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

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The first- and second-order magnetoelastic coefficients of the metastable 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 metastable phases of materials by growth 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. It became even possible to synthesize the bcc phase of Co on various substrates (Ref. 3 and references therein), and most recently 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 $\mu_B$ (Co) and 0.54 $\mu_B$ (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 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 (-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. 6,68): When changing the direction of the magnetization in the epitaxial film by changing the direction of the external magnetic field, the magnetostrictive stress $\Delta \sigma^m$ 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 $\Delta \sigma^m$ 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 with the ab-initio density functional electron theory it has been shown (see, for example, Refs. 10,11,12,13 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 to suggest a complete set of six cantilever experiments to determine the first-order ($B_1$ and $B_2$) and the second-order ($m_1^{c2}$, $m_2^{c2}$, $m_1^{c2}$, $m_2^{c2}$, $m_3^{c2}$, $m_3^{c2}$) magnetoelastic coefficients of a cubic material. Thereby ($m_1^{c2}$, $m_2^{c2}$) is related to pure tensile strains, ($m_1^{c2}$, $m_2^{c2}$) to tensile and shear strains, and ($m_3^{c2}$, $m_3^{c2}$) to pure shear strains. The first- and second-order coefficients have been calculated by the ab-initio electron theory for Fe, fcc Co, Ni, Ni$_3$Fe and CoFe.

The determination of the magnetoelastic coefficients is especially difficult for metastable 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 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.

According to Ref. 11 the magnetoelastic coefficients may be obtained by exposing the cubic material to certain strain modes $\epsilon_i$. Then the difference $\Delta \epsilon_i$ in the total energy per atom when changing the direction of the magn-
netization from \( \alpha_i^1 \) to \( \alpha_i^2 \) is calculated:

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

\[ \Delta \epsilon_1 = B_1 \epsilon_0 + \left( B_1 + \frac{1}{2} m_{1}^{\gamma,2} \right) \epsilon_0^2 \quad (1) \]

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

\[ \Delta \epsilon_2 = -B_1 \epsilon_0 + \frac{1}{2} \left( -B_1 - m_1^{\gamma,2} + m_2^{\gamma,2} \right) \epsilon_0^2 \quad (2) \]

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

\[ \Delta \epsilon_3 = \left( \frac{B_1}{2} + B_2 \right) \epsilon_0 + \frac{1}{2} \left( \frac{1}{2} B_1 + m_1^{\gamma,2} + B_2 + m_2^{\gamma,2} \right) \epsilon_0^2 \quad (3) \]

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

\[ \Delta \epsilon_4 = B_2 \epsilon_0 + \frac{1}{2} m_{2}^{\gamma,2} \epsilon_0^2 \quad (4) \]

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

\[ \Delta \epsilon_5 = -B_2 \epsilon_0 + \frac{1}{2} \left( m_3^{\gamma,2} - B_1 \right) \epsilon_0^2 \quad (5) \]

\[ i = 6 : \epsilon_6 = \epsilon_{yz} = \epsilon_{xx} = \epsilon_0, \alpha_i^1 = \langle 112 \rangle, \alpha_i^2 = \langle 111 \rangle \]

\[ \Delta \epsilon_6 = \frac{8}{3} B_2 \epsilon_0 + \frac{1}{12} \left( B_1 + 2B_2 - m_3^{\gamma,2} + 2m_3^{\gamma,2} \right) \epsilon_0^2 \quad (6) \]

The coefficient \( B_1 \) and the pair \( \left( m_1^{\gamma,2}, m_2^{\gamma,2} \right) \) of second-order coefficients are obtained from eqs.(1,2) by fitting parabola to the data points for \( \Delta \epsilon_1(\epsilon_0) \) and \( \Delta \epsilon_2(\epsilon_0) \). Similarly, the coefficients \( B_2, \left( m_3^{\gamma,2}, m_4^{\gamma,2} \right) \) are obtained from eqs.(3,4) by parabolic fits using the already determined coefficient \( m_1^{\gamma,2} \). As long as the parabolic fits represent the calculated data points \( \Delta \epsilon_i(\epsilon_0) \) well, we can conclude that third-order magnetoelastic effects can be neglected for the considered range of \( \epsilon_0 \).

The calculations of \( \Delta \epsilon_i(\epsilon_0) \) 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. Furthermore, we use the WIEN97 code which adopts the full-potential linearized-augmented-plane-wave method (FLAPW) as well as the local-spin-density approximation (LSDA) and the generalized-gradient approximation (GGA) for the exchange-correlation functional. The strains \( \epsilon_i \) 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)\( \mu_B \) for bcc Co and of 0.47(0.53)\( \mu_B \) for bcc Ni are in agreement with the experimental values of 1.53\( \mu_B \) and 0.54\( \mu_B \), respectively.

For the case of bcc Co all the data points \( \Delta \epsilon_i(\epsilon_0) \) could be perfectly fitted by parabola in the range \(-0.03 \leq \epsilon_0 \leq 0.03 \), i.e., third-order effects can be neglected. Like for other materials\(^{10,11,12,13} \) the discrepancy between LSDA and GGA may be quite large. 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 \( B_2 \) which magnitudes are considerably larger and of opposite sign for bcc Co as compared to fcc Co. In Ref.\(^3 \) 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 \( \Delta \epsilon_i(\epsilon_0) \) show a drastic deviation from a parabolic behavior in the range \(-0.03 \leq \epsilon_0 \leq 0.03 \). To the best of our knowledge, bcc Ni therefore represents the first known 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 \((-1.3 \text{ MJ/m}^3)\), much smaller than the one for fcc Ni \((10.2 \text{ MJ/m}^3)\). 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\(^{14} \) 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.

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 \( \epsilon_0 \) which may be controlled with the film thickness\(^{15} \) and then the change \( \Delta \sigma^m \) 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:

\[ \Delta \sigma^m = a + D_1 \epsilon_0 + D_2 \epsilon_0^2 \quad (7) \]

As discussed above, a linear dependence has been observed experimentally already for several materials. The observation of a quadratic contribution would mean that for the first time 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 $\epsilon_0$. 
TABLE I: The calculated magnetoelastic coefficients (in MJ/m$^3$), elastic constants $C_{11}$, $C_{12}$ and $C_{44}$ (in $10^{11}$ N/m$^2$), the magnetostrictive coefficients $\lambda_{100} = -2B_1/(3(C_{11} - C_{12})$ and $\lambda_{111} = -B_2/3C_{44}$ (in units $10^{-5}$) Our calculated elastic constants for bcc Co and Ni agree nicely with those given in Ref. 19.

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