Europhysics News (2003) Vol. 34 No. 6

Magnetostriction: fundamental principles and novel magneto-strictive materials

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Magnetostriction and magnetoelastic coupling

When any magnetic solid acquires a magnetization, $\vec{M}$under the application of a magnetic field, $\vec{H}$at the same time its crystalline (or morphous) lattice is deformed, a phenomenon called *magnetostriction* (MS from hereinafter). Therefore this is a general effect suffered by all solid matter, once it becomes magnetized. It therefore appears for any kind of magnetic materials: diamagnetic, pramagnetic, ferromagnetic, anti-ferromagnetic, ferrimagnetic, superconductors, etc. It does not matter if the solid is a metal or an insulator, what only matters is if the atoms either acquire an induced magnetic moment or they have a permanent one. By Hokes's law the deformation suffered is proportional to the material size, say *l*, and therefore it is convenient to express the deformation by the relative variation $\lambda = \Delta 1/1$, called the *linear* MS. In a crystal λ is *doubly anisotropic:* it depends on the crytallographic direction of measurement and on the direction along which the magnetization, $\vec{M}$is oriented by the applied magnetic field, $\vec{H}$. Because MS depends on the crystalline directions of λ and $\vec{M}$($\vec{\beta}$ and $\vec{\alpha}$, respectively) we have to specify MS as $\lambda(\vec{\alpha}, \vec{\beta})$ (Fig.1). But also in a poly-crystalline material, formed by small crystallites more or less randomly distributed, the MS measured along $\vec{H}$, called the parallel MS, $\lambda_{\|}$ is quite different from that measured along the perpendicular direction, termed te perpendicular MS, $\lambda_{\perp}$. It can happen that $\lambda_{\perp} \cong - \lambda_{\|}/2$, but usually this is not the case and then the volume of the solid changes by the amount $\Delta V / V = \lambda_{\|} + 2\lambda_{\perp}$, called the *volume* MS. On the other hand, the shape (or form of the unit cell) of the mateial is modified if $\lambda_{\|} \neq \lambda_{\perp}$, and a measure of this effect is the difference $\lambda t = \lambda_{\|} - \lambda_{\perp}$, called the shape MS. Both MS's appear in crystalline, polycrystalline and amorphous solids.

The key questions are: why is this phenomenon so general, and what is te physical mechanism of MS? We know that atoms possess two main sources of magnetic moment: the orbital electronic motion around the nucleus, which gives rise to the orbital angular momentum, $\vec{L}$and the spin, $\vec{S}$. Classically the orbital current produces amagnetic field, $\vec{B}_L$ where the spin magnetic moment, $\vec{m}_S = -\vec{s}\mu_B$ has an energy $-\vec{m}_S \cdot \vec{B}_L$. This is a relativistic effect, and because $\vec{B}_L \sim \vec{L}$, the energy becomes $E_{SO} = \xi \vec{L} \cdot \vec{S}$, which links the spins to the orbits, e.g. in a ferromagnet. This interaction iscalled the *spin-orbit coupling*, and is the first ingredient of MS. By it when $\vec{S}$(or $\vec{M}$) is rotated by the torque $\Gamma = \vec{m}_s x \vec{B}$, $\vec{L}$is dragged on. In a ferromagnet this is very important because the spins become ordered below the Curie temperature. But there s another equally important ingredient, the interaction which couples the orbit to the lattice, if we want the lattice to deform when their atoms are magnetized.

Let us consider a ferromagnet (FM) or antiferromagnet (AF) in order to fix ideas. For transiion metals (TM), notably Fe, Co, Ni and Mn, or their ions (in FM insulators) and for rare earth (RE) metals and insulators, their respective atomic 3d and 4f shells are incompletely filled, and therefore they possess spin and orbital momenta, giving rise o a magnetic moment, $m = m_L + m_S$. In magnetic solids there exists a strongly inhomogeneous electric field, called the *crystal electric field* (CEF), whose gradients interact with the ion magnetic electrons, giving rise to a splitting of the ion energy leves while still keeping some degeneracy (CEF energy levels). The splitting is purely dictated by symmetry: e.g. in cubic symmetry the levels are e_g (x2) and $t2_g g$ (x3). This remaining degeneracy is quite important for MS, because otherwise $\langle L \rangle$ is zero. In thesolid, 3d and 4f electrons have orbital wave-functions, φ_{orb}, rather different from those of the free atoms (or ions), as a consequence of the CEF potential which admixes the free atom states $|M_L>$ (M_L is the quantized projection of $\vec{L}$along some crystal smmetry axis, OZ). So the electronic charge distribution (i.e. $|\varphi_{orb}|^2$) is determined by the symmetry entourage of the ionic site and is very anisotropic. Most importantly is that φ_{orb} changes when $\vec{L}$rotates, with the result that the CEF energy changes, gving rise to the *magnetocrystalline anisotropy* (MCA), by which the crystal energy changes with the $\vec{M}$direction. 3d shells are little screened from the CEF ("medium" to "strong" CEF) whereas 4f ones are much more screened ("weak" CEF). For the former, theground state φ_{orb} is formed by *almost* pure 50% admixtures of $|\pm M_L>$ states and therefore the quantum mechanics expectation value $\langle L_Z \rangle$ is rather weak (an effect called quenching). Contrarily for the RE metals quenching is very weak, i.e., $\langle L_L \rangle$ is weakly reuced. FM or AF order in solids is the result of the exchange interaction between electrons, with energy $E_{ex} = - 2J\, S_i \cdot S_j$, which is clearly isotropic. Very important for MS is the fact that J (exchange integral) depends on the atom distance. This interactin gives rise to the spontaneous magnetization, $\vec{M}_S$. Therefore when $\vec{M}_S$ (or $\vec{S}$) is rotated within the crystal by the $\vec{\Gamma}$torque, the ionic charge cloud is dragged on and also distorted, with the consequent distortion of the ion entourage: so *the crystal isdeformed*. This deformation is the so-called single-ion *CEF magnetostriction* and it requires the rotation of $\vec{M}_S$ in some way: either by rigid vector rotation or by domain wall displacements that entrain $\vec{M}_S$ rotation. This happens for 70.5°, 90°, and 109.5°domain walls (DW) in cubic crystals (180° DW displacement can not produce MS, which is the case for uniaxial crystals when they are magnetized along the easy magenetization direction). In elasticity theory we know that the deformation, or more properly th *strain*, ϵ_{ij} (a tensor), is the medium response to an external stress, σ_{ij}, the relationship being governed by Hooke's linear law, $\sigma_{ij} = c_{ijkl}\epsilon_{kl}$ (repeated indices summation assumed), c_{ijkl} being the elastic constants. Therefore we can equally think that n internal magnetoelastic (MEL) stress, B_{ij}, develops under $\vec{L}$rotation and gives rise to the MS and, in accordance with elasticity theory, the MEL energy gain is given by $F_{ms} = - B_{ij}\epsilon_{ij}$. When the solid deforms under the MEL stress, there is a cost of elatic energy, $E_{el} = (1/2)c_{ijkl}\, \epsilon_{ik}\, \epsilon_{jl}$, and an equilibrium MS, for short $\lambda = B / c$, is attained when the full energy is minimized. The B_{ij} are also called *MEL* constants and the magnetic moment, **m**, coupling to the strained lattice, is called *MEL coupling*. Th number of strains, and so of *MEL* constants, is dictated by the crystal symmetry alone. The *irreducible* strains are the minimum number of them, spanning all unit cell deformations compatible with symmetry. They are four for cubic symmetry (see fig. 2): voume dilation ϵ^{α} (a), tetragonal distortion along a cubic axis ϵ^{γ} (b), orthorhombic deformation of {100} planes ϵ[illegible](not shown in Fig. 2), and shear of the <100> axes ϵ^{ε} (c).

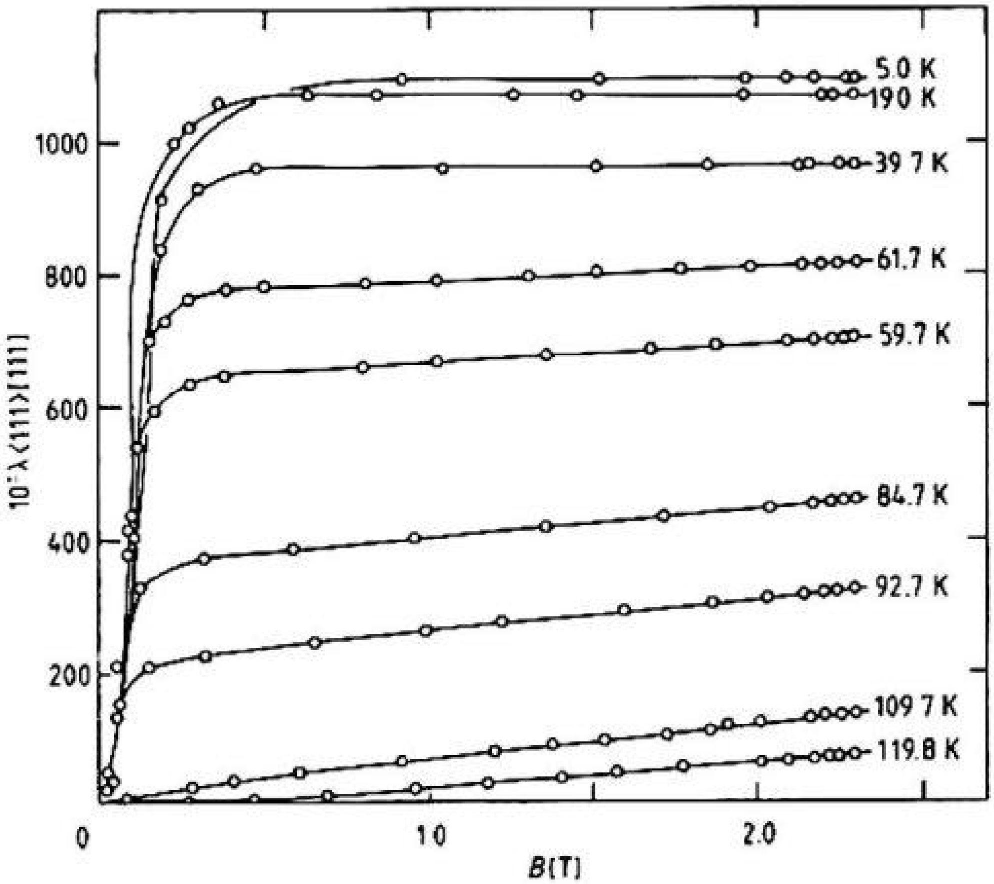

Fig 1 1**([111], (111)) MS isotherms vs. applied magnetic field for the cubic intermetallic ferromagnet $TbAl_2$ (Tc =115K) [1].**

Invar effect, magnetostrictive superlattices and "giant" magnetostriction materils

There is a second kind of MS due to the spatial dependence of the exchange (EX) interaction, i.e. $\partial J(r)/\partial r$, called *exchange* (*EX*) MS. Because $J\, S_i{\cdot}S_j$ is isotropic, EX MS produces just a *volume* deformation (VMS), given by $\omega ex \sim (\partial J(r)/\partial r)/B$, where B isthe bulk modulus. Since this VMS shows up *spontaneously* in the thermal expansion (THE) when the temperature approaches from above the Curie temperature, Tc it gives an additional contribution to the lattice THE, that manifests itself as an anomaly of the HE coefficient, $\alpha^v = 3\,(1/L)(\partial L/\partial T)$. It also manifests itself in an increase of λ beyond its technical saturation, λ_S, a phenomenon called *forced* MS,where fundamental many-body electron repulsion is involved and which varies with H linearly. It can happenthat the spontaneous VMS is positive and of such a magnitude that it fully or partially cancels the lattice THE, a phenomenon known as the *invar effect*, and by which the material does not contract below Tc. Obviously this effect has enormous consequences or applications, where materials with "null" effective a are required (watches, optical interferometers, condensers, standards, etc.). Examples of invar materials are the Fe-Ni, Fe-Ni-Cr, Fe-Pt alloys, certain compositions of Fe-Ni-Mn alloys, Ni3Al, MnSi,amorphous Fe-B and intermetallics $RECo_2$, $RE_2Fe_{14}B$ and Dy_2Fe_{17} (Fig.3). Theoretically it can be shown that $\omega_{mag} \sim T^2$, precisely the same dependence as that for the lattice thermal contraction, ω_{latt} (T). Therefore compensation of the lattice THE takes plae, or in other words $\alpha\nu \approx 0$.

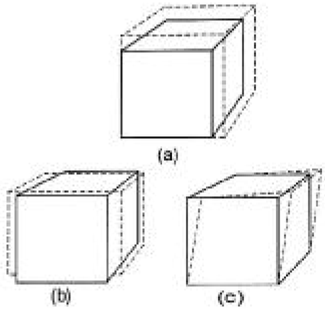

Fig 2 **Three of the possible "irreducible"MS strains for a cubic crystal (the orthorhombic one is not shown) [1].**

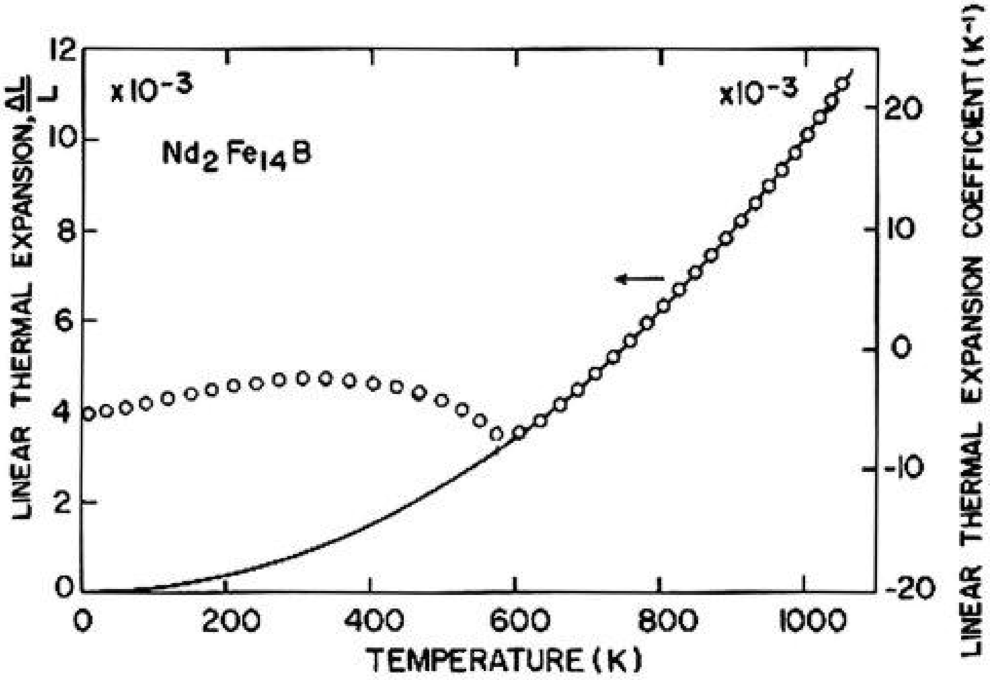

Fig 3 **Invar effect in the thermal expansion (THE) of the tetragonal hard ferromagnet $Nd_2Fe_{14}B$ .Also shown is the lattice THE extrapolation (line) [1].**

Currently vigorous research is been undertaken on MS in artificially periodic structures known as multilayers (ML) and superlattices (SL), formed by alternative deposition of thin layers ($\approx$5-10^3 Å) of two magnetic materials. ecause of the modification of the electronic structure, an *interface* MS is manifested at the interface (IF) of the two layers (in the SL the layers grow coherently, forming an artficial macroscopic lattice). Usually only one layer is magnetic (m), the m-mexchange being transmitted by the non-magnetic (nm) one. Because ML and SL are grown upon a substrate, this restrains the MS distortion, and therefore experimentally one measures the MEL stress. Néel (1954) showed that the IF MEL stress has the form B_{IF}/t_m, where t_m is the layer thickness. Therefore this stress becomes as *important* as the bulk contribution for very thin m-layers, and can be of opposite sign. When the growth is epitaxial (i.e. the lattice constants $a_m = a_{nm}$ in the SL), which can be achieve using sophisticated fabrication techniques (molecular beam epitaxy, laser ablation and sputtering), the m-layer is under mechanical stress, which induces enormous *misfit* strains (up to≈ 10%), when the bulk materials have different lattice constants. In sch a situation the magnetoelastic coupling becomes *non-linear*, and B_{vol} is modified by a stress of the form $B_{NL}\epsilon_{mf}$ (the MEL free energy is now quadratic in the strain), which can be as substantial as B_{vol}.

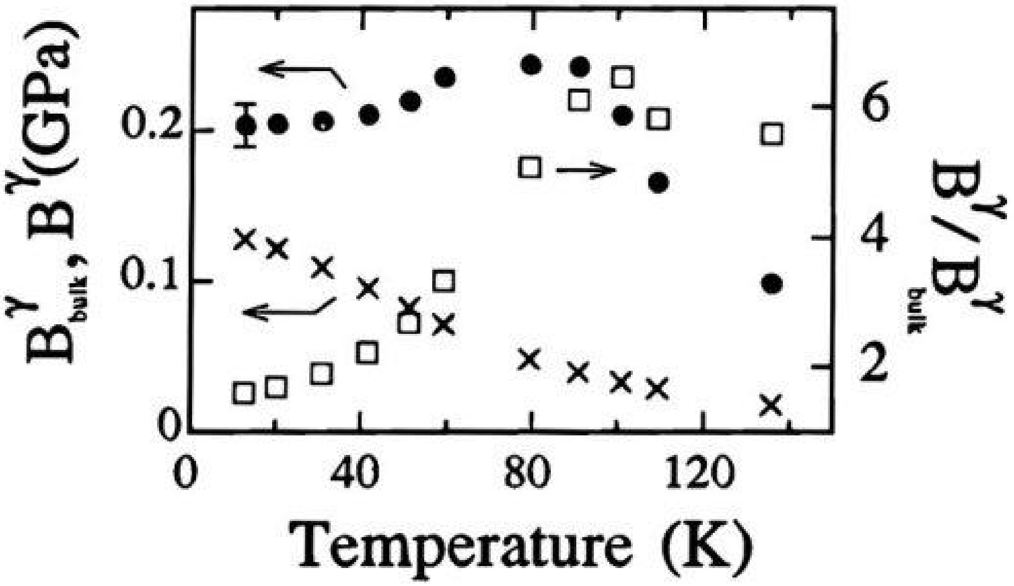

Fig 4 **Tetragonal MEL stress, B^g vs. temperature (• points) for the ultrathin hexagonal superlattice $(Ho_6/Y_6)_{x\,100}$ .The difference from the bulk Ho one, B^{γ}_{bulk}(x points) is due to interface and nonlinear MEL stresses. Also shown is the quotient B^g/B^g bulk (€ points) [1].**

The NL MEL coupling is also well manifested in te *elastic constants*, where it gives rise to the appearance of a non-symmetric or rotational strains, ω_{ij}, which differ from the usual symmetric ones ($\epsilon_{ij} = \epsilon_{ji}$) in that $\omega_{ij} \neq \omega ji$. These strains give rise to the rotational invariance of the MEL energy, in he sense that the MEL anisotropy that originates from the $\vec{M}$rotation is the same as the one produced by an imposed external rotational deformation of opposite sense. The existence of the three MEL stresses has been experimentally observed in hexagonal symetry, SL, RE/SP (SP= Y, Lu, Sc) (see Fig.4) and also in SL and ML made of cubic transition metal, such as TM/M, where TM= Fe,Co,Ni and M is a noble metal or Cu. The RE/SP SL are important from a basic point of view, because RE magnetic moments are well lcalized and CEF MS is very amenable to study. However, because RE metals are FM or helically ordered only at rather low temperatures (Gd, Dy, Tb and Ho, although more complex modulated magnetic structures do appear), they are of not much use for applicatins. Instead the ferromagnetics TM/M ML, with high Tc and weak magnetic anisotropy, may have wide application as MEL transducers (actuators and sensors) within nanostructured devices.

Fig 5 **A Terfenol magnetostrictive device for generation of seismic waves (from Handbook of Giant Magnetostrictive Materials, G. Engdahl, Academic,New York,2000).**

MS in soft transition metals and their alloys is relatively small (exprssed in 10^{-6} = 1μst units), with $\lambda_s \equiv \lambda_t \cong$ - 8, -34 for Fe and Ni, ~22 for $(FeCo)_{80}B_{20}$, and ~-30 for spinel ferrites ($TM_xFe_{1-x}O_4$, TM = Fe,Ni,Mn). An exception is the Co ferrite that has the very large value $\lambda([100], [100]) \cong -700$, where all values are at T. However, for the RE metals and their intermetallic compounds, MS is generally ***very large***, up to $\approx$ 1% (thousands of 1 μst) at low temperatures, because of the RE^{3+} ion large quadrupolar electric moment [Q ~ J(J-1/2)], which interacts with the inhomogeneus CEF, albeit this is more more shielded than in TM. The intermetallics, $RETM_2$ (TM=Fe,Co,Ni and Mn) with Laves-phase structure (the RE^{3+} form a diamond lattice), show the largest lt ever found (the so-called "giant" MS). Among these are, for example: TbN_2 (0.23% at 4.2K), NdCo2 (-0.17% at 77K) and $TbFe_2$, with the largest ***room-temperature*** λ_t known (0.2%), reaching 0.4 % at 4.2K The problem for use in applications of these intermetallics is the large saturation magnetic field required, of $\approx$ 1T. However, cobining the opposite sign anisotropies of Tb^{3+} and Dy^{3+} in the compound $Tb_{0.27}Dy_{0.73}Fe_2$ (a material commercially known as ***Terfenol***®), the saturation field is reduced down to $\approx$ 2kOe , still with a room temperature MS of 0.12%. This material is currently tha most used for MEL transducers, and can be prepared in engineering amounts (Fig.5). But the largest MS ever measured is in $TbMn_2$, where λ_t(40K, H=15T)=0.6% and , which ω_t(20K, H=15T)=-1.6%are certainly huge values. But other alloys not containing RE also how giant volume MS values of 0.5-1%, at near room temperature and at rather modest fields ($\approx$1-5 kOe), notably FeRh, $FeRh_{1-x}Pt_x$ and $Hf_{1-x}Ta_xFe_{2-y}$, when the applied magnetic field induces an AF to FM transition. This makes them also very promising for applcations.

As concluding remarks, we have seen that the most fundamental interactions in solids (exchange, CEF and spin-lattice), taken together with electronic structure, are involved in magnetostriction, and from which fundamental information can be extacted. Magnetostriction is a ubiquitous phenomenon in solid matter, spanning a wide range of values, between $\approx 10^{-8}$ (strongly correlated systems) and $\approx 10^{-2}$ (rare earth and other intermetallics). Applications in sensors and actuators, among many others, is n active reality and their use in nanostructured devices is also promising.

References

[1] A.del Moral, *Magnetostriction;Basic Principles and Applications*, Inst.of Physics Pub.,Bristol,to appear (2004) (references to original papers are given here).