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Temperature and magnetic field dependence of permeability in amorphous magnetic materials

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Abstract. — In this paper, the temperature and magnetic field dependence of permeability, $\mu$, in Fe$_{100-x}$B$_x$, ($x = 18, 20, 22$), Fe$_{70}$B$_{14}$Si$_8$ and Fe$_{62}$Ni$_{16}$B$_{12}$Si$_8$ are studied. After annealing up to Curie temperature, Fe$_{90}$B$_{10}$ and Fe$_{70}$B$_{30}$ alloys become crystalline, while other alloys remain amorphous. The Curie temperatures were measured. The DC magnetic field dependence of the permeability shows that the approach to saturation is modified by annealing.

1. Introduction.

The new measurement method [1, 2] is based on the reflection pulse by a coil which contains the material sample. The coil is connected to a nanosecond pulse generator with a coaxial cable. The same generator output is connected to a sampling oscilloscope. The amplitude of the reflection pulse is given by the expression [3].

$$U = \left\{ \rho + (1 - \rho) \exp \left[ - (t - 2T)/\tau \right] \right\} U_0 \theta(t - 2T) -$$

$$- \left\{ \rho + (1 - \rho) \exp \left[ - (t - t_0 - 2T)/\tau \right] \right\} U_0 \theta(t - t_0 - 2T)$$

$$\rho = \frac{R - Z}{R + Z} \quad \tau = \frac{L}{R + Z} \quad \theta(t - x) = \begin{cases} 0, & t < x \\ 1, & t > x \end{cases}$$

$\tau$ is the propagation delay of the pulse in the coaxial cable and $Z$ the impedance. $U_0$ is the reflected pulse amplitude defined in figure 1. It was assumed that the coil has a resistance $R$ and a series inductance $L$. If $R \ll Z$ (in our experiment $R \approx 0.1 \Omega$ and $Z = 80 \Omega$), $\rho = -1$ and the first term from relation (1) becomes

$$U_1 = U_0 \left\{ 2 \exp \left[ - (t - 2T)/\tau \right] - 1 \right\}$$

so that

$$\tau = \frac{t - 2T}{\ln \left[ \frac{2U_0}{U_1 + U_0} \right]}$$
For the direct pulse width, \( t_2 = t_0 \), we measure \( U_1 \) and \( U_0 \) (defined in Fig. 1a), and determine \( \tau \) from relation (3) and therefore

\[
L = \tau Z.
\]

(4)

The permeability may be evaluated from the relation

\[
\mu_r = \frac{L}{L_0}
\]

(5)

where \( L_0 \) is the empty coil inductance and \( L \) is the coil inductance when the coil contains a sample which fills the coil core. If \( U_1 = 0 \), then from the relation (3) we obtain

\[
\tau = \frac{t_0'}{\ln 2}
\]

(6)

where \( t_0' \) is defined in figure 1b. This situation appears when \( \tau < t_0 \), while the situation presented in figure 1a appears when \( \tau > t_0 \).

The pulse permeability is usually less than the conventional incremental permeability, since the skin effects limits penetration into the material at high frequencies. When the skin depth

\[
\delta = \left( \frac{2}{\omega \mu \sigma} \right)^{1/2}
\]

(7)

is much smaller than the sample thickness, \( d \), we have \([1, 2]\)

\[
L = \frac{N^2 W \delta}{\ell} \mu, \quad R_s = \varnothing L
\]

where instead of the cross-section area of the sample appears \( W \delta \), \( W \) is the sample width, \( \ell \) is the length of the coil with \( N \) turns, \( \omega \) is the frequency, \( \sigma \) is the conductivity, \( R_s \) is a surface resistance due to the sample which appears at high temperatures and must be added to the coil resistance; the sample fills all the core.
When the skin depth is much larger than the sample thickness and if the sample does not fill all the core, then denoting the fill factor by $F$,

\[ L = \frac{\mu_0 \mu_r N^2 S}{q} \left( F + \frac{\mu_0 N^2 S}{q} (1 - F) \right) \]

$\mu_0 = 4 \pi \times 10^{-7} \text{Hm}^{-1}$, $S$ is the cross-sectional area of the coil.

The pulse repetition time is $\tau_r = 8 \mu s$ so that repetition frequency is of the order of 125 kHz.

2. Errors.

From equations (3)-(5) we may derive the total error as follows. Incorporating equation (3) with (4) leads to

\[ \mu_r = \frac{t_0 Z}{L_0 \ln \left[ \frac{2}{(U_1/U_0) + 1} \right]} \] (8)

By definition we may write

\[ \Delta \mu_r = \left[ \left( \frac{\partial \mu_r}{\partial t_0} \Delta t_0 \right)^2 + \left( \frac{\partial \mu_r}{\partial L_0} \Delta L_0 \right)^2 + \left( \frac{\partial \mu_r}{\partial Z} \Delta Z \right)^2 + \left( \frac{\partial \mu_r}{\partial U_1} \Delta U_1 \right)^2 + \left( \frac{\partial \mu_r}{\partial U_0} \Delta U_0 \right)^2 \right]^{1/2} \] (9)

Manipulating the derivation of equation (8) given us that

\[ \frac{\Delta \mu_r}{\mu_r} = \left[ \left( \frac{\Delta t_0}{t_0} \right)^2 + \left( \frac{\Delta L_0}{L_0} \right)^2 + \left( \frac{\Delta Z}{Z} \right)^2 + \left( \frac{\Delta U_0}{U_0} \right)^2 + \left( \frac{\Delta U_1}{U_1 + U_0} \ln X \right)^2 \right]^{1/2} \] (10)

For $U_1 = 0$, $\tau$ is given by equation (6) and this relation becomes

\[ \frac{\Delta \mu_r}{\mu_r} = \left[ \left( \frac{\Delta t_0}{t_0} \right)^2 + \left( \frac{\Delta Z}{Z} \right)^2 + \left( \frac{\Delta L_0}{L_0} \right)^2 \right]^{1/2} \] (11)

In the experiment presented here the reflected pulse length $t_0$ is smaller than the direct pulse length $t_0$ (the case presented in Fig. 1b) so that it is given by equation (6) and the errors are given by equation (11).

The error in measuring the time $t_0 = 20$ ns on the oscilloscope time base for a 5 ns/cm sensitivity is of the order of $(\Delta t_0/t_0) = 0.01$. Because of the large errors that can arise in the calculation of the empty coil from dimensions of only modest uncertainties it was decided to use a measured value of $L_0$. By measuring $L_0$ with a bridge at 125 kHz the error was $(\Delta L_0/L_0) = 0.02$. We measure the cable impedance $Z$ by connecting at the end of the cable a resistance $R$ for which the amplitude of the reflected pulse is equal to zero, that is $R = Z$. The error $(\Delta Z/Z) = 0.01$. Therefore, the error in measuring $\mu_r$ is $(\Delta \mu_r/\mu_r) \approx 0.03$. Evidently, there are intrinsic noise factors to be considered which are related to the skin effect [1].
A single pulse is composed of a continuous frequency spectrum, i.e. a Fourier spectrum. The various frequency components add at the input end to give the applied pulse wave form. If these components all propagate at the same velocity with no attenuation they will add at any given point to produce the identical waveform. Distortion comes about when attenuation is present and/or when the velocity of propagation is different for the various frequencies, i.e. dispersion. If the attenuation and velocity are the same for all the frequencies, the waveform at any point on an infinitely long line will be identical in the shape, but will decrease in amplitude with distance from the input end. This case is not usually in practice because the mechanism which give raise to the attenuation also introduces a small but finite dependence of velocity on frequency, thus causing a distortion of waveform in addition to a reduction in amplitude.

The distortion appears due to series resistance losses and the skin effect in the conductor. The error due to the distortion of the pulse shape by frequency dependence of the permeability is small if the rise time is smaller than the pulse length, \( t_0 \). In order to estimate this error we measure the impedance \( L_i \) of the coil which contains the sample by using a bridge at 125 kHz. Then we connect the end of the cable to an empty coil with the value \( L_i = L_1 \) and measure \( t_0 \). By comparing these values with \( t_0 \) obtained for the coil \( L_1 \) which contains the sample we determine \( \Delta t_0/t_0 \) and therefore the error due to the distortion of the pulse shape. If the skin effect is negligible this error is very small.

3. Results and discussion.

Amorphous ribbons of Fe-B, Fe-B-Si and Fe-Ni-B-Si alloys \( \sim 1 \) mm in width and \( \sim 30 \) \( \mu \)m in thickness were prepared using a conventional melt-quenching apparatus consisting on a single roll. The result of X-ray diffraction and Mossbauer effect confirmed the amorphous state of the ribbons [4]. By using the reflection pulse method we measured the temperature and the DC bias field dependence of the permeability of this ribbons. The repetition time of the pulse is \( \tau_r = 8 \) \( \mu \)s and by using \( \mu_r = 100 \) (see below), \( \rho = 1/\sigma = 200 \) \( \mu \)\Omega cm, \( \omega = 2\pi/\tau_r \), one obtains \( \delta = 200 \) \( \mu \)m from equation (7). This value is larger than the ribbon half-thickness (~15 \( \mu \)m) so we neglected the skin effect. The current pulse in the coil is \( I_{\text{max}} \approx 1 \) mA, so that in a coil with \( N/\ell = 1 \) turn/mm a pulse magnetic field of \( \sim 10 \) mOe was obtained.

3.1 Curie temperatures. — Figure 2 shows the temperature dependence of the permeability \( \mu_r \) of Fe\(_{100-x}\)B\(_x\) \( (x = 18, 20, 22) \). As seen in the figure just below the Curie temperature \( \mu_r \) drops rapidly as temperature is raised through \( T_c \). The Curie temperature evaluated from these curves (shown by an arrow in the figure) are the following: 631, 654, and 683 K respectively.

In figure 3 the temperature dependence of the permeability is presented for Fe\(_{78}\)B\(_{22}\) (curve 1), Fe\(_{78}\)B\(_{14}\)Si\(_8\) (curve 2) and Fe\(_{62}\)Ni\(_{18}\)B\(_{14}\)Si\(_8\) (curve 3). The Curie temperatures are the following: 683 K, 700 K and 715 K respectively. The Curie temperature for some of these alloys are measured in the past by using standard methods and they are in a good agreement with our results [5]. The following conclusions appear. In Fe rich alloys, the increase in the metalloid concentration leads to an increase in \( T_c \). Silicon is more effective than boron [6].

In the phenomenological Bethe-Slater curve argument this may appear to be related to the increase in Fe-Fe average distance in terms of the first peak position in RDF [7]. Also, in Fe rich alloys the replacement of Fe by Ni increases \( T \). This in most because the presence of Ni at the nearest neighbour fills up the majority spin bands of Fe up to the antiresonance minimum of the electronic density of states and at the same time increasing the exchange interaction, even through Ni itself may have only a small moment. There is an analogous situation in crystalline
Fig. 2. — The variation of the permeability near the Curie temperature in Fe$_{100-x}$B$_x$ ($x = 18, 20, 22$).

Fig. 3. — The variation of the permeability near the Curie temperature in Fe$_{78}$B$_{22}$ (curve 1), Fe$_{78}$B$_{14}$Si$_8$ (curve 2) and Fe$_{60}$Ni$_{16}$B$_{14}$Si$_8$ (curve 3).
Fe-Ni alloys in γ phase [7]. In all samples, annealing up to the Curie temperature increases little $T_c$ value.

3.2 THE FIELD DEPENDENCE OF THE PERMEABILITY. — In figures 4-8 the field dependence of the permeability is presented for Fe$_{82}$B$_{18}$, Fe$_{80}$B$_{20}$, Fe$_{78}$B$_{22}$, Fe$_{78}$B$_{14}$Si$_8$ and Fe$_{62}$Ni$_{16}$B$_{14}$Si$_8$ respectively. The circles curve is for as-quenched state and the crosses curve is for the state after the sample heating up the Curie temperature. The coercivity of amorphous ribbons is smaller than that of crystalline state and is of 1-10$^2$ mOe. It is determined by surface effect and by internal stresses. By heating up to the Curie temperature the samples of Fe$_{80}$B$_{20}$ and Fe$_{78}$B$_{22}$ alloys become crystalline, because their crystallisation temperatures are smaller than the Curie temperatures. (For Fe$_{80}$B$_{20}$ Curie temperature $T_c = 654$ K and crystallisation temperature $T_x = 652$ K and for Fe$_{78}$B$_{22}$ $T_c = 683$ K, $T_x = 654$ K). As appears from figures 5 and 6 in these cases, after annealing (the heating up to the Curie temperature) at high field the permeability decreases slower with field increase than before annealing. In other cases the Curie temperature is smaller than the crystallisation temperature and the annealing up to the Curie temperature induces a more rapidly decrease of $\mu_r$ with field increase (Figs. 4, 7, 8). In the other word, the annealing modifies the approach to saturation. Also, the annealing reduces the value of the coercivity somewhat.

It is well-known that when the applied field is increased to larger values, domain-well movement becomes relatively unimportant and magnetisation changes occur primarily by domain rotation. It has been stated that the approach to the saturated state may be described by the empirical relationship [8].

$$M = M_s \left( 1 - a \frac{b}{H} - b H^2 \right) + cH \quad (12)$$

where $M_s$ is the saturation magnetisation and $M$ is the magnetization (along the field direction) in the field $H$. We measured the induction $B_r$ at saturation by hysteresis loop visualisation and obtains the following values (in the as-quenched state).

<table>
<thead>
<tr>
<th>Material</th>
<th>$B_s$, kGs</th>
<th>$B_r$, kGs</th>
<th>$H_c$, Oe</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe$<em>{82}$B$</em>{18}$</td>
<td>13.03</td>
<td>6.12</td>
<td>0.123</td>
</tr>
<tr>
<td>Fe$<em>{80}$B$</em>{20}$</td>
<td>14.53</td>
<td>8.5</td>
<td>0.103</td>
</tr>
<tr>
<td>Fe$<em>{78}$B$</em>{22}$</td>
<td>12.07</td>
<td>6.34</td>
<td>0.114</td>
</tr>
<tr>
<td>Fe$<em>{78}$B$</em>{14}$Si$_8$</td>
<td>11.06</td>
<td>6.43</td>
<td>—</td>
</tr>
<tr>
<td>Fe$<em>{62}$Ni$</em>{16}$B$_{14}$Si$_8$</td>
<td>12.5</td>
<td>5.87</td>
<td>0.112</td>
</tr>
</tbody>
</table>

Also in this table are presented the obtained values of the remanent induction $B_r$ and the values of the coercivity $H_c$.

From the relation (12) appears that

$$\frac{dM}{dH} = \mu_r, - 1 = M_s \left( \frac{a}{H^2} + \frac{2 b}{H^3} \right) + c \quad (13)$$
Fig. 4. — The field dependence of the permeability in Fe₈₂B₁₈. 1) as-quenched state, 2) annealed up to the Curie temperature.

Fig. 5. — The field dependence of the permeability in Fe₈₀B₂₀. 1) as-quenched state, 2) annealed up to the Curie temperature.
\[ b = b_1 + b_2 \] depends on the anisotropy constant \( K_1 \), that is \( b_1 \sim K_1^2 \) [8] and also on the internal stresses, that is \( b_2 \sim \lambda^2 \sigma_i^2 \), where \( \lambda \) is the saturation magnetostriction constant and \( \sigma_i \) is the internal tension. The origin of the \( 1/H \) term in (12) is less well understood. When

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**Fig. 6.** — a) The field dependence of the permeability in Fe\textsubscript{78}B\textsubscript{22}, b) logarithmic scale: 1) as-quenched state, 2) annealed up to the Curie temperature.
imperfections such as dislocations, nonmagnetic inclusions and nonuniformities of the ribbon are present, the $1/H$ term predominates for intermediate fields, whereas $1/H^2$ term predominates for very large fields.

The magnetic domain structure of the amorphous ribbons was the subject of recent interest [9]. In the as-quenched state the internal stresses are usually non-zero, since during the fabrication of the ribbon the freezing often occurs non-uniformity as aerodynamic oscillations of the molten alloys puddle are difficult to avoid. Then the portion of the ribbon frozen early is left in compression. When the magnetostriction of the alloys is non zero, these inhomogeneous stresses produce local anisotropies with the easy axis out of the plane of the ribbon in about 10-30% of the volume. This anisotropy can be relaxed to a large extent by annealing. Therefore, in an applied field, parallel with a ribbon length, the annealing induces a more rapidly saturation with the field increase, as it appears in figures 4, 7, 8. However, the anisotropy due to more microscopic stress inhomogeneities of the scale $10^2$-$10^3$ Å, described in terms of the dislocation dipoles, cannot be readily annealed out. In figure 8b are presented, in the logarithmic scale, $\log (\mu_r - 1)$ versus $\log H$, the data from Fe$_{78}$B$_{14}$Si$_6$. It results that in the as-quenched state, at high fields

$$n = \frac{\Delta \log (\mu_r - 1)}{\Delta \log H} = -2.5$$

while after annealing $n = -3$. Therefore, in annealed samples the $1/H^3$ term predominates in $\mu_r(H)$ dependence at high fields. The other is the behaviour when the annealing up the Curie temperature the sample become crystalline. In figure 6a is presented $\log (\mu_r - 1)$ versus $\log H$ for Fe$_{78}$B$_{22}$. In the as-quenched state $n = -2.5$ at high fields, while after annealing (in the
crystalline state) in the same of the DC magnetic field variation, \( n = -2 \) is obtained. The crystalline state has a larger anisotropy (a larger induction at saturation) than the amorphous state.


We used a new method for permeability measurements. This method is based on a pulse reflection by a coil containing the sample. Also, we present the error calculus for the method.
The Curie temperature were determined. In Fe rich alloys the Curie temperature increases with increasing metalloid content. Also, the replacement of Fe by Ni increasing $T_c$. The field dependence of permeability was measured. The annealing up to Curie temperature modifies this dependence. If the sample becomes crystalline during the annealing process, the field dependence of $\mu_r$ is slower than before annealing. If the sample remains in amorphous state $\mu_r$ decreases more rapidly with $H$ increases than before annealing. This effect is due to the structural relaxation.

References