Nonlinear magnetoelastic behavior of the bcc phases of Co and Ni: Importance of third-order contributions for bcc Ni

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The first- and second-order magnetoelastic coefficients of the bcc phases Co and Ni are calculated by using a combination of the phenomenological theory of nonlinear magnetoelasticity with the *ab initio* density functional electron theory. The magnetoelastic behavior of the bcc phases is drastically different from that of the corresponding fcc phases. The recently synthesized bcc phase of Ni appears to be an example of a material for which third-order magnetoelastic effects are essential.

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In recent years it became possible to stabilize some of the phases of those materials that do not exist in bulk by growing them on appropriate substrates. For the transition metals Fe, Co, and Ni this is especially interesting because in these systems magnetism and structure are closely related. Using molecular beam epitaxy, the fcc phases of Fe and Co could be stabilized on substrates at room temperature.[1] It became even possible to synthesize the bcc phase of Co[2] on various substrates
(Ref. 3 and references therein), and most recently[4] the bcc phase of Ni. Both of these materials turned out to be ferromagnetic at room temperature, with a magnetic moment per atom of 1.53 μ_B (Co) and 0.54 μ_B (Ni).

From the viewpoint of technological applications of ultrathin magnetic films the most important feature is the magnetic anisotropy. Because in general there will be a lattice mismatch between the substrate and the magnetic film, magnetoelastic contributions to the magnetic anisotropy may be important. For instance, it has been suggested[5] that the in-plane anisotropy of bcc Co on GaAs is dominated by the magnetoelastic contribution, although the epitaxial strains in this material are rather small, about 0.25%. For comparison, for bcc Co on Pt(001) the epitaxial strains are considerably larger[3] (−1.8% in plane and 5.1% out of plane). It is well known that for considerable epitaxial strains nonlinear contributions to the magnetoelastic energy become essential. This has been demonstrated experimentally by cantilever-bending-beam experiments (see, e.g., Refs. 6–8): When changing the direction of the magnetization in the epitaxial film by changing the direction of the external magnetic field, the magnetostrictive stress $\Delta\sigma^{\mathrm{m}}$ along the cantilever axis changes, resulting in a detectable change of the bending of the film-substrate composite. In the framework of linear magnetoelastic theory, this change should be independent of the magnitude of the epitaxial strain and should be determined by the first-order magnetoelastic coefficients, i.e., B_1 and B_2 for cubic materials. Experimentally, however, a linear dependence of $\Delta\sigma^{\mathrm{m}}$ on the strain was found, which was ascribed to nonlinear magnetoelastic effects.

For a proof of this conjecture a knowledge of the first- and second-order magnetoelastic coefficients of the respective bulk material is required. The standard method to determine them is the ultrasonic pulse echo experiment. Because the attainable strains in these experiments are very small, it is, however, nearly impossible to explore the second-order magnetoelastic coefficients by these experiments. The first confirmation of the conjecture therefore was supplied by theory. By a combination of the phenomenological theory of nonlinear magnetoelasticity[9] with the *ab initio* density functional theory it has been shown (see, for example, Refs. 10–13 and references therein) that the second-order magnetoelastic contribution indeed may be very large, especially for the case of Fe. The theory was also able[11] to suggest a complete set of six cantilever experiments to determine the first-order (B_1 and B_2) and the second-order ($m_1^{\gamma,2}, m_2^{\gamma,2}, m_1^{\epsilon,2}, m_2^{\epsilon,2}, m_3^{\gamma,2}, m_3^{\epsilon,2}$) magnetoelastic coefficients of a cubic material. Thereby ($m_1^{\gamma,2}, m_2^{\gamma,2}$) is related to pure tensile strains, ($m_1^{\epsilon,2}, m_2^{\epsilon,2}$) to tensile and shear strains, and ($m_3^{\gamma,2}, m_3^{\epsilon,2}$) to pure shear strains. The first- and second-order coefficients have been calculated[12] by the *ab initio* electron theory for Fe, fcc Co, Ni, Ni_3Fe, and CoFe.

The determination of the magnetoelastic coefficients is especially difficult for phases which can be synthesized only as epitaxial films on substrates, like fcc Co, bcc Co, and bcc Ni, and in these cases the help of electron theory is very important. For the case of fcc Co, the theory has shown[13] that the nonlinear magnetoelastic coupling coefficients are essential for the magnetostrictive strain but have only little influence on the strain-induced out-of-plane anisotropy. In the present paper we apply the theory to the case of bcc Co and bcc Ni. It will be shown that in these systems the nonlinear magnetoelastic coefficients are again very large. Furthermore, it will be shown that bcc Ni is the first example of a system for which third-order magnetoelastic effects become relevant.

It has been shown by the *ab initio* density functional theory[14,15] that free bulk bcc Co and Ni have a negative shear modulus C_{11}-C_{12}, i.e., these phases are elastically instable with respect to the volume-conserving tetragonal strains which transform the system along the Bain path from bcc to fcc (the elastic energy as function of this strain mode exhibits a saddle point at the bcc configuration). Therefore, these phases need to be stabilized by elastic boundary conditions supplied by the interfacial interaction with the substrate. In

the following we suppose that the only effect of the substrate is to stabilize the bcc phase but that the influence of the substrate on the magnetic and magnetoelastic properties of the bcc film can be neglected, i.e., we consider these properties as intrinsic properties of the bulk bcc phase. Indeed the instability of these phases does not affect the calculation of their magnetic properties and of their magnetoelastic coefficients. The calculational procedure of the magnetoelastic coefficients described below depends only on the symmetry of the reference state (here the bcc state) used for the definition of the strain modes, irrespective of the question whether the reference state is stable or not. With the so-obtained values of the magnetoelastic coefficients and the *ab initio* data for the elastic constants of the bcc phase we can also determine without any conceptual problems the magnetostrictive stresses $\Delta\sigma^{\mathrm{m}}$ for the cantilever-bending-beam experiments along the lines described, for example, in Ref. 11. The reason is that in these calculations the strains in the plane parallel to the interface are fixed, i.e., the stabilization of the film by the substrate is implicitly taken into account. However, it does not make sense to define magnetostriction constants for elastically unstable phases. The reason is that the magnetostrictive constants describe for a free system without surface stresses the difference in geometry between the demagnetized stable phase and the magnetized stable phase. If we want to describe magnetostrictive properties in the presently considered situation, we have to consider the respective whole system consisting of the bcc film and the substrate, at least via constraints imposed by the substrate (as we do for the calculation of the magnetostrictive stress).

According to Ref. 11 the magnetoelastic coefficients may be obtained by exposing the cubic material to certain strain modes ϵ_i. Then the difference Δe_i in the total energy per atom when changing the direction of the magnetization from α_i^1 to α_i^2 is calculated for

$$i=1{:}\,\epsilon_1=\epsilon_{xx}=\epsilon_0,\quad \alpha_i^1=\langle 100\rangle,\quad \alpha_i^2=\langle 001\rangle$$

$$\Delta e_1=B_1\epsilon_0+\left(B_1+\frac{1}{2}m_1^{\gamma,2}\right)\epsilon_0^2, \tag{1}$$

$$i=2{:}\,\epsilon_2=\epsilon_{yy}=\epsilon_{zz}=\epsilon_0,\quad \alpha_i^1=\langle 100\rangle,\quad \alpha_i^2=\langle 001\rangle$$

$$\Delta e_2=-B_1\epsilon_0+\frac{1}{2}(-B_1-m_1^{\gamma,2}+m_2^{\gamma,2})\epsilon_0^2, \tag{2}$$

$$i=3{:}\,\epsilon_3=\epsilon_{xx}=\epsilon_{xy}=\epsilon_0,\quad \alpha_i^1=\langle 010\rangle,\quad \alpha_i^2=\langle 110\rangle$$

$$\Delta e_3=\left(\frac{B_1}{2}+B_2\right)\epsilon_0+\frac{1}{2}\left(\frac{1}{2}(B_1+m_1^{\gamma,2})+B_2+m_2^{\epsilon,2}\right)\epsilon_0^2, \tag{3}$$

$$i=4{:}\,\epsilon_4=\epsilon_{zz}=\epsilon_{xy}=\epsilon_0,\quad \alpha_i^1=\langle 010\rangle,\quad \alpha_i^2=\langle 110\rangle$$

$$\Delta e_4=B_2\epsilon_0+\frac{1}{2}m_1^{\epsilon,2}\epsilon_0^2, \tag{4}$$

$$i=5{:}\,\epsilon_5=\epsilon_{xy}=\epsilon_0,\quad \alpha_i^1=\langle 110\rangle,\quad \alpha_i^2=\langle 001\rangle$$

$$\Delta e_5=-B_2\epsilon_0+\frac{1}{2}(m_3^{\gamma,2}-B_1)\epsilon_0^2, \tag{5}$$

$$i=6{:}\,\epsilon_6=\epsilon_{yz}=\epsilon_{zx}=\epsilon_0,\quad \alpha_i^1=\langle 11\bar{2}\rangle,\quad \alpha_i^2=\langle 111\rangle$$

$$\Delta e_6=\frac{8}{3}B_2\epsilon_0+\frac{1}{12}(B_1+2B_2-m_3^{\gamma,2}+2m_3^{\epsilon,2})\epsilon_0^2. \tag{6}$$

The coefficient B_1 and the pair $(m_1^{\gamma,2},m_2^{\gamma,2})$ of second-order coefficients are obtained from Eqs. (1) and (2) by fitting parabola to the data points for $\Delta e_1(\epsilon_0)$ and $\Delta e_2(\epsilon_0)$. Similarly, the coefficients B_2 $(m_3^{\gamma,2},m_3^{\epsilon,2})$ are obtained from Eqs. (5) and (6) by parabolic fits. Finally, the pair $(m_1^{\epsilon,2},m_2^{\epsilon,2})$ is obtained from Eqs. (3) and (4) via parabolic fits using the already determined coefficient $m_1^{\gamma,2}$. As long as the parabolic fits represent the calculated data points $\Delta e_i(\epsilon_0)$ well, we can conclude that third-order magnetoelastic effects can be neglected for the considered range of ϵ_0.

The calculations of $\Delta e_i(\epsilon_0)$ were performed by applying the *ab initio* density functional theory taking into account the spin-orbit coupling which is responsible for magnetoelasticity in a perturbative manner using the second-variational method.[16] Furthermore, we use the WIEN97 code[17] which adopts the full-potential linearized-augmented-plane-wave method (FLAPW)[18] as well as the local-spin-density approximation (LSDA)[19] and the generalized-gradient approximation (GGA)[20] for the exchange-correlation functional. The strains ϵ_i were applied with respect to the theoretically determined equilibrium lattice parameters $a=0.273(0.281)$ nm for bcc Co and $a=0.273(0.279)$ nm for bcc Ni in LSDA (GGA). The resulting LSDA (GGA) magnetic moments per atom of 1.63(1.74) μ_{B} for bcc Co and of 0.47(0.53) μ_{B} for bcc Ni are in agreement with the experimental values of 1.53 μ_{B} and 0.54 μ_{B}, respectively.

For the case of bcc Co all the data points $\Delta e_i(\epsilon_0)$ could be perfectly fitted by parabola in the range $-0.03\leqslant\epsilon_0\leqslant 0.03$, i.e., third-order effects can be neglected. As for other materials,[10–13] the discrepancy between LSDA and GGA may be quite large (see Table I). Because for the experimentally well-investigated B_1 of bcc Co, fcc Ni, and fcc Co the agreement with the GGA values was better than the agreement with LSDA, we concentrate in the following on the GGA results. For bcc Co the values of B_1 and B_2 are quite large as compared to bcc Fe, fcc Ni, and fcc Co. The second-order coefficients are also large. It is interesting that there is a very large difference between bcc Co and fcc Co. This holds even for the first-order coefficients B_1 and B_2 whose magnitudes are considerably larger and of opposite sign for bcc Co as compared to fcc Co. In Ref. 5 it has been assumed that for bcc Co the first-order magnetoelastic coefficients can be approximated by those of fcc Co, in contrast to the results of our calculation.

The case of bcc Ni is even much more interesting because, as shown in Fig. 1, the data points for $\Delta e_i(\epsilon_0)$ show a drastic deviation from a parabolic behavior in the range $-0.03\leqslant\epsilon_0\leqslant 0.03$. This deviation indicates that bcc Ni repre-

TABLE I. The calculated magnetoelastic coefficients (in MJ/m^3), elastic constants C_{11}, C_{12}, and C_{44} (in 10^{11} N/m^2). Our calculated elastic constants for bcc Co and Ni agree nicely with those given in Ref. 15.

		B_1	B_2	$m_1^{\gamma,2}$	$m_2^{\gamma,2}$	$m_1^{\epsilon,2}$	$m_2^{\epsilon,2}$	$m_3^{\gamma,2}$	$m_3^{\epsilon,2}$	C_{11}	C_{12}	C_{44}
fcc Co[12]	LSDA	−15.9	3	243	−53	81	102	759	796	3.85	2.26	
	GGA	−9.8	4.5	184	3	59	−41	862	1681	3.13	1.8	
bcc Co	LSDA	61.5	−35.2	−672	575	357	−108	−363	336	2.1	2.95	1.78
	GGA	28.6	−39.6	−1013	973	148	51	−826	611	1.74	2.33	1.4
fcc Ni[12]	LSDA	12.6	16.9	−117	23	168	−47	−2	388	3.63	2.2	
	GGA	10.2	11.1	−95	71	90	−4	108	96	2.95	1.75	
bcc Ni	LSDA	−0.2	19.1	−116	−100	917	634	≈0	−308	1.99	2.62	1.9
	GGA	−1.3	19.7	−28	−3	87	590	−75	−1347	1.52	2.32	1.63

sents a material for which third-order magnetoelastic effects become very important. Another surprising result is that for bcc Ni the magnitude of B_1 is very small (-1.3 MJ/m^3), much smaller than the one for fcc Ni (10.2 MJ/m^3). As in the case of Co, the magnetoelastic properties of the bcc phase are drastically different from those of the fcc phase. This is in line with the experimental observations[4] that the cubic magnetic anisotropy constant K_1 of bcc Ni is drastically different from the one of fcc Ni, and this was attributed to the different electronic band structures as found by angle-resolved photoemission.

In the past, the magnetic anisotropy of various materials has been analyzed successfully in terms of the electronic band structure,[21] and the magnetoelastic coefficients are of course also related to the properties of the band structure. However, what matters for magnetoelasticity is the change in the band structure when exerting strain to the material. Thus the analysis of the magnetoelasticity in terms of the band structure is much more involved, and we think that it is outright impossible to figure out from the properties of the band structure why the third-order magnetoelastic contributions are so important for the case of bcc Ni.

We hope that our prediction of strong third-order contributions to the magnetoelastic properties of bcc Ni will initiate an experimental investigation by cantilever-bending-beam experiments. To do this one has to grow epitaxial films of bcc Ni with various average epitaxial strains ϵ_0 which may be controlled with the film thickness[6] and then the change $\Delta\sigma^{\mathrm{m}}$ of the magnetostrictive stress due to a change of the magnetization direction has to be measured. For the case that third-order effects are relevant we expect a parabolic dependence

$$\Delta\sigma^{\mathrm{m}} = a + D_1\epsilon_0 + D_2\epsilon_0^2. \tag{7}$$

As discussed above, a linear dependence has been observed experimentally already for several materials. The observation of a quadratic contribution would mean that a material was found for which the third-order magnetoelastic contribution is relevant.

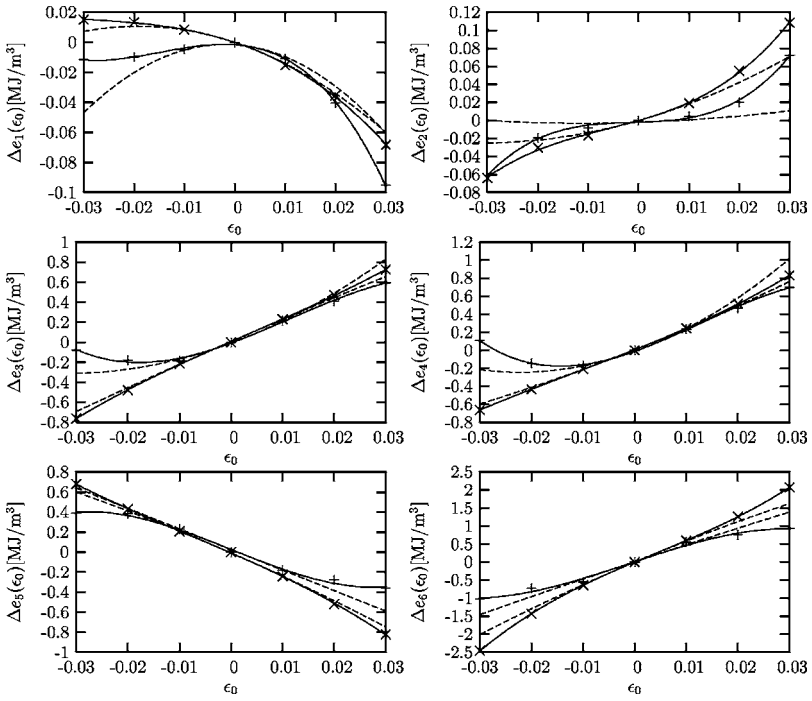

FIG. 1. The calculated functions $\Delta e_i(\epsilon_0)$ by applying LSDA (+) and GGA (×). The solid lines are the third-order polynomials fitted to the *ab initio* calculated data points, whereas the dashed lines represent the contribution up to the second order in ϵ_0.

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