn)As proposed in Ref. [18]. In this equation the terms of the series are expressed by the coordinates n_x, n_y, and n_z of the unit vector $\hat{\boldsymbol{M}} \equiv \boldsymbol{M}/M$ oriented along the magnetization $\boldsymbol{M}$ of the sample: $n_x = \cos\varphi \sin\vartheta, n_y = \sin\varphi \sin\vartheta$, and $n_z = \cos\vartheta$, where the angles φ and ϑ are measured with respect to the [100] and [001] axes, respectively. The equation reads

$$\begin{aligned} F(\hat{\boldsymbol{M}}) = F_0 &+ K_{c1}\left(n_x^2 n_y^2 + n_x^2 n_z^2 + n_y^2 n_z^2\right) \\ &+ K_{c2}\left(n_x^2 n_y^2 n_z^2\right) \\ &+ K_{c3}\left(n_x^4 n_y^4 + n_x^4 n_z^4 + n_y^4 n_z^4\right) \\ &- K_{[001]_1} n_z^2 - \tfrac{1}{2} K_{[001]_2} n_z^4 \\ &- K_{[100]} n_y^2 - \tfrac{1}{2} K_{[110]} (n_y - n_x)^2, \end{aligned} \tag{2.1}$$

where F_0 is an isotropic term expressed by an angle-independent constant. The next three terms represent an expansion of the cubic anisotropy energy related to the crystal symmetry of the zinc-blende structure of (Ga,Mn)As. The cubic anisotropy is described by terms invariant under permutation of the coordinate indexes x, y, and z. The independent first-, second-, and third-order cubic terms read $K_{c1}(n_x^2 n_y^2 + n_x^2 n_z^2 + n_y^2 n_z^2)$, $K_{c2}(n_x^2 n_y^2 n_z^2)$, and $K_{c3}(n_x^4 n_y^4 + n_x^4 n_z^4 + n_y^4 n_z^4)$, respectively, where K_{c1}, K_{c2}, and K_{c3} are cubic energy density coefficients.

Along with the cubic anisotropy, (Ga,Mn)As has different types of uniaxial anisotropy, originating in lattice strains; these uniaxial anisotropies are described by the last four terms of Eq. (2.1). The terms $-K_{[001]_1} n_z^2$ and $-\frac{1}{2} K_{[001]_2} n_z^4$ refer to the perpendicular-to-plane uniaxial anisotropy energy, which is due to the growth strain induced in the sample by the lattice mismatch between the substrate and the film; $-K_{[001]_1} n_z^2$ refers to the lowest-order anisotropy component, while $-\frac{1}{2} K_{[001]_2} n_z^4$ is the second-order contribution. The last two terms in Eq. (2.1) are contributions to the free energy brought by the in-plane uniaxial anisotropy, originating in strains which occur in the sample along particular in-plane crystal axes. Thus, the term $-K_{[100]} n_y^2$ refers to the uniaxial anisotropy along the *main* crystal axes, since positive $K_{[100]}$ favors an in-plane easy axis aligned close to the [010] axis; the last term, $-\frac{1}{2} K_{[110]}(n_y - n_x)^2$, refers to the uniaxial anisotropy along the *diagonals*, positive $-\frac{1}{2} K_{[110]}$ favoring an easy axis aligned close to the $[1\bar{1}0]$ axis.

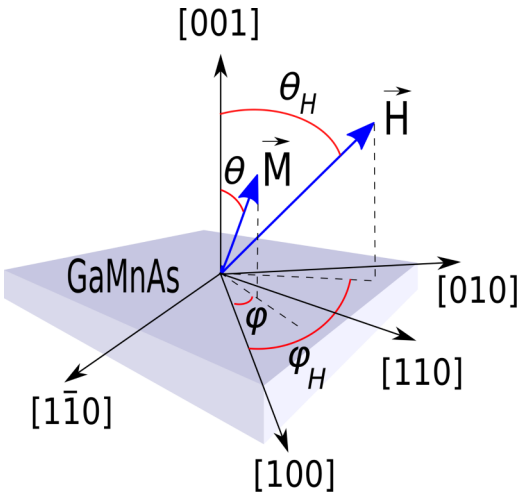

FIG. 1. Coordinate system used in this paper to describe sample configuration. The orientation of the applied magnetic field $\boldsymbol{H}$ is described by angles ϑ_H and φ_H, whereas the equilibrium orientation of the sample magnetization $\boldsymbol{M}$ is given by ϑ and φ.

The model of magnetocrystalline anisotropy developed in Ref. [18] predicts that the anisotropy coefficients in Eq. (2.1) strongly depend on the hole density, Mn local-moment concentration, and temperature, which is confirmed by numerous experiments. Another prediction implied by this model is that the cubic anisotropy coefficient K_{c1} should be proportional to M^4, and the uniaxial anisotropy coefficients $K_{[001]}$, $K_{[100]}$, and $K_{[110]}$ to M^2.

Usually in the literature the free energy is not represented directly by formula (2.1), but by an equivalent equation with anisotropy fields in place of the energy coefficients. The anisotropy fields H_a are related to the energy coefficients K_a by the identity $H_a \equiv 2K_a/M$. Also, the coordinates of the magnetization unit vector $\hat{\boldsymbol{M}}$ in Eq. (2.1) are expressed directly by the angles defining its direction in space, i.e., the angles φ and ϑ measured with respect to the [100] and [001] axes, respectively (see Fig. 1):

$$n_x = \cos\varphi \sin\vartheta; n_y = \sin\varphi \sin\vartheta; n_z = \cos\vartheta. \tag{2.2}$$

The resulting formula for the free energy is

$$\begin{aligned} F(\hat{\boldsymbol{M}}) = F_0 &+ \frac{1}{8} M H_{c1} \sin^2\vartheta (\sin^2\vartheta \sin^2 2\varphi + 4\cos^2\vartheta) \\ &+ \frac{1}{8} M H_{c2} \sin^4\vartheta \cos^2\vartheta \sin^2 2\varphi \\ &+ \frac{1}{32} M H_{c3} [\sin^8\vartheta \sin^4 2\varphi \\ &+ 4\cos^4\vartheta \sin^4\vartheta (3 + \cos 4\varphi)] \end{aligned}$$

$$- \frac{1}{2} M H_{[001]_1} \cos^2\vartheta - \frac{1}{4} M H_{[001]_2} \cos^4\vartheta - \frac{1}{2} M H_{[100]} \sin^2\vartheta \sin^2\varphi - \frac{1}{2} M H_{[110]} \sin^2\vartheta \sin^2\left(\varphi - \frac{\pi}{4}\right). \quad (2.3)$$

The anisotropy fields in Eq. (2.3) have the following sense: H_{c1}, H_{c2}, and H_{c3} are the first-, second-, and third-order cubic anisotropy fields, respectively; $H_{[001]_1}$ and $H_{[001]_2}$ denote the first- and second-order perpendicular uniaxial anisotropy fields, respectively; $H_{[100]}$ and $H_{[110]}$ are independent in-plane uniaxial anisotropy components along the [100] and [110] axes, respectively.

Some information on the properties of these anisotropy fields is available in the literature; the reported properties were either established in experimental studies or predicted theoretically (for an exhaustive review of these data please refer to paper [18]). For example, H_{c1}, $H_{[110]}$, and $H_{[100]}$ are known to be oscillatory functions of the hole density, and there exist critical hole densities where the anisotropy fields change sign; these critical values depend on the Mn local-moment concentration. Moreover, the in-plane uniaxial anisotropy fields oscillate with a longer period than the cubic anisotropy field H_{c1}. It has been established that the extreme values of $H_{[110]}$ are an order of magnitude smaller than the extreme values of $H_{c1} \sim 10^3$ Oe and weakly dependent on the hole density. In general, the amplitude of oscillations decreases with decreasing Mn local-moment concentration [18]. It has also been established [15] that in some (Ga,Mn)As thin film samples the out-of-plane uniaxial anisotropy field $H_{[001]_1}$ varies linearly with the hole/Mn concentration (changing from positive to negative values), however, it is suggested [18] that the extreme values of $H_{[001]_1}$ are of the same order as those of H_{c1}. The higher-order anisotropy terms are believed to be small, or even negligible, in (Ga,Mn)As thin films. For example, H_{c2} and H_{c3} have not been definitely resolved experimentally to date.

Ferromagnetic resonance (FMR) and spin-wave resonance (SWR) studied in thin films are major sources of information on the anisotropy fields in (Ga,Mn)As. However, it has been underestimated in the literature so far that SWR studies can provide information not only on bulk anisotropy fields, but also, and perhaps first of all, on surface anisotropy fields. Note by the way that the surface properties of (Ga,Mn)As have been studied very scarcely, and in light of recent findings [19] they can be of crucial importance for a deeper understanding of the essence of the ferromagnetism of diluted ferromagnetic semiconductors [20–28]. In the present paper we propose a simple model for surface anisotropy characterization independent of the bulk anisotropy. The model describes SWR in (Ga,Mn)As thin films on the basis of the above-specified phenomenological formula expressing the anisotropic part of the free energy in this material. For this purpose we are going to introduce a separate concept of surface free-energy density $F^s(\hat{\boldsymbol{M}}) \equiv F^{\rm surf}(\hat{\boldsymbol{M}})$, a surface characteristic of a ferromagnetic sample expressed by surface anisotropy fields $H_a^s \equiv H_a^{\rm surf} = 2K_a^{\rm surf}/M$, as opposed to the bulk free-energy density $F^b(\hat{\boldsymbol{M}}) \equiv F^{\rm bulk}(\hat{\boldsymbol{M}})$, a bulk characteristic expressed by bulk anisotropy fields $H_a^b \equiv H_a^{\rm bulk} = 2K_a^{\rm bulk}/M$. We will use these quantities in the next section, in which we propose a general formula for the surface pinning parameter, a key quantity in the analysis of SWR spectra; it is precisely the surface pinning parameter that will be expressed by the above-introduced surface and bulk free energies.

III. SURFACE PINNING ENERGY IN TERMS OF FREE-ENERGY DENSITY

In a ferromagnetic thin film with magnetic properties homogeneous along the direction perpendicular to the surface of the sample this homogeneity is only disturbed structurally at the surfaces. Thus, the magnetic properties of such a sample can be described using the surface inhomogeneity (SI) model, which in the molecular field approximation assumes that an effective magnetic field $\boldsymbol{H}_{\rm eff}^{\rm bulk}$ uniform across the sample acts on spins in its bulk, whereas surface spins experience another effective magnetic field, which we will denote as $\boldsymbol{H}_{\rm eff}^{\rm surf}$. The difference between these two fields is referred to as the effective surface anisotropy field [29,30] $\boldsymbol{K}_{\rm eff}^{\rm surf}$:

$$\boldsymbol{K}_{\rm eff}^{\rm surf} \equiv \boldsymbol{H}_{\rm eff}^{\rm surf} - \boldsymbol{H}_{\rm eff}^{\rm bulk}. \quad (3.1)$$

Thus, in relation to bulk spins, surface spins have an additional pinning that is due to the effective surface anisotropy field, which in general consists of surface single-ion and surface exchange anisotropies.

As we have demonstrated in our earlier papers [29,30,33], the precession of surface spins under this additional anisotropy field $\boldsymbol{K}_{\rm eff}^{\rm surf}$ can be fully described by introducing into the corresponding equations of motion a *surface pinning parameter* A, defined

$$A = 1 - \frac{d^2}{D_{\rm ex}} \boldsymbol{K}_{\rm eff}^{\rm surf} \cdot \hat{\boldsymbol{M}}, \quad (3.2)$$

where d is the lattice constant, $D_{\rm ex}$ is the exchange constant, and $\hat{\boldsymbol{M}}$ denotes a unit vector oriented along the magnetization $\boldsymbol{M}$ of the thin film. Another quantity used in the literature for quantitative description of the surface pinning is the surface anisotropy energy $E_{\rm surf}$, which, in contrast to the above-defined *dimensionless* surface pinning parameter, is expressed in energy density units (erg/cm^2). These two approaches are equivalent, since A and $E_{\rm surf}$ are related as follows:

$$E_{\rm surf} = \frac{1}{d} M D_{\rm ex} (A - 1). \quad (3.3)$$

Note that in the surface parameter model the lack of surface anisotropy, $E_{\rm surf} = 0$, implies $A = 1$; by Eq. (3.2) this, in turn, means that the surface anisotropy field is either zero ($\boldsymbol{K}_{\rm eff}^{\rm surf} \equiv 0$), or nonzero but perpendicular to the magnetization of the sample, and thus of no effect on the surface pinning. In this situation, which we will refer to as the *natural surface pinning*, surface spins only feel energetically the natural lack of that part of their neighbors of which they have been deprived by the formation of the surface, and do not experience at all the influence of the surface anisotropy field. This interpretation results from the way in which formula (3.2) is derived in the SI model [29,30]: The breaking of the interaction between the surface spins and their eliminated neighbors is contained in the "unity" in Eq. (3.2), whereas all other surface perturbations

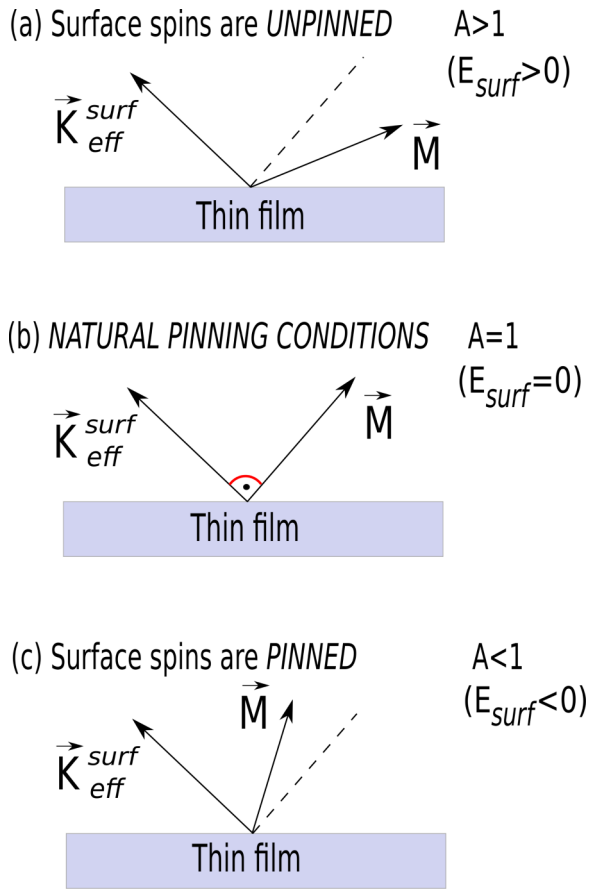

FIG. 2. Schematic representation of three surface spin pinning regimes which prevail in a thin film depending on the configuration of its magnetization $\boldsymbol{M}$ with respect to the effective surface anisotropy field $\boldsymbol{K}_{\text{eff}}^{\text{surf}}$ [see (3.2)]. When aligned as in (b), the surface spins do not feel the anisotropy field and $A = 1$, which corresponds to their natural freedom. In the configurations (a) and (c) the surface spins are unpinned ($A > 1$) and pinned ($A < 1$), respectively, due to the anisotropy field. E_{surf} denotes the surface anisotropy energy density [see (3.3)].

are contained in the anisotropy field $\boldsymbol{K}_{\text{eff}}^{\text{surf}}$. The natural pinning is a reference point with respect to which we consider other surface spin pinning situations (see Fig. 2). When $E_{\text{surf}} > 0$ (i.e., $A > 1$), we say that surface spins and *unpinned*; when $E_{\text{surf}} < 0$ ($A < 1$), the freedom of surface spins is constrained, and therefore we say they are *pinned*. (The boundary conditions corresponding to the natural pinning are particular and require additional discussion, which we have included in Appendix.)

The resonance intensity I of a given spin wave in the SI model with symmetric boundary conditions is described by the equation [29]:

$$I(k) \sim (A-1)^2[u_{\text{surf1}}(k) + u_{\text{surf2}}(k)]^2 \sin^{-4}\left(\frac{k}{2}\right), \quad (3.4)$$

where k is the "wave vector" component perpendicular to the surface of the film describing the character of the standing spin wave assigned to the given resonance excitation; $u_{\text{surf1}}(k)$ and $u_{\text{surf2}}(k)$ are the *surface* amplitudes of the standing spin wave at the bottom and top surfaces, respectively. As we can see, nonzero resonance intensities are only possible when $A \neq 1$, i.e., when the surface spin pinning is different from natural; in other words, a surface anisotropy is necessary for the occurrence of SWR in a thin film. However, even when this condition is fulfilled, antisymmetric modes (in which $u_{\text{surf1}} = -u_{\text{surf2}}$) will not be excited and the resonance spectrum will only consist of symmetric modes. The spectrum of symmetric modes k_n allowed by the boundary conditions results from the following characteristic equation (discussed in detail in Refs. [29,30]):

$$A = \frac{\cos\frac{L+1}{2}k}{\cos\frac{L-1}{2}k}, \quad (3.5)$$

where L is the thickness of the film (in units of lattice constant d). A careful analysis of Eqs. (3.4) and (3.5) leads to the conclusion that the intensities of spin-wave resonance peaks depend *solely* on the *surface* parameter A and the *surface* amplitudes of the corresponding spin-wave modes. Therefore, SWR opens a number of doors, which we will point out in the next sections, to the investigation of some exceptional surface properties.

In this context it is particularly important to relate the surface pinning parameter of a material with the expression for its free energy. Note that the standard method for the description of ferromagnetic resonance in thin films is always based on an appropriately constructed expression for the free energy of the sample. However, its boundary conditions are commonly formulated with the aid of something that we can call a kind of "prosthesis": a variously defined, depending on the context, quantity referred to as the surface anisotropy, by no means related to the basic characteristic of the bulk material, which is its free energy. In the present study we have managed to fill this conceptual gap by finding a relatively simple bridge connecting the surface free energy with the surface boundary, or pinning, conditions.

Let us rewrite Eq. (3.2) taking account of relation (3.1). We obtain

$$A = 1 - \frac{d^2}{M D_{\text{ex}}}\left[\boldsymbol{H}_{\text{eff}}^{\text{surf}} \cdot \boldsymbol{M} - \boldsymbol{H}_{\text{eff}}^{\text{bulk}} \cdot \boldsymbol{M}\right]. \quad (3.6)$$

Note that the terms in square brackets are locally defined free-energy densities:

$$F^{\text{bulk}} = -\boldsymbol{M} \cdot \boldsymbol{H}_{\text{eff}}^{\text{bulk}};$$
$$F^{\text{surf}} = -\boldsymbol{M} \cdot \boldsymbol{H}_{\text{eff}}^{\text{surf}}. \quad (3.7)$$

Thus, the formula for the surface pinning parameter becomes

$$A = 1 + \frac{d^2}{M D_{\text{ex}}}[F^{\text{surf}} - F^{\text{bulk}}]. \quad (3.8)$$

Accordingly, from Eq. (3.3) we obtain the following expression for the surface anisotropy energy:

$$E_{\text{surf}} = d[F^{\text{surf}} - F^{\text{bulk}}]. \quad (3.9)$$

Considering this, we can again rewrite formula (3.8) as

$$A = 1 + \frac{E_{\text{surf}}}{M D_{\text{ex}}/d} = 1 + \frac{E_{\text{surf}}}{2A_{\text{ex}}/d}, \quad (3.10)$$

where $A_{\text{ex}} \equiv \frac{1}{2} M D_{\text{ex}}$ denotes the exchange stiffness parameter. This expression of the surface parameter allows one

to identify [34] our surface anisotropy energy $E_{\rm surf}$ as the surface anisotropy constant figuring in the Rado-Weertman (RW) boundary equation, the earliest boundary condition to have been proposed in the theory of surface magnetism [35]:

$$2A_{\rm ex}\left(\frac{\partial m}{\partial n}\right)_{\rm surf} - E_{\rm surf} m_{\rm surf} = 0, \tag{3.11}$$

where m is the amplitude of the transversal (dynamic) component of the magnetization and n denotes the direction normal to the surface of the film. Since our surface anisotropy energy density $E_{\rm surf}$ is identical with the RW surface anisotropy constant by the above-derived formula (3.10) the RW equation acquires a general character, becoming suitable for studying the configuration effects contained explicitly in the dependence $A(\vartheta,\varphi)$:

$$d\left(\frac{\partial m}{\partial n}\right)_{\rm surf} - (A-1)m_{\rm surf} = 0. \tag{3.12}$$

A major advantage of Eqs. (3.8) and (3.9) we have derived above is that by using expression (2.3) for the free energy in these equations we will obtain full information on the angular configuration dependence of the surface pinning. Note that this information was not provided directly by the initial formula (3.2), expressing the surface pinning parameter by the unspecified effective surface anisotropy field $\boldsymbol{K}_{\rm eff}^{\rm surf}$.

Now, if we use Eq. (2.3) for expressing both the bulk and surface free energies in (3.8), we obtain the following expression for the surface pinning parameter:

$$\begin{aligned} A(\vartheta,\varphi) = {} & 1 + a_{iso} + \frac{1}{8}a_{c1}\sin^2\vartheta(\sin^2\vartheta\sin^2 2\varphi + 4\cos^2\vartheta) \\ & + \frac{1}{8}a_{c2}\sin^4\vartheta\cos^2\vartheta\sin^2 2\varphi \\ & + \frac{1}{32}a_{c3}[\sin^8\vartheta\sin^4 2\varphi \\ & + 4(3+\cos 4\varphi)\cos^4\vartheta\sin^4\vartheta] \\ & - \frac{1}{2}a_{[001]_1}\cos^2\vartheta - \frac{1}{4}a_{[001]_2}\cos^4\vartheta \\ & - \frac{1}{2}a_{[100]}\sin^2\vartheta\sin^2\varphi \\ & - \frac{1}{2}a_{[110]}\sin^2\vartheta\sin^2\left(\varphi - \frac{\pi}{4}\right), \end{aligned} \tag{3.13}$$

where the dimensionless *surface pinning coefficients* a_a are related to the respective *surface anisotropy fields* by

$$\begin{aligned} a_{\rm anis} &= \frac{d^2}{D_{\rm ex}}\left(H_{\rm anis}^{\rm surf} - H_{\rm anis}^{\rm bulk}\right) \\ &= \frac{2d^2}{M D_{\rm ex}}\left(K_{\rm anis}^{\rm surf} - K_{\rm anis}^{\rm bulk}\right). \end{aligned} \tag{3.14}$$

The general idea for using Eq. (3.13) in SWR studies is the following: The experiment allows one to establish the configuration dependence of SWR spectra on either ϑ or φ; this provides the basis for the determination of the dependence of the pinning parameter on both angles, $A = A(\varphi,\vartheta)$. In the next step, by numerical fitting of the experimental data to Eq. (3.13) we can determine the set of surface pinning coefficients that figure in this equation. Finally, in the third step, the surface pinning coefficients can be used for the determination of the corresponding surface anisotropy fields from Eq. (3.14) (the bulk anisotropy fields are assumed to be known from other measurements). Our recent papers [33,36] provide an example in which this procedure is used for studying very special configurations considered in an experimental study by Liu *et al.* [14].

For practical reasons, Eq. (3.13) still needs some adjustment to be adapted to the experimental conditions. Usually in an SWR experiment only one angle is varied (either ϑ or φ), the other one being fixed. Thus, two configurations are considered: the in-plane configuration, in which the static magnetic field lies in the plane of the film ($\vartheta = 90^\circ$) and rotates around the z axis, and the out-of-plane configuration, with the magnetic field rotating in a plane perpendicular to the surface of the film. In the out-of-plane configuration the plane of rotation is defined by the z axis and a straight half-line $\varphi = {\rm const}$; the magnetic field is tilted with respect to the film surface and its orientation can vary from perpendicular ($\vartheta = 0^\circ$) to parallel ($\vartheta = 90^\circ$) to the surface. In the next section each of these two configurations will be discussed in detail separately.

IV. SPECIAL RESONANCE ARRANGEMENTS: IN-PLANE VS OUT-OF-PLANE SWR

In-plane SWR occurs when the static magnetic field $\boldsymbol{H}$ lies in the plane of the film; consequently, the magnetization $\boldsymbol{M}$ of the sample is forced to be oriented in the same plane, i.e., its polar angle $\vartheta = 90^\circ$. In the in-plane configuration formula (3.13) for the surface pinning parameter simplifies to the following equation, in which the azimuthal angle φ is the only angle variable:

$$\begin{aligned} A_{\rm in-plane}(\varphi) = {} & 1 + a_{\rm iso} + \frac{1}{8}a_{c1}\sin^2 2\varphi + \frac{1}{32}a_{c3}\sin^4 2\varphi \\ & - \frac{1}{2}a_{[100]}\sin^2\varphi - \frac{1}{2}a_{[110]}\sin^2\left(\varphi - \frac{\pi}{4}\right). \end{aligned} \tag{4.1}$$

Note that in the considered in-plane configuration the surface pinning parameter does not include contributions from the second-order cubic anisotropy (the term with a_{c2} has vanished) and from the first- and second-order perpendicular uniaxial anisotropy (the terms with $a_{[001]_1}$ and $a_{[001]_2}$ have vanished, too). Only the first- and third-order cubic anisotropies and both in-plane uniaxial anisotropies still contribute to the surface pinning.

Out-of-plane SWR is realized when the azimuthal angle φ is fixed and only the polar angle ϑ varies. For example, in the SWR study of (Ga,Mn)As thin films by Liu *et al.* [14] the azimuthal angle is fixed at $\varphi = -45^\circ$. This means that the applied magnetic field is rotated in the plane perpendicular to the surface of the film defined by two axes: the [001], or z axis, and the $[1\bar{1}0]$ axis, which lies in the film plane. Thus, in this experiment the magnetic field rotates from the direction normal to the surface of the film ($\vartheta = 0$, perpendicular SWR) to the in-plane direction ($\vartheta = 90^\circ$, parallel SWR). For this particular experimental setup the formula for the surface

pinning parameter becomes

$$\begin{aligned} A^{[1\bar{1}0]}_{\text{out}-\text{of}-\text{plane}}(\vartheta) &= 1 + a_{\text{iso}} + \tfrac{1}{8}a_{c1}\sin^2\vartheta(\sin^2\vartheta + 4\cos^2\vartheta) \\ &+ \tfrac{1}{8}a_{c2}\sin^4\vartheta\cos^2\vartheta + \tfrac{1}{32}a_{c3}\sin^4\vartheta(\sin^4\vartheta + 8\cos^4\vartheta) \\ &- \tfrac{1}{2}a_{[001]_1}\cos^2\vartheta - \tfrac{1}{4}a_{[001]_2}\cos^4\vartheta \\ &- \tfrac{1}{4}a_{[100]}\sin^2\vartheta - \tfrac{1}{2}a_{[110]}\sin^2\vartheta. \end{aligned} \tag{4.2}$$

As we can see, in this out-of-plane configuration all the anisotropies present in (Ga,Mn)As contribute to the surface pinning parameter.

For better illustration of the role the chosen azimuthal angle φ plays in the out-of-plane SWR let us consider the situation in which $\varphi = 45^\circ$, i.e., the plane of rotation of the magnetic field $\boldsymbol{H}$ is perpendicular to the surface of the film and determined by the in-plane [110] axis. For $\varphi = 45^\circ$ the last term in (3.13) vanishes; consequently, the surface pinning parameter in this case will differ, by this very term, from the surface pinning parameter determined for $\varphi = -45^\circ$. Thus,

$$A^{[110]}_{\text{out}-\text{of}-\text{plane}}(\vartheta) = A^{[1\bar{1}0]}_{\text{out}-\text{of}-\text{plane}}(\vartheta) + \tfrac{1}{2}a_{[110]}\sin^2\vartheta. \tag{4.3}$$

The vanishing of the term with $a_{[110]}$ in the configuration with $\varphi = 45^\circ$ implies that the uniaxial in-plane anisotropy related to the diagonal [110] axis ("diagonal" in-plane anisotropy) does not contribute to the surface pinning parameter in this configuration; the formula for $A^{[110]}_{\text{out}-\text{of}-\text{plane}}(\vartheta)$ simply *does not include* the corresponding term. Consequently, on the basis of two measurements of the surface pinning parameter, one for $\varphi = 45^\circ$ and one for $\varphi = -45^\circ$, with the same polar angle ϑ, it is possible to determine, from relation (4.3), the *surface* diagonal uniaxial in-plane anisotropy expressed by $a_{[110]}$.

Let us consider yet another out-of-plane configuration, corresponding to the azimuthal angle $\varphi = 0$. In this case Eq. (3.13) becomes

$$\begin{aligned} A^{[100]}_{\text{out}-\text{of}-\text{plane}}(\vartheta) &= 1 + a_{\text{iso}} + \tfrac{1}{2}a_{c1}\sin^2\vartheta\cos^2\vartheta \\ &+ \tfrac{1}{2}a_{c3}\cos^4\vartheta\sin^4\vartheta - \tfrac{1}{2}a_{[001]_1}\cos^2\vartheta \\ &- \tfrac{1}{4}a_{[001]_2}\cos^4\vartheta - \tfrac{1}{4}a_{[110]}\sin^2\vartheta. \end{aligned} \tag{4.4}$$

Thus, in this particular azimuth orientation the surface pinning parameter formula loses two terms, related to the cubic anisotropy a_{c2} and the uniaxial in-plane anisotropy $a_{[100]}$.

From the general formula (3.13) for the surface pinning parameter, which applies to any polar and azimuth orientation, it follows that in (Ga,Mn)As thin films the surface pinning parameter is a sum of isotropic and anisotropic contributions: The isotropic term a_{iso} goes along with three terms related to the cubic anisotropies a_{cub} and four terms related to the uniaxial anisotropies a_{uni}. This gives a total of eight separate energy contributions, all of which should be determined for a full insight into the pinning of surface spins. In the next section we will propose an optimal experimental arrangement for SWR measurements to enable the determination of the complete set of pinning coefficients figuring in Eq. (3.13). Namely, we will specify optimal angles ϑ in particular out-of-plane configurations, as well as optimal angles φ in the in-plane configuration.

V. FURTHER EXPERIMENTAL CONJECTURES

We propose two types of SWR studies for the exploration of the surface anisotropy of (Ga,Mn)As thin films. Experiments of the first type are aimed at the determination of the critical angles, i.e., the angles at which a multipeak SWR spectrum will reduce to a single-peak FMR spectrum. In our earlier papers [29,30] we have demonstrated that in the SI model critical SWR corresponds to the surface pinning parameter value $A_{\text{surf}} \equiv 1$ in such a particular (critical) configuration; as a consequence, and as implied also by Eq. (3.4) cited in the present paper, the resonance intensities of all the spin-wave modes vanish in this case, with the exception of the intensity of the fundamental mode $k = 0$ (see also Appendix). The SWR study by Liu *et al.* [14] shows that at least one critical angle ϑ_c for which $A_{\text{surf}} \equiv 1$ should exist in any out-of-plane configuration. Thus, we can expect to determine experimentally three such angles, one for each of the out-of-plane configurations considered above. Let us denote these critical angles as $\vartheta_{c[1\bar{1}0]}$, $\vartheta_{c[110]}$, and $\vartheta_{c[100]}$. Conditions (4.2), (4.3), and (4.4) lead to the respective equations:

$$A^{[1\bar{1}0]}_{\text{out}-\text{of}-\text{plane}}\left(\vartheta_{c[1\bar{1}0]}\right) = 1, \tag{5.1}$$

$$A^{[110]}_{\text{out}-\text{of}-\text{plane}}\left(\vartheta_{c[110]}\right) = 1, \tag{5.2}$$

$$A^{[100]}_{\text{out}-\text{of}-\text{plane}}\left(\vartheta_{c[100]}\right) = 1. \tag{5.3}$$

The experiment by Liu *et al.* [14] indicates also the existence of two critical angles, φ_{c1} and φ_{c2}, in the in-plane configuration. If these angles can be determined experimentally, then for each of them we can write

$$A_{\text{in}-\text{plane}}(\varphi_{c1}) = 1, \tag{5.4}$$

$$A_{\text{in}-\text{plane}}(\varphi_{c2}) = 1, \tag{5.5}$$

using (4.1) in both equations.

Thus, we have already five equations with eight unknown pinning coefficients. We need three more equations, which will be provided by SWR studies of the second type. In these experiments it is necessary to *determine the surface pinning parameter* from the SWR spectrum measured in a specific angular configuration. (For the idea and details of the method for the conversion of an SWR spectrum into the corresponding value of the surface pinning parameter please refer to papers [30–32].) We propose to measure the SWR spectrum and determine the corresponding surface pinning parameter in *extreme* orientations in each of the three out-of-plane configurations considered above. Thus, for the orientation with $\vartheta = 0$, which we will henceforth refer to as the *perpendicular* configuration, we obtain [see Eq. (3.13)]:

$$A_\perp = 1 + a_{\text{iso}} - \tfrac{1}{2}a_{[001]_1} - \tfrac{1}{4}a_{[001]_2}. \tag{5.6}$$

For the orientations with $\vartheta = \frac{\pi}{2}$, henceforth referred to as the *parallel* configurations, conditions (4.3) and (4.4) imply

$$A^{[110]}_{\parallel} - A^{[1\bar{1}0]}_{\parallel} = \tfrac{1}{2}a_{[110]}, \tag{5.7}$$

$$A^{[100]}_{\parallel} = 1 + a_{iso} - \tfrac{1}{4}a_{[110]}. \tag{5.8}$$

Note that two of all eight pinning coefficients $a_{[110]}$ and $a_{\rm iso}$ can be determined unambiguously from the latter two equations alone. Thus, for the determination of the other six pinning coefficients it is enough to add Eq. (5.6) to the five equations (5.1)–(5.5) obtained previously from critical angle measurements. To sum up, the complete set of pinning coefficients figuring in the general formula (3.13) for the surface pinning parameter can be determined by measuring (1) the SWR spectrum in four special configurations: the perpendicular configuration (along the [001] axis) and three parallel configurations (along the [100], [110], and $[1\bar{1}0]$ axes), and (2) five critical angles: three polar critical angles $\vartheta_{c[100]}$, $\vartheta_{c[110]}$, and $\vartheta_{c[1\bar{1}0]}$, and two azimuthal critical angles φ_{c1} and φ_{c2}.

Now, let us see what interesting information can be acquired from pinning coefficient values determined in this way. For example, let us consider the coefficient $a_{[110]}$, determined from Eq. (5.7). Its experimental value, as established in the SWR study by Liu *et al.* [14], is $a_{[110]} = -0.054$. Now, let us refer to the definition (3.14) of a pinning coefficient, from which it follows that

$$a_{[110]} = \frac{2d^2}{M D_{\rm ex}}\left(K^{\rm surf}_{[110]} - K^{\rm bulk}_{[110]}\right). \tag{5.9}$$

The determined *negative* value of $a_{[110]}$ implies

$$K^{\rm surf}_{[110]} < K^{\rm bulk}_{[110]}, \tag{5.10}$$

which means that the diagonal in-plane uniaxial anisotropy on the surface of the sample is *weaker* than in its bulk. Moreover, if we knew the bulk value of the anisotropy constant, then (knowing the *numerical* value of $a_{[110]}$) from Eq. (5.9) we could estimate also the value of this constant on the surface.

Obviously, the same procedure can be applied to all the other surface anisotropy coefficients determined from SWR spectra taken in (at least) eight angular configurations. However, we suggest to seek first of all the critical angles, since the determination of the surface pinning parameter from the relative intensities of peaks in an SWR spectrum measured in a noncritical angular configuration is subject to some error, which is substantially reduced when the measurement is aimed at a critical angle.

VI. OUTLOOKS

To date, measurements of the surface pinning parameter based on experimental SWR spectra of (Ga,Mn)As thin films in various angular configurations have only been reported by Liu *et al.* [14]. For analyzing the SWR spectra obtained in the out-of-plane configuration the authors of that experimental study use the phenomenological formula for the free energy density of the sample:

$$F^{[1\bar{1}0]}_{\rm out-of-plane}(\boldsymbol{M}) = (2\pi M^2 - K_{2\perp})\cos^2\vartheta - \tfrac{1}{2}K_{4\perp}\cos^4\vartheta - \tfrac{1}{4}K_{4\parallel}\sin^4\vartheta - K_{2\parallel}\sin^2\vartheta; \tag{6.1}$$

note that the term $2\pi M^2\cos^2\vartheta$, related to the shape anisotropy, does not figure in our Eq. (2.3). Now, if we compare Eq. (6.1) with Eq. (2.3) for $\varphi = -45^\circ$, assuming an additive constant $F_0 = -\frac{1}{2}K_{4\parallel}$, we find the following relations between the anisotropy constants figuring in (6.1) and the anisotropy constants we have defined in (2.1):

$$K_{4\parallel} \equiv K_{c1}, \quad K_{[001]_2} \equiv K_{4\perp} - K_{4\parallel}, \tag{6.2}$$

$$K_{2\perp} \equiv K_{[001]_1}, \quad K_{2\parallel} \equiv K_{[110]}. \tag{6.3}$$

This implies that Eq. (6.1) does not include the second- and third-order cubic anisotropy terms K_{c2} and K_{c3}; neither does it contain the uniaxial in-plane anisotropy described by $K_{[100]}$. (Considerations similar to those above, leading to an identical conclusion, can be found in the paper [18].) In our earlier studies [33,36] we analyzed SWR measurements reported by Liu *et al.* [14], assuming, by Eq. (6.1), that the only types of surface anisotropy in a (Ga,Mn)As thin film are those figuring in this formula; however, it turned out that the model of surface anisotropy in (Ga,Mn)As thin films needs to be enhanced by the addition of other types of anisotropy, not yet postulated in Eq. (6.1). We leave this issue for in-depth consideration in another paper, since the procedure of determination of the pinning coefficients that we propose in Sec. V still requires careful modification to be used for analyzing the measurements reported by Liu *et al.* [14].

Before closing, let us remark that the above-discussed SWR studies proposed for the determination of surface anisotropy will actually provide information on more than just the surface, because of the existing certain correlation between surface and bulk properties of a thin film. Specifically, if some type of anisotropy is found in the bulk, the same type can be anticipated on the surface; and vice versa, if an anisotropy of a type not yet observed in the bulk is found on the surface, it should be expected that more thorough studies will reveal it also in the bulk (this property may be called *a surface-bulk anisotropy affinity*). Although in our considerations here we have referred to (Ga,Mn)As thin films magnetically homogeneous throughout the bulk (and therefore described by the surface inhomogeneity model), we believe that the expected correspondence between bulk and surface in terms of magnetocrystalline anisotropy applies as well to *volume-inhomogeneous* (Ga,Mn)As thin films in which SWR is observed. This is the case of the samples studied by Goennewein *et al.* [4,7,8] and Khazen [37], which use the volume inhomogeneity model for the interpretation of their results; we believe that also their SWR spectra bear a significant imprint of the surface anisotropy too. Thus, it can be expected that the surface anisotropy of such samples can be studied also by a method similar to that proposed in the present paper, based on SWR spectra measured in various carefully chosen angular configurations.

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APPENDIX: THE SURFACE INHOMOGENEITY MODEL AND NATURAL PINNING CONDITIONS

The concept of surface pinning is related to the description of the energy status of surface spins, specifically to the degree

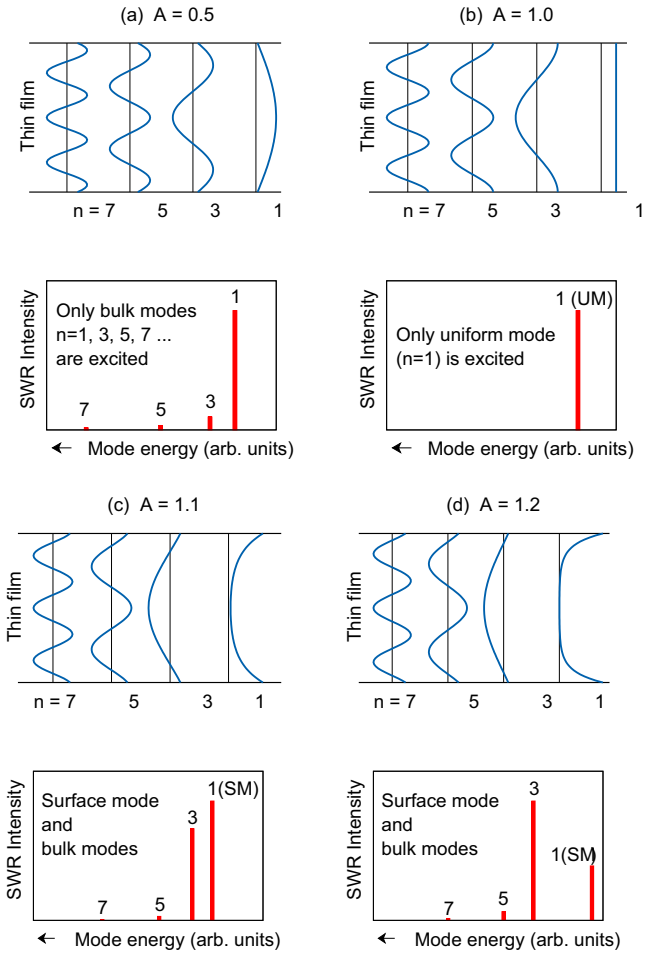

FIG. 3. Profiles of the lowest spin-wave resonance modes (top) and the corresponding SWR spectra (bottom) depicted separately for various values of the surface pinning parameter A (i.e., for various pinning of the surface spins) within the surface inhomogeneity (SI) model. The spectra only exhibit peaks corresponding to symmetric modes of odd number, $n = 1,3,5,7$. The calculations are performed for the case of $L = 75$ (75 layers in the film); UM denotes uniform mode, and SM-surface mode. Two very peculiar effects are observed: first, the multipeak SWR spectrum reduces to a single-peak FMR spectrum at $A = 1$, second, for $A > 1$ the spectrum includes a surface-localized resonance peak.

of freedom of their precession. In a simplified image besides the effective magnetic field present throughout the sample an additional magnetic field $\boldsymbol{K}_{\text{eff}}^{\text{surf}}$, referred to as the effective surface anisotropy field, acts on the surface spins. As we have shown in Refs. [29,30] the boundary conditions to be fulfilled by the precession of the surface spins can be expressed by the surface pinning parameter defined by Eq. (3.2). Note that a complete lack of anisotropy field on the surface corresponds to the surface parameter value one; the freedom of the surface spins in this situation will be referred to as the *natural freedom*. As already we have illustrated in Fig. 2 in the case of nonzero anisotropy field three situations, substantially different from the physical point of view, may occur depending on the angle between the magnetization $\boldsymbol{M}$ and the surface anisotropy field $\boldsymbol{K}_{\text{eff}}^{\text{surf}}$. If the surface spins are aligned perpendicularly to $\boldsymbol{K}_{\text{eff}}^{\text{surf}}$, their freedom remains *natural* ($A = 1$); otherwise, the surface spins are pinned (and $A < 1$) or unpinned (and $A > 1$) for the above-mentioned angle acute or obtuse, respectively.

Note now that in the special case in which the bulk and surface values of the free-energy density are equal [see Eq. (3.8)] the surface pinning parameter is equal to one, which corresponds to the natural pinning. This case is special enough to deserve a separate discussion.

From Eq. (3.5) for $A = 1$ we obtain the following set of allowed wave numbers k of symmetric modes:

$$k = 0, 2\frac{\pi}{L}, 4\frac{\pi}{L}, \ldots, (L-1)\frac{\pi}{L}; \tag{A1}$$

for convenience we have assumed above that L is an odd number.

On the other hand, the surface amplitude of a symmetric mode is expressed by the following equation [29]:

$$u_{\text{surf}}(k) = \sqrt{2}\left(L + \frac{\sin Lk}{\sin k}\right)^{-1/2}\left(\cos\frac{L-1}{2}k\right). \tag{A2}$$

Now, if we substitute Eqs. (3.5) and (A2) into (3.4), we obtain the following expression for the resonance intensity of symmetric modes:

$$I(k) \sim \left(L + \frac{\sin Lk}{\sin k}\right)^{-1}\left(\frac{\sin\frac{1}{2}Lk}{\sin\frac{1}{2}k}k\right)^2, \tag{A3}$$

from which it follows that for all nonzero wave numbers in the set (A1) the resonance intensity is zero, whereas for $k = 0$ (uniform mode) (A3) becomes a 0/0 type undefined expression. However, its limit for $k \to 0$ is nonzero:

$$\lim_{k\to 0} I(k) = L. \tag{A4}$$

Thus, the natural pinning excludes a multipeak resonance excitation, and the SWR spectrum in this case only consists of a single peak corresponding to the excitation of the symmetric uniform mode $k = 0$ (see Fig. 3). In the literature this is referred to as the critical resonance, and the angles ϑ and φ for which this kind of resonance occurs are known as the critical angles (ϑ_c and φ_c , respectively).

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