

METHODS OF
Experimental Physics

VOLUME 6 - PART B

SOLID STATE PHYSICS

Volume 6


Solid State Physics

Edited by

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Purdue University, Lafayette, Indiana

PART B
Electrical, Magnetic,
and
Optical Properties

1959 

ACADEMIC PRESS • New York and London

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ACADEMIC PRESS INC.
111 FIFTH AVENUE
NEW YORK 3, N. Y.

United Kingdom edition published by
ACADEMIC PRESS INC. (London) LTD.
40 PALL MALL, LONDON, S. W. 1

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Library of Congress Catalog Card Number: 59-7686

PRINTED IN THE UNITED STATES OF AMERICA

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FOREWORD TO VOLUME 6

A few months ago, Volume I of this series, entitled "Classical Methods," came off the press. In rapid succession, we are presenting now Volume VI (A and B) of this series, devoted to solid state physics. It may be worth repeating some of the aims set for these publications.

When starting on the task of creating this new series, we—and by we, I mean the general editors and the volume editors—defined the aims as follows: "The book should be a concise, well-illustrated presentation of the most important methods, or general principles, needed by the experimenter, complete with basic references for further reading. Indications of limitations of both applicability and accuracy are an important part of the presentation. Information about the interpretation of the experiments, about the evaluation of errors, and about the validity of approximations should also be given. The book should not be merely a description of laboratory techniques, nor should it be a catalogue of instruments." Part of our aim was also to organize a series in such a manner that they should provide not only the advanced research worker but also the graduate student and the teacher with a good means for carrying out his experimental research or for starting him on a new task when he is already familiar with the basic principles. Also, we believe, the title of the series should really mean what it says, in the sense that the method is more important than gadgetry.

The task of converting these ideas into solid fact was taken over by Professors Karl Lark-Horovitz and Vivian A. Johnson. The untimely death of Professor Lark-Horovitz did not alter in any appreciable manner the scope and outline of the work as it had been planned by him, together with Professor Johnson. In fact, it is a great tribute to the planning of these two outstanding scientists that the death of one of them did not stop the work at all, but that it continued in the original spirit with unabated vigor. I am convinced that as these volumes stand now, they will constitute a lasting memorial to that great physicist, Karl Lark-Horovitz.

It is my pleasant duty to thank here publicly all those who contributed to creating these volumes. Besides the basic work of the volume editors, it is a pleasure to mention all the authors, the officials and staff of the Academic Press Inc., and Mrs. Claire Marton. The wonderful cooperative spirit which animated all these individuals finds its reward in the books we are presenting herewith.

L. MARTON

Washington, D. C.
June, 1959

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7. ELECTRICAL PROPERTIES

7.1. Dielectric Measurement Procedures* †

7.1.1. Introduction and Definitions

In the eighteenth century Franklin and Cavendish recognized that the amount of charge which could be induced upon the plates of a capacitor by an applied potential depended upon the nature of the insulation between the plates. But it remained for Faraday in the early nineteenth century to formulate the quantitative relations between induced charge, applied potential, electrode geometry, and dielectric material in a capacitor. Faraday pointed out that the amount of charge per unit of applied potential (i.e., the capacitance C) of a system with arbitrary electrode geometry was directly proportional to a characteristic property of the insulation material which is now called permittivity. When normalized with respect to air or vacuum he called this property the "specific inductive capacity." The normalized quantity will be called the dielectric constant or relative permittivity ϵ_r . Thus $C/C_0 = \epsilon/\epsilon_0 = \epsilon_r$, where C_0 is the capacitance of the same capacitor with air or vacuum instead of dielectric between its electrodes. Likewise ϵ is the permittivity of the dielectric and ϵ_0 is that of vacuum. The exact meaning of the above definition of ϵ_r will become clearer when its measurement has been explained.

Since it is now known that the dielectric constant of a material may depend strongly on the frequency of the applied potential or on the temperature, it might perhaps seem preferable not to use the word "constant" to describe this property. However, the name dielectric constant is so well established that its use continues. The names permittivity and relative permittivity are often used interchangeably without making the distinctions noted above.

If a sinusoidal voltage of frequency $\omega/2\pi$ cycles per second is applied to a capacitor, the relation between current I and voltage V may be written:

$$I = C dV/dt = i\omega VC = i\omega V\epsilon_r C_0. \quad (7.1.1)$$

In a simple lossless capacitor, having air or vacuum in lieu of dielectric between its plates, the current would lead the voltage by 90° and $C(= C_0)$ would be a pure real number. However, in a capacitor containing a solid dielectric, the phase difference is somewhat less than 90° , say $90^\circ - \delta$.

† See also Vol. 1, Chapter 8.5 and Vol. 2, Section 10.6.3.

* Chapter 7.1 is by A. H. Sharbaugh and S. Roberts.

Then, according to Eq. (7.1.1), ϵ_r would be a complex number which could be resolved into its real and imaginary components as follows:

$$\epsilon_r = \epsilon_r' - i\epsilon_r''; \quad \tan \delta = \epsilon_r''/\epsilon_r'. \quad (7.1.2)$$

Here, the total current may be resolved into two components: a charging current in quadrature with the voltage, and a conduction current in phase with the voltage. The vector resolution of the current is shown schematically in Fig. 1. The loss tangent $\tan \delta$ is identical to the power factor $\cos \theta$ for small values of δ , since the angles θ and δ are complementary.*

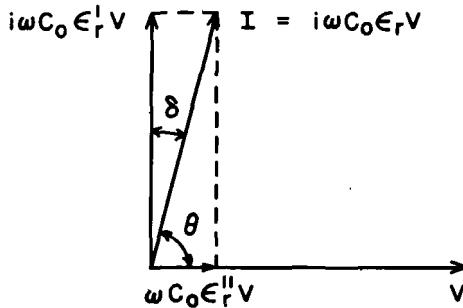


FIG. 1. Vector resolution of the ac current in a capacitor.

The conduction component of the current may be used to define the effective conductivity of the dielectric σ according to the relation:

$$\sigma = \omega \epsilon_r'' \epsilon_0. \quad (7.1.3)$$

The effective conductivity defined in this manner depends upon frequency and is almost always greater than the dc conductivity. The difference is often called the ac conductivity.

The "lumped" circuit interpretation used above becomes inappropriate in the microwave frequency range, although the concept of a complex dielectric constant applies just as well. It describes the physical fact that a dielectric in a periodic electromagnetic field carries an effective current density,

$$J = \sigma E + \partial D/\partial t = i\omega(\epsilon' - i\epsilon'')E \quad (7.1.4)$$

where E and D are the electric intensity and the electric displacement, respectively. The current density in this case may be resolved into its real and imaginary components. The imaginary component J_c represents a charging current density, while the real component represents a loss—or conduction current density J_l .

$$J_c = i\omega \epsilon_r' \epsilon_0 E; \quad J_l = \omega \epsilon_r'' \epsilon_0 E = \sigma E. \quad (7.1.5)$$

* See also Vol. 2, Section 2.5.2.

Important electrical properties of solid dielectrics include dielectric constant (real part), loss tangent, electric strength, surface resistivity, tracking resistance, and volume resistivity. Here will be discussed the measurement of the first three of these; the last being covered in Chapter 7.2 of the present volume. To meet the practical requirements of the electrical industry and the need for fundamental studies of the mechanism of these phenomena, the measurement of these dielectric parameters has become a very important and highly specialized branch of measurements which includes many different techniques and methods. It would be neither possible nor desirable in this limited treatment to cover all of the many techniques that have been developed through the years. Certain classical methods will not be treated here. These include, for example, the measurement of capacitor charge and discharge rates, calorimetric techniques, ponderomotive methods, and refraction of plane waves by prisms. However, those methods which are exemplary of the general principles which will enable an experimenter to select a method to meet his special requirements will be discussed.

7.1.2. Specimens and Electrode Configurations

7.1.2.1. Uniform Electric Field. The choice of specimen dimensions and the arrangement of the electrodes are perhaps the most critical matters connected with measurements of the complex dielectric constant. Considerable ingenuity has been exercised in devising a variety of techniques to cope with the various problems. Practically every method has some disadvantages as well as advantages. Only some of the principles involved will be discussed.

The primary requirement is to choose a dielectric specimen of such shape and with suitable electrodes that the dielectric constant and loss may be calculated from the dimensions and the measured electrical admittance. The simplest and most common shape for this purpose is a flat plate or disk, of uniform thickness t . Electrodes may be applied in the form of metal foil or as painted, sprayed, or evaporated metal to the surfaces of the plate. Suitable techniques for doing this are described in the ASTM Standards on Electrical Insulating Materials¹ and in an article by Hartshorn *et al.*² If circular electrodes of radius r are applied on opposite sides of the plate, then the admittance Y , at a frequency $\omega/2\pi$ cycles/sec, is given in reciprocal ohms approximately by the formula:

$$Y = i\omega\pi r^2\epsilon_0\epsilon_r/t \quad (7.1.6)$$

¹ *Am. Soc. Testing Materials, ASTM Standards 1955, Part 6, pp. 500-547 (1955); Designation D150-54T.*

² L. Hartshorn, W. H. Ward, B. A. Sharpe, B. J. O'Kane, *J. Inst. Elec. Engrs. (London)* **75**, 730 (1934).

where r and t are expressed in meters and ϵ_0 has the value 8.854×10^{-12} farad/meter. If r and t are given in centimeters, ϵ_0 would be 8.854×10^{-14} . The ϵ_r stands for the complex dielectric constant. If the admittance is equated to that of a capacitance C_x in parallel with a resistance R_x , one obtains:

$$Y = \frac{1}{R_x} + i\omega C_x = \frac{\omega\pi r^2\epsilon_0}{t} (\epsilon_r'' + i\epsilon_r').$$

Hence,

$$\epsilon_r' = \frac{C_x t}{\pi r^2 \epsilon_0}; \quad \epsilon_r'' = \frac{t}{\pi r^2 \omega \epsilon_0 R_x}; \quad \tan \delta = \frac{\epsilon_r''}{\epsilon_r'} = \frac{1}{\omega C_x R_x}. \quad (7.1.7)$$

If C_x and R_x are determined from experiment, the above relations show how to calculate ϵ_r' , ϵ_r'' , and $\tan \delta$ from them. Instruments for measuring C_x and R_x of dielectric specimens are described in Section 7.1.3.

The above equations are a good approximation if the electric field is concentrated in the dielectric in the region between the electrodes and is of negligible intensity elsewhere. Since this is ordinarily not the case, it is often necessary to make a correction for the nonuniform electric field near the edge of the electrodes. This correction is discussed in detail by Scott and Curtis.³

There are at least two methods of avoiding the need for edge corrections. One of these makes use of so-called "micrometer" electrodes. These are plane parallel circular electrodes whose spacing may be controlled and measured by a fine micrometer adjustment. The dielectric sample is a flat disk smaller in diameter than the electrodes by at least twice its thickness. The capacitance and conductance are measured with the electrodes in contact with the sample and then with the sample removed but with the same electrode spacing. The difference in admittance, $1/R_x + i\omega \Delta C_x$, is equal to the difference between the admittance of the sample and that of the air which it displaces. Consequently the formula for ϵ_r' is somewhat different from Eq. (7.1.7).

$$\epsilon_r' = 1 + \Delta C_x t / \pi r^2 \epsilon_0; \quad \epsilon_r'' = t / \pi r^2 \omega \epsilon_0 R_x. \quad (7.1.8)$$

In the above arrangement the electric field is uniform in the sample, and the constant edge capacitance of the micrometer electrodes cancels out in the difference in capacitance. This method should be used with caution at low frequencies, since it in no way compensates for conduction across the edge surface of the sample.

The second method of eliminating the effects of edge capacitance makes use of a guard electrode. This may be an annular electrode concentric with the main guarded electrode on one side of the sample. The

³ A. H. Scott and H. L. Curtis, *J. Research Natl. Bur. Standards* **22**, 747-775 (1939).

unguarded electrode on the other side overlaps both the guard and guarded electrodes. In measurements with this three-electrode arrangement, both guard and guarded electrodes are held at the same potential. Only the direct capacitance between the main electrodes is measured. Suitable dimensions for three-electrode systems are given in the ASTM Standards.¹

7.1.2.2. Nonuniform Electric Field. A parallel, approximately uniform electric field can be maintained in a dielectric plate only if its dimensions are small compared with the wavelength. In order to work with samples of reasonable size at microwave frequencies, it becomes necessary to insert them in resonators or waveguides where the electric field is not uniform. For example, the dielectric sample may be included as a short section at the short-circuited end of a coaxial line or a waveguide. The experimental problem, then, is to measure the admittance or impedance of the front surface of the dielectric plug. Instruments for doing this are described in Section 7.1.3.3.1.* The purpose of the present section is to show how the impedance, if known, may be used in a calculation of the complex dielectric constant.

A detailed discussion of the propagation of electromagnetic waves in various types of waveguides is to be found in appropriate texts and will not be repeated here. Ramo and Whinnery⁴ have classified the modes of transmission and have explained how the concept of impedance may be applied to waveguides. Their notation and definitions of the characteristic impedance Z_2 , and propagation constant γ_2 , of the dielectric-filled guide will be adopted. Transmission line formulas are used to determine the impedance Z_a of the front surface of a dielectric plug of thickness d in the closed end of the waveguide. The impedance in ohms is

$$Z_a = Z_2 \tanh \gamma_2 d. \quad (7.1.9)$$

Note that both Z_2 and γ_2 are complex quantities.

The relations between the characteristic impedance, the propagation constant, the permeability μ , the permittivity $\epsilon = \epsilon_0 \epsilon_r$, and the dimensions of the guide are likewise derived by Ramo and Whinnery.⁴ If Z_1 and γ_1 are the characteristic impedance and propagation constant for the empty waveguide, the following relations are shown to be valid for transverse electric (TE) waves in which the electric field is normal to the direction of propagation.

$$\gamma_2 Z_2 = i\omega\mu_2; \quad \gamma_1 Z_1 = i\omega\mu_0. \quad (7.1.10)$$

* See also Vol. 2, Chapter 10.2.

⁴ S. Ramo and J. R. Whinnery, "Fields and Waves in Modern Radio," 2nd ed. Wiley, New York, 1953.

Since it is true for most dielectric materials that $\mu_2 = \mu_0$, the permeability of free space, one obtains $\gamma_2 Z_2 = \gamma_1 Z_1$, and

$$\frac{Z_a}{Z_1 \gamma_1 d} = \frac{\tanh \gamma_2 d}{\gamma_2 d} \quad (7.1.11)$$

Now, Z_a/Z_1 is the impedance ratio that is determined by experiment, d is the measured thickness of the dielectric, and γ_1 is equal to $i2\pi/\lambda_1$, where λ_1 is the wavelength in the empty waveguide. Hence the left side of Eq. (7.1.11) may be evaluated completely. Then $\gamma_2 d$ may be derived from charts of the function on the right.^{5,6}

For TE waves the dielectric constant may then be found from the relation:

$$\epsilon_{r2} = \frac{\epsilon_2}{\epsilon_0} = \frac{1}{\omega^2 \epsilon_0 \mu_2} \left[\left(\frac{2\pi}{\lambda_c} \right)^2 - \gamma_2^2 \right] \quad (7.1.12)$$

where λ_c is the cut-off wavelength of the waveguide. The evaluation of the expression on the right in Eq. (7.1.12) is perfectly straightforward if it is assumed again that $\mu_2 = \mu_0$.

Since there is some ambiguity in the evaluation of $\gamma_2 d$ from charts, one needs at least two measurements to determine ϵ_{r2} uniquely. The second measurement may be done either with a different thickness of dielectric or with an air space between the dielectric and the closed end of the guide. In either case the results of two measurements also suffice for calculating ϵ_{r2} uniquely, when $\mu_2 = \mu_0$, even without the aid of charts.

Suppose, for example, that the second measurement is done with the rear surface of the dielectric plug located a quarter wavelength from the end of the guide.⁷ The impedance at the front surface may then be shown to be

$$Z_b = Z_2 \coth \gamma_2 d. \quad (7.1.13)$$

By combining Eqs. (7.1.9) and (7.1.13), one obtains:

$$Z_2 = \sqrt{Z_a Z_b}. \quad (7.1.14)$$

When $\mu_2 = \mu_0$, γ_2 may be calculated unambiguously, according to Eq. (7.1.10). This result should also agree with that obtained by dividing Eq. (7.1.9) by Eq. (7.1.13).

$$\gamma_2 = \frac{1}{d} \tanh^{-1} \sqrt{\frac{Z_a}{Z_b}} = \frac{1}{2d} \ln \frac{\sqrt{Z_b} + \sqrt{Z_a}}{\sqrt{Z_b} - \sqrt{Z_a}} \quad (7.1.15)$$

⁵ S. Roberts and A. von Hippel, *J. Appl. Phys.* **17**, 610-616 (1946).

⁶ W. B. Westphal, in "Dielectric Materials and Applications" (A. von Hippel, ed.), pp. 63-122. Wiley, New York, 1954.

⁷ W. H. Surber, Jr. and G. E. Crouch, Jr., *J. Appl. Phys.* **19**, 1130 (1948).

Some ambiguity will be encountered when Eq. (7.1.15) is used for evaluation of γ_2 for dielectrics in which $\mu_2 \neq \mu_0$. Finally, ϵ_2 or ϵ_{r2} may be determined by using Eq. (7.1.12).

Another method involves a second measurement with twice the thickness of dielectric used in the first.⁸ The principles involved in the calculation by this method are similar to those already described.

In the above equations the waveguide losses in the section containing the sample have been neglected. For low-loss materials it may be necessary to make a correction for these losses. Formulas suitable for this correction are summarized by Westphal.⁶ Special considerations for measurement of materials with extra high dielectric constants are discussed by Powles and Jackson.⁹

7.1.2.3. Miscellaneous Arrangements. In the preceding discussion it has been tacitly assumed that the dielectric could be shaped into a specimen with an accurately known thickness and with plane parallel surfaces to which electrodes could be properly applied. If, for some reason, it is undesirable to apply electrodes directly to such a sample, then the series impedance of the sample and an accurately known air gap may be measured in a micrometer test cell. The complex dielectric constant ϵ_r of the sample is then computed to a good approximation by the series capacitor relation. Should the sample be rough and of indeterminate thickness, the capacitance and loss of a series arrangement of the sample immersed in any two fluids of known dielectric constant at constant electrode spacing may be made. The two simultaneous equations for the series impedance may then be solved to obtain the complex dielectric constant of the solid and its fractional thickness without making any thickness measurement on the sample.¹⁰ In the case of high dielectric constant solids, certain methods of plotting the data may be useful.¹¹

On the other hand, the sample may be in the form of a low-loss powder, crystal, or fiber, which cannot be formed into a sheet sample at all. Then it is necessary to use the "method of mixtures"^{10,12,13} which consists of mixing two low-loss liquids whose dielectric constants bracket that anticipated for the solid (ϵ_r') until a liquid composition is found with which the capacitance of a liquid filled cell is the same with and without the immersed solid. This composition, at which the dielectric

⁸ K. V. G. Krishna, *Trans. Faraday Soc.* **52**, 1110-1111 (1956).

⁹ J. G. Powles and W. Jackson, *Proc. Inst. Elec. Engrs. (London)*, Pt. III **96**, 383 (1949).

¹⁰ H. S. Endicott and W. F. Springgate, *Ann. Rept., Conf. on Elec. Insul.* **1950**, p. 43 (1951).

¹¹ E. Kleinke, *Physik. Z.* **36**, 565 (1935).

¹² H. Starke, *Ann. Physik* [3] **60**, 629 (1897).

¹³ S. Whitehead and W. Hackett, *Proc. Phys. Soc. (London)* **51**, 173 (1939).

constant of the liquid–solid mixture (ϵ_m) equals that of the liquid (ϵ_l), is usually obtained by interpolation from a plot of ($\epsilon_m - \epsilon_l$) against liquid composition. From a knowledge of the value of ϵ_l as a function of composition, ϵ_r' is determined by the relation $\epsilon_l = \epsilon_m = \epsilon_r'$. The use of this method requires that the dielectric losses in both the solid and immersion liquid be negligible and that the solid be electrically homogeneous. Sometimes the dielectric constant of the immersion liquid is matched to that of the solid by changing the temperature of the liquid–solid mixture.¹⁴ In this way the use of a single liquid whose dielectric constant depends strongly on the temperature makes it easier to avoid disturbing air bubbles and moisture when ϵ_l is changed in the usual manner by mixing two liquids.

With irregularly shaped solids having high dielectric constants, it may be difficult to obtain a matching liquid of sufficiently high dielectric constant. Furthermore, such a liquid may have a large dielectric loss and high chemical reactivity toward the specimen. However, it is still possible to estimate the dielectric constant (ϵ_r') by a measurement of the composite dielectric constant (ϵ_m) of the particles mixed with a liquid of known dielectric constant (ϵ_l). The value of ϵ_r' is computed¹⁵ by the use of an equation such as

$$\epsilon_r' = \epsilon_l \frac{(2 + v)\epsilon_m - 2(1 - v)\epsilon_l}{(1 + 2v)\epsilon_l - (1 - v)\epsilon_m} \quad (7.1.16)$$

where v is the fractional volume of the solid between the electrodes. Since this equation is derived with the assumptions that the solid particles are spherical in shape and small compared to the size of the measuring cell, it is advisable to calculate ϵ_r' from measurements made with several immersion media to establish confidence in the computed value. With values of dielectric constant larger than about five, the shape of the particles must be taken into account.^{16,17}

7.1.3. Instruments for Measuring Admittance

7.1.3.1. Bridges and Other Null Devices. 7.1.3.1.1. INTRODUCTION. The capacitance or complex admittance of a dielectric specimen may be measured in a variety of ways by appropriate instruments. The purpose of this section is to outline briefly the general types of instruments which may be used in these measurements and to point out their principal advantages and limitations.

¹⁴ K. Hojendahl, *Z. physik. Chem. (Leipzig)* **B20**, 54 (1933).

¹⁵ S. Roberts, *J. Opt. Soc. Am.* **42**, 850 (1952).

¹⁶ J. C. Van Vessel and J. M. Bijvoet, *Rec. trav. chim.* **67**, 191 (1948).

¹⁷ D. Polder and J. H. Van Santen, *Physica* **12**, 257 (1946).

The generally preferred method in a wide range of frequency is based on the null-balance principle.* Instruments may be designed on this basis for operation at any frequency from subaudio to the shortest microwaves. The most effective applications of this principle are in the audio- and moderate radiofrequency range, within which circuit elements may be regarded as having lumped parameters. Three types of instruments have been selected as representative of this group; namely, the Schering

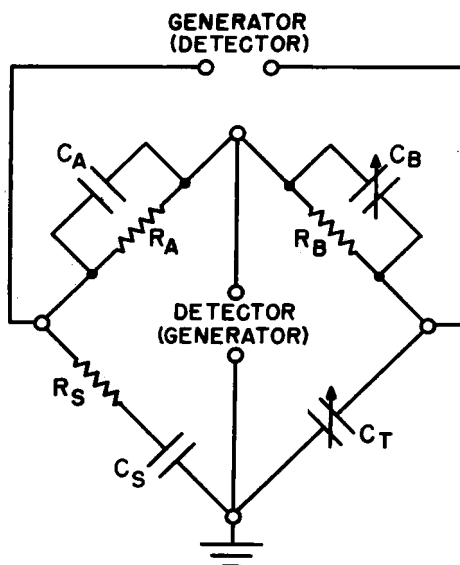


FIG. 2. Basic Schering bridge circuit.

bridge, the transformer or hybrid-coil bridge, and the parallel-T. A more thorough treatment of impedance bridges is given by Hague.¹⁸

Impedance bridges and other equivalent devices developed for microwave frequencies will not be reported here in detail. A general discussion of microwave impedance bridges is given by Young.¹⁹ † A microwave bridge employing two hybrid waveguide junctions and designed specifically for dielectric measurements is described by Beljers and Van de Lindt.²⁰

7.1.3.1.2. SCHERING BRIDGE. The basic schematic circuit of the Schering bridge is shown in Fig. 2. In this circuit R_A and R_B are fixed

* See also Vol. 2, Section 9.5.2.4.

† See also Vol. 2, Section 10.5.1.2.

¹⁸ B. Hague, "Alternating Current Bridge Methods." Pitman, London, 1946.

¹⁹ L. B. Young, in "Technique of Microwave Measurements" (C. G. Montgomery, ed.), Chapter 9, pp. 515-560. McGraw-Hill, New York, 1947.

²⁰ H. G. Beljers and W. J. Van de Lindt, *Philips Research Rept.* **6**, 96 (1951).