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SWR Studies of Higher-Order Surface Anisotropy Terms in (Ga,Mn)As Thin Films

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We extend the theory of spin-wave resonance (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. Comparing our theory with the reported experimental studies of SWR in thin films of the ferromagnetic semiconductor (Ga,Mn)As, we find that besides the first-order cubic anisotropy, higher-order cubic anisotropies (in the second and third orders) as well as uniaxial anisotropies (perpendicular in the first and second orders, and in-plane diagonal) occur on the surface of this material. To our best knowledge this is the first report of the existence of higher-order surface anisotropy fields in (Ga,Mn)As thin films.

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1. 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 [1, 2]. Gallium manganese arsenide, (Ga,Mn)As, is a promising material in this class, hence the recent intensification of studies of its properties. 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) [3–22]. 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 [9] or exchange constant [16] in the studied material. Paradoxically, this leaves the main potential of SWR unexploited, since the main message of SWR studies provides information on magnetic characteristics of the *surface* (see e.g. Ref. [23]). We suggest here to use SWR first of all for probing the surface magnetic anisotropy in (Ga,Mn)As thin films.

2. Surface pinning model 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 mean field approximation assumes that an effective magnetic field $\boldsymbol{H}_{eff}^{bulk}$ uniform across the sample acts on spins in its bulk, whereas surface spins experience another effective magnetic field, which we will denote as $\boldsymbol{H}_{eff}^{surf}$. The difference between these two fields is referred to as the effective surface anisotropy field [24, 25] $\boldsymbol{K}_{eff}^{surf}$:

$$\boldsymbol{K}_{eff}^{surf} \equiv \boldsymbol{H}_{eff}^{surf} - \boldsymbol{H}_{eff}^{bulk}. \tag{2.1}$$

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 anisotropies.

As we have demonstrated in our earlier papers [24–26], the precession of surface spins under this additional anisotropy field $\boldsymbol{K}_{eff}^{surf}$ can be fully described by introducing into the corresponding equations of motion a *surface pinning parameter* A, defined:

$$A = 1 - \frac{d^2}{D_{ex}} \boldsymbol{K}_{eff}^{surf} \cdot \hat{\boldsymbol{M}}, \tag{2.2}$$

where d is the lattice constant, D_{ex} is the exchange constant, and $\hat{\boldsymbol{M}}$ denotes a unit vector oriented along the magnetization $\boldsymbol{M}$ of the thin film; the coordinates of the magnetization unit vector $\hat{\boldsymbol{M}}$ 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):

$$n_x = \cos\varphi \sin\vartheta; n_y = \sin\varphi \sin\vartheta; n_z = \cos\vartheta. \tag{2.3}$$

Let us rewrite equation (2.2) taking account of relation (2.1). We obtain:

$$A = 1 - \frac{d^2}{M D_{ex}} \left[\boldsymbol{H}_{eff}^{surf} \cdot \boldsymbol{M} - \boldsymbol{H}_{eff}^{bulk} \cdot \boldsymbol{M} \right]. \tag{2.4}$$

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

$$F^{bulk} = -\boldsymbol{M}\cdot\boldsymbol{H}^{bulk}_{eff}; \quad F^{surf} = -\boldsymbol{M}\cdot\boldsymbol{H}^{surf}_{eff}. \quad (2.5)$$

Thus, the formula for the surface pinning parameter becomes:

$$A = 1 + \frac{d^2}{MD_{ex}}\left[F^{surf} - F^{bulk}\right]. \quad (2.6)$$

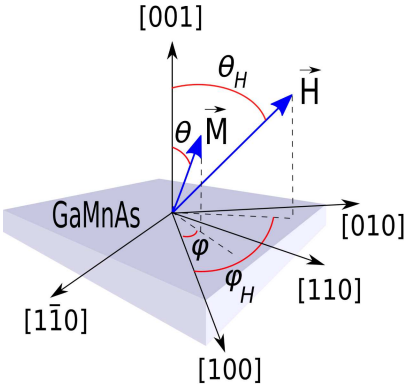

Fig. 1. Coordinate system used in this paper to describe sample configuration. The orientation of the applied magnetic field $\boldsymbol{H}$ is described by angles ϑ_H and φ_H, whereas the equilibrium orientation of the sample magnetization $\boldsymbol{M}$ is given by ϑ and φ.

Now, if we use equation (2.1) of the paper [27] for expressing both the bulk and surface free energies in (2.6), we obtain the following expression for the surface pinning parameter:

$$\begin{aligned} A(\vartheta,\varphi) = 1 &+ a_{iso} + \tfrac{1}{8}a_{c1}\sin^2\vartheta\left(\sin^2\vartheta\sin^2 2\varphi + 4\cos^2\vartheta\right) \\ &+ \tfrac{1}{8}a_{c2}\sin^4\vartheta\cos^2\vartheta\sin^2 2\varphi + \tfrac{1}{32}a_{c3}\left[\sin^8\vartheta\sin^4 2\varphi\right. \\ &\left. + 4\,(3+\cos 4\varphi)\cos^4\vartheta\sin^4\vartheta\right] - \tfrac{1}{2}a_{[001]_1}\cos^2\vartheta \\ &- \tfrac{1}{4}a_{[001]_2}\cos^4\vartheta - \tfrac{1}{2}a_{[100]}\sin^2\vartheta\sin^2\varphi \\ &- \tfrac{1}{2}a_{[110]}\sin^2\vartheta\sin^2(\varphi - \tfrac{\pi}{4}), \end{aligned} \quad (2.7)$$

where the dimensionless *surface pinning coefficients* a_{anis} are related to the respective *surface and bulk anisotropy density coefficients* by:

$$a_{anis} = \frac{2d^2}{MD_{ex}}\left(K^{surf}_{anis} - K^{bulk}_{anis}\right). \quad (2.8)$$

The surface pinning coefficients in equation (2.7) correspond to different anisotropies: a_{c1}, a_{c2} and a_{c3} are related to the first-, second- and third-order cubic anisotropies, respectively; $a_{[001]_1}$ and $a_{[001]_2}$ to the first- and second-order perpendicular uniaxial anisotropies, respectively; $a_{[100]}$ and $a_{[110]}$ to the respective in-plane uniaxial anisotropies along the [100] and [110] axes, respectively; ϑ and φ denote the spherical magnetization angles.

The general idea for using equation (2.7) in SWR studies is the following: The experiment allows 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 equation (2,7) 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 equation (2.8).

3. Determination of surface pinning energies

By calculating the surface pinning parameter values corresponding to each measured SWR spectrum the study by Liu *et al.* [16] opens the door to the exploration of the properties of the surface magnetic anisotropy, which is the prime cause of the observed SWR. The spherical surface pinning model provides a bridge which will allow us to turn the experimental SWR spectra obtained by Liu *et al.* [16] into specific knowledge of the surface magnetic anisotropy in (Ga,Mn)As thin films.

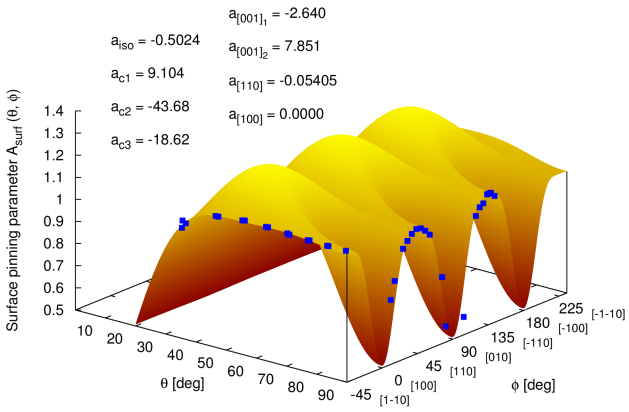

Fig. 2. Fitting of the theoretical pinning hypersurface resulting from our spherical surface pinning model, Eq. (2,7), to the experimental points of Liu *et al.* [16]. The magnetization angle dependence of the surface pinning parameter A_{surf} by Eq. (2,7) fits very well the experimental data with the set of surface pinning coefficient values specified in the graph.

The result of our fitting procedure is presented in Fig. 2, in which, along with the determined pinning coefficients, we show the hypersurface representing the surface pinning parameter versus both ϑ and φ. The theoretical predictions fit surprisingly well the experimental data, and detailed analysis of the shape of the hypersurface depicted in Fig. 2 gives an exceptionally complete insight into the properties of the surface magnetic anisotropy. The fitting yields values of the pinning coefficients figuring in our general formula (2,7). Omitting the isotropic surface pinning term $a_{iso} = -0.5024$, of no importance for our further considerations, the other seven coefficients can be classified in three sets, related to the pinning due to three types of anisotropy, namely, to cubic anisotropy:

$$a_{c1} = 9.104, \quad a_{c2} = -43.68, \quad a_{c3} = -18.62, \quad (3.1)$$

perpendicular uniaxial anisotropy:

$$a_{[001]_1} = -2.640, \quad a_{[001]_2} = 7.851, \tag{3,2}$$

and in-plane uniaxial anisotropy:

$$a_{[110]} = -0.05405, \quad a_{[100]} = 0.000. \tag{3.3}$$

What information on the surface anisotropy can we deduce from these numerical values? Let us recall that, by definition (2.8), each of these coefficients measures the difference between the surface and bulk values of the given anisotropy constant, by the equation:

$$K_{anis}^{surf} - K_{anis}^{bulk} = \frac{MD_{ex}}{2d^2} a_{anis}. \tag{3.4}$$

Unfortunately, we are unable to evaluate quantitatively the difference between the surface and bulk values of the anisotropy constant from this equation, even if we manage to determine the value of the relevant pinning coefficient, mainly because of the lack of reports on the evaluation of the effective lattice constant d in the factor on the right hand side of Eq. (3.4). Still, we can extract some qualitative data, which will, after all, prove valuable in the light of our so far nearly complete lack of knowledge of the surface anisotropy. These qualitative data will be mainly based on the signs of the determined pinning coefficients. Thus, the positive value of a_{c1} implies *strengthening* of the first-order cubic anisotropy on the surface of the (Ga,Mn)As thin film. The negative values of a_{c2} and a_{c3} may imply that the second- and third-order cubic anisotropies are reduced on the surface. Analogically, we will interpret the negative values of $a_{[110]}$ and $a_{[001]_1}$ as an evidence that the respective uniaxial anisotropies (diagonal in-plane and perpendicular in the first order) are reduced on the surface in relation to their bulk values. Somewhat surprising in this context is the positive value of $a_{[001]_2}$, implying that the perpendicular uniaxial anisotropy in the second order is *increased* on the surface. From the zero value of the coefficient $a_{[100]}$ of in-plane anisotropy related to the main axis we conclude that this type of anisotropy is absent both in the bulk and on the surface, since it is hard to believe that, if this anisotropy were present in the bulk, it would remain unchanged on the surface.

Let us now present the results obtained so far in the spherical coordinate system; this will provide a new insight into the nature of surface anisotropy, complementary to that acquired previously by presenting our results in the Cartesian coordinate system. Figure 3 shows the surface pinning parameter $A(\vartheta, \varphi)$ plotted in the spherical coordinate system by Eq. (2,7). The pole of the system, from which the surface parameter value is measured, is in the center of the depicted figure; the indicated crystal axes are draw from this center, and the azimuthal and polar angles φ and ϑ are defined traditionally with respect to the [100] and [001] axes, respectively (see also Fig. 1).

The hypersurface shown in Fig. 3, representing the configuration dependence of the surface pinning in a (Ga,Mn)As thin film, is an exact equivalent of the hypersurface plotted in the Cartesian coordinate system in Fig. 2. Presenting the pinning hypersurface in the spherical coordinate system has an advantage of enabling characterization of the crystal axes in terms of surface pinning. Note that the surface parameter value corresponding to the [100] direction is much below the value $A = 1$ that corresponds to the natural freedom of surface spins. Thus, the [100] axis can be characterized as a *strong* surface pinning axis; obviously, for the same reason, also the [010] axis is a direction of strong surface pinning. By contrast, the [110] axis corresponds to a surface pinning $A > 1$, which means that the freedom of surface spins is greater than natural in this direction. Thus, the [110] axis is a *weak* surface pinning axis. Such pinning characterization of the crystal axes is of practical use, since it allows us to easily assess whether a given axis favors the occurrence of surface modes. This is only a property of a weak pinning axis, as surface modes require that the surface spins have more freedom than in the natural pinning conditions, which implies weak pinning.

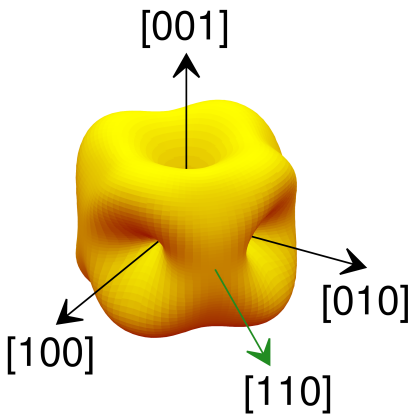

Fig. 3. Configuration dependence of the surface pinning parameter $A(\vartheta, \varphi)$, defined by Eq. (2,7), represented in the spherical coordinate system. The pole of the spherical system is in the center of the depicted figure, and the azimuthal and polar angles φ and ϑ are defined as in Fig. 1. The [100], [010] and [001] crystal axes are *strong* surface pinning axes, and the [110] axis is a *weak* surface pinning axis.

4. Outlooks

Before concluding, 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 a 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 thorougher studies will reveal it also in the bulk (this 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.* [6, 9, 10] and Khazen [28], 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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