

THE COMPLEX DIELECTRIC CONSTANT AND PERMEABILITY OF Ni-Cd FERRITE SYSTEM AT MICROWAVE FREQUENCIES*

BY

Susumu TAKEMOTO**

(Received December 17, 1957)

ABSTRACT

The complex dielectric constant $\epsilon = \epsilon' - j\epsilon''$ and permeability $\mu = \mu' - j\mu''$ of many ferrites have already been studied in low and radio frequency regions, and excellent ferrites have successfully been utilized for many electrical devices. The present author measured the dielectric constant and permeability of the polycrystalline sintered ferrite system $\text{Ni}_{1-x}\text{Cd}_x\text{Fe}_2\text{O}_4$ at 9300 MC and 24000 MC at room temperature. Such experiments as those by the present author have been made only seldom, and especially for the Ni-Cd ferrite system, no experiment seems to have been carried out yet so far as the author is aware of. In this paper, the mechanisms of conductivity and of dispersion of permeability at microwave frequencies will be discussed, and the relationship between conductivity and dielectric constant will also be discussed.

1. Introduction

The dielectric and magnetic properties of ferrites are greatly improved by making the solid solution of normal spinel ferrite in inverse spinel ferrite. This fact is very interesting from both theoretical and practical points of view, and recently a great number of studies (1) on such mixed ferrites have been carried out in low and radio frequency regions, but the studies (2) in the microwave range have been rather inactive. While zinc ferrite has usually been employed as a normal spinel, cadmium ferrite has been used seldom due to some reasons.

Both cadmium ferrite and zinc ferrite have the normal spinel structure, and it is known that the radii of Cd ion and Zn ion are 1.03 Å and 0.83 Å respectively, while the radius of oxygen ion is 1.32 Å. So, it is expected that the electric and magnetic properties will be influenced by this and other differences between Cd ion and Zn ion. The author measured the complex dielectric constant and permeability of Ni-Cd ferrite system in the microwave region simultaneously by the so-called short-circuit and open-circuit method. According to the experimental data obtained, the conductivity σ and the dielectric constant ϵ have a close relation with each other and especially a

* Communicated by Professor Isao TAKAHASHI.

** Department of Physics, Faculty of Literature and Science, Shimane University.

remarkable reduction appears on both at the 70% Cd concentration. The results of measurement shows that the values of μ' are all less than unity owing to the dispersion phenomenon in the microwave region, and the orders of ϵ' and μ' are 15 and 0.6 at 9300 MC and those of ϵ' and μ' are 10 and 0.4 at 24000 MC.

2. Preparation of ferrite samples

The Ni-Cd ferrite system, $\text{Ni}_{1-X}\text{Cd}_X\text{Fe}_2\text{O}_4$ was prepared by the following process. The values of X were varied from 0 to 1.0 successively. The raw materials NiO, CdO, and Fe_2O_3 of high purity were milled in steel ballmills for about 24 hours, and the mixture of these very fine particles obtained was dried and pre-fired in order to obtain a homogeneous sample. After pre-firing, the material is milled. Then, the powder was pressed in steel dies at the pressure of 1 ton/cm² into a final shape. The final sintering was performed at about 1300°C in the air atmosphere, and the end-product was annealed slowly in the air.

3. Theory of measurement

The dielectric constant and permeability of ferrite are complex quantities, having the resonance absorption in the microwave region by the spin relaxation phenomena. We denote them by $\epsilon = \epsilon' - j\epsilon''$ and $\mu = \mu' - j\mu''$ respectively. Hitherto the following two methods have mainly been adopted for the simultaneous measurements of ϵ and μ in the microwave range. One is the resonant cavity method, in which we can determine separately the imaginary and real parts of ϵ and μ by measuring the change of quality factor and the shift of resonance frequency of the cavity resonator, respectively. Healy (3), Birnbaum (4), Okamura (5), Ogasawara (6) and other researchers discussed the resonant cavity method and attained valuable results in practice. The other is the so-called short-circuit and open-circuit method, and by using it we can obtain the complex ϵ and μ from the measurement of the standing wave ratio and the change of the position of the voltage minimum. Since no special considerations are needed in the theory of the second method, it is a reliable and orthodox method so far as the precise measurement of the standing wave ratio is possible. Birks (7), Nishioka (8), Mori (9) and others studied ϵ and μ of various ferromagnetic substances by this method. Since there are some practical difficulties which limit the first method when applied at the frequencies above 9000 MC,

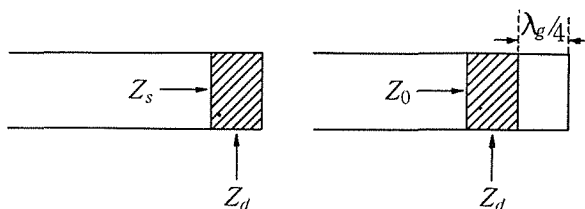


Fig. 1. Schematic representation of the sample at the wave guide termination.

the author adopted the second method. The outline of the theory of this measurement is as follows.

As shown in Fig. 1 the sample is inserted tightly into the wave guide with the free surfaces perpendicular to the guide axis while only the H_{10} wave is excited. The normalized short-circuit and open-circuit impedances are denoted by Z_s and Z_0 respectively in Fig. 1. Then the normalized impedance Z_d and the propagation constant γ in the sample become

$$Z_d = \sqrt{\frac{1 - (\lambda/\lambda_c)^2}{(\epsilon' - j\epsilon'')(\mu' - j\mu'') - (\lambda/\lambda_c)^2}} (\mu' - j\mu''), \quad (1)$$

and

$$\gamma = \frac{2\pi}{\lambda} \sqrt{(\lambda/\lambda_c)^2 - (\epsilon' - j\epsilon'')(\mu' - j\mu'')} \equiv \alpha + j\beta, \quad (2)$$

where λ is the wave length in free space and λ_c is cutoff wave length in the empty guide.

It is well known that

$$Z_s = Z_d \tanh \gamma l, \quad (3)$$

and

$$Z_0 = Z_d \coth \gamma l, \quad (4)$$

where l is the thickness of the sample.

From equations (3) and (4), the following equations are obtained:

$$(Z_s/Z_0)^{1/2} = \tanh \gamma l = \tanh (\alpha + j\beta)l \equiv r e^{j\theta}, \quad (5)$$

where r and θ are the absolute value and the phase angle respectively.

From equation (5), we obtain:

$$\alpha = \frac{1}{2l} \tanh^{-1} \frac{2r \cos \theta}{1 + r^2}, \quad (6)$$

and

$$\beta = \frac{1}{2l} \tan^{-1} \frac{2r \sin \theta}{1 - r^2} + \frac{n\pi}{2}. \quad (7)$$

The integer n is determined from the data obtained on two samples of the same material but of different thickness.

Now we put

$$(\epsilon' - j\epsilon'')(\mu' - j\mu'') = \epsilon_1 - j\epsilon_2, \quad (8)$$

and

$$(\mu' - j\mu'')^2 = \mu_1 - j\mu_2. \quad (9)$$

Then from equations (8), (2), (6) and (7), ϵ_1 and ϵ_2 can be calculated, and from (3) and (4) the following formula is obtained:

$$Z_s Z_0 = Z_a^2 \equiv a + jb. \quad (10)$$

When the left-hand side of the above equation is known, we can calculate a and b , and then from equations (1) and (10), μ_1 and μ_2 are obtained. Therefore from equation (9) μ' and μ'' are determined and by substituting these values into equation (8), ϵ' and ϵ'' are obtained.

4. Description of equipment

A block diagram of the equipment is shown in Fig. 2. Similar equipments were used at both 9300 MC and 24000 MC.

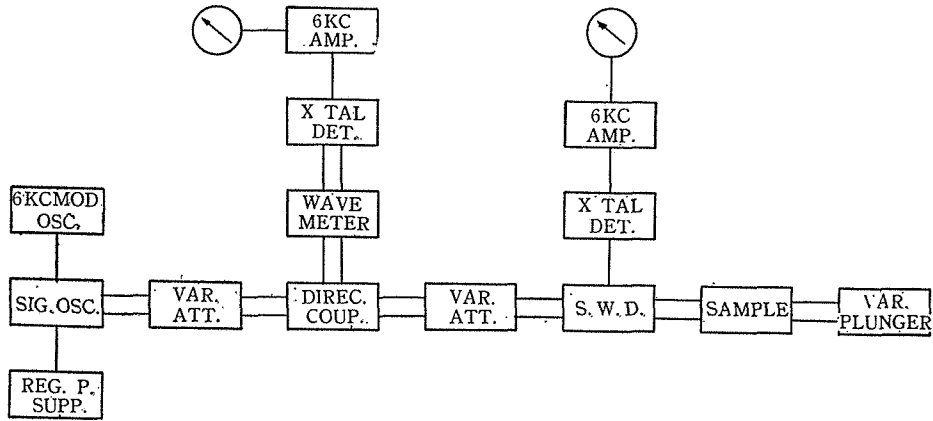


Fig. 2. Block diagram of the equipment used for measuring the complex dielectric constant and permeability of Ni-Cd ferrite system at 9300 MC and 24000 MC.

For the oscillators, Raytheon klystron 2K33 and Sylvania klystron 2K25 were used. The oscillator was modulated by a 6 kc square wave and the output from the detector was amplified by a linear narrow band amplifier. Since the precision of the reading of V. S. W. R. has a serious influence on the final result, the author used the standing wave detector manufactured by Shimada Rika-Kogyo Co. for both frequency ranges. The probe of the detector is movable by a screw micrometer along the main wave guide in such a way that it moves 1/100 mm per revolution with the 24000 MC detector and 1/10 mm with the 9300 MC detector. 1/10 mm was not precise enough for our purpose. From each ferrite, samples were formed to be square-shaped and to have two different thicknesses of 2 mm and 4 mm. Under the present conditions, the skin depth seems to be greater than 10 mm and so the EHF magnetic field is uniform throughout the sample. The output from the oscillator should always be monitored, in order to keep constant the output and frequency. It must be done with care to measure the voltage standing wave ratio, since it has a large effect on the precision of the whole measurement.

5. Result and discussion

A) Dielectric constant and conductivity

The ferrites are naturally semiconductors, and therefore the conductivities of ferrites are greatly affected by the presence of impurities. According to Verwey and de Boer (10) and others (11), the electronic conduction mechanism in oxide semiconductors can be associated with the simultaneous occurrence of metal ions of different valencies of a certain element on the identical lattice sites. Ferrous ions are substituted by other divalent metal ions in ferrites other than magnetite, and therefore these ferrites are practically insulators. But they show a small conductivity owing to the presence of a few ferrous ions which inevitably enter them in the course of heat treatment. The conductivity of ferrites are partly explained by the simultaneous presence of ferrous and ferric ions on the same sublattice in the spinel structure. The dielectric constant ϵ (real) and the resistivity ρ follow the same relaxation formulas (12) as a function of frequency :

$$\left. \begin{aligned} \epsilon &= \epsilon_{\infty} + \frac{\epsilon_0 - \epsilon_{\infty}}{1 + \omega^2 T_e^2}, \\ \rho &= \rho_{\infty} + \frac{\rho_0 - \rho_{\infty}}{1 + \omega^2 T_p^2}, \end{aligned} \right\} \quad (11)$$

where ϵ_0 and ρ_0 are the values obtained by static measurement and ϵ_{∞} and ρ_{∞} are those at EHF; T_e and T_p denote the respective relaxation times. From these expressions ϵ and ρ at extremely high frequencies should tend to the limiting values ϵ_{∞} and ρ_{∞} respectively. As shown in Fig. 3 the high frequency limit of ϵ' seems to approach 10. As to the value of σ , it is known that the high frequency limit (13) becomes several times greater than its DC value. The result of measurement of DC resistivity ρ of the Ni-Cd ferrite system is given in Fig. 4, and this curve shows that one can obtain the ferrite of increased resistivity by properly mixing Ni-ferrite and Cd-ferrite.

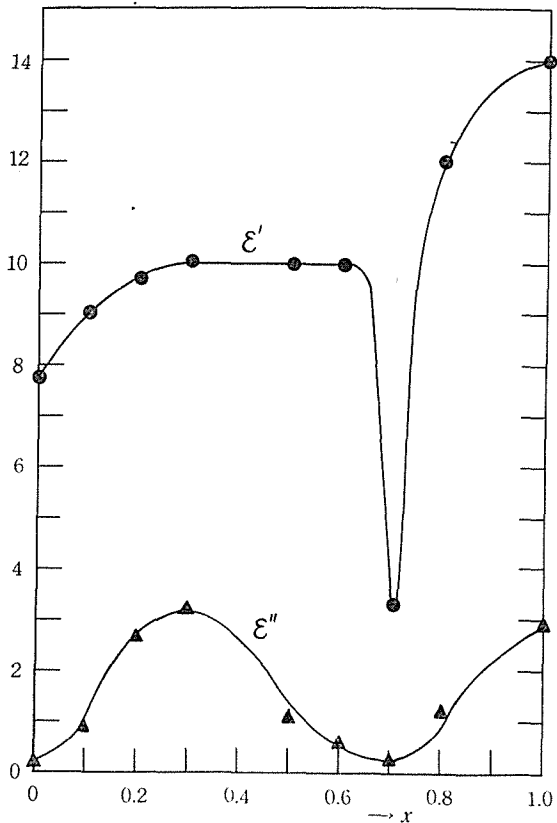


Fig. 3. ϵ' and ϵ'' measured respectively for various contents of Cd ion at 24000 MC.

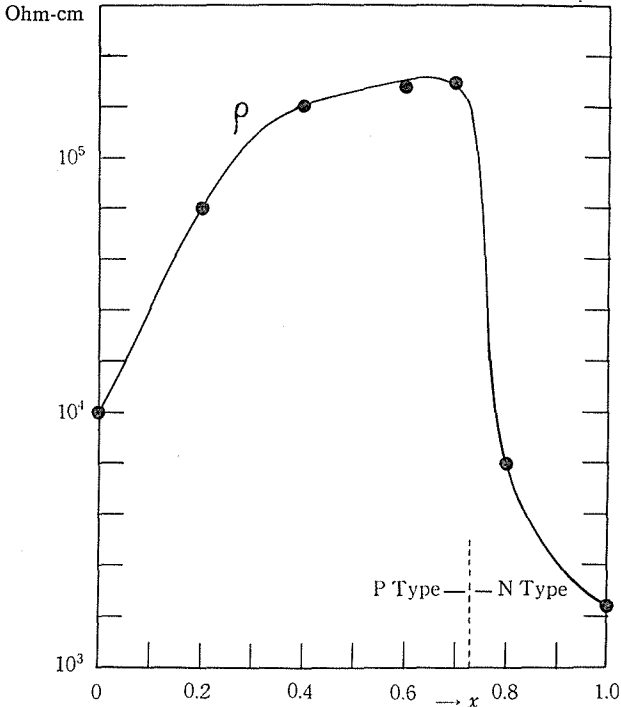


Fig. 4. The resistivity ρ of Ni-Cd ferrite system before surface grinding.

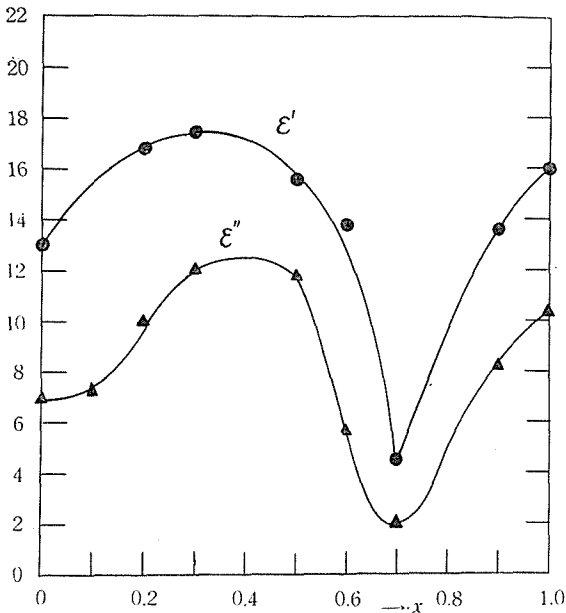


Fig. 5. ϵ' and ϵ'' measured respectively for various contents of Cd ion at 9300 MC.

It is remarkable that as seen in Figs. 3, 4 and 5, both ρ and ϵ' show a sharp reduction at the 70% Cd content. As seen in Figs. 3, 4 and 5 ϵ' increases together with ρ at the compositions between $X=0$ and $X=0.7$ and decreases above $X=0.7$. The break of the ρ curve at $X=0.7$ may be considered partly due to the cancelling effect of possible P-type (14) and N-type conduction mechanisms since Cd-ferrite is an N-type conductor. From these results it can be said that dielectric constant ϵ' has a close relation with conduction electrons, but the precise explanation of these phenomena should be postponed to future studies.

B) *The complex initial permeability and magnetic loss*

The magnetic structure of ferrite was first explained by Néel (15) in 1948 and the name 'ferrimagnetism' was proposed.

Fig. 6 shows that the saturation magnetization increases with the concentration of Cd-ferrite in Ni-ferrite up to $X=0.4$ contrary to the opposite expectation since it is non-magnetic, and then it decreases. This tendency was already observed on Ni-Zn ferrite which has the maximum saturation at $X=0.5$. As the interionic distances among the magnetic ions of Ni-Cd ferrite are somewhat

larger than those of Ni-Zn ferrite, it seems reasonable that the maximum saturation of Ni-Cd ferrite occurs at the smaller value of X . Further, in Fig. 6 the sharp reduction of magnetization is seen at the Cd content of $X = 0.7$. As is well known, these phenomena can be explained by the theory of ferrimagnetism. It is conspicuous that ρ , ϵ' and saturation magnetization M_s have all a break at $X = 0.7$. Notwithstanding the high resistivity, ferrites generally show pronounced ferromagnetic absorptions and large magnetic losses at high frequencies. This resonance absorption was first found in 1947 by Snoek (16) in the experiments on some mixed ferrites and named 'natural ferromagnetic resonance' or rotational resonance. The width of this resonance curve is very large owing to its origin and $(\mu' - 1)$ becomes negative due to the dispersion phenomena in the microwave region. The curves of μ' and μ'' versus Cd content are shown in Figs. 7 and 8. The mean values of μ' and μ'' are about 0.4 and 0.35, respectively at 24000 MC. These values are smaller than those at 9300 MC. When we discuss the initial permeability

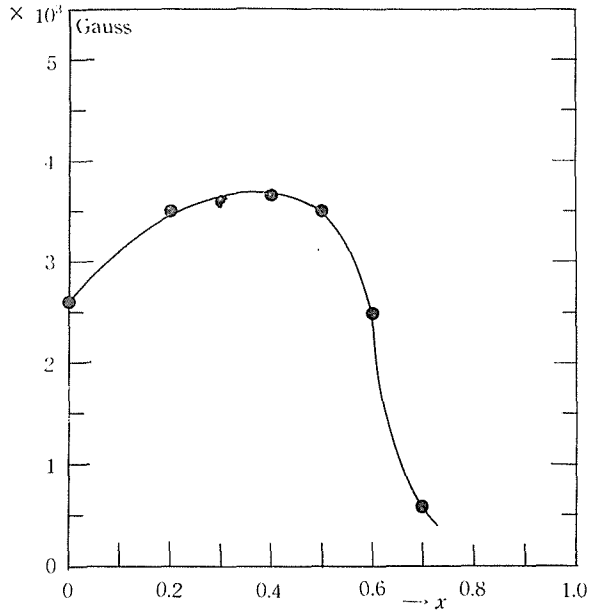


Fig. 6. Dependence of saturation magnetization $4\pi M_s$ for Ni-Cd ferrite on concentration of Cd ion.

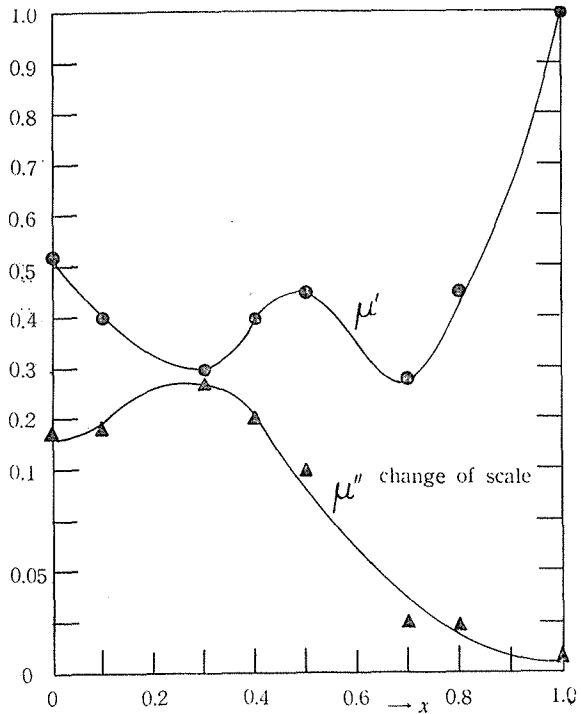


Fig. 7. μ' and μ'' of ferrite at 24000 MC.

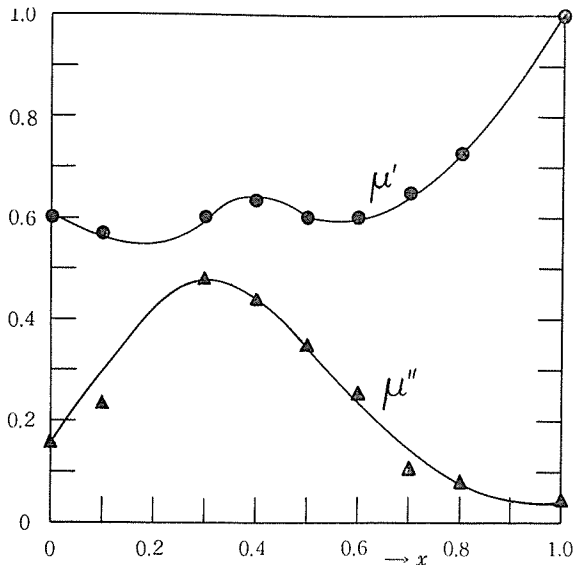


Fig. 8. μ' and μ'' of Ni-Cd ferrite at 9300 MC.

internal anisotropy field. When there are the demagnetization fields in addition to the internal effective field, the condition of the resonance and the value of permeability are naturally affected. In such cases the maximum resonant frequency (17) is

$$\omega_r = \gamma(H_A + 4\pi M_s), \quad (12)$$

where γ is the gyromagnetic ratio and H_A denotes the anisotropy field. For a free electron the numerical value of γ is $1.76 \times 10^7 \text{ sec}^{-1} \cdot \text{oersted}^{-1}$ and usually H_A can be neglected as compared with $4\pi M_s$. In the present case the maximum resonant frequency exists in about 10,000 MC region which is the important frequency range in practice, and therefore the energy loss (μ'') shows a decrease with higher frequencies, which explains the experimental results shown in Figs. 7 and 8. Because of the dispersion due to the broad rotational resonance absorption, the ferrite of high μ' value will be difficult to obtain at microwave frequencies, but the mixed ferrites such as those used in this experiment may be utilized as low loss attenuator for microwave devices.

Acknowledgment

The author wishes to express his sincere thanks to Professor I. Takahashi of Kyoto University for his valuable advice and discussion. He is also indebted to Assistant Professor H. Mori of Shimane University for his help in the experiment and Tokyo Denki Kagaku-Kogyo Co. for preparing the excellent ferrite samples used for the present experiment. The present investigation has been partly supported by a grant-in-aid for scientific researches of the Ministry of Education.

μ of ferrites at microwave frequencies, the hysteresis loss and eddy current loss can be neglected, but the residual loss cannot be neglected. The residual loss at the frequencies higher than 1 MC is composed of the losses corresponding to rotational, Bloch wall, and dimensional resonances in general, but in the present experiment, only the rotational resonance might be considered. The rotational resonance loss is ascribed to the absorption of energy due to the spin rotation in the

REFERENCES

- 1, 2. Solid-state Electronics, Proc. I. R. E. **43** (1955), 245 ;
Advances in Electronics and Electron Physics, Vol. 2, p. 251 & Vol. 4, p. 70 (Academic Press, 1954) ;
J. L. Snoek, *New Developments in Ferromagnetic Materials* (Elsevier, 1949) ;
Rev. Mod. Phys. **25**, No. 1 (1953).
Ferrites Issue, Proc. I. R. E. **44**, No. 10 (1956).
3. D. W. HEALY, Phys. Rev. **86** (1952), 1009.
4. G. BIRNBAUM, J. Appl. Phys. **20** (1951), 817.
5. T. OKAMURA, T. FUJIMURA and M. DATE, Scientific Reports of the Research Institute, Tohoku Univ., **73** (1952), 191.
6. N. OGASAWARA, Rev. E. E. Japan, **74** (1954), 795.
7. J. B. BIRKS, Proc. Phys. Soc. **60** (1949), 282.
8. A. NISHIOKA, Reports Res. Inst. Comm., No. 277 (1952), 5.
9. H. MORI, Bulletin Shimane Univ. (Natural Science), No. 6 (1956), 34.
10. E. J. W. VERWEY and J. H. de BOER, Recueil des travaux chimiques des Pays-Bas, **55** (1936), 531.
11. E. J. W. VERWEY and P. W. HAAYMAN, Physica **8** (1941), 979.
E. J. W. VERWEY and P. W. HAAYMAN, Philips Research Report, **5** (1950), 173.
12. C. G. KOOP, Phys. Rev. **83** (1951), 121.
13. J. SMIT and H. P. J. WIJN, *Advances in Electronics and Electron Physics*, Vol. 4 (1955), 119.
14. L. G. VAN UITERT, J. Chem. Phys. **23** (1955), 1183.
15. L. NÉEL, Ann. de Phys., **3** (1948), 137.
16. J. L. SNOEK, Nature **160** (1947), 90 ; Physica **14** (1947), 207.
17. D. POLDER and J. SMIT, Rev. Mod. Phys., **25** (1953), 89.