BAND THEORY OF THE LINEAR MAGNETOSTRICTION FOR FERROMAGNETIC TRANSITION METALS*

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It is shown that for ferromagnetic transition metals with cubic symmetry the change of one-electron state occupation numbers, caused by the influence of the spin-orbit interaction and lattice deformation on initially doubly degenerate electronic bands which intersect the Fermi level, may be considered as the mechanism of linear magnetostriction. Expressions for magnetostriction constants for the [100] and [111] directions are calculated. The crude estimation for Ni shows that the proposed mechanism gives the correct result for the [111] direction, but not for the [100] direction. In the last case, other details of the electronic band structure must be taken into account.

It is well known, that linear magnetostriction in ferromagnets is related to a dependence of the magnetocrystalline anisotropy energy on lattice deformation. Therefore, similarly as in the case of magnetocrystalline anisotropy, the spin-orbit interaction is usually treated as a physical reason for magnetostriction. For itinerant ferromagnets, like transition metals (e.g. nickel), the main contribution to the anisotropy energy comes from the energy bands, which intersect the Fermi level and are doubly degenerate in the absence of spin-orbit interaction [1–3]. With the spin-orbit coupling taken into account, these bands split and the change of one-electron state occupation numbers near the Fermi level, connected with this splitting, leads to a lowering of the energy, dependent on the magnetization direction, and contributes to the anisotropy energy. It is quite possible, that the same mechanism is responsible for the linear magnetostriction. However, in the previous investigations of the linear magnetostriction within a framework of the band model, the energy degeneration [4, 5] and the variation of occupation numbers near the Fermi level [4] were completely neglected. Moreover, in the previous papers [4, 5] very simple band structures were assumed. Therefore, it seems worth-while to check whether the described mechanism does lead to the linear magnetostriction and try to estimate the role of this mechanism for constants of

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linear magnetostriction. That is the purpose of this paper; calculations are performed for nickel.

We consider a simple model of the electronic band structure of nickel and calculate the change of the mean energy of the system caused by the lattice deformation and spin-orbit interaction. For this purpose, we assume that in the absence of both perturbations the essential features of band structure of nickel, which are important for magnetocrystalline anisotropy and magnetostriction effects, can be described by the two-band model proposed by Mori [3]. In this model, the doubly degenerate bands are partially occupied by electrons with spin ↑, whereas the bands for electrons with spin ↓ are empty.

Bloch functions which diagonalize the one-electron Hamiltonian H in the absence of perturbations are denoted here by $\psi^0_{\lambda k\sigma}(r)$ and the corresponding one-electron energies by $E^0_{\lambda k\sigma}$, where λ is the band index, k — the wave vector and $\sigma = \uparrow$ or $\downarrow$ is the spin index. In the tight-binding method, the spin-independent part of the wave function, $\psi^0_{\lambda k}(k)$, can be expressed as a linear combination of atomic orbitals φ_i, namely

$$\psi^0_{\lambda k}(r) = \frac{1}{\sqrt{N}} \sum_h e^{ikR_h^0} \sum_i c_{\lambda i}(k)\varphi_i(r-R_h^0), \tag{1}$$

where R_h^0 represents a vector in the crystal lattice before deformation and N is the number of lattice sites.

Let us assume now, that the crystalline lattice is submitted to some deformation. This deformation leads to a change of crystalline potential and to a variation of eigenfunctions, too. So, the one-electron Hamiltonian in $\psi^0_{\lambda k}(r)$ representation becomes non-diagonal. When the lattice deformation is very small, energy and wave function corrections can be calculated by means of the perturbation method. With only two degenerate bands intersecting the Fermi level taken into account, the following expressions for one-electron energies and corresponding Bloch functions are obtained:

$$E_k^{\pm} = E^0_{\lambda k} + \tfrac{1}{2}(\Delta E_{11k} + \Delta E_{22k}) \pm \tfrac{1}{2}\sqrt{4\Delta E_{12k}^2 + (\Delta E_{11k} - \Delta E_{22k})^2}\,, \tag{2}$$

$$\psi_k^+(r) = \cos\frac{\alpha_k}{2}\psi^0_{1k}(r) + \sin\frac{\alpha_k}{2}\psi^0_{2k}(r), \tag{3}$$

$$\psi_k^-(r) = -\sin\frac{\alpha_k}{2}\psi^0_{1k}(r) + \cos\frac{\alpha_k}{2}\psi^0_{2k}(r), \tag{4}$$

where

$$\operatorname{tg}\alpha_k = \frac{2\Delta E_{12k}}{\Delta E_{11k} - \Delta E_{22k}}, \tag{5}$$

$$\Delta E_{\lambda\lambda' k} = \sum_{ij} c^*_{\lambda i}(k) c_{\lambda' j}(k) \sum_h e^{-ikR_h^0} [\nabla E_{ij}(R_h^0)(\vec{e}R_h^0) - E_{ij}(R_h^0) ik (\vec{e}R_h^0)] \tag{6}$$

with

$$E_{ij}(R_h^0) = \int d^3 r \varphi_i^*(r - R_h^0) H(r) \varphi_j(r), \tag{7}$$

and $\hat{e}$ represents here a deformation tensor. In formula (6) for $\Delta E_{\lambda\lambda' k}$ only linear terms of the expansion with respect to the lattice deformation are included.

Next, we consider the spin-orbit coupling. In the presence of this interaction, the one-electron Hamiltonian should be completed by the following term:

$$H_{so} = \frac{\hbar}{4m^2c^2}\,\sigma(\nabla V \times p), \tag{8}$$

where σ is the Pauli spin operator, $V(r)$ is the crystalline potential and p is the momentum operator.

According to Ricodeau [6] the corrections to spin-orbit coupling and magneto-crystalline anisotropy connected with the deviation of the crystalline potential V from spherical symmetry for transition metals, especially for nickel, are negligibly small. Therefore, it is justified to calculate the matrix elements of the Hamiltonian, representing the spin-orbit interaction, with use of the expression $\sum_h \xi(r-R_h^0)SL(r-R_h^0)$ (L is the angular momentum operator at the site R_h^0, $S = \frac{1}{2}\hbar\sigma$ and ξ is a function depending only on the absolute magnitude of the radius vector) instead of the general formula (8). Moreover, it was shown by Wang [7] that the only non-negligible matrix elements of H_{so} are those calculated with the use of atomic d-orbitals, centered on the same atomic site. They can be expressed in the form:

$$[(H_{so})_{ij}^{\sigma\sigma'}] = \xi \begin{matrix} \\ \uparrow \\ \downarrow \end{matrix} \begin{matrix} \begin{matrix} \uparrow & \downarrow \end{matrix} \\ \begin{bmatrix} M & N \\ -N^* & M^* \end{bmatrix} \end{matrix}, \tag{9}$$

where ξ is a spin-orbit coupling parameter, M and N are 5×5 matrices describing the LS interaction in atomic d-orbital representation, which are given, for example, by Abate and Asdente [8]. In formula (9) the considered spin states are indicated by arrows.

Now, we can calculate the change in band structure for ferromagnetic nickel, caused both by the lattice deformation and the spin-orbit coupling. For this purpose, matrix elements of the LS interaction are expressed in $\psi_{k\sigma}^{\pm}$ — function representation, in which the one-electron Hamiltonian is diagonalized in the presence of the lattice deformation. Next, we use the perturbation method for the case of two nearly situated energy levels (according to formula (2) the lattice deformation leads only to a very small modification of the assumed band structure). Then, taking into account that the spin-orbit splitting is small in comparison to the exchange splitting and that the contribution of the lattice deformation is small even in comparison to the LS interaction (terms proportional to the square of the deformation tensor are neglected), we obtain

$$E_{nk} = E_k^0 + \tfrac{1}{2}(\Delta E_{11k} + \Delta E_{22k}) - \hat{n}|E_{so}^{12}|, \qquad \hat{n} = \pm 1, \tag{10}$$

where

$$E_{so}^{12} = \sum_{ij} c_{1i}^*(k)c_{2j}(k)\,(H_{so})_{ij}^{\uparrow\uparrow}. \tag{11}$$

The change of the energy of the system of electrons in the one-electron approximation, ΔE, caused by the lattice deformation and the spin-orbit interaction can be expressed in a form:

$$\Delta E = \sum_{nk} E_{nk}\Theta(E_F - E_{nk}) - \sum_{nk} E^0_{nk}\Theta(E^0_F - E^0_{nk})$$

$$\simeq \sum_{nk} (E_{nk} - E^0_{nk})\Theta(E^0_F - E^0_{nk}) + \sum_{nk} E_{nk}[\Theta(E^0_F - E_{nk}) - \Theta(E^0_F - E^0_{nk})]$$

$$+ \sum_{nk} E_{nk}[\Theta(E_F - E_{nk}) - \Theta(E^0_F - E_{nk})], \tag{12}$$

where Θ is the step function, E^0_{nk}, E^0_F denote the one-electron energy and the Fermi energy prior to perturbations, respectively, and E_F is the Fermi energy with lattice deformation and spin-orbit coupling taken into account. E_F can be calculated according to the condition that the number of occupied states does not change when perturbations are introduced:

$$\sum_{nk} [\Theta(E_F - E_{nk}) - \Theta(E^0_F - E^0_{nk})] = 0. \tag{13}$$

Similarly as in the case of the magnetocrystalline anisotropy (see, e.g. [2]) the first term in the expression for ΔE can be interpreted as the volume contribution and the two remaining terms as the surface contribution (related to the change of the Fermi surface shape caused by perturbations). If only two degenerate bands are taken into account and the change of the Fermi energy is neglected, we can expect that analogically as in the case of the magnetocrystalline anisotropy [2], the main contribution to magnetostriction is given by the second term. Then, the change in the system energy ΔE is first of all a result of variation of one-electron state occupation numbers. Considering only states very close to the Fermi level we can express ΔE approximately in the form:

$$\Delta E \simeq E^0_F \sum_{nk} [\Theta(E^0_F - E_{nk}) - \Theta(E^0_F - E^0_{nk})]. \tag{14}$$

The investigations of the band structure of ferromagnetic nickel show that the doubly degenerate bands intersecting the Fermi level appear along the ΓL and ΓX directions of the Brillouin zone and these bands are not influenced by $s-d$ hybridization. Analogically as in Mori's paper [3] we assume that these regions of the Brillouin zone give the main contribution to ΔE. In Eq. (14), the expression $\sum_{nk} [\Theta(E^0_F - E_{nk}) - \Theta(E^0_F - E^0_{nk})]$ is proportional to this part of the volume of the Brillouin zone where the variation of occupation numbers takes place. Due to the fact that in the presence of spin-orbit interaction and lattice deformation, the directions [111], [1$\bar{1}$1], [$\bar{1}$11] and [11$\bar{1}$] as well as [100], [010] and [001] are non-equivalent, we can approximately write:

$$\sum_{k} [\Theta(E^0_F - E_{nk}) - \Theta(E^0_F - E^0_{nk})] \simeq \frac{2v}{(2\pi)^3}\left[\sum_{i=1}^{4} (\Delta k_n)^3_{L_i} + \sum_{j=1}^{3} (\Delta k_n)^3_{X_j}\right], \tag{15}$$

where i, j — indices characterize the non-equivalent ΓL and ΓX directions in the Brillouin zone, respectively. v is the crystal volume and Δk_n is calculated according to condition (analogically to [9]):

$$E_n^0(k_F) = E_n(k_F + \Delta k_n). \tag{16}$$

We obtain

$$\Delta k_n = \left(\frac{\partial E}{\partial k}\bigg|_{k=k_F}\right)^{-1} [\hat{n}|E_{so}^{12}| - \tfrac{1}{2}(\Delta E_{11k_F} + \Delta E_{22k_F})], \tag{17}$$

then

$$\Delta E = -\frac{6v}{(2\pi)^3 a^3} E_F^0 \left\{\left(\frac{\partial E^0}{\partial ak}\bigg|_{k=k_F}^{111}\right)^{-3} \sum_{i=1}^{4} [|E_{so}^{12}|^2(\Delta E_{11k_F} + \Delta E_{22k_F})]_{L_i}\right.$$

$$\left. + \left(\frac{\partial E^0}{\partial ak}\bigg|_{k=k_F}^{100}\right)^{-3} \sum_{j=1}^{3} [|E_{so}^{12}|^2(\Delta E_{11k_F} + \Delta E_{22k_F})]_{X_j}\right\}, \tag{18}$$

where a is the lattice constant. To calculate the matrix elements of spin-orbit coupling and lattice deformation which appear in the above expression, we assume that the Bloch functions for electrons in the degenerate energy bands, which cross the Fermi level in the ΓL and ΓX directions of the Brillouine zone, can be expressed in the tight-binding approximation by use of linear combinations of atomic d-orbitals $\varphi_1 \ldots \varphi_5$ in the following way: along ΓL axis ([111] direction of the crystal)

$$\psi_1 = (\varphi_1 - \varphi_2)/2 + (\varphi_4 - \sqrt{3}\,\varphi_5)/2\sqrt{2},$$

$$\psi_2 = (\varphi_1 + \varphi_2 - 2\varphi_3)/2\sqrt{3} - (\sqrt{3}\,\varphi_4 + \varphi_5)/2\sqrt{2}, \tag{19}$$

and along ΓX axis ([100] direction of the crystal)

$$\psi_1 = \varphi_1, \quad \psi_2 = \varphi_3. \tag{20}$$

In expression (18) for ΔE one can find the usual contributions corresponding to magnetoelastic energy. This energy (per unit volume) for crystals with cubic symmetry can be written phenomenologically as follows:

$$\Delta E_{mgel} = -\tfrac{3}{2}(c_{11} - c_{12})(\alpha_1^2 e_{11} + \alpha_2^2 e_{22} + \alpha_3^2 e_{33})\lambda_{100}$$

$$-3c_{44}(\alpha_1\alpha_2 e_{12} + \alpha_2\alpha_3 e_{23} + \alpha_1\alpha_3 e_{13})\lambda_{111}, \tag{21}$$

where λ_{100}, λ_{111} are magnetostriction constants in the [100] and [111] directions, respectively; c_{ij} are elastic constants, α_i denote direction cosines of the magnetization vector with respect to the crystallographic axes and e_{ij} are components of the deformation tensor. To calculate the magnetostriction constants we find in formula (18) all terms of the form

such as those in Eq. (21). Then, we obtain

$$\lambda_{100} = \frac{\xi^2 E_F^0}{2\pi^3 a^3 (c_{11}-c_{12})} \left(\frac{\partial E^0}{\partial ak}\bigg|_{k=k_F}^{100}\right)^{-3} \left[(A_1 - A_2) ak_F \sin\frac{ak_F}{2}\right.$$

$$\left. + (\tfrac{1}{2} A_1' + A_2'') \cos\frac{ak_F}{2} - (A_2' + A_2'')\right], \tag{22}$$

$$\lambda_{111} = \frac{(2\sqrt{2}-1)^2 \xi^2 E_F^0}{9\pi^3 a^3 c_{44}} \left(\frac{\partial E^0}{\partial ak}\bigg|_{k=k_F}^{111}\right)^{-3} \left[(A_1 - 2A_2 + A_3 - A_4 + 2A_5 + 2\sqrt{2} A_6)\right.$$

$$\left. \times \frac{ak_F}{2\sqrt{3}} \sin\frac{ak_F}{\sqrt{3}} - (\tfrac{1}{2} A_1' + A_2' + A_2'' - A_3' + 2A_4' - 2A_5' + \sqrt{2} A_6') \sin^2\frac{ak_F}{2\sqrt{3}} - A_3' + \sqrt{2} A_6'\right], \tag{23}$$

where

$$A_1 = -\int d^3 r \varphi_1 \left(x - \frac{a}{2}, y - \frac{a}{2}, z\right) H(r) \varphi_1(x, y, z) \tag{24}$$

and the other A_m are defined analogically (see, e.g. [10]). Here A_m', A_m'' include the integrals of the type $\int d^3 r \nabla_{R_h^0} \varphi_i(r - R_h^0) H(r) \varphi_j(r)$ and are given in the appendix.

One can see that the described mechanism really contributes to the linear magnetostriction, but it is especially interesting how important this contribution is. Therefore, we try to estimate the values of magnetostriction constants λ_{100} and λ_{111} for Ni. The following data are taken: $E_F = -0.5342$ Ry, $ak_F^{100} = 4.7171$, $ak_F^{111} = 2.5553$, $\left.\frac{\partial E}{\partial ak}\right|_{k=k_F}^{100} = 0.1271$ Ry, $\left.\frac{\partial E}{\partial ak}\right|_{k=k_F}^{111} = 0.0313$ Ry. They were found by Laurent according to Wang and Callaway [11] calculations of band structure of nickel with the use of the von Barth–Hedin potential. The expressions in square brackets in formulae for magnetostriction constants are estimated approximately by taking into account only three d-orbitals φ_1, φ_2, φ_3. Then, the values of the integrals A_1, A_2, A_3can be taken from paper [12] and of integrals A_1', A_2', A_2'', A_3' from paper [4]. We put also: $\xi = 6.7 \cdot 10^{-3}$ Ry [7], $a = 3.508 \cdot 10^{-8}$ cm, $c_{11} = 2.5 \cdot 10^{12}$ erg/cm^3, $c_{12} = 1.6 \cdot 10^{12}$ erg/cm^3, $c_{44} = 1.185 \cdot 10^{12}$ erg/cm^3 (these elastic constants for Ni were measured at 300 K) [13].

Then, the calculated magnetostriction constants are equal:

$$\lambda_{100} = -7.13 \cdot 10^{-6}, \quad \lambda_{111} = -29.5 \cdot 10^{-6}. \tag{25}$$

The obtained results show that there is quite good agreement between the calculated magnetostriction constant and experimental data for the [111] direction ($\lambda_{111}^{exp} = -25 \cdot 10^{-6}$) and a rather significant discrepancy for the [100] direction ($\lambda_{100}^{exp} = -46 \cdot 10^{-6}$). The calculated value of λ_{100} is much lower than the experimental one. A reason for such a disagreement becomes clear when one realizes that the obtained magnetostriction con-

stants first of all depend on the reversal of the gradient of energy at the Fermi level and for the [100] direction this gradient is much higher than for the [111] direction.

Therefore, we can state that the change related to spin-orbit coupling and lattice deformation of one-electron state occupation numbers in the doubly degenerate bands which cross the Fermi level has an important influence on the magnetostriction constant λ_{111} of Ni, but for accurate calculation of λ_{100} it is necessary to take into account other details of the electronic band structure which have not been considered in the above calculations.

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APPENDIX

The A'_m and A''_m integrals are defined as follows:

$$A'_1 = -a\int d^3r\left(y-\frac{a}{2}\right)\left[F\left(x-\frac{a}{2},\ y-\frac{a}{2},\ z\right)\right.$$

$$\left.+\left(x-\frac{a}{2}\right)^2 F'\left(x-\frac{a}{2},\ y-\frac{a}{2},\ z\right)\right]H(r)xyF(x,y,z),$$

$$A'_2 = \frac{a}{6}\int d^3ry\left[F\left(x-\frac{a}{2},\ y,z-\frac{a}{2}\right)+\left(x-\frac{a}{2}\right)^2 F'\left(x-\frac{a}{2},\ y,z-\frac{a}{2}\right)\right.$$

$$\left.-2\left(x-\frac{a}{2}\right)\left(z-\frac{a}{2}\right)F'\left(x-\frac{a}{2},\ y,z-\frac{a}{2}\right)\right]H(r)xyF(x,y,z),$$

$$A''_2 = \frac{a}{6}\int d^3ry\left[-2F\left(x-\frac{a}{2},\ y,z-\frac{a}{2}\right)-2\left(x-\frac{a}{2}\right)^2 F'\left(x-\frac{a}{2},\ y,z-\frac{a}{2}\right)\right.$$

$$\left.-\left(x-\frac{a}{2}\right)\left(z-\frac{a}{2}\right)F'\left(x-\frac{a}{2},\ y,z-\frac{a}{2}\right)\right]H(r)xyF(x,y,z),$$

$$A'_3 = -\frac{a}{4}\int d^3ry\left[F\left(x-\frac{a}{2},\ y,z-\frac{a}{2}\right)+\left(x-\frac{a}{2}\right)^2 F'\left(x-\frac{a}{2},\ y,z-\frac{a}{2}\right)\right.$$

$$\left.+\left(x-\frac{a}{2}\right)\left(z-\frac{a}{2}\right)F'\left(x-\frac{a}{2},\ y,z-\frac{a}{2}\right)\right]H(r)yzF(x,y,z),$$

$$A'_4 = -\frac{a}{8}\int d^3r\left(x-\frac{a}{2}\right)\left\{2F\left(x-\frac{a}{2},\ y-\frac{a}{2},\ z\right)\right.$$

$$\left.+\left[\left(x-\frac{a}{2}\right)^2-\left(y-\frac{a}{2}\right)^2\right]F'\left(x-\frac{a}{2},\ y-\frac{a}{2},\ z\right)\right\}H(r)\,(x^2-y^2)F(x,y,z),$$

$$A_5' = \frac{a}{8}\int d^3r \left\{\left(z-\frac{a}{2}\right)\left[2F\left(x-\frac{a}{2},\, y,\, z-\frac{a}{2}\right)+\left(\left(z-\frac{a}{2}\right)^2-y^2\right)\right.\right.$$

$$\left.\times F'\left(x-\frac{a}{2},\, y,\, z-\frac{a}{2}\right)\right]+\left(x-\frac{a}{2}\right)\left[z^2-\left(y-\frac{a}{2}\right)^2\right]$$

$$\left.\times F\left(x-\frac{a}{2},\, y-\frac{a}{2},\, z\right)\right\} H(r)\,(z^2-y^2)F(x,y,z),$$

$$A_6' = \frac{a}{8}\int d^3r \left\{\left(y-\frac{a}{2}\right)\left[F\left(x-\frac{a}{2},\, y-\frac{a}{2},\, z\right)\right.\right.$$

$$\left.+\left(x-\frac{a}{2}\right)^2 F'\left(x-\frac{a}{2},\, y-\frac{a}{2},\, z\right)\right] H(r)\,[2z^2-x^2-y^2]F(x,y,z)$$

$$-\left(x-\frac{a}{2}\right)\left[2F\left(x-\frac{a}{2},\, y-\frac{a}{2},\, z\right)-\left(2z^2-\left(x-\frac{a}{2}\right)^2\right.\right.$$

$$\left.\left.\left.-\left(y-\frac{a}{2}\right)^2\right)F'\left(x-\frac{a}{2},\, y-\frac{a}{2},\, z\right)\right] H(r)xyF(x,y,z)\right\},$$

$$F(x,y,z) \equiv F(r) = \left(\frac{15}{4\pi}\right)^{1/2} f(r)/r^2,$$

$$F'(r) = \frac{d}{dr}F(r).$$

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