

# Magnetic permeability behavior in single crystal Mn-ferrites

**Citation for published version (APA):**

Simsa, Z., Zounova, F., & Brabers, V. A. M. (1988). Magnetic permeability behavior in single crystal Mn-ferrites. *IEEE Transactions on Magnetics*, 24(2, Pt. 2), 1841-1843. <https://doi.org/10.1109/20.11620>

**DOI:**

[10.1109/20.11620](https://doi.org/10.1109/20.11620)

**Document status and date:**

Published: 01/01/1988

**Document Version:**

Publisher's PDF, also known as Version of Record (includes final page, issue and volume numbers)

**Please check the document version of this publication:**

- A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher's website.
- The final author version and the galley proof are versions of the publication after peer review.
- The final published version features the final layout of the paper including the volume, issue and page numbers.

[Link to publication](#)

**General rights**

Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the public portal.

If the publication is distributed under the terms of Article 25fa of the Dutch Copyright Act, indicated by the "Taverne" license above, please follow below link for the End User Agreement:

[www.tue.nl/taverne](http://www.tue.nl/taverne)

**Take down policy**

If you believe that this document breaches copyright please contact us at:

[openaccess@tue.nl](mailto:openaccess@tue.nl)

providing details and we will investigate your claim.

MAGNETIC PERMEABILITY BEHAVIOUR IN SINGLE CRYSTAL Mn-FERRITES

Z. Šimša, F. Zounová and V.A.M. Brabers (1)

Institute of Physics, Czechoslovak Acad. Sci., 180 40 Prague 8, Czechoslovakia  
 (1)Dept. of Physics, Technical University, 5600 MB Eindhoven, The Netherlands

Measurements of the initial permeability  $\mu$  of single crystal ferrites  $Mn_xFe_{3-x}O_4$  with  $0 < x < 1.25$  are performed at audio frequencies at temperatures from 10 to 550 K. Large dispersionless changes in  $\mu$  occur near the Verwey transition of magnetite whereas two relaxation type anomalies are observed at 40 K and 80 K. Similar anomalies are found at 20 K and 60 K for the  $x=0.5$  sample. They are followed by a broad feature persisting up to room temperature. The main relaxation processes in the case of samples containing larger amounts of manganese occur above 100 K and shift to higher temperatures with the increase of  $x$ . The activation energies determined for particular processes indicate the electronic origin of these effects.

Introduction

The initial permeability of magnetic materials,  $\mu = \mu' - j\mu''$ , relates directly to the movement of the domain walls and to the relaxation processes encountered in the samples studied. The observed relaxation processes in ferrites appear to be mostly of electronic or of ionic character. They manifest themselves in a variety of measured phenomena as e.g. disaccommodation of permeability, induced anisotropy and magnetic losses (for reviews see e.g. [1-6]). Only few papers were devoted to the initial permeability measurements on single crystals. Bickford [7] ascribed the anomalous behaviour of  $\mu$  near 120 K in single crystals of magnetite to both the crystallographic phase transition (Verwey transition) and to the passage of the magneto-crystalline anisotropy constant  $K_1$  through zero at  $T_K$ . On the other hand, Enz [8] argued that the maxima of  $\mu$  below room temperature in some Mn- and Mn-Zn ferrites were due to disaccommodation phenomena. However, Ohta [9] demonstrated that the second maxima in  $\mu$  in the system of Mn-Zn ferrites are primarily caused at  $K_1=0$  whereas the disaccommodations bring about only small temperature shifts.

It seemed worthwhile to investigate the behaviour of the initial permeability of Mn-ferrite single crystals over a broad temperature range and to find out the main underlying mechanisms responsible for the observed anomalies.

Experimental part

**Samples.** Single crystals in the ferrite system  $Mn_xFe_{3-x}O_{4+\gamma}$  were prepared by a travelling molten zone technique as described elsewhere [10]. After the growth process the single crystals were additionally heat treated in controlled atmospheres to improve homogeneity and to relieve mechanical stress. The final chemical compositions as well as some other sample parameters are given in Table 1. From cylindrical rods the toroid cores 3 mm in height and 5 mm of outer and 2mm of inner diameter were tubed bored with the help of grinding powder. Before the appropriate winding was put on, the samples were etched to remove the damaged surfaces.

Table 1  
Parameters of  $Mn_xFe_{3-x}O_{4+\gamma}$  crystals

Sample	x	$\gamma$	toroid plane	$T_C$ (K)	$T_K$ (K)
1	0	0.007	(110)	858	126
2	0.52	0.009	(100)	741	340
3	0.95	0.002	(110)	563	380
4	1.10	-0.004	(100)	545	>400
5	1.25	-0.011	(100)	527	>400

**Method.** To determine real and imaginary parts of the permeability of toroidal ferrite samples, the two-coil method described earlier [11] was used. Using the internal built-in oscillator for the ac driving field in the 100 Hz to 10 kHz frequency region a Two Phase Lock-in Analyzer EG&G model 5206 served for the measurements of both the in-phase and quadrature components of a signal induced in the secondary coil. In order to measure the true initial permeability values in the Rayleigh region of the magnetisation curve, the amplitudes of the magnetic driving fields were kept below 1 A/m ( $< 10$  mOe). Low temperature measurements were realized using a liquid helium continuous-flow cryostat; higher temperatures were attained in a furnace. More details about the method and the set-up used can be found in [11].

**Results.** In Figs. 1 to 5, the temperature dependences of the real and imaginary parts of the initial permeability measured at 1 kHz are represented. All samples were first heated above their Curie temperatures  $T_C$  to attain demagnetized state. Curves in Figs. 1 to 5 were then obtained by measuring  $\mu$  in decreasing temperatures. The high-temperatures behaviour of  $\mu'$  and  $\mu''$  indicates the presence of the maxima in  $\mu$  occurring just below the Curie temperatures [12].

In magnetite (see the right hand part of Fig.1) the other maximum lies near the Verwey transition temperature,  $T_V=121$  K. A more

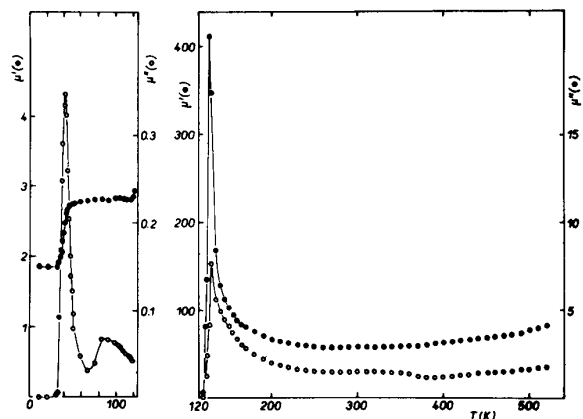


Fig 1. Temperature dependences of the real ( $\mu'$ ) and the imaginary ( $\mu''$ ) parts of the initial permeability of magnetite (sample 1).

detailed investigation showed [11] that the maxima of  $\mu'$  and  $\mu''$  are reached at  $T_k = 126$  K (independent of the frequency) followed at  $T_v$  by a two orders of magnitude decrease. At still lower temperatures (see the left hand part of Fig.1)  $\mu'$  shows a small kink at 90 K followed by a sharp decrease near 40 K. At the same temperatures  $\mu''$  passes through maxima: a small and broad one at 90 K and a sharp one at 40 K. Both maxima are frequency dependent, pointing out the presence of relaxations.

Sample 2 ( $x=0.52$ ) displays a somewhat similar behaviour - see Fig. 2a. After passing the high temperature maxima just below  $T_c$  both components of  $\mu$  display a second narrow dispersionless maximum near 340 K followed by broad features persisting to low temperatures. Other sharp maximum and a shoulder in  $\mu''$ , both frequency dependent, may be observed (see the arrows in Fig. 2a) at 20 K and 60 K respectively.

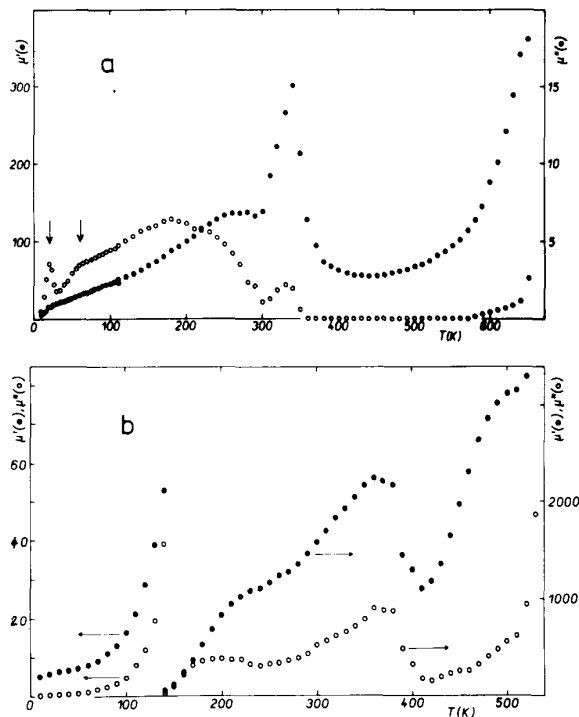


Fig. 2a,b. Temperature dependences of the real and the imaginary parts of the initial permeability of the sample 2(a) and 3(b). See Table 1 for sample details.

One peak and two shoulders in the temperature dependences of  $\mu$  for sample 3 ( $x=0.95$ ) are seen in Fig. 2b. Shoulders at 200 K and 500 K shift with frequency while the maximum near 360 K is found to be frequency independent whereas its magnitude appears to be time dependent.

Only one additional maximum (besides that just below  $T_c$ ) can be recognized in the temperature dependence of  $\mu$  of samples 4 and 5 - see Fig. 3a,b. The maxima in both samples were found to be frequency dependent. No other anomalies were found in the temperature behaviour of  $\mu$  except a slight increase of  $\mu''$  below 30 K for sample 5.

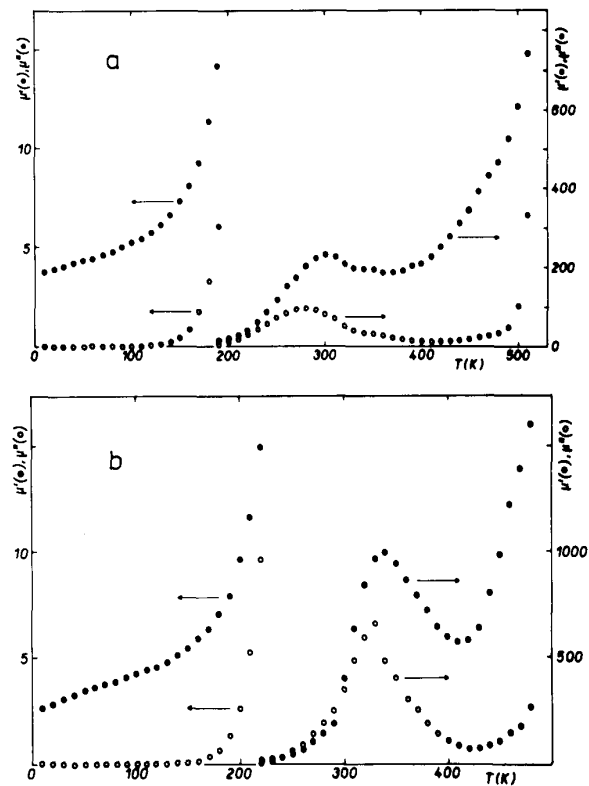


Fig. 3a,b. Temperature dependences of the real and the imaginary parts of the initial permeability of the sample 4(a) and 5(b). See Table 1 for sample details.

### Discussion

**Initial permeability behaviour.** The increase of the initial permeability near  $T_c$  (i.e. the primary maximum) is well described [1,2]. More interesting is the occurrence of the second maximum in  $\mu$  at temperatures well below  $T_c$ . One of the possible reasons is the passing of the anisotropy constant  $K_1$  through zero. Sharp maxima in  $\mu'$  and  $\mu''$  in magnetite (sample 1) near  $T_v$  could indeed be conceived in terms of  $K_1=0$  occurring at  $T_k=126$  K [7]. However, the steep decrease by two orders of magnitude is more likely to be connected with the crystallographic and magnetic transition occurring at  $T_v=121$  K. The two other anomalies below  $T_v$  have a quite different character and cannot be explained by supposing  $K_1=0$ .

In the case of sample 2 ( $x=0.52$ ), sharp maxima near 340 K are also in agreement with the estimated temperature range for  $K_1=0$  values [14]. The same type of explanation is also valid in the case of sample 3 ( $x=0.95$ ) where the extrapolated temperature for  $K_1=0$  is about 370 K [14] in good agreement with the observed dispersionless maxima of  $\mu$  at 360 K. The broad shoulder in  $\mu'$  and a maximum in  $\mu''$  below 200 K have obviously a different, frequency dependent character.

No reliable data on the high temperature behaviour of  $K_1$  in manganese-rich part of the system are available. The extrapolation following [14] would yield  $T_k$  approaching  $T_c$ .

