Goddard Space Flight Center
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ON THE ANALYSIS OF THE DIELECTRIC DISPERSION OF FERRITES

by

P. H. Fang

ABSTRACT

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Several models which have been used to analyze the low frequency dielectric dispersion of Ni-Zn or Mn ferrites are discussed. Based on the analytic nature of the dispersion functions corresponding to these models, it is shown what experimental data are required to decide the validity of specific models.

Einige Modelle, die zur Analyze der dielektrischen Niederfrequenzdispersion von Ni-Zn oder Mn Ferriten untersucht wurden, werden besprochen. Auf Grund der analytischen Natur der diesen Modellen entsprechenden Dispersionfunktionen wird gezeigt, welche experimentellen Daten erforderlich sind, um die Echtheit von spezifischen Modellen zu bestimmen.

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I. INTRODUCTION

In a recent work, Krotzsch proposed a new model of dielectric relaxation of ferrites at low frequencies.[1] Since the discovery of an anomulous increase of the dielectric constant at low frequencies of some Mn-ferrites,[2] a large number of publications have appeared on this subject, 18 references are given in Ref. 1, to which we will repeat the work of Koops[3] and of Kamiyoshi,[4] which we will discuss. We also would like to add an interesting reference of Grant.[5] In fact, the work of Grant is not quite complete because he has not analyzed explicitly the mathematical implications. Therefore, we will devote Part II to discuss the underlying mathematical formulations. In Part III, we will compare different models and point out some decisive tests of those models.

II. COMPLEX CONDUCTIVITY REPRESENTATION

A convenient representation to analyze the dielectric relaxation data is the so-called Cole-Cole diagram[6] where the trace of the frequency is plotted in the complex plane of the dielectric permittivity. Recently Grant[5] has shown that another representation has some desirable features, that is, plotting the trace of the frequency in the complex plane of the dielectric conductivity. The relation between the complex permittivity, ϵ^*, and the complex conductivity, σ^*, is given by,

[redacted]

$$\sigma^* - \sigma_0 = i\omega(\epsilon^* - \epsilon_\infty), \tag{1}$$

where

$$\sigma^* = \sigma' + i\sigma'',$$

$$\epsilon^* = \epsilon' - i\epsilon'',$$

and subscripts are to be identified with the value of ω, the frequency. Grant has shown that such a plot is particularly useful at low frequencies where d.c. conduction becomes prominent. Some time ago Skanavi[7] also has discussed the complex conductivity representation, but his treatment is limited to the case of Debye relaxation[8] with a single relaxation time. Grant analyzed data which show a continuous distribution of relaxation times. In particular, he has treated a datum of Koops[3] on theNi-Znferrite dielectric dispersion, Grant's analytical diagram is reproduced in Fig. 1. The complex conductivity diagram suggests strongly the so-called Cole-Cole arc. Of course, the Cole-Cole arc represents a specific dispersion function, and is not to be confused with the Cole-Cole diagram, except as an interesting coincidence: the Cole-Cole dispersion function has a very simple geometrical form in the Cole-Cole diagram; an arc of a circle with the center depressed an angle of $\pi/2\alpha$ from the ϵ'-axis. Since α is the only adjustable parameter, and the arc can be rigorously and readily drawn, a test to find whether the relaxation data fit the Cole-Cole arc is particularly simple. This leaves lesser ambiguity than to fit the data by two simultaneously adjustable parameters, such as the dispersion function of Krotzsch.[1]

The most important observation is that the dispersion function of the ferrite under discussion is not represented by the Cole-Cole dispersion function of complex permittivity, but by a Cole-Cole type of dispersion function of complex conductivity. Mathematically, we mean instead of the ordinary Cole-Cole function,[6]

$$\epsilon^* = \epsilon_\infty + (\epsilon_0 - \epsilon_\infty)\left[1 + (i\omega\tau)^{1-\alpha}\right]^{-1}, \tag{2}$$

the result of Grant implies that the dispersion function should be,

$$\sigma^* = \sigma_0 + (\sigma_\infty - \sigma_0)\left[1 + (i\omega\tau)^{1-\alpha'}\right]^{-1}, \tag{3}$$

By using Eq. (1), one can show that functions given by Eq. (2) and (3) are equal if and only if $\alpha = \alpha' = 0$, that is the case of Debye dispersion. Figs. 2 and 3 show the two representations of the function given by Eq. (2). By interchanging the symbols σ and ϵ, and simultaneously changing the sense of the increase of ω, those figures also represent the function given by Eq. (3). We want to point out a divergence of σ' and σ'' for Eq. (2) at infinity frequency, and of ϵ' and ϵ'' at zero frequency. The difference between Figs. 2 and 3 is contributed by the different value of α; Fig. 2 is for $\alpha < 0.21$ and Fig. 3 for $\alpha > 0.21$, that is, only when $\alpha < 0.21$, σ'' of corresponding Eq. (2) will show a maximum.

In the case of Fig. 1, we found a value of 0.13 for α' in the complex conductivity representation; therefore, according to the above argument, a maximum should be observed in the complex permittivity representation. The contradicting result with respect to Fig. 1 is due to the fact a d.c. conductive effect has not been subtracted. In fact, after we subtract the respective d.c. conductive and a.c. capacitive effect from Koops data, we obtain the result of Fig. 4. Here the close fitting of the data points with the curves computed from Eq. (3) strongly support the new dispersion function given by Eq. (3).

III. TEST OF MODEL

Although there are three or four models referred to in Ref. 1 which show some difference in mathematical forms, the original model of Koops[3] can be discarded because it fails to produce the feature of a broadening relaxation in the absorption peak – broader then that of the Debye dispersion.[8] The mathematical analytic aspect of other models can be included in the generalized model of Krotzsch,[1] and, therefore, we will limit our discussion to the dispersion functions of Krotzsch and of Eq. (3).

It is known[9] in general, one can adjust the two parameters, b and c of Krotzsch such that the curve computed from his formula can be made very close to that of Eq. (3), without significant difference, especially in the region of frequency where the dispersion quantities undergo a rapid change. But actually, from analytical point of view, it is more important to know the manner for these quantities to approach the zero frequency and the infinite frequency. Since the paper of Krotzsch is very recent, and mathematical procedure to establish the analytic properties are very simple, we will not repeat the equations of Krotzsch and our derivations. Table 1 lists the qualitative comparison. As frequency spans from 0 to ∞, we listed some asymptotic properties. f stands for finite, inc for increasing, dec for decreasing, M for an existence of a maximum. Since the maximum should occur at a finite frequency, a close examination of σ'' would be most fruitful to test the validity of models.

Another critical region to be examined is the low frequency behavior of ϵ' and ϵ''. We note that specimen 2 of Ref. 1, and several data of Kamiyoshi[4] shows that the dielectric constant and loss at low frequency does not seem to have a saturated value. In Ref. 1, a work of Habel is referred where a more complicated conductivity state is used as an interpretation. It might be interesting to investigate Eq. (3) and the resulting Fig. 2 and 3, before introducing a new mechanism.

Although there are always experimental difficulties and reproducibilities at extreme frequencies, perhaps more extensive measurement at moderately high temperature is interesting. That is based on the consideration of the rapid decrease of the relaxation time at high temperature, therefore, "d.c. values" of dispersion quantities can be obtained at conveniently attainable frequencies. In this respect, however, complicated problem of electron diffusion as envisaged by Norwick[10] must be taken into account.

In conclusion, it seems rather important to examine completely the mathematical analysis before elucidating and proving the validity of a serious physical model based on a comparison between the experimental data and mathematical numbers at some finite temperature or frequency region.

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TABLE I

Dispersion	σ'	σ''	ϵ'	ϵ''
Frequencies	0 ∞	0 ∞	0 ∞	0 ∞
Krotzsch	0 inc f	0 inc ∞	f dec 0	0 M 0
Eq. (3), $\alpha' < .21$	0 inc f	0 M 0	∞ dec 0	∞ M 0
Eq. (3), $\alpha' > .21$	0 inc f	0 M 0	∞ dec 0	∞ dec 0

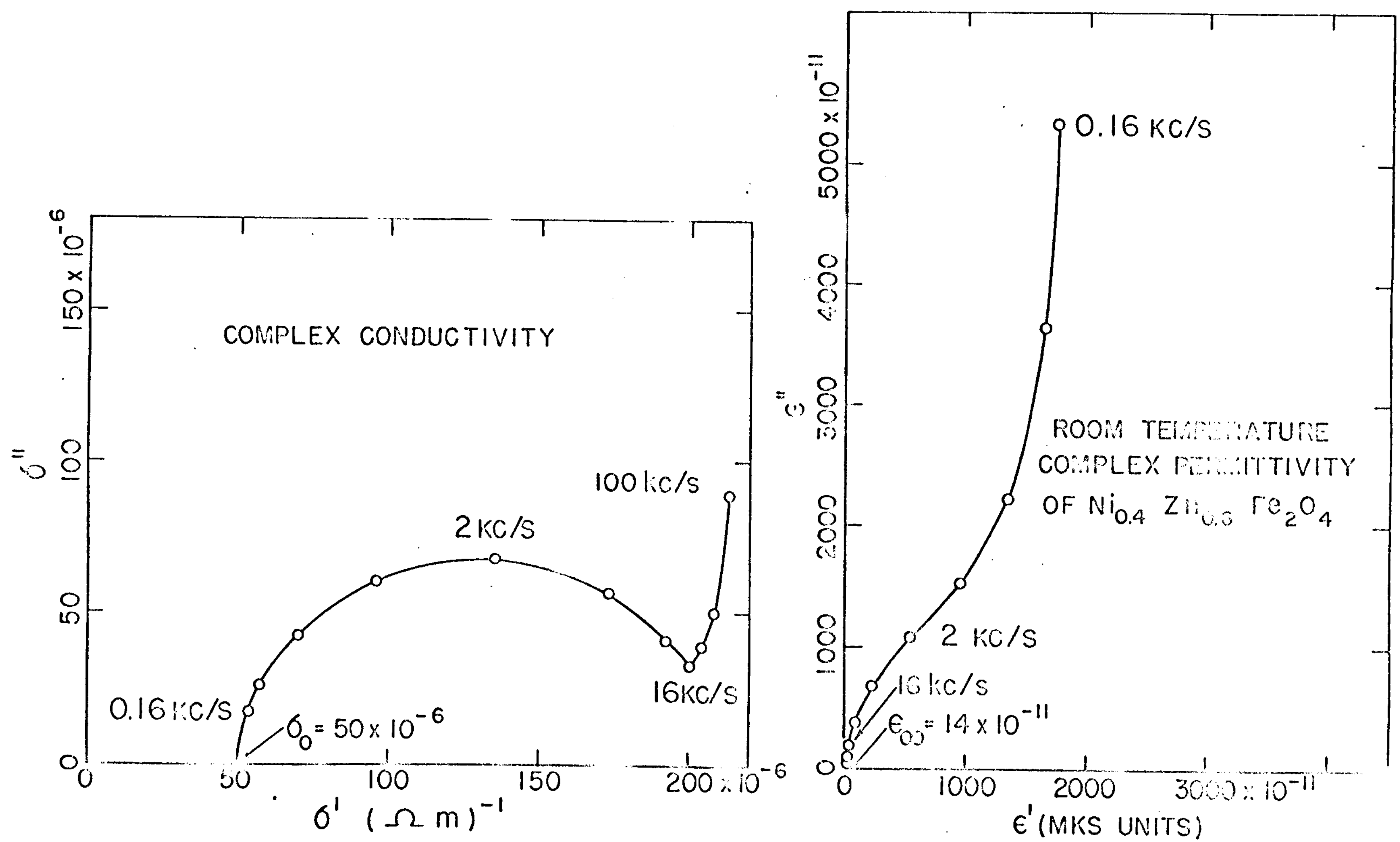
LIST OF FIGURES

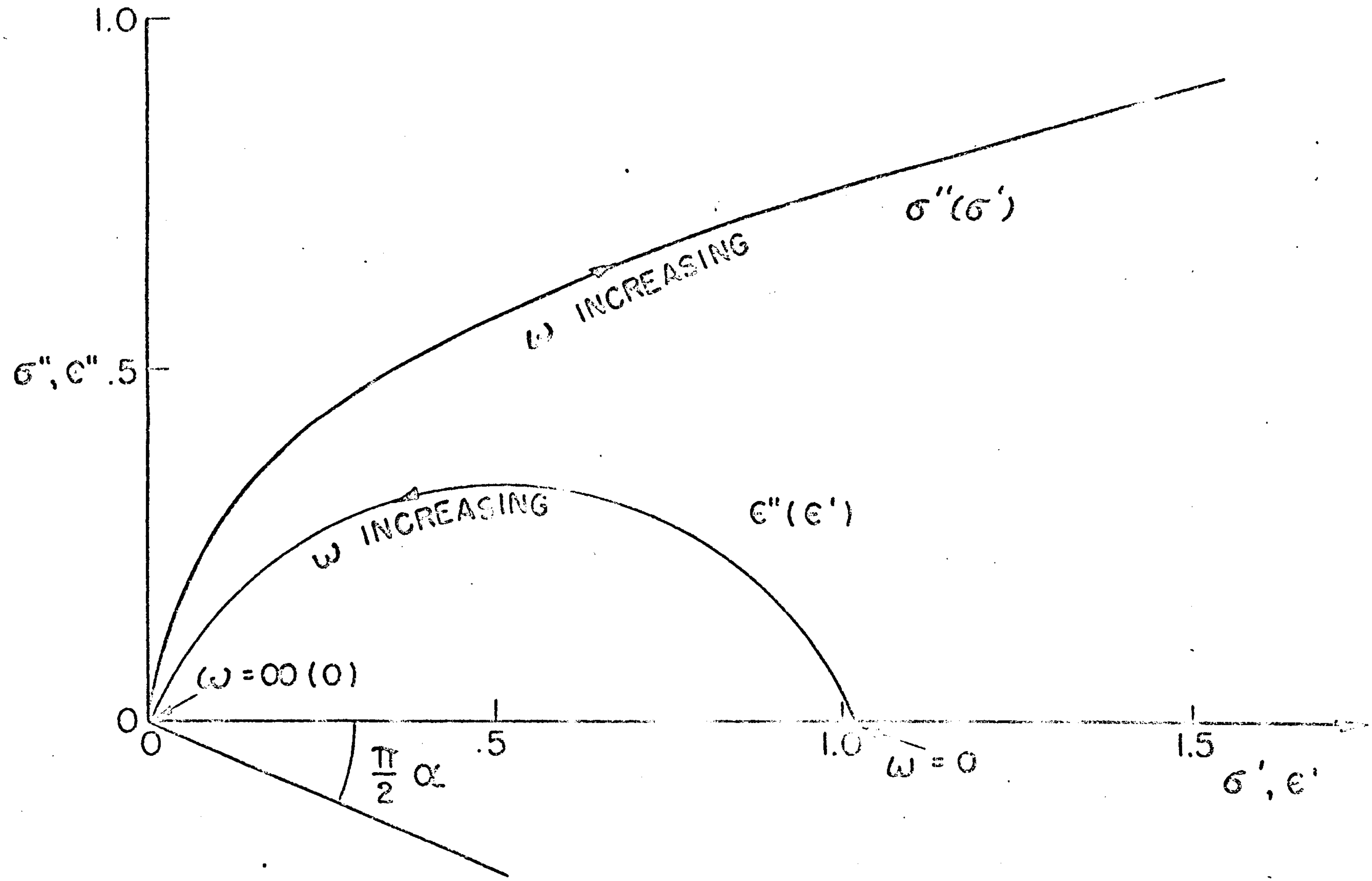
Figure 1—Argand diagram of Ni-Zn ferrite dielectric dispersion from Koop's datum, (a) complex conductivity representation, (b) complex permittivity representation (after Grant).

Figure 2—Complex permittivity and complex conductivity representations of Cole-Cole function, Eq. (2), for $\alpha < 0.21$.

Figure 3—Complex permittivity and complex conductivity representations of Cole-Cole function, Eq. (2), for $\alpha > 0.21$.

Figure 4—Complex permittivity and complex conductivity representation of the data of Fig. 1. Experimental points are based on Koop's datum. Curves are computed from Eq. (3).


COMPLEX CONDUCTIVITY
σ''
150 x 10^-6
100
50
0
0
50
100
150
200 x 10^-6
σ' (Ω m)^-1
0.16 KC/S
σ0 = 50 x 10^-6
2 KC/S
100 kc/s
16KC/S
ROOM TEMPERATURE
COMPLEX PERMITTIVITY
OF Ni0.4 Zn0.6 Fe2O4
ε''
5000 x 10^-11
4000
3000
2000
1000
0
0
1000
2000
3000 x 10^-11
ε' (MKS UNITS)
0.16 KC/S
2 KC/S
16 kc/s
ε∞ = 14 x 10^-11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.5
1.0
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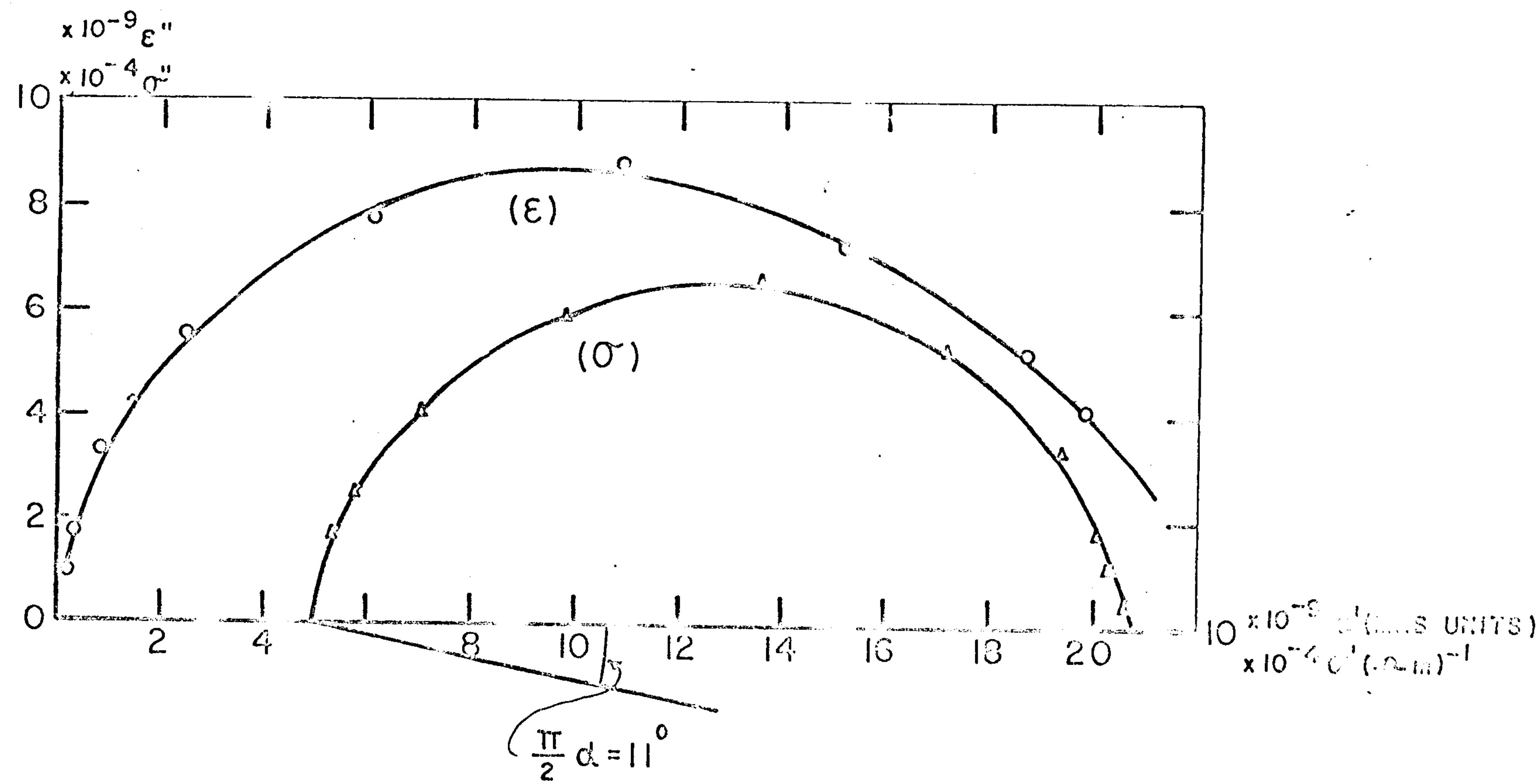
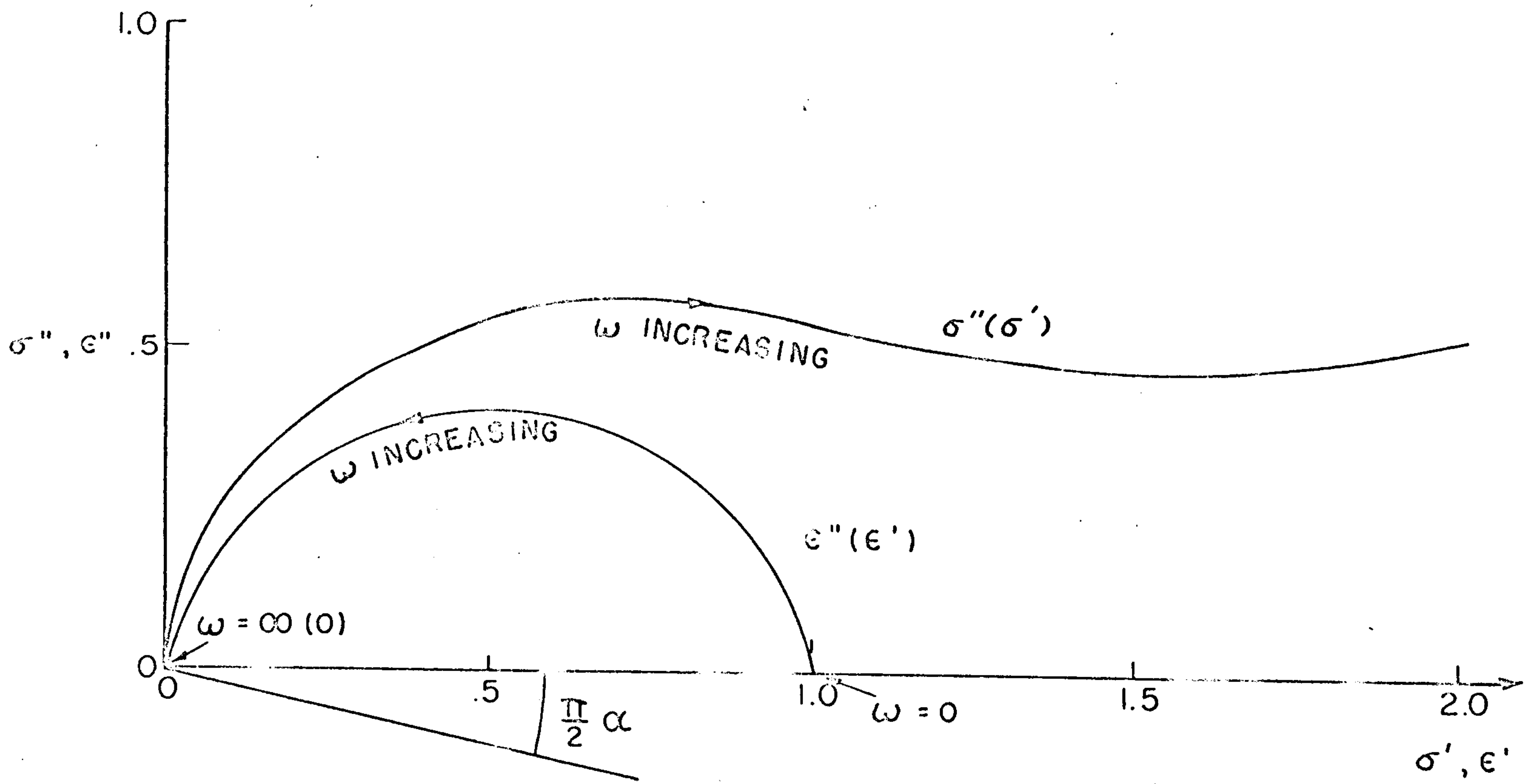
σ"(σ')
ω INCREASING
ϵ"(ϵ')
ω = ∞ (0)
ω = 0
π/2 α



x 10^-9 ε''
x 10^-4 σ''
10
8
6
4
2
0
2
4
8
10
12
14
16
18
20
10
(ε)
(σ)
π/2 α = 11°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σ", ε" .5
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σ', ε'
ω = ∞ (0)
ω = 0
ω INCREASING
ω INCREASING
σ"(σ')
ε"(ε')
π/2 α