Ferromagnetic resonance in thin films – cross-validation analysis of numerical solutions of Smit-Beljers equation. Application to GaMnAs

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The new method of numerical analysis of experimental ferromagnetic resonance (FMR) spectra in thin films is developed and applied to (Ga,Mn)As thin films. Specifically, it starts with the finding of numerical solutions of Smit-Beljers (SB) equation and continues with their subsequent statistical analysis within the cross-validation (CV) approach taken from machine learning techniques. As a result of this treatment we are able to reinterpret the available FMR experimental results in diluted ferromagnetic semiconductor (Ga,Mn)As thin films with resulting determination of magnetocrystalline anisotropy constants. The outcome of CV analysis points that it is necessary to take into account terms describing the bulk cubic anisotropy up to the fourth order to reproduce FMR experimental results for (Ga,Mn)As correctly. This finding contradicts the wide-spread conviction in the literature that only first order cubic anisotropy term is important in this material. We also provide numerical values of these higher order cubic anisotropy constants for (Ga,Mn)As thin films resulting from SB-CV approach.

I. INTRODUCTION: THE EXPERIMENTAL DATA

Gallium manganese arsenide, (Ga,Mn)As, is probably one of the most thoroughly studied diluted ferromagnetic semiconductors. Simultaneous presence of magnetism and conductivity in this material makes it possible to control of both the charge and the spin degrees of freedom of the charge carriers. This creates potential spintronic applications. Another reason for the intense research on (Ga,Mn)As are its remarkable magnetic properties between which magnetic anisotropy plays an important role. It determines, among others, the orientation of magnetization in the absence of an applied magnetic field \( H \). Although its understanding is important for prospective applications such as e.g., memory devices, its origins are far from being fully explained. Magnetocrystalline anisotropy in (Ga,Mn)As, usually described by the single-domain model,\(^2\)\(^3\) is being investigated by various experimental techniques, such as ferromagnetic resonance (FMR) and spin-wave resonance (SWR)\(^4\). Most of these methods have been used to obtain information on anisotropy bulk properties of this material. Recently we have proposed\(^5\) that one can use the SWR to get information on such magnetic properties as surface anisotropy and surface pinning energy of (Ga,Mn)As thin films and their dependence on the orientation of magnetization in the material.

The ferromagnetic resonance spectroscopy has long been a good tool for examining magnetocrystalline anisotropy, see e.g., recent review on FMR in (Ga,Mn)As thin films\(^6\). In FMR experiment, since the equilibrium position of the total magnetic moment \( M \) of the sample does not coincide with the direction of magnetic field \( H \) due to the presence of magnetocrystalline anisotropy, \( M \) precesses around its equilibrium position with a specific (microwave) frequency \( \omega \). By changing the magnetic field \( H \) one hits a resonance field \( H_r \): the precession frequency of \( M \) is equal to the frequency of the spectrometer. The value of the resonance field \( H_r \) strongly depends on its orientation with respect to the examined sample, which is determined by angles \( \vartheta_H \) and \( \varphi_H \), see Fig. 1 due to magnetocrystalline anisotropy.

![FIG. 1: The coordinate system in which an orientation of the applied magnetic field \( H \) is described with respect to the sample in the FMR experiment. The field direction is characterized by angles \( \vartheta_H \) and \( \varphi_H \) measured relative to the sample [001] and [100] axes.](image-url)

This article presents the results of the analysis of bulk magnetocrystalline anisotropy in (Ga,Mn)As based on examination of the uniform mode in SWR resonance in
Fig. 2: Resonance field $H_r$ of the uniform SWR mode as a function of the magnetic field orientation for the out-of-plane configuration (plane $H_r \vartheta_H$) and for the in-plane configuration (plane $H_r \varphi_H$).

(Ga,Mn)As thin film. The motivation to carry out this analysis was two-fold: First – on the basis of examination of surface mode in the same SWR experiment, we have shown\cite{6} that magnetocrystalline surface anisotropy in (Ga,Mn)As thin films contains cubic terms up to third order, which is not commonly found among ferromagnets. We wonder if this is also true for bulk magnetocrystalline anisotropy. Second – it was originally shown\cite{6} that in order to reproduce the experimental dependence of the resonance field on the orientation of the magnetic field with respect to the sample, only the first order term of cubic anisotropy should be taken into account, which, to some extent, is contradictory to the analysis carried out for the surface\cite{6}. In the meantime, numerical tools have been developed that allow a thorough analysis of this problem. That is why we have considered the old problem again.

At the beginning let us recall the angular dependencies of resonance field for the uniform mode in ferromagnetic (Ga,Mn)As thin film\cite{6}. They are shown in Ref. [6] in Fig. 5 for the out-of-plane geometry and in Fig. 6 for the in-plane geometry, respectively. We show them again in Fig. 2 to clearly emphasize the difference between resonance field resulting from the uniaxial anisotropy (plane $H_r \vartheta_H$) and that resulting from the cubic anisotropy (plane $H_r \varphi_H$). We focus on the interpretation of this experiment because the authors very carefully identified resonance from uniform SWR modes and distinguished it from that for surface modes.

II. PHENOMENOLOGICAL FREE ENERGY

The starting point for the interpretation of experimental data of ferromagnetic resonance in (Ga,Mn)As is the phenomenological formula for the free energy of the investigated sample. We assume that there exists a single homogeneous magnetic domain within the sample and that the free energy of unit volume of the sample consists of Zeeman term $F_Z$, demagnetization term $F_D$, and magnetocrystalline anisotropy terms (cubic $F_C$ and uniaxial $F_U$):

$$ F = F_Z + F_D + F_C + F_U. \quad (1) $$

Expressing free energy in terms of fictitious fields one obtains

$$ f(\vartheta_H, \varphi_H, \vartheta, \varphi) = \frac{F}{M} = H_Z + H_D + H_C + H_U, \quad (2) $$

$M$ stands here for the value of homogeneous magnetization. Dependence of fictitious anisotropy fields in Eq. (2) on the direction in space is expressed by unit vectors along the applied magnetic field $\mathbf{H}$ and along the magnetization of the sample $\mathbf{M}$. Their spatial orientation is determined by angles $\vartheta_H$, $\varphi_H$ and $\vartheta$, $\varphi$ with respect to the [001] and [100] axes, see Fig. 1. The unit vectors are given by

$$ \mathbf{H} = [n_x^H, n_y^H, n_z^H] = [\cos \varphi_H \sin \vartheta_H, \sin \varphi_H \sin \vartheta_H, \cos \vartheta_H], \quad (3a) $$

$$ \mathbf{M} = [n_x, n_y, n_z] = [\cos \vartheta \sin \varphi, \sin \varphi \sin \vartheta, \cos \vartheta]. \quad (3b) $$

Zeeman field is given by

$$ H_Z(\vartheta_H, \varphi_H, \vartheta, \varphi) = -H(n_x n_x^H + n_y n_y^H + n_z n_z^H). \quad (4) $$

Demagnetization field of a thin film may be approximated by the expression describing demagnetization field of an infinite plane

$$ H_D(\vartheta) = 2\pi M n_z^2. \quad (5) $$

The field $H_C(\vartheta, \varphi)$ should be invariant under the cubic symmetry transformations. It follows that it is possible to expand it into basis functions with the same symmetry. Typically this expansion is limited to some low-order terms of systematically decreasing basis functions. The way of constructing such basis functions is presented, e.g., in Ref. [7]: they are chosen from all terms of the expansion of the identity $(n_x^2 + n_y^2 + n_z^2)^n = 1 (n = 2, 3...)$ which are invariant under permutation of the indices $x, y,$ and $z$. Expansion up to $n = 7$ is used in what follows:

$$ H_C(\vartheta, \varphi) = H_{c1}(n_x^2 n_y^2 n_z^2 + n_y^2 n_z^2 + n_z^2 n_x^2) + H_{c2}(n_z^2 n_y^2 n_x^2) + H_{c3}(n_x^4 n_y^4 + n_y^4 n_z^4 + n_z^4 n_x^4) + H_{c4}(n_x^4 n_y^2 n_z^2 + n_x^2 n_y^4 n_z^2 + n_x^2 n_z^4 n_y^2) + H_{c5}(n_z^4 n_y^2 n_x^2) + H_{c6}(n_x^6 n_y^6 n_z^2 + n_y^6 n_z^6 n_x^2 + n_z^6 n_x^6 n_y^2). \quad (6) $$

All terms included in the expansion of cubic field $H_C(\vartheta, \varphi)$ are shown in Fig. 3(a)-(f). Let us emphasize that every next term is smaller than the previous one - the expansion [6] is convergent. Let us note, however, that the set of six basis functions used in expansion [6]
The right side of Eq. (2) depends on 15 variables: 
$H_r, \theta_H, \varphi_H, \varphi, H_{c1}, ..., H_{c6}, H_{\text{eff}[001]}, H_{[110]}$. The first three will be considered as independent variables that are measured in the experiment and are shown in Fig. 2. The pairs of angles $(\theta_H, \varphi_H)$ and $(\varphi, \varphi)$ are not independent. The angles determining the equilibrium orientation of $M, \varphi$ and $\varphi$, should minimize free energy. For the set of fixed parameters 
$H_r, \theta_H, \varphi_H, H_{c1}, ..., H_{c6}, H_{\text{eff}[001]}, H_{[110]}$, one finds them from the equilibrium condition

$$\frac{\partial f}{\partial \theta} = 0, \quad \frac{\partial f}{\partial \varphi} = 0. \quad (8)$$

Thus the right-hand side of Eq. (2) really depends on 10 fictitious anisotropy fields $H_{c1}, ..., H_{c6}, H_{\text{eff}[001]}, H_{[110]}$ which we collectively denote by the vector 
$h = (H_{c1}, ..., H_{c6}, H_{\text{eff}[001]}, H_{[110]}). \quad (9)$

The questions should then be asked: how to find anisotropy fields and how many of them are necessary to reproduce the experimental dependence $H_r(\theta_H, \varphi_H)$ well? These questions are answered in the next section.

III. WHICH ANISOTROPY FIELDS ARE IMPORTANT FOR (GA,MN)AS?

The resonance condition (in the case of uniform magnetization) is given by Equations 10 and 11:

$$\left(\frac{\omega}{\gamma}\right)^2 = \frac{1}{\sin^2 \theta} (f_{\theta \theta} f_{\varphi \varphi} - f_{\theta \varphi}^2), \quad (10)$$

where $f_{\theta \varphi} = \frac{\partial f}{\partial \theta \partial \varphi}, \gamma = \frac{g \mu_B}{\hbar}$ with $g$ being the spectroscopic splitting factor, $\mu_B$ the Bohr magneton and $h$ the Planck constant. The 9.46 GHz spectrometer was used in the considered experiment, thus Eq. (10) reads

$$45.6834 = \frac{g^2}{\sin^2 \theta} (f_{\theta \theta} f_{\varphi \varphi} - f_{\theta \varphi}^2), \quad [\text{Oe}^2]. \quad (11)$$

At resonance Eq. (11) should be met for any given direction of $H_r(\theta_H, \varphi_H)$, i.e., in the case under consideration, for all points shown in Fig. 2. Let us treat $g$ and components of the vector $h$ as not known parameters and denote the right-hand side of the Eq. (11) calculated at $i$-th experimental point by $R_i$,

$$R_i(g, h) = \frac{g^2}{\sin^2 \theta} (f_{\theta \theta} f_{\varphi \varphi} - f_{\theta \varphi}^2). \quad (12)$$

Note that free energy derivatives (e.g., $f_{\theta \varphi}$) are calculated at $i$-th the experimental point — for given values $H_r, \theta_H, \varphi_H$.

One should find such values of unknown coefficients $g, h$ that the Eq. (11) is met as accurately as possible for...
TABLE I: Models of cubic magnetocrystalline anisotropy in (Ga,Mn)As for which cross-validation was carried out. In the second column the terms are given included in the expansion of the free energy \( g \) for models C1 - C6. Uniaxial anisotropy for C1 - C6 models does not change — only fields \( H_{c1}^{001} \) and \( H_{110} \) are present.

| Model | Cubic anisotropy | Uniaxial anisotropy |
|-------|------------------|---------------------|
| C1    | \( H_{c1} \)     | \( H_{c1}^{001}, H_{110} \) |
| C2    | \( H_{c1}, H_{c2} \) | \( H_{c1}^{001}, H_{110} \) |
| C3    | \( H_{c3} - H_{c3} \) | \( H_{c1}^{001}, H_{110} \) |
| C4    | \( H_{c4} - H_{c4} \) | \( H_{c1}^{001}, H_{110} \) |
| C5    | \( H_{c5} - H_{c5} \) | \( H_{c1}^{001}, H_{110} \) |
| C6    | \( H_{c6} - H_{c6} \) | \( H_{c1}^{001}, H_{110} \) |

Each experimental point. So the following error function, being the positive square root of the sum of squares of residuals,

\[
E_{RMS}^{N}(g, h) = \sqrt{\frac{1}{N} \sum_{i} \left( R_{i}(g, h) - 45.6834 \right)^{2}}, \quad (13)
\]

should be minimized in 11-dimensional parameter space \((g, h)\). The sum in Eq. (13) runs over all \( N \) experimental points shown in Fig. 2. This least squares approach to finding the unknown parameters represents a specific case of maximum likelihood approach\(^{12,13}\). Actual values of the spectroscopic splitting factor \( g \) and magnetocrystalline anisotropy fields are those for which Eq. (13) has a minimum close to zero.

We have included 10 magnetocrystalline anisotropy fields into the formula for free energy. Now we will check which ones are really essential to describe the experimental results well using a simple cross-validation scheme\(^{12}\). For this purpose anisotropy models are defined in Tables I and II. For example, in the model C3 (third row of Table I) the cubic anisotropy is expanded up to 3rd order, the uniaxial anisotropy along \( z \) axis up to 1st order, the uniaxial anisotropy along \( [110] \) axis up to 1st order and similarly for other models.

The cross-validation, within leave-one-out technique, runs as follows: We divide the \( N \) (= 55) element set of experimental data into two subsets: the training one and the test one. The first one contains \( N - 1 \) elements, the second one — 1 element. One can do it in \( N \) possible ways. Subsequently the \( N \) subsets obtained in this way are used to train, i.e., to determine the values of the unknown parameters \((g, h)\) by minimizing the error function \( E_{RMS}^{N-1}(g, h) \), defined in Eq. (13) for each model under consideration. Simultaneously the error function \( E_{RMS}^{N}(g, h) \) is calculated for one left test point for each model. Note that its value informs us how well we are doing in predicting the values of anisotropy fields for a particular model. After \( N \) minimizations one examines how averages \( \langle E_{RMS}^{N-1} \rangle \) and \( \langle E_{RMS}^{1} \rangle \) depend on the model, i.e., on the order of expansion \( g \) or \( h \). We use the following criterion to assess the quality of the model: the model describes magnetocrystalline anisotropy well if taking into account higher order terms in the expansions \( g \) and \( h \) does not improve its predictive ability given by the average \( \langle E_{RMS} \rangle \).

The procedure described above allowed to find the average values of anisotropy fields, \( g \)-factor and error functions \( \langle E_{RMS}^{N-1} \rangle \) and \( \langle E_{RMS}^{1} \rangle \) for each model after \( N \) minimizations for the sample in the experiment under consideration. They are collected in Tables I and II. The values error functions were calculated for the corresponding field values taken from the Tables III and IV.

TABLE II: Models of uniaxial magnetocrystalline anisotropy in (Ga,Mn)As for which cross-validation was carried out. In the second column the terms are given included in the expansion of the free energy \( g \) for models U1 - U3. Cubic anisotropy for U1 - U3 models does not change — only fields \( H_{c1} - H_{c4} \) are present.

| Model | Uniaxial anisotropy | Cubic anisotropy |
|-------|---------------------|-----------------|
| U1    | \( H_{c1}^{[001]}, H_{110} \) | \( H_{c1} - H_{c4} \) |
| U2    | \( H_{c2}^{[001]}, H_{2[001]}, H_{110} \) | \( H_{c1} - H_{c4} \) |
| U3    | \( H_{c3}^{[001]}, H_{2[001]}, H_{3[001]} H_{110} \) | \( H_{c1} - H_{c4} \) |
TABLE III: Anisotropy fields [Oe] in bulk (Ga,Mn)As related to cubic and uniaxial symmetry and values of $g$-factor calculated for models C1-C6 according to the procedure described in the text. In the last two columns error functions $\langle E_{RMS}^{N-1} \rangle$ and $\langle E_{RMS}^1 \rangle$ are shown.

| Model | $H_{c1}$ | $H_{c2}$ | $H_{c3}$ | $H_{c4}$ | $H_{c5}$ | $H_{c6}$ | $H_{c6}^{eff}$ | $H_{[110]}$ | $g$ | $\langle E_{RMS}^{N-1} \rangle$ | $\langle E_{RMS}^1 \rangle$ |
|-------|---------|---------|---------|---------|---------|---------|----------------|-------------|-----|-------------------------------|------------------|
| C1    | 91.81   | -87.94  | 4765    | 65.53   | 1.978   | 0.95    | 0.69           |             |     |                               |                  |
| C2    | 92.21   | 4774    | 70.72   | 1.979   | 0.88    | 0.68    |                |             |     |                               |                  |
| C3    | 77.06   | -419.0  | 4780    | 61.70   | 1.982   | 0.75    | 0.61           |             |     |                               |                  |
| C4    | 78.07   | -534.1  | 43.93   | 1405    | 66.29   | 1.985   | 0.59           | 0.50        |     |                               |                  |
| C5    | 79.57   | -583.4  | 41.68   | 1790    | 2080    | 4814    | 64.50          | 0.58        | 0.48 |                               |                  |
| C6    | 78.78   | -419.0  | 41.82   | 436.1   | 2841    | 2700    | 63.81          | 0.57        | 0.51 |                               |                  |

Note that definition of cubic anisotropy in Ref. [6] is slightly different. It takes into account tetragonal distortion in (GaMn)As thin films. The value of cubic anisotropy field calculated from this definition should be approximately equal to $2H_{c1}$.

TABLE IV: Anisotropy fields [Oe] in bulk (Ga,Mn)As related to cubic and uniaxial symmetry and values of $g$-factor calculated for models U1-U3 according to the procedure described in the text. In the last two columns error functions $\langle E_{RMS}^{N-1} \rangle$ and $\langle E_{RMS}^1 \rangle$ are shown.

| Model | $H_{c6}^{eff}$ | $H_{2}[001]$ | $H_{3}[001]$ | $H_{[110]}$ | $H_{c1}$ | $H_{c2}$ | $H_{c3}$ | $H_{c4}$ | $g$ | $\langle E_{RMS}^{N-1} \rangle$ | $\langle E_{RMS}^1 \rangle$ |
|-------|----------------|---------------|---------------|-------------|---------|---------|---------|---------|-----|-------------------------------|------------------|
| U1    | 4811           | 66.28         | 78.07         | -534.1      | 43.93   | 1405    | 1.985   | 0.59    | 0.50 |                               |                  |
| U2    | 4779           | 21.83         | 66.08         | 80.12       | -449.3  | 40.76   | 1222    | 1.987   | 0.59 |                               |                  |
| U3    | 4778           | 21.99         | 6310          | 66.13       | 80.15   | -449.3  | 40.59   | 1221    | 1.987 |                               |                  |

roughly constant for the C4 - C6 models. It means that higher order terms used in free energy expansion [2] for C5 and C6 models do not improve their ability to predict cubic magnetocrystalline anisotropy on the basis of experimental data from Fig. 2. Similarly we see that taking into account higher order terms in uniaxial anisotropy expansion for models U2 and U3 does not improve their predictive ability. It follows that the correct description of magnetocrystalline anisotropy in (GaMn)As requires that the free energy expansion should include terms describing cubic anisotropy up to fourth order and that is enough to take into account uniaxial anisotropies up to first order.

One can also see the result of minimization in Fig. 6 in the form of a collapse: for the real minimum of error function at $g^*, h^*$ its values $R_i(g^*, h^*)$ for all experimental points fall onto a line 45.6834. By comparing the scattering of points for C1 model (a) and C4 model (b) we note the important thing: the addition of the higher orders of cubic anisotropy fields improves fitting not only for in-plane experimental points but also for out-of-plane experimental points. This is due to the occurrence of partial mixed derivatives of the free energy in the determinant from Hessian (representing a local curvature of free energy in $\vartheta_H, \varphi_H$ space) in Smit-Beljers equation [10] which we solve numerically[13] for all experimental points simultaneously treating them on equal footing. Therefore, to get as accurate as possible anisotropy field values it is important to measure resonance fields in different geometries.
Fulfilling condition (8) while solving Eq. (13) leads to finding dependences \( \vartheta(\vartheta_H, \varphi_H) \) and \( \varphi(\vartheta_H, \varphi_H) \). for all models from Tables III and IV. They are shown for C4 model in Figs. 7 and 8. Function \( \vartheta(\vartheta_H, \varphi_H) \) for a given \( \varphi_H \) it always is a concave function. For angle \( \varphi_H = 45^\circ \) and \( 135^\circ \) we see a ripple, which is the result of the presence of cubic symmetry. Note also, that function \( \varphi(\vartheta_H, \varphi_H) \) for a given \( \vartheta_H \) is for all \( \vartheta_H \) a linear function \( \varphi \propto \varphi_H \) (does not depend on \( \vartheta_H \)).

Let us summarize this section by stating that for the correct description of magnetocrystalline anisotropy in (GaMn)As, the free energy expansion should include terms describing cubic anisotropy up to fourth order and that is enough to take into account uniaxial anisotropies up to first order.

IV. BACK TO THE EXPERIMENT: WHAT IS THE EFFECT OF INCORPORATING OF ANISOTROPY FIELDS OF HIGHER ORDERS?

Let us now examine the dependence of the resonance field \( H_r \) on \( \vartheta_H \) and \( \varphi_H \). The problem can be stated in the following way: Given the values of \( H_r \) on the boundary of the box presented in Fig. 2 determine the resonance field inside the box. One finds a solution in two stages. First, one determines the anisotropy fields, for which Eq. (11) is satisfied on the boundary of the box. This stage has been described in the Section III. Second, to get the resonance field for each \( \vartheta_H \) and each \( \varphi_H \) one should solve Eq. (11) numerically for the anisotropy fields determined in the first stage (collected in Table III) with condition (8) met for each tentative point obtained during the numerical solving procedure. In Figs. 9 and 10 one can see dependencies \( H_r(\vartheta_H, -45^\circ) \), i.e. for out-of-plane geometry and \( H_r(90^\circ, \varphi_H) \) — for in-plane geometry, respectively. Uniaxial anisotropy is the most visible for out-of-plane geometry (Fig. 9) and although the use of higher order cubic terms does improve the agreement of the calculated \( H_r \)-values with the experimental data, this improvement is not particularly visible in the scale of Fig. 9 because the cubic anisotropy is much smaller than the uniaxial one. The improvement, however, can be seen for in-plane geometry: the use of higher order terms in of cubic anisotropy expansion given by Eq. (6) becomes necessary to describe dependence \( H_r(90^\circ, \varphi_H) \) more precisely.

Spatial dependence of the resonance field on angles \( \vartheta_H \) and \( \varphi_H \) is shown in Fig. 11. For small angle \( \vartheta_H \) we see resonance field whose source is mainly uniaxial [001] anisotropy (with two-fold symmetry), whereas for angle \( \vartheta_H \approx 90^\circ \) the resonance field with four fold sym-
The values of fictitious anisotropy fields are important in that they allow to reproduce the spatial dependence of the energy of the sample in a magnetic field due to magnetocrystalline anisotropy and to find easy and hard axes. To find this spatial dependence one needs to know the saturation magnetization. Then the magnetic anisotropy constants can be easily expressed by corresponding anisotropy fields, see e.g., very clearly written Ref. [15]. We have found the saturation magnetization of the considered sample: it amounts $M_s = 30.5$ emu/cm$^3$, which is a typical value for (Ga,Mn)As containing a few percent of Mn atoms.

Returning to the Eq. (6) and multiplying it by $M_s$ one obtains the spatial distribution of energy $F_C(\theta, \varphi)$ stored in bulk (Ga,Mn)As and related to its cubic magnetocrystalline anisotropy for the C4 model

$$F_C(\theta, \varphi) = M_s H_{c1}(n_x^2 n_y^2 + n_y^2 n_z^2 + n_z^2 n_x^2) +$$

$$M_s H_{c2}(n_x^2 n_y^2 n_z^2) +$$

$$M_s H_{c3}(n_x^4 n_y^4 + n_y^4 n_z^4 + n_z^4 n_x^4) +$$

$$M_s H_{c4}(n_x^4 n_y^4 n_z^2 + n_y^4 n_z^4 n_x^2 + n_z^4 n_x^4 n_y^2) =$$

$$K_{c1}(n_x^2 n_y^2 + n_y^2 n_z^2 + n_z^2 n_x^2) +$$

$$K_{c2}(n_x^2 n_y^2 n_z^2) +$$

$$K_{c3}(n_x^4 n_y^4 + n_y^4 n_z^4 + n_z^4 n_x^4) +$$

$$K_{c4}(n_x^4 n_y^4 n_z^2 + n_y^4 n_z^4 n_x^2 + n_z^4 n_x^4 n_y^2),$$

and similarly for the C1 model. $K_{c1} - K_{c4}$ stand in Eq. (14) for cubic anisotropy constants. Taking the numerical values of anisotropy fields $H_{c1} - H_{c4}$ from Table III one obtains the numerical values of anisotropy constants for C1 and C4 models – they are collected in Table V. Let us note that the values of first order cubic anisotropy for (Ga,Mn)As are several dozen to several hundred times smaller than the corresponding values for such ferromagnets as Ni or Fe. Perhaps this is why anisotropies of higher orders become visible in resonance experiments only for weak ferromagnets.

To assess the accuracy of the present method we used the bootstrap method to evaluate errors for C1 and C4 models. To do this we assumed that the error probability distribution of experimental results was normal, and
FIG. 13: Spatial dependence of cubic magnetocrystalline energy in spherical coordinate systems for C1 (a) and C4 (b) models.

FIG. 14: Spatial dependence of the difference of cubic magnetocrystalline energy between C1 and C4 models in spherical coordinate systems.

V. SUMMARY AND OUTLOOKS

The article presents how to determine bulk magnetocrystalline anisotropy in (Ga,Mn)As thin film by numerical solution of the Smit-Beljers equation for all data collected in one FMR experiment, i.e., for different spatial orientations of the magnetic field with respect to the sample, on equal footing. To avoid essential drawbacks of fitting procedures (lack of information which fitted constants are relevant and possibility of overfitting) by finding anisotropy constants we cross-validated the numerical solutions of Smit-Beljers equation for six models (C1-C6). The results of this cross-validation, i.e., the values of the function $\langle E_{RMS}^1 \rangle$ displaying predictive ability for models C1-C6 point that it is necessary to expand bulk cubic anisotropy up to the fourth order to reproduce spatial dependence of the resonance field correctly — that is increasing the order of expansion of anisotropy does not change predictive ability of the model under consideration. Such cubic anisotropy (up to fourth order) is visible in the resonant experiment. It means that the models of first order cubic anisotropy applied so far to (Ga,Mn)As overestimated the value of this anisotropy. Let us stress that this description of the bulk anisotropy is consistent with the presented earlier description of the surface anisotropy (both descriptions require higher order expansion of cubic anisotropy). We also have shown that FMR data allow one to find the spectroscopic splitting factor with high accuracy. We intend to confirm the usefulness of this new approach by applying it to other available resonance experiments in the near future.

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TABLE V: Cubic anisotropy fields ($H_c$) in Gaussian units [Oe] and SI units [kA/m] and cubic anisotropy constants ($K_c$) in [erg/cm$^3$] and [J/m$^3$] for bulk (Ga,Mn)As calculated for models C1 and C4.

|       | $H_{c1}$   | $H_{c2}$   | $H_{c3}$   | $H_{c4}$   | $K_{c1}$   | $K_{c2}$   | $K_{c3}$   | $K_{c4}$   |
|-------|------------|------------|------------|------------|------------|------------|------------|------------|
| Model C1 |            |            |            |            |            |            |            |            |
| Gaussian | 91.81 ± 55 |            |            |            |            |            |            |            |
| SI      | 7.306 ± 0.044 |            |            |            |            |            |            |            |
| Model C4 |            |            |            |            |            |            |            |            |
| Gaussian | 78.07 ± 40 | -534 ± 28  | 43.9 ± 1.2 | 1410 ± 70  | 2381 ± 13  | -1628 ± 86 | 1330 ± 37  | 4290 ± 220 |
| SI      | 6.213 ± 0.032 | -42.5 ± 2.3 | 3.493 ± 0.096 | 112.2 ± 5.6 | 238.1 ± 1.3 | -1628 ± 86 | 133.0 ± 3.7 | 4290 ± 22 |

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Details of this calculation will be published in a separate paper.