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Abstract. A study is made of the reflectance anisotropy of martensitic and magnetic domains in ferromagnetic shape memory alloys (FSMA) Ni-Mn-Ga and Co-Ni-Ga. The reflectance of metallographic sections of these alloys was measured in the visible with the aid of standard inverted polarized light microscope with a 360° rotatable specimen stage. Calculations are presented for the estimation of image contrast values between neighboring martensite twins. Qualitative and quantitative observations and angular measurements in reflected polarized light proved to be useful for the analysis of specific features of the martensite microstructure of multiferroic materials.

1. Introduction

Ferromagnetic shape memory Heusler-type alloys Ni-Mn-Ga and Co-Ni-Ga are a relatively new class of ferroic materials which may change their shape and magnetization by a magnetically induced martensite transformation or by a magnetically induced reorientation of martensite twin structure [1-3]. The behaviour of these materials is determined by the magnetoelastic interaction of correlated ensembles of structural (martensite) and ferromagnetic domains. The details of this interaction in real materials depend on a large number of microstructural parameters and crystal defects [4-6]. In this view it is important to gain detailed experimental information on both static configurations of coexisting martensite and magnetic domain structure (DS) and their dynamic behavior under the action of external magnetic, mechanical and thermal influences.

The studies of the martensite and magnetic domain structure (DS) were attempted by a number of techniques including Bitter patterns [7,8], interference optical microscopy [9], magnetooptic indicator films (MOIF) [10-13], scanning electron microscopy [14], magnetic force microscopy [15], Lorentz transmission electron microscopy and electron holography [16], and some others.

In spite of rather large number of publications the problem of DS observation appeared to be more difficult than expected, so the necessary experimental data are still lacking. This is explained partly by the fact that in addition to the known limitations of existing DS techniques [17] the FSMA put forward a number of additional problems. First of all one faces the situation of revealing in a single experiment the coexisting DS's of different physical origin – martensitic and magnetic. Another problem is the proper preparation of defect-free metallographic sections [4-6]. Even if the FSMA

surface is successfully planarized by thorough polishing it readily becomes corrugated during martensite variant rearrangement. Among other peculiarities is the "ghost" martensite-like residual relief observed by optical contrast in a sample heated well above the martensite-austenite phase transition point where no martensite should exist (S J Murray [18]). One more unexpected feature mentioned by Y.Ge et al. is the formation of a relief delineating the magnetic DS and making it observable in ordinary optical microscope even without the use of polarized light illumination [19].

Thinking about the further development of martensite and magnetic DS observation techniques for FSMA our attention was attracted by the polarized light studies of twin structure of optically anisotropic YBaCuO high temperature superconductors [20]. In the present work we examine the possibility of exploiting this technique to the case of MSMA.

2. Experimental

Polycrystalline samples of the Ni-Mn-Ga and Co-Ni-Ga alloys examined earlier by the method of MOIF [12] were prepared by arc melting in argon followed by vacuum annealing. The composition of the alloys was determined by X-ray energy dispersive microanalysis (Oxford Instruments). Various abrasive, chemico-mechanical and electrolytic polishing were employed to prepare the metallographic sections of the samples. The martensite and austenite start and finish temperatures were estimated by the temperature dependence of AC initial susceptibility. The angular variations of the reflectivity data were measured in the visible with the aid of a silicon photodiode or a CCD video camera (integral and local light intensity studies, respectively) in conjunction with a light polarizing microscope with a 360° rotatable specimen stage.

3. Results and discussion

Figure 1 shows the low magnification grain structure image of a longitudinal cross section of polycrystalline $Ni_{2.12}Mn_{0.88}Ga$ button prepared by ordinary metallographic abrasive polishing routine. The angular orientation of the sample on the specimen stage is arbitrary while analyzer and polarizer of the microscope are set at nearly crossed position. The existence of uniaxial crystallographic texture is clearly evidenced by the elongation of the grains in a direction perpendicular to the copper hearth. Rotation of the sample results in a complicated interplay of the image contrast between the individual grains and (at higher magnifications) martensite twins. At the same time the polarized light observations generally lead to a great enhancement of the images of noninformative artifacts (scratches, etch pits, pores) which may remain unseen under bright field illumination.

Figure 1. Grain structure of the longitudinal cross section of polycrystalline $Ni_{2.12}Mn_{0.88}Ga$ button prepared by arc melting on a copper hearth observed in polarized light

To understand the mechanism of optical polarization contrast formation it is convenient to consider a plane parallel plate splitted into two martensite domains with different optical indicatrices having ellipse projections on the plane of observation with the same semiaxes n' and n'' but different orientation. For such an optical system there is a known solution given by Born and Wolf [21]:

$$I_{1,2} = I_0 \left(\cos^2 \theta - \sin(2\varphi \pm \chi)\sin(2\varphi \pm \chi - 2\theta)\sin^2\frac{\delta}{2} \right), \tag{1}$$

where θ is the orientation of the transmission axis of the analyzer with respect to the plane of the incident light, and δ is the phase difference between the output rays, χ is the angle between the large semiaxes of the ellipses. For a plate of thickness h the phase difference is $\delta = 2\pi h (n'' - n')/\lambda$, where λ is the light wavelength. From this the contrast C is written as

$$C \equiv \frac{I_1 - I_2}{I_1 + I_2} = \frac{\sin(4\varphi - 2\theta)\sin 2\chi \sin^2\frac{\delta}{2}}{\left(\cos 2\theta - \cos(4\varphi - 2\theta)\cos 2\chi\right)\sin^2\frac{\delta}{2} - 2\cos^2\theta} \qquad (2)$$

It is seen that C is zero for the angles φ and θ related as $\theta = 2\varphi - (\pi n/2)$, thus resulting in fourfold change of the contrast with the rotation of the analyzer by 360° and eight-fold changes for the sample rotation by the same angle:

$$C \equiv \frac{I_1 - I_2}{I_1 + I_2} = \frac{\sin(4\varphi - 2\theta)\sin 4\omega \sin^2\frac{\delta}{2}}{\left(\cos 2\theta - \cos(4\varphi - 2\theta)\cos 4\omega\right)\sin^2\frac{\delta}{2} - 2\cos^2\theta} \cdot \qquad (3)$$

To check the predictions of the calculations detailed angular measurements of the light intensities were performed experimentally for a pair of adjacent martensitic variants (twins). Figure 2 shows a local area of a flat sample surface containing the twin made visible by polarized light observation.
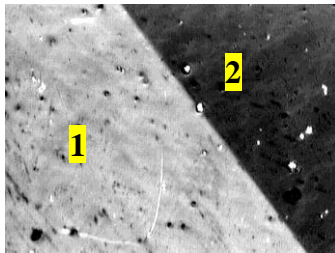

Figure 2. The optical polariztion contrast during observation of martensite twin in polarized light

The experiments have shown that the contrast between the martensite variants labeled above as 1 and 2 may varies in a regular way. The detailed data of the measurements made in a localized field of view separately for the two adjacent martensite variants are presented by the graphs of figure 3.
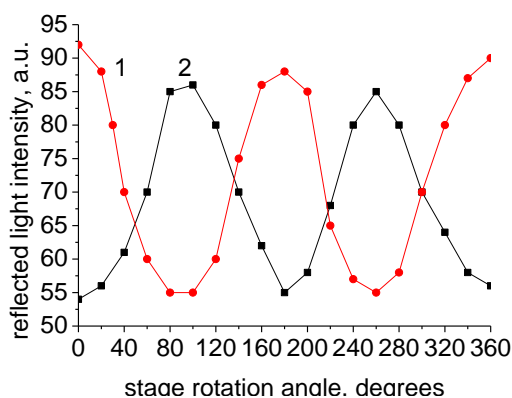

Figure 3. Dependence of the intensity of the polarized light reflected from two adjacent martensite variants of a $Ni_{2.16}Mn_{0.84}Ga$ sample (curves 1 and 2). The points of curves intersection correspond to the condition of zero contrast between the variants.

It is seen from figure 3 that the optical contrast $C = (I_1 - I_2)/(I_1 + I_2)$ may be set to zero, be maximized or inverted simply by rotation of the sample on the microscope stage. It is also evident that the symmetry of this optical effect differs from that of the magnetooptic polar Kerr or Faraday effect which are invariant to sample rotation. This feature opens the way of enhancing the conditions for both martensite and magnetic domain structure observations by the digital methods of differential

polarized microscopy [17, 22]. The differential procedure of [17, 22] starts with a digitized image of the magnetically saturated state. This domain-free image serves as a background which is subtracted from a state containing domain information, so that in the difference image a clear micrograph of the domain pattern is obtained. This procedure is not applicable to FSMA because of great difficulties in obtaining the saturation. Alternatively, for FSMA our data show that it is possible to produce two images of the same field of view and invert the contrast of one of them through rotation of the sample by an angle multiple to 45°. In this way it becomes possible to form a differential pair of images. The resulting effect of such background subtraction is shown figure 4.
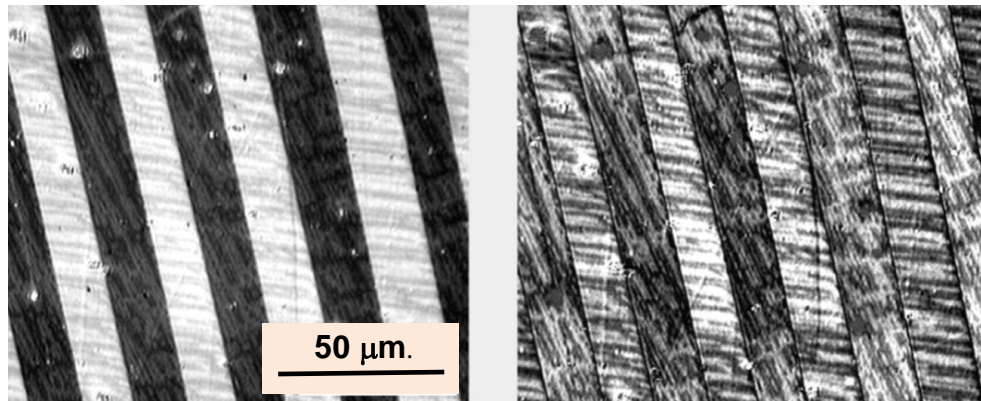

Figure 4. Polarized optical image of martensitic and magnetic domain structure on a flat surface of $Co_2Ni_{0.85}Ga_{1.15}$ sample. (*a*) original image, (*b*) after background compensation

The left part of figure 4 demonstrates the original image of $Co_2Ni_{0.85}Ga_{1.15}$ sample with band structure of martensite variants and hardly discernible internal structure. Background subtraction (at the right) demonstrates many-fold increase in the contrast of this internal structure bringing into evidence its relation to the magnetic DS of 180° degree type which is invariant to sample rotation.

4. Conclusion

Polarized light microscopy was applied to study the martensite twin domain structure of ferromagnetic shape memory alloys of the Heusler type. It is shown that due to anisotropic character of the reflection of light in these alloys the polarization optical contrast depende on a number of factors among which is the angular orientation of the sample on the microscope stage. By proper choice of these factors the optical contrast of martensite twins may be either maximized or set to zero or intermediate values. This behaviour is in contrast to that of the magnetooptic polar Kerr effect which is invariant to the rotation of the sample about the normal to the plane of observation. This peculiarity opens ways for manipulating the contrast of polarized light images by their segmentation and using the principles of differential polarized microscopy.

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