Photoinduced Irreversible Effects on Magnetic Properties and Allied Phenomena in Magnetic Oxides. IV

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I. INTRODUCTION

This article is the fourth part of the review which concerns with the main theme of photoinduced magnetic effects (PME) in YIG and intimately related problems. The purpose of the present review is twofold: (i) to discuss high energy radiation induced magnetic effect on permeability \( \mu \), with special emphasis on our measurements; (ii) to discuss the models proposed by our research group for a possible mechanism of PME in single crystal of YIG (Ga : 0.0625) with some new results obtained up to Sep. 1990. It will be shown that the experimental result can be satisfactorily explained in a manner which is generally consistent with our model based on the electronic structure of the \( F \)-center.

II. HIGH ENERGY RADIATION INDUCED MAGNETIC EFFECT ON PERMEABILITY

2. 1 X-ray-irradiation effect on real part of permeability (\( \mu' \))

One of us (K. H) presented, for the first time, X-ray induced \( \mu \) effect\(^{1-3}\), although it had been suggested that X-ray is also effective as well as near infrared light for the PME in Si-doped YIG\(^4\). Then Würlitzer et al. followed the author's experiments and found the reproducibility, developing them more in a sophisticated way.\(^{5-7}\) Fig. 1 shows a representative remarkable decay curve of \( \mu \) during X-ray irradiation, slowly recovered after cessation of irradiation for the sample of single crystal of YIG (Pb : 0.89, Si \( \leq \) 0.001).\(^8\) In polycrystalline \((Y_{2.6}Ca_{0.4})[Fe_{1.4}In_{0.6}] (Fe_{2.8}V_{0.2}) O_{12}\) garnet, he obtained an interesting result associated with these experiments. Some of the samples exhibit a detectable acceleration of disaccommodation (DA), but any of them shows no irreversible PME in near infrared light at 77 K.\(^3\) However, all of these samples show, more or less, an X-ray induced \( \mu \) effect as shown in Fig. 2. In this case he assumed that an X-ray quantum gives rise to electronic transitions from the oxygen \( sp \)-valence band state to empty Fe (3d) levels or the Fe (4s, 4p) band.\(^9\) Although this charge transfer transition applies rigorously only to molecular complexes, the situation of localization will be nearly realized at low temperature by analogy with the exciton–like picture, and a simple expression may
be given:

\[ \text{Fe}^{3+} - \text{O}^{2-} + \text{X-ray quantum} \rightarrow \text{Fe}^{2+} - \text{O}^{-} \]

Fig. 3 shows schematically the situation, in which model an existence of divalent iron is not always necessary. The reason why this effect is not sensitive to near infrared light but to X-ray may be explained based on the assumption that a quantum of the before could not penetrate into the sample but the latter do. It is known that soft X-ray is attenuated more strongly than infrared radiation in the range of the optical window in YIG. For example, the intensity of a soft X-ray (0.7 A) has dropped to 18% of the incident radiation after penetrating a 0.2 mm thick single crystal layer, while the intensity of infrared radiation (wavelength ; 1.06 µm ; absorption constant ; \(1.3 \times 10^4 \text{m}^{-1}\)) remains still 90% of the incident light. PME depends on the intensity of light or the current density of photons within the sample and the amounts of doping. When the absorption depth of the irradiation is approximately equal to the thickness of the sample, the maximum of PME could be expected. Since the X-ray mass absorption coefficient of YIG is estimated to be approximately 400 cm\(^{-1}\) for the Mo target, the most effective thickness for the effect is approximately 0.03 mm, corresponding to one tenth of the sample employed. Fig. 4 indicates that the most efficient thickness extrapolated from the experimental points agrees well with the expectation. Qualitatively, this tendency agrees well with Würlitzer’s result in YIG (Si). Fig. 5 shows the decay of \(\mu(t)\) as a function of time for YIG (Pb=0.02) single crystal with different sources of irradiation of X-ray and optical light (Near infrared), which is normalized and approximately represented by the Richter function. The distribution function of relaxation time is assumed to be flat between the lower limit \(\tau_1\) and the upper \(\tau_2\). When optically excited, the relaxation time is short and its distribution is narrow ; \(\tau_1=0.6\) min and \(\tau_2=1.7\) min. On the other hand, when excited by X-rays, a broad distribution is found ; \(\tau_1=0.3\) min and \(\tau_2=173\) min. Even though an X-ray quantum is absorbed at a certain depth, a number of secondary electrons is estimated to be \(10^{4-6}\) from \(h\nu/\lambda\), where \(h\nu\) is the energy of the X-ray quantum and \(\lambda\) is the mean energy required to produce one secondary electron with an energy range of 0.5-1.0 eV. If No photons of a monochromatic radiation are incident on a sample and \(N(x)\) is the amount of photons arriving after penetrating the layer thickness \(x\),

\[ N(x) = N_0 \exp(-ax) \quad (1) \]

\(a\) is denoted as the linear attenuation coefficient. The mass attenuation coefficients of yttrium, iron and oxygen is given as shown below:

\[ \frac{\alpha}{\rho_{\text{YIG}}} = C_Y \left( \frac{\alpha}{\rho} \right)_Y + C_{\text{Fe}} \left( \frac{\alpha}{\rho} \right)_{\text{Fe}} + C_{\text{O}} \left( \frac{\alpha}{\rho} \right)_{\text{O}} \quad (2) \]

where \(C_i\) is the fractions of the elements constituting the total mass of YIG. Taking into account the above relations, the analysis of the experimental results was carried out and self-consistent explanation was obtained.

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Fig. 1 Decrease of permeability upon X-ray irradiation in YIG with some amounts of Pb.(8)

Fig. 2 Decrease of $\mu$ upon X-ray irradiation in CaV In-YIG at 77 K. (3)

Fig. 3 Schematic representation of the cation octahedral site in the garnet lattice where Fe$^{3+}$-O$^{2-}$ is supposed to occur from Fe$^{3+}$-O$^{2-}$ under X-ray excitation. (3)

Fig. 4 Dependence of effectiveness $\Delta \mu/\mu$ on the thickness of the sample of YIG. (8)
Fig. 5 Normalized $\Delta \mu / \mu$ as a function of time for the sample of YIG (No. 2)

2.2 Gamma-ray irradiation effect on the real part of permeability ($\mu'$)

In this section, it is presented that the phenomenon is, for the first time, found to be reproducible also with irradiation of gamma-ray in the same sample employed by the authors in a previous study of PME.\textsuperscript{13} This work is still a preliminary stage and our unambiguous experimental result is limited to the change of the $\mu'$ and $\mu^{\prime 2-3}$ under the gamma-ray irradiation (1.25 MeV) 77 K. The YIG crystal was grown by the floating zone method, as mentioned already in the initial part of the review.\textsuperscript{4} For a source of emitting gamma-ray, two kilocurie cobalt-60 teletherapy unit for a medical use was employed. The spectra of the gamma-ray involve the two dominant peaks of 1.17 Mev and 1.33 MeV, respectively.\textsuperscript{5} If the absorption by the Dewar vessel or the cooling liquid nitrogen in which the sample was directly immersed is neglected, the gamma-ray intensity on the sample surface may be estimated as $2.3 \times 10^{-2}$ Ckg$^{-1}$ min$^{-1}$. The half-value thickness ($d_{1/2}$) of the sample for the gamma-ray can be calculated by using the mass attenuation coefficient ($\alpha / \rho$; $\alpha$ = the linear attenuation coefficient, $\rho$ = density) of YIG.\textsuperscript{6,7} As a result, $d_{1/2}$ is 26.5 mm. Correspondingly, only 2.5% of incident gamma-ray is absorbed in this sample with the thickness of 1.1 mm. The PME effect in visible wavelength region, on the other hand, seems to be more complicated by the strong absorption or light influencing only a thin surface layer of the specimen and the scattering due to pores. If the magnetocrystalline constant $K_1$ is very low, incoherent magnetization which should be classified between wall displacement and coherent rotation is thought to be predominant. For simplicity, however, we assume that the mechanism of magnetization in this situation consists entirely of the rotation of magnetization. Hence in analogy with the process of disaccommodation, the effectiveness of the gamma-ray induced phenomenon $R$ ($\Delta \mu / \mu$) is given by

$$R = \frac{K_{\text{gi}}}{K_1 + K_{\text{gi}}} \frac{L^2}{3 \mu_o K_1 + L^2} = \frac{K_{\text{gi}}}{K_1} \text{ when } K_1 \gg K_{\text{gi}} \text{, and } 3 \mu_o K \ll L^2$$ \hspace{1cm} (1)$$

where $L$ denotes the magnetization, $K_{\text{gi}}$ the gamma-ray induced anisotropy and $\mu_o$ the permeability of vacuum. In other words, measurement of $R$ may provide for a simple and sensitive method available for obtaining $K_{\text{gi}}$, although it may not be a direct means. In addition, this phenomenon may not be attributed to a trivial heating effect. Only a slight DA was observed at 77 K even before gamma-ray irradiation, which is safely neglected. As expected,\textsuperscript{8,9,10} a remarkable decrease of $\mu_i$ is observed to be induced by the gamma-ray in this sample at 77 K and $R$ is plotted against exposure time ($t$) or an amount of doses of gamma-rays ($D$) in a logarithmic form in Fig. 1. At 77 K, the $\mu$ is irreversibly kept lowered, but it is brought back to the original magnitude at room temperature. This
suggests that the gamma-ray irradiation gives rise to a simple multiple ionization and is
unlikely to lead to the long range displacement of the atom. It is, however, rigorously
not determined that this is the case since a complex electronic and ionic processes are
involved in this behavior. As is seen in this figure, $R$ or $K_{el}$ is proportional to $D^{1.5}$ at low
dose ($0 < t < 4 \text{ min}$), while it is to $D^{0.17}$ at high dose ($t > 5 \text{ min}$). Concerning the previous
data of X-ray irradiation on YIG including Pb impurity grown by a flux method, $R$ is
proportional to $D^{0.7}$, as is often the case in photoconductivity at usual light wavelength.\(^\text{[11]}\)
Experiments for studying the discrepancy are now in progress along with measurements
of complex permeability\(^\text{[5]}\) and much more microscopic details of the mechanism.

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![Graph](image)

Fig. 1. Logarithm of $R$ ($\Delta n/\mu$) vs logarithm of irradiation time (amounts of gamma-
ray) at 77 K in undoped YIG with some deficiency of oxygen.

2. 3 Gamma ray-irradiation effect on an imaginary part of permeability ($\mu''$) in
undoped YIG
As described in the previous section, the authors found, for the first time, the gamma ray (1.25 Mev) induced effect on \( \mu'' \) in undoped YIG.\(^1-v\) In the preliminary stage, however, we would like to mention several results obtained. The detail will be reported again in the next issue. On warming a gamma-ray irradiated sample, an imaginary part of permeability \( \mu'' \) below 10 kHz has been found to show the double peaks versus temperature. Each of the activation energy is 0.19 and 0.35 ev, respectively. Besides, it has been observed that the dependence of height of peaks is contrasted with each other on the gamma-ray irradiation time. These features have been discussed within a simple phenomenological model. Next, the measurement of \( \mu'' \) has been carried out as a function of temperature after the irradiation is switched off, as shown in Fig. 1. In this figure, it turns out that the double peaks of \( \mu'' \) occur in the ranges 77 to 300 K and 0.5 to 10 kHz, whereas only one \( \mu'' \) peak is visible before irradiation. Here, the peak at lower temperature is termed \( P_1 \) and another one \( P_\Pi \) at higher temperature. On the other hand, Würlitzer et al.\(^5\) reported the double peaks of \( \mu'' \) between these temperatures even in an unirradiated polycrystalline YIG with a higher concentrations of Si-dopants \((x \geq 0.02)\). The result is suggestive of a similar contribution to \( \mu'' \), independent on whether the \( Fe^{3+} \) is induced by gamma-ray or nonstoichiometry or doping with tetravalent cations. From the Arrehnius plot of the frequency associated with each peak, the activation energies are obtained as \( E_1 = 0.19 \) eV for \( P_1 \) and \( E_\Pi = 0.35 \) eV for \( P_\Pi \), whereas the near-infrared light excitation measurements which were previously carried out indicated the slightly lower activation energies, 0.10 and 0.30 eV, respectively. In Fig. 2, the detail of the results is fed to a computer which presents them as a three dimensional view of \( \mu'' \), gamma-ray irradiating time and temperature \( T \), based on the experimental results. In this figure an interesting behavior is seen much more clearly. The observation may be explained in terms of the one dimensional model of an \( F \)-center of oxygen vacancy which forms a ground state and two excited levels. Each of these levels is assumed to have two magnetically equivalent potentials separated by barriers of activation energies \( U_{ex1} \) and \( U_{ex2} \) which are associated with \( P_1 \) and \( P_\Pi \), respectively. These excited states could be sufficiently close to conduction band for the electrons to be ionized thermally. If these levels may be associated with “near” or “far” site, our observation could give an evidence to support the two center model.\(^6\) At low temperature an irreversible behavior or complex permeability is suggestive of a long lived trapped state. In order to explain the maximum of \( P_1 \) and a monotonic increase of \( P_\Pi \) with intensity of the irradiation before saturation, we give a set of consecutive equations of the simple form

\[
\frac{dN_g}{dt} = -W_1 N_g, \quad (1a)
\]

\[
\frac{dN_{ex1}}{dt} = W_1 N_g - W_2 N_{ex1}, \quad (1b)
\]

\[
\frac{dN_{ex2}}{dt} = W_2 N_{ex1}, \quad (1c)
\]

where \( N_g \) is the number of ground state centers, \( N_{ex1} \) the number of the first excited ones and \( N_{ex2} \) the number of the second excited states, \( t \) the exposure time of the gamma-ray, \( W_1 \) the probability of transition from the ground state, to the first excited state and \( W_2 \) the one from the first to the second excited state. These have been presented in Fig. 3. In this case, a thermal recovery process may be approximately neglected at low tempera-
ture. Thus we have found how the concentrations of $N_e$, $N_{ox1}$ and $N_{ox2}$ vary with $t$. By integration of eq. (1a) we find $N_e$ to be

$$N_e = N_{eo} \exp(-W_1 t)$$  \hspace{1cm} (2a)

where $N_{eo}$ is the concentrations before irradiation. Similarly, the final expression for $N_{ox1}$ may be deduced to

$$N_{ox1} = N_{eo} W_1 (W_2 - W_1)^{-1} \exp(-W_1 t) + (W_1 - W_2)^{-1} \exp(-W_2 t)$$  \hspace{1cm} (2b)

Noting that there is no change in $N_{eo}$, the stoichiometry relates the concentrations of each component by

$$N_{eo} = N_e + N_{ox1} + N_{ox2}$$

which with eqs. (2a) and (2b) gives

$$N_{ox2} = N_{eo} (1 - W_2 (W_1 - W_2)^{-1} \exp(-W_1 t) + W_1 (W_2 - W_1)^{-1} \exp(-W_2 t))$$  \hspace{1cm} (2c)

By differentiating (2b) and setting $dN_{ox1}/dI = 0$, the intensity at which the maximum of $N_{ox1}$ occurs is thus

$$I_{max} = (W_1 - W_2)^{-1} \ln(W_1/W_2)$$  \hspace{1cm} (2b')

On the other hand, it is always found to be

$$dN_{ox2}/dI > 0.$$  \hspace{1cm} (2c')

Therefore the behavior of $P_1$ may be ascribed to that of $N_{ox1}$ and a monotonic increase of $P_{II}$ to that of $N_{ox2}$. Furthermore, it is proposed that a gamma-ray induced decrease of $P'$ may be caused by a decrease of $N_e$ of which anisotropy will be neglected. Figure 4 shows that the measured values of each curve may be fitted satisfactorily with the parameters based on the model as well as in a white light illuminated sample. Further work with a precisely determined concentration of oxygen vacancy is clearly required in order to obtain a better understanding of the origin of these phenomena.

References
III PME (WHITE LIGHT) ON COMPLEX PERMEABILITY IN Ga-DOPED YIG

3. 1 Optical light effect on a real part of permeability ($\mu'$)

Photoinduced decrease in magnetic initial permeability $\mu$ of over 60% below 20 kHz is observed at 77 K in yttrium iron garnet (YIG) single crystal with a small amounts of Ga (0.0625) and some deficiency of oxygen. On warming the previously illuminated sample with a white light, an imaginary part of permeability $\mu''$ below 10 kHz was found to show the triple peaks as a function of temperature, the activation energies of which are 0.095–0.22 eV, 0.50 eV and 0.81 eV, respectively. Besides, it has been observed that the area under each peak depends on the illuminating time, as expected in our earlier measurement in undoped YIG crystal. These features have been discussed and interpreted in terms of the electronic transfer between octahedral sites at the nearest, the next
nearest and further remote neighbor of oxygen vacancy or $F$-center based on a modified "two center" model. A drastic photoinduced change of $\mu$ in polycrystalline YIG with a small contents of Fe$^{2+}$ induced by Si$^{\text{ii+}}$ was firstly found by U. Enz et al.\(^1\) And an original "two center" model explaining for the observation was proposed.\(^2\) This model assumes that the anisotropy of Fe$^{2+}$ ion near Si$^{\text{ii+}}$ is much larger than that of photoinduced Fe$^{2+}$ ion far Si$^{\text{ii+}}$, resulting in a photoinduced pinning of domain wall or decrease of $\mu$, since $\mu$ is inversely proportional to the summation of crystalline anisotropy $K_i$ and photoinduced anisotropy $K_\text{p}$, besides the concentration fluctuations of strong anisotropic ions.\(^3\) In this context, not only a decrease but also an increase in $\mu$ could be expected, depending on illuminating time or a sign of $K_i$ before illumination since the contribution of Fe$^{2+}$ ion in garnets is large, of opposite sign to $K_i$.\(^4,5\) There are no data available for the observation of photoinduced increase of $\mu$. Nevertheless, the most obvious success of this model is that it explains the irreversible behavior at low temperature. In this review, our attention is mainly concerned with an observation of the complex permeabilities during and after illumination in YIG (0.0625 Ga) single crystal and an explanation based on a modified two center model. The definition of thermally stimulated process\(^6\) has an application to this measurement. The Ga ion enters both octahedral site and tetrahedral site in YIG, which favors quality of crystalline character, giving no influence on a valency of iron ion\(^7\) Single crystals of YIG (Ga) were grown by a modified floating zone method, the details of which can be found elsewhere.\(^8\) The sample set in a copper case immersed in liquid nitrogen was illuminated with white light (5mW/cm$^2$) and then warmed at a controlled heating rate. The experimental assembly is further given in Ref. 9. Fig. 1 displays a decrease of $\mu$ on illumination in a YIG with a small amount of Ga (0.0625) and some degree of oxygen deficiency. we would like to attribute the decrease in $\mu$ to a photoinduced local distortion since a remarkable decrease in $\mu$ has been observed on applying an inhomogeneous stress to the sample at 77 K in a preliminary experiment; the removal of an electron by absorption of photon from $F$-center or oxygen vacancy could lead to a movement of the surrounding ions and a considerable distortion, while the released electron will be caught at several states mentioned below. On single and or double photoionization, a domain wall pinning could be enhanced through an interaction between a photoinduced distortion and domain wall.

**References**

3.2 Optical light irradiation effect on an imaginary part of permeability $\mu''$ in Ga-doped YIG

Next, a measurement of $\mu''$ has been carried out as a function of temperature after the illumination is switched off. As shown in Fig. 1, the triple peaks of $\mu''$ occur in ranges 77 to 300 K for an hour illumination at 77 K, the sample temperature being raised at a constant rate (0.5 K/min). In this figure, a background is subtracted, where only one broad and low maxima of $\mu''$ is visible near 100 K. Here the lowest temperature peak is termed $P_1$, the intermediate temperature peak, $P_\Pi$ and the highest temperature peak, $P_\Pi$. From the Arrhenius plot of the frequency associated with each peak, the activation energies are obtained as 0.095-0.22 eV for $P_1$, 0.50 eV for $P_\Pi$ and 0.81 eV for $P_\Pi$, whereas our earlier measurement in undoped YIG single crystal indicated the slightly lower activation energies, 0.10 and 0.30 eV, respectively.\textsuperscript{11} Next, a thermal variation of $\mu''$ has been studied as a function of illuminating time $t$. Figure 2 shows the result as expected in undoped YIG. We would like to introduce a new mechanism based on an F-center in order to explain our experiment. As is well known, the F band arises from the optical transitions of an electron trapped at a negative ion vacancy. The interest in the study of the F-center in alkali halides has not been decayed, since it became clear that the F-center possesses several higher excited states at energies corresponding to those of the conduction band of the crystal. At large distances from the vacancy, the field is Coulombic. Since the Coulomb potential does not present discrete levels at positive energies to excited states of positive energies, it will cause some objections. The observation may be explained in terms of the one dimensional model of an F-center or oxygen vacancy which forms a ground state and three metastable excited levels in which an excited electron may spontaneously go to these levels via so called “excited” level. Each of these metastable levels is assumed to have two magnetically equivalent potentials separated by barriers of activation energies $U_{exc1}$, $U_{exc2}$ and $U_{exc3}$ which are associated with $P_1$, $P_\Pi$ and $P_\Pi$, respectively. These states could be sufficiently close to the conduction band for the electrons to be ionized thermally. Based on the “two center” model, these levels could be associated with “near” “intermediate” or “far” site. A question is, however, left poorly understood why it is possible for the clear-cut sharp peaks to occur under an assumption of the dependence on a gradual continuous separation $r$ from oxygen vacancies. More likely explanation is
that a local distorted structure could be identified with a Fe^{2+} perturbed in a discrete way by a nearest- and a next nearest neighbour of oxygen vacancy and an isolated Fe^{2+} ion or far from the vacancy. In order to explain the intensity dependence of these peaks, we postulate a set of consecutive equations of the simple form¹

\[
\frac{dN_e}{dt} = -W_1 N_e, \quad \text{(1a)}
\]
\[
\frac{dN_{\text{exc1}}}{dt} = W_1 N_e - W_2 N_{\text{exc1}}, \quad \text{(1b)}
\]
\[
\frac{dN_{\text{exc2}}}{dt} = W_2 N_{\text{exc1}} - W_3 N_{\text{exc2}}, \quad \text{(1c)}
\]
\[
\frac{dN_{\text{exc3}}}{dt} = W_3 N_{\text{exc2}} \quad \text{(1d)}
\]

where \(N_e, N_{\text{exc1}}\) and \(N_{\text{exc2}}\) is the fractional number of electrons at ground state, the first, the second, and the third metastable excited states, respectively. \(W_i\) is the optical probability for an electron to go from state \(i\) to more excited state \(i\). Dillon et al.² shows that such a relation of inequality as \(W_1 > W_2 > W_3\) could be applied to our case. The relation between photoexcitation and raising temperature is complex and sometimes a competing process.³ However, a thermal recovery process may be, at present, ignored at low temperature. Thus we try to find how the fractional concentrations of \(N_e, N_{\text{exc1}}, N_{\text{exc2}}\) and \(N_{\text{exc3}}\) vary with \(t\). By integration of eq. (1a) we find \(N_e\) simply to be

\[
N_e = N_{e0} \exp(-W_1t) \quad \text{(2a)}
\]

where \(N_{e0}\), is the total concentration of Fe^{2+}. Similarly, the other expressions in each level may be,

\[
N_{\text{exc1}} = N_{e0} W_1 (W_1 - W_2)^{-1} \{\exp(-W_1t) - \exp(-W_2t)\} \quad \text{(2b)}
\]

\[
N_{\text{exc2}} = N_{e0} W_1 W_2 \{((W_1 - W_2)(W_2 - W_3)(W_1 - W_3))^{-1} \}
\]
\[
((W_2 - W_3) \exp(-W_1t) + (W_3 - W_1) \exp(-W_3t) + W_1(W_2 - W_3) \exp(-W_3t)) \quad \text{(2c)}
\]

\[
N_{\text{exc3}} = N_{e0} \{((W_2 - W_1)(W_2 - W_3)(W_2 - W_3))^{-1} \}
\]
\[
((W_2 - W_3) - W_2 W_3(W_2 - W_3) \exp(-W_1t) - W_1 W_3(W_2 - W_1) \exp(-W_3t)
\]
\[
- W_1 W_2(W_2 - W_1) \exp(-W_3t)) \quad \text{(2d)}
\]

Differentiating and setting zero, the illuminating time \(t\) at which the maximum of \(N_{\text{exc2}}\) of \(N_{\text{exc3}}\) occurs may be simply deduced. And a relation which implies a monotonic increase of \(N_{\text{exc3}}\) with \(t\) may be obtained. The illuminating time \(t\) dependence of these peaks in Fig. 3 may be rather well explained simply under an assumption that each peak is in proportional to the fractional concentration of these electrons occupied in each level. In this context, a photoinduced decrease of \(\mu'\) in Fig. 1 may be primarily caused by a decrease of \(N_e\) with less contribution to distortion, or conversely an increase in a rest of \(N_e\). In the discussion so far is not included an explicit temperature dependence of each concentration. As shown in Fig. 2, the curves obtained may be fitted with the parameters taking into consideration the Boltzmann activation process; decay rate of each concentration is assumed to be governed by thermal decay in proportional to \(\exp(-U_1/kT)\).²⁻⁴ Although the data are rather scattered, a trend is observable. In order to obtain more information, we investigated the effect of heating sample to a certain temperature after illumination at 77 K. Fig. 2 (A) shows the change in \(\mu'\) and \(\mu''\) when illuminated sample was heated to 180 K in the dark and then cooled to 77 K, followed by another warming
process without illumination. It is clearly seen that a separated triple peaks in $\mu''$ in the first run is coagulated into one broad maximum in the second run, retaining the total area under the peaks, whereas $\mu'$ retraces itself almost with fidelity. In Fig. 3 (B), a similar is presented except for a turning temperature at 200 K. The same sort of thing is seen in $\mu''-T$ curves but $\mu'-T$ curve is irreversibly changed, probably due to thermal relaxation. This result could be explained by assuming that the thermal potential barriers for a photoexcited electron to be recaptured by oxygen vacancy is higher than those between excited electrons and only excited Fe$^{2+}$ ion contributes to a decrease of $\mu'$. Fig. 4 shows a schematic representation of the mechanism of the behavior shown in Fig. 3.\textsuperscript{3)} New development of PME may be expected, linked with a concept of deep level in semiconductor physics. These works by the authors in the review were in part supported by Grant-in Aid for Scientific Research from the Ministry of Education, Science and Culture in Japan under Contract No. 01550025.

References

![Fig. 1 Thermal variation of $\mu''$ after exposure of white light illumination at 77 K. The frequency and field intensity are 0.5 kHz and 1 mOe, respectively.](image1)

![Fig. 2 Illuminating time dependence of $\mu''$, $P_1$, $P_\Pi$, and $P_\Sigma$. The points are experimental values measured at frequency of 0.5 kHz as a function of $t$. The solid lines are calculated from the relations (2b), (2c) and (2d) with the values of the fitting parameters including thermal activation process.](image2)
Fig. 3 Behaviors of $\mu'$ and $\mu''$ after illuminated at 77 K in the temperature cycle. (A) Turning temperature : 180 K and (B) Turning temperature : 200 K.

Distance

Fig. 4 Schematic representation of a proposed mechanism of the behavior shown in Fig. 3.