

Dielectric constant of liquid helium films

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The dielectric constant of a thin planar film of liquid He adsorbed on the surface of a solid is calculated as a function of the film thickness, the characteristic energy of the substrate, and the thickness of the frozen He layer. The effect was found to be observable and useful in the determination of the aforementioned parameters.

The principal parameter of a planar film of liquid helium adsorbed on the surface of a solid is its thickness l . The measurement of the thickness of thin films to within the monatomic-layer thickness (3.6 Å) remains a difficult task. In the analysis of the results obtained by different methods it is generally assumed that the film is uniform and that its dielectric constant is the same as that of the bulk material.

We will show that the dielectric constant of thin films of liquid He actually contains an appreciable nonuniform part, $\Delta\epsilon^B$, which depends on the properties of the substrate and the thickness of the frozen He, and that it decreases slowly, as $1/l$, with increasing l . The latter behavior accounts for the pronounced effect in films with l much greater than the thickness of the monatomic layer.

The appearance of a nonuniform contribution to the dielectric constant at distances from the substrate greater than the atomic dimensions stems primarily from the fluctuational long-wave electromagnetic field in the system. The contribution of the field to $\Delta\epsilon^B$ is proportional to the mean square of its nonuniform part. The latter quantity and hence $\Delta\epsilon^B$ are nonuniform along the length of the film and depend on l and the properties of the substrate.

The strictional origin of the effect under discussion and the "gas-like" nature of the density and dielectric properties of the liquid He help one understand an appreciable part of the effect in these films. Specifically, the compressibility of liquid He is $\beta_T \approx 1.2 \times 10^{-8}$ esu, which is 1 or 2 orders of magnitude greater than that of other liquids, and its striction is high. On the other hand, the customarily measured quantity $\epsilon - 1$ (ϵ is the dielectric constant of He in the volume) here is $\approx 6 \times 10^{-2}$ in comparison with unity for typical liquids. As a result, the measured relative deviation of the dielectric constant, $\Delta\epsilon^B/(\epsilon - 1)$, should be reasonably large.

Let us examine a film with planar interfaces at $x = 0$ at the boundary of a solid and at $x = 1$ at the boundary of a vapor. We take into account that the film is separated from the substrate by a layer of solid He of thickness $D_0 \sim 1-2$ atomic layers. We use the following model for the dielectric constant of a film $\epsilon(x)$: At $x < 0$, $\epsilon(x)$ is equal to the dielectric constant of a metal (ϵ_1), for $0 \leq x \leq D_0$ it is equal to the dielectric constant of solid He, and in the liquid part of the film with $D_0 \leq x \leq l$ $\epsilon(x) = \epsilon + \Delta\epsilon^B(x)$. We assume that for $x \geq D_0$ $\Delta\epsilon^B(x)$ is determined primarily by the

long-wave field, and the general theory of the van der Waals forces holds.¹

Because of the linear dependence of the dielectric constant of He on the density, we can write $\Delta\epsilon^B(x)$ as

$$\Delta\epsilon^B(x) = \frac{\epsilon - 1}{\rho} \Delta\rho^B(x), \quad (1)$$

where ρ is the density of the bulk He, and $\Delta\rho^B(x)$ is the density profile of the liquid part of the film which is determined by the van der Waals forces. For an arbitrary inhomogeneous insulator $\Delta\rho^B(x)$ is²

$$\Delta\rho^B(x) = \frac{T}{4\pi} \rho^2 \beta_T \sum_{n=0}^{\infty} \omega_n^2 \frac{\partial\epsilon(i\omega_n)}{\partial\rho} D_{II}(\mathbf{r}, \mathbf{r}; \omega_n), \quad (2)$$

where T is the temperature, $\omega_n = 2\pi nT$, n are integers, and $D_{II}(\mathbf{r}, \mathbf{r}; \omega_n)$ is the inhomogeneous part of the Green's function of a photon when the arguments are the same. For thin films ($l < 30-40$ atomic layers) the retardation and thermal effects can be disregarded. After a transformation in (2) from $\sum_{n=0}^{\infty}$ to $\int_0^{\infty} (d\xi/2\pi)$, we find from (1)

$$\Delta\epsilon^B(x) = \frac{(\epsilon - 1)\beta_T}{8\pi^2} \int_0^{\infty} \xi^2 \left(\rho \frac{\partial\epsilon(i\xi)}{\partial\rho} \right)_T D_{II}(\mathbf{r}, \mathbf{r}; \xi) d\xi. \quad (3)$$

In the calculation of the integral in (3) we restrict the analysis to the first-order terms in $\epsilon - 1$. In this approximation the liquid He-vapor interface does not contribute to D_{II} (Ref. 3), so that D_{II} can be written, as in the case of a two-phase system, liquid He-solid, in the form³

$$D_{II}(\mathbf{r}, \mathbf{r}; \xi) = \frac{\Delta(i\xi)}{2x^3 \epsilon(i\xi) \xi^2} \quad \Delta(i\xi) = \frac{\epsilon_1(i\xi) - \epsilon(i\xi)}{\epsilon_1(i\xi) + \epsilon(i\xi)} \quad (4)$$

where ϵ_1 is the dielectric constant of a solid. Substituting (4) into (3), we find

$$\Delta\epsilon^B(x) = \frac{(\epsilon - 1)\beta_T}{16\pi^2 x^3} \int_0^{\infty} d\xi \frac{\epsilon(i\xi) - 1}{\epsilon(i\xi)} \Delta(i\xi). \quad (5)$$

Since absorption in liquid He occurs at higher frequencies than in the substrate, we can write (5) in the following form in the approximation in $\epsilon - 1$:

$$\Delta\epsilon^B(x) = \frac{(\epsilon - 1)^2 \beta_T \hbar \bar{\omega}}{16\pi^2 x^3} \quad \hbar\omega = \hbar \int_0^{\infty} \frac{\epsilon_1(i\xi) - 1}{\epsilon_1(i\xi) + 1} d\xi \quad (6)$$

The dielectric properties of the liquid part of the film are determined by the function $\Delta\epsilon^B(l)$, which is the mean value of $\Delta\epsilon^B(x)$ over the film thickness

$$\Delta\epsilon^B(l) = (l - D_0)^{-1} \int_{D_0}^l \Delta\epsilon^B(x) dx. \quad (7)$$

From (6) and (7) we find

$$\Delta\epsilon^B(l) = \frac{(\epsilon - 1)^2 \beta_T \hbar \bar{\omega}}{32\pi^2 D_0^3} \left[\frac{D_0}{l} + \left(\frac{D_0}{l} \right)^2 \right] \quad (8)$$

within the error of third-order terms in the small parameter D_0/l . For $l \gg D_0$ the dielectric constant of the film thus decreases as $1/l$. For various solid substrates we have $\hbar\omega \sim 10^{-11} - 10^{-10}$ (Ref. 4), and for metals we have typically $\sim 3 \times 10^{-11}$. For l and D_0 in the atomic layers we have

$$\Delta\epsilon^B(l) \approx \frac{10^{-1}}{l D_0^2} \left(1 + \frac{D_0}{l} \right). \quad (9)$$

For the films with $l \sim 5 - 30$ we have $\Delta\epsilon^B(l) \sim 10^{-2} - 10^{-3}$, which is comparable with the measured value, $\epsilon - 1 \sim 6 \times 10^{-2}$. Because of this circumstance, and because of the particular dependence of $\Delta\epsilon^B(l)$ on l and D_0 , the effect under discussion must be taken into account in the measurement of the dielectric constant of thin films.

We assume that the He film is adsorbed at the metal surface, which doubles as one of the capacitor plates, where the spacing between the plates is $d \gg l$. The difference between the capacitances of the capacitor considered here $[C(l, d)]$ and the hollow capacitor $[C(d)]$ is, within the terms $\sim D_0/d$,

$$\frac{C(l, d) - C(d)}{C(d)} \approx 6 \cdot 10^{-2} \frac{l}{d} + \frac{10^{-1}}{D_0^2 d} + \frac{10^{-1}}{D_0 d l} \quad (10)$$

Ignoring $\Delta\epsilon^B(l)$, the difference under consideration is a linear function of l , which vanishes at $l = 0$. The second term in (10) accounts for the fact that the difference in capacitances is a linear function of l . This function does not, however, go through the coordinate origin. The third term, which is important only for extremely thin films, increases the divergence of the capacitance from its values for a homogeneous film. With $l \sim 10$ and $d \sim 10^3$, both of these terms are observable. Expression (10) can be written in the form

$$\frac{C(l, d) - C(d)}{C(d)} \approx 6 \cdot 10^{-2} \frac{\tilde{l}}{d}, \quad \tilde{l} = l + b, \quad b = \frac{10}{6D_0^2} \quad (11)$$

Here \tilde{l} is the film thickness which is determined by the capacitive method and which is greater than its true value (l) by the amount $b \sim 1 - 2$ atomic layers.

The second term in (9) can be observed by measuring the difference in capacitances of the capacitors with identical spacing between the plates and with film thickness l_1 and l_2 ,

$$\frac{C(l_1, d) - C(l_2, d)}{C(d)} = \frac{\epsilon - 1}{d} (l_1 - l_2)(1 - \sigma), \quad \sigma = \frac{10}{6D_0} \frac{1}{l_1 l_2} \quad (12)$$

For $l_1 l_2 \sim 10^2 - 10^3$ we have $\sigma \sim 5 \times 10^{-2} - 10^{-3}$ and the contributions to the capacitance, $\sim 1/l$, are observable.

In summary, if the nonlinear properties of liquid He (the striction) are taken into account, the fluctuational long-wave electromagnetic field of the substrate causes the dielectric constant of the film to be nonuniform and to depend on its thickness, on the characteristic energy of the substrate, and on the parameter D_0 . Because of the "gas-like" nature of liquid He, and because of the high accuracy of the capacitive measurements, the effect can be observed and used to determine the values of l , $\hbar\omega$, and D_0 very accurately.

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