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Direct X-ray observation of lattice parameter changes due to magnetostriction in nickel single crystal

Till now the measurements of magnetostriction in single crystal or polycrystalline ferromagnetic materials have been performed by electrical (capacity or resistivity variations, strain gauges), mechanical or optical methods [1, 2], i.e. through an evaluation of the macroscopic effect of dimensional alteration produced in a suitably shaped specimen by the applied magnetic field.

Since a tested and reliable X-ray diffractometric technique has recently become available, which allows the measuring of lattice parameters in single crystals with a precision at least one order of magnitude better than that attainable by the previous most sophisticated powder procedures (up to ten years ago the best tool for the purpose), we thought of trying to analyse magnetostrictive effects on a microscopic scale (changes in the unit cell dimensions) in this way.

The instrument employed was the APEX automatic precision X-ray goniometer, designed and manufactured in the UK [3] which, through the Bond's method of symmetric equivalent reflections [4], allows one to obtain in standard working conditions a precision of about 2 ppm in the measurement of Bragg angles relative to single crystal reflections.

The examined material was a high-purity (99.995%) nickel single crystal of cylindrical shape (produced by Materials Research Corporation, Orangeburg, N.Y. 10962, USA), Czochralski-grown along the [1 1 0] direction. We selected Cr$K\alpha_1$ X-radiation, with $\lambda = 2.28976 \pm 0.00002$ Å (this value derived from Bearden [5] and modified according to Deslattes and Henins [6]), because among the commonly used wavelengths it provides the highest Bragg angle pertaining to reflections from crystallographic planes parallel to the flat surface of the sample, namely about 66.78° for Ni (2 2 0), and also made allowance for the three fundamental cubic orientations.

The side to be studied was carefully polished by mechanical and chemical treatments, to remove any surface damage. Employing a cylindrical X-ray beam (generated by a fine-focus tube working at 30 kV and 30 mA) with a collimator diameter of 0.5 mm, becoming 0.8 mm on the crystal owing to the divergence, we performed several Bragg angle measurements for the (2 2 0) reflection in different points of the crystal surface fitted on a two-dimensional scanner (see Fig. 1), at first without a magnetic field and afterwards laying a suitable permanent magnet in contact with the opposite side of the crystal (local value of $H \approx 300$ Oe), and averaging five readings in each position.

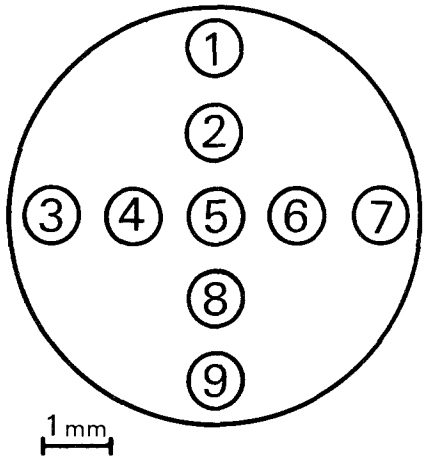

Figure 1 Schematic disposition on the Ni (1 1 0) crystal surface of different X-ray measurement points.

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The application of this magnet produced a mean magnetization value of nearly 2500 gauss* on the X-ray reflecting face of the sample, corresponding to about 40% of the saturation value ($M = 0.4M_s$, with $M_s = 6300$ gauss). In this preliminary phase of investigation, we adopted the permanent magnet as a source of magnetic field to avoid any trouble connected to sample heating: moreover the temperature within the thermal-insulating enclosure of the specimen holder was recorded during each measurement and the values of the lattice parameter a_0 converted to the reference conditions (25° C) using the linear thermal expansion coefficient of nickel (13.3 ppm $^\circ C^{-1}$ between 0 and 100° C [7]).

In regard to the correction factors applied to the a_0 values obtained with this procedure, we now recall that Bond's method [4] allows one to measure the Bragg angle θ free from errors due to sample eccentricity, absorption and zero location. The crystal tilt error (i.e. condition with the plane of diffraction not exactly parallel to the plane of angular measurement) can be adjusted instrumentally by the method of the maximum angle [8]. The wavelength dispersion error is overwhelmed by referring the λ values to peaks.

The axial divergence in our experimental arrangement produces $\Delta a_0/a_0 \sim 1$ ppm, while the angular displacement due to the Lorentz and polarization factors [9] affects a_0 for less than 2 ppm: both these errors are included in the mean standard deviation of measurements (~ 3 ppm, with no set exceeding 4 ppm). The only correction of any consequence in absolute value remains that for refraction, which involves, allowing for our diffraction geometry, an a_0 increase of more than 71 ppm.

The corrected values obtained in the different positions for lattice parameter a_0 are listed in Table I, together with the relative variations due to the magnetic field and coinciding with the magnetostriction of the sample in the particular zone, evaluated for a real penetration depth normal to the surface of about

$$\frac{2}{\pi^2}\xi_{220} = 0.75\,\mu m$$

(where ξ_{hkl} = extinction distance for (hkl) reflection).

As shown in Table I, the magnetostrictive changes of a_0 differ from point to point because of the magnetization inhomogeneity due to domain structure. By means of the well known Bitter powder technique [10], we analysed such a structure on the X-ray reflecting surface. This surface is almost parallel to a (1 1 0) plane and contains two of the easy directions of magnetization, $[\bar{1}\,1\,1]$ and $[1\,\bar{1}\,1]$: therefore, in the absence of magnetic field, the domain directions lie mainly in the same surface plane, so as to avoid the formation of free magnetic poles. For this reason they appear faintly visible at the optical microscope (see Fig. 2a), as already observed by various techniques [11, 12].

On application of a magnetic field perpendicular to the sample surface, these domains tend to rotate out of the surface plane without appreciable wall displacements, owing to the field intensity insufficient to saturate the sample; in consequence we have an increase of surface

*10^4 gauss = 1 T.

TABLE I

Position number	Lattice parameter $\bar{a}_0$ (Å) at 25° C		Relative variation (ppm)
	$M \simeq 0$	$M \simeq 0.4\,M_s$	
1	3.523898 (5)†	3.523839 (12)	−16.7
2	3.523885 (4)	3.523823 (8)	−17.6
3	3.523872 (10)	3.523804 (14)	−19.3
4	3.523867 (14)	3.523783 (13)	−23.8
5	3.523878 (7)	3.523815 (13)	−17.9
6	3.523893 (12)	3.523805 (11)	−25.0
7	3.523895 (6)	3.523823 (10)	−20.5
8	3.523904 (13)	3.523851 (9)	−15.0
9	3.523900 (9)	3.523837 (11)	−18.0
Mean values on the whole crystal surface	3.523888 (13)	3.523820 (21)	−19.3

† (Standard deviations for measured average values are reported in parentheses, expressed in units of 10^{-6} Å).

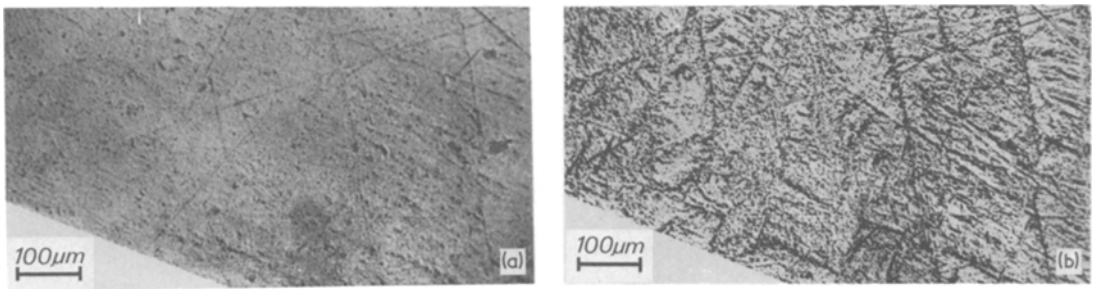

Figure 2 Optical micrographs by Bitter powder technique of representative details of domain structure: (a) without magnetic field; (b) with magnetic field applied perpendicular to the studied surface.

magnetic poles. Such an increment in the number of free poles induced by the applied magnetic field, allows a better domain observation in the powder pattern (see Fig. 2b). The reversible rotation of domain magnetic moments is connected to the magnetostrictive lattice deformation: it should be pointed out that in regard to such a deformation, the magnetization sense is unimportant; only the change in direction.

Since the penetration depth of incident radiation is less than 1 μm, only one domain inside the crystal is analysed by X-rays: however the beam diameter is about 0.8 mm and it certainly covers some domains on the surface. The measured a_0 value provides therefore an average of the lattice parameter contractions typical of several surface domains. As seen in Table I, the values determined on the various zones of crystal with $M = 0.4M_s$ give a mean magnetostriction value (at surface) of -19.3 ppm, to be compared with -35 ppm reported at saturation for [1 1 0] crystal axis [1].

These preliminary results allow us to ascertain the capability to follow directly by X-ray diffraction techniques the lattice deformations and consequent dimensional changes which occur in magnetostriction phenomena, generated by magnetization rotation in the surface domains of a sample, and it is time to modify some discouraging statements on this subject [13].

We also think that it will be possible later to link the magnetization rotation and the lattice deformation on a single domain, and to be feasible to operate properly with adequate magnetic fields on single crystals arranged so as to favour the formation of highly extended domains, i.e. comparable with the X-ray beam width necessary to have enough reflected intensity; further work is in progress.

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