THE SMALL LOCAL MAXIMA AND MINIMA IN THE MAGNETIC LOSS VERSUS TEMPERATURE CURVES OF IRON AND ITS ALLOYS

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The paper deals with the appearance of small local maxima and minima in the curves of the temperature dependence of magnetic losses of iron and some iron alloys. A number of factors which may bear some effect on this phenomenon are looked into.

1. *Introduction*

Small local maxima and minima in the curves of the temperature dependence of magnetic losses were observed for the first time during experiments described in Ref. [1], which dealt with permeability disaccommodation in silicon steels containing from 2.5 to 3 per cent Si and up to 0.02 per cent C in solid solution form. In those experiments the samples were magnetized by a field of about 5 mOe having a frequency from 40 to 200 Hz. Since there were some doubts as to whether these maxima and minima actually did exist (*e. g.*, see [2]), measurements of magnetic losses were repeated for yet another sample of silicon steel (2.73% Si, 0.023% C) with much better accuracy and smaller temperature increments [3]. The outcome was very distinct, small maxima and minima lying above the temperature of the Snoek peak. At the same time, this phenomenon was observed in the case of several samples of pure iron (99.9% Fe) containing about 0.02 per cent carbon [4]. In all three quoted papers it was believed that the small local maxima and minima originate from the relaxation peaks shifting towards the higher temperatures with increasing frequency of the magnetizing current [1, 3, 4]. It was found not long ago, however, that the position of the examined peaks and points of inflection are, in practice, independent of temperature. In consideration of this unequivocal fact, new measurements were performed for several samples of pure iron and iron alloys containing C, or C and N, in solid solution form.

2. *Samples*

Investigations involved three samples of electrolytic iron (99.99% Fe, not counting C, N and O), described in detail in Ref. [5], one sample made of the same iron with 0.12% aluminum content, and three samples of silicon steel. Of the latter, two were made from 99.8% pure iron and contained 0.90% or 2.73% of silicon, while the third, of semitechnical origin,

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contained 3.2% of silicon. In the pure iron and silicon steel samples (the latter made from 99.8% pure iron) there was about 0.02% C and smaller quantities of N. The Fe-Al samples, on the other hand, contained 0.02% C, 0.011% N and traces of Al_2O_3.

The average grain size of all samples, except Fe —3.2% Si, was from 500 to 1000 μ^2. The sample of semitechnical origin had very large grains, of an average area of 70 mm^2.

In a majority of cases the samples were investigated in the stress-relieved state. For this purpose, the Fe and Fe—Si were heat treated in a vacuum furnace at a temperature of about 700°C for several hours, and then cooled slowly together with the furnace (in the 700°C to 300°C range the mean cooling rate was approximately 1°/min). The Fe-Al sample was initially annealed at 900°C for five hours, then cooled with the furnace to 700°C and annealed at this temperature for two hours, and finally cooled together with the furnace to ambient temperature. It is common knowledge that after such heat treatment an Fe-Al sample does not contain nitrogen in the form of solid solution [6, 7].

3. Procedure

The measurements of the temperature dependence of magnetic losses were performed by means of a Maxwell-Wien type bridge, described in Ref. [5], in the temperature range from +45° to +170°C. The temperature of the samples was stabilized with an accuracy better than ±0.1°C. in a thermostat with air circulation. In most cases, once the temperature of the sample became settled, the sample was demagnetized during several seconds by a variable field of frequency 50 Hz and maximum amplitude of the order of several oersteds; this field was generated by means of the electronic device described in Ref. [8]. The sample was magnetized by a field of acoustic frequency and strength of 0.5 to 8 mOe. The bridge was balanced in a continuous manner and 45 seconds after initiation of demagnetization its indications were read. Each measurement was repeated several times. The resistance of the sample's windings was determined by means of a precision Wheatstone bridge at each temperature.

The classical discussion of error of a single measurement showed that the absolute error of the tangent of the angle of losses does not exceed ±0.0002. Experience with many measurements shows that this quantity is not grater than ±0.0001.

4. Results of measurements

Investigated was the influence on the small local maxima and minima of a number of factors, either associated with the procedure of the experiment, or enabling the determination of the kind of magnetic losses which are being dealt with. Moreover, an attempt was made to find the relationship between the extrema and the actual structure of the examined samples.

4.1. Influence of experimental procedure

As already mentioned, owing to the necessity of achieving good reproducibility of measurements, the sample was demagnetized before each measurement. Therefore, the effect of this operation on the small local maxima and minima was investigated first.

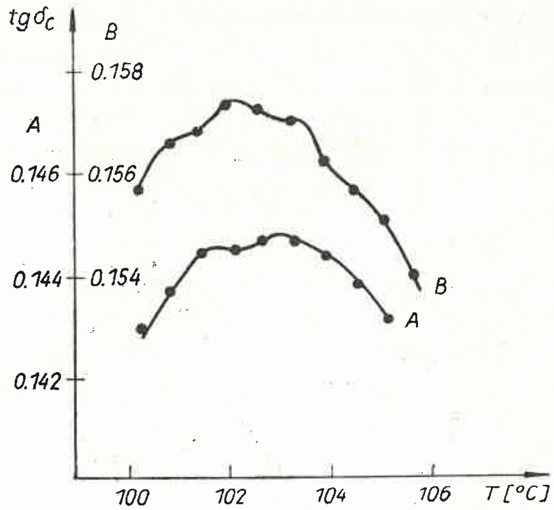

Fig. 1. Graph of temperature dependence of magnetic losses: Fe—Al sample; $f = 165.5$ Hz; $H = 2.8$ mOe; A – without demagnetization; B – with demagnetization

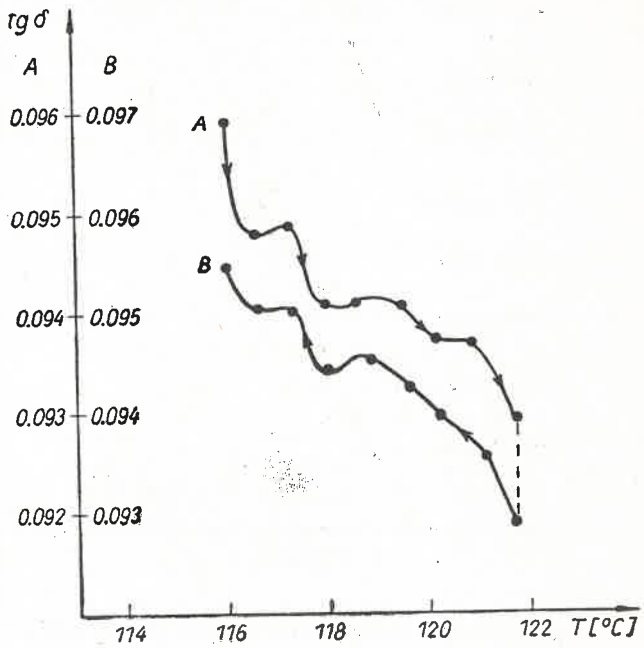

Fig. 2. $\tan\delta = f(T)$ in the case of rising (curve A) and dropping (curve B) temperature; Fe sample; $f = 65.7$ Hz; $H = 5.4$ mOe

For this purpose tan $\delta = f(T)$ curves were determined in a definite temperature range in two cases:

1. with the sample demagnetized only once, half an hour before the first measurement, and
2. with the sample demagnetized before every measurement.

In the former case the ohmic resistance was measured only at the lowest and highest temperatures. For the intermediate temperatures the value obtained by linear interpolation was assumed. It is seen in Fig. 1 that loss curves obtained thus have quite similar shapes.

The loss measurements were usually made at increasing temperatures. With this in mind, a check was made to see whether the small local maxima and minima also appear in

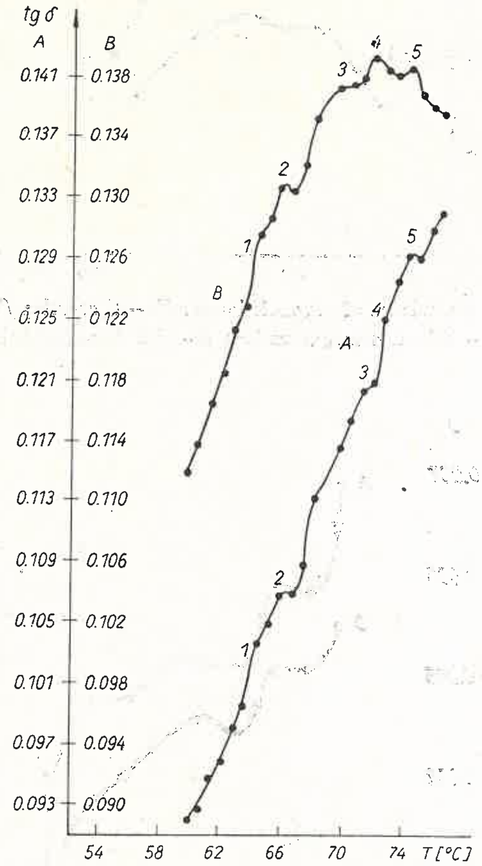

Fig. 3. tan $\delta = f(T)$ curves for two frequencies of magnetizing current; A – 165.1 Hz; B – 65.8 Hz; Fe sample; $H = 5.4$ mOe

curves taken during temperature decreases. The results, presented in Fig. 2, show that small local extrema do appear on both curves. The distinct maximum occurring at 117.3°C is seen to appear in both procedures.

4.2. Influence of frequency and strength of magnetic field

The losses which are determined in weak magnetic fields may have their origin among the following four phenomena: 1. magnetic hysteresis, 2. eddy currents, 3. diffusion after-effects, and 4. fluctuation after-effects. In order to establish the origin of the small local maxima and minima, the effect of the frequency and strength of the magnetic field on this phenomenon was looked into.

For this purpose, the dependence tan $\delta = f(T)$ was determined for one of the pure iron samples in the range from +46° to +76°C at two frequencies of the field. Figure 3 depicts a part of the obtained curves. It is seen that the first extremum, at +64°C, appears identically in both cases. The other maxima and minima also mutually repeat themselves at the same temperatures.

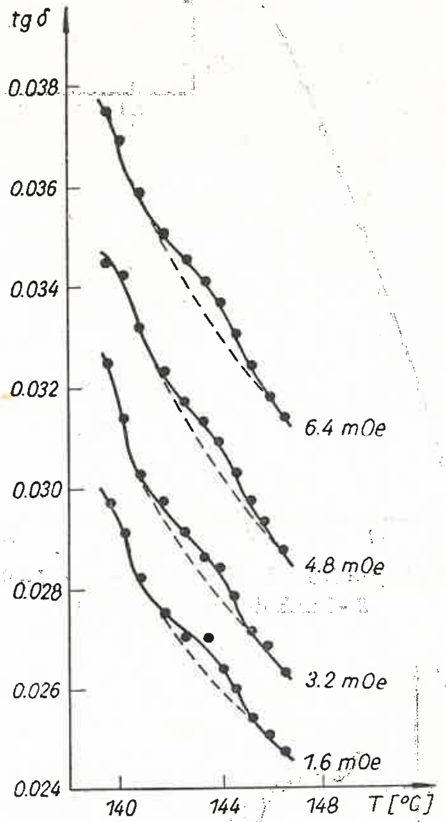

Fig. 4. Loss curves for four magnetizing fields; Fe—Si sample (2.73% Si); f = 65.8 Hz

The same result was obtained for another iron sample and for the iron-aluminum sample. In consideration of this, all previous measurements were inspected, and it was found that in the case of more densely distributed temperatures and in the more accurate measurements the maxima an minima in the curves taken at various frequencies fall at temperatures differing from each other, in general, by less than one degree Celsius.

All of these results imply that the position of a small extremum does not depend on the frequency of the magnetizing field.

Very much attention was devoted to theinfluence of the magnetic field strength. In all cases, even at the lowest applied strengths (of the order of 0.5 mOe), small local maxima and minima

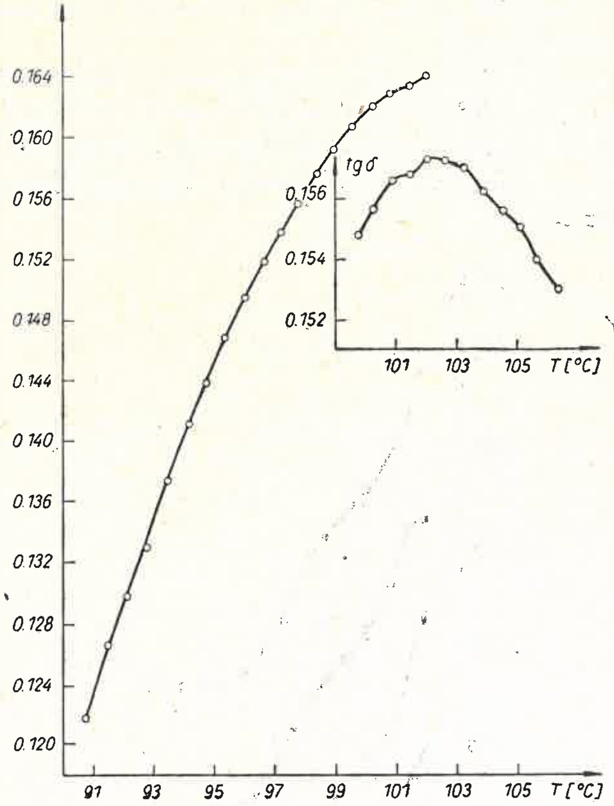

Fig. 5. tan $\delta = f(T)$; Fe—Al sample; $f = 165.5$ Hz, $H = 2.8$ mOe; curve *1* – third day of measurement; curve *2* – fourth day of measurement

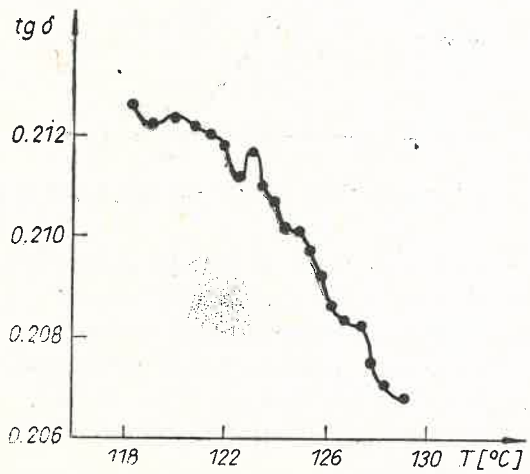

Fig. 6. Temperature dependence of magnetic losses for Fe–Si sample (3.2% Si) of very large grain; $f = 165.8$ Hz; $H = 2.8$ mOe

were observed at temperatures differing by 0.5°C at most (*e. g.*, Fig. 4). Unfortunately, a satisfactory extrapolation of the tangent of the angle of losses to zero field has not been achieved yet. In the examined temperature range there is disaccommodation of magnetic permeability in very low-strength fields which, in the case of Fe and Fe−0.1% Al, had been studied in detail earlier [9, 10, 11]. It gives rise to a non-linear character of the tan δ vs H dependence in weak fields [12]. At the same time the slow, but distinct, structural changes taking place in the sample during measurements at a fixed temperature did not permit the use of a greater number of measuring fields.

The only results which seem to indicate a linear dependence of tan δ on H in the 0.44 to 2.64 mOe range were obtained for the Fe−0.1% Al sample. In this case, linear extrapolation to zero field by the least squares method gave a curve with distinct local extrema at the same temperatures as for the individual measuring fields. It should be stated here, however, that in a sample containing aluminum deviation from linearity will infallibly appear at fields lower than 0.44 mOe.

It was also found that subtraction of losses associated with eddy currents does not affect the phenomenon of small local maxima and minima. The results discussed in the sub-

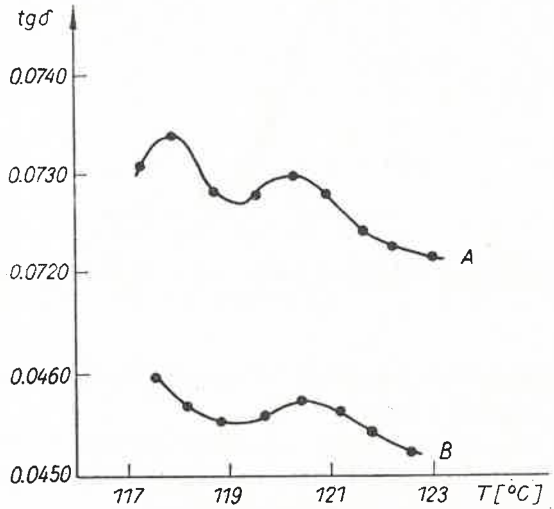

Fig. 7. Losses before (curve *A*) and after rolling (curve *B*); plastic working 0.4%; iron sample; $f = 65.8$ Hz; $H = 5.0$ mOe

sequent section also provide evidence that this phenomenon is not caused by fluctuation after-effects, for losses due to this type of effect depend only very slightly on the presence of carbon and nitrogen in the solid solution.

4.3. Influence of actual structure of examined samples

With an eye to the determination of the origin of the small local maxima and minima the effect of a number of structural factors on this phenomenon was investigated.

In the case of the sample of pure iron for which the first extremum was obtained at 64 °C (Fig. 3) an analysis of the relaxation losses associated with Snoek relaxation, performed by the method described in Ref. [10], showed that in the solid solution there are approximately the same quantities of nitrogen and carbon. Figure 5 present a part of the curve obtained for Fe-Al. In this case, the first extremum appeared at 101°C. In the range from +60°C to +101°C (every 0.6°C) not even the least deviation from a monotonic curve was seen.

Also looked into was the influence of the mean size of the grain. All of the results presented hitherto were obtained for fine grained samples. The earlier studies also made use of this type of material [1, 3, 4]. Figure 6 depicts the results of one of two measurement series obtained for an Fe-Si sample which had an average grain size of 70 mm². We see that the

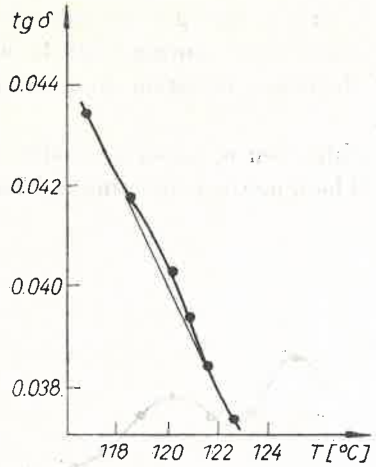

Fig. 8. Graph of tan $\delta = f(T)$; iron sample hardened in water with ice after heat treatment at 500°C; $f = 65.8$ Hz, $H = 8.2$ mOe

small local extrema also appear in the almost monocrystalline sample. (The sharp maximum at 123°C showed up in both series.)

It was also found that extrema appear after plastic working by cold rolling (Fig. 7) and after hardening the sample in water with ice following heat treatment at 500°C (Fig. 8); the sample was placed in a pumped off quartz tube.

5. *Discussion of results*

The results of the present work are a full corroboration that the small local maxima and minima in the curve of the temperature dependence of magnetic losses are an excellently reproducible physical phenomenon which occurs in samples of iron and iron alloys crystallizing in the body-centered cubic system. For example, in the case of Fe–0.1% Al the first maximum at 101°C (Fig. 5) was obtained many times. The extrema appear independently of demagnetization of the sample before the measurements (Fig. 1). They appear when the temperature is decreasing as well as when it is increasing (Fig. 2). Experience with a multitude of measurements indicates, however, that at longer measuring times, and also when the measurement is a repeat, the extrema are less dinstinctly separated from the monotonic loss curve. This suggests that there undoubtedly is a certain effect of long-duration heating on this phenomenon.

The results presented in Fig. 3 imply that the small local maxima and minima are not directly relaxation peaks with time constants obeying Arrhenius' law. For extrema of relaxation origin shift considerably with a change in the frequency of the magnetizing field.

Investigations on the influence of the magnetizing field strength did not give unambiguous results. At the present moment, the data from measurements indicate the possibility of one of the two following eventualities:

1. The curve obtained by means of proper extrapolation to $H = 0$ does not have any local extrema. In this case the examined phenomenon would consist in a change of losses originating from magnetic hysteresis.

2. Small extrema appear on the curve extrapolated to zero field. Then, the losses corresponding to the extrema would be indirectly associated with relaxation processes taking place in the same interval of temperatures.

In the latter case, as ascertained for Fe—0.1% Al, the difference between the losses at a definite field and without field, representing the losses dependent of field, hence, primarily losses linked with hysteresis, also show small local maxima and minima; they are, however, shifted with respect to the extrema in mention up to now. It is seen thus that in the discussed case the phenomenon under examination would be simultaneously caused by relaxation and hysteresis.

Very interesting conclusions are arrived at from the data discussed in Sec. 4.3. As it does not seem plausible that the shift by almost 40°C in the position of the first peak (Figs 3 and 5) is caused by the 0.1 per cent addition of Al, it is seen that the small local maxima and minima are closely linked with the presence of carbon and nitrogen in solid solution form. Nitrogen gives rise to an extremum already at +64°C, whereas carbon at +101°C.

Small local extrema of nearly the same intensity appear in both fine grained and almost monocrystalline samples. They are observed also after plastic working, and in hardened samples.

Results obtained up to now do not allow a detailed mechanism of the formation of the small local extrema to be given yet. Notwithstanding, it seems that the new results corroborate the hypothesis put forth in Refs [3, 4] that the examined phenomenon is associated with the presence of Guinier–Preston type precipitation zones, being segregations of C or N atoms, but with a lattice cohesive with the lattice of the matrix. These results show, however, that the nature of the phenomenon is not just the appearance of new relaxation peaks associated with jumps of interstitial atoms between vacant octahedral sites located in the vicinity of the zones.

Recently, in order to give an explanation of the non-monotonic shape of the curves of coercive field strength against ageing time in hardened Fe—N samples, a hypothesis was put forth that in this material the nitrogen atoms become deposited in whole layers on the existing plate-shaped coherent zones [13]. It appears that in the samples examined in this work this kind of phenomenon may be, within narrow temperature intervals, a positive or negative source of C or N atoms which constitute relaxation centers in the relaxation process, *e.g.* of the Snoek type. On the other hand, it may at the same time cause discontinuous changes in losses linked with magnetic hysteresis.

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