THE TEMPERATURE DEPENDENCE OF THE PHOTOMAGNETIC EFFECT IN YIG(Si^{4+})

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Experimental results are given on the dependence of the irreversible photomagnetic effect on temperature for various polycrystalline samples of $Y_3Fe_{5-x}Si_xO_{12}$, and they are interpreted in terms of a previously introduced model. According to this it is possible to explain the behaviour after finishing the irradiation (recombination phase) as well as the vanishing of the effects above 200 K by a number of relaxation processes with different activation energies which simultaneously occur after disturbing the thermal equilibrium by irradiation.

ТЕМПЕРАТУРНАЯ ЗАВИСИМОСТЬ ФОТОМАГНИТНОГО ЭФФЕКТА В YIG(Si^{4+})

В работе приведены экспериментальные данные о температурной зависимости необратимого фотомагнитного эффекта в разных поликристаллических образцах системы $Y_3Fe_{5-x}Si_xO_{12}$ и дается их интерпретация на основе ранее сформулированной модели. На основе этой модели, которая учитывает несколько релаксационных процессов с различными энергиями активации, протекающих одновременно после нарушения термического равновесия облучением, удалось объяснить не только поведение материала после облучения (фаза рекомбинации), но также исчезновение данного эффекта при температурах свыше 200 K.

1. INTRODUCTION

The existence of small amounts of Fe^{2+} in polycrystalline YIG leads to some effects which have not been understood in detail until now. This is above all the case of the photomagnetic effect (PME) and of the magnetic after-effect (MA). Both influence the initial permeability and depend on several parameters in a complex way:

— The irradiation sensitivity of the material increases with the growing Fe^{2+}-content up to a maximum at $x_m \approx 0.014$. x is the content of Fe^{2+} per formula unit. In Si-doped samples large loss factor maxima spontaneously (without irradiation) arise at higher dopings ($x > x_m$) and therefore the initial permeability is

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reduced drastically at 77 K. Simultaneously the tendency of the material to assume the original state before irradiation diminishes with the increasing Fe^{2+} content (reduced recombination).

— The irradiation effects are not only a function of the transmitted energy but they depend on how the irradiation takes place (intensity, irradiation time).

— The effects mentioned become sensitive to temperature changes above 77 K and vanish on warming the sample to 200 K.

One cannot expect that the whole complicated behaviour can be deduced from simple assumptions. The concept that there are only two clearly distinguishable types in the arrangment of the divalent iron as neighbours of the Si^{4+} or of oxygen vacancies is not sufficient for the YIG. This was expressed already in [1]. The existence of Fe^{2+} influences the anisotropy in YIG as is well known from a series of papers [2]. An extremely large change of initial permeability in the order of magnitude of 80 % due to light can be presumed in the concentration range ($x \approx 0.005$) if the resulting anisotropy deviates from the cubic type. The major fraction of the light-induced change of permeability in YIG(Si) can be attributed to the formation of a diffusion anisotropy if very large effects are observed [3]. On account of the variety of possible arrangements of the Fe^{2+} with respect to donor centres in the range of low concentrations a formal distinction between types with and without uniaxial symmetry (K_u) is possible, the formation of K_u influencing the permeability essentially only if the time constants for the rearrangement of the particles are small enough, that means, if the diffusion for the rearrangement is not frozen in. The existence of such processes in YIG in the low temperature range was proved already in [4], and it was found, for example, in [5], that even at 77 K a rearrangement of particles with an average relaxation time of 10^{-4} s takes place manifesting itself in a magnetic after-effect.

If the irradiation causes an electron transfer which results in a rearrangement of the Fe^{2+}, then positions are temporarily occupied due to the irradiation which do not correspond to the thermal equilibrium state and which give a higher contribution to the energy of the crystal. In [6] we tried to explain the effects with the following assumptions for the population of a-sites, which will be occupied under the influence of irradiation:

1. The potential energy W_i of an a-site (i) increases with the growing distance r_i ($r_{i+1} > r_i$) from the donor centre (by $-1/r_i$), but the potential depth diminishes. $i = 1 \ldots k$ denotes the No. of the type of site with a distance r_i from the centre.

The more Fe^{2+} is present the larger is the probability that in the equilibrium state a-sites with larger distances ($i > 1$) from the centre are occupied.

2. The population of the sites ($i = k$) with a maximum distance r_k from the donor centre, the smallest potential depth, is connected with the formation of a MA with a small time constant. That means, the energy level of the k-sites is split by the instantaneous direction of the spontaneous magnetization.

The irradiation causes an occupation of the sites of maximum potential energy ($i = k$) together with a decrease of the population at the sites which are occupied in the thermal equilibrium state (for example, $i = 1$). After switching off the irradiation the stepwise reoccupation of the sites $i < k$ occurs spontaneously with a population rate which strongly diminishes with decreasing i, because the depth of the potential wells grows, that means, the migration from sites k to sites $k-1$ takes place more quickly than from $k-1$ to $k-2$, etc. Denoting the population rates by $\dot{n}_i$ (n_i number of particles on the level i) we have taken as a basis the following equations assuming thermally activated electron hopping:

$$\dot{n}_i = -n_i (N_{i-1} - n_{i-1}) C_i + n_{i+1} (N_i - n_i) C_{i+1} + \\ + [(N_i - n_i) n_{i-1} C_{i-1} - n_i (N_{i+1} - n_{i+1}) C_i] \tag{1}$$

$$C_i = C \exp\left\{-\frac{1}{kT}(W_b - W_i)\right\} \tag{2}$$

for the recovery process, where N_i represents the number of available sites (i), C is a phenomenological constant and W_i the height of the i-th potential minimum above an arbitrary level. The potential maxima W_b (or barriers) were assumed to be independent of r after the example of [4]. C_i denotes the diffusion velocity by electron hopping which results in a migration of the Fe^{2+} from the i-th level to the other levels. At a constant temperature $C_i > C_{i-1}$, because $W_i > W_{i-1}$. That means, after switching off the irradiation the migration of the particles from the k-level to the $(k-1)$-level takes place very quickly at first but slows down very soon because the migration from the level $k-1$ to level $k-2$ occurs much more slowly. The particles accummulate on the upper level before they go to their equilibrium positions more slowly.

For the irradiation process we assume the equation:

$$\dot{n}_i = -\beta_i I (N_k - n_k) n_i + \text{all terms from (1)}$$

$$\dot{n}_k = \sum_{i=1}^{k-1} \beta_i (N_k - n_k) n_i + \text{all terms from (1)} \qquad i = 1 \ldots k-1 \tag{3}$$

I is the intensity of the light and the constants β_i are a measure for the sensitivity to irradiation depending on the potential depth, but they will be regarded as temperature independent.

This picture will come the closer to reality the higher the temperature, while for low temperatures the probability of transitions due to tunnelling increases and does not correspond to equation (2). On this basis we could describe the measured dependences semi-quantitatively in [6] and [7] by the system of differential equations (3) for the population rates. The dependence of the effects on temperature results from varying the constants C_i after equation (2). In this paper such

dependences are shown as measured on YIG and are compared with the predictions of the model just mentioned.

II. METHOD AND RESULTS

As in former papers [8, 9] small, thinly ground polycrystalline toroids with a varying Si concentration were used for the experiments. The permeability was determined in a weak, constant alternating magnetic field of 20 kHz or 10 kHz by measuring the coefficient of mutual induction with a bridge or by automatically recording the secondary voltage.

We carried out two kinds of experiments:

(a) experiments with gradually changing temperature from room temperature down to 77 K (Figs. 1, 2),

(b) experiments under isothermal conditions (Figs. 3, 4, 6, 8).

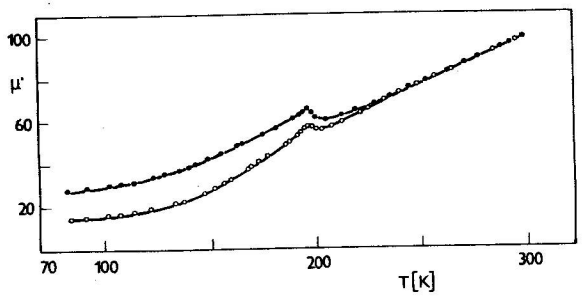

Fig. 1. Gradual cooling down of an YIG sample, $x = 0.012$, from 300 K to 80 K. × — intial permeability μ_d in the dark, O — initial permeability μ_{irr} under irradiation with incandescent light.

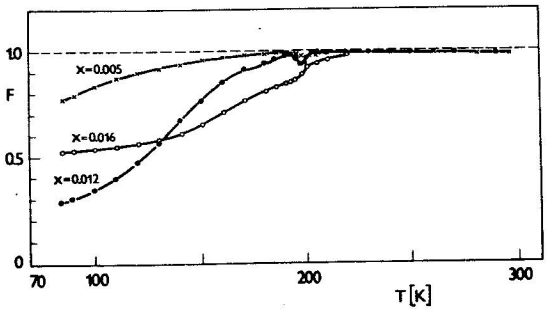

Fig. 2. Cooling down as in Fig. 1. Permeability divided by its value in the dark, $F(T) = \mu_{irr}(T)/\mu_d(T)$, for samples with different Si contents. For all these samples no appreciable influence by irradiation can be found above 200 K.

The variation of initial permeability by cooling down to low temperatures is shown in Fig. 1 for a sample with a strong PME ($\chi = 0.012$) with and without irradiation (μ_{irr} and μ_d, respectively). Under the given conditions below 210 K an influence of irradiation is found and a secondary maximum of permeability which is due to disaccommodation brought about by thermal demagnetization, but which has only little to do with the origin of the PME [10]. Qualitatively, the $\mu(T)$-curve does not

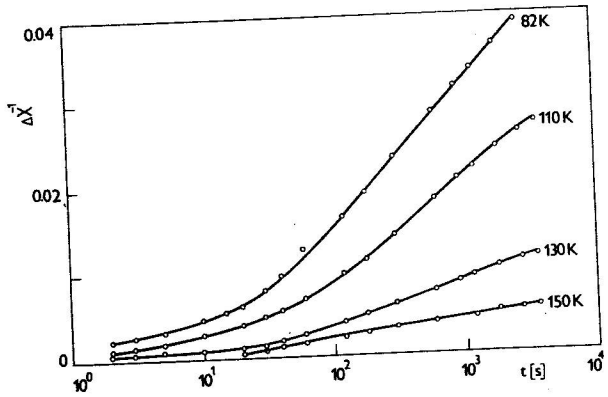

Fig. 3. Change of magnetic stiffness, $\Delta(1/\chi)$, during irradiation of a sample with $x = 0.014$ at constant temperatures. t = irradiation time.

differ for samples with different x' s. In Fig. 2 the ratio μ_{irr}/μ_d was used for an understandable representation in order to eliminate the influence of individual sample properties as much as possible and thus to enable a comparison of the different samples. At the lowest temperature, 85 K, one finds the sample with $x = 0.012$ to be the most sensitive of the series shown, while weaker or stronger dopings lead to a smaller PME [9]. In the range of higher temperatures the conditions are different. It can be clearly seen that with increasing doping a shift of the temperature range takes place where the PME vanishes. The reason for the breakdown of the PME at higher temperatures is the more and more rapid dissociation of the photomagnetically active centres (recombination), which was already studied for samples with varying doping [6] and which takes place more slowly with increasing doping.

In fig. 3 the change of magnetic stiffness, $\Delta(1/\chi)$, is given as a function of the irradiation time t for a sample with a strong PME ($x = 0.014$). In order to get this the sample was gradually cooled down from room temperature to the desired temperature and then irradiated under isothermal conditions. It can be seen that the effect is not finished even after a long time. We must suppose that a series of elementary processes are superimposed which are distributed over more than

3 orders of magnitude in time. Applying higher temperatures the influence of irradiation diminishes essentially and cannot be determined above 200 K in the present experiments.

The behaviour of the material after switching off the irradiation is demonstrated in Figs. 4a and b, which are directly correlated to the experimental conditions applying to Fig. 3. Here the increase of permeability after removing the irradiation

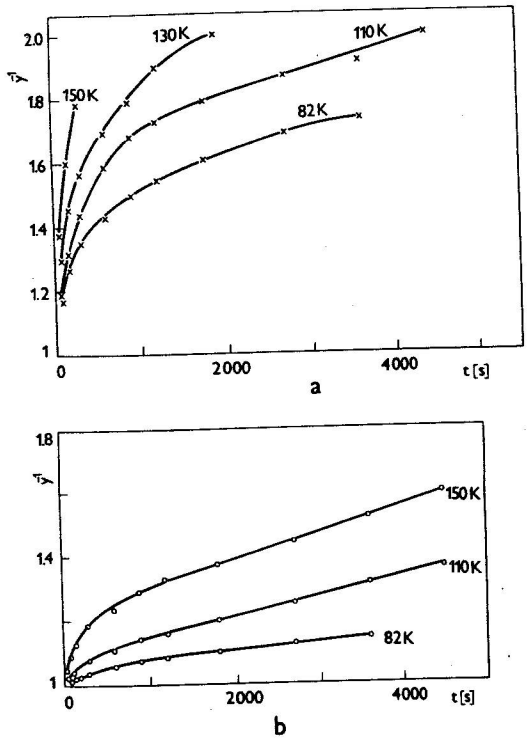

Fig. 4. Recovery phase. Ratio $(\Delta 1/\chi)_{irr}/(\Delta 1/\chi)_{rec} = y^{-1}$ after switching off the irradiation which had been carried out under the conditions of Fig. 3. a) irradiation time $t_{irr} = 30$ s b) irradiation time $t_{irr} = 1$ h.

was measured (with the temperature kept further constant). The irradiation time was only 30 s in the one series after Fig. 4a and 120 times longer in the other one after Fig. 4b. For the representation of the recovery process (recombination) we used the quantity y^{-1}:

$$y = \frac{\Delta(1/\chi)_{rec}}{\Delta(1/\chi)_{irr}}, \qquad (4)$$

$$\Delta(1/\chi)_{rec} = 1/\chi_{irr} - 1/\chi(t)$$

denotes the change in stiffness during the recovery phase and

$$\Delta(1/\chi)_{irr} = 1/\chi_{irr} - 1/\chi_d$$

is the change in stiffness reached before by irradiation. The deviation from linearity of the function $y^{-1}(t)$ was already discussed in former papers [7, 11]. The diffusion after-effect character of recombination finds its expression in the strong temperature dependence. Thus it is obvious that the breakdown of the irradiation effect by warming up to about 200 K is of dynamic origin. That means, the diffusion effects which cause a dissociation of the photomagnetic centres take place more quickly if warming progresses until a state is reached where the diffusion is finished in a shorter time than the recording of the permeability occurs and finally the diffusion time constants become smaller than the period of the applied measuring field.

With short irradiation times (Fig. 4a) processes with time constants smaller than at a longer irradiation (Fig. 4b) govern the initial phase of the recovery. It is not possible to explain the experimental results by a single process with only one time constant [7], but a large number of elementary processes are involved.

III. DISCUSSION AND CONCLUSIONS

The explanation of the results is possible on the basis of the dynamic model summarized in the introduction. For the temperature dependence it is necessary to determine the activation energies W_i after equation (2). The fitting of the other constants of the differential equation system (3) for a possibly quantitative description of the processes was assumed from our former results.

— We neglect the terms in square brackets in equation (1), that means transitions from the lower levels i to the higher levels $i+1$.

— We limit our considerations to only $k = 11$ different elementary processes (with 11 time constants). (The choice $k = 11$ is an arbitrary one and k was only limited by the storage capacity of the available computer.)

— The irradiation sensitivites β_i increase with the growing distance from the centre (for example in a geometric series $\beta_i/\beta_{i-1} = 2$).

— The intensity parameter I and the sensitivities β_i were fitted to the experiments in such a way that the results (Fig. 5) are qualitatively well given in a large interval of irradiation time ($\beta_{k-1} I/C_k = 0.3$ was appropriate for 77 K).

In order to deal with the temperature dependence we determined the W_i as follows:

— The height of the potential bottom varies as $1/r$ (more exactly as the potential of two point charges with the distance d, which is due to an 0 defect and a Si^{4+}).

— The potential barrier is constant for simplicity.

— The maximum depth of the wells is $W_1 = 0.5$ eV, the minimum $W_k = 0.1$ eV.

For simplicity it was assumed that before the beginning of the irradiation, in the equilibrium state, the Fe^{2+} is equally distributed among all levels (1 ... $k-1$), the level k is not occupied.

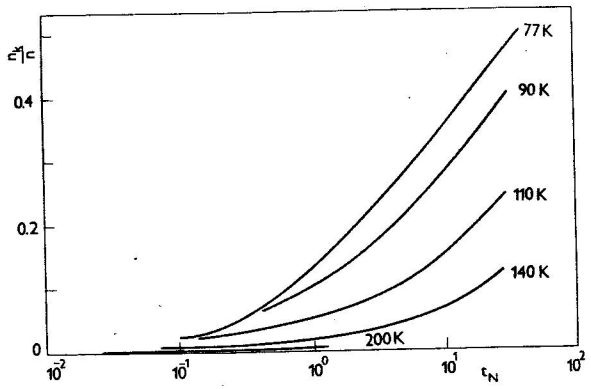

Fig. 5. Calculated function $n_k(t_N)/n$, which is proportional to the change in stiffness. t_N is a measure for the irradiation time.

— The constants of light sensitivity, β_i, do not depend on temperature.

The numerical solution of the system of differential equations (3) gave the results according to Fig. 5. The change in stiffness is proportional to the population n_k on the k level

$$\Delta(1/\chi) = C\frac{n_k}{n} \tag{5}$$

(n = total Fe^{2+} content). The quantity n_k/n therefore can be regarded as a "normalized" change in stiffness. $t_N = C_k n t$ represents a time scale which is proportional to the time t and the choice of which has no influence on the form of the graphs (Fig. 5). The graphs in Fig. 5 are essentially similar to those found in the experiments (Fig. 3). The great influence of temperature on the irradiation curves between 77 K and 200 K arises from the drastic diminishing of the intensity parameter $\beta_{k-1} I/C_k$ with an enormously growing migration velocity C_k (β_i all assumed to be temperature independent).

This shows that the vanishing of the PME towards higher temperatures is caused by the quicker migration of the Fe^{2+} from the photomagnetically active centres even if the constants of light sensitivity (β_i) are assumed to be temperature independent.

The same situation finds its expression in Fig. 6. The reduction of the susceptibility $\Delta\chi$ was measured on a sample ($x \doteq 0.0075$) under the influence of irradiation

similar to that in Fig. 1 but under isothermal conditions, that means, the sample was cooled down to the low temperature in the dark each time before it was irradiated. In order to construct the solid line the theoretical results in Fig. 5 were used, where the fitting of the χ-change was carried out at 77 K (about 50 % of the value in the dark). A qualitatively good agreement is seen above 77 K, demonstrating the break-down of the irradiation effect above 200 K, as could already been seen in Figs. 1, 2. Below 77 K the deviation becomes larger because the effects do not freeze in as quickly as equation (2), which underlies the calculations predicts.

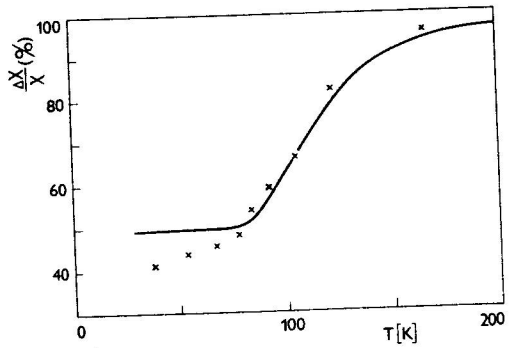

Fig. 6. Relative change of permeability $\Delta\chi/\chi$ under the influence of irradiation between 40 K and 200 K. 100 % corresponds to the permeability without irradiation μ_d. × — experimentally found dots for a sample $x = 0.0075$. The solid line was calculated.

The differences which can be seen in Fig. 2 for various dopings also confirm the conclusions from Fig. 6.

Solutions of the differential equation system were analysed further for the case when the irradiation had been finished ($I = 0$) after a time t_N^{irr} and the recovery of the material began. Figs. 7a, b, c show the results for three different assumed normalized irradiation times t_N^{irr}. Since equations (4) and (5) give the connection between the calculated n_k and the experimentally determinable y^{-1}, for the recombination process the ratio

$$y^{-1} = \frac{n_k(t_N^{irr})}{n_k(t_N)} \tag{6}$$

was plotted versus the normalized time t_N. The scale for t_N which had been arbitrarily fixed for the assumed preliminary irradiation (Fig. 5) is identical for both processes. If one compares Fig. 4 with Fig. 7 one finds that the total behaviour is qualitatively well given by the calculated functions. The large number of simplifications which underly the calculations, does not enable an exact agreement

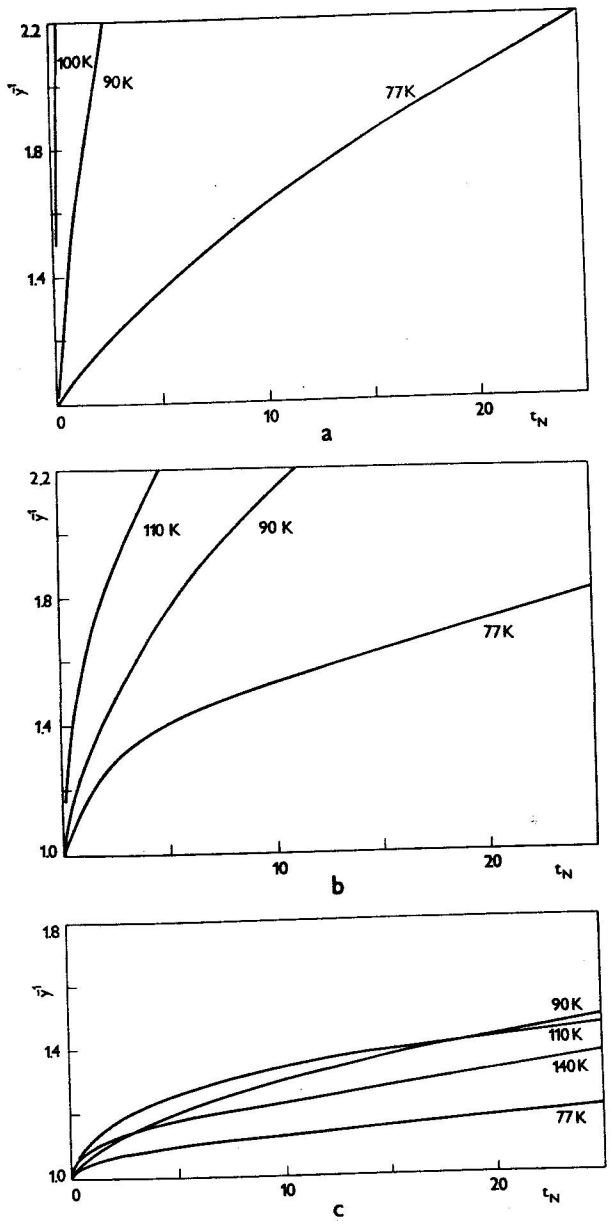

Fig. 7. Calculated function $y^{-1}(t_N)$ which characterizes the recovery process. t_N is a measure for the time which passed since the assumed irradiation was finished. For the initial ($t_N = 0$) the set of calculated functions n_i was used which resulted from an irradiation time t_N^{irr}. a) $t_N^{irr} = 0.3$, b) $t_N^{irr} = 2.5$, c) $t_N^{irr} = 25$.

with the experimental results. Especially the marked bending off of the experimentally found curves, which determines the course for large times t_N is striking (Figs. 4a, b; 7a, b). One reason for this may be that we assume all the N_i in equation (1) to be equal, which is not the case in reality.

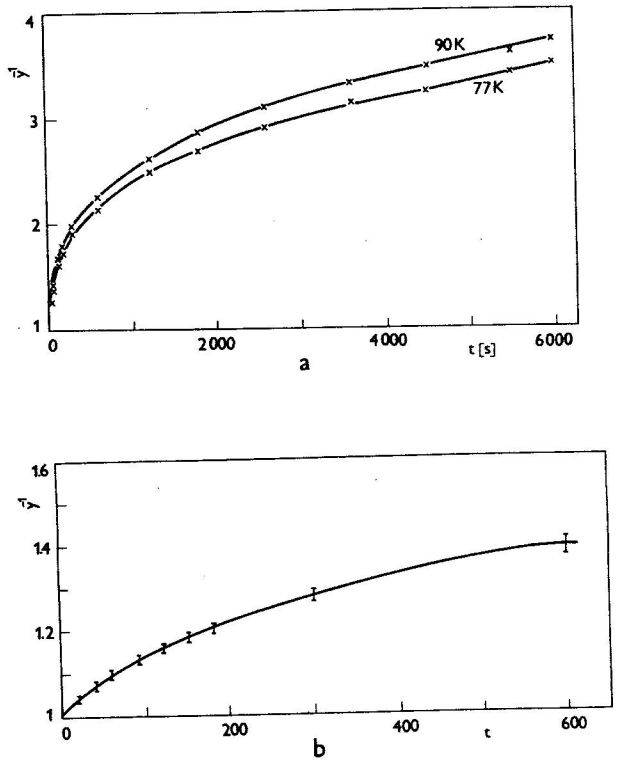

Fig. 8. Recovery phase as in Fig. 4 (experimental). Sample $x = 0.005$. a) irradiation time $t_{irr} = 60$ s, sample-lamp distance 10 cm; b) irradiation time $t_{irr} = 600$ s, sample-lamp distance 60 cm. *I* marks the limits within which the plots vary if the temperature as the same constant parameter is 80 K or 110 K in the irradiation and in the recovery phase.

Fig. 7c shows that no or only a weak dependence of y^{-1} on temperature is to be expected under special irradiation conditions which could not be observed with the sample (Fig. 4a, b). Fig. 8 confirms this possibility. No relevant dependence of y^{-1} on temperature was found for a sample $x = 0.005$. Thus we have shown that the model described in the introduction is able to explain the temperature dependence of the effects in the range above 77 K besides the other relationships which were mentioned in the introduction and proved elsewhere [6, 7]. Especially it becomes clear that a series of relaxation effects contribute to the whole complex which are

effective simultaneously with different diffusion velocities for the Fe^{2+}, changing their arrangement by electron hopping.

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