ELECTRICAL NON-LINEAR PARAMETERS OF ROCKS

Pre-Print (Short Version)

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ABSTRACT

As the I.P. added a second dimension to resistivity mining exploration surveys by allowing finding of disseminated sulphides, measurement of non-linear electrical properties may add a third dimension to such surveys by allowing a direct diagnosis of the minerals causing the anomalies. So it is important to simplify and to standardize the concepts of non-linearity in order to develop instrumentation and to experiment on non-linear behavior of rocks.

The following mathematical operation defines the first order N^{th} harmonic small current non-linear parameter of a specific rock sample at a given frequency, $Z_{1N}(f)$: "The N^{th} harmonic of an A.C. electric field observed through the material divided by the N^{th} power of the A.C. amplitude of the excitation current density. This ratio is measured at the limit where the current amplitude tends to zero". Such value is a complex number and is nearly independent of the current density, as long as this density is small.

Experimental studies show that Z_{11}, Z_{12} and Z_{13} are easily measurable at current density as low as 0.1 to 1 uA/cm^2 at 1 Hz on suitable saturated polarized rock samples and on outcrops.

INTRODUCTION

Studies on the non-linear behavior of electrical conductivity of various rocks and minerals have been made for the last ten years; among others by Anderson and Keller (1964), Scott and West (1969), Katsube, Ahrens and Collett (1973), Bertin (1968) and Shaub et Al. (1974) (71a) (71b). The experimental conditions were, in general, so that a strong non-linear behavior was observed when using large current density. The results were so complex that only a few generalized conclusions could be made, mostly based only on experimental observations.

By combining mathematics, curcuit theory, electrochemical theory and experimental proof, it was possible for the first time, to step further into a more generalized theory of non-linear conduction laws of materials, as it will be shown hereinafter.

THEORY

First, we will define the non-linear conductivity of rocks. We can say that this material does not obey Ohm's Law, saying that the electrical field in the sample is linearly proportional to the current density. For a pure cosine periodic excitation, say a current density of $J(t) = J_o \cos \omega t$, we observe as electrical field a "distorted" signal, this means a signal with harmonics present. This is described by the Fourier series as:

(1) $E = E_o + E_1 \cos(\omega t + \theta_1) + E_2 \cos(2\omega t + \theta_2) + \ldots,$

where the electric field amplitude E_o, E_1, E_2, E_3 ..., and the phase shifts θ_1, θ_2, θ_3 ... are funtions of the current density amplitude J_o as well as of the frequency ω.

Results are generally shown in Lissajeous figures for a specific frequency ω, as shown in Figure 1. Curve 1 represents the observed signal, curve 2 the linear (or first harmonic) component of the signal, and curve 3 represents its distortion and is the voltage difference between curve 1 and curve 2. This non-linear component may be divided into its Fourier component, in our example curves 4 and 5. These latter represent respectively a 2nd harmonic sine signal and a 3rd harmonic cosine signal.

Using Fourier series combined with Taylor series, it is possible to show, for different conduction mechanisms and circuit networks, the following generalized conclusion:

Let $J = J_o \cos \omega t$ be the electrical current or current density of the excitation. If J_o is small and if there is no discontinuity given in any part of the system in its voltage to current relation (V-I), then the voltage or the electric field may be represented by

$$
\begin{aligned}
(2) \qquad E \simeq\ & E_o{}^{*} \\
& + Z_1 J_o \cos(\omega t + \theta_1) \\
& + Z_2 J_o{}^2 \cos(2\omega t + \theta_2) \\
& + Z_3 J_o{}^3 \cos(3\omega t + \theta_3) \\
& + \ldots
\end{aligned}
$$

where $Z_1, Z_2, Z_3, \ldots$ and $\theta_1, \theta_2, \theta_3 \ldots$ are parameters independent of the current or current density.

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[*] E_o represents the natural S.P. value; its variations by non-linear rectifications is considered to be small with respect to the natural S.P. value

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In this pre-print, no theoretical proofs are developed, but only experimental proofs are presented. The relations (2) apply on all electrical circuit networks, where one or many components are "continuously" non-linear, and on all known electrochemical overvoltage mechanisms.

Technical details of non-linear calculations for circuit networks are clearly shown by D.G. Tucker (1964).

Now, we will illustrate the relation (2). Figure 2 is based on Figure 1 and represents the observations expected for two different current excitations. In Figure 2, below, the non-linear component is enlarged five (5) times, in order to be better able to recognize it. Curve 3 is the non-linear component of curve 1, and curve 4 is the non-linear component of curve 2, the latter representing the observed signal with only half of the preceding excitation current amplitude. Two main observations can be made: a) At a smaller current, the non-linear component has a different shape, its third harmonic component being smaller than the second one; b) The non-linear component has decreased at a faster rate than the current excitation amplitude.

EXPERIMENTS

Experimental proof has been made by using the circuit shown in Figure 3. Design and component have been chosen very critically, so as to obtain an as high as possible linear circuit; non-linear effects caused by the instrument were measurable down to ± 5 ppm. This design was chosen in order to avoid non-linear effects due to electrodes at their contact on the sample. In fact, if the samples were replaced by salted water, no parasitic non-linear effects were observed as long as the potential between

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$C_1 - C_2$ of Figure 3 was below 12 volts. It was possible to choose a current amplitude J_o of between 0 and 10 μA.

Some measurements were made outside, in the field, more precisely on rock outcrops. A Wenner configuration was used, with one foot separation, as illustrated in Figure 4. Such a small scale was necessary because of the actual instrument limitations. A 100 foot Wenner electrode configuration would require a 10 kW power transmitter, well controlled down to ± 1 ppm.

Experimental proof of the equation (2) has been made 1° on electronic circuits; 2° on electrochemical systems and 3° on rock samples. On figures 5 and 6 it can be seen that below a maximum current value, the amplitude of harmonics and their respective phase shifts behave as predicted in equation (2). Suitable current density lies usually below 1 $\mu A/cm^2$ for rock samples at 0.1 Hz.

In Figure 5 we observe that the second harmonic signal is proportional to the square of the excitation current, and the third harmonic signal proportional to the cube of the excitation current. Experimental points are here near the theoretical line.

On figure 6 we see that the observed second and third harmonic phase shifts Θ_2 and Θ_3 are nearly independent of the current density, as predicted.

Frequency effects of non-linear parameters can also be predicted by using the method described by Tucker (1964). Interesting conclusions can thus be obtained.

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This preprint shows a simple example, i.e. the non-linear behavior of a parallel R-C circuit, where the resistive component is non-linear, the circuit being excited by a current source. The dependence of Z_1, Z_2 and Z_3 on the frequency, as well as θ_1 vs ω, is illustrated in Figure 7. Figure 8 shows the relation of θ_3 and θ_2, compared to θ_1. Such an R-C circuit represents very roughly a metal-eletrolyte interface as found in some mineral bearing rocks. The most important observations are:

1° Z_3 and Z_2 vary much faster than Z_1

2° θ_3 and θ_2 vary much faster than θ_1

3° Z_2 and Z_3 increase when the frequency decreases down to a critical frequency ω_0

Similar but not identical conclusions are obtained on electrochemical systems (see example in Appendix).

An application of the genralized equation (2) is shown in the spectrum of Figure 9. A non-linear measurement has been made on a disseminated bornite outcrop. The voltages E_2 and E_3 (see equation (1)), where the current amplitude J_o is 1 milliampere, are shown with plain lines. When the value J_o is halved, E_2 and E_3 will behave as illustrated by the dotted lines, but the phase shift of θ will remain unchanged. It is important to note that the ratio E_3/E_2 is halved if the excitation current is halved.

CONCLUSION

It was shown that at a small current density, non-linear effects on geological materials (as well as on others) become simpler. When a non-linear spectrum is made at a certain small current density, this spectrum is predictable for any other small enough current density.

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It is the opinion of the author that such definitions will be most easily applicable for the purpose of developing non-linear geophysical exploration techniques. Chances for getting more information on rock chemistry by non-linear electrical means are better, since more than the two usual I.P. parameters (resistivity and chargeability or phase shift) are measurable by non-linear exploration techniques. In this paper, six (6) parameters have been considered.

ACKNOWLEDEGEMENTS

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The encouragement and advices of Dr Alex Becker, my thesis adviser, were of fundamental help for the realization of this paper.

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APPENDIX

It has been shown by Marshall and Madden (1959) that the electrochemical Warbourg impedance is an important mechanism in I.P. effects, observed on some rocks. This impedance is well described on an electrochemical basis by Vetter (1967).

It was possible to use the non-linear analysis and the theory of a pure Warbourg overvoltage* to predict the frequency effect on non-linear parameters Z_1 (ω), Z_2 (ω), Z_3 (ω) ... and $\theta_1(\omega)$, θ_2 (ω), θ_3 (ω) ... of this electrochemical interface. This is shown in Figure 10. It is well-known that the Warbourg impedance Z_1 varies as $\omega^{-\frac{1}{2}}$, and its phase shift θ is independant of frequency with a value of 45°.

On Figure 10 we see that the phase shifts θ_1, θ_2 and θ_3 are independent of frequency and take place at 45°, 90° and 135°, respectively. But the values of Z_1, Z_2 and Z_3 vary as $\omega^{-\frac{1}{2}}$, ω^{-1} and $\omega^{-3/2}$, respectively. Non-linear effects become larger as ω gets smaller.

Katsube, Ahrens and Collett (1973) have also observed that the non-linear effect on their samples become larger as the frequency ω gets smaller.

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* A pure Warbourg overvoltage effect is practically observed on any system, where this overvoltage mechanism is the only one controlling its own and other's rate for the frequency range used for the above pure Warbourg effect, and when no convection of active ions is taking place (Vetter, 1967).

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DESCRIPTION OF FIGURES

Figure No 1

Non-linear behavior observation expressed in Lissageous figure form, curve 1. This observation may be analysed by Fourier's method; the components are:

Curve 2: first harmonic or linear component
Curve 3: higher harmonics or non-linear component
Curve 4: second harmonic, shown here in quadrature
Curve 5: third harmonic, shown here in phase

Curve 3 is the sum of curves 4 and 5.

Figure No 2

Curve 1 is the same as curve 1 in Figure 1
Curve 3 is the same as curve 3 in Figure 1, but enlarged 5 times
Curve 2 is the predicted Lissageous figure with smaller current excitation (here it is halved)
Curve 4 is the predicted non-linear component of curve 2, but also enlarged five times.

Figure No 3

Block diagram of the non-linear analyses.

Figure No 4

Electrode arrangement for non-linear measurements on rock outcrop.

DESCRIPTION OF FIGURES (cont'd)

Figure No 5

Normalized relation between the current excitation amplitude J_o and the harmonic components of the observed signal;

fundamental E_1 = curve 1
second harmonic E_2 = curve 2
third harmonic E_3 = curve 3

Plain curves are theoretical curves; triangles, dots and ellipses are experimental observations.

Figure No 6

Dependence of the observed signal harmonic's component phase shifts on the normalized excitation current amplitude J_o.

Figure No 7

Normalized values of the conduction parameters Z_1, Z_2 and Z_3 with varying frequency ω.
Plain curves are from non-linear circuit network theory squares; circles and triangles are experimental observations.

Figure No 8

Relations between the fundamental component phase shift θ_1 and their higher harmonics counterparts, θ_2 and θ_3.
Plain curves come from non-linear circuit network theory; dots and triangles are experimental observations.

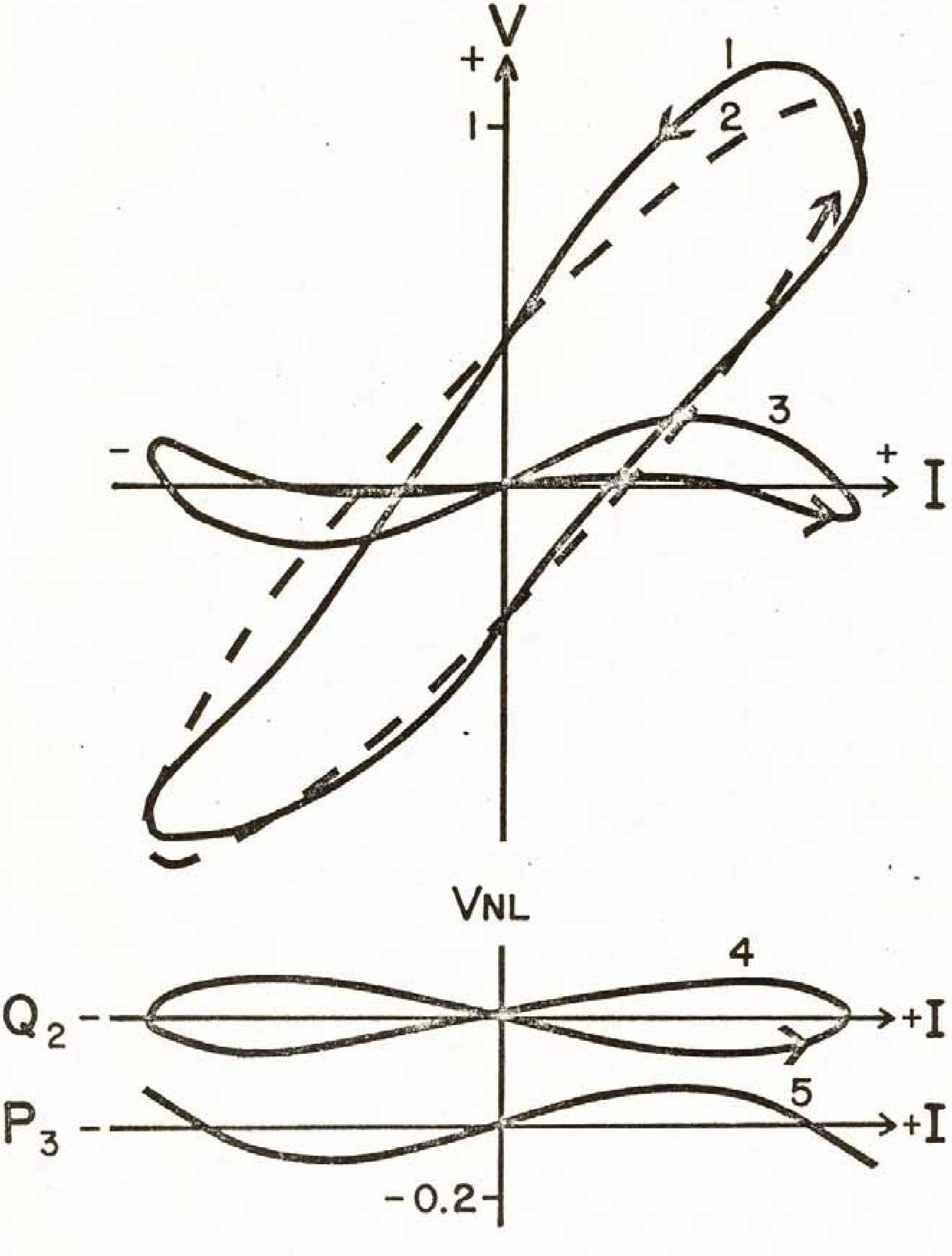
DESCRIPTION OF FIGURES (cont'd)

Figure No 9

Field results on disseminated bornite outcrop; plain lines are field results; dotted lines represent results which can be predicted, if a smaller current was used.

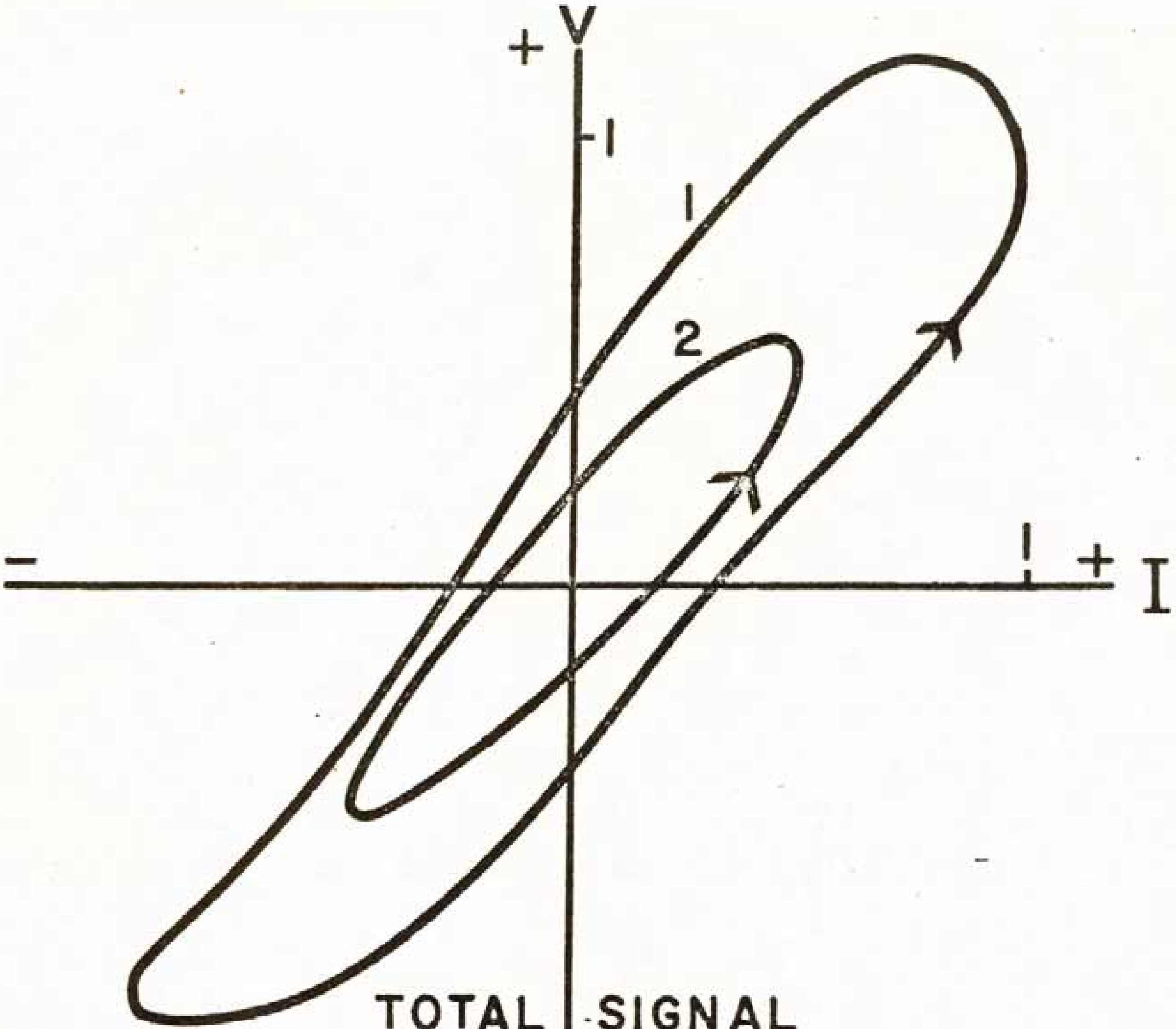
Figure No 10

Normalized non-linear overvoltage parameters of a pure Warbourg impedance.



ELECTRICAL AC VOLTAGE VS CURRENT RELATION
AT SMALL CURRENT LIMIT

FIG I



TOTAL SIGNAL

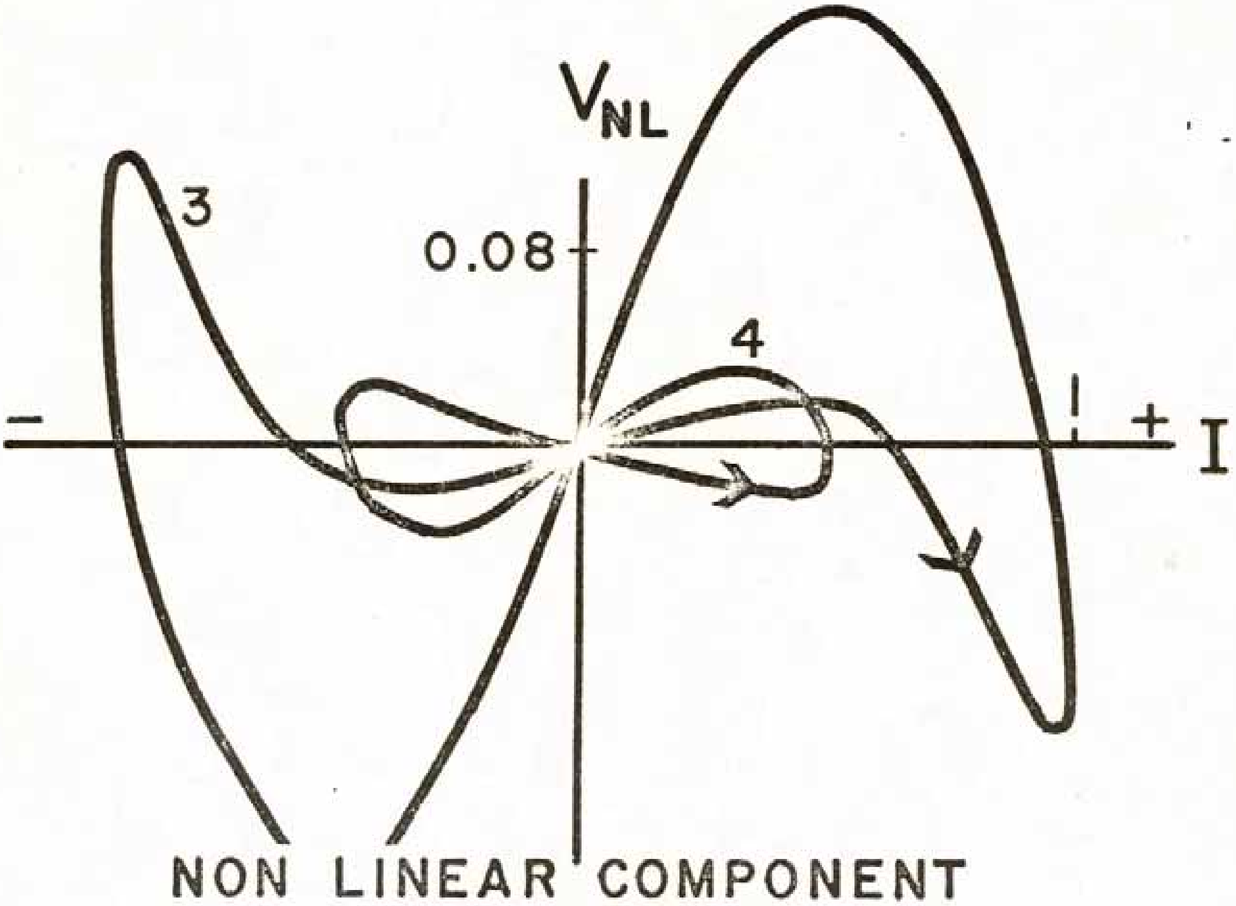


FIG. 2 NON LINEAR COMPONENT

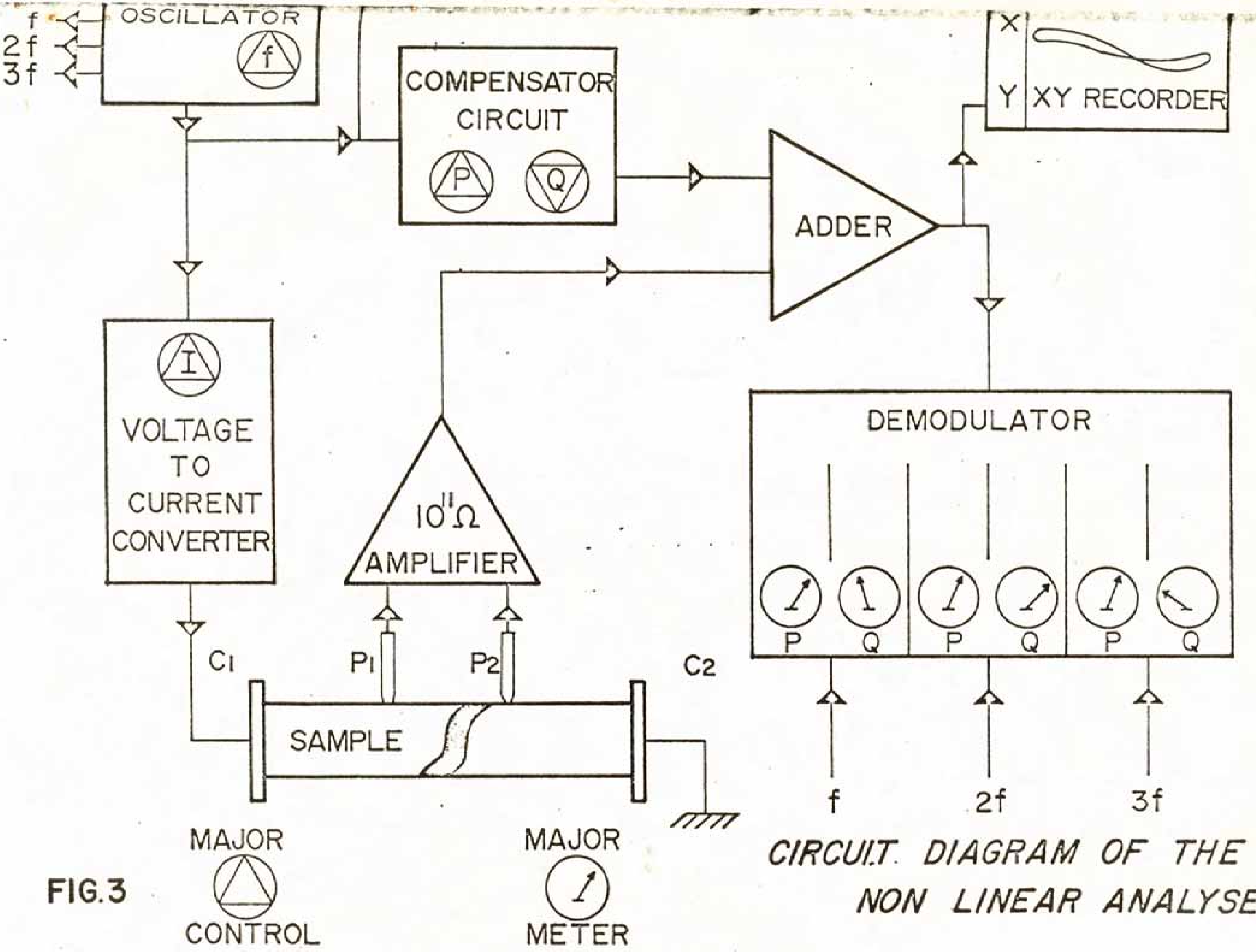
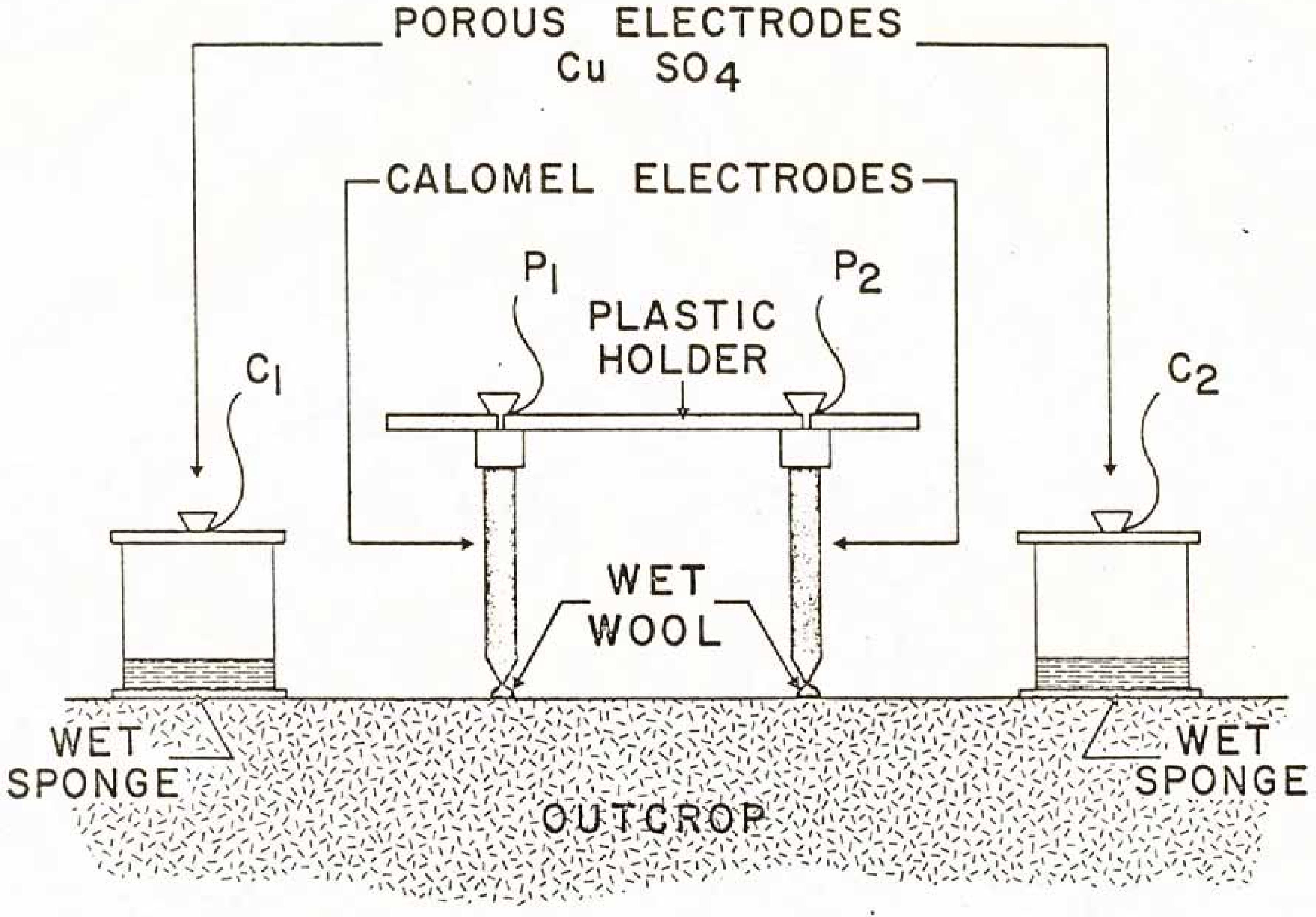


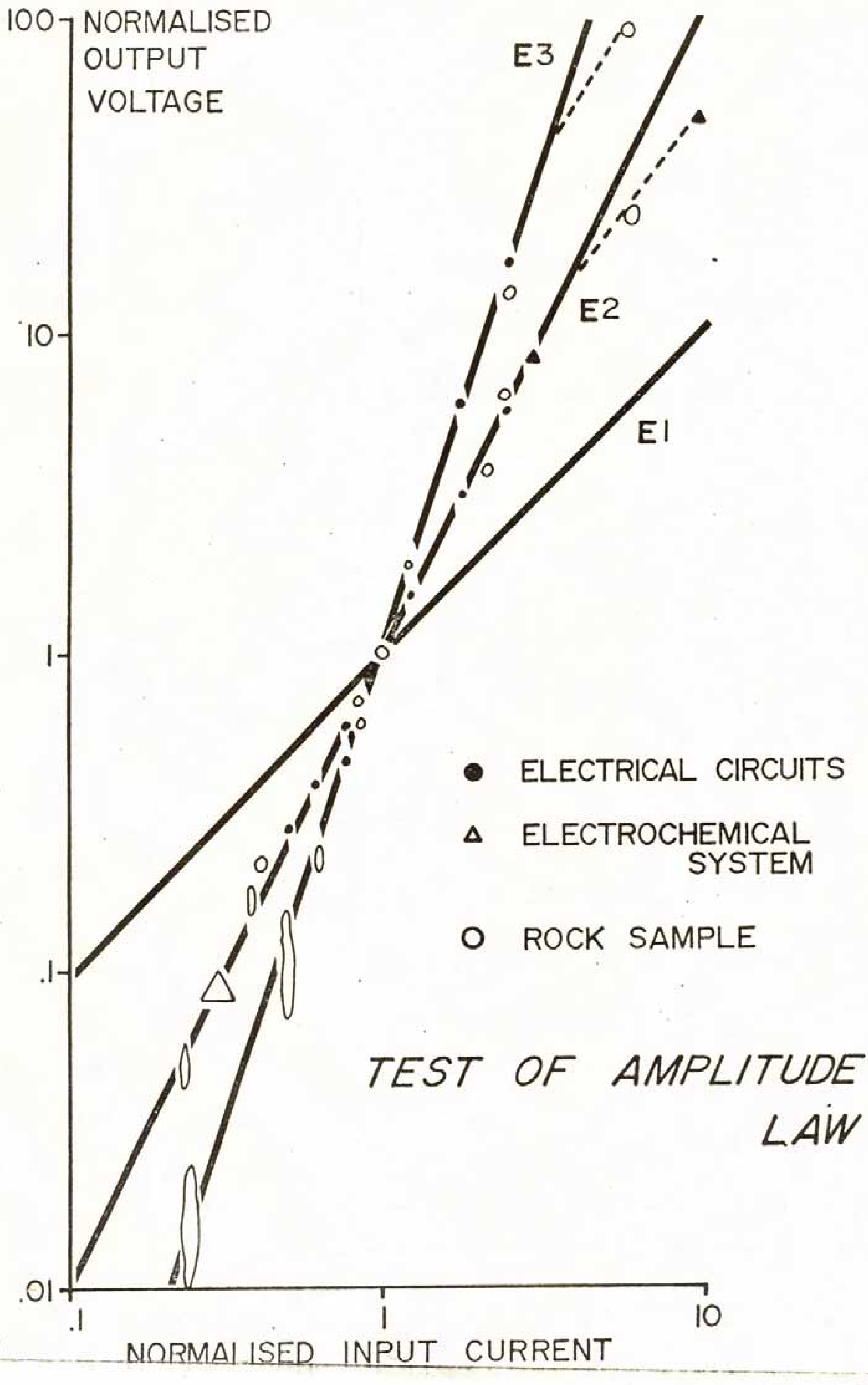
FIG.3

CIRCUIT. DIAGRAM OF THE NON LINEAR ANALYSE



ELECTRODES' CONFIGURATION FOR OUTCROP FIELD TEST

FIG. 4



TEST OF AMPLITUDE LAW

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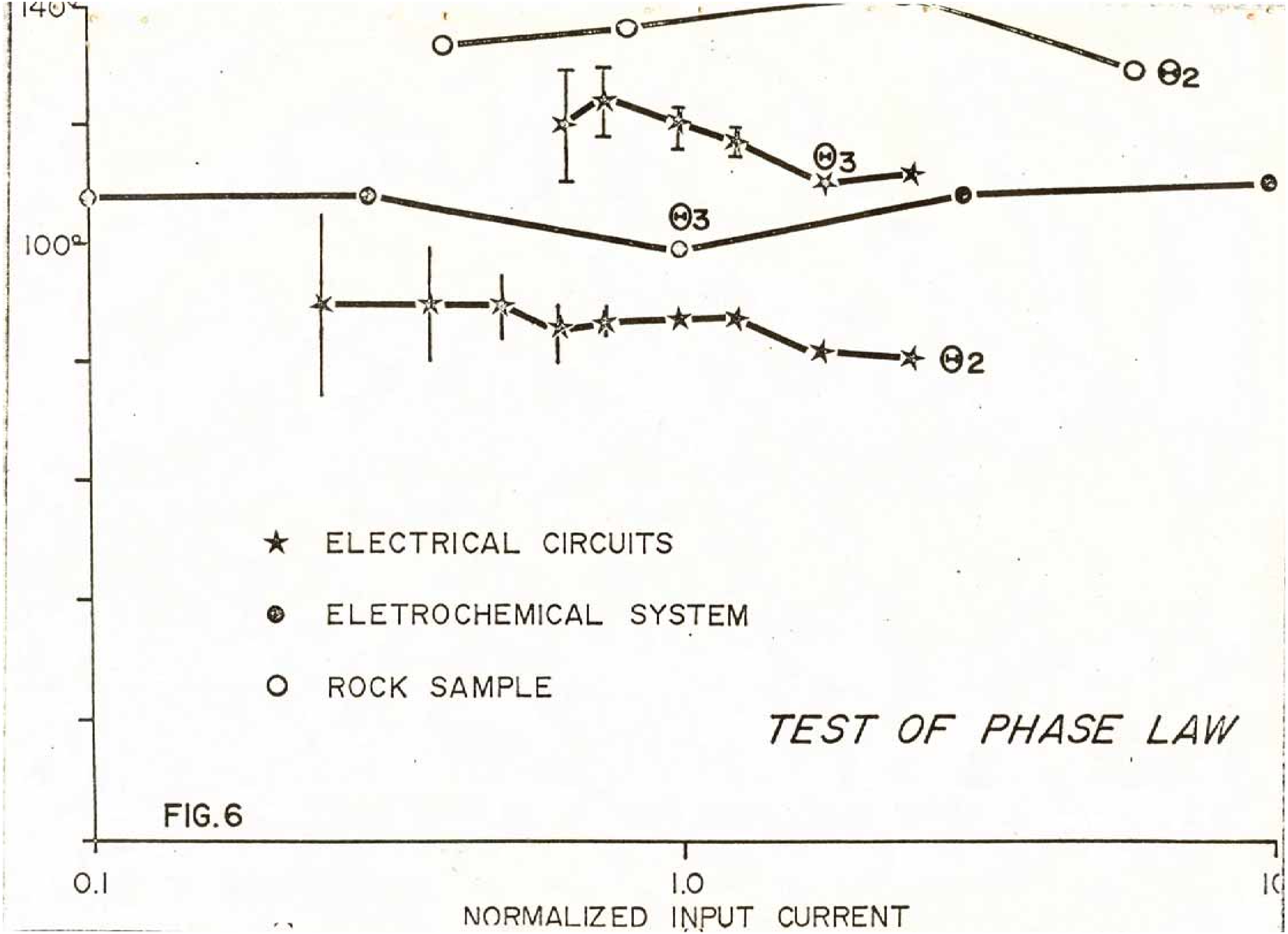
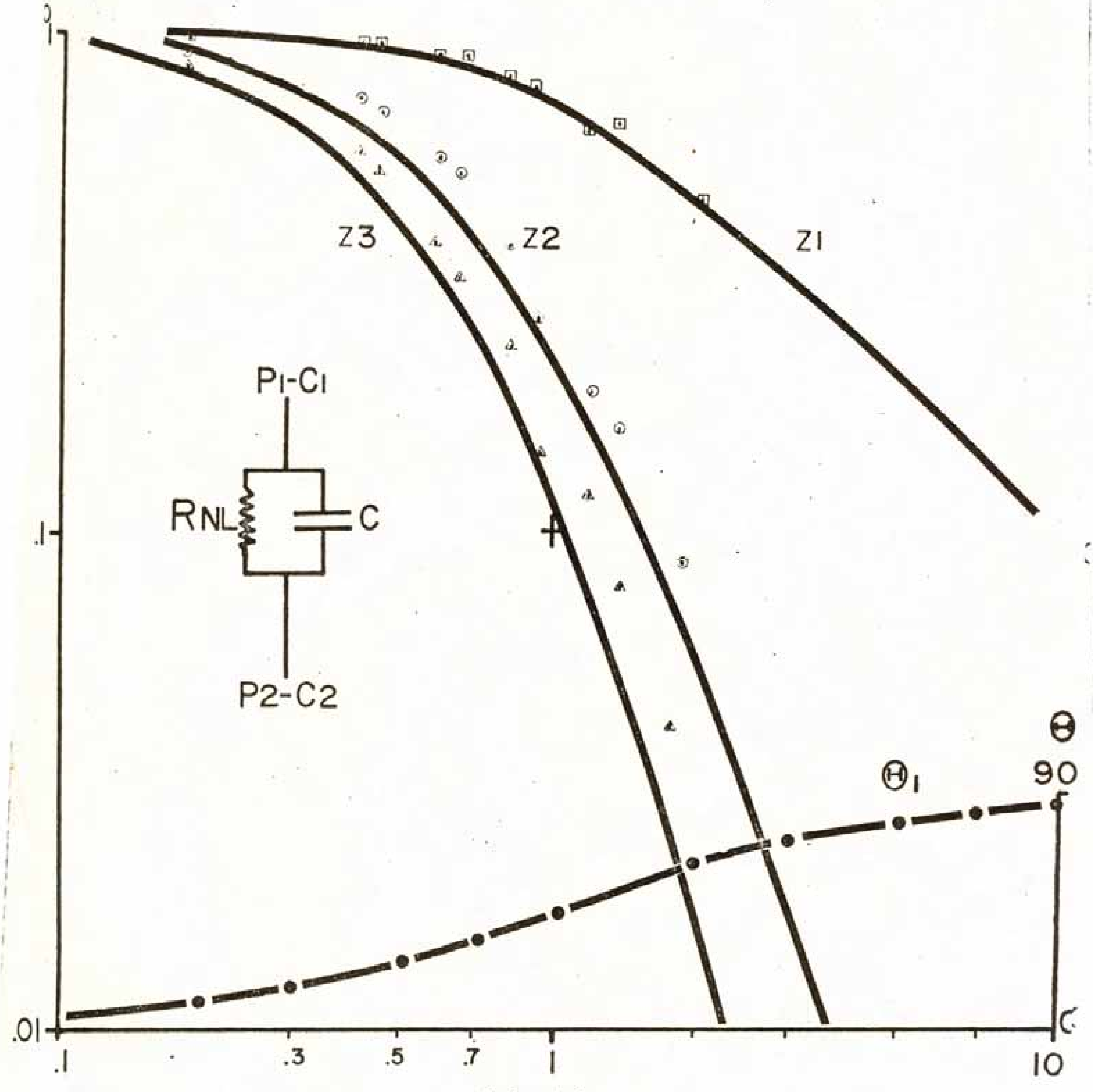


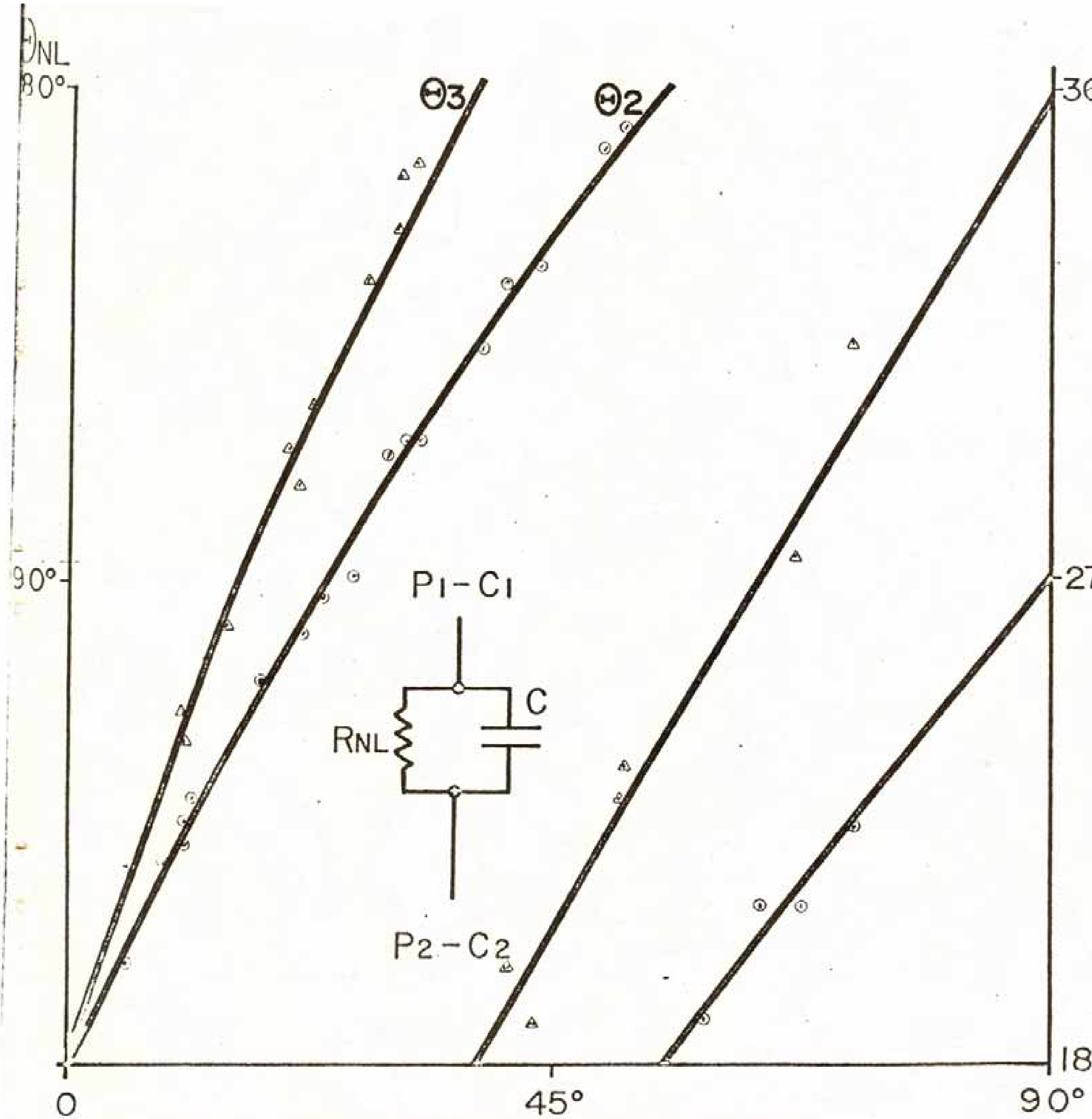
TEST OF PHASE LAW

FIG.6



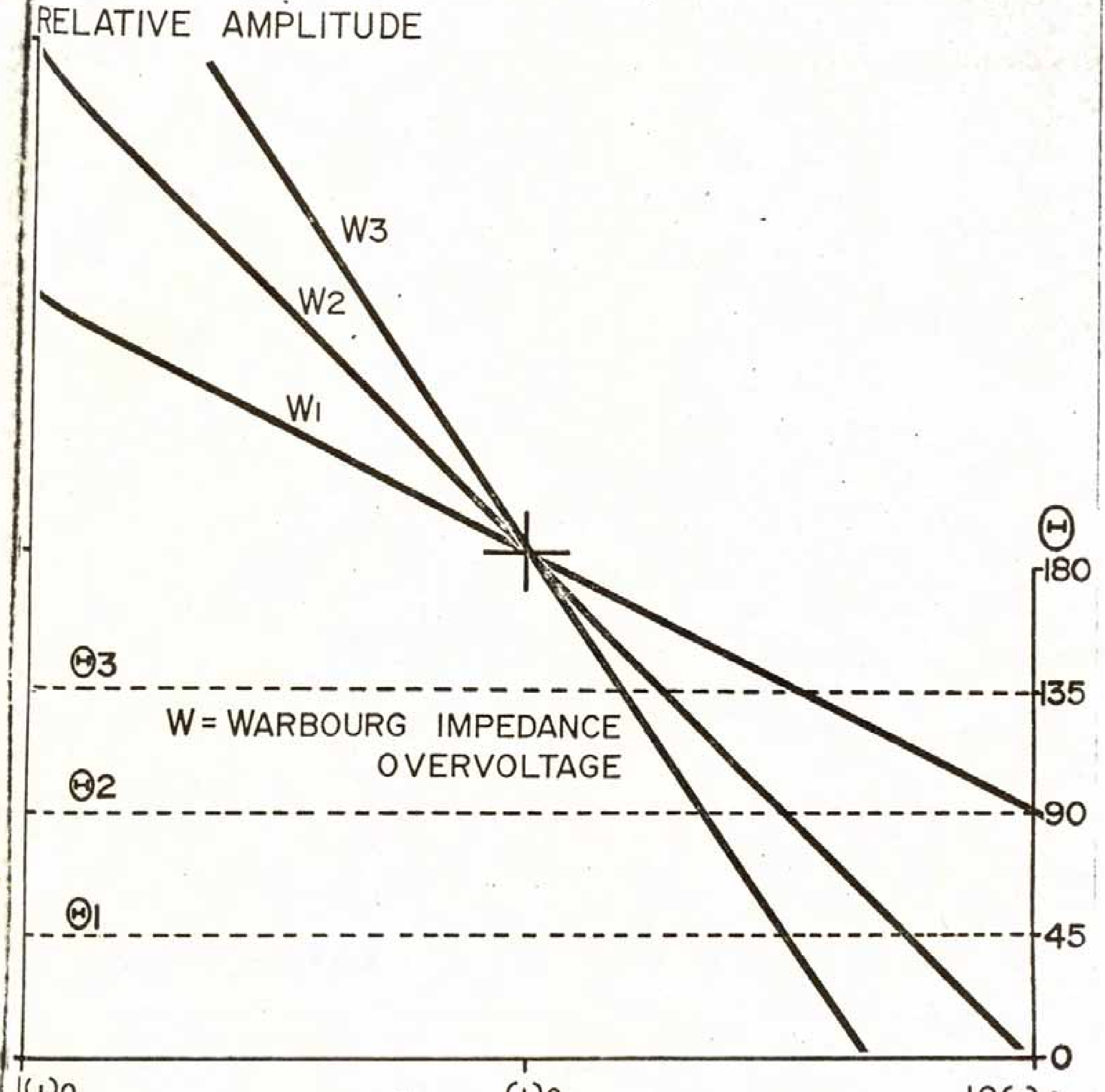
RELATIVE AMPLITUDE OF HARMONIC TO THEIR VALUE AT ω ⟶ 0

FIG 7 *FOR NON LINEAR RC CIRCUIT*



RELATION BETWEEN PHASE OF
FONDAMENTAL vs PHASE OF HARMONIC
FOR NON LINEAR RC CIRCUIT

FIG. 8



FREQUENCY EFFECT ON ELECTROCHEMICAL SYSTEM

FIG. 10