

A second look at electrokinetic phenomena in boiling

Trevor J. Snyder

School of Mechanical and Materials Engineering, Washington State University, Pullman, Washington 99164

John B. Schneider

School of Electrical Engineering and Computer Science, Washington State University, Pullman, Washington 99164

Jacob N. Chung^{a)}

School of Mechanical and Materials Engineering, Washington State University, Pullman, Washington 99164

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In the article by V. Asch [J. Appl. Phys. **37**, 2654 (1966)], experiments were performed to study the influence of an electrostatic field on nucleate boiling of Freon-113(R-113). We have found that Asch might not have properly considered the effects of his experimental setup and therefore came to incorrect conclusions concerning electrophoretic and dielectrophoretic forces. Asch's analysis of the electric field distribution led him to conclude that the dielectrophoretic forces were small, however, we show in this article that, in general, there are strong dielectrophoretic forces in the vicinity of the heater wire. This article presents the results from a set of experiments performed with an apparatus similar to that of Asch's with test fluids of R-113 and FC-72. The experimental results show that vapor bubbles can be attracted to either the anode or cathode depending on the potentials with respect to the heater wire on which the boiling takes place. This is contrary to the results obtained by Asch which led him to conclude that the bubbles were always attracted to the anode. The bubble movement appears to be the result of a combination of dielectrophoretic forces (which are very strong but highly localized), electrophoretic forces, and bulk electroconvective flow. Furthermore, the combined effects of these forces away from the wire can lead to unstable behavior. © 1996 American Institute of Physics. [S0021-8979(96)08608-7]

I. INTRODUCTION

In 1966, Asch¹ presented the results from two experiments. The first experiment consisted of two ball electrodes with a chromel wire placed midway between them. The working fluid was R-113. The wire was electrically heated with 4.5 amps of direct current which produced boiling on the wire. This experiment was designed to study the trajectories of the vapor bubbles from the heater wire when a 30 kV potential was placed across the ball electrodes. It was observed that placing the heater wire near the anode resulted in vapor bubbles being drawn towards the anode. The results were the same when the heater wire was placed near the cathode, i.e., bubbles still traveled to the anode. According to Asch "the test thus proved that motion was due to the charging of particles rather than the nonuniformity of field, i.e., that the observed phenomenon is due to electrophoresis and not to dielectrophoresis, else the bubble pattern would have been directed to the cathode."¹ The following is the explanation of the phenomena as given by Asch:¹

The existence of a charge on the bubbles and their motion is best explained by the Helmholtz-Lamb hypothesis of the electric "double layer." This treats the charging phenomenon as a parallel to electrification in frictional electricity. By this theory, two layers of charges are created at the boundary between the bubble and the surrounding liquid, molecules of one charge lining up on the vapor side matched with molecules of opposite

charge on the liquid side (see Fig. 1). The hypothesis further allows for the existence of "slip" between the two layers. In slipping apart, the positive charges in the liquid are transferred away to adjoining molecules while the bubbles, lacking such continuity of their phase, retain their negative charges and are attracted by the anode.

The second experiment consisted of a similar arrangement; however, the wire was placed between circular-disk electrodes and the peak heat flux was measured for various applied potentials.

Other publications have questioned the conclusions of this work. In a review, Jones² states that "Asch's observations of bubble motion were confined to regions of extremely weak electric field gradient, so his comparison of 'electrophoretic' and 'dielectrophoretic' forces for nucleate boiling is somewhat questionable. Further, the paper did not recognize the possible presence of bulk electroconvection, which could cause bubble motion." In a more recent review of Asch's results, Marco and Grassi³ stated ". . . some other masking effect [other than a net electrical charge] might be present, such as electroconvection."

Based on the results from another experiment performed by the present authors it was found that, among other things, the conclusions of Asch were in error due to the improper characterization of the electric field distribution near the heater wire. Therefore, a replica of the experiment used in the original 1966 article was constructed. The results from these tests show that the overall problem is extremely complex and that there are competing interactions from both dielectrophoresis and electrophoresis.

^{a)}Electronic mail: chung@mme.wsu.edu

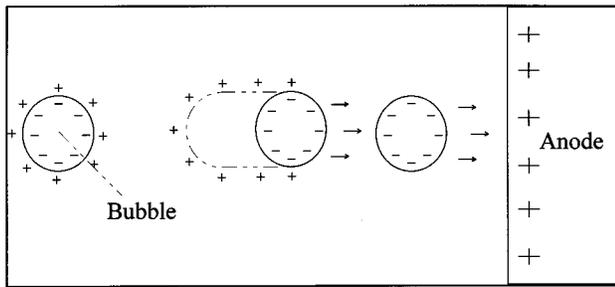


FIG. 1. Electric double layer analysis used in Asch's original work.

II. THEORY

The electrohydrodynamic (EHD) force that is produced by an electric field acting on a unit volume element of dielectric has no unique formulation. One generally accepted expression is (Jones,² Marco and Grassi,³ Stratton⁴)

$$\mathbf{F}^e = \rho_f \mathbf{E} - 0.5 |\mathbf{E}|^2 \nabla \epsilon - \nabla \left(\frac{1}{2} \rho \left(\frac{\partial \epsilon}{\partial \rho} \right) |\mathbf{E}|^2 \right), \quad (1)$$

where \mathbf{F}^e is the force per unit volume, ρ_f is the free charge density, \mathbf{E} is the electric field, ϵ is the dielectric permittivity of the dielectric fluid, and ρ is the dielectric density. The first term is the force per unit volume on a medium containing free electric charge of density ρ_f . The direction of this force depends on the polarities of the free charge and the direction of the electric field. The second term describes the force exerted on a dielectric fluid due to spatial gradients in the permittivity. Since ϵ is not differentiable at a liquid-vapor interface, it is appropriate to formulate directly the dielectrophoretic force acting on a dielectric spherical vapor bubble or liquid droplet in a two-phase mixture. The resulting DEP force for a vapor bubble is given by Pohl⁵ and Jones and Bliss⁶

$$\mathbf{F}_{\text{DEP}} = 2 \pi R^3 \epsilon_o \epsilon_l \left(\frac{\epsilon_v - \epsilon_l}{\epsilon_v + 2 \epsilon_l} \right) \nabla (|\mathbf{E}|^2), \quad (2)$$

where R is the radius of the bubble and ϵ_l and ϵ_v are the relative permittivity of the liquid and vapor, respectively. The direction of this force is such that the medium with relatively smaller permittivity will be driven away from regions of high electric field towards regions of low electric field. Therefore, for both R-113 and FC-72, the vapor phase will move away from the higher field regions while the continuous liquid phase will be drawn into these regions. The third term in Equation (1) describes electrostrictive effects and is important when considering application of a nonuniform electric field to a compressible dielectric. The exact physical significance of this term for fluids has been the subject of some conjecture^{2,3,7} and has been likened to an electrical pressure. Since the compressibility of a fluid is small, the electrostriction effect has no practical influence on hydrodynamics for an incompressible flow.^{2,3}

Equations (1) and (2) have been used extensively in the analysis of EHD induced phenomena for boiling.^{2,3} These equations show that high-voltage electric fields affect both the bulk fluid flow (electroconvection) and the movement and growth of bubbles (in addition to many other aspects of

the ebullition cycle). Equation (2) has been used successfully to determine dielectrophoretic force on vapor bubbles.⁶ Either, or both, of the first two terms in Equation (1) can be significant in controlling the resulting electroconvective flow.

III. EXPERIMENT

The experiments performed here are almost identical to that of Asch.¹ A rectangular boiling chamber (12.5 cm × 7.5 cm × 12.5 cm) was fabricated from 1.27 cm (0.5 in.) Plexiglas and was vented to the atmosphere. For the first experiment (the visualization experiment), two 1.27 cm (0.5 in.) brass-ball electrodes were mounted at opposite ends of the chamber and were separated by 6.02 cm (2.38 in.). A platinum wire 3.17 cm long and 0.025 cm in diameter ($\approx 0.09 \Omega$) was placed at one of three locations between the ball electrodes. The first and second locations were 1.27 cm (0.5 in.) from the left and right electrodes (similar to Asch), and the third was midway between the electrodes. Asch used a BWG 28 chromel wire that had a diameter of 0.035 cm, a resistance of 4.113 Ω/ft , and an unknown length. However, from the pictures provided, it appeared to be approximately 3 to 4 cm long. The wire was electrically heated with a dc power supply. "Alligator clips" similar in dimension to those observed in the original work were used to hold and position the platinum wire within the boiling chamber. The voltage across the heater was measured at the connection to the clips, and the current was also recorded. This allowed the calculation of the mean heat flux from the platinum wire (neglecting the small conduction losses due to end effects). For the figures shown later, a current between 8 and 10 amps was run through the wire which produced a sufficient amount of vapor for visualization. In addition, the power was varied over the complete range of the boiling curve ($\approx 10\text{--}35 \text{ W/cm}^2$), and it was verified that the observed results were not dependent on the amount of power supplied (Asch apparently used about 15 to 30 W/cm^2). Various dc high-voltage potentials were applied to the ball electrodes while the motion of the vapor generated from the wire was filmed.

The second experiment was similar to the first except the ball electrodes were replaced with 2.54 cm (1 in.) circular-disk electrodes spaced 2.54 cm (1 in.) apart. The platinum wire was placed between the electrodes and the maximum heat flux was measured for various dc high-voltage potentials applied across the electrodes. For most of the tests the bulk fluid temperature was the same as ambient conditions ($25 \pm 2 \text{ }^\circ\text{C}$), however, it was verified that similar trends were obtained using different subcoolings. The bulk fluid temperature in Asch's experiments was not given. Spectrophotometric grade R-113 was used as one of the working fluids, since this was the fluid of the original work, and FC-72 was chosen as the second. Both fluids are highly insulating dielectrics and have boiling points of 47.6 $^\circ\text{C}$ and 56 $^\circ\text{C}$, respectively, at standard atmospheric conditions. The relative permittivity of the liquid and the vapor for R-113 and FC-72 are $\epsilon_l = 2.44$, $\epsilon_v = 1.01$, and $\epsilon_l = 1.76$, $\epsilon_v = 1.00$, respectively. In the original work of Asch there is no mention of degassing procedures; however, the high solubility of gases in both these fluids is well established. Therefore, two separate tests

were performed: one without degasing and a second with the fluid degased by boiling vigorously at low pressure while venting through a reflux condenser.

IV. RESULTS AND DISCUSSION

It appears that Asch was mistaken in his analysis of the electric field distribution near the heated wire. Specifically, Asch states “in visualizing the general pattern of [electric] flux lines between two ball electrodes one can well recognize that field intensity increases with proximity to the electrodes.”¹ However, Asch apparently did not take into account the potential of the electrically heated wire. This potential is dictated by the power supply used to heat the wire and, assuming a grounded power supply, is generally very small with respect to the applied high-voltage field. In the experiments the wire was placed between the electrodes at points where, if the wire were not present, the potential would be several thousand volts. Since the potential of the wire was fixed by its power supply, this caused a local disturbance of the field that resulted in a very large field gradient which, in turn, produced large dielectrophoretic forces. Therefore, the electric field in the fluid depended not only on the ball electrodes, as Asch assumed, but on the entire system of conductors (including the wire itself). It is not completely clear from the original manuscript what the potential of each ball electrode was. It was stated for the visualization experiment that a 30 kV difference was used, and for the heat flux experiment that the potential difference was varied between zero and 50 kV. It is therefore assumed that one ball electrode, identified by Asch as the cathode, was at ground potential; the other was at the stated positive potential; and the heated wire was at a potential near that of the cathode.

For simplicity, we will refer to any conductor at a potential between +20 V and -20 V as grounded (thus, the heater wire is always described as grounded). If an electrode has a potential significantly greater than that of the heater wire, it will be referred to as positively charged. Conversely, if an electrode has a potential significantly less than the heater wire, it will be referred to as negatively charged. When one of the electrodes is grounded, the other will be described as charged (whether positively or negatively charged).

Our experiments showed that results similar to Asch's can be obtained by grounding the left ball electrode and applying +30 kV to the right ball electrode, i.e., the flow was towards the anode. However, grounding the right electrode and placing -30 kV on the left electrode we found that the motion of the vapor is towards the cathode. Repeated runs with both fluids showed the general trend that, for one ball electrode grounded and the heater wire grounded, the bubbles moved towards the charged ball electrode regardless of its polarization or the location of the wire in the boiling chamber. Sample results are shown in Fig. 2 for R-113. Although the vapor flow to the charged electrode was not as strong for FC-72, similar trends were observed.

Due to the large subcooling, the vapor flow towards the charged electrode was easier to visualize when the fluid was not degased. In fact, given that Asch did not mention a degasing procedure and, in view of the pictures from the original manuscript, it appears that Asch did not degas his fluid.

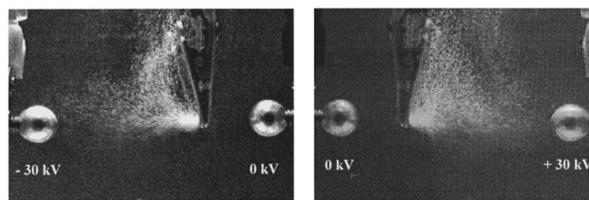


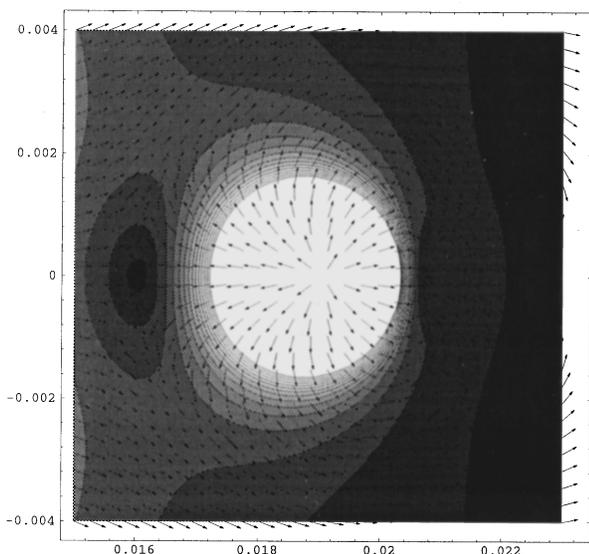
FIG. 2. Dominant vapor trajectory towards the anode or the cathode, i.e., towards the electrode with potential different from that of the wire itself.

Therefore, much of what he describes as “vapor” was actually some mixture of air and vapor. This can also be recognized by the fact that the very small diameter bubbles did not quickly condense. Vapor bubbles can recondense extremely rapidly (the rate being dependent on the subcooling level). On the other hand, while dissolved gas will come out of solution quickly when heated at the wire surface, it takes the gas a considerably longer time to diffuse back into solution. We found similar bubble flow patterns for the degased and nondegased tests (air has approximately the same permittivity as FC-72 and R-113 vapor and would be expected to experience approximately the same dielectrophoretic force). For simplicity we will refer to any air bubble, vapor bubble, or air/vapor mixture as simply the vapor, as it was impossible to determine any relative contributions except for the degased tests in which all the bubbles are composed of the vapor phase of the respective fluid.

There are two phenomena which, considered together, are key to understanding the observed results. The first is the force which acted to detach the vapor bubbles with extremely small sizes from the heater surface as observed in both Asch's work and the present work. Asch¹ correctly stated in the original manuscript that “it was apparent that the forces of the electric field were acting to detach the bubbles from the heated surface early in their process of growth.” It is the origin of the force very near the wire, which acted to detach the bubbles, that is in dispute. The second phenomenon came into play away from the wire, after the bubble detached, and controlled the vapor bubbles trajectories.

A numerical solution would be required to accurately find the dielectrophoretic force present in this experimental setup. This is due, in part, to the proximity of the alligator clips to the region of interest along the heater wire and the finite length of the wire. However, given the small diameter of the wire and the complexity of the shape of the clips, a complete model of the experiment is not feasible. Alternately, to gain a better insight into the qualitative behavior of the system, the dielectrophoretic force in a two-ball system was studied. In this simplified model, one ball had a radius equal to that of one of the electrodes (1.27 cm) while the other had a radius equal to that of the heater wire (0.025 cm). An analytic solution to the dielectrophoretic forces present in this system is easily obtained and serves to illustrate the behavior one might expect in the more complicated experimental setup. Figure 3(a) shows the resulting DEP force distribution and Fig. 3(b) shows the resulting DEP+buoyancy force distribution in a 4 mm by 4 mm window with the

(a) DEP Forces



(b) DEP + Buoyancy Forces

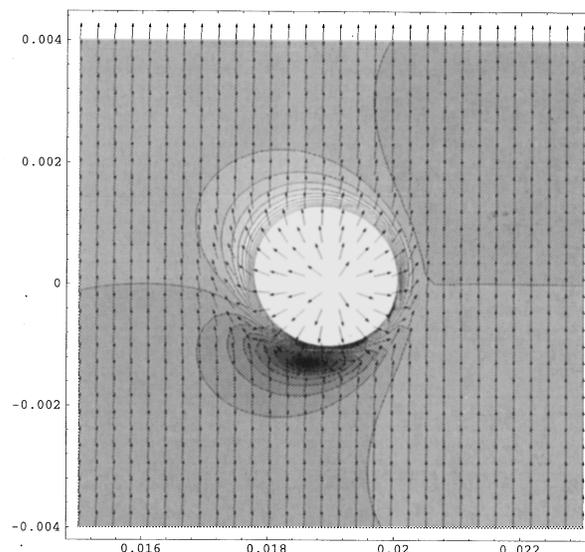


FIG. 3. (DEP) and (DEP + buoyancy) forces for the simplified two-ball system. The figures represent a 4 mm by 4 mm window around the small-ball electrode (the wire) located 1.27 cm from the diameter of the large ball electrode (the charged ball electrode).

heater wire located at the center of the window and R-113 as the working fluid. The center of the small ball was located 1.27 cm away from the diameter of the large ball and the potential of the large ball was 30 kV while the small ball was grounded. This geometry and potential roughly correspond to the first wire position in the ball-electrode visualization study described earlier and provides a qualitative view of the relative magnitudes of the DEP and buoyancy forces in the vicinity of the wire. The arrows show the direction and the contour plots represent ranges of force magnitudes. For simplicity, the magnitudes were normalized by the buoyancy force. In both Figs. 3(a) and 3(b), there are 21 evenly distributed contours. These contours span the range between 0 and 0.4 g for Fig. 3(a), and between the range 0 to 2 g for Fig. 3(b) (black representing zero for each plot). Therefore, within the white section the forces are larger than 0.4 g and 2 g for the two plots, respectively. In fact, it was found that the DEP force increased rapidly in this region near the small ball and was on the order of 90 000 times buoyancy at the surface of the small ball. Because the DEP force is independent of the polarity of the field, these plots represent either a positive- or negative-charged electrode. Also, similar trends were observed when the wire was placed at the second and third positions in the ball-electrode visualization study, i.e., midway between the two ball electrodes and at 1.27 cm away from the right ball electrode. From these results, it appears that the electric field and electric-field gradients are intense near the extremely small radius wire and resulted in very large dielectrophoretic forces on the vapor bubbles. This is consistent with previously published results for EHD enhanced boiling when a small diameter wire serves simultaneously as an electrode and the heat source.^{2,3} Away from the wire, in the regions where buoyancy and DEP forces were comparable, there exists a region below the wire where the DEP and buoyancy forces compete, resulting in a rela-

tively smaller total force; above the wire the two forces add to produce a larger force. Most importantly, it is obvious that the DEP force is stronger towards the charged electrode and the combined effects from DEP and buoyancy forces result in a dominant force up and towards the charged electrode. A comparison of Fig. 2 with Fig. 3(b) reveals that the combined DEP and buoyancy forces, at least qualitatively agree with much of the observed results.

In support of his conclusions concerning the dominance of electrophoresis, Asch cites Pohl⁸ who states that dielectrophoresis “generally requires a large difference in dielectric constant between solvent and solute, $(\epsilon_l - \epsilon_v) = 2-100$.” Pohl⁵ further states that “dielectrophoresis usually requires a substantial difference in the relative permittivities of the particle and the surrounding medium, e.g., $(\epsilon_l - \epsilon_v) \geq 1$.” This criterion is satisfied with R-113 and not FC-72; however, dielectrophoretic phenomena has been observed for both these fluids with electric fields and field gradients much smaller than those of the present study. Pachosa⁹ used a pair of diverging plate electrodes to show that bubbles can be moved against gravity for both R-113 and FC-72. Pohl⁵ also states “dielectrophoresis usually requires quite divergent fields for strong effects and requires relatively high field strengths (10^4 V/m for media of low dielectric constant as in the case of R-113 and FC-72).” These results leave no doubt that, given the magnitude and gradient of the electric field in this study, both of these fluids will be strongly affected by dielectrophoresis as represented in Fig. 3. It was observed for both fluids that the bubbles near the heater wire moved radially outward (even if only for a short distance), and only moved towards the charged electrode after detachment.

By setting one ball electrode to -15 kV and the other to $+15$ kV, a 30 kV field is present between the ball electrodes. If the wire is placed at the midpoint between the two balls,

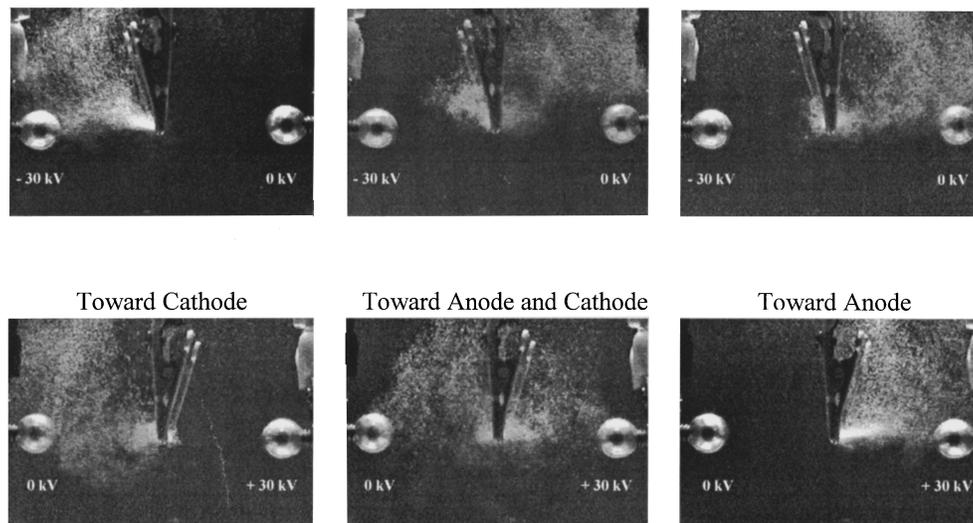


FIG. 4. Vapor trajectories toward the anode or the cathode or both.

the DEP forces should be very small. At this location the potential from the ball electrodes is nearly zero (approximately equal to that of the wire) and the electric field and electric-field gradients are small (given the small radius of the heater wire used in this study, it is reasonable to assume that when the grounded heater wire is centered at a point where the potential is zero the DEP force due to local disturbance of the field is essentially zero). This was the exact result obtained in our experiment. For this case, the bubbles all consistently move up under the action of buoyancy with sizes similar to the tests without the electric field, i.e., there was no noticeable difference when the electric field was turned on.

Consistent with Asch's results, increases in heat flux as high as threefold were measured (over that obtained when no field was present) when the heated wire was placed at a location where there was a significant difference between the potential established by the disk electrodes and that of the wire itself (for example, 0.127 cm away from the negative- or positive-charged disk electrode with the other disk electrode grounded). However, when the heated wire was located where the potential established by the electrodes was nearly zero—one disk electrode had a negative potential, the other had a positive potential, and the wire was located at the point of zero potential between the electrodes—there was virtually no change in the measured heat flux.

From the experimental results and this simplified analysis, all evidence supports the conclusion that the dielectrophoretic force was the dominant force controlling the detachment of extremely small bubbles for both R-113 and FC-72. The DEP force distribution also appears to account for much of the observed vapor flow pattern (up and towards the charged electrode). However, the results from the experiments were complicated by two other observed phenomena. First, as shown in Fig. 2, vapor was also observed to continue to move towards the charged electrode at points where the DEP force should be opposing the vapor movement and the buoyancy force would cause them to rise. In our experiments the EHD-induced secondary motions created a con-

vective flow towards the charged electrode and this could have carried the vapor bubbles towards the charged electrode. In fact, this is the explanation suggested by Jones² and Marco and Grassi.³ However, because of the extremely large electric field and electric-field gradient near the wire, and the fact that bubbles formed on and emanated from the heater wire (which was essentially one of the electrodes), it is difficult to discount the possibility of free charge forces (electrophoretic effects) on the vapor bubbles in accordance with Asch's observations. The wire could have provided a source or sink for charge while the large electric field provided the potential for charge movement. The fact that the bubbles, which appeared to act as charge carriers, are capable of either acquiring charge (when moving towards the negative electrode) or giving up charge (when moving towards the positive electrode) cannot be ignored. In fact, this charging mechanism, commonly known as induction charging, is one of the two principal methods of applying electric charge to objects (the other being corona charging). In induction charging the electrostatic field induces a surface charge on a dispersed phase which, lacking continuity of its phase, can then travel to and charge an object (in this case the object is the charged electrode). In addition to the explanation given previously by Asch,¹ the bubbles may exchange charge with the heater wire during the growth/detachment stage. Also, after being removed with extremely small sizes by extremely large DEP forces, the vapor bubbles experience small DEP and buoyancy forces away from the wire since these forces are proportional to the bubble volume. The drag force is also small on a small diameter vapor bubble.¹⁰ Therefore, after detaching, the bubbles trajectory could be dominated by small amounts of charge and would move freely towards the charged electrode as observed.

As shown in Fig. 4, the second phenomena we observed was that, for any individual boiling test, the vapor may move in either direction and sometimes moved in both directions simultaneously; however, the dominant direction was towards the charged electrode. As the wire was placed closer to the charged electrode, the system was more stable and

quickly settled on vapor moving towards the charged electrode. Note that when a vapor bubble flow is established towards the charged electrode, the resulting vapor itself can distort the field near the heater wire. This may result in the dominant vapor bubble flow towards the charged electrode being overtaken by a flow in the opposite direction towards the grounded electrode. In a similar manner, after the flow is established towards the grounded electrode, and the vapor towards the charged electrode has been removed by buoyancy and EHD effects, the original dominant flow towards the charged electrode can be reestablished. Again the very small DEP, buoyancy, and viscous drag forces on the small vapor bubbles could have contributed to this observed instability.

V. CONCLUSIONS

In Asch's article generalizations were made in support of observed experimental results which we have shown to be incorrect. Concerns about the interpretation of the results in this work have been raised in at least two review papers.^{2,3} The main error in Asch's interpretation was probably caused by neglecting the wire itself as an electrode. We found that the electric field in the vicinity of the grounded heater wire created intense dielectrophoretic forces, which explains the observed small bubble detachment diameters. Asch's statement that the bulk vapor flow was always towards the anode was also shown to be incorrect. We observed that the bulk vapor flow was dominantly towards the electrode with the potential different from that of the wire itself.

Using a simplified analysis, it was shown that much of the observed vapor flow was qualitatively consistent with combined effects from buoyancy and the dielectrophoretic force produced in the vicinity of the wire. However, the dielectrophoretic forces on the vapor bubbles did not suffi-

ciently explain the tiny bubbles observed to move all the way to the charged electrode. These small diameter bubbles were effected by the electroconvective bulk liquid flow. In addition, the bubbles appeared to act as charge carriers, apparently exchanging charge during the ebullition detachment process and/or during their trajectory. This was further justified by the fact that after detachment the dielectrophoretic, buoyancy, and viscous drag forces were very small on the extremely small bubbles. Therefore, away from the wire, the bubbles could be dominated by a very small electrophoretic force. This, coupled with the fact that the bubbles themselves could have distorted the electric field distribution, resulted in a non-periodic oscillatory vapor flow. However, the dominant flow direction was towards the charged electrode and was strengthened as the wire was placed closer to the charged electrode. Finally, while we have corrected some mistakes made in the past, the observed hydrodynamics appear to be controlled by complex interactions between numerous and competing EHD forces on both the liquid and vapor and further research into this problem is required to determine the relative contributions of the various forces and the resulting instabilities due to their interactions.

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