Determination of the complete set of second-order magnetoelastic coupling constants on epitaxial films

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(Received 26 February 2002; published 5 June 2002)

We propose a combination of cantilever-bending-beam experiments on cubic epitaxial films required to measure the coupling constants describing all possible second-order magnetoelastic effects. In addition to previous theoretical predictions of some of these quantities for elementary $3d$-transition metals, the missing couple of the constants is calculated *ab initio*. The results indicate a possibility for the third-order effects in bcc Fe.

DOI: 10.1103/PhysRevB.65.212410 PACS number(s): 75.80.+q, 71.15.Mb

The available results of cantilever-bending-beam experiments clearly imply the presence of nonlinear magnetoelastic (ME) effects in epitaxial films,[1–6] which are cubic in the unstrained state (in the following denoted as cubic epitaxial films). These results, originally described in terms of effective first-order ME coupling constants, have been interpreted[7,8] in terms of intrinsic second-order ME constants by applying the nonlinear phenomenological theory of magnetoelasticity.[9] The key quantity of this interpretation is the part of the total-energy density, which depends on the magnetization direction $\boldsymbol{\alpha}=(\alpha_1,\alpha_2,\alpha_3)$ and strain-tensor components ϵ_{ij}. This contribution $e=e_{\mathrm{el}}+e_{\mathrm{me}}^{(1)}+e_{\mathrm{me}}^{(2)}$ consists of the elastic energy density with the elastic constants C_{ij}

$$e_{\mathrm{el}}=\frac{1}{2}C_{11}(\epsilon_{11}^2+\epsilon_{22}^2+\epsilon_{33}^2)+2C_{44}(\epsilon_{12}^2+\mathrm{cycl})+C_{12}(\epsilon_{11}\epsilon_{22}+\mathrm{cycl}), \tag{1}$$

the first-order ME energy density with the constants B_1, B_2

$$e_{\mathrm{me}}^{(1)}=B_1(\epsilon_{11}\alpha_1^2+\epsilon_{22}\alpha_2^2+\epsilon_{33}\alpha_3^2)+2B_2(\epsilon_{12}\alpha_1\alpha_2+\epsilon_{23}\alpha_2\alpha_3+\epsilon_{31}\alpha_3\alpha_1), \tag{2}$$

and the second-order ME energy density

$$\begin{aligned}e_{\mathrm{me}}^{(2)}=&\frac{1}{2}(B_1+m_1^{\gamma,2})(\epsilon_{11}^2\alpha_1^2+\epsilon_{22}^2\alpha_2^2+\epsilon_{33}^2\alpha_3^2)\\&+\frac{1}{2}m_2^{\gamma,2}(\epsilon_{11}\epsilon_{22}\alpha_3^2+\mathrm{cycl})+m_1^{\epsilon,2}(\epsilon_{11}\epsilon_{23}\alpha_2\alpha_3+\mathrm{cycl})\\&+(B_2+m_2^{\epsilon,2})[(\epsilon_{11}+\epsilon_{22})\epsilon_{12}\alpha_1\alpha_2+\mathrm{cycl}]\\&+\frac{1}{2}(m_3^{\gamma,2}-B_1)(\epsilon_{12}^2\alpha_3^2+\mathrm{cycl})\\&+(B_2+m_3^{\epsilon,2})(\epsilon_{12}\epsilon_{23}\alpha_1\alpha_3+\mathrm{cycl}),\end{aligned} \tag{3}$$

where $m_i^{\gamma,2}$ and $m_i^{\epsilon,2}$ are intrinsic second-order ME coupling constants. The expression $e_{\mathrm{me}}^{(2)}$ is just an approximation for the second-order ME energy density up to the second order in harmonic polynomials of α_i and therefore the first-order constants B_1 and B_2 appearing in the phenomenological expression (2) are identical to the first-order coupling constants $b^{\gamma,2}$, $b^{\epsilon,2}$ from the phenomenological theory based on symmetrized strain variables.[9]

Three different types of the second-order ME effects follow from Eq. (3): the pure tensile-strain related, described by the diagonal strain-tensor components ϵ_{ii}; the mixed-strain related, described by the diagonal as well as the off-diagonal components $\epsilon_{ij}, i\neq j$; and the pure shear-strain related, described just by the off-diagonal components. Each effect corresponds to a different pair of coupling constants: $m_1^{\gamma,2}, m_2^{\gamma,2}$ for the tensile strain, $m_1^{\epsilon,2}, m_2^{\epsilon,2}$ for the mixed strain, and $m_3^{\gamma,2}, m_3^{\epsilon,2}$ for the shear strain. So far, just the tensile-strain[2–4,6] and mixed-strain[3,5] effects have been investigated in cantilever-bending-beam experiments on cubic epitaxial films. In these experiments the nonlinear magnetoelastic effects result in the existence of effective magnetoelastic constants B_1^{eff} and B_2^{eff}, which depend linearly on the epitaxial strain. Within the phenomenological magnetoelastic theory these effective constants could be represented[7,8] by linear combinations of $B_1, m_1^{\gamma,2}, m_2^{\gamma,2}$ and $B_2, m_1^{\epsilon,2}, m_2^{\epsilon,2}$, respectively. The same theory provides the background to determine $B_1, m_1^{\gamma,2}$, and $m_2^{\gamma,2}$ separately by measuring simultaneously the magnetostrictive stress in a cantilever-bending-beam experiment and the strain-induced magnetic anisotropy energy.[7] In the present paper we propose a set of cantilever-bending-beam experiments on epitaxial films to determine B_1, B_2, and all six second-order constants appearing in Eq. (3) without additional experiments on the strain-induced magnetic anisotropy. So far, these six constants have been measured only for the case of fcc Ni by bulk ultrasound-pulse echo experiments,[9] but the experimental errors are huge, see Table I, most likely due to the very small available strains in bulk materials. We hope that the proposed experiments on epitaxial films, where the strains are much larger, will provide more reliable data.

The coupling constants B_1, B_2, $m_1^{\gamma,2}$, $m_2^{\gamma,2}$, $m_1^{\epsilon,2}$, and $m_2^{\epsilon,2}$ have been determined also by the *ab initio* electron theory[8,10] for bcc Fe, fcc Co, fcc Ni, and LI_2-Ni_3Fe, and in the present paper the two remaining constants $m_3^{\gamma,2}$ and $m_3^{\epsilon,2}$ are also calculated *ab initio*.

TABLE I. Theoretical values of B_2 and the pure shear-strain-related second-order ME coupling constants in MJ/m^3 using LSDA and GGA. The last column represents the measured values of B_2, $m_3^{\gamma,2}$, and $m_3^{\epsilon,2}$ together with the experimental errors for fcc Ni from Refs. 1 and 9.

	bcc Fe		fcc Co		fcc Ni		
	LSDA	GGA	LSDA	GGA	LSDA	GGA	Expt.
B_2	-7.0	-3.9	3.0	4.5	16.9	11.1	10
$m_3^{\gamma,2}$	-10.9	-462.8	759.1	861.9	-2.3	108.5	210 ± 302
$m_3^{\epsilon,2}$	-77.4	-868.3	795.9	1681.1	387.7	96.3	46 ± 28

I. A COMPLETE SET OF CANTILEVER-BENDING-BEAM EXPERIMENTS

For an ultrathin magnetic film grown epitaxially on a substrate the average epitaxial strain $\epsilon_0(d)$ in the film is determined[1] by the lattice mismatch between the film and the substrate and by the film thickness d. On changing the direction of the magnetization in the film, the magnetoelastic effects tend to change the lateral extensions of the material. This is not possible due to the bond to the substrate, and a change in the magnetostrictive stress $\boldsymbol{\sigma}^{\mathrm{m}}$ appears instead. The result is a bending of the film and the substrate which is used in a cantilever-bending-beam-experiment monitoring the change in magnetostrictive stress $\sigma_{ll}^{\mathrm{m}} \equiv \sigma_l^{\mathrm{m}}$ along the long cantilever axis $\boldsymbol{l}$. Three different coordinate systems are required to calculate this change (Fig. 1).

(a) The internal frame $\mathbf{r}=(x,y,z)$ with axes parallel to the cubic axes.

(b) The frame $\mathbf{r}'=(x',y',z')$ describing the geometry of the film, where the (x',y') plane coincides with the film plane and the z' axis is parallel to the film normal.

(c) The laboratory frame $\mathbf{r}''=(x'',y'',z''=z')$ with the cantilever axis $\boldsymbol{l}$ parallel to either x'' or y'' axis.

The various frames are interrelated by $r_i=a_{ik}r_k'$ and $r_i'=b_{ik}r_k''$, the direction cosines of the magnetization, α_i, α_i', and α_i'' with $i=1,2,3$ are interrelated in the same way and the tensor components transform like $\epsilon_{ij}=a_{ik}a_{jl}\epsilon_{kl}'$ and $\epsilon_{kl}'=b_{km}b_{ln}\epsilon_{mn}''$, where the a_{ik} and b_{ik} are the elements of the well-known rotation matrices between two Cartesian frames, respectively. The in-plane strain components ϵ_{11}' and ϵ_{22}' are assumed to be equal to the average epitaxial strain ϵ_0, while the perpendicular strain follows from $\partial e/\partial\epsilon_{33}'=0$. The change of the magnetostrictive stress upon rotation of the magnetization from the orientation $\boldsymbol{\alpha}_1'$ to the orientation $\boldsymbol{\alpha}_2'$ is then calculated from

$$\Delta\sigma_l^{\mathrm{m}}\Big|_{\boldsymbol{\alpha}_1'}^{\boldsymbol{\alpha}_2'}=\frac{\partial e}{\partial\epsilon_{ll}''}\Bigg|_{\boldsymbol{\alpha}_1'}^{\boldsymbol{\alpha}_2'}=\frac{\partial e}{\partial\epsilon_{ij}}\frac{\partial\epsilon_{ij}}{\partial\epsilon_{kl}'}\frac{\partial\epsilon_{kl}'}{\partial\epsilon_{ll}''}\Bigg|_{\boldsymbol{\alpha}_1'}^{\boldsymbol{\alpha}_2'}. \tag{4}$$

To simplify the final equations we assume that the absolute values of the second-order ME coupling constants are much smaller than the elastic constants, which is consistent with the results of the *ab initio* calculations. For instance, in Fe the elastic constants are of the order of 10^5 MJ/m^3 whereas the second-order constants are of the order of 10^2 MJ/m^3.

In the following, we define one example of a complete set of cantilever-bending-beam experiments for (001) and (110) films, which requires rotations of the magnetization direction in and out of the film plane. Alternative complete sets exist, among them also sets involving additional film geometries which require only respective in-plane rotations. This may be important if it turns out to be inconvenient to rotate the magnetization out of the plane in the experiment. We, therefore, plan to derive a general expression for the change of the magnetostrictive stress for any kind of rotation of the magnetization direction, for arbitrary surface orientation and for arbitrary orientation of the cantilever axis.

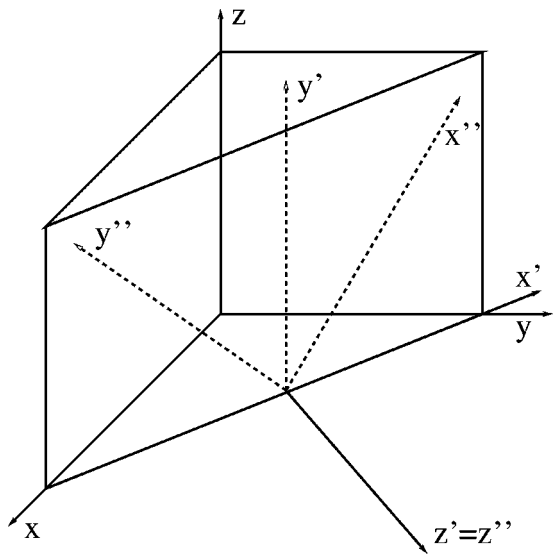

FIG. 1. The coordinate systems describing the geometry of the considered experiments.

A. Measurements on a (001) film for which all three coordinate frames coincide

$$\Delta\sigma_{(100)}^{\mathrm{m}}\Big|_{(010)}^{(100)}=B_1+D_{\mathrm{I}}^{\mathrm{eff}}\epsilon_0, \tag{5}$$

$$D_{\mathrm{I}}^{\mathrm{eff}}=B_1+m_1^{\gamma,2}-\frac{1}{2}m_2^{\gamma,2}\frac{\epsilon_{33}'}{\epsilon_0}, \tag{6}$$

$$\epsilon_{33}'=-2\frac{C_{12}}{C_{11}}\epsilon_0, \tag{7}$$

$$\Delta\sigma_{(100)}^{\mathrm{m}}\Big|_{(001)}^{(100)}=B_1+D_{\mathrm{II}}^{\mathrm{eff}}\epsilon_0, \tag{8}$$

$$D_{\mathrm{II}}^{\mathrm{eff}}=B_1+m_1^{\gamma,2}-\frac{1}{2}m_2^{\gamma,2}. \tag{9}$$

Measuring these two quantities as functions of the average film strain $\epsilon_0(d)$ enables one to separate $B_1, m_1^{\gamma,2}$ and $m_2^{\gamma,2}$. It should be noted that according to our former *ab initio* calculations, $|B_1|$ is often considerably smaller than $|m_1^{\gamma,2}|$, $|m_2^{\gamma,2}|$, and therefore B_1 has been neglected in the equation given formerly[7,10] for $D_{\mathrm{I}}^{\mathrm{eff}}$.

B. Measurements on (110) films for the case that the film frame (x',y',z') coincides with the laboratory frame (x'',y'',z'')

$$\Delta\sigma^{\mathrm{m}}_{(100)'}|^{(100)'}_{(001)'}=B_2+D_{\mathrm{III}}^{\mathrm{eff}}\epsilon_0, \tag{10}$$

$$D_{\mathrm{III}}^{\mathrm{eff}}=B_2+m_2^{\epsilon,2}+\frac{1}{2}m_1^{\epsilon,2}, \tag{11}$$

$$\Delta\sigma^{\mathrm{m}}_{(100)'}|^{(001)'}_{(010)'}=\frac{1}{2}(B_1-B_2)+D_{\mathrm{IV}}^{\mathrm{eff}}\epsilon_0, \tag{12}$$

$$D_{\mathrm{IV}}^{\mathrm{eff}}=\frac{1}{4}(B_1+m_1^{\gamma,2})\frac{\epsilon_0+\epsilon'_{33}}{\epsilon_0}-\frac{1}{4}m_2^{\gamma,2}\frac{\epsilon'_{33}}{\epsilon_0}$$
$$-\frac{1}{4}(m_3^{\gamma,2}-B_1)\frac{\epsilon_0-\epsilon'_{33}}{\epsilon_0}-\frac{1}{2}\left(B_2+m_2^{\epsilon,2}+\frac{1}{2}m_1^{\epsilon,2}\right), \tag{13}$$

$$\epsilon'_{33}=-\frac{C_{11}-2C_{44}+3C_{12}}{C_{11}+2C_{44}+C_{12}}\epsilon_0, \tag{14}$$

$$\Delta\sigma^{\mathrm{m}}_{(010)'}|^{(100)'}_{(001)'}=D_{\mathrm{V}}^{\mathrm{eff}}\epsilon_0, \tag{15}$$

$$D_{\mathrm{V}}^{\mathrm{eff}}=\frac{1}{2}m_1^{\epsilon,2}\frac{\epsilon_0-\epsilon'_{33}}{\epsilon_0}. \tag{16}$$

Measuring these three quantities as functions of $\epsilon_0(d)$ and using the already obtained results for $B_1, m_1^{\gamma,2}$ and $m_2^{\gamma,2}$ yield separately $B_2, m_3^{\gamma,2}, m_1^{\epsilon,2}$ and $m_2^{\epsilon,2}$.

C. Measurement on a (110) film for the case that the (x'',y'') plane of the laboratory frame is rotated against the (x',y') plane by 45° aligning the x'' axis along the diagonal in the (x',y') plane, $(100)''=(110)'$

$$\Delta\sigma^{\mathrm{m}}_{(100)''}|^{(1\bar{1}0)'}_{(110)'}=B_2+D_{\mathrm{VI}}^{\mathrm{eff}}\epsilon_0, \tag{17}$$

$$D_{\mathrm{VI}}^{\mathrm{eff}}=\frac{1}{4}m_1^{\epsilon,2}\frac{\epsilon_0+\epsilon'_{33}}{\epsilon_0}+\frac{1}{4}(B_2+m_2^{\epsilon,2})\frac{3\epsilon_0+\epsilon'_{33}}{\epsilon_0}$$
$$+\frac{1}{4}(B_2+m_3^{\epsilon,2})\frac{\epsilon_0-\epsilon'_{33}}{\epsilon_0}. \tag{18}$$

From this quantity as a function of $\epsilon_0(d)$ we can also determine $m_3^{\epsilon,2}$.

Note, that the cantilever-bending-beam experiments of Refs. 1–6 and 11 have been performed on (001) films providing the quantity $\Delta\sigma^{\mathrm{m}}_{(100)}|^{(100)}_{(010)}$ from Eqs. (5)–(7) (in Ref. 10 this quantity was denoted as $\Delta\sigma_1^{\mathrm{m}}=B_1+D^{\mathrm{eff}}\epsilon_0$) and the quantity $\Delta\sigma^{\mathrm{m}}_{(100)''}|^{(110)'}_{(010)'}$ denoted as $\Delta\sigma_2^{\mathrm{m}}=\frac{1}{2}B_2+\frac{1}{4}(D_{12}-D_{11})\epsilon_0$ in Ref. 8. From these quantities linear combinations of $m_1^{\gamma,2}, m_2^{\gamma,2}$ and $m_1^{\epsilon,2}, m_2^{\epsilon,2}$, respectively, can be obtained.

II. *AB INITIO* CALCULATION OF THE PURE SHEAR-STRAIN RELATED ME CONSTANTS

In the experiments, the epitaxial film geometry is used to determine the second-order ME constants because it is nearly impossible to impose similar large strains to a bulk material. In contrast, such strains in bulk materials can be easily simulated in theoretical models. Instead of the more complicated film geometry, we consider the differences in the total-energy density for different magnetization directions in suitably deformed crystals,

$$\Delta e_5=e(\epsilon_{12}=\epsilon_0)|^{(001)}_{(110)}=-\epsilon_0B_2+\frac{1}{2}\epsilon_0^2(m_3^{\gamma,2}-B_1), \tag{19}$$

$$\Delta e_6=e(\epsilon_{23}=\epsilon_{31}=\epsilon_0)|^{(111)}_{(11\bar{2})}$$
$$=\frac{8}{3}\epsilon_0B_2+\frac{1}{12}\epsilon_0^2(B_1+2B_2-m_3^{\gamma,2}+2m_3^{\epsilon,2}) \tag{20}$$

omitting the constant terms due to the cubic magnetocrystalline anisotropy since these are negligible compared to the contribution of the ME energy density. Using similar calculations we have already calculated B_1 as well as the pure tensile-strain-related constants[10] $m_1^{\gamma,2}$, $m_2^{\gamma,2}$, and B_2 as well as the mixed-strain-related constants[8] $m_1^{\epsilon,2}, m_2^{\epsilon,2}$ (in Ref. 8 erroneously denoted as shear-strain-related quantities). Fitting Eqs. (19) and (20) to the *ab initio* results for Δe_5 and Δe_6 as functions of ϵ_0 and using B_1 from our former calculations[10] yield $B_2, m_3^{\gamma,2}$ and $m_3^{\epsilon,2}$. The calculations were performed using the *ab initio* density functional theory in a standard way, described elsewhere,[8,10] applying the WIEN97 code,[12] which adopts the full-potential linearized-augmented-plane wave method.[13]

The results obtained within the local-spin-density approximation[14] (LSDA) and the generalized gradient approximation[15] (GGA) are presented in Fig. 2 and in Table I. The results for B_2 are identical to our previously calculated results.[8] The calculated values of Δe_5 and Δe_6 for fcc Co and fcc Ni are nearly perfectly fitted by harmonic polynomials according to Eqs. (19) and (20), while third-order polynomials describe the data for bcc Fe better, as already found for another pure shear-strain-related mode.[16] This deviation of the calculated results from the prediction of the second-order phenomenological theory of magnetoelasticity might indicate the presence of third-order ME effects in shear-strained bcc Fe. However, one should take into account that there seem to be problems to describe magnetoelasticity of Fe with the LSDA and GGA version of the density-

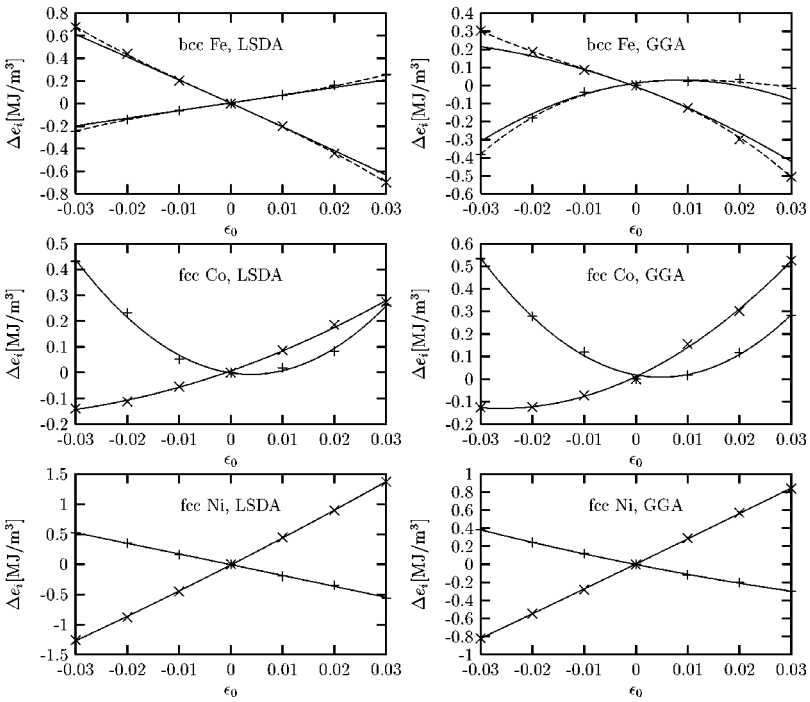

FIG. 2. The calculated differences in the total-energy density as functions of the average epitaxial strain ϵ_0. In the case of fcc Co and fcc Ni, the solid lines represent fits to quadratic polynomials according to Eqs. (19) and (20); $+$, Δe_5; $\times$, Δe_6. In the case of bcc Fe, the dashed lines represent fits to cubic polynomials, while the solid lines correspond to the quadratic contributions, predicted analytically by Eqs. (19) and (20).

functional electron theory. First, both versions give a wrong sign of B_2 as compared to the experimental value of $B_2 = 7.62$ MJ/m^3, see also Refs. 8 and 16. Second, the LSDA and GGA values differ very significantly (Table I). The absolute values of the GGA $m_3^{\gamma,2}$ and $m_3^{\epsilon,2}$ are one order of magnitude larger than the corresponding LSDA quantities, while the ratios between the third-order coefficients, obtained from the fitting polynomials, and $\frac{1}{2}(m_3^{\gamma,2}-B_1)$ in the case of Δe_5 and $\frac{1}{12}(B_1+2B_2-m_3^{\gamma,2}+2m_3^{\epsilon,2})$ in the case of Δe_6 are about -4292 and 180 for LSDA compared to about just -11 and 31 for GGA. The results for fcc Co seem to be more reliable because the difference between the LSDA and GGA values is less significant. So far, a comparison with an experiment[9] is possible just for fcc Ni (see Table I) but due to the large measuring errors and discrepancy between LSDA and GGA, it cannot yield any solid conclusions until the quantities are measured more precisely.

Following the guidelines given in this paper, experimentalists should be able to measure the second-order ME coupling constants of cubic materials with a reasonable accuracy and also answer the question about the possible existence of the third-order effects.

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