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THE INFLUENCE OF HIGH TEMPERATURE PLASTIC DEFORMATION ON MAGNETIC PROPERTIES OF Ni_2MnGa TYPE SINGLE CRYSTALS

WPŁYW WYSOKOTEMPERATUROWEGO ODKSZTAŁCENIA PLASTYCZNEGO NA WŁASNOŚCI MAGNETYCZNE MONOKRYSZTAŁÓW TYPU Ni_2MnGa

The work presents experimental results on the influence of high temperature plastic deformation on the magnetic properties of Ni_2MnGa type single crystals. It has been shown that room temperature plasticity of the Ni_2MnGa single crystals is limited to very small strains (lower than 10^{-2}), whereas at 1073K these single crystals could be plastically compressed to strains greater than $5x10^{-1}$. Magnetic properties of deformed and undeformed samples have been studied in low and high magnetic field. The analysis of the obtained magnetic hysteresis at different temperatures indicate a significant impact of the plastic deformation on the coercive field and the remanent magnetization (an increase by a factor of 3) and a lack of the effect on the saturation magnetization.

Keywords: Ni_2MnGa single crystals, plastic deformation, magnetic properties

W pracy przedstawiono wyniki doświadczalne dotyczące wpływu wysokotemperaturowej deformacji plastycznej na własności magnetyczne monokryształów typu Ni_2MnGa. Wykazano, iż plastyczność badanych monokryształów Ni_2MnGa w temperaturze otoczenia jest ograniczona do bardzo małych odkształceń, nie większych niż 10^{-2}, natomiast w 1073K badane monokryształy można ściskać do odkształceń większych niż $5x10^{-1}$. Przeprowadzono badania własności magnetycznych w małym oraz w dużym polu na próbkach przed i po procesie wysokotemperaturowego ściskania. Analiza pętli histerezy magnetycznej uzyskanych przy różnych temperaturach wskazuje na istotny wpływ odkształcenia plastycznego na zwiększenie wartości pola koercji oraz namagnesowania resztkowego (około trzykrotne) i jego brak na wartość namagnesowania nasycenia.

1. Introduction

The intermetallic Ni_2MnGa single crystals show giant magnetostriction effect which may reach the value of about 6% of linear strain [1]. In the polycrystalline alloy this effect is much smaller, because random distribution of crystallographic orientations of grains strongly reduces the deformation anisotropy on a macroscopic level. Recently, there have been much attention paid to the possibility of forming this alloy by plastic deformation at elevated temperatures, since at room temperature even the Ni_2MnGa single crystal becomes brittle due to the presence of well ordered internal structure. The mechanical investigation that was carried on compressed Ni_2MnGa single crystals at room temperature show the appearance of cracks already formed at about 10^{-2} of relative reduction of sample height [2]. Till now, high temperature plastic deformation and its effect on the crystal structure and magnetic properties has been studied only on Ni_2MnGa polycrystals, where, as it was mentioned above, the magnetostriction effect is very limited already in the undeformed samples. Moreover, high temperature plastic deformation cannot form strong texture in polycrystalline material, and therefore it is unable to cause the deformation anisotropy. The present work brings experimental results on the influence of high temperature compression on the mechanical and magnetic properties of Ni_2MnGa type single crystals. The temperature of deformation (1073K) has been chosen above the temperature of the order-disorder transformation, which in these alloys, depending on the chemical composition, is in the range of 823K - 1071K [3].

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2. Experimental procedure

Polycrystalline ingots of Ni_2MnGa were obtained in arc furnace from pure elements of nickel, manganese and gallium (Ni-99,999%, Mn-99,98%, Ga-99,999%). They were re-melted three times in order to get good homogeneity before the single crystal growth. Single crystals of a shape of rectangular prism of dimensions 3x3x30 mm^3 were grown in a resistance furnace with a natural vertical temperature gradient, at a vacuum better than 10^{-5} hPa. Crystals were grown inside a boron nitride crucible which is non-reactive to the used elements. Chemical composition of the as grown crystals (Ni-51,6%, Mn-28,3%, Ga-20,1%) was checked using the Hitachi S-3400N scanning electron microscope equipped with an x-ray spectrometer, EDS. The crystallographic orientations of the main axis (compression direction, CD) and the two lateral surfaces, (LS1 and LS2) were checked by the Bruker/Siemens D8 Advance x-ray diffractometer (see Fig.1). The compression tests of the single crystals with dimensions 3x3x4.5 mm^3 were performed at 1073 K and also, for comparison, at room temperature using Instron 5566 testing machine. The initial strain rate was set to $10^{-4}s^{-1}$. The measurements of magnetic properties of the samples before and after compression at 1073 K were performed in the temperature range between 4K - 400 K using a Quantum Design physical properties measuring system (PPMS) equipped with a 9 Tesla superconducting magnet.

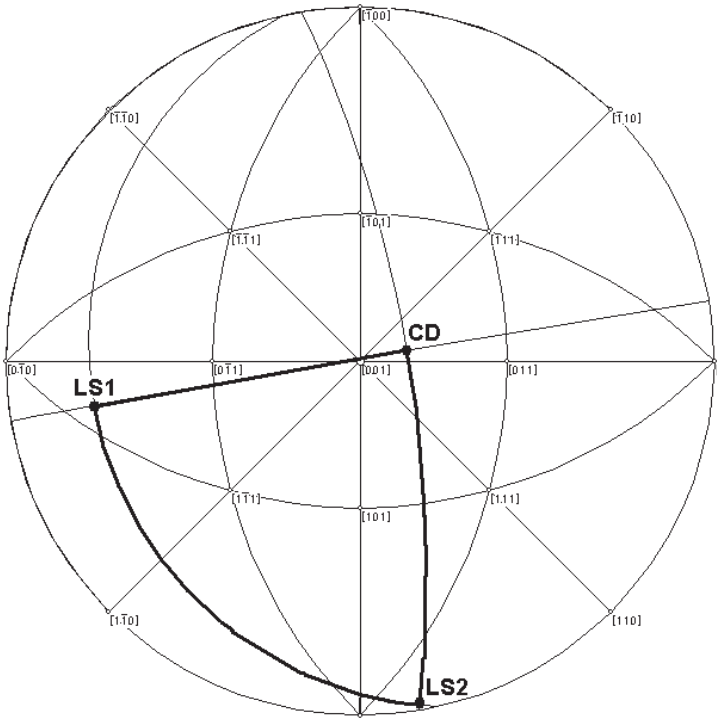

Fig. 1. Crystallographic orientation of the undeformed sample shown on a stereographic projection

3. Results and discussion

3.1. Mechanical properties

A comparison between mechanical properties of the investigated Ni_2MnGa single crystals compressed at 1073K and at room temperature is presented in figure 2. It is worth noting that due to the presence of a martensite structure the initial 293K stress-strain curve reveals in a range of the applied stress between 25MPa - 50MPa an intermediate region of both of elastic and plastic deformation of martensite which seperates the primary and secondary elastic regions of the tested sample. Measuring of the sample height before and after the compression up to the applied stress of 250 MPa it was established that the total sample deformation was not greater than $5x10^{-3}$. Further increase of the stress magnitude up to 1GPa generates plastic instabilities which lead rapidly to the sample failure [2]. The stress – strain curves obtained at 1073K show quite different mechanical properties of the Ni_2MnGa single crystals in comparison with those obtained at room temperature. As it was already mentioned the 1073K compression tests were performed above the temperature range, where the order – disorder transformation of the $L2_1 \rightarrow B2'$ type is to occur [3]. In this case the material plastic deformation

begins already at the applied stress of 40MPa and accordingly to the classical hot compression properties of f.c.c. structures after reaching maximum stress of about 50MPa the steady state flow continues at somewhat lower stress level. The 1073K mechanical characteristics of the Ni_2MnGa single crystals compressed up to the level of true plastic strains of about $5x10^{-1}$ is shown in figure 3.

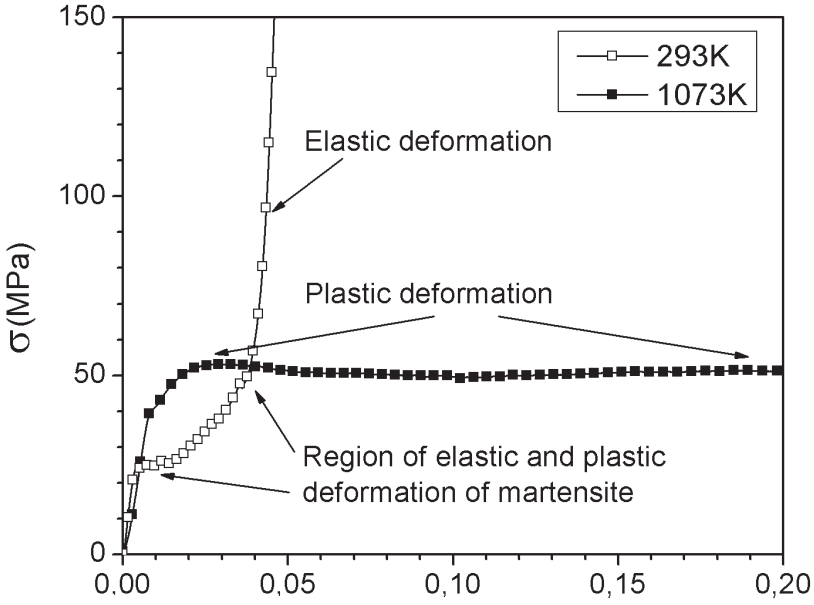

Fig. 2. Stress – strain curves of the compression tests performed at room temperature and 1073K with specific deformation regions marked

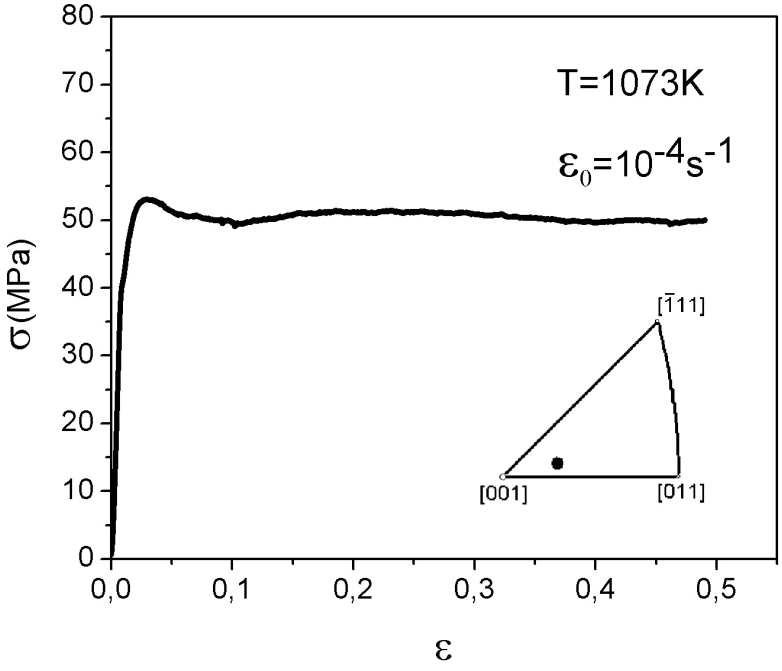

Fig. 3. The 1073K stress – strain curve of the Ni_2MnGa single crystals deformed plastically up to the total plastic strain of 5 x 10^{-1}. Insert shows the initial orientation of the single crystal located within the basic stereographic triangle

3.2. Physical properties

3.2.1. Specific heat measurements

The undeformed sample was measured in with the relaxational method in the Ouantum Design PPMS apparatus in the temperature range 4-400K. The results are plotted in Fig. 4. The dominant peak at 333.4K is attributed to the martensitic transition. Smaller peaks at 345.6K and 353.6K, coinciding with the anomalies at

the temperature dependence of the low field magnetisation discussed below are attributed to the pre-martensitic transformation and Curie point, T_C respectively.

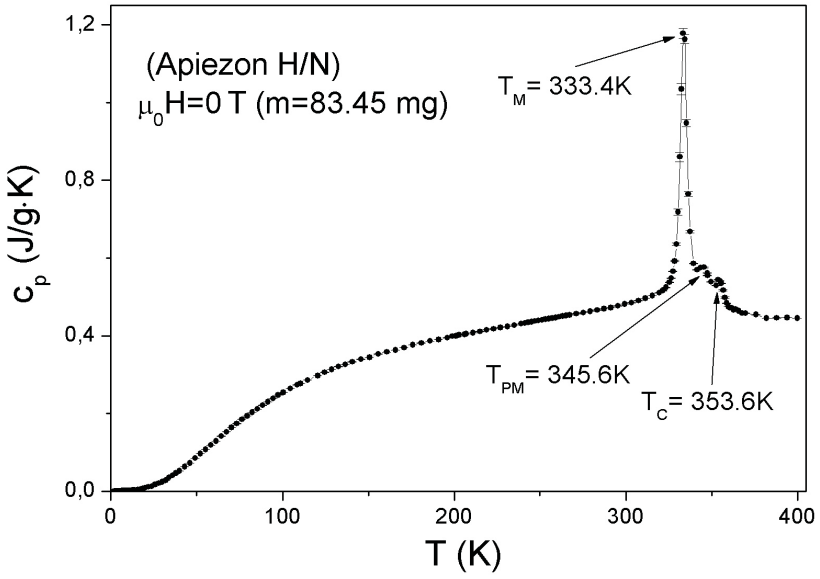

Fig. 4. Temperature dependence of the specific heat of the undeformed sample in the range of 4- 400K

3.2.2. Low field magnetisation measurements

Low field (0.01 T) magnetisation of the samples has been measured in the 4-400K range in the zero field cooled (ZFC) state and field cooled state (FC). The samples were aligned with their [1 -6 2] axes along the direction of the applied field. The results are plotted in figures 5 and 6.

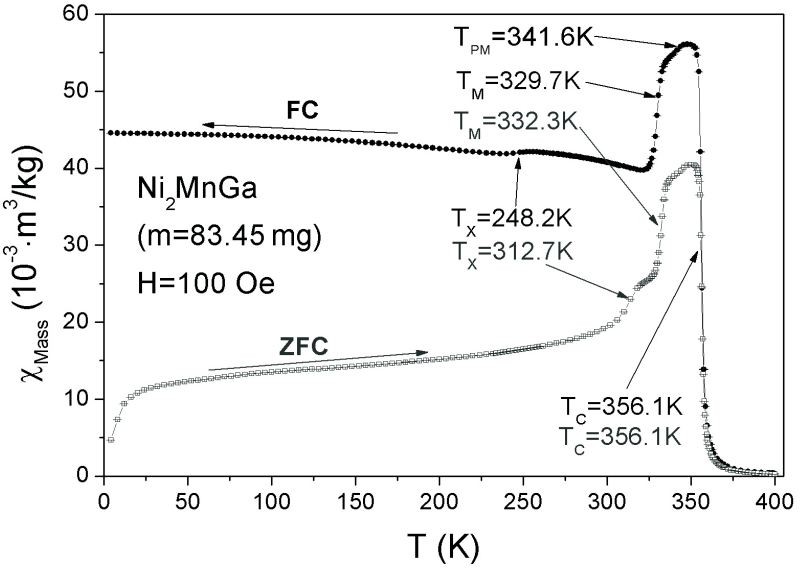

Fig. 5. Temperature dependence of the magnetization in low field (0.01 T) measured in the zero field cooled (ZFC) and the field cooled (FC) state for the undeformed sample

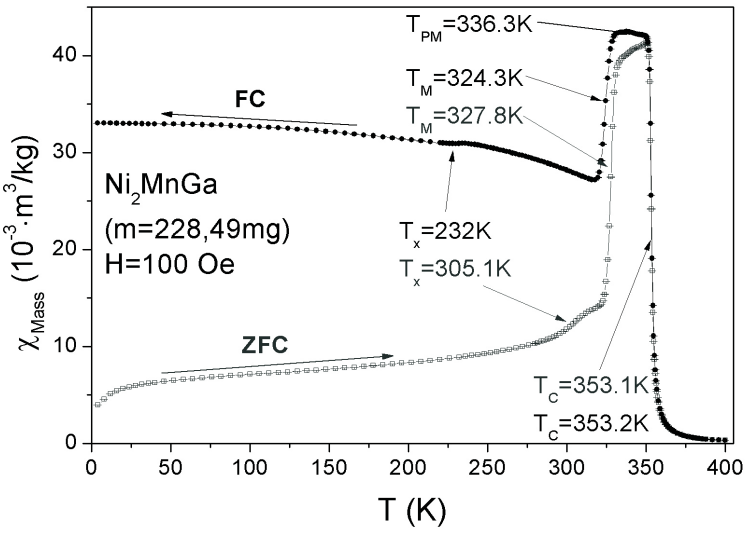

Fig. 6. Temperature dependence of the magnetization in low field (0.01 T) measured in the zero field cooled (ZFC) and the field cooled (FC) state for the deformed sample

The characteristic points of the curves of the pristine and deformed samples correspond respectively to their T_C of 356.1 K and 353.1 K, the pre-martensitic transformation, 341.6 K and 336.3 K and martensitic transition, 329.7 K and 324.3 K. An additional characteristic point corresponding to a kink of the magnetisation curve at 248.2 K and 232 K can possibly be attributed to the modulation of the new-created martensitic structure [4].

It is worth noting that the low field magnetisation which is a measure of the initial susceptibility is larger in the high temperature cubic phase than in the martensite region. Plastic deformation 5 x 10^{-1} causes an overall decrease of the initial susceptibility by a factor of 0.75 and the ratio of the susceptibilities of the cubic and martensitic phases changes from 1.4 before, to 1.6 after the deformation.

A comparison of the ZFC and FC curves shows that they diverge considerably in the martensitic phase, whereas they are similar for the cubic phase. This indicates a much larger coercivity of the martensitic phase than that of the cubic one, which will be discussed in more details in the next paragraph concerning the high field magnetisation measurements.

3.2.3. High field magnetisation measurements

Measurements of the magnetisation versus field (up to 9 T) of the two samples have been carried out near the characteristic points of the low field magnetisation curve, at the temperatures (see Fig.7 and 8). The samples were aligned with their [1, -6, 2] axes along the direction of the applied field, as in the low field measurements.

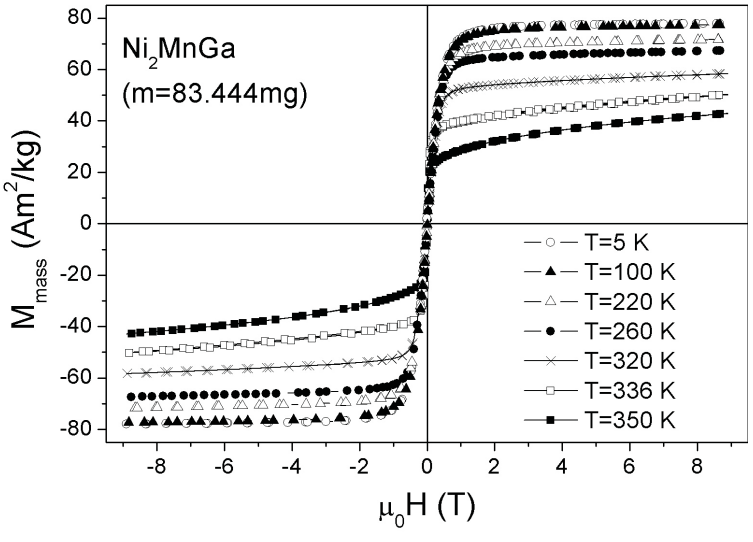

Fig. 7. Hysteresis loops at different temperatures for the undeformed sample

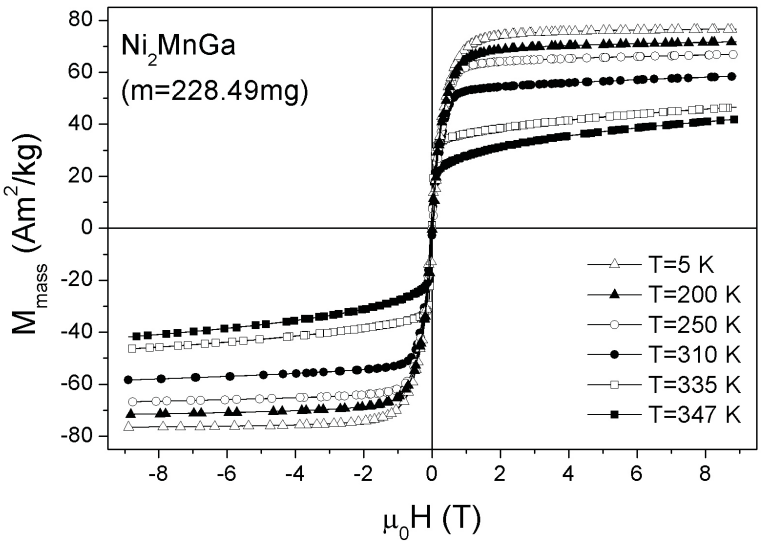

Fig. 8. Hysteresis loops at different temperatures for the deformed sample

Virgin magnetisation curves have been measured after de-magnetising samples by heating them up to 400 K, i.e. above their T_C values. Subsequently, after measuring each virgin curve, the hysteresis loops were measured. The enlarged part of the diagrams around their centres are shown in figures 9 and 10. The negative curvature of the initial parts of the virgin curves shows that the dominating mechanism of the coercivity is the domain wall pinning.

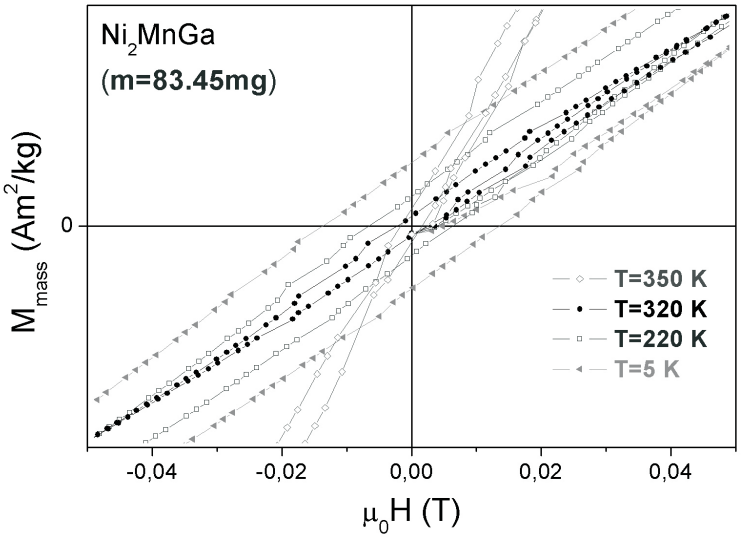

Fig. 9. Virgin magnetization curves and hysteresis loops at different temperatures for the sample before deformation

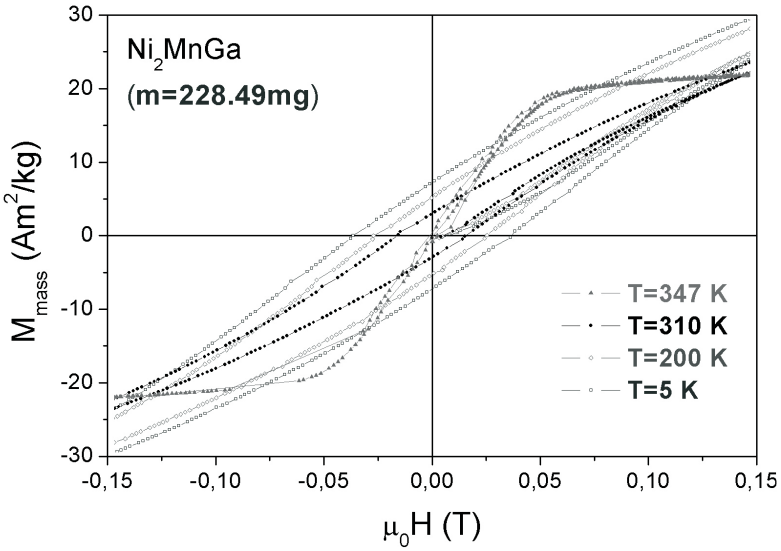

Fig. 10. Virgin magnetization curves and hysteresis loops at different temperatures for the sample after deformation

The temperature dependences of the characteristic parameters of the hysteresis loops, i.e the coercive force, B_c and remanent magnetisation, B_r are presented in figure 11 and 12. It shows a large difference in the coercivity and remnance between the cubic and the martensitic phases and their considerable increase upon deformation. Although the coercivity generally decreases with increasing temperature and vanishes at T_C, a step in its temperature dependence ocurring between 320 K (martensite) and 340 K (cubic phase) shows that it is considerably lower in the cubic phase. Also the remanent magnetisation is much lower in the cubic phase, which is mostly related to the difference of the coercivity.

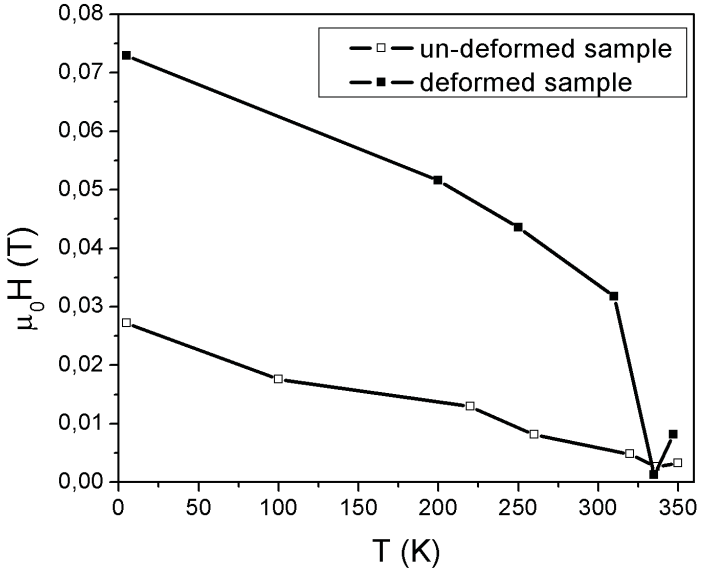

Fig. 11. Coercive field vs. temperature curves for the undeformed and deformed samples

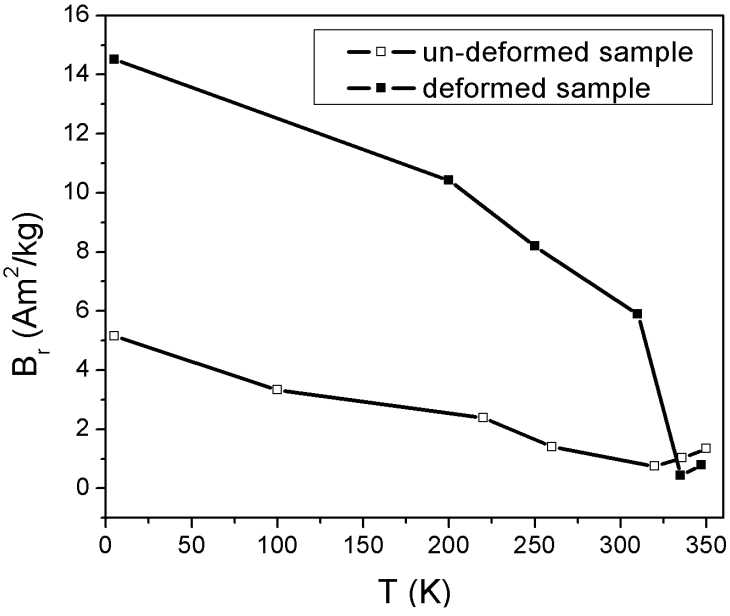

Fig. 12. Remanent magnetization vs. temperature curves for the undeformed and deformed samples

A 3 fold overall increase of the coercivity upon deformation can be attributed to the introduction of dislocations, which number is dependent on the square of the shear stress. They act as the pinning sites and the coercive force is proportional to the square root of their number. Thus, comparing the coercive forces in the martensite phase for the pristine sample and the deformed sample one arrives by an order of magnitude increase of the number of dislocations upon deformation.

As the magnetisation curves nearly saturate at the highest applied field, 9 T the corresponding magnetisation values could be taken as these of saturation magnetisation. Except for the overall decrease of the saturation magnetization with increasing temperature, as expected for a Brillouin dependence, a difference between the cubic and martensitic phases can be noticed. This indicates different values of manganese moments in the two phases or a possible appearance of a magnetic moment on the nickel atoms.

4. Conclusions

On the basis of the obtained experimental results, the following conclusions can be drawn:

(i) The critical temperatures of the martensitic and another pre-martensitic transformation of the undeformed Ni_2MnGa single crystals are after the relaxational method measurements 333.4K and 345.6K respectively, whereas the Curie point Tc is at 353.6K. The low field magnetisation measurements confirm fully the above results. The differences observed between the FC and ZFC data show additionally the phase transformation hysteresis, which is most significant for the Tx temperature.

(ii) The mechanical effect of martensite deformation revealed on the 293K compression curve within the applied stress region from 25MPa to 50 MPa (Fig.2) also confirms that the TM temperature for the tested single crystals is above room temperature.

(iii) The Ni_2MnGa single crystals, very brittle at room temperature, become very plastic at 1073K and soft, with the yield point of about 50 MPa, as well as, no plastic strain hardening effect is also observed.

(iv) Large plastic strain of 5 x 10^{-1} causes an overall decrease of the initial magnetic susceptibility of the Ni_2MnGa single crystals by a factor of 0.75 and the ratio of the susceptibilities of the cubic and martensitic phases changes from 1.4 before, to 1.6 after the deformation.

(v) The negative curvature of the initial parts of the virgin magnetization curves (Figs. 9,10) suggest that the dominating mechanism of the coercivity of the single crystals is the domain wall pinning.

(vi) The high field magnetization measurements revealed that the value of the coercive field and the remanent magnetization changes about three times due to the plastic strain of the Ni_2MnGa single crystals. Since the coercive force is assumed to be proportional to the square root of the dislocation density,

therefore the number of dislocations is expected to increase by an order of a magnitude.

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