Ferromagnetic resonance in Ni-Mn based ferromagnetic Heusler alloys

Aksoy S., Posth O., Acet M., Meckenstock R., Lindner J., Farle M., Wassermann E.F.
Experimentalphysik AG-Farle, Universität Duisburg-Essen, 47048 Duisburg, Germany
E-mail: seda.aksoy@uni-due.de

Abstract.
Ferromagnetic Ni-Mn based Heusler alloys undergo martensitic transformations leading to properties such as magnetic shape memory, magnetic field induced strain and magneto-caloric effects. The occurrence of such effects are closely related to the nature of magnetic interactions around the transition. These interactions can be closely examined by the ferromagnetic resonance (FMR) technique. Here, we report on the results of FMR studies performed at various temperatures in the martensite and austenite states of powder samples and discuss the mixed nature of the magnetic interactions in the martensitic state.

1. Introduction
Ternary non-stoichiometric Ni-Mn-X (X: In, Sn, Sb) Heusler alloys exhibit a martensitic transformation from a high symmetry austenite to a low symmetry martensite state [1]. The structural transition involves changes both in structural and magnetic properties of the materials. The rapid change in temperature dependence of the magnetization $M(T)$ occurs in a narrow temperature range around martensitic transition, where also both austenite and martensite states coexist giving rise to complex magnetic exchange. Magnetically inhomogenity related with different magnetic sublattices in the martensite state in Ni-Mn-Ga ribbons were reported previously [2]. Also, and more recently, the results of polarized neutron diffraction studies on Ni$_{50}$Mn$_{37}$Sn$_{13}$ and Ni$_{50}$Mn$_{40}$Sb$_{10}$ have verified the presence of complex antiferromagnetic (AF) correlations in the neighborhood of the transition [3]. Since polarization analysis is only possible in non-ferromagnetic environments, this technique does not provide information on the nature of any mixed magnetic interactions at lower temperatures because of the development of ferromagnetic (FM) long-range order in the martensitic state. Here, we complement these studies using the FMR technique and extract closer information on the mixed nature of FM and AF coupling in Ni$_{49.1}$Mn$_{35.4}$In$_{15.5}$ and Ni$_{49.9}$Mn$_{37.6}$Sn$_{13.1}$.

2. Experimental
Samples were prepared by arc melting under argon atmosphere. They were encapsulated under argon in quartz glass and annealed at 1073 K for 2 hours and then quenched in ice-water. The composition of the samples were determined by energy dispersive x-ray analysis. The temperature dependence of the magnetization $M(T)$ in 5 mT magnetic field was measured in the temperature range $5 \leq T \leq 380$ K with a superconducting quantum interference device
magnetometer. The data were first taken with zero-field-cooled (ZFC) and field-cooled (FC) protocols. As a last step \( M(T) \) was measured with a field-heating (FH) protocol back up to 380 K. FMR measurements were carried out using a 10 \( \mu \)m powder sample at a microwave frequency of 9.29 GHz in the temperature range 100 K \( \leq T \leq 300 \) K. The sample was first cooled down to 100 K, and then, data were taken on increasing temperature up to 300 K. At each temperature, the external field was increased up to 1.6 T and resonance spectra were recorded as a function of temperature. The signal was detected using a lock-in technique so that the derivative of the absorption signal was recorded \([4, 5, 6]\). An estimated error of resonance field \((\mu_0 H)\) is up to \( \pm 20 \) mT for AF signals and around \( \pm 80 \) mT for FM signals.

3. Result and Discussion

\( M(T) \) in an applied field of 5 mT for \( \text{Ni}_{49.1}\text{Mn}_{35.4}\text{In}_{15.5} \) and \( \text{Ni}_{49.9}\text{Mn}_{37.0}\text{Sn}_{13.1} \) are shown in Figs. 1a) and b). The Curie temperatures of austenite and martensite phases (\( T_C^A \) and \( T_C^M \)), the martensite start and finish \((M_s \text{ and } M_f)\) and austenite start and finish \((A_s \text{ and } A_f)\) are listed in Table 1. Only \( A_s \) and \( A_f \) are shown in Fig. 1 for clarity, since these are relevant to the discussion on the FMR results. Mainly due to the remanent field of the magnetometer of about 0.5 mT, the ZFC data at 5 K do not begin at \( M = 0 \). Nevertheless, the merging point of the FC and ZFC data at higher temperatures allows for a good estimation of \( T_C^M \). A thermal hysteresis is observed between the FC and FH (or ZFC) data around the structural transition. The samples are FM above \( A_f \) and paramagnetic (PM) above \( T_C^A \).

| Sample                  | \( T_C^A \)(K) | \( M_s \)(K) | \( M_f \)(K) | \( A_s \)(K) | \( A_f \)(K) | \( T_C^M \)(K) |
|-------------------------|----------------|--------------|--------------|--------------|--------------|----------------|
| \( \text{Ni}_{49.1}\text{Mn}_{35.4}\text{In}_{15.5} \) | 310            | 250          | 219          | 220          | 260          | 205            |
| \( \text{Ni}_{49.9}\text{Mn}_{37.0}\text{Sn}_{13.1} \) | 310            | 305          | 255          | 265          | 305          | 230            |

Fig. 2 shows the FMR spectra for both samples in the martensite and austenite states at 180 K and 300 K, respectively. The isotropic value of the resonance field is given as \( \omega/\gamma \approx 330 \) mT, where \( \omega \) is the microwave frequency and \( \gamma \) is the gyromagnetic ratio. The resonance equation for a ferromagnetically coupled lattice is given by \( B_{res} = \omega/\gamma - B_A \) where \( B_{res} \) is the resonance field and
$B_A$ is the internal anisotropy field. In a FM polycrystalline sample, $B_A$ is randomly oriented over the sample volume, and $B_{res}$ shifts to lower field values with respect to $\omega/\gamma$. For a paramagnetic state, $B_A = 0$, and $B_{res} = \omega/\gamma$. When the magnetic sublattices in an antiferromagnet are coupled by an exchange field $B_E$, the resonance equation is given by $B_{res} = \omega/\gamma \mp \sqrt{B_A(2B_E + B_A)}$ [8]. In this case, the equation has two solutions, and the resonance field is shifted below and above $\omega/\gamma$. The spectra at 300 K shows a single resonance signal up to 800 mT. However, in the martensite state at 180 K, three signals are seen. The signal at $\mu_0 H \leq 300$ mT (labeled as $I_{FM}$) lying below the isotropic value is related to the FM component and is observed up to 300 K. The signals which are observed above $\omega/\gamma$ for both samples, labeled $I_{AF}$, are related to AF components according to the resonance equation. $I_{AF}$ is present in the temperature range $100 \leq T \leq 180$ K for Ni-Mn-In and in the range $100 \leq T \leq 200$ K for Ni-Mn-Sn. They disappear above $T_C^{M}$ for both samples. For $400 \leq \mu_0 H \leq 450$ mT an additional AF signal appears for Ni-Mn-Sn that disappears at 230 K. The signal between $300 \leq \mu_0 H \leq 400$ mT in Ni-Mn-In lies nearly on the isotropic value and is related to PM components.

![FMR spectra](image)

Figure 2. FMR spectra in the martensite state at 180 K for both samples. AF ($I_{AF}$) and a FM ($I_{FM}$) contributions from the magnetic phase are observed. The inset shows the spectra for the austenite state at 300 K. $\omega/\gamma$ is marked by the dashed lines.

The temperature dependence of the resonance fields $\mu_0 H_{res}(T)$ extracted from the signals according to Dyson’s treatment for bulk specimens are shown in Fig. 3 and Fig. 4 [7]. In Fig. 3, FM signals are observed in the temperature range $100 \leq T \leq 300$ K. The isotropic value $\omega/\gamma \approx 330$ mT is shown as the horizontal filled area. Values larger than $\omega/\gamma$ are related to AF components of the magnetic phase. For $100 \leq T \leq 180$ K a signal labeled as $H_{res}^{I}$ having a higher resonance field than $\omega/\gamma$ is observed that decreases monotonically with increasing temperature. Since the sample is first cooled to the lowest temperature, and the data are taken on increasing temperature, the behavior of $\mu_0 H_{res}(T)$ can be understood by comparing it to $M(T)$ in the ZFC state. The splitting of the ZFC and FC curves below $T_C^{M}$ is understood to be due to the pinning of the FM components by coexisting AF components. The AF exchange weakens with increasing temperature so that the FC and ZFC branches of $M(T)$ merge at $T_C^{M}$. This is reflected as a decrease in $\mu_0 H_{res}(T)$ with increasing temperature. The FMR intensity which is a measure of the magnetic moment is shown in the inset of Fig. 3. Here the intensity of the ferromagnetic signal is estimated from $\mu_0 H_{res}(T)$ data lying below the isotropic value. Starting at the low temperature end, the intensity is nearly constant up to a temperature close to $A_s$ with a low value and then increases with increasing temperature as FM austenite develops in the sample.

In Fig. 4a) for the Ni-Mn-Sn sample, two magnetic contribution are observed below and above $\omega/\gamma$. Below $T_C^{M}$, two signals, $H_{res}^{I}$ and $H_{res}^{II}$, appear which are related to weak AF ordering. $H_{res}^{I}$ is related to AF exchange which is consistent with the behavior of $M(T)$, as in the case for Ni-Mn-In. $H_{res}^{II}$ is particularly significant for $100 \leq T \leq 230$ K; the range which is essentially below the splitting point of the ZFC and FC curves in $M(T)$. It approaches the isotropic value close to $T_C^{M}$. The intensities of $H_{res}^{II}$ (open circles) and the FM part $H_{res}^{III}$ in

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Figure 3. Temperature dependence of the resonance field in Ni$_{49.1}$Mn$_{35.4}$In$_{15.5}$. AF coupling appears below about $T^M_C$ denoted by the data labeled with $H^M_{res}$. $\omega/\gamma$ indicates the isotropic value. Inset shows the temperature dependence of the intensity for the FM phase.

Fig. 4b) are temperature independent at low temperatures and both begin to increase with increasing temperature at about 200 K. They reach a maximum at 250 K, after which the FM part diminishes, reflecting an overall decrease of the magnetic moment as the temperature approaches $T_C$. At 270 K magnetization starts to increase as FM ordering sets in. The sharp decrease in $M(T)$ for the Ni-Mn-Sn sample just below $M_s$ was shown to be related to the loss of ferromagnetism and the appearance of AF correlations [3]. Analogously, one can observe the development of such correlations in the present FMR data particularly through the behavior of $H^I_{res}$.

4. Conclusion

Around and below the martensitic transformation in Ni$_{49.1}$Mn$_{35.4}$In$_{15.5}$ and Ni$_{49.9}$Mn$_{37.0}$Sn$_{13.1}$, the coupled FM and AF interactions can be distinguished by FMR studies. We show directly that AF coupling is present within an essentially FM state below $T^M_C$, which is otherwise inferred indirectly from ZFC-FC $M(T)$ data as well as the observation of exchange-bias [9]. This study complements to temperatures $T < T^M_C$ the results of earlier polarization analysis studies, which show the presence of AF correlations for $T > T^M_C$ and around the martensitic transition.
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