THEORETICAL ADMITTANCE VARIATION WITH FREQUENCY IN INSULATORS HAVING TRAPS SUBJECT TO CHARGE INJECTION*

by

Richard S. Muller

Electrical Engineering
University of California
Berkeley, California

ABSTRACT

The admittance function for an insulating crystal with monoenergetic traps subject to space-charge-limited electron injection is considered. Frequency limitations imposed by the trap-emptying sequence are shown to result in frequency-dependent conductance and capacitive susceptance values. An explicit solution for the admittance variation is not possible, but the results obtained from a computer solution to the representative equations are given. Two distinct physical situations are shown to apply, depending on the frequency range investigated.

Application of the theoretical analysis presented here should permit the use of comparatively easy and accurate measuring techniques (ac admittance bridge measurements) to obtain the attempt-to-escape frequency for monoenergetic trapping centers. Observation of the admittance variation derived here would confirm the trap-influenced, space-charge-limited current theory of Rose.

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INTRODUCTION

The theory of the effects of volume-distributed trapping centers upon the electronic properties of single-crystal insulators, as proposed by Rose,[1] has been successful in explaining much experimental data.[2-3] Some authors have, however, disagreed over the applicability of the physical model employed by Rose,[4] and others have had only partial success in using the theory to explain their measurements.[5]

Basically, the theory proposed by Rose states that trapping centers distributed uniformly in the insulator bulk may exist in various energetic configurations, both discrete and distributed. As a consequency, the volt-ampere behavior may vary widely among crystals of the same material, dependent upon the position of the interior Fermi level with respect to these trapping centers.

Under the assumptions of Rose's theory, ac bridge measurements of the admittance of thin single crystals should show a frequency-sensitive behavior which would be indicative of fundamental properties of the trapping states. This frequency dependence comes about due to the rate limitations of trap-filling and emptying. Actually, the measured admittance of crystals subject to charge injection and having trapping states is a function of three rate processes. First, there is the rate of charge transfer at the contact. Second, there is the transport rate for electrons either from the contact to the trapping site, or to the collecting electrode. Finally, there is the rate of emission and capture at the trapping site.

The first rate process has a counterpart in vacuum-tube theory, emission from the contact leading to the Richardson equation. Likewise, for vacuum tubes, transit time from emitter to collector results in the high-frequency admittance variations given by the Llewellyn-Peterson analysis.[6] The effects of transit time in solids carrying space-charge-limited currents has been considered by Brojdo.[7] The third rate process mentioned above is, however, unique to solids because it concerns the interchange between free and immobile electron states in the space-charge region. For typical insulating crystals subject to charge injection this interchange between the conduction band and the trapping states is very likely to be the lowest limiting rate process, and therefore to be the cause of a detectable ac impedance variation in the crystal. When this occurs, ac bridge measurements can be used to provide information about fundamental properties of the trapping states.

ANALYSIS

The following analysis will calculate the expected variation in admittance due to electron trapping under the assumption that contact and transit-time limitations occur at higher frequencies than does the trapping-rate limitation. Calculations are simplest in the case that a voltage-independent Boltzmann factor, designated θ by most authors, relates free and trapped charge in the crystal at thermal equilibrium. Under this condition, the volt-ampere characteristic, potential profile and current-voltage law are modified from the trap-free case only by the factor θ appearing in either numerator or denominator.[8] The frequent observation of square-law currents in insulators is usually interpreted to imply the existence of this condition, which is postulated for the following analysis. Hence, the contacts are taken to be instantaneously capable of space-charge-limited emission of electrons into the crystal and subsequent instantaneous transfer of these electrons either to the trapping sites or to the collecting electrode is assumed. In addition to a dc bias of magnitude V_o there is postulated an ac driving function of radian frequency ω and magnitude V_m, small enough to ensure continuous charge injection from a given contact. Then, one can write:

$$Q_i = \text{Re}\,[\,C(V_o + V_m\, e^{j\omega t})] \ . \tag{1}$$

In Eq. (1), the real part of Q_i is the injected charge and C is the geometric capacitance. Consideration of the spatial distribution of electrons from the space-charge-limited current analysis[8] shows that in the dc case C is given by $(3/2)\, C_e$ where $C_e = A\epsilon\epsilon_o/a$ is the electrode capacitance. The crystal is assumed to have thickness a, and cross-sectional area A.

If the crystal has a very high resistivity without any charge injection, the average values (over the length of the crystal) for the free- and trapped-charge densities will be the real components of the solutions to:

$$e\,a\,A(\overline{n} + \overline{n}_t) = C(V_o + V_m\, e^{j\omega t}) \,. \tag{2}$$

It is convenient to rewrite Eq. (2) in the form

$$\overline{n} = K(V_o + V_m\, e^{j\omega t}) - \overline{n}_t \tag{3}$$

where $\overline{n}$ and $\overline{n}_t$ denote respectively average values of the free- and trapped-charge densities over the crystal length and $K = C/eaA$.

For trapping centers more than a few kT above the steady-state Fermi level, the rate equation for the trapped-charge density $\overline{n}_t$ is given by the difference between the rates of filling and emptying, and is easily derived[9] to be of the form:

$$\frac{d\overline{n}_t}{dt} = -(\overline{n}_t - \gamma\overline{n})\,\nu_e \tag{4}$$

where $\overline{n}_t$ is the trapped-charge density, $\overline{n}$ is the free-charge density, ν_e is the probability-of-escape frequency for a trapping center, and γ is the equilibrium ratio between trapped and free charge.

If a thermal equilibrium prevails between the conduction-band and trapping states, then γ will be θ^{-1}, the reciprocal of the Boltzmann factor which specifies the ratio of the populations in the conduction band and trapping levels. In most cases of interest in which appreciable space charge is injected into crystals, γ will be much greater than unity.[2]

The probability-of-escape frequency ν_e may be expressed in terms of fundamental parameters, provided that it describes a thermal equilibrium situation for shallow trapping levels. In this case, it can be shown[8] that ν_e is given by:

$$\nu_e = N_c \, v \, S_t \exp\left[-(W_c - W_t)/kT\right] \tag{5}$$

where N_c is the effective density of conduction-band states, v is the electron thermal velocity (10^7 cm/sec at 300°K), S_t is the capture cross-section of an empty trap for a conduction-band electron, and $(W_c - W_t)$ is the trap-depth below the conduction band.

The use of Eq. (3) in Eq. (4) leads to the form:

$$\frac{d\bar{n}_t}{dt} = -[\bar{n}_t - \gamma K(V_o + V_m e^{j\omega t}) + \gamma \bar{n}_t] \, \nu_e \tag{6}$$

Eq. (6) is recognized as describing the charge storage on a capacitor in a simple voltage-source-driven RC circuit, provided $\gamma \nu_e$ is analogous to the circuit time constant, and the voltage source is proportional to $K(V_o + V_m e^{j\omega t})$. Thus, the steady-state solution to the differential equation is familiar. It will be convenient to express this result in terms of complex numbers which will be signified by hats over the variables and accompanied by reference angles. The complex angles denote phase differences from a reference vector as is done conveniently in ac circuit analysis. With the voltage V equal to $\hat{V}_m \angle 0^\circ$ as the reference, and $\gamma \nu_e$ represented by the reciprocal time constant τ^{-1}, the solution to Eq. (5) is the real part of:

$$\bar{n}_t = \frac{\gamma K}{\gamma + 1} \left(V_o + \frac{\hat{V}_m \angle{-\psi_1}}{(1 + \omega^2 \tau^2)^{1/2}} \right) \tag{7}$$

with $\psi_1 = \tan^{-1}(\omega\tau)$ and $\tau = (\gamma \nu_e)^{-1}$. Hence, from Eq. (1), the solution for $\bar{n}$ is:

$$\bar{n} = \frac{K}{\gamma + 1} \left[V_o + \frac{\hat{V}_m}{(1 + \omega^2 \tau^2)} + \frac{\gamma \hat{V}_m \omega \tau \angle{\varphi_1}}{(1 + \omega^2 \tau^2)^{1/2}} \right] \tag{8}$$

where $\varphi_1 = \tan^{-1}(\omega\tau)^{-1}$. At this point, a further physical restriction becomes evident. This restriction is the fact that the free-electron density $\overline{n}$ is constrained to be positive. The result obtained in Eq. (8) is not constrained, since the magnitude of the third term becomes comparable to the magnitude of the second term as ω approaches $\omega_a = (\gamma\,\tau)^{-1}$, and the third term becomes comparable to the first term as ω approaches $\omega_b = (V_o/V_m)(\gamma\,\tau)^{-1}$. An exact expression for the frequency at which $\overline{n}$, as given in Eq. (8), may become negative can be derived. The frequency at which this can occur, and therefore at which Eqs. (7) and (8) cease to be valid will be between ω_a and ω_b, and will approach ω_b as the ratio V_o/V_m becomes large. Hence at frequencies roughly above ω_b, the physical treatment of the problem must be modified to include the constraint of a positive value for $\overline{n}$. Because of this constraint, the trapping and trap-emptying sequence which does take place above ω_b is asymmetric.

The physical result of this asymmetry will be a dc buildup in the value of $\overline{n}_t$. When the net voltage applied to the crystal favors electron injection, the conduction band becomes flooded with electrons, and trap filling takes place at a very rapid rate because thermal equilibrium conditions require that $\overline{n}_t$ greatly exceed $\overline{n}$. When the net voltage applied to the crystal favors charge extraction, the conduction band is immediately emptied and the trap density decays with the characteristic time constant of the trapping sites. Instead of the simple RC equivalent circuit which was the analogue to the low frequency situation, the analogue circuit now becomes the asymmetric capacitor-changing circuit shown in Fig. 1. The time constant R_1C in the circuit of Fig. 1 represents the characteristic trap-filling time, while R_2C in the same circuit represents the trap-emptying time. The trap density $\overline{n}_t$ is analogous to the capacitor voltage in the circuit of Fig. 1.

If γ (the ratio of trapped to free charge at equilibrium) is large, it is apparent from Eq. (2) that trap filling will take place at a much faster rate than will trap emptying ($R_1C << R_2C$ in Fig. 1). A plot of the average trapped-charge density through one cycle of applied voltage under this condition is given in Fig. 2. The trap occupancy is capable of following the applied voltage through a complex angle $\varphi_1 = \omega t_1$, which is given by the expression $\varphi_1 = \tan^{-1}(\omega\tau)^{-1}$.

This result is most easily visualized by considering the complex angle at which the diode opens in the analogue circuit of Fig. 1. The traps then decay exponentially (solution to Eq. (2) with $\bar{n} = 0$) until free-electron injection occurs again at $\omega t_2 = \varphi_2$. The injected free charge after this time is assumed to fill the traps immediately. The equations governing the trapped-charge density through one cycle of $\varphi = \omega t$ under these conditions are:

$$\bar{n}_t = \frac{\gamma K}{\gamma + 1} (V_o + \hat{V}_m) \qquad \left.\begin{array}{l} 0 < \omega t < \varphi_1 \\ \varphi_2 < \omega t < 2\pi \end{array}\right\} \tag{9}$$

$$n_t = \frac{\gamma K}{\gamma + 1} \left[V_o + \frac{V_m \omega \tau}{(1 + \omega^2 \tau^2)^{1/2}} \right] \exp \left(\frac{\varphi - \varphi_1}{\omega \tau} \right) \varphi_1 < \omega \tau < \varphi_2 \tag{10}$$

where $\varphi_1 = \tan^{-1} (\omega \tau)^{-1}$, and φ_2 is the solution to the equation:

$$\left[V_o/V_m + \frac{\omega \tau}{(1 + \omega^2 \tau^2)^{1/2}} \right] \exp (-\varphi_2/\omega \tau)$$

$$= \left[\frac{V_o}{V_m} + \cos \varphi_2 \right] . \tag{11}$$

The solution for $\bar{n}_t$ in Eq. (9) applies provided $\omega > \omega_b = (V_o/\gamma V_m)$. If this inequality is not true, then the solution of Eq. (7) is applicable. Because γ in real crystals has turned out to be of the order of 1000 or greater,[2] the frequency dependence of $\bar{n}_t$ embodied in Eq. (9) is, therefore, the one considered in the following section in which admittance variation is discussed.

ADMITTANCE VARIATION

The frequency dependence for the trapped-charge density as considered here will result in a frequency dependence for the admittance function which characterizes the crystal. At all frequencies, the equivalent admittance to the crystal will consist of a parallel conductance and a capacitive susceptance. If measured on a frequency-variable bridge circuit, the equivalent capacitance will decrease from its low-frequency value of $3/2\ C_e$ (where C_e is the capacitance of a parallel-plate capacitor of equal dimensions) to C_e. This relaxation corresponds to the difference between the modulation of interior charge by the applied voltage which occurs at low frequencies and the modulation of charge stored on the electrodes, which occurs at high frequencies. The frequency dependence for this capacitance relaxation will, therefore, be the same as the frequency dependence of the Fourier component of $\bar{n}_t$ which is in phase with the applied voltage.

Fig. 3 is a plot of the in-phase component of $\bar{n}_t$ vs frequency, normalized to the frequency represented by the reciprocal-relaxation time τ^{-1}. The curves in Fig. 3 are drawn for five values of $\beta = V_o/V_m$, the dc to ac voltage ratio. The dependence of β for the relaxation phenomenon apparent in Fig. 3 occurs because the trap-emptying rate is a function of the total trap density. The emptying rate will, therefore, be increased by increasing the applied dc bias.

As the measured capacitance decreases because of an inability to modulate the trap density in the crystal, the measured conductance will increase because of the increased free-charge density. The free-charge density will remain in phase with $\bar{n}_t$, and will, therefore, increase from $KV_o/(\gamma + 1)$ to $K(V_o + V_m)/(\gamma + 1)$ as frequency increases. The specific dependence on frequency for $\bar{n}$ and, therefore, the dependence of conductance on frequency will be that of the dc component to the trapped-charge density. Curves of the calculated dependence of $\bar{n}$ on frequency are given in Fig. 4 for the same values of β the dc to ac ratio that were used in calculating the curves of Fig. 3. The crystal conductance will therefore increase from $e\mu KV_0/(\gamma + 1)$ to $e\mu K(V_o + V_m)/(\gamma + 1)$.

From these expressions, it is apparent that the total conductance variation is limited to the ratio $\beta/(\beta + 1)$ of the high-frequency value, and hence conductance variations become very slight as dc bias is increased. This behavior is clearly seen in Fig. 4.

The admittance variation considered here can be expected to occur at fairly low frequencies; in typical crystals in which space-charge-limited currents have been observed, the frequency τ^{-1} should be less than a few Mc. As an example, in a crystal having a trap density of 10^{14} cm^{-3}, the trapping time constant τ would range from about 100 ms to 1 μs for centers with capture cross sections in the normal range (10^{-15} to 10^{-20} cm^2).

This low-frequency range is particularly suitable for admittance-bridge measurements, so that positive identification of the phenomenon analyzed here would permit a sensitive and convenient means for the determination of trapping relaxation times. This determination, if coupled with dc space-charge-limited current measurements, would allow one to obtain trapping capture cross sections much more easily than by such measuring techniques as thermally-stimulated trap emptying.

Although other mechanisms can lead to admittance variations with frequency in the low-frequency range, the derived dependence on bias voltage for the effect analyzed here should allow a positive identification of the trap-emptying mechanism. An experimental program is now underway to see if admittance variations, already observed on crystals believed to conform to the model considered here, can be explained by this theory. Applicability of the distributed trap theory of Rose[1] demands that such a variation will take place.

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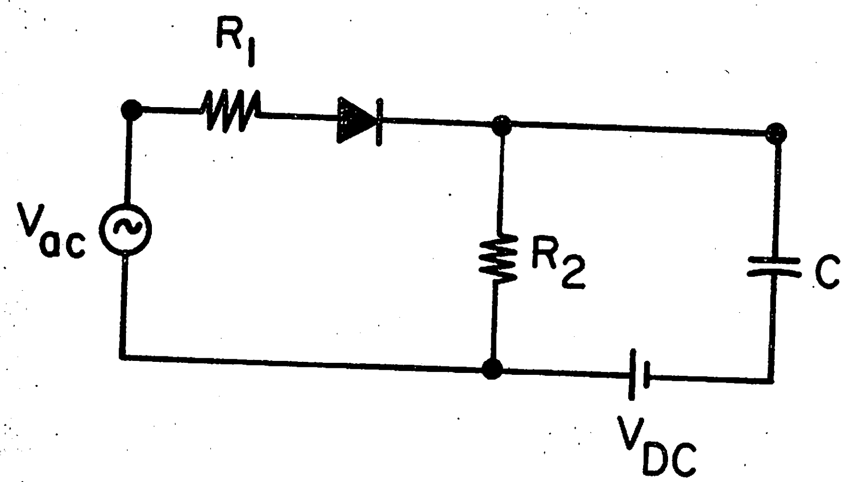
FIGURE CAPTIONS

Fig. 1 Analogue circuit to the trap-filling and trap-emptying sequence considered in the text. The voltage across the capacitance is analogous to the volume-distributed, trapped charge. The trap-emptying time constant has its analogue in R_2C while the trap-filling time constant is analogous to $R_1R_2C/(R_1 + R_2)$ which is roughly R_1C for $R_2 >> R_1$.

Fig. 2 Average trapped-charge density through one cycle of ωt at frequencies such that trap-emptying limitations result in a build-up of $\bar{n}_t$. The trap density $\bar{n}_t$ follows the applied frequency for $\varphi = \omega t < \varphi_1$ and at $\varphi > \varphi_2$. From φ_1 to φ_2 the density decays exponentially. Equations for φ_1 and φ_2 are given in the text.

Fig. 3 Decay of $\bar{m}_t$ (the Fourier component of $\bar{n}_t$ that is in phase with the applied voltage) as frequency is increased. The frequency scale $\alpha = \omega\tau$ is normalized to τ^{-1}, the attempt-to-escape frequency. The dependence on dc bias is embodied in the curve shift with changes in the ratio of dc to ac bias voltage $\beta = V_o/V_m$. As described in the text, capacitive relaxation should follow these curves.

Fig. 4 Calculated variation of the free-carrier density $\bar{n}$ with normalized frequency $\alpha = \omega\tau$. The density is normalized to its high-frequency value $\bar{n} = K V_m(\beta + 1)/(\gamma + 1)$. The crystal conductance will be proportional to this variation. The five curves represent five values of the dc to ac voltage ratio β.



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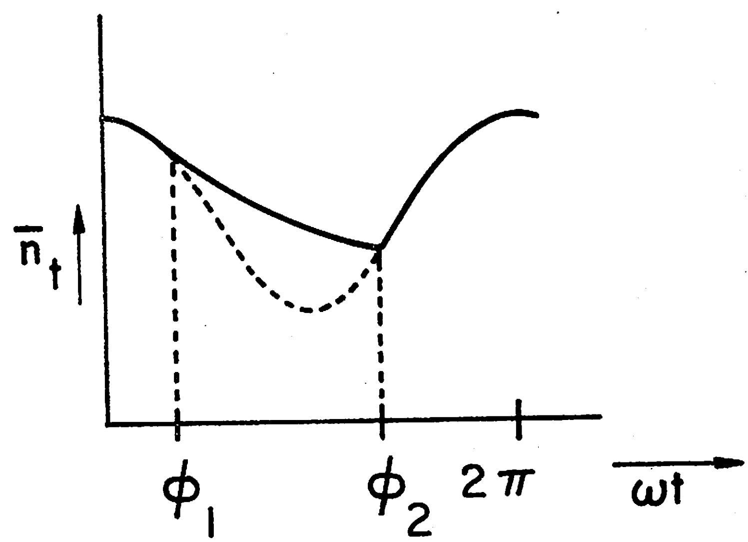
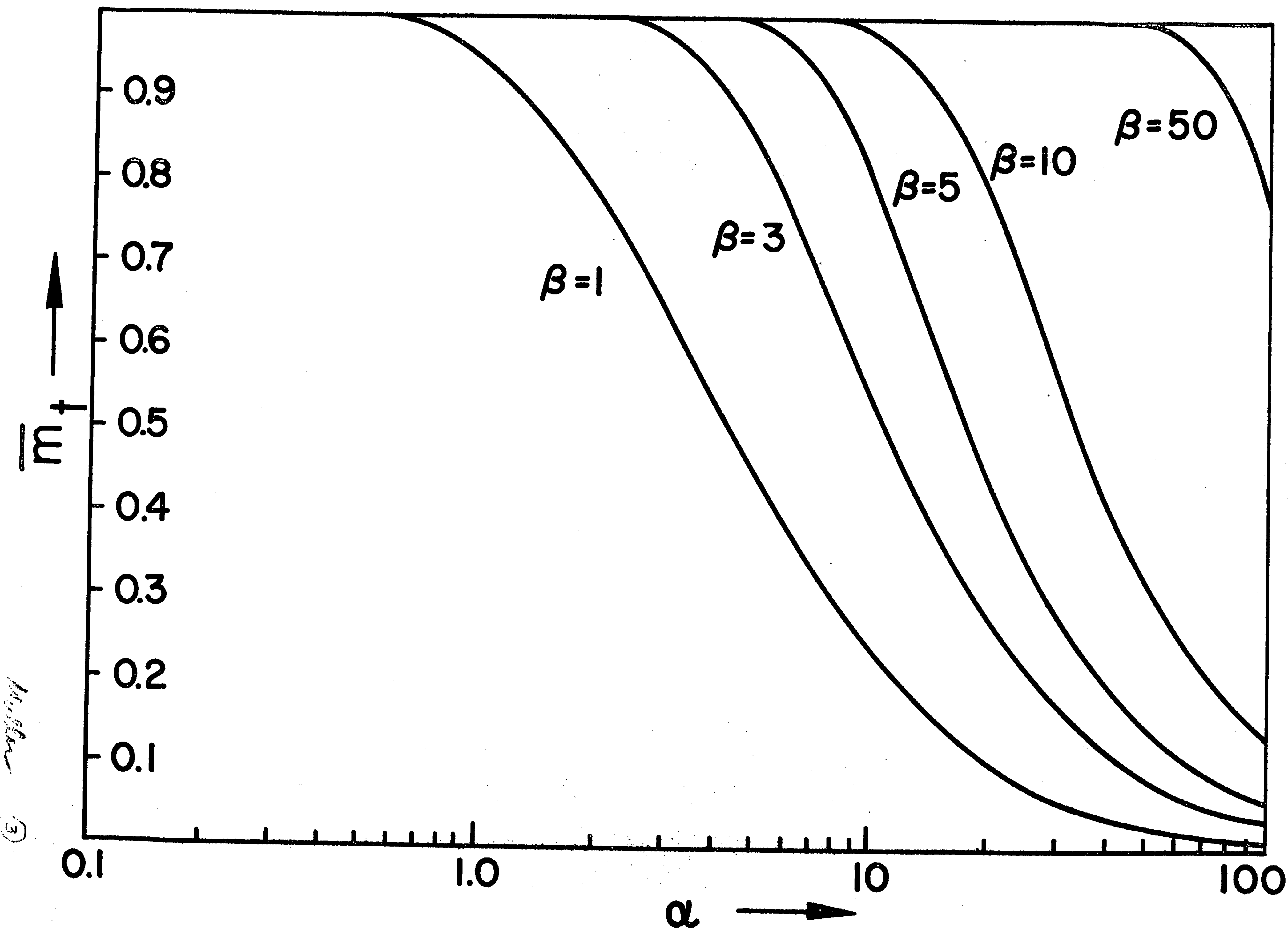
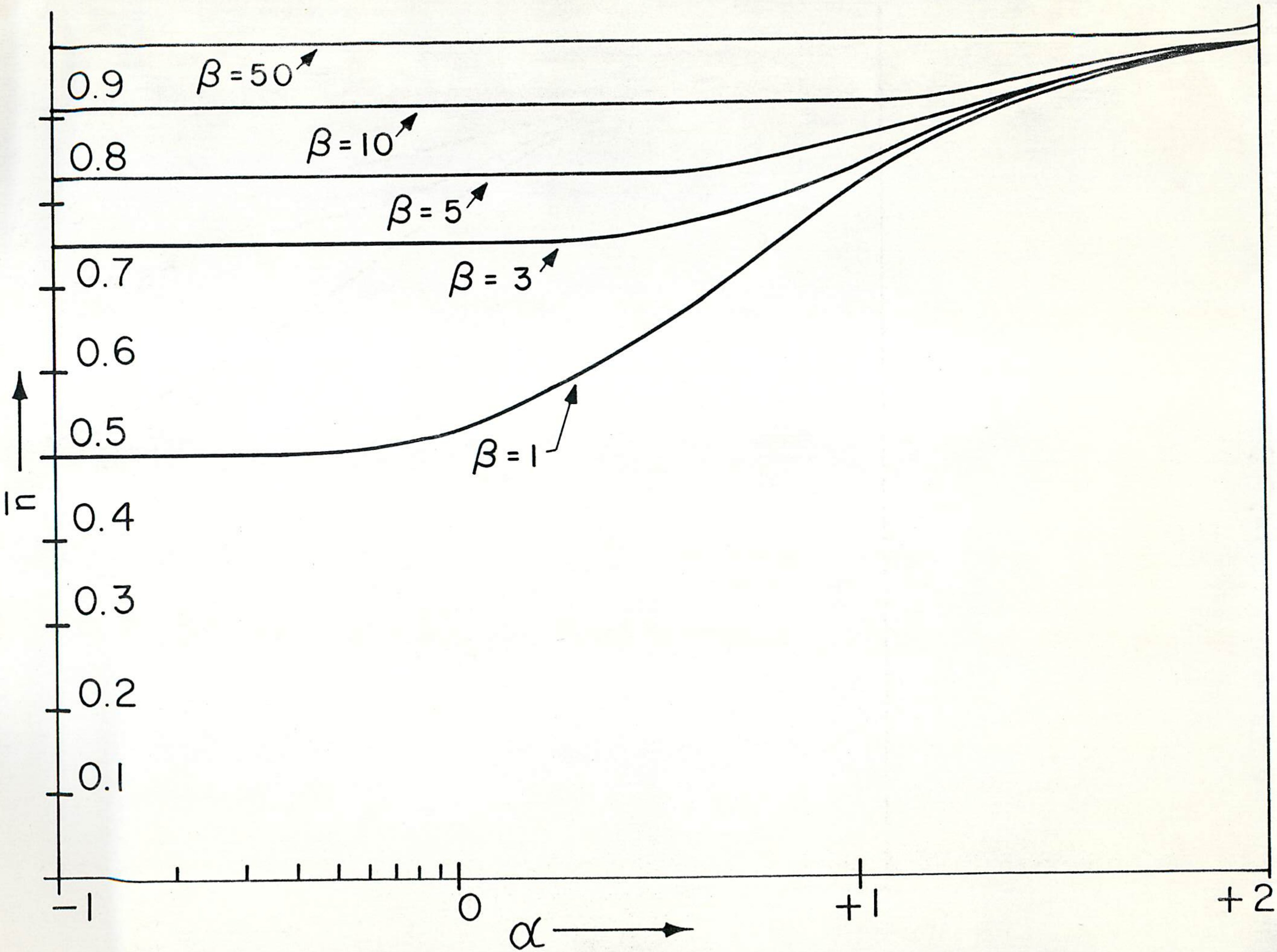


$\bar{n}_t$
ϕ_1
ϕ_2
2π
ωt



0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
$\overline{m}_t$
0.1
1.0
10
100
α
$\beta = 1$
$\beta = 3$
$\beta = 5$
$\beta = 10$
$\beta = 50$

Müller ③



β = 50
β = 10
β = 5
β = 3
β = 1
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
−1
0
+1
+2
α