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Magnetic domain structure in a ferromagnetic shape memory alloy $Ni_{51}Fe_{22}Ga_{27}$ studied by electron holography and Lorentz microscopy

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Behaviors of magnetic domains with cooling in a $Ni_{51}Fe_{22}Ga_{27}$ ferromagnetic shape memory alloy were examined by electron holography and Lorentz microscopy. A peculiar meshy pattern was observed in the Lorentz microscope image of the parent phase, being concurrent with the anomaly in the thermomagnetization curve. The meshy pattern was found to stem from the heavily bent lines of magnetic flux. The dramatic change in the magnetic domains is presumably due to some intrinsic magnetic instability that is pronounced by cooling, rather than a phenomenon triggered by the lattice modulation as the precursor effect of martensitic transformations or formation of the intermediate phase as observed in other systems. © *2003 American Institute of Physics.*
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Ferromagnetic shape memory alloys (SMAs) have attracted considerable attentions of researchers because of the potential applications to actuators driven by the applied magnetic field. There are several ferromagnetic SMAs developed so far; Ni_2MnGa,[1–3] Ni_2MnAl,[4,5] Co_2NiGa,[6,7] Co–Ni–Al,[7–9] Fe–Pd,[10,11] Fe–Pt,[12] etc. Recently, some of the present authors reported an alloy system Ni–Ga–Fe with superior performance.[13,14] $Ni_{73-x}Ga_{27}Fe_x$ ($20<x<22$) alloys exhibit martensitic transformations from the $L2_1$ parent phase to the 14 or 10 M martensite. Both T_C (Curie temperature) and M_s (martensitic transformation start temperature) can be controlled by the heat treatment for the quenched specimens, via enhancement of the $L2_1$ order, as well as the choice of appropriate composition. Moreover, the ductility in the polycrystalline state can be dramatically improved by introducing the γ phase into grain boundaries. Besides these aspects, this alloy is also attractive from a viewpoint of the transformation mechanism. Figure 1 shows a thermomagnetization curve (magnetization versus temperature curve) of a $Ni_{51}Fe_{22}Ga_{27}$ alloy subjected to the heat treatment at 773 K. The magnetization monotonously increases upon cooling in the parent phase as observed in other ferromagnetic SMAs. However, the magnetization is depressed in the wide temperature range between room temperature and M_s (see the inset of Fig. 1) although it has not yet reached to the saturated magnetization. The nature of this anomaly is not well understood yet, although this is an essential problem relevant to both the fundamentals and applications of the Ni–Ga–Fe alloys. The purpose of the present work is to examine the temperature dependence of the magnetic domains in a $Ni_{51}Fe_{22}Ga_{27}$ alloy by electron holography and Lorentz microscopy.

The $Ni_{51}Fe_{22}Ga_{27}$ alloys were heat treated at 1473 K to obtain the homogeneous single phase and then quenched into ice water. The specimens were subsequently heat treated at 773 K for 1 h to develop the $L2_1$ order in the parent phase followed by quenching into ice water. M_s is about 172 K, which was determined by differential scanning calorimetry. T_C was evaluated at 369 K, which was defined as the minimum point of the temperature derivative of magnetization in the thermomagnetization curve. The magnetic domains were observed by Lorentz microscopy and electron holography[15] using a transmission electron microscope JEM-3000F, to which a special pole piece producing a low magnetic field (0.2 mT at the specimen position) was attached.

Figure 2 provides change in the Lorentz microscope image upon cooling, where the magnetic domain walls are visualized as dark or bright lines. Large magnetic domains

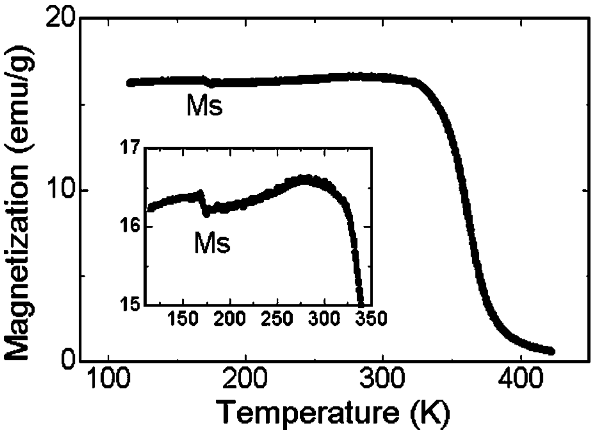

FIG. 1. Thermomagnetization curve for a $Ni_{51}Fe_{22}Ga_{27}$ alloy subjected to the heat treatment at 773 K for 1 h, measured in the magnetic field of 1000 Oe. The inset shows an enlarged part, which manifests the depressed magnetization between room temperature and M_s.

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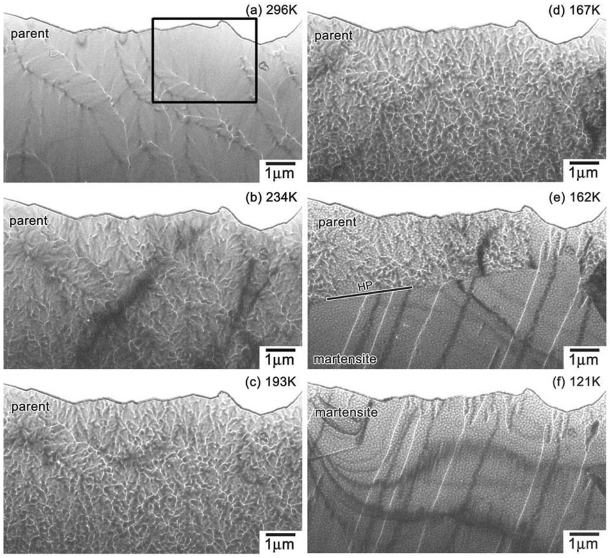

FIG. 2. Change in the Lorentz microscope image with cooling. The martensitic transformation occurs at 162 K in this field of view.

exceeding 1 μm are observed at 296 K [Fig. 2(a)] as in the case of other ferromagnetic SMAs.[9,10,16–18] The magnetic domains appear to be subdivided into finer parts (100–200 nm) with cooling to 234 K resulting the peculiar meshy pattern [Fig. 2(b)]. The subdivision proceeds with further cooling to 193 K [Fig. 2(c)]. The Lorentz microscope image at 167 K [Fig. 2(d)] is similar to that of 193 K. At 162 K, the martensite has formed in the lower part of Fig. 2(e), where the habit plane separates two distinct magnetic domain structures. The martensite shows usual plate-like magnetic domains as displayed in Fig. 2(f). The reverse transformation upon heating (from 121 to 296 K) proceeded in a converse fashion to that of the forward transformation as described earlier.

Figure 3 provides the reconstructed phase images of the holograms obtained from the rectangular area as shown in Fig. 2(a). The white lines represent the lines of magnetic flux projected along the incident electrons. The arrows indicate the directions of lines of magnetic flux. The lines of magnetic flux are smooth inside the large magnetic domain at 296 K [Fig. 3(a)], although they are steeply bent near the magnetic domain walls. The lines of magnetic flux become fluctuated with cooling to 234 K [Fig. 3(b)]. The fluctuation is much pronounced at the lower temperature 193 K [Fig. 3(c)]. Thus, it is found that the meshy pattern of the Lorentz microscope images originates from the heavily bent lines of magnetic flux. Here we mention the process of the change in the magnetic microstructure, focusing on the lines of magnetic flux within the circled area in Fig. 3. Inside the circle 1, the lines of magnetic flux are almost straight at 296 K, but they are heavily bent at 234 K. The feature of the lines is almost unchanged by the subsequent cooling to 193 K. By contrast, inside the circle 2, the lines are not strongly modified by cooling from 296 to 234 K, but they show prominent change by the subsequent cooling to 193 K. The observation indicates that the fluctuated lines are stable once they are produced by cooling, and the subsequent cooling makes the unmodified area fluctuated. This process continues until the whole area of the specimen is filled with the fluctuated lines. Since the image of Fig. 3(d) is similar to that of Fig. 3(c), the fluctuation is presumably completed at 193 K in this field of view. At 162 K, where the interface between the parent phase and the martensite is represented by the white broken lines, the fluctuated lines in the parent phase is somewhat modified due to the accommodation of magnetic flux between the parent phase and the martensite with distinct magnetocrystalline anisotropy [Fig. 3(e)]. In fact the lines of magnetic flux are almost straight in the martensite with the higher anisotropy [Fig. 3(f)], being in sharp contrast to those in the parent phase.

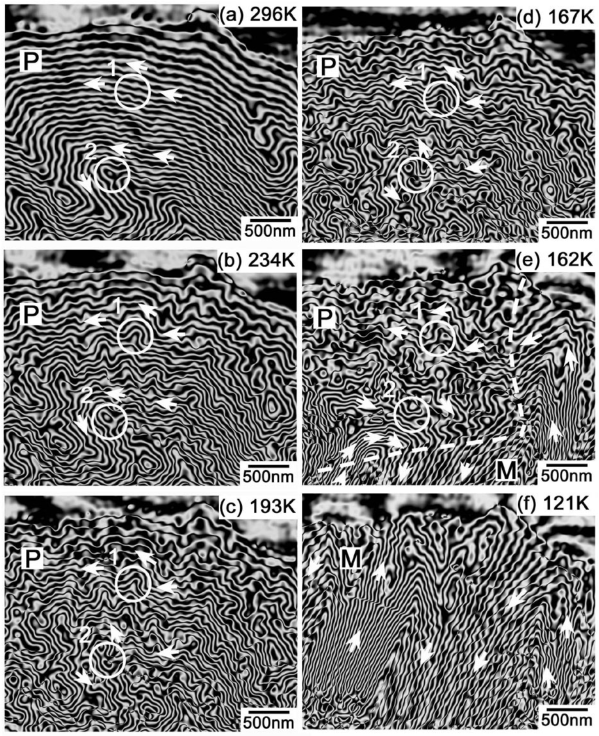

FIG. 3. Reconstructed phase images observed at (a) 296, (b) 234, (c) 193, (d) 167, (e) 162, and (f) 121 K. *P* and *M* stand for the parent phase and martensite, respectively.

Several SMAs exhibit lattice modulation in the parent phase near M_s, which is called the precursor effect of martensitic transformations.[19–21] In $\mathrm{Ni_2MnGa}$ alloys, an intermediate phase that has a distinct lattice symmetry to the martensite is produced above M_s.[3,21] These phenomena can be detected as definite change in the electron diffraction patterns. However, in the present specimen, the electron diffraction pattern did not show remarkable temperature dependence between room temperature and M_s. Thus, it is reasonable to consider that the dramatic change in the magnetic domains is due to some magnetic instability, which is enhanced by cooling, rather than a phenomenon triggered by the pronounced lattice modulation (development of the precursor effect) or the formation of an intermediate phase. At this stage, identification of the magnetic instability is difficult. But it should be noted that the size of the antiphase domains in the $L2_1$-ordered parent phase (about 200 nm)[14] is comparable to that of the meshy pattern observed in the Lorentz microscope images (Fig. 2). This microstructure (antiphase domains) may play an important role to stimulate the magnetic instability that is manifested with cooling. In fact the anomaly of the thermomagnetization curve becomes obscure if the population of the antiphase domains decreases.[14]

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One notices weak spotty contrast in the martensite (Fig. 2), which may be also related with the modulation of magnetization distribution by the microstructure, but careful observations are likely to be necessary to rule out any artificial effects. Note that the fluctuation of the lines of magnetic flux (Fig. 3) is an intrinsic phenomenon, since this is concurrent with both the change in the Lorentz microscope image and the suppression of magnetization.

To summarize, the anomaly in the thermomagnetization curve (suppressed magnetization in the parent phase) is accompanied by the formation of the peculiar meshy pattern in the Lorentz microscope image. This meshy pattern is found to originate from the heavily bent lines of magnetic flux. The dramatic change in the magnetic domain structure is thought to be due to some magnetic instability that is pronounced by cooling, rather than a phenomenon triggered by the pronounced lattice modulation (the precursor effect of martensitic transformations) or formation of the intermediate phase as observed in a Ni_2MnGa alloy.

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