

# Transformation of twinned $\text{Ni}_{52.0}\text{Mn}_{24.4}\text{Ga}_{23.6}$ martensite in a rotating magnetic field: Theory and experiment

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The influence of a rotating magnetic field on disk-shaped twinned Ni-Mn-Ga single crystals is studied theoretically and experimentally. A magnetoelastic model of ferromagnetic martensite is used for the comprehension of experimental results. The model considers the magnetic field influence on each twin component in terms of the magnetically induced mechanical stress (magnetostress). The angular dependence of magnetostress and the correspondence between the directions of the magnetic field and magnetization vector are obtained. The magnetically induced transformation of twin structure of the specimen is observed experimentally in a  $\text{Ni}_{52.0}\text{Mn}_{24.4}\text{Ga}_{23.6}$  single-crystalline disk by magnetic measurements performed in a two-dimensional vibrating sample magnetometer. The threshold character of the transformation process is stated. The threshold angles between the [100] crystallographic direction and the directions of magnetic field and magnetic vector of the transformed twin component were measured for the different magnetic field values. The comparison of experimental values with the theoretical ones points to the comparatively low value of the magnetic anisotropy constant ( $50 \text{ kJ m}^{-3}$ ). The obtained results disclose the possibility of obtaining large magnetically induced strains in ferromagnetic-shape memory alloys with reduced magnetocrystalline anisotropy.

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## I. INTRODUCTION

Ferromagnetic-shape memory effect (FSME) typically exhibited by Ni-Mn-Ga Heusler alloys is due to magnetic field-induced twinning/detwinning in the martensitic phase, resulting in recoverable strains on the order of the martensitic spontaneous distortion (up to 10%).<sup>1-6</sup> Abnormally soft crystal lattice and sufficiently large ordinary magnetostriction are crucial ingredients of FSME.<sup>7-12</sup>

By comparing the mechanical and magnetic energies necessary to detwin the martensitic structure, it is possible to establish an equivalence between the applied magnetic field  $\mathbf{H}$  and mechanical stress  $\sigma$ .<sup>13-17</sup> The equivalent stress (magnetostress) value  $\sigma_{\text{eq}}(\mathbf{H})$  can be expressed through a magnetic anisotropy constant<sup>14,15</sup> or a constant of magnetoelastic coupling.<sup>7,13,16</sup> So far, only stress that is induced by the field applied in the  $\langle 100 \rangle$  crystallographic direction was measured<sup>15-19</sup> and evaluated theoretically.<sup>13-16,18</sup> This is, probably, because in practice the actuation of FSME element is usually performed by the orthogonal application of magnetic field and mechanical stress. Nevertheless, the alternative contactless loading of the FSME element by a rotating magnetic field appeared to be a useful tool to study the magnetomechanical performances of these materials.<sup>18-23</sup> However, the complementary measurements of magnetostress in rotating magnetic field and the corresponding theoretical analysis have not been carried out yet.

In the present paper, theoretical and experimental results related to the action of a rotating magnetic field on the twin structure of ferromagnetic Ni-Mn-Ga martensite are presented. The generalization of the concept of magnetostress to

this situation is proposed. The magnetostress values are estimated for the different orientations of the magnetic field in the twinned single crystal. A universal criterion for the start of the martensite reorientation, applicable to an arbitrarily oriented magnetic field, is derived and applied to the magnetic measurements that reflect the reorientation process.

## II. THEORETICAL CONSIDERATIONS

Disk-shaped specimens are considered with the magnetic field applied in the disk plane ( $XOY$  coordinate plane) so that the magnetostatic contribution to the free energy is isotropic with respect to rotation in the disk plane and does not need to be taken into account. Let the twin structure of martensite be formed by two alternating variants of the tetragonal crystal lattice with the principal axes ( $c$  axes) aligned with the [100] and [010] (cubic) crystallographic directions, lying along the  $x$  and  $y$  coordinate axes, respectively. These are the equilibrium directions for the unit magnetic vectors of the variants:  $\mathbf{m}^{[100]}(\mathbf{H})$  and  $\mathbf{m}^{[010]}(\mathbf{H})$ . The transformation of the twin structure is caused by the difference in diagonal stress components  $\sigma(\mathbf{H}) = \sigma_{xx}(\mathbf{H}) - \sigma_{yy}(\mathbf{H})$ , which breaks the physical equivalence of the martensite variants and induces the twin boundary motion. A magnetically induced stress  $\sigma(\mathbf{H})$  arises due to the rotation of the magnetic vectors. A magnetic field applied parallel to [100] or [010] direction rotates the magnetic vector of only one of the martensite variants. Therefore, the average magnetostress value is equal to  $\sigma(\mathbf{H})/2 \equiv \sigma_{\text{eq}}(\mathbf{H})$  (for more details see Ref. 16). The generalization of this formula to the case of an arbitrarily directed field is

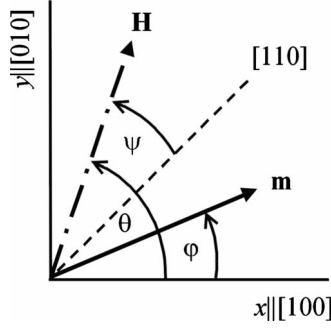


FIG. 1. Definition of angular variables used for the problem solution.

$$\sigma(\mathbf{H}) = \frac{1}{2}[\sigma^{[100]}(\mathbf{H}) + \sigma^{[010]}(\mathbf{H})], \quad (1)$$

where

$$\sigma^{[100]}(\mathbf{H}) = \sigma_{xx}^{[100]}(\mathbf{H}) - \sigma_{yy}^{[100]}(\mathbf{H})$$

and

$$\sigma^{[010]}(\mathbf{H}) = \sigma_{xx}^{[010]}(\mathbf{H}) - \sigma_{yy}^{[010]}(\mathbf{H})$$

are the stresses induced in the  $x$  and  $y$  variants with  $c\|[100]\|x$  and  $c\|[010]\|y$ , respectively. These stresses depend on the values of the dimensionless magnetoelastic constant  $\delta$ , the magnetization  $M$ , and the angles  $\varphi^{[100]}(\mathbf{H})$  and  $\varphi^{[010]}(\mathbf{H})$  between the  $x$  axis and the unit magnetic vectors of the  $x$  and  $y$  variants, respectively,<sup>13,16</sup> (see Fig. 1). Hereafter the magnetically induced stress is expressed as

$$\sigma(\mathbf{H}) = 6\delta M^2\{\cos[2\varphi(\mathbf{H})] - \cos[2\varphi(0)]\}, \quad (2)$$

where  $\varphi(\mathbf{H}) \equiv \varphi^{[100]}(\mathbf{H})$  stands for the  $x$  variant and  $\varphi(\mathbf{H}) \equiv \varphi^{[010]}(\mathbf{H})$  for the  $y$  variant. The functions  $\varphi^{[100]}(\mathbf{H})$  and  $\varphi^{[010]}(\mathbf{H})$  can be obtained by a minimization of the magnetic energy

$$F = \pm K_u m_x^2 - (\mathbf{m} \cdot \mathbf{H})M, \quad (3)$$

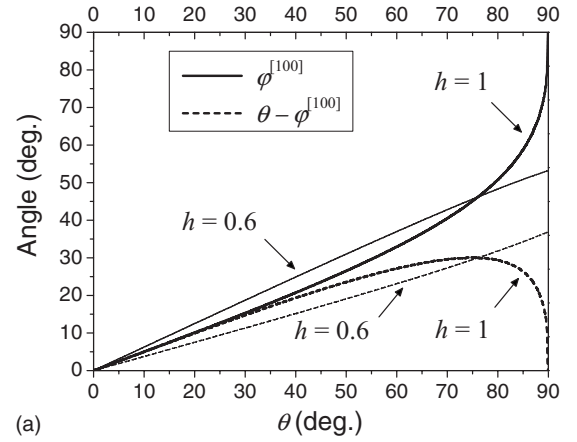
where  $K_u$  is the uniaxial anisotropy constant.

The anisotropy constant  $K_u$  is positive and so the sign “+” in Eq. (3) applies to the  $y$  variant and the sign “−” to the  $x$  variant. In this case  $\mathbf{m}^{[100]}(0)\|x$  and  $\mathbf{m}^{[010]}(0)\|y$ , as it was assumed above.

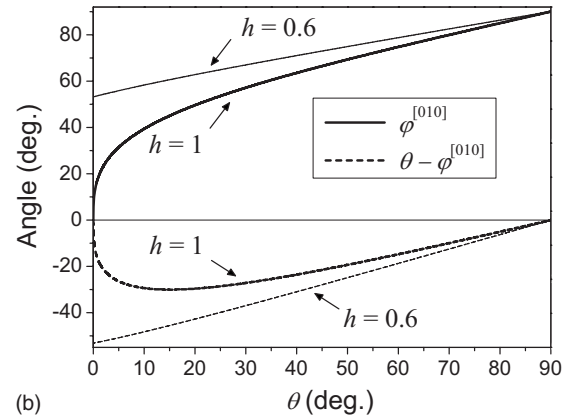
Now, the equilibrium directions of the magnetic vectors can be found from the condition  $\partial F/\partial\varphi=0$ , which results in the equation

$$\sin 2\varphi \pm 2h \sin(\theta - \varphi) = 0, \quad (4)$$

where  $h=H/H_A$  denotes a dimensionless field,  $H_A=2K_u/M$  is the anisotropy field. When  $H=0$ , the Eq. (4) has the solutions  $\varphi(0) \equiv \varphi^{[100]}(0)=0$  and  $\varphi(0) \equiv \varphi^{[010]}(0)=\pi/2$ , corresponding to magnetic moment oriented along  $[100]$  in the  $x$  variant and along  $[010]$  in the  $y$  variant. The solutions computed for nonzero magnetic field values are shown in Fig. 2. In a saturating magnetic field,  $h=1$ , applied along the  $[100]$  or  $[010]$  direction (i.e., when  $\theta=0^\circ$  or  $\theta=90^\circ$ , respectively) the magnetic moment vector of each variant is aligned with the field. The magnetic vector can be strictly parallel to the



(a)



(b)

FIG. 2. Solid lines show the angles between the  $[100]$  direction and magnetic vectors of (a)  $x$  variant and (b)  $y$  variant of martensite for the different orientations of magnetic field. Dashed lines show the angles between the magnetic vectors of martensite variants and magnetic field. The curve  $h=1$  is characterized by a fast continuous change before it terminates at  $\varphi^{[100]}=90^\circ$  and  $0^\circ$  in the graphs (a) and (b), respectively.

magnetic field only when the latter one is directed along  $\langle 100 \rangle$  directions, in all other cases, the magnetic vector approaches asymptotically the field direction, but never reaches it. The difference  $\theta - \varphi$  is positive for the  $x$  variant, and negative for the  $y$  variant. It means that the vector  $\mathbf{m}^{[100]}$  moves behind  $\mathbf{H}$  when the latter rotates from the direction  $[100]$  to the  $[010]$  and the vector  $\mathbf{m}^{[010]}$  stays ahead of the rotating field.

Now, the Eqs. (2) and (4) enable the computation of the equivalent stress value as a function of the angle  $\psi$  between the magnetic field and the  $[110]$  direction, that was our main purpose in this section. The equivalent stress functions computed for different magnetic field values, using a magnetoelastic constant  $\delta=-23$  and a magnetization value  $M=0.5$  T, typical for Ni-Mn-Ga alloys, are shown in Fig. 3. When  $\psi=\pm 45^\circ$ , the magnetic field is parallel to  $[010]$  (upper sign) or  $[100]$  (bottom sign) and the magnetostress is maximum. The largest magnetostress is limited by magnetic saturation and therefore the value  $\sigma(\pm 45^\circ) \approx 3.5$  MPa cannot be further increased by applying magnetic fields larger than  $h=1$ .

The martensite reorientation starts when the axial stress reaches the threshold value  $\sigma_{th}$ , i.e., when

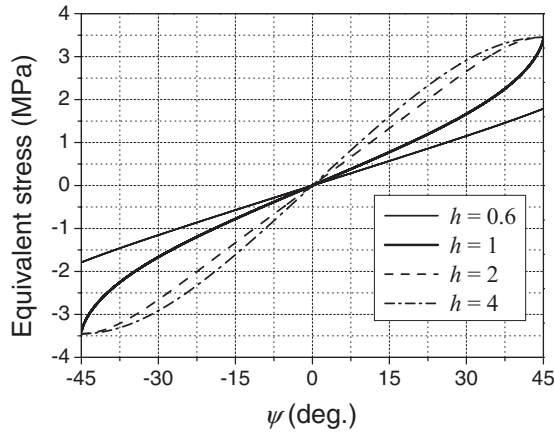


FIG. 3. Magnetostress value as a function of the magnetic field direction.

$$\sigma(\mathbf{H}) = \sigma_{th}. \tag{5}$$

The threshold stress value depends on the alloy composition, defects, and thermomechanical treatment. Twinned Ni-Mn-Ga specimens with  $\sigma_{th} \sim 1$  MPa are typical (see, e.g., Refs. 7, 16, and 24).

In the majority of published works, the threshold value of magnetic field, which provides the fulfillment of Eq. (5), was measured or theoretically evaluated only for a magnetic field aligned with one of the principal  $\langle 100 \rangle$  crystallographic directions. Now the problem has been fully explored for a rotating magnetic field (or rotating specimen).

The theoretical model can be used for the comprehension of experiments with rotating specimen. Let the single-crystalline disk-shaped specimen be preliminary “trained” to create a twinned state with a dominant  $x$  variant and a residual  $y$  variant of martensite. The application of the magnetic field in the  $[100]$  direction will not change this state noticeably but field or specimen rotation will start the transformation of the  $x$  variant into a  $y$  variant ( $x \rightarrow y$  transformation) when the rotation angle  $\theta$  reaches the threshold value predetermined by the Eq. (5). The threshold value of the rotation angle and the corresponding angle between the  $[100]$  direction and the magnetic vector of the  $x$  variant are plotted in Fig. 4 as a function of  $h$ . The values  $K_u = 200$  kJ m<sup>-3</sup> and  $\sigma_{th} = 1$  MPa were chosen for computations. The anisotropy constant corresponds to an anisotropy field  $H_A = 635$  kA m<sup>-1</sup> (see, e.g., Ref. 7 and references therein) whereas the threshold stress is typical for alloys with a large magnetostrain<sup>1,2,16</sup> although substantially larger values were also observed.<sup>15,25</sup>

Figure 4 shows that if the rotating field is smaller than  $0.55H_A$ , it cannot induce the  $x \rightarrow y$  transformation even when the field is perpendicular to the easy axis of the  $x$  variant since the magnetostress never exceeds the  $\sigma_{th}$ . Above  $0.55H_A$ , the larger the magnetic field is, the earlier the  $x \rightarrow y$  transformation starts. Note that the threshold value of the angle between the magnetic vector of the  $x$  variant and the  $[100]$  direction depends weakly on the field. Such dependence is due to the fact that the equivalent stress of Eq. (1) depends on the orientation of the magnetic vectors in both  $x$  and  $y$  variants. If the second summand in Eq. (1) is omitted,

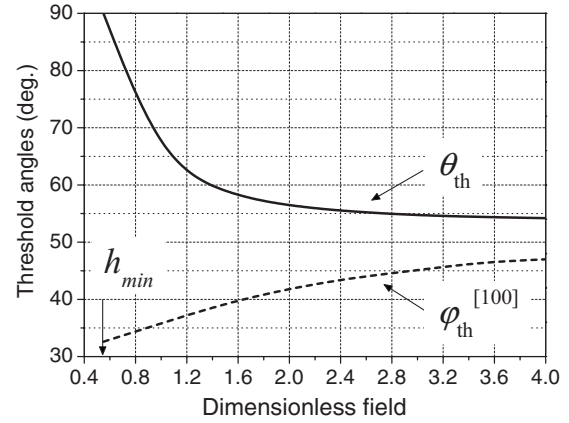


FIG. 4. Threshold values of the angles between the  $[100]$  direction and magnetic field vector ( $\theta_{th}$ , solid line), as well as between the  $[100]$  direction and the magnetic vector of the  $x$  variant of martensite ( $\varphi_{th}^{[100]}$ , dashed line). These values correspond to the start of the transformation of  $x$ —into  $y$  variant and depend on the magnetic field magnitude.

the condition  $\sigma^{[100]}(\mathbf{m}^{[100]}) = 2\sigma_{th}$  will predetermine the unique direction of the magnetic vector.

Once a dominating variant has been selected in the specimen, for instance, the  $y$  variant, Eqs. (1), (2), and (5) can be used to determine the maximum misalignment of a saturating magnetic field with respect to  $[100]$  direction in order to get variant reorientation. In this case the  $y \rightarrow x$  transformation can be started only if the angle between the field vector and the  $[100]$  direction does not exceed some critical value, which depends both on the threshold stress and magnetic field values. The critical angle values are shown in Fig. 5 as the functions of threshold stress. These graphs correspond to the equivalent stress functions shown in Fig. 3, and were computed using the aforementioned values of saturation magnetization and magnetoelastic constant.

The magnetically induced transformation of the twin structure is possible in the wide range of angles  $0 \leq \theta < 30^\circ$  if the absolute value of the threshold stress is less than 1.6

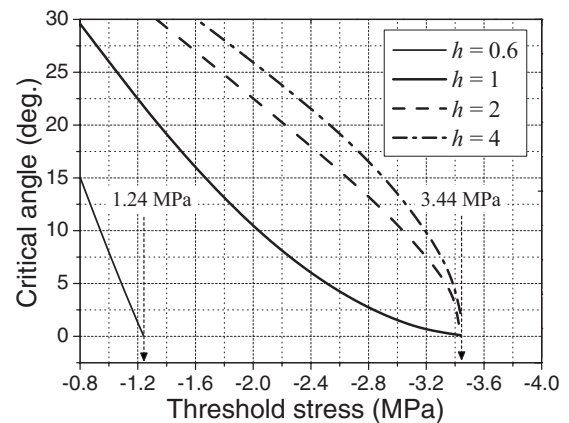


FIG. 5. Critical magnitudes of the angle  $\theta_{th}$  between the magnetic field and  $[100]$  direction, which render impossible a magnetically induced reorientation of martensite variants in the specimens with different threshold stresses. Different curves correspond to different values of dimensionless field values.

MPa (for  $h=4$ ), 1.3 MPa (for  $h=2$ ), and 0.8 MPa (for  $h=1$ ). However, the transformation is impossible if the threshold stress exceeds 3.44 MPa. All curves plotted in Fig. 5 for the fields exceeding the saturation value finish at the same threshold stress because this stress can be created only by the field applied along the  $[100]$  direction (see Fig. 3). In this case the magnetic saturation is completed at  $h=1$  and further increase of the field does not change the saturated value of magnetostress. Smaller threshold values of magnetostress can be overcome by a magnetic field directed outside the  $[100]$  direction. In this case the magnetic saturation state is not attainable so, the magnetostress smoothly grows even for  $h > 1$ .

### III. EXPERIMENT

The experiments have been carried out using the  $\text{Ni}_{52.0}\text{Mn}_{24.4}\text{Ga}_{23.6}$  disk-shaped sample of 5-mm diameter and 1.5-mm thickness which was cut from a single crystal studied in Ref. 16. This crystal was chosen because it exhibited a highly mobile twin structure in the stress-strain experiments.<sup>16</sup>

The single-crystalline sample was oriented as to present the  $[100]$  and  $[010]$  crystallographic directions in the disk plane and the  $[001]$  direction out of plane. The magnetization measurements have been made by a two-dimensional vibrating sample magnetometer.<sup>26</sup> Before each magnetic measurement, a quasingle-variant state with the easy magnetization axis aligned with the  $[100]$  crystallographic direction was prepared by a proper mechanical training of the specimen. Then, the specimen was rotated in a constant magnetic field (case A). In the second sequence of measurements, the specimen was magnetized at chosen angles with respect to the easy magnetization axis of the selected variant (case B). The magnetic vector direction was determined by the measurement of the parallel- and perpendicular-to-the-field components of the magnetic moment.

The magnetic field induces an equivalent mechanical stress in the crystal and starts the transformation of the dominating martensite variant into the other one when the increasing magnetostress reaches the threshold value. In the case A the magnetostress increases due to the deviation of the magnetic vector of the dominating  $x$  variant from the  $[100]$  axis and the threshold stress value is reached at the certain (threshold) value of the deviation angle  $\varphi^{[100]}$ . The experimental threshold values of this angle are shown in Fig. 6 together with the appropriate values of the angle  $\theta$  (see also Fig. 1).

The experiment shows that the magnetically induced martensite reorientation is possible only if the magnetic field exceeds the value  $H_{\text{start}} \approx 130 \text{ kA m}^{-1}$ . For the minimal field, the threshold angles are  $\theta_{\text{th}} \approx 78^\circ$  and  $\varphi_{\text{th}}^{[100]} \approx 56^\circ$ . Theoretically, the starting field is minimal if  $\theta_{\text{th}} = 90^\circ$  (due to the obvious physical reasons explained in the Sec. II). The discrepancy between the theoretical and experimental values may be partially caused by the fact that the magnetic field value was changed in the course of experiment with the rather large step of  $\Delta H > 50 \text{ kA m}^{-1}$ , and so, the  $H_{\text{start}}$  value was slightly underestimated and the initial segment of the

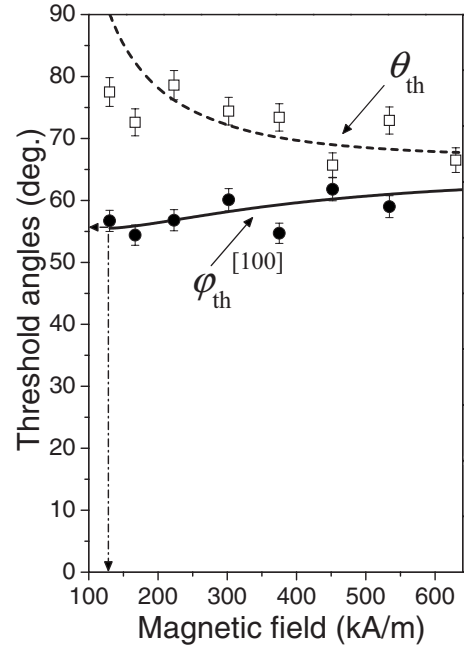


FIG. 6. Experimental (symbols) and theoretical (lines) dependences of the threshold angles determined for the different values of magnetic field. The vertical arrow shows the minimal field value, which can start the martensite reorientation; the horizontal one indicates the angle between the field and magnetic vector in the minimal field.

$H$ - $\theta$  curve was omitted consequently. Using the Eq. (4), a theoretical value of the rotation angle  $\varphi^{[100]}$  can be fitted to the value of  $56^\circ$  by the appropriate choice of the parameter  $2h = H_{\text{start}}M/K_u$  without any reference to the threshold stress. Physically, it means that the rotation angle value is prescribed by the competition of the external magnetic field with the magnetic anisotropy field. The best fit was observed for the magnetic anisotropy constant  $K_u = 50 \text{ kJ m}^{-3}$  determined using value  $M = 0.48 \text{ T}$  measured in the present work. The threshold stress value was obtained, then, from the condition  $\sigma_{\text{th}} = \sigma(H_{\text{start}})$  and proved to be equal to 2.3 MPa. Finally, the dependences of the threshold angles on the external magnetic field were computed from Eqs. (4) and (5) indicating a reasonable agreement with the experimental data (see Fig. 6).

In the case B the  $x \rightarrow y$  martensite reorientation is caused by the magnetic field directed along  $(\pi/2) - \theta$ . The martensite reorientation manifests itself through a jump of magnetization observed when the applied field reaches the value  $H_{\text{start}}$  and through the difference in the initial slope of the magnetization curves during increasing and decreasing the applied magnetic field. Figure 7(a) illustrates that this difference is almost equally pronounced for the deviation angles from  $0^\circ$  to  $20^\circ$ . Further deviation of the field from the  $[010]$  direction results in the quick convergence of the forward and reverse magnetization curves which denotes the absence of variant reorientation.

The above estimated magnetic anisotropy constant makes possible the conversion of magnetic field values to dimensionless field  $h$ . Figure 7(b) shows the dependence of magnetic moment on the dimensionless field computed for the



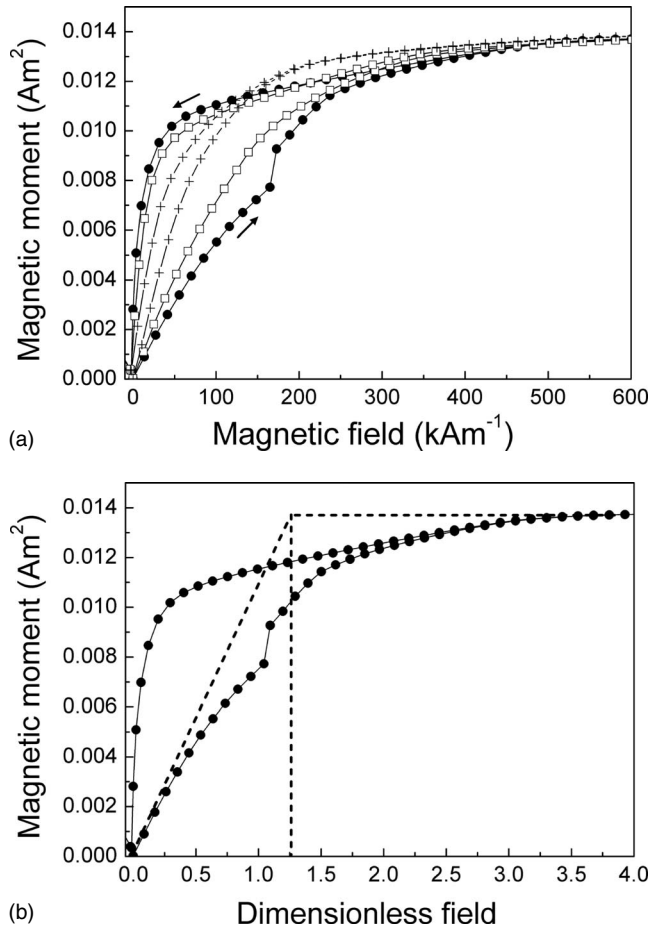


FIG. 7. (a) Magnetization loops measured under the field directed at the angle  $\theta=0^\circ$  (circles),  $20^\circ$  (squares), and  $30^\circ$  (crosses) to the  $[100]$  crystallographic directions. (b) Magnetization loop measured in the field aligned with the  $[100]$  direction and plotted versus the dimensionless magnetic field,  $h$ . The dashed lines illustrate the method of graphic determination of the saturating field value.

zero value of the deviation angle. This dependence confirms the reasonability of the estimation of the magnetic anisotropy constant: the graphic determination of the saturating dimensionless field results in the value  $h=1.25$  which is rather close to the  $h=1$  prescribed by the Eq. (4). This procedure can be considered as a tentative estimation since the magnetization curve exhibits so-called “technical saturation,” typi-

cal for the ferromagnets possessing microstructure. In spite of the uncertainty in the value of saturating field, it is seen now that the maximum magnetic field magnitude of  $600 \text{ kA m}^{-1}$  used to magnetize the sample corresponds to  $h=4$ . Figure 5 shows that at  $h=4$ , the martensite with threshold stress  $\sigma_{th}=2.3 \text{ MPa}$  can be reoriented even when the deviation angle exceeds the value of  $20^\circ$ . This conclusion is in agreement with Fig. 7(a) where at  $\theta=20^\circ$  the variant reorientation is still present and vanishes between  $20^\circ$  and  $30^\circ$ .

#### IV. DISCUSSION AND CONCLUSION

In the present work, we have studied the influence of a rotating field on the reorientation of martensitic twin variants in a Ni<sub>52.0</sub>Mn<sub>24.4</sub>Ga<sub>23.6</sub> FSM single crystal. We have demonstrated that the experimental results with the disk-shaped sample of the aforementioned alloy are satisfactorily described by the properly upgraded magnetoelastic model of ferromagnetic martensite elaborated in Refs. 13–16. The theoretical treatment of the experimental results leads to a reasonable estimation of the twinning stress ( $\sigma_{th} \approx 2.3 \text{ MPa}$ ) and points to the comparatively low value of the magnetic anisotropy constant  $K_u \approx 50 \text{ kJ m}^{-3}$ . This value corresponds to the magnetic anisotropy field  $H_A \approx 160 \text{ kA m}^{-1}$ , which is about one-fourth of the values reported for prismatic specimens (see, e.g., Refs. 27 and 28). On the other hand, this estimation is close to the values that were obtained for Ni-Mn-Ga films by a ferromagnetic resonance method.<sup>29–31</sup>

It may be concluded that a magnetostress, which exceeds the value of  $2.3 \text{ MPa}$ , was induced in the Ni-Mn-Ga single crystal with a reduced constant of magnetocrystalline anisotropy. This result (i) points to the expediency of the purposeful search for compositions and thermomechanical treatment of the FSM alloys, which could provide a further reduction in the magnetic anisotropy constant and (ii) indicates the possibility of observing a large magnetically induced strain in low magnetic fields by using FSM samples with enhanced twinning stress.

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