Dielectric constant of Ne near its liquid-vapor critical point

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We report high-resolution measurements of the static dielectric constant ϵ of neon near its critical point along nine isochores with densities ranging from $0.85\rho_c$ to $1.15\rho_c$, ρ_c being the critical density. These measurements were conducted to search for the predicted divergence in $(\partial\epsilon/\partial T)_{\rho_c}$ near T_c. We did not find any anomaly in the dielectric constant in either the one-phase ($T > T_c$) or the two-phase ($T < T_c$) regions within our experimental uncertainty of $\delta\epsilon = 3\times10^{-7}$ and 5×10^{-6}, respectively.

I. INTRODUCTION

The Clausius-Mossotti (CM) equation and the companion Lorentz-Lorenz (LL) equation allow high-resolution determination of the fluid density, especially near the critical point, by measuring the dielectric constant ϵ or the refractive index n. According to the CM equation,

$$\frac{\epsilon-1}{\epsilon+2}=\frac{4\pi}{3}\rho\theta_0 \ , \tag{1}$$

where ρ is the density and θ_0 is the polarizability of an isolated atom. It is well known that this equation is not exact[1]; away from the critical point there are small correction terms. These correction terms are generally expressed as density-dependent polarizability. If these correction terms are included, the dielectric constant is still expected to be constant for a sample of constant density over a narrow temperature range near T_c.

Recently the validity of this equation near the critical point has been questioned. The derivations of the CM and LL equations are based on the assumption that the medium is homogenous, a condition not satisfied near the critical point where the density fluctuation is significant. Various groups[2–5] have developed theories predicting the functional form of the deviation for nonpolar fluids. Stell and Høye found that the static dielectric constant should show the singularity of the internal energy; i.e., along the critical isochore,

$$\epsilon(\omega)-\epsilon_{\rm CM}=A\theta_0^2\omega^2 f(k\xi)+B\theta_0^3 t^{1-\alpha} \ , \tag{2}$$

where $\epsilon_{\rm CM}$ is the CM contribution to $\epsilon(\omega)$, θ_0 is the molecular polarizability, α is the specific-heat exponent, ξ is the correlation length, t is the reduced temperature, and $k=\omega/c$. In the low-frequency limit,

$$\epsilon(0)-\epsilon_{\rm CM}=B\theta_0^3 t^{1-\alpha} \ . \tag{3}$$

Bedeaux and Mazur[3] found that the dielectric constant behaves as,

$$\epsilon(\omega)-\epsilon_{\rm CM}=C\omega^2\psi(k\xi)+D\epsilon^{-1} \ .$$

Therefore, in the low-frequency limit

$$\epsilon(0)-\epsilon_{\rm CM}=D\xi^{-1}\propto t^{\nu} \ , \tag{4}$$

since the correlation length ξ diverges with exponent ν.

Anomalous behavior of the real part of the refractive index near T_c were first reported by Hocken and Wilcox[6] in their light-scattering experiment; their results are not in agreement with either of the two existing theorems. The anomaly was found to be roughly ten times too large. Hocken, Horowitz, and Greer[7] found a very large anomalous change of the static dielectric constant $\delta\epsilon$ in SF_6. They found $\delta\epsilon$ to be more than ten times the size predicted by Bedeaux and Mazur[3]; they also found the behavior of the slope $(\delta\epsilon/\partial t)_{\rho_c}$ to be more complicated than that of a simple power law. Doiron and Meyer[8] carried out a high-precision capacitance measurement in He^3 near the critical point. Doiron and Meyer, however, were not able to detect any anomaly within their experimental resolution. Although they did see a change in the measured dielectric constant ϵ, this change was highly dependent on the ^4He bath temperature, which affected the temperature of the needle valve that was used to shut off the sample volume. At a bath temperature of 3.5 K, a change of ϵ no more than 1.5×10^{-7} was observed at a reduced temperature of $t=(T-T_c)/T_c=1.5\times10^{-4}$. At a bath temperature of 4.2 K, however, the change in ϵ was found to be about 5×10^{-6} at the same reduced temperature.

Anomaly in dielectric constant and refractive index near the liquid-liquid consolute point has also been proposed in theories[9] and reported in a number of experiments.[9,10] But the experimental evidence is not conclusive at this point.[9,11]

The experiment to be described here aimed to clarify several discrepancies and uncertainties that exist among the various experiments and theories on nonpolar fluids.

II. EXPERIMENTAL SETUP

The sample cell is situated inside a copper vacuum can that is immersed in liquid helium. Two copper heat shields at temperatures intermediate to 4.2 K and the critical temperature of neon (44 K) are used to enclose the sample cell inside the vacuum can. The 4.2 K vacuum can, heat shields, and the sample cell are connected in stages through weak thermal links (0.003 cm thick and 0.5 cm wide copper foils.) The temperature of the inner (close to sample cell) heat shield is regulated with a carbon resistor thermometer-heater servo system. It is maintained to within 5 mK at 35 K. The temperature of the outer heat shield is not regulated; its equilibrium temperature is around 16 K and has drifts on the order of 25 mK. All electrical leads from outside the cryostat to the sample cell are anchored at each of the two shields. The copper sample filling capillary (o.d. 0.08 cm, i.d. 0.025 cm) from outside the cryostat terminates inside the inner heat shield, and it is connected to the low-temperature needle valve with a small copper-nickel capillary with an inner diameter of 0.01 cm. The lower end of the copper capillary is weakly attached to the inner heat shield. We estimate the temperature of the copper capillary at the lower end to be about 60 K. At this temperature the sample is prevented from freezing during filling.

A. Sample cell

The sample cell is made of oxygen-free high-conductivity (OFHC) copper (Fig. 1). The two active plates are held in position and insulated from the cell body with epoxy and thin (0.003 cm) mylar strips. The diameter and the height of the capacitor gap are 2.4 and 0.086 cm, respectively, giving a volume of 0.390 cm^3. Care has been taken to limit the volume available to the fluid outside the cell; the volume of low-temperature needle valve and the 0.01 cm i.d. capillary connecting the cell and the needle valve are 0.3% and 0.05% of the cell volume, respectively. We have also been careful in limiting the volume outside the capacitor gap within the sample cell. In the present design this excess volume lies entirely on the side of the indium O ring, "above" the capacitor gap. In this cell the excess volume is approximately 3% of the cell volume, and we shall see that this is responsible for most of the observed instrumental effect. In order to understand and separate this effect, we have performed experiments with the sample cell in both

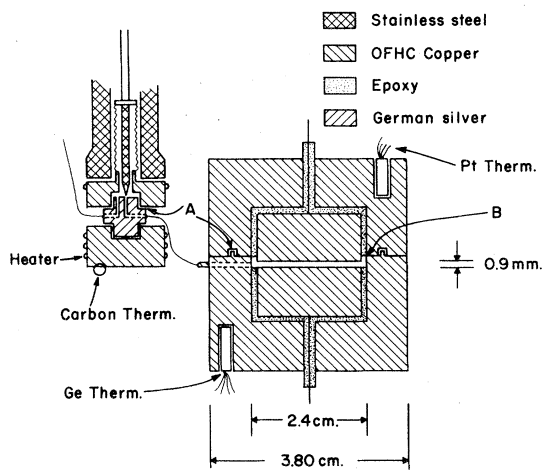

FIG. 1. Sample cell and needle valve. (a) The valve seat is made of German-silver. (b) 3 cm long, 1×10^{-2} cm i.d. Cu-Ni capillary is used to connect needle valve with sample cell. (c) Valve-seat system is regulated at temperature far from T_c with carbon thermometer and heater system. (d) Letter A indicates positions of indium O ring. (e) Letter B indicates position of possible excess volume. (f) The spacing for epoxy between the capacitor plates and sample-cell wall is greatly exaggerated for clarity; the actual spacing is 0.005 cm.

the upright (indium O ring above capacitor gap) and the inverted positions (indium O ring below the capacitor gap). We shall use this convention throughout this paper. The capacitor plates and the bottom and top side of the sample cell were machined and polished so that they are parallel to each other to within 0.5°.

B. Low-temperature needle valve

The most important difference of this experiment and that of Doiron and Meyer on He^3 is that the low-temperature needle valve used to shut off the sample in this experiment is regulated at a temperature far away from T_c. In Doiron and Meyer's experiment their needle valve is thermally anchored to the sample cell. It is, however, also weakly connected to the helium bath through the stem of the needle valve. The heat leak from the helium bath creates a small but important temperature gradient between the valve and the sample cell. As T_c is approached, the fluid becomes highly compressible, so that a small temperature gradient causes a non-negligible amount of fluid flow between the sample cell and the needle valve.[8] Doiron and Meyer estimate this effect accounts for all the observed changes in the dielectric constant $\delta\epsilon$. They found $\delta\epsilon$ to be dependent on the temperature of the helium bath and changes from 2×10^{-8} to 5×10^{-6} at $t = 1.5 \times 10^{-4}$ as the bath temperature is varied from 3.5 to 4.2 K.[8]

In our experiment the needle valve volume is approximately 0.3% of the sample-cell volume. The temperature of the needle valve is regulated at a constant value far away from T_c (typically at a temperature of 2 K above T_c) by means of a carbon resistance-thermometer bridge and servo heaters. This arrangement maintains a constant amount of fluid in the valve and therefore a constant amount of fluid in the sample cell even when the temperature of the sample cell is varied near T_c. The amount of fluid that flows in and out of the capillary leading to the needle valve is negligible: The total volume of the capillary is only 0.05% of that of the sample cell, and only a small fraction of that contains fluid of large compressibility.

C. Temperature measurements and control of sample cell

The temperature of the sample cell is measured with two "4" leads ac resistance bridges using germanium and platinum resistors as sensors (Fig. 2). The bridge is balanced when the ac ratio standard[12] is set at a value equal to the ratio of the sensor resistor to that of the standard resistor. The unbalanced signal from the lock-in is used to drive a commercial temperature controller[13] that supplies current to the heater wound around the sample cell. The standard resistors are made of bifilarly wound Evanohm wires thermally anchored at the bottom of the copper vacuum can (4.2 K). The platinum resistor[14] does not have the necessary sensitivity in this temperature range; it is used primarily to cross calibrate the germanium resistor.[15] The platinum temperature scale is based on the IPTS-68 scale[16] with calibration points at 273.15 and 77.3 K. Both resistance thermometers have rather large shifts upon thermal cycling to room temperature. We estimate the critical temperature T_c of neon to be at 44.450 ±0.020 K. The relative temperature of the sample cell, however, can be determined with much higher precision. The precision of germanium thermometer bridge is 4 μK and the sample cell temperature can be held constant to within ±10 μK for at least 5 hours.

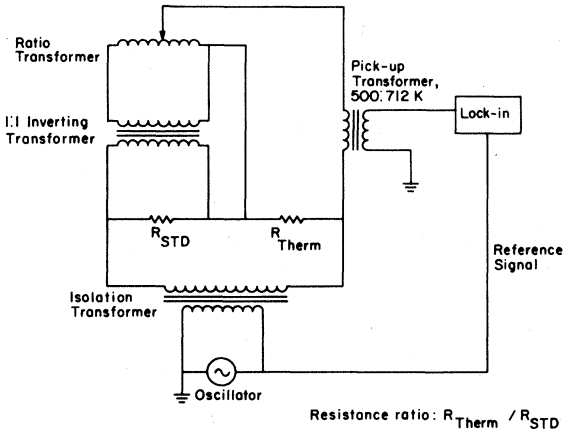

FIG. 2. Schematic for "4" leads resistance bridge.

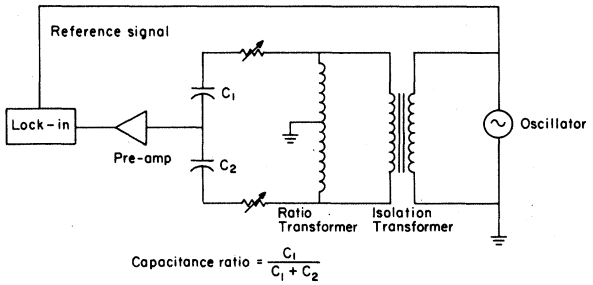

FIG. 3. Schematic for capacitance bridge.

D. Capacitance bridge

The dielectric constant is measured with a capacitance bridge set up similar to that used by Chan, Ryschkewitsch, and Meyer[17] (Fig. 3). This bridge measures a ratio (R_c),

$$R_c = \frac{C_{\text{sam}}}{C_{\text{sam}} + C_{\text{ref}}} , \tag{5}$$

where C_{sam} and C_{ref} are, respectively, the capacitances of the sample cell and reference capacitors. A number of reference capacitors were used in the various stages of this experiment; they include (i) a commercial 100-pf standard capacitance made by General Radio, (ii) a 7-pf silver-mica capacitor thermally anchored at the sample cell, and (iii) a parallel-plate capacitor in a separate experimental cell designed for a separate experiment. The capacitance bridge is normally operated with an excitation voltage of 16 V rms and a frequency of 1000 Hz. All connections to the various capacitors were made with coaxial connectors and cables. With the latter two reference capacitors, the resolution in the measured capacitance ratio, $\delta(R_c)/R_c$ is better than 1.4×10^{-8} at this temperature range.

III. EXPERIMENTAL PROCEDURE

At each sample filling, the sample cell is first cleaned by repeatedly filling it with neon gas and evacuated to low pressure (1×10^{-6} Torr) at room temperature. The neon gas used is supplied by Linde Specialty Gas and has a purity of 99.996%. The sample cell is regulated at 45 K during filling. The capacitance ratio with and without the sample is used to

find out the dielectric constant of the sample,

$$\epsilon = \frac{(R_{ce})^{-1} - 1}{(R_{cs})^{-1} - 1} \quad , \tag{6}$$

where R_{ce} is the capacitance ratio as defined in Eq. (5) with the sample cell empty and R_{cs} is the capacitance ratio with a filled sample cell. The density of the sample is determined by means of the Clausius-Mossotti equation. By comparing the measured dielectric constant and that of the "critical" dielectric constant, we can determine how close is the sample density from the critical density. The critical dielectric constant and the critical density for neon are determined in a separate experiment[18] in our laboratory. They are respectively,

$$\rho_c = 0.484 \text{ gm cm}^{-3} \pm 0.0005 \quad ,$$

$$\epsilon_c = 1.07250 \pm 7 \times 10^{-5} \quad .$$

The sample cell is shut off by closing the low-temperature needle valve when the desired sample density is obtained.

IV. RESULTS

Capacitance ratio as a function of temperature are determined by controlling the sample cell at each specific temperature. The equilibrium time ranges from less than 10 minutes far away from T_c ($t \geq 1 \times 10^{-3}$) to over 4 hours as T_c is approached. Figure 4 is a plot of the result of a run with $\bar{\rho} = 1.01\rho_c$ and the sample cell in the upright position. The needle valve, situated at 1.5 cm above the capacitor gap, is regulated at 47 K or $t = 6 \times 10^{-2}$. At temperatures sufficiently higher than T_c, we found the capacitance ratio changes linearly with temperature, with a temperature coefficient $\Delta R_{cs}/(R_{cs})\Delta T = 1 \times 10^{-4}\ \text{K}^{-1}$. The variation of capacitance are due to two effects: (i) Both the empty sample cell capacitance and the reference capacitance are temperature dependent. (ii) The pressure of the neon fluid sample which affects the capacitor spacing is also temperature dependent.

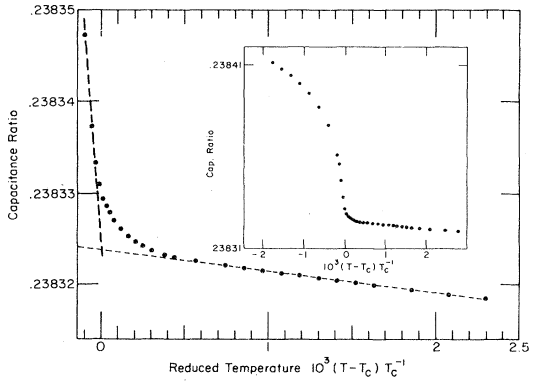

FIG. 4. Capacitance ratio vs temperature for a run with sample cell in "upright" position. Inset is a plot over a larger temperature range showing more clearly the different form in the one- and two-phase regions; T_c is determined by smooth extrapolation from $T > T_c$ and $T < T_c$. We estimate the uncertainty in $t = (T - T_c)/T_c$ to be 5×10^{-5}.

The simple linear relationship between the capacitance ratio and temperatures shown in Fig. 4 extends to $t = 1 \times 10^{-2}$ from 1×10^{-3}. We would expect this linear relationship to be applicable in the narrow temperature range of $-1 \times 10^{-3} < t < 1 \times 10^{-3}$ if the dielectric constant is not changing. The pressure of a fluid along an isochore is expected to change nearly linearly in the vicinity of T_c.

A clear deviation from the linear relationship near T_c is seen in Fig. 4, indicating that the measured dielectric constant in the capacitor gap is not constant. The rapid rise in the capacitance ratio (R_{cs}) is also an excellent signature of the critical temperature. The change in R_{cs} in the two regions is shown more clearly in the inset over a larger temperature range. The uncertainty in T_c is about 2 mK which corresponds to a reduced temperature of 4.5×10^{-5}. The position of T_c is also confirmed by the lengthening of the equilibrium time just below T_c.

Changes in the dielectric constant can be calculated from the measured capacitance ratio and the expected capacitance ratio according to linear extrapolation from high temperature,

$$\frac{\delta\epsilon}{\epsilon_N} = \frac{R_{cs} - R_{cn}}{(R_{cn})(1 - R_{cs})} \quad , \tag{7}$$

where R_{cn} is the expected or normal capacitance ratio, and R_{cs} is the actual measured capacitance ratio. The closed circles in Fig. 5 translate the observed capacitance ratio in Fig. 4 into apparent dielectric-constant changes. An increase in the measured dielectric constant of 3×10^{-5} is seen from $t = 5 \times 10^{-4}$ to $t = 0$ and a much larger change in ϵ is seen for $T < T_c$. The inset in Fig. 5 shows the change in ϵ more clearly in the one phase region.

We have considered two likely instrumental effects that may be responsible for the observed increase in $\delta\epsilon$ near T_c:

First, near T_c, the density of fluid is not homogeneous: The divergent compressibility in the gravitational field induces a density gradient along the capacitor gap for $T > T_c$, and the fluid separates into liquid and vapor phases below T_c. We have calculated the expected change in dielectric constant with density gradient from that of homogeneous fluid using critical amplitudes and exponents determined in our experiment on the equation of state.[18] We found this effect tends to *reduce* the effective dielectric con-

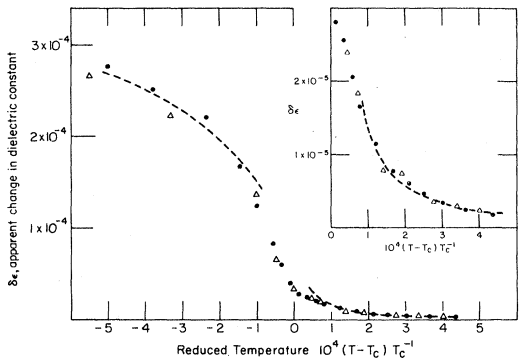

FIG. 5. Magnitude of the apparent change in dielectric constant, $\delta\epsilon$, as a function of reduced temperature. Closed circles, representing positive $\delta\epsilon$ are data obtained with sample cell in upright position. Open triangles, representing negative $\delta\epsilon$, are data obtained with sample cell in inverted position. Dash lines show the estimated $\delta\epsilon$ with a 3% excess volume not at the capacitor gap. The inset shows more clearly $\delta\epsilon$ for $T > T_c$.

stant for both $T < T_c$ and $T > T_c$ which is contrary to our measurement and that the magnitude of the expected change is only 2% of the observed $\delta\epsilon$. At $t = 1 \times 10^{-5}$, when the difference in density at top and the bottom of capacitor gap is 6% of the critical density, the effective dielectric constant is only 6×10^{-7} smaller than that of homogenous density. At $t = -1 \times 10^{-5}$, the *decrease* in ϵ is 3.2×10^{-6}, which is also small compared with the observed increase in ϵ.

Second, excess volume in the sample cell above or below the capacitor gap also causes changes in the measured dielectric constant in the presence of density gradient near T_c: The measured dielectric constant reflects the average density inside the capacitor gap and not the average density of the entire sample. The dashed line in Fig. 5 is an estimate of this effect assuming the excess volume corresponds to 3% of the total sample-cell volume, and it is at a height 0.17 cm above the midpoint of the capacitor gap. A simple model calculation of this effect is presented in the Appendix. We should point out that the 3% excess volume is possible with our cell design. An excess volume on this order is difficult to avoid since the top capacitor plate must clear the cell wall and that an excess amount of indium subtending a volume on top is needed to seal the sample cell (see Fig. 1). We can see from Fig. 5 that the calculated values are in excellent agreement with the observed $\delta\epsilon$ both below and above T_c. The agreement is not good inside $|t| \leq 5 \times 10^{-5}$ since we have not included the gravitational rounding effect in our calculation.

In order to separate this instrumental effect and any true anomaly in the dielectric constant we invert the sample cell and repeat the measurements. The excess 3% volume, if it exists, is now below the capacitor gap. In this series of experiments, the low-temperature needle valve is at the same height as the capacitor gap, and it is controlled at 47 K. The sample density is $1.02\rho_c$. The dielectric constant is found to *decrease* upon approach to T_c in the one-phase region and continue to decrease in the two-phase region. In order to compare with the result of the upright cell, we have plotted the magnitude of $\delta\epsilon$ as open triangles in Fig. 5. The observed decrease in ϵ is again consistent with the presence of a 3% excess volume. The magnitude of $\delta\epsilon$ is identical for the upright and inverted cell over the entire temperature range both above and below T_c. Had we added the change in ϵ in the two different cell configurations we would see an almost perfect cancellation in $\delta\epsilon$.

By inspection of Fig. 5, we may conclude that any critical anomaly in ϵ, if real, is less than 3×10^{-7} for $T > T_c$ and less than 5×10^{-6} for $T < T_c$. The larger uncertainty in $\delta\epsilon$ for $T < T_c$ reflect experimental scatters that may be caused by liquid droplets in the vapor side of the sample cell.

We have repeated the above experiment with the needle valve regulated at 45.5 K ($t = 2 \times 10^{-2}$) and 43.5 K ($t = -2 \times 10^{-2}$). Identical results were found.

We have also performed the above experiment with sample densities equal to $0.85\rho_c$, $0.97\rho_c$, $1.04\rho_c$ with the sample cell in the upright position and with sample densities equal to 0.95, 1.05, 1.08, and $1.12\rho_c$ with the sample cell in the inverted position. No anomaly in ϵ was found. The geometry-induced $\delta\epsilon$ as shown in Fig. 5 was found to be almost density independent for $|(\rho - \rho_c)/\rho_c| \leq 0.05$ and decreases rapidly for sample densities outside the 5% range. This is due to the fact that at densities far from ρ_c the density gradient of the fluid in and outside the capacitor gap is greatly reduced.

We have made measurements with the entire cryostat tilted at 6° from vertical. The measured $\delta\epsilon$ near T_c is only marginally different (about 10% smaller) from that shown in Fig. 5. This confirms our estimate that density gradient of the sample inside the capacitor gap is not the dominating instrumental effect.

Other experimental parameters varied includes the excitation voltage (1 and 16 V rms) and the frequency (100, 1000, and 2000 Hz) used in the capacitance bridge. No difference in the observed $\delta\epsilon$ from that shown in Fig. 5 were found.

V. CONCLUSIONS

We have carried out systematic high-precision studies of the dielectric constant of neon near the critical point. Our experiment excludes any anomalous change in ϵ larger than 3×10^{-7} for $T > T_c$ and larger than 5×10^{-6} for $T < T_c$. Our results in the one-phase region are in agreement with the result of Doiron and Meyer in He^3 and the most recent experi-

ments on SF_6 that are in progress at the National Bureau of Standards.[19]

These results are not in agreement with the earlier results on SF_6 by Hocken, Horowitz, and Greer.[7] Our results are not necessarily in contradiction with the theories of those presented by Stell and Høye[2] and Bedeaux and Mazur,[3] since they gave no estimate for the amplitude of the singular part of the dielectric constant.

Experiment on CO, a polar fluid, is currently in progress in our laboratory.

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APPENDIX

We wish to calculate the average dielectric constant of the fluid inside the capacitor gap as a function of temperature if there is an excess volume (3%) at a height 0.17 cm above midpoint of the capacitor gap. We shall make the following assumptions: (i) The excess volume V_2 is equal to $0.03\,V_0$, where V_0 is the total volume of the cell, connected through a narrow tube to the capacitor gap region of V_1, $V_1 = 0.97\,V_0$. (ii) Average density of the entire sample is ρ_c. (iii) The density of fluid in V_2 is different from that at V_1, the difference is described by critical parameters. (iv) There is no density gradient inside the capacitor gap itself, even in the two-phase region. This simplification causes changes in ϵ much smaller than the observed effect (see text.)

We shall calculate the expected $\delta\epsilon$ with the excess volume on top of the sample cell, or in the upright position.

One-phase ($T > T_c$) *region.* The average density in the excess volume V_2 is,

$$\rho_2 = \rho_c - [\rho_c g(\Delta h)/p_c]\Gamma(t)^{-\gamma}\rho_c \ .$$

where

$$\rho_c = 0.484 \text{ gm/cm}^3, \quad g = 980 \text{ cm/s}^2 \ ,$$

$$p_c = 2.72 \times 10^7 \text{ dyn/cm}^2, \quad \Delta h = 0.17 \text{ cm} \ .$$

and we have found $\gamma = 1.25$ $\Gamma = 0.02$ in our experiment on the equation of neon.[18]

$$\therefore \ \rho_2 = \rho_c - 5.929 \times 10^{-8}(t)^{-\gamma}\rho_c \ . \tag{8}$$

Once ρ_2 is determined, the density of fluid in the capacitor gap can be determined,

$$\rho_c V_0 = \rho_1(0.97)\,V_0 + \rho_2(0.03)\,V_0 \ . \tag{9}$$

The change in ϵ from that with no excess volume can be obtained through the Clausius-Mossotti equation,

$$\delta\epsilon = \frac{\rho_1 - \rho_c}{\rho_c}(\epsilon_c - 1) \ , \tag{10}$$

when $\epsilon_c = 1.0725$. Using Eqs. (8), (9), and (10) we found

$$\delta\epsilon = 3.16 \times 10^{-5} \quad \text{at } t = 5 \times 10^{-5} \ ,$$

$$\delta\epsilon = 1.33 \times 10^{-7} \quad \text{at } t = 1 \times 10^{-4} \ ,$$

and

$$\delta\epsilon = 7.48 \times 10^{-7} \quad \text{at } t = 1 \times 10^{-3} \ .$$

These results are plotted in Fig. 5.

Two-phase region ($T < T_c$). The average density in the excess volume is the vapor density, or

$$\rho_2 = \rho_c[1 - B(t)^{-\beta}] \ , \tag{11}$$

where $B = 1.72$ and $\beta = 0.35$.[18] Making use of Eqs. (9) and (10), we may again calculate the average density of the fluid and the change in dielectric constant in the capacitor gap region. Results are also shown in Fig. 5. Similar calculations can be carried out for an inverted sample cell. The decrease in ϵ is equal in magnitude to that of an upright cell at each temperature.

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