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## Detection of weak-order phase transitions in ferromagnets by ac resistometry

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It is shown that ac resistometry can serve as an effective tool for the detection of phase transitions, such as spin reorientation or premartensitic phase transitions, which generally are not disclosed by dc resistivity measurement. Measurement of temperature dependence of impedance,  $Z(T)$ , allows one to unmask the anomaly, corresponding to a weak-order phase transition. The appearance of such an anomaly is accounted for by a change in the effective permeability  $\mu$  of a sample upon the phase transition. Moreover, frequency dependence of  $\mu$  makes it possible to use the frequency of the applied ac current as an adjusting parameter in order to make this anomaly more pronounced. The applicability of this method is tested for the rare earth Gd and Heusler alloy  $\text{Ni}_2\text{MnGa}$ . © 2003 American Institute of Physics. [DOI: 10.1063/1.1594840]

It is well known that impedance  $Z$  of a magnetic conductor depends on the effective permeability  $\mu$  of the material. In particular, this is evident from the fact that impedance of soft ferromagnetic materials changes considerably in a magnetic field, which is accounted for by a drastic change of  $\mu$  upon application of the field. This phenomenon, called giant magnetoimpedance, is most pronounced in soft ferromagnetic wires with a peculiar configuration of magnetic domains (Refs. 1–3 and references therein), but generally it seems to be an intrinsic property of soft ferromagnets and recently it was observed in several Heusler alloys.<sup>4,5</sup>

The fact that  $Z$  depends on the effective permeability  $\mu$  gives rise to suggest that it can be used for studying weak-order phase transitions, such as spin reorientation, order disorder, and so on, occurring in the ferromagnetic state. Indeed, since dc resistivity does not allow detection or accurate determination of a temperature of the transition, upon which no significant change in the Fermi surface, mean-free path, or electron concentration occurs, such transitions are usually studied by magnetic measurements. The existence of the anomaly corresponding to a weak-order phase transition at a temperature dependence of magnetization evidences that such a transition is accompanied by a change in permeability of the sample, therefore, measurements of  $Z(T)$  could also be effectively used for this aim. Moreover, in certain cases, this method can have an advantage compared to the magnetic measurements, for example, if a weak-order phase transition is located in the proximity to another phase transition, characterized by a drastic change in the magnetization.

It is shown in this article, that ac resistometry can serve as an effective tool for this purpose. The validity of this method is demonstrated for the case of the rare-earth Gd, which undergoes a spin reorientation transition at a low tem-

perature, and for a  $\text{Ni}_2\text{MnGa}$  alloy, which undergoes a weak-order premartensitic phase transition.

Impedance  $Z$  of a cylindrical conductor can be derived using Maxwell equations:<sup>6</sup>

$$Z = R + iX = \frac{1}{2} R_{\text{dc}} k a \frac{J_0(ka)}{J_1(ka)},$$

where  $R_{\text{dc}}$  is dc resistance of the conductor at a frequency  $f = \omega/2\pi = 0$ ,  $a$  is the conductor radius,  $J_i$  are Bessel functions of the first kind, and  $k = (1 + i)/\delta$ , where  $\delta$  is the skin depth,

$$\delta = c \sqrt{\frac{\rho}{2\pi\omega\mu}}.$$

The skin depth  $\delta$  is determined by resistivity  $\rho$  and effective permeability  $\mu$  of the conductor, and by the frequency  $\omega$  of applied to the conductor ac current. In the case of a slab geometry, the expression for the impedance is<sup>7</sup>

$$Z = R_{\text{dc}} i k d \coth(ikd),$$

where  $d$  is the half thickness. Both of these equations show that at a constant frequency, the temperature dependence of  $Z$  is determined by the temperature dependence of  $\rho$  and  $\mu$  as  $Z(T) \sim \sqrt{\rho(T)\mu(T)}$ .

The experimental setup for the measurement of temperature dependencies of the impedance  $Z$  consists of a lock-in amplifier, a function synthesizer, a reference resistor connected in a serial way with the sample, and a computer. Alternating voltage of amplitude  $V_0$  at a frequency  $f$  was applied to the reference resistor using the function synthesizer. The voltage drop on the sample and the phase shift between the voltage and the reference signal were measured by the lock-in amplifier.

Measurements of  $\rho(T)$  and  $Z(T)$  were performed on a Gd sample measuring  $8 \times 2 \times 1 \text{ mm}^3$  and a  $\text{Ni}_2\text{MnGa}$  sample measuring  $10 \times 1.5 \times 0.5 \text{ mm}^3$ . Using typical values of  $\rho$  and

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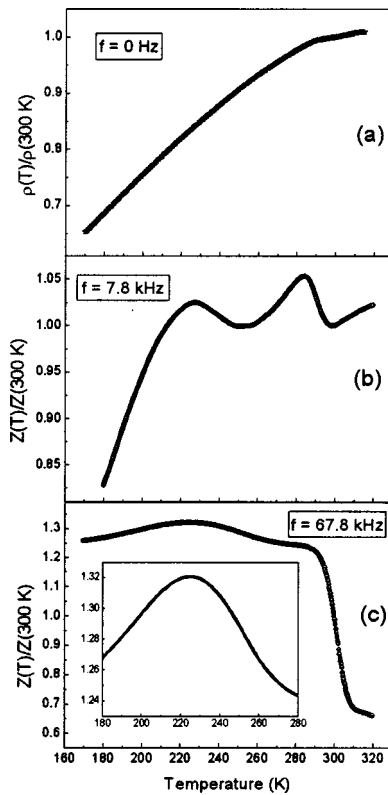


FIG. 1. Temperature dependencies of resistivity  $\rho$  and impedance  $Z$  measured on the Gd sample.

$\mu$  for Gd,  $\rho \approx 130 \times 10^{-6} \Omega \text{ cm}$  and  $\mu \approx 200$ , and  $\text{Ni}_2\text{MnGa}$ ,  $\rho \approx 30 \times 10^{-6} \Omega \text{ cm}$  and  $\mu \approx 200$  (Ref. 4), the skin depth for  $7 \text{ kHz} < f < 100 \text{ kHz}$  is  $1500 \text{ \AA} < \delta < 5000 \text{ \AA}$  for the Gd sample and  $700 \text{ \AA} < \delta < 2600 \text{ \AA}$  for the  $\text{Ni}_2\text{MnGa}$  sample. These values are much smaller than typical sample thickness ( $\geq 0.5 \text{ mm}$ ), so that the skin effect is essential at these frequencies.

### A. Spin reorientation transition in Gd

It is known from literature that Gd orders ferromagnetically at the Curie temperature  $T_C \approx 293 \text{ K}$  and undergoes a spin reorientation transition at a lower temperature,  $T_s \approx 225 \text{ K}$  (Refs. 8 and 9 and references therein).

Temperature dependencies of  $\rho$  and  $Z$ , measured upon heating, are presented in Fig. 1. As evident from Fig. 1(a),  $\rho(T)$  has an anomaly at Curie temperature  $T_C$ , equal to 290 K for this sample. No anomaly, corresponding to the spin reorientation transition is seen in the curve in Fig. 1. This result is in agreement with early transport measurements of Gd.<sup>10</sup>

Temperature dependencies of the impedance  $Z$ , measured on this sample [Figs. 1(b) and 1(c)], drastically differ from the temperature dependence of the resistivity  $\rho$ .  $Z(T)$ , measured at a frequency  $f = 7.8 \text{ kHz}$  has a complex temperature dependence [Fig. 1(b)], namely, two marked peaks are clearly seen on this curve. The first peak is observed at the temperature of spin reorientation phase transition,  $T_s = 226 \text{ K}$ . This temperature, determined from the  $Z(T)$  measurement, is in good agreement with the temperature deter-

mined by another experimental techniques.<sup>8,9</sup> Since  $\rho(T)$  does not exhibit anomaly at  $T_s$ , the local maximum of  $Z(T)$  at this temperature is accounted for by a larger permeability  $\mu$  at the spin reorientation transition temperature. The origin of the second peak observed near the ferromagnetic phase transition is presumably the same because Gd has a pronounced peak of susceptibility  $\chi = \mu - 1/4\pi$  in the vicinity of  $T_C$  (Ref. 9). The result of  $Z(T)$  measurement performed at a higher frequency,  $f = 67.8 \text{ kHz}$ , is shown in Fig. 1(c). This curve also has a pronounced peak at the spin reorientation temperature  $T_s$ . Since upon transition from the ferromagnetic-to-paramagnetic state, the impedance  $Z$  drastically decreases, this peak is shown in the inset in Fig. 1(c). The existence of this prominent and well-defined peak permits an accurate determination of the spin reorientation transition temperature from the  $Z(T)$  measurements.

Therefore, these results have shown that measurement of impedance  $Z$  can be effectively used, along with magnetic measurements, for the study of spin reorientation transitions and construction of phase diagrams of compounds undergoing such transitions.

### B. Premartensitic transition in $\text{Ni}_2\text{MnGa}$

An interesting property of near stoichiometric  $\text{Ni}_2\text{MnGa}$  alloys is that they undergo a so-called premartensitic phase transition, which is characterized by a modulation of the Heusler cubic structure.<sup>11</sup> Resistivity measurement of these alloys showed<sup>12-14</sup> that it is difficult to determine the temperature of premartensitic and martensitic phase transition from  $\rho(T)$  data because the resistivity has weak and broad anomalies at these phase transformation temperatures. Measurement of magnetization is an effective tool to determine the martensitic transformation temperature,<sup>15</sup> but as for the premartensitic transition,  $M(T)$  measurement is sometimes not sufficient to determine the temperature of the premartensitic transition in the case of polycrystalline samples.<sup>16</sup> In order to prove that the premartensitic transition can be disclosed by  $Z(T)$  measurements, temperature dependencies of  $\rho$  and  $Z$  were measured on a polycrystalline sample of stoichiometric  $\text{Ni}_2\text{MnGa}$  composition.

Measurement of  $\rho(T)$ , shown in Fig. 2(a), was typical for the stoichiometric  $\text{Ni}_2\text{MnGa}$  behavior of resistivity.<sup>12-14</sup> A marked anomaly is seen only at the ferromagnetic transition temperature  $T_C = 380 \text{ K}$ , whereas the martensitic and premartensitic phase transitions are accompanied by a broad change in the slope of the curve at  $T_m \approx 200 \text{ K}$  and  $T_p \approx 260 \text{ K}$ , respectively.

Unlike the resistivity  $\rho$ , the impedance  $Z$  of this sample has quite a different temperature dependence [Figs. 2(b) and 2(c)].  $Z(T)$  measured at  $f = 7.8 \text{ kHz}$  [Fig. 2(b)] exhibits a peak in the vicinity of Curie temperature  $T_C$ , which is accounted for by a high susceptibility of the sample. Contrary to  $\rho(T)$ , the temperature dependence of the impedance  $Z$  exhibits a jump-like behavior at the martensitic transition temperature  $T_m$ . This is due to the fact that the martensitic phase has a lower permeability  $\mu$  as compared to the austenitic cubic phase. Finally, the premartensitic transition appears at this curve in Fig. 2 as a small dip at  $T_p = 260 \text{ K}$ .

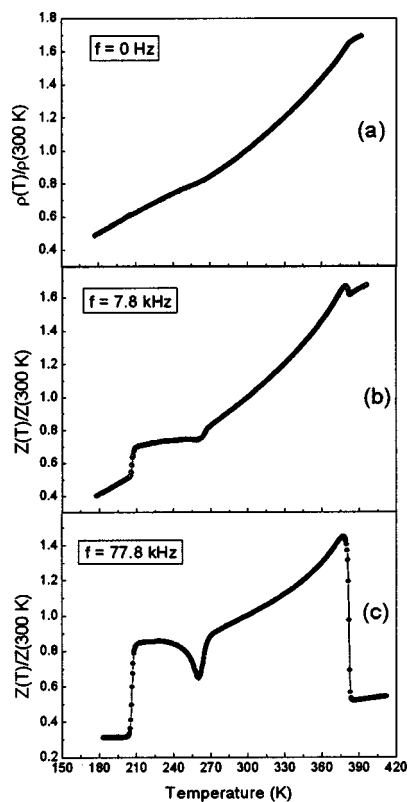


FIG. 2. Temperature dependencies of resistivity  $\rho$  and impedance  $Z$  measured on the  $\text{Ni}_2\text{MnGa}$  sample.

Measurement of  $Z(T)$  at a higher frequency  $f = 77.8$  kHz [Fig. 2(c)] indicates that the anomalies corresponding to the phase transitions are enhanced. The most interesting finding is that the increase in the frequency of the current results in the appearance of a pronounced dip at the premartensitic phase transition. This observation allows an accurate determination of the premartensitic transition temperature  $T_p$ . Since the impedance  $Z$  decreases and then increases smoothly around  $T_p$ , whereas the drastic drop of  $Z$  is observed at  $T_m$ , it can be suggested that by measuring  $Z(T)$  one can easily distinguish these transitions even if they are close to each other.

In conclusion, the results presented in this article have shown that, by measuring the temperature dependence of im-

pedance  $Z$ , it is possible to unmask anomalies which are generally not observed on dc resistivity curves. This has been confirmed for the case of gadolinium, which exhibits a spin reorientation transition at  $T_s = 226$  K, and for the case of  $\text{Ni}_2\text{MnGa}$ , which exhibits a premartensitic phase transition at  $T_p = 260$  K. Moreover, the results of this study indicated that by adjusting the frequency of the ac current, one can observe a sharp anomaly at the temperature of such a transition. Therefore, this method can be used, along with magnetic measurements, as a simple and effective tool for the study of spin reorientation transitions and construction of phase diagrams in intensively studied rare-earth alloys and in other magnetic alloys and compounds.

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