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MÖSSBAUER SPECTROSCOPY ON NICKEL-ZINC FERRITES⁺

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Résumé.- La structure magnétique hyperfine du système Zn_xNi_{1-x}Fe₂O₄ (ferrite de nickel et de zinc) a été le sujet de maintes études par effet Mössbauer. Le sujet de cette étude concerne le même type de ferrite avec x égal à 0,3; 0,5 et 0,65 à différentes températures en-dessous du point de Néel. On en déduit le temps de relaxation longitudinal en fonction de la température pour les sites d'ions ferriques A et B.

Abstract.- The magnetic hyperfine structure of the nickel-zinc ferrite system, Zn_xNi_{1-x}Fe₂O₄, have been the subjects of Mössbauer effect studies for many years /1-6/. It is generally found that the Mössbauer spectra of this material at higher temperature are characterized by the presence of ferromagnetic relaxation /3-6/. Bhargava and Iyengar /6/ fitted the spectra of Ni-Zn ferrites (with x=0.25, 0.5, 0.75) with the relaxation-shape calculated according to the stochastic model /7/. The calculated line shape agrees with experimental one qualitatively at all temperature except near the Néel point. Here we report our work on the same ferrite with x=0.3, 0.5 and 0.65 at various temperatures near and below the Néel point. As the result of the successful fitting of the Mössbauer spectra with perturbation theory of Levinson and Luban /8/, we extracted the longitudinal relaxation rates of the A- and B-sites ferric ions in the ferrites as function of temperature.

1. Experimental.- Our nickel-zinc ferrites (Zn)_x(Ni)_{1-x}Fe₂O₄ (x = 0.30, 0.50 and 0.65, were prepared by sintering stoichiometric amounts of ZnO, NiO, and Fe₂O₃ at a temperature of 1200 ± 10°C in air for 10 hours and then allowed to cool slowly. X-ray diffraction analyses /9/ confirmed the spinel structure and the absence of other phases. Chemical analyses /10/ confirmed the chemical compositions. A Mössbauer absorber of about 15 mg/cm² was prepared from the fine ground powder of each ferrite. The Mössbauer spectrometer was a constant acceleration type. The spectrometer was calibrated using an α-Fe₂O₃ powder absorber.

2. Line-shape calculation and data fittings.- According to Levinson and Luban (equation 29 in /8/), we obtain that, for a particular Mössbauer transition |M_g⁺), to |M_e⁻), absorption probability as a function of ω

$$I_{M_g M_e}(\omega) \propto \sum_{P=1}^{+1} |(M_g^+ | \rho^{-P} | M_e^-)|^2 \exp(C_L A_{M_g M_e} / R_L^2 + C_T B_{M_g M_e} / R_T^2) \int_{-\infty}^{\infty} dt \exp \{it[\bar{\omega} + (\omega_g^z M_g^+ - \omega_e^z M_e^-)]\} \cdot [-(\Gamma_{M_g M_e} + C_L A_{M_g M_e} / R_L + C_T B_{M_g M_e} / R_T) |t|] \cdot \exp \{- [C_L A_{M_g M_e} / R_L^2] \exp(-R_L |t|) + [C_T B_{M_g M_e} / R_T^2] \exp(-R_T |t|)\}$$

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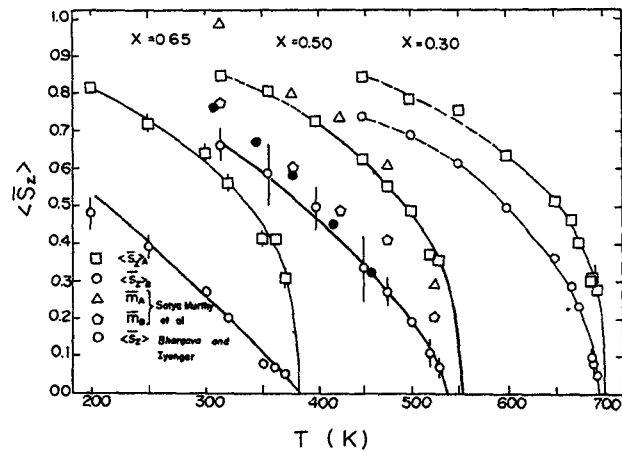


Fig. 1 : Averaged spins of ferric ions in Zn_xNi_{1-x}Fe₂O₄ (x=0.65, 0.5 and 0.3) vs. temperature.

We have assumed : (a) the transversal relaxation broadening $C_T B_{M_g M_e} / R_T \ll C_L A_{M_g M_e}$ so that the transversal term was insignificant for the overall calculation of the Mössbauer spectrum. (b) Upper bounds of $\Gamma_{M_g M_e}$'s were estimated from experimental data given by Leung et al. /5/. While lower bounds of $\Gamma_{M_g M_e}$'s (0.11 mm/s) were taken according to the resolution of our spectrometer. The upper bounds enable us to estimate the maximum errors of our reduced parameters after fittings. (c) The Mössbauer spectrum consisted of two absorption patterns corresponding to A- and B-site ferric ions and weighted according to the chemical composition x. Therefore the number of fitting parameters for the shape of the Mössbauer spectrum has been reduced to four,

the Boltzman factors p_A and p_B and the longitudinal relaxation rates R_{LA} and R_{LB} .

3. Results and discussions.- In figure 1, we show the temperature dependences of $\langle S_z \rangle_A$ and $\langle S_z \rangle_B$ with $x = 0.30, 0.50$ and 0.65 . (The $\langle S_z \rangle$ was calculated from p). Also presented in figure 1 are $\langle S_z \rangle$'s determined by Bhargava and Iyengar /6/ and the reduced sublattice magnetizations, \bar{m}_A and \bar{m}_B , determined with the neutron diffraction method by Satya Murthy et al. /11/ for the ferrite $(Zn)_{0.50}(Ni)_{0.50}Fe_2O_4$. In Ni-Zn ferrite system, all contribution to A-site sublattice magnetization comes from ferric ions so that $\bar{m}_A = \langle S_z \rangle_A$. As indicated in figure 1, for the ferrite with $x = 0.50$, the values of $\langle S_z \rangle_A$ of our result agree remarkably with the values of \bar{m}_A . For B-site, both Fe^{3+} and Ni^{2+} ions contribute to the sublattice magnetization \bar{m}_B . Thus the discrepancy between the values of $\langle S_z \rangle_B$ with those of \bar{m}_B is expectable. The Néel temperatures for $x = 0.30, 0.50$ and 0.65 are respectively $700 \pm 3, 553 \pm 11, \text{ and } 381 \pm 6$ K. These values agree with the results of Leung et al. /5/ (and Satya Murthy et al. /11/ for the sample with $x = 0.50$).

In figure 2, we present the temperature dependences of R_{LA} and R_{LB} with $x = 0.50$. It is obvious that for this composition of ferrite, R_{LA} is considerably greater than R_{LB} at any given temperature in the temperature range under consideration. This means that the A-site ferric ions transfer out their excitation energy easier than B-site ferric ions do.

In figure 3 we show respectively the zinc content dependences of R_{LA} and R_{LB} when $p_A = p_B = 0.60$ (i.e. $kT = 3.9 \mu_B H_{eff}$). As shown in this figure, for both sites, R_L decreases when the zinc content x increases. However, R_{LA}/R_{LB} increases when x increases. The values of R_{LA}/R_{LB} when $p_A = p_B = 0.60$ are $2.1 \pm 0.4, 7.1 \pm 3.5, 10.3 \pm 3.5$ for $x = 0.30, 0.50, \text{ and } 0.65$ respectively. The difference in the values of R_{LA}/R_{LB} becomes very suggestive after one has compared the numbers of A-O-B superexchange bonds connected with each A-site ferric ion, n_A , and that connected with each B-site ferric ion n_B . The values of n_A/n_B are 2.86, 4.00, and 5.71 for $x = 0.30, 0.50, \text{ and } 0.65$ respectively. It seems that the values of R_{LA}/R_{LB} are rather close

to the values of n_A/n_B for the three compositions of Ni-Zn ferrite. This suggests that the dominant relaxation mechanism in these ferrites should involve the A-O-B superexchange interaction between the neighboring magnetic ions situated on different sites.

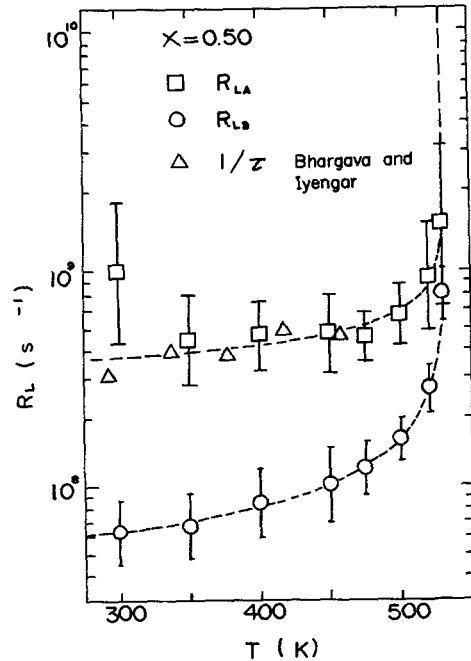


Fig. 2 : Longitudinal relaxation rates of ferric ions in $Zn_{0.5}Ni_{0.5}Fe_2O_4$ vs. temperature.

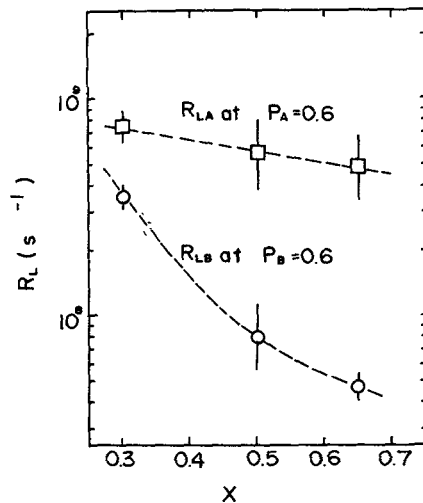


Fig. 3 : R_{LA} and R_{LB} vs. x at $p_A=p_B=0.6$

4. Conclusion.- We have analysed the Mössbauer spectra of $Zn_xNi_{1-x}Fe_2O_4$ for $x = 0.30, 0.50$ and 0.65 . We have reduce the $\langle S_z \rangle_{A,B}$ $R_{LA, LB}$ as functions of temperature which agree with results of others. Difference in R_{LA} and R_{LB} are suspected to be originating to that in the number of magnetic neighbors in A and B sites.

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