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Nonlinear oscillations of a polar-liquid column under unipolar-ion injection

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A nonlinear oscillation of the level and of the current is theoretically predicted and experimentally observed in a U-tube system containing a polar liquid with a capillary whose diameter is as large as 1 mm, and which is equipped with an electrode injecting unipolar ions. The observed critical potential difference, amplitude, period, and waveforms of the level oscillation agree qualitatively and quantitatively with the theoretical results. The experimental amplitude of the current oscillation is larger than the theoretical one, and it is suggested that the discrepancy might be due to the high-field dissociation current of neutral impurities.

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I. INTRODUCTION

Many reports have been made on nonlinear phenomena caused directly or indirectly by the action of electro-osmosis.¹⁻⁴ Especially in studies on nerves and living cells, Teorell first indicated that in nerve excitation the phenomena could be simulated by a system in which a charged membrane separates two electrolytic solutions of different concentrations.¹ Analyses of such systems were made both by him and also extensively by Kobatake and Fujita, who took into account the action of electro-osmosis in each capillary of the membrane.³ Their results were confirmed in Meares and Page.⁴ For more recent studies on the stability of charged membrane systems the reader is referred to Dickel and Kretner⁵ and Srivastava *et al.*⁶ In their system, space charge, which is of course necessary in each capillary, is produced as a double layer induced by the charged capillary wall. Here we consider a U-tube system which consists of two level-free cylindrical reservoirs for a polar liquid connected by a horizontal capillary. One reservoir is equipped with a unipolar injector and the other with a collector. One may expect different characteristics in the electro-osmosis from the charged-membrane system, especially concerning nonlinear oscillatory states of the liquid flow. These problems may arise not only from pure physical interest but also from the viewpoint of the practical application for ion-drag pumping.⁷ In this paper we show how the nonlinear oscillations appear in our U-tube system both theoretically and experimentally. From theoretical investigations the criteria for the generation of oscillations of the level and the current were obtained. The waveform, amplitude, and period are found to depend on the applied potential difference V and the injected charge density q_0 at the injector. The experimental results essentially support the theory both qualitatively and quantitatively. However, several points are left open for future studies.

II. THEORY

To simplify this discussion we employ the following assumptions. (1) The cross-sectional area of the capillary is so small compared to that of the reservoir, where the injecting or collecting electrode is placed, that the potential drop around the electrode is negligible compared to that in the capillary, but still strong enough for charge injection or col-

lection. (2) The radius a of the capillary is much smaller than the length l . (3) The contribution to the electric current of the ion density dissociated from molecular impurities is not considered. This assumption is not critical for the occurrence of the level oscillation, since the compensated bipolar current does not contribute to the electro-osmotic pressure. However, the real current would be the sum of the calculated current and the bipolar current. (4) In the capillary the radial changes of the pressure, charge density, electrical potential, and radial component of the liquid velocity are neglected. So the spatial dependence of these variables, except for the liquid velocity v and the electric current j is only on the axial coordinate x .

A. Steady-State Analysis

In the capillary the basic equations for the charge density $q(x)$, electrical potential $\phi(x)$, current density $j(r)$, pressure $p(x)$, and axial velocity of the liquid $v(r)$ are written in steady states as

$$\frac{\partial^2 \phi}{\partial x^2} = -\frac{q}{\epsilon} \quad (1)$$

$$\frac{dj}{dx} = 0, \quad j = -\mu q \frac{\partial \phi}{\partial x} + qv, \quad (2)$$

$$-\frac{\partial p}{\partial x} + \frac{\eta}{r} \frac{\partial}{\partial r} \left(r \frac{\partial v}{\partial r} \right) - q \frac{\partial \phi}{\partial x} = 0, \quad (3)$$

where ϵ , μ , and η are the dielectric permittivity, ion mobility, and viscosity of the liquid, respectively. Equation (1) is the Poisson equation, and Eq. (2) is the current-continuity law, where the first term in j expresses the drift motion of the space charge with mobility μ and the second term in j is a convective current. Here the diffusion term is neglected since we consider only the case where the electric field is strong enough. Equation (3) is the Navier-Stokes equation, where the inertia term has been dropped, assuming the motion to be sufficiently slow.

The analysis for the very narrow charged capillary, which is a model for a cell membrane, started from similar equations.³ The electro-osmotic force in that case arises from the space charge near the wall in Debye-Hückel approximation, while in our case the electro-osmosis arises from the whole volume of the capillary. Since p , q , and ϕ are indepen-

dent of the radial coordinate r , Eq. (3) is easily integrated with respect to r , then averaged over the cross section to give

$$u = -A \frac{\partial p}{\partial x} - Aq \frac{\partial \phi}{\partial x}, \quad (4)$$

where u is the liquid velocity averaged over the cross section and

$$A = a^2/8\eta. \quad (5)$$

Equation (2) then becomes

$$j_m = -\mu q \frac{\partial \phi}{\partial x} + qu, \quad (6)$$

where j_m is the average current density over the cross section. Equations (1), (4), and (6) may be represented with nondimensional primed variables by the following transformations:

$$\begin{aligned} x' &= x/l, & \phi' &= \phi/V, & u' &= (l/\mu V)u, \\ j'_m &= (l^3/\epsilon\mu V^2)j_m, & p' &= (l^2/\rho\mu^2 V^2)p, \\ q' &= (l^2/\epsilon V)q, \end{aligned} \quad (7)$$

where V is the potential difference between the electrodes, and ρ is the density of the liquid. The transformed equations are, after omitting the primes,

$$\frac{\partial^2 \phi}{\partial x^2} = -q, \quad (8)$$

$$j_m = -q \frac{\partial \phi}{\partial x} + qu, \quad (9)$$

$$u = -A_p \frac{\partial p}{\partial x} - A_e q \frac{\partial \phi}{\partial x}, \quad (10)$$

where

$$A_p = \rho\mu\xi^2 V/8\eta, \quad A_e = \epsilon\xi^2 V/8\eta\mu, \quad \xi = a/l. \quad (11)$$

by eliminating ϕ from Eqs. (8) and (9) one can obtain the result

$$q^2 = j_m/2(x+c), \quad (12)$$

where c is a constant of integration to be determined by the boundary condition, which should be given at $x=0$ when the ion velocity $v_i = E + u$ is positive. When $V_i = 0$, $j_m = qV_i = 0$, and then, from Eq. (12), q in turn is zero everywhere. By use of Eq. (8), electric field E becomes unity. Therefore $v_i = 0$ corresponds to $u = -1$. Thus Eq. (12) will be (i) for $u > -1$ ($v_i > 0$), $q = q_0$ at $x = 0$,

$$q = \left(\frac{j_m}{2(x + j_m/2q_0^2)} \right)^{1/2}, \quad (13)$$

or (ii) for $u < -1$, ($v_i < 0$), since no carrier can be injected from the collector,

$$j_m = 0 \text{ and } q = 0. \quad (14)$$

To obtain the static relation between the flow velocity u and the negative pressure difference across the capillary $-\Delta p$ (hereafter called the u - p curve) for $u > -1$, we substitute q of Eq. (13) into (9) and we obtain

$$\partial \phi / \partial x = - \left[2j_m \left(x + \frac{j_m}{2q_0^2} \right) \right]^{1/2} + u. \quad (15)$$

The integration with respect to x from 0 to 1 gives

$$u = \frac{2}{3}(2j_m)^{1/2} \left[(1 + j_m/2q_0^2)^{3/2} - (j_m/2q_0^2)^{3/2} \right] - 1. \quad (16)$$

Here we have used the condition $\phi(1) - \phi(0) = -1$. The elimination of $q(\partial \phi / \partial x)$ from Eqs. (4) and (6) and the integration with respect to x from 0 to 1 gives

$$\Delta p = (A_e/A_p)j_m - (1/A_p) \left\{ 1 + A_e(2j_m)^{1/2} \times [(1 + j_m/2q_0^2)^{1/2} - (j_m/2q_0^2)^{1/2}] \right\} u. \quad (17)$$

Here we have used the condition $p(1) - p(0) = \Delta p$.

Thus we have obtained the results, Eqs. (16) and (17), which represent the u - p characteristics of the capillary system as a function of the average current density j_m or q_0 . From Eqs. (16) and (17) one can, in principle, eliminate j_m or q_0 . But still one of them remains as a parameter for the u - p curve. The charge density q_0 at the injection side of the capillary could in general depend on the liquid-flow velocity u . However from condition (1), we assume that q_0 is maintained constant. The differentiation of Eq. (17) with respect to u gives

$$\frac{d(-\Delta p)}{du} = -2q_0 \frac{A_e}{A_p} G + \frac{1}{A_p}, \quad (18)$$

where

$$G = q_0 \frac{\partial J}{\partial u} - u \frac{\partial f}{\partial u} - f, \quad J = \frac{j_m}{2q_0^2} \quad (19)$$

and

$$f = J^{1/2}[(1+J)^{1/2} - (J)^{1/2}]. \quad (20)$$

A negative slope in the u - p curve may appear for a large positive value of $q_0 A_e G$, where A_e includes the potential difference V linearly. It is seen from Eq. (19) that, roughly speaking, negative resistance in u - p curves is realized if the first term of the equation is relatively larger than the second and third terms. Here we note that f , $\partial J / \partial u$, and $\partial f / \partial u$ are positive.

It is difficult in general to eliminate j_m analytically. First we consider two extreme cases.

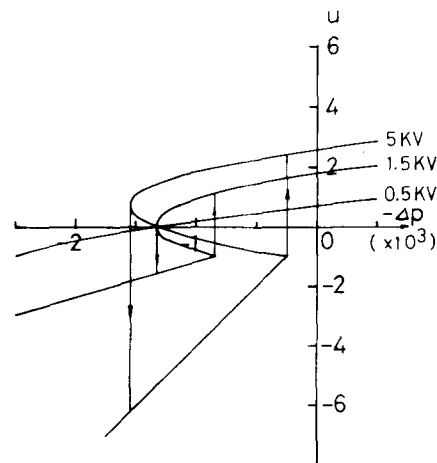


FIG. 1. u vs $-\Delta p$ characteristics obtained from Eq. (24) for the SIL case, where $A_p = 8.0 \times 10^{-7} V$ and $A_e = 9.4 \times 10^{-4} V$, where V is the potential difference in volts.

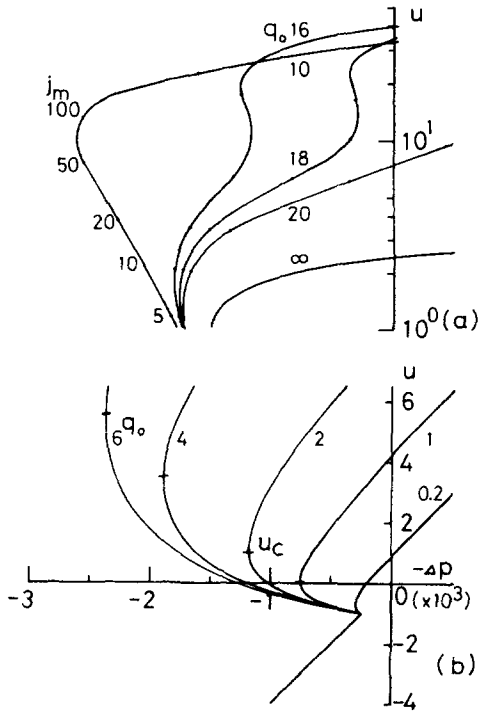


FIG. 2. u vs $-\Delta p$ characteristics obtained from Eqs. (16) and (17), where the values of A_p and A_e are same as in Fig. 1, with $V = 5$ kV. Curves are shown (a) for $u > 1$, (b) for relatively small q_0 's.

1. Weak Injection Limit (WIL): $j_m/2q_0^2 \gg 1$

In the first-order approximation Eq. (16) becomes

$$j_m = q_0(u + 1) \quad \text{for } u > -1, \quad (21a)$$

and from Eq. (14)

$$j_m = 0 \quad \text{for } u < -1. \quad (21b)$$

From Eq. (17) we find

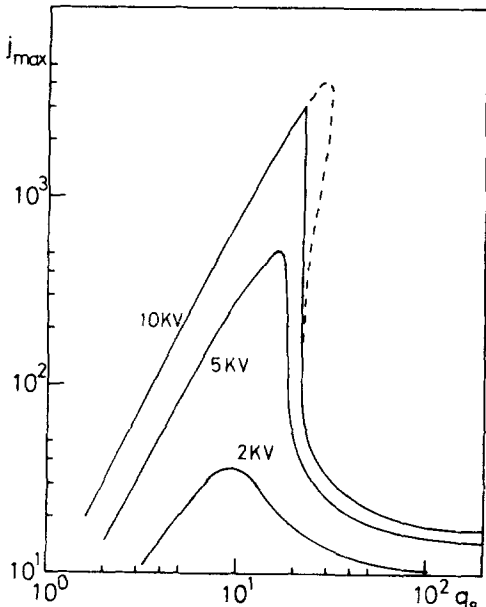


FIG. 3. q_0 dependence of j_{max} , obtained from Eqs. (16) and (17) for various potential differences, where the values of A_p and A_e are same as in Fig. 1, with $V = 5$ kV.

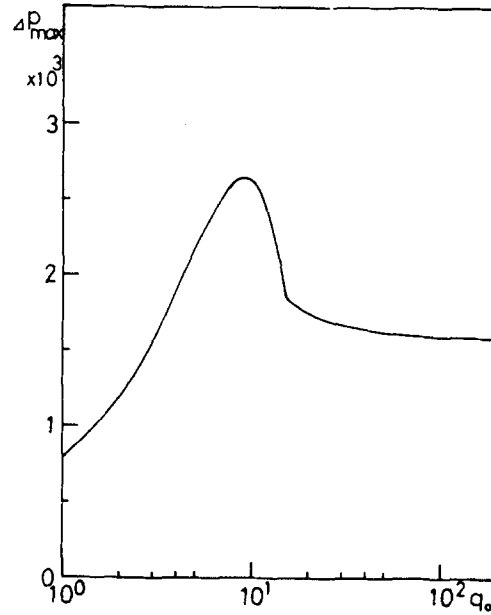


FIG. 4. q_0 dependence of Δp_{max} . The conditions are the same as in Fig. 1.

$$u = -A_p \Delta p + A_e q_0 \quad \text{for } u \geq -1, \quad (22a)$$

and from Eq. (14)

$$u = -A_p \Delta p \quad \text{for } u < -1. \quad (22b)$$

Equations (22) shows that u is related to Δp linearly. Therefore there is no nonlinear phenomenon.

2. Strong Injection Limit (SIL): $j_m/2q_0^2 \ll 1$

Neglecting the term $j_m/2q_0^2$ in Eq. (16), we find that

$$j_m = \frac{3}{8}(u + 1)^2 \quad \text{for } u \geq -1, \\ = 0 \quad \text{for } u < -1, \quad (23)$$

and from Eqs. (17) and (14)

$$\Delta p = -(3A_e/8A_p)(u - 3)(u + 1) - u/A_p \quad \text{for } u \geq -1, \\ = -u/A_p \quad \text{for } u < -1. \quad (24)$$

The maximum of Δp is easily obtained from Eq. (24):

$$\Delta p_{max} = (3A_e/8A_p)[(1 - 4/3A_e)^2 + 3]. \quad (25)$$

At the maximum of Δp ,

$$u = u_c = 1 - 4/3A_e. \quad (26)$$

Hereafter u_c is called the critical velocity. In this limit one obtains from Eq. (19)

$$G = -\frac{1}{2}J^{1/2} + 3/4q_0. \quad (27)$$

The condition of $G > 0$ is satisfied when

$$J < 9/4q_0^2, \text{ namely, } j_m < 9/2. \quad (28)$$

From Eq. (24) the u - p curves are shown in Fig. 1 for the several potential differences. It is obvious that the nonlinear level oscillation occurs when

$$u_c > 0, \quad (29)$$

namely,

$$A_e \geq 4/3. \quad (30)$$

When the slope of the u - p curve at $u = 0$ is positive, the system can be at the stationary state. The critical condition can be represented by Eq. (29).

3. Intermediate Region of $j_m/2q_0^2$

For the intermediate region of $j_m/2q_0^2$ numerical computations reveal an additional negative-resistance region in the u - p curve as shown in Fig 2: Fig 2(a) is $u > 1$ in a semilog graph, with corresponding current values j_m noted on the curves; Fig 2 (b) shows the u - p curve for relatively small values of q_0 , with critical flow velocity u_c on each curve. Figures 3 and 4 show the dependence of maximum value of j_m and Δp on q_0 . The maximum of the electric current strongly depends on q_0 . For a large q_0 ($> 10^2$), j_{max} and Δp_{max} become constant for a given potential difference V . This means that the property of the charge injector is not important when q_0 is larger than 10^2 . Single maximum of pressure difference is found for relatively small values of q_0 , the upper limit of which depends on the applied potential difference. For example, the limiting value of q_0 is about 15 at $V = 5$ kV. The nonlinear oscillatory state appears when

$$u_c \geq 0, \quad (31)$$

which is given by the same argument as in the SIL case, where u_c is the critical velocity causing a maximum of pressure difference. The nonlinear oscillation can be initiated by increasing the potential difference V , since V changes the critical velocity u_c from negative to positive.

It was found that there are two mechanisms for occurrence of the negative resistance. For a given liquid of a finite mobility, electro-osmotic pressure is caused by qE , which is proportional to the conductive current. The larger the ratio

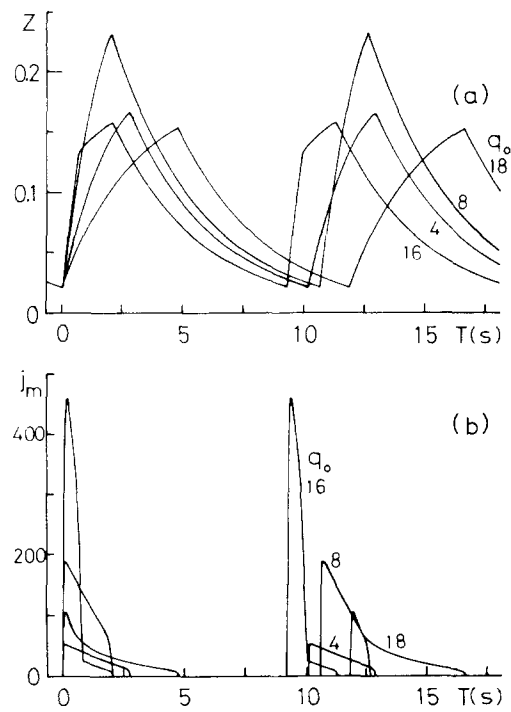


FIG. 6. Waveforms numerically obtained from Eq. (34) for intermediated q_0 , for the (a) level and (b) current. The numerical parameters are the same as in Fig. 5.

of the conductive current to the convective current, the greater the electro-osmotic pressure. The negative resistances in the u - p relation therefore occur in the region where the convective current is relatively small. A possible explanation for this is that the flow itself is small, or that the charge increment for a given flow increment is relatively small.

B. Analysis of Dynamic Behavior

When the condition expressed in Eq. (31) is satisfied, the liquid in the U-tube could be in the state of nonlinear level oscillation. Here we show the waveform analysis. The equation of motion for the liquid in the U-tube is

$$\rho(\lambda l + L) \frac{d^2 z}{dt^2} = \Delta p - 2\rho g z, \quad (32)$$

where λ is the ratio of the cross section of the vertical column (reservoir) to that of the capillary. Here z expresses the liquid level in the collecting reservoir measured from the equilibrium point at zero potential difference. For Δp in Eq. (32) we use the expression from Eq. (17) for the static condition. This approximation is valid if the time constants of the velocity distribution $\tau_v = 8\eta/\rho a^2$ and the charge distribution $\tau_q = \epsilon/\mu$ are smaller compared to the typical period $T = l^2/\mu V$ of the level oscillation. We estimate $\tau_v \sim 0.01$, $\tau_q \sim 0.02$, whereas $T \sim 10$ (for 1 kV), which proves the validity of our approximation. The primed variables are nondimensional and are determined as

$$z' = z/l, \quad t' = (\mu V/l^2)t,$$

and

$$\Delta p' = (l^2/\rho\mu^2 V^2)\Delta p. \quad (33)$$

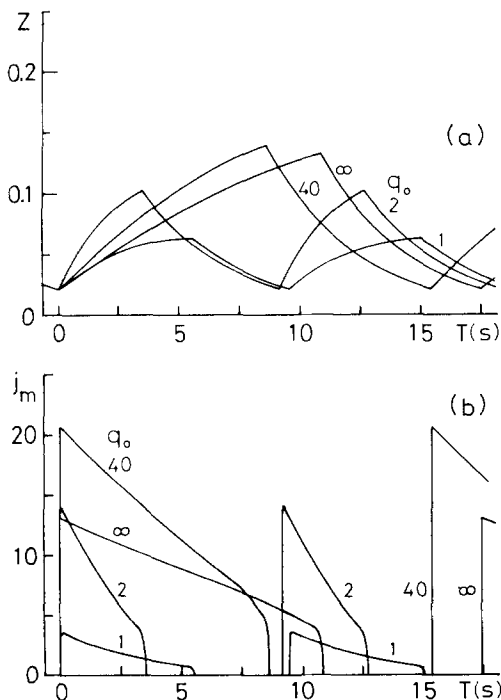


FIG. 5. Waveforms numerically obtained from Eq. (34) for various q_0 's for the (a) level and (b) current. Numerical parameters are the same as in Fig. 1, with $V = 5$ kV.

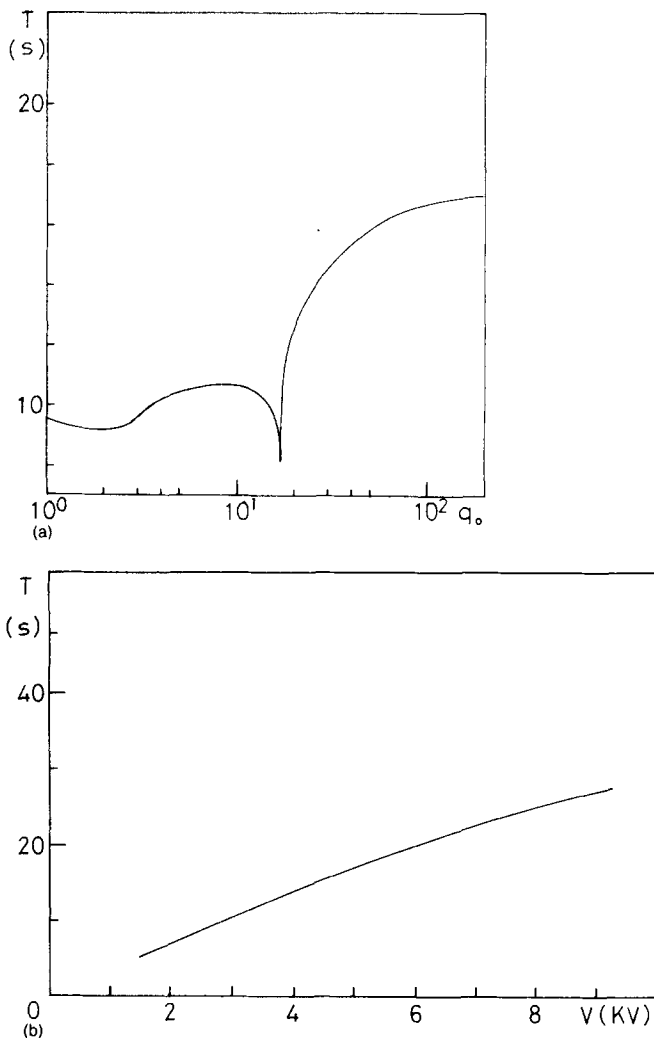


FIG. 7. Oscillation period: (a) q_0 dependence for $V = 5$ kV, (b) dependence on potential difference for the SIL case.

The equation of motion becomes, after omitting the primes and neglecting L compared to $l\lambda$,

$$\frac{d^2z}{dt^2} = \alpha\Delta p - \beta z, \quad (34)$$

where $\alpha = 1/\lambda$, $\beta = 2gl^3/\lambda\mu^2V^2$, and g is gravitational acceleration.

Waveforms of the level and of the current have been computed from Eqs. (34) and (17) by the Runge-Kutta method at the typical experimental condition $V = 5$ kV for various values of q_0 . The results are shown in Fig. 5 (a) and 6 (a). The waveforms of the level oscillation can be classified into four types: as (1) the region of $1 < q_0 < 2$ where the rising period is comparable to the falling period, (2) the region of $2 < q_0 < 15$ where the rising period is shorter than the falling period, (3) the region of $15 < q_0 < 17$ where there are two rising rates, one larger than the falling rate and the other. The rising rate changes discontinuously from first one to the second, comparable to the falling rate, and (4) the region of $q_0 > 17$ where the rising period is comparable to or longer than the falling period. The values of q_0 which separate these four regions were obtained for 5 kV.

In contrast to the dramatic changes of the waveforms for the region of $q_0 < 20$, for $q_0 > 20$ the waveforms are rather simple. The experimental observation of the waveform variation may yield data necessary to estimate the strength of the injection. The oscillation period has a complicated dependence on q_0 as shown in Fig. 7(a). The period increases when q_0 is above 18. In the region of $10 < q_0 < 18$ the intense electro-osmotic force acts to decrease the period sharply. The period also depends on the potential difference. Calculated periods are shown in Fig. 7 (b) for the SIL case. From the critical potential difference of 1.4 kV the period increases with the applied potential difference. The corresponding waveforms of current oscillation are shown in Figs. 5(b) and 6(b). The current flows only in the rising period and increases with the flow speed.

We summarize here the present theoretical findings.

(1) Negative resistance can appear in u - p curves for the U-tube system when the injected charge density is sufficiently strong. The critical condition for the nonlinear oscillation is, for the SIL case,

$$V > V_c = 32\mu\eta/3\zeta^2\epsilon.$$

(2) Two types of negative resistance were found in u - p curves for the intermediate region of q_0 . They cause two different rising rates in the waveform of level oscillation (Fig. 20).

(3) Analysis of dynamic behavior has given detailed characteristics of the system: (i) the waveforms for various q_0 (Figs. 5 and 6), (ii) the dependence of the amplitude of oscillation of Δp and j_m on q_0 (Figs. 3 and 4), and (iii) to q_0 and V dependence of the period of oscillation (Fig. 7).

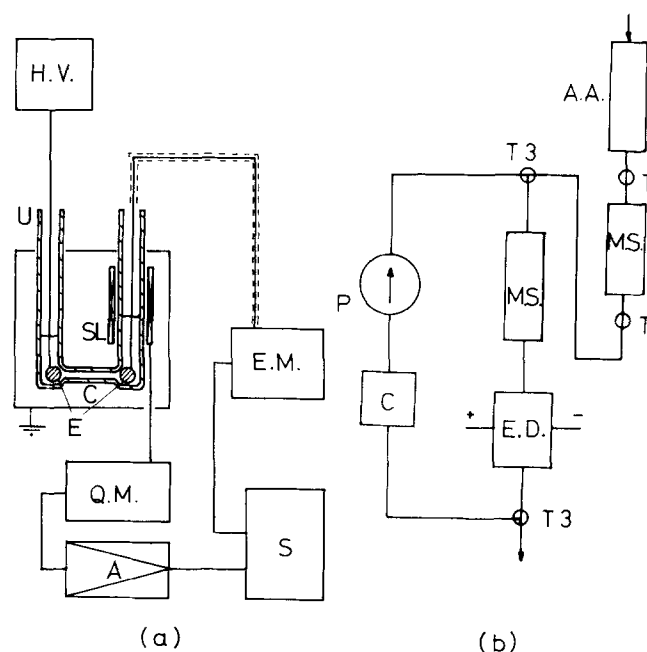


FIG. 8. (a) Experimental arrangement: HV, high-voltage power supply; U, Pyrex U tube (the inner radius of the vertical-column reservoir is 3 mm; E, electrodes for unipolar ion injection and collection; EM, electrometer for current measurement; C, capillary of inner radius $a = 0.40 \pm 0.05$ or 0.27 ± 0.03 mm and length $l = 15$ mm; SL, single-layer solenoid; Q.M., Q meter; A, amplifier; S, oscilloscope or recorder. (b) Arrangement for liquid purification: AA, activated alumina; T and T3, Teflon cocks; MS, molecular sieves (4 Å); ED, electro-dialyzer; C, test cell; and P, pump.

III. EXPERIMENT

We have fabricated a U-tube system with unipolar ion-injection electrodes and measured the static and dynamic behavior of the level and current for various impurity concentrations and applied potential differences. Experiments were carried out with the arrangement shown in Fig. 8(a). The U tube was made of Pyrex and had a capillary section at the base of the U. In carrying out these experiments it is important to avoid pumping due to mechanisms other than ion emission from one of the paired electrodes, e.g., dielectrophoretic effects or space-charge generation due to high-field dissociation.⁷ The dielectrophoretic effect can be avoided if the paired electrodes are symmetrical, and the space-charge generation can be neglected if the test liquid is sufficiently purified. In advance of the experiment the pressure head was tested and found negligible by utilizing a noninjecting electrode pair which was replaced for an injecting pair. The noninjecting electrode pair was produced by coating metallic electrodes with ion-exchange material of reverse polarity to the applied potential, while the injecting electrode was coated with material of the same polarity.⁸ In the present experiment potassium polyvinyl sulphate was used for the cation exchanger. The test liquid used was nitrobenzene with the relative permittivity $\epsilon = 36$, the viscosity $\eta = 2cp$, the liquid density $\rho = 1.2 \times 10^3 \text{ kg/m}^3$, the capillary length $l = 1.5 \times 10^{-2}$, radius $a = 2.7 \pm 0.3 \times 10^{-4}$ or $4.0 \pm 0.5 \times 10^{-4} \text{ m}$. The liquid was left for several days in activated alumina and subsequently put through molecular sieves (4Å), following which it was led into a loop having an electro dialyzer, with paired noninjecting electrodes, and an electric field of 4 kV/cm was applied as shown in Fig. 8 (b). The liquid was circulated for hours. The electrical resistivity of the liquid was varied by changing the period of electro dialysis from 10^5 to $10^8 \Omega \text{ m}$.

To compare the experimental observation with theoretical calculation, we need to know q_0 . q_0 is known to increase with increasing impurity concentration,⁹ though the mechanism is not yet very clear. The dependence of q_0 on the potential difference V may be estimated experimentally, for example, by the waveform of the oscillation. The direct measurement of q_0 by means of the Kerr effect—and so on—

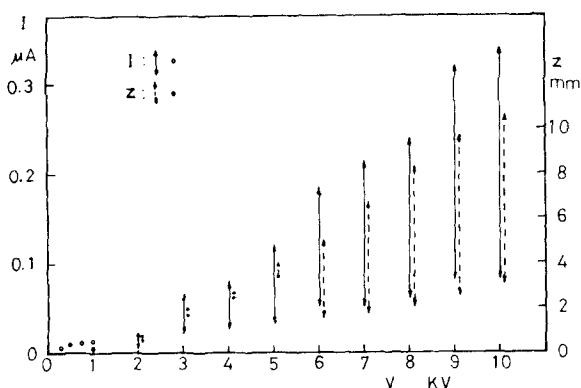


FIG. 9. I - V characteristics: solid arrows indicate the peak-to-peak variation of the current; dotted arrows indicate the level variation; capillary radius a equals 0.40 mm.

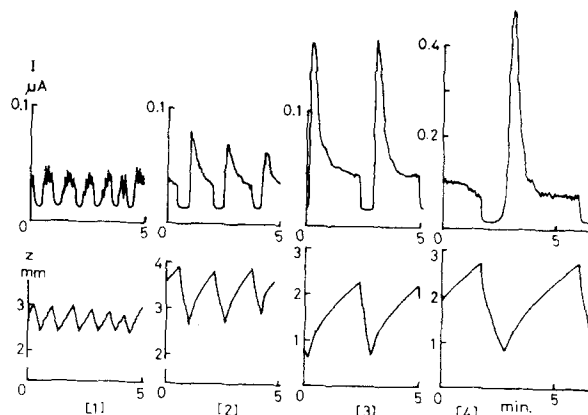


FIG. 10. Waveforms of the level and current for various conductivities of nitrobenzene, (1) 8×10^{-7} , (2) 9×10^{-7} , (3) 1.2×10^{-6} , and (4) $2 \times 10^{-6} (\Omega \text{ m})^{-1}$ with capillary radius $a = 0.27 \text{ mm}$, at 4 kV.

would be more plausible but we did not try it in the present work. It is expected that the electrical resistivity of the liquid, which depends on the concentration of the electrolytic impurities, affects the injected charge density. Therefore the purity of the liquid, as measured by the current, was considered a parameter which has a strong correlation with the theoretical parameter q_0 .

The liquid level in the U tube was detected with a single-layer solenoid. The floating capacitance between coils changes with the liquid level, and was measured by a Q meter.

IV. EXPERIMENTAL RESULTS AND DISCUSSION

A. Amplitude and critical condition

Figure 8 shows a typical example of the range of temporal variations of level and current versus the potential difference in the level-free condition for a liquid of given conductivity. The level and current oscillate between the arrow heads. The stationary current and level values increase with the increasing potential difference up to 1.5 kV in this particular case. For a potential difference greater than this one sees that the stationary state of the level and the current becomes unstable against nonlinear oscillating states. The critical value for the oscillation is in good quantitative agreement with the theoretical prediction $V_c = 32\eta\mu/3\xi^2\epsilon = 1.4 \text{ kV}$ for the SIL case according to Eq. (30). The amplitude of the level and current oscillation increase with the applied potential difference. The level amplitude at 5 kV for this liquid is about 4 mm, whereas the theoretical value for SIL case is 2.0 mm using a value of the ion mobility of $1.5 \times 10^{-8} \text{ m}^2/\text{Vs}$ which had been obtained by conductometry.¹⁰ The theory and the observation fall into the approximately same quantitative range, although we see below how the amplitude changes by liquid conductivity. The amplitude of the current at 4 kV for the case shown in Fig. 9 is on the order of 10^{-7} A , while the theory in the model liquid predicts that the current due to injected ions is only of the order of 10^{-11} A . This is the only quantitative disagreement between simple theory and observation. We shall come back to this point later.

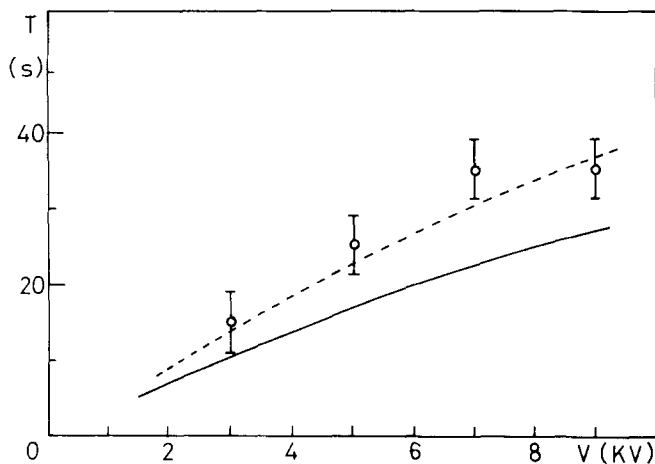


FIG. 11. Dependence of oscillation period on the potential difference. Experimental results are shown (open circles) together with predicted results (solid line) from Fig. 7(b); capillary radius a equals 0.40 mm. The dashed line was predicted theoretically for $a = 0.35$ mm.

B. Waveforms

The waveforms of level and current for a fixed value of the potential 5 kV, but for various liquid conductivities are shown in Fig. 10. Generally speaking, the waveform of the level is very similar to the theoretically predicted one. The waveform of the current is similar for small amplitudes, but it departs from the theoretical prediction with increasing amplitude. The waveform of level oscillation varied gradually from case (1) to (4). The conductivity of the liquid increases as one goes from case (1) to (4). The waveform of the level for case (1) is almost symmetric; that is, the rising period is close to the falling period. As the conductivity increases, the rising period becomes longer than the falling period. For case (4) the rising period is almost three times longer than the falling period. If we go back to the theoretical waveform of Figs. 5 and 6, we see that for small q_0 the ratio of the two periods is about unity, and for $q_0 \rightarrow \infty$, the ratio becomes about two. However for intermediate values of q_0 , the rising period becomes much shorter than the falling period. Therefore it is most natural to consider that case (1) corresponds to the value of $q_0 \approx 20$, and that value increases as we go to (2), (3), and (4). Thus the general feature of level oscillation is quite well explained by the theory. The magnitude and waveform of the current are not completely explained by the present theory. The current observed might include the effect of high-field dissociation of impure molecules. They do not, however, contribute to the electro-osmotic force since they are compensated for in this situation, as was examined in the first paragraph of Sec. III. This would not play an important role in level oscillation, only increasing the measured current in some fashion. The magnitude of the additive current and its dependence on flow speed u are not known at the moment. We have analyzed waveforms of current oscillation in theory, assuming that the injected charge density q_0 at the injector is constant for the entire period of oscillation, neglecting the u dependence of q_0 . But if the u dependence of q_0 is not negligible, the waveform of the current can be influenced by the oscillating part of q_0 in a com-

plicated manner. In addition, we have assumed a homogeneity of all the physical quantities other than the flow velocity in the radial direction of the capillary. But inhomogeneity, for example, darts or turbulence might have some effect on the static and dynamic behavior of the U-tube system. These problems are left for future studies.

C. Period

Figure 11 shows an example of the observed period of oscillation as a function of the applied potential difference. The period increases gradually with the increasing value of potential difference. The theoretically calculated values for the SIL case are shown in the same figure. The general qualitative features and quantitative values show good agreement. Figure 12 shows an example of the observed period of oscillation for a constant applied potential difference, but with varied conductivities of the liquid. As we have discussed before, q_0 is a function of the conductivity of the liquid for a given potential difference. Therefore by increasing the conductivity, the q_0 value is also considered to increase.

The experimentally observed tendency agrees with the q_0 dependence of period T numerically calculated and shown in Fig. 7.

V. CONCLUDING REMARKS

A nonlinear oscillation of the level and the current is theoretically predicted and experimentally observed in a U-tube system with electrode injecting unipolar ions containing polar liquid.

This new oscillation has many counterparts in the physical origin with the level oscillation in a very narrow charged capillary filled with electrolytic solutions proposed by Kobatake and Fujita. However this nonlinear oscillation takes place in a tube of a diameter of the order of a millimeter and the capillary is not charged. The oscillation in electrolytic solutions arises from a thin, charged double layer near the capillary wall, whereas the present oscillation is caused by the electric force acting on the whole volume of the tube.

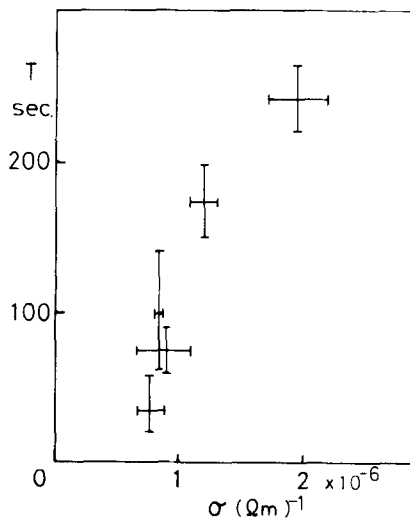


FIG. 12. Conductivity dependence of oscillation period with capillary radius $a = 0.27$ mm, at 4 kV.

The level and the current oscillations were experimentally observed. The critical potential difference, amplitude, period, and waveforms of the level oscillation agreed qualitatively and quantitatively with the theoretical predictions. The amplitude of the current oscillation was larger than the predicted, and it is suggested that the discrepancy might be explained by high-field dissociation current of neutral impurities.

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