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# MÖSSBAUER POLARIMETER AND MÖSSBAUER FARADAY EFFECT

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## ABSTRACT

A Mössbauer polarimeter consists of a gamma ray source (polarizer), transmitter (sample to be analyzed), analyzer and automatic counting system. This equipment was used to observe the Mössbauer Faraday effect in non-stoichiometric $Fe_3O_4$. Experimental results demonstrated that electronic hopping above the Verwey temperature between $Fe^{2+}-Fe^{3+}$ ions on the octahedral sites is a only localized phenomenon and the recoilless fractions of $^{57}Fe$ nuclei in $Fe_{3-\upsilon}O_4$ ($\upsilon = 0.02$) are 0.71 for A sites and 0.62 for B sites, respectively.

**Keywords:** Mössbauer polarimeter Faraday effect

## 1 INTRODUCTION

Mössbauer polarization effect has gradually been recognized both experimentally and theoretically based on the interaction between the gamma rays and matter. Some researchers pointed out that the dispersion phenomenon of gamma rays associated with Mössbauer resonance absorption would have an important applications as well as the magneto-and electro-optics effects soon after the discovery of Mössbauer effect. Although only few materials have been observed the magneto-optics effect up to now[1], we believe that further studies in the field will bring on new development in the fundamental and applied aspects.

A Mössbauer polarimeter consists of a gamma ray source (polarizer), transmitter (sample to be analyzed), analyzer and automatic counting system as shown in Fig.1. The rotation of linearly polarized gamma rays transmitted through a measured medium (transmitter) with a magnetic field applied along the propagation direction of the gamma rays is called the Mössbauer Faraday effect. According to the theory of electromagnetic radiation, the rotation angle of the polarization plane for monochromatic radiation is given by:[1,2]

$$\Phi_{\mathrm{i}} = (-\tau_{\mathrm{T}}\Gamma/32)\{\Sigma_{\mathrm{l}=1}^{4}[(-1)^{\mathrm{l}}a_{\mathrm{l}}^{2}(E_l - E_i)]/[(E_l - E_i)^2 + (\Gamma/2)^2]\} \qquad (1)$$

where $E_l$ and $E_i$ are the energies of spectral lines in transmitter and polarizer, respectively. Γ is the full line width and $\tau_T$ is the effective thickness of the transmission sample. The rotation angle caused by each lattice site in the transmitter is different and has to be evaluated. Taking into account all the six radiation lines, the net rotations of the radiation passed through the transmitter is then determined by

$$\Phi = tg^{-1}\{\Sigma\,[a_i^2 \sin(2\Phi_i)]/\Sigma\,[a_i^2 \cos(2\Phi_i)]\} \qquad (2)$$

In the equations above, the $a_1^2$ and $a_i^2$ indicate the relative contributions of the different lines to each polarization in transmitter and polarizer.

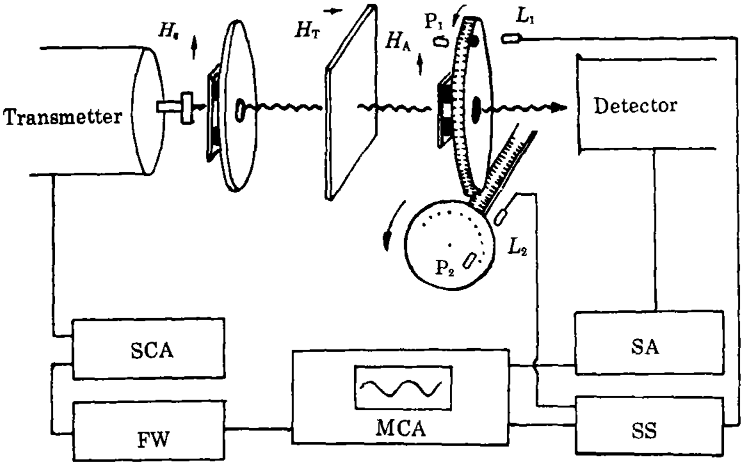

**Fig.1 Mössbauer polarimeter equipment**

SCA: Single channel analyser FW: Flyback wave form MCA: Multi-channel analyser

SS: Synchro-signal SA: Signal amplifier $H_S$: Applied magnetic field for the source

$H_A$: Applied magnetic field for the absorber $H_T$: Applied magnetic field for the transmitter

L: Lamp P: Photoresistance

In this work, we first observed the Mossbauer Faraday effect in non-stoichiometric magnetite. Experimental results demonstrated that the electron hopping between $Fe^{2+}-Fe^{3+}$ ions at the octahedral sites above Verwey temperature $T_v$ is a only localized phenomenon and the recoilless fractions of the two lattice sites, $f_A$ and $f_B$, were obtained in terms of the Debye model.

## 2 RESULTS AND DISCUSSION

In Fig.2 the room-temperature Mössbauer spectra for non-stoichiometric $Fe_{3-\upsilon}O_4$, sample 1 ($\upsilon = 0.02$) and sample 2 ($\upsilon = 0.09$) prepared using chemical reduction of $\alpha-Fe_2O_3$, are shown. Each spectrum has the two components corresponding to A and B sites in $Fe_{3-\upsilon}O_4$, the ratios of areas $S_A/S_B$ are 0.67 for sample 1 and 1.28 for sample 2.

The relative transmission as a function of the angle between source and absorber magnetic fields is shown in Fig.3. The Mössbauer Faraday rotations corresponding to sample 1 ($\tau$ =24.9 mg/cm$^2$, 83.7% enriched in $^{57}$Fe) are 13.1° and 21.4°, respectively. Although $S_B > S_A$ for sample 1 and $S_A > S_B$ for sample 2, the net Faraday rotations observed are in the same direction.

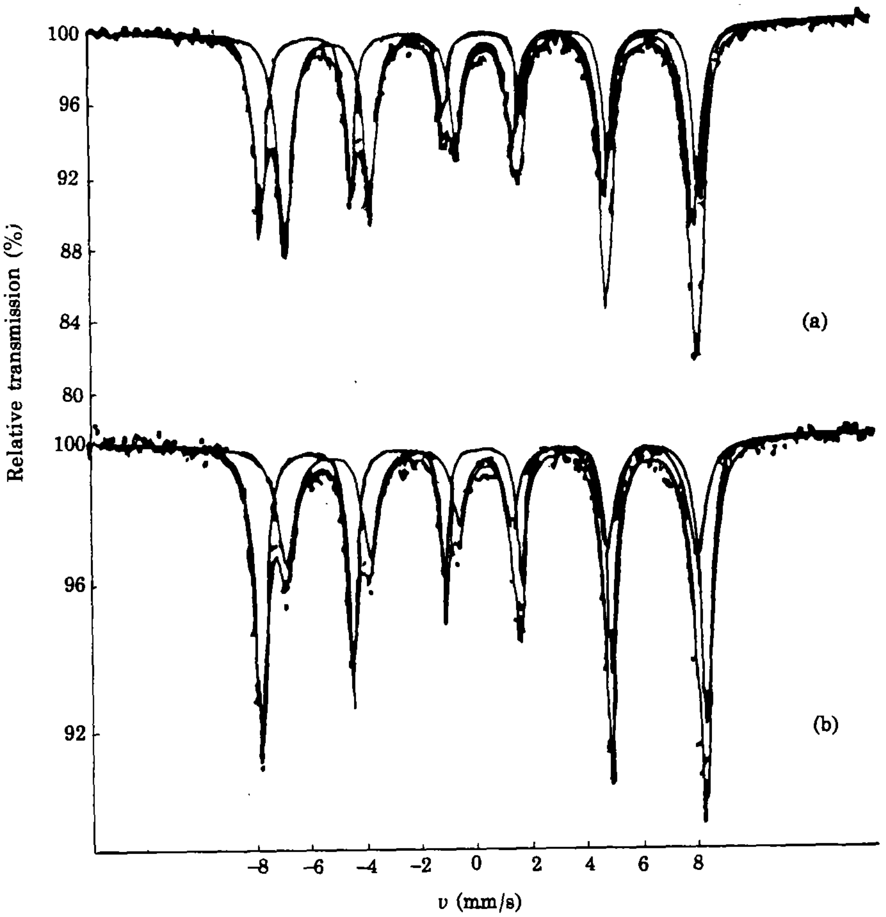

**Fig.2 Mössbauer spectra of $Fe_{3-v}O_4$ at room temperature**

a) Sample 1 ($v=0.02$) b) Sample 2 ($v=0.09$)

It is well known that there is a rapid electron exchange between the Fe$^{2+}$ and Fe$^{3+}$ ions on octahedral sites for $Fe_3O_4$ at room temperature (above $T_v$), but whether all or part of the Fe$^{3+}$ ions in the B sites participate in the electron exchange is still a controversy. If non-stoichiometric $Fe_{3-v}O_4$ may be formulated as (Fe$^{3+}$) [$Fe_{1-3v}^{2+}$ $Fe_{1-3v}^{3+}$ $Fe_{5v}^{3+}$]$O_4$ and assumed that Fe$^{3+}$ in B sites partly participate in the exchange process, then the areas $S_B$ in Fig.2 only indicate the contributions of the $Fe_{1-3v}^{2+}$ and $Fe_{1-3v}^{3+}$ ions in the B sites, whereas the areas $S_A$ reperesent that of the unpaired $Fe_{5v}^{3+}$ ions in B sites besides the all Fe$^{3+}$ in the A sites, it is as similar as $\gamma$ $-Fe_2O_3$. In the measurement of the Mössbauer Faraday effect for $Fe_3O_4$, because the two sites are antiferromagnetically coupled, the rotations by the two sites are in opposite directions. Although $S_A > S_B$ for sample 2, the contribution of $Fe^{3+}$ ions at A sites is a part in $S_A$ only, the rest belonging to B sites. Furthermore, the occupation probability of the $Fe^{3+}_{5v}$ at the B sites increases with increasing $v$. Therefore, the total rotation caused by the B sites is still dominant.

In order to determine $f_A$ and $f_B$, we can use the methods both of the Mössbauer Faraday effect and of the temperature dependence of the recoilless fraction[3]. In the Debye approximation for lattice vibrations, the recoilless fraction $f$ can be written as

$$f = \exp\left(-6 E_R T/k\theta^2\right) \qquad T > 0.5\theta \tag{3}$$

where $E_R$ is the recoil energy for $^{57}Fe$ nuclei, $\theta$ denotes the Debye temperature, $k$ is the Boltzmann constant and $T$ is the absolute temperature. For the thin absorber of non-stoichiometric $Fe_{3-v}O_4$, the ratio of areas $S_A/S_B$ does not directly indicate the ratio of iron ions in the above model, but it can be written as

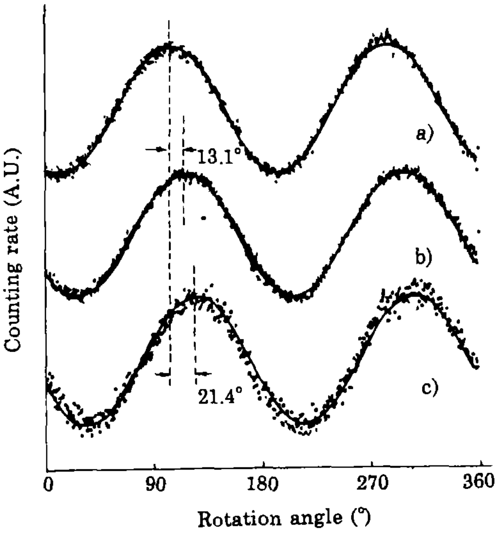

**Fig.3 The relative transmission of the linear radiation as a function of the rotation angle between the polarizer ($^{57}Co(\alpha - Fe)$) and analyser ($\alpha - Fe$)**

*a*) Without a transmitter *b*) With the transmitter $Fe_{2.98}O_4$($\tau$ = 15.3 mg/cm$^2$, 83.7% enriched in $^{57}Fe$) *c*) With the transmitter $Fe_{2.91}O_4$ ($\tau$ = 24.9 mg/cm$^2$, 83.7% enriched in $^{57}Fe$)

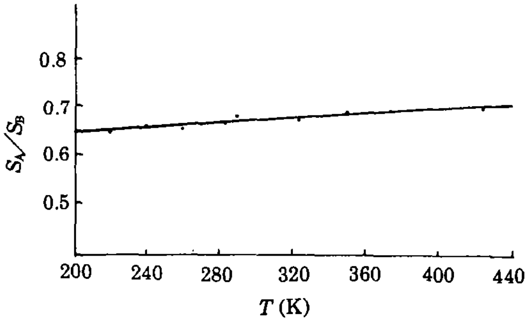

**Fig.4 The area ratio of the spectra for $Fe_{2.98}O_4$ as a function of temperature**

The points are the experimental data, the straight line is a result of the least square fitting

$$S_A/S_B = [1 \cdot f_A + 5v \cdot f_B]/[2(1-3v) \cdot f_B] \approx [1+5v]/[2(1-3v)] - \{[6E_R/2(1-3v)k]\,[1/\theta_A^2 - 1/\theta_B^2]\}\,T \tag{4}$$

Fig.4 shows the temperature dependence of the area ratio $S_A/S_B$ for sample 1, the solid line can be plotted by using the least square fits to Eq.(4), and from this we find $v=0.02$ and $f_A/f_B=0.88$. Inserting the values and other parameters of sample 1 to Eq. (1) and (2), the values, $f_A=0.71$ for A sites and $f_B=0.61$ for B sites are obtained. The recoilless fraction for octahedral sites is smaller than that for tetrahedral sites, this implies that the bond of the $^{57}$Fe nuclei in B sites is slightly weak due to the presence of lattice vacancies.

## 3 CONCLUSIONS

a. The Mössbauer Faraday effect can be taken as one method for determining recoiless fractions of nuclei in the sublattice sites associated with other way.

b. The electron exchange (hopping) between $Fe^{3+}$ and $Fe^{2+}$ ions in B sites above Verwey temperature is a localized phenomenon for the non-stoichiometric magnetite. This means that the relative occupation ratio in $Fe_{3-v}O_4$ can not simply be attributed to the area ratio of the sublattice spectra from Mössbauer measurement. However, the information can be obtained by using the above calibrations.

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