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THE ELECTRIC CAPACITANCE OF A RIGID DIELECTRIC STRUCTURE

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Abstract—There is experimental evidence to support the idea that the inverse capacitance of very thin structures tends toward a constant value instead of zero despite the prediction of the classical linear theory (Mead, 1961, *Phys. Rev. Lett.* **6**, 545–546; 1962, *Phys. Rev.* **128**, 2088–2093). Motivated by Mindlin's explanation (1969, *Int. J. Solids Structures* **5**, 1197–1208) of this anomalous behaviour, the analysis of this effect is here revisited. Specifically, new boundary conditions are proposed and discussed, accounting for the metal–dielectric interface effect.

1. INTRODUCTION

In 1961 C. A. Mead observed a remarkable anomaly concerning the behaviour of very thin structures such as Ta-Ta_2O_5-Au and Ta-Ta_2O_5-Bi. Afterwards, Mead and Maserjian (1967) observed a similar anomaly in thin layers so composed: Al-TiO_2-Al. These structures, once arranged in plane parallel layers, behave like plane parallel capacitors where the Ta_2O_5 and TiO_2 play the roles of the dielectric materials. The classical prediction is that the capacitance should become infinite (and, of course, the inverse of the capacitance zero) as the distance between the electrodes approaches zero. Despite this prediction the experiments show that the inverse of the capacitance per unit surface tends toward a constant value which is approximately 0.5–5 μF cm^{-2} (Ku and Ullman, 1964; Mead and Maserjian, 1967). The explanation of such a phenomenon surely involves the metal–dielectric interface effect which becomes dominant for such short distances; Mead himself (1961, 1962) conjectured that the penetration of the electric field into the metal could be a possible explanation. The occurrence of such a phenomenon as the penetration is extraneous to the classical theory while it is foreseen by quantum mechanics and the estimated depth of penetration is of the order of one Å (Mead, 1961; Mott, 1936, 1982). Following Mead's conjecture Ku and Ullman (1963) stated the problem within the framework of the semi-classical solid state physics. They considered the free electron density in the metal, which is known by the Fermi–Dirac distribution function, and the corresponding energy known as the Fermi energy. They argued that the electron density and the corresponding energy were not uniform under the effect of an applied potential between the two faces of one of the electrodes. From the mathematical point of view they solved the Poisson equation within the metal and the Laplace equation within the dielectric, for the potential ϕ. They assumed the usual classical condition for ϕ at the outer faces of the metal electrodes and at the metal–dielectric interfaces, but the *unusual condition* at the metal–dielectric interface for the electric field $E = -\phi'$: this was required to behave as at the interface between two dielectric materials (Ku and Ullman, 1961). Finally, a formula for the capacitance was deduced by them making use of the solution found for ϕ. In this formula the inverse of the capacitance consists of the sum of two parts: one is the classical form, the second one a correction term independent of the distance. Afterward, Maserjian and Mead (1967) proposed a general theory for the electric conduction across thin layers of crystallized structures, a theory which is framed within the microscopic theory of semiconductors. In this broader context they also proposed the explanation of the anomaly of the capacitance.

There is a general agreement that a quantity such as the macroscopic dielectric constant is physically meaningful for short distances down to a tenth of an angstrom; one is then encouraged to revisit the problem within the macroscopic description. This is what Mindlin

(1969) did by re-stating the problem within the linearized theory of the polarization gradient for deformable bodies. He offered a brilliant answer to the problem of the anomalous capacitance and proposed a challenge to the experimentalists by showing the following: with reference to the solution he had found for ϕ and for the polarization field P, the inverse of the capacitance C_a^{-1} tends effectively toward zero with x_0, where $2x_0$ is the distance between the electrodes. But the graph of C_a^{-1} versus x_0, approaches to an asymptote as $x_0 \to \infty$. The intercept of the asymptote with the ordinate axis is positive. Mindlin (1969) concludes that it is very likely that the experimentalists had found the asymptotic behaviour of C_a^{-1}; in fact appreciable deviations of the function $C_a^{-1}(x_0)$ from the asymptote are quite negligible down to the interested distances (≈ 30 Å). It is worth noticing that the macroscopic theory of the polarization gradient takes into account the microscopic structure of the crystalline lattice of the dielectric, to some extent (Maugin, 1988; Mindlin, 1969, 1972). In this sense Mindlin's approach is not so far from Maserjian's and Mead's point of view. In the present note the problem is re-stated in a general form. The solution for the electric potential and for the electric field is found in terms of the polarization field P which is not specified at the beginning. With reference to this solution we define a quantity which seems not to play any role in the classical linear case. To this quantity we attribute the physical meaning of an extra charge at the metal–dielectric interface. Within the theory of the polarization gradient (Maugin, 1988; Mindlin, 1972), this quantity specializes in a form that furnishes the required boundary conditions at the metal–dielectric interfaces. These boundary conditions involve the polarization field as well as its first spatial derivative and Mindlin's condition for P at the interface is recovered from them as a particular case. Then Mindlin's argument is re-proposed, though in a revised form and within the context of the electrostatics of rigid bodies. The anomalous behaviour of the capacitance is explained within this context, provided that the proposed boundary conditions hold true. It should be emphasized that while Mindlin was concerned with the elastic dielectric, we restrict ourselves to the rigid dielectric. The formulation of the problem is rather simplified and, in addition, is devoid of the concepts of strains and stresses which are inherent to deformable bodies. Nevertheless, the new boundary conditions could be reasonably extended to the electro–elastic problem.

2. STATEMENT OF TWO PROBLEMS AND OF A CONJECTURE

We will consider the following problem: a homogeneous and isotropic dielectric material is placed in a capacitor. Then we distinguish two cases: the dielectric fills the whole capacitor, as shown in Fig. 1; second, the dielectric fills it only partially, as shown in Fig. 2. The position of the metal plates will be denoted by x_0, respectively, and the position of the edges of the dielectric by $\pm x_1$, respectively. Thus, the inequality $x_1 \leqslant x_0$ holds true. We assume that the system is of infinite transversal extent and we look for a solution ϕ in terms of the polarization field P which is assumed to be known. Both problems are one-dimensional, x being the independent coordinate. We also assume that the polarization has no transversal components.

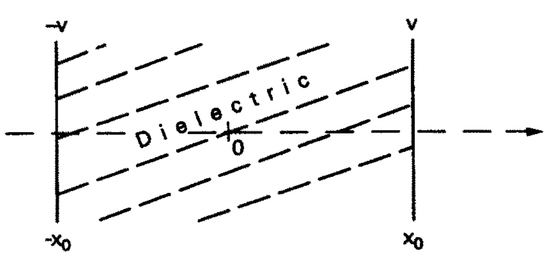

Fig. 1. The capacitor filled by the dielectric.

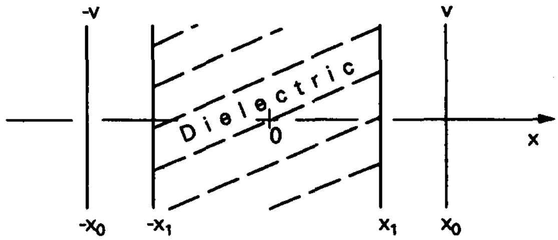

Fig. 2. The capacitor partially filled by the dielectric.

First problem

$$\phi'' = \begin{cases} 0 & \text{in } \mathbb{R}/[-x_0, x_0] \\ \dfrac{P'(x)}{\varepsilon_0} & \text{in } (-x_0, x_0) \end{cases} \tag{1}$$

$$\phi^- |_{-x_0} \equiv \lim_{x \to -x_0^-} \phi = 0; \quad \phi^+ |_{-x_0} \equiv \lim_{x \to -x_0^+} \phi = -V \tag{2}$$

$$\phi^- |_{x_0} \equiv \lim_{x \to x_0^-} \phi = V; \quad \phi^+ |_{x_0} \equiv \lim_{x \to x_0^+} \phi = 0 \tag{3}$$

$$\phi |_{\pm\infty} \text{ bounded} \tag{4}$$

$$\phi \in \mathscr{C}^3(\mathbb{R}/\{-x_0, +x_0\}) \quad \text{and} \quad P \in \mathscr{C}^2(-x_0, x_0).$$

The solution vanishes out of $[-x_0, x_0]$ and

$$\phi(x) = \frac{1}{\varepsilon_0} \int_{-x_0}^{x} P \, \mathrm{d}x + \frac{V}{x_0} x - \frac{\langle P \rangle}{\varepsilon_0}(x + x_0) \tag{5}$$

$$\phi'(x) = \frac{V}{x_0} + \frac{1}{\varepsilon_0}(P(x) - \langle P \rangle), \quad x \in (-x_0, x_0) \tag{6}$$

where

$$\langle f \rangle \equiv \frac{1}{2x_0} \int_{-x_0}^{x_0} f \, \mathrm{d}x \equiv \text{the mean value of } f \text{ across the capacitor}$$

$\varepsilon_0 \equiv$ dielectric constant of vacuum.

Second problem

$$\phi'' = \begin{cases} 0 & \text{in } \{\mathbb{R}/[-x_0, x_0]\} \cup (-x_0, -x_1) \cup (x_1, x_0) \\ \dfrac{P'(x)}{\varepsilon_0} & \text{in } (-x_1, x_1) \end{cases} \tag{7}$$

$$[\phi]_{\mp x_1} \equiv \phi^+ |_{\mp x_1} - \phi^- |_{\mp x_1} = 0 \tag{8}$$

$$-\varepsilon_0 [\phi']_{\mp x_1} = P^- |_{\mp x_1}. \tag{9}$$

Conditions for ϕ at $\mp x_0$ and at infinity as in the first problem.

$$\phi \in \mathscr{C}^3(\mathbb{R}/\{\pm x_0, \pm x_1\}) \cup \mathscr{C}^0(-x_0, x_0); \quad P \in \mathscr{C}^2(-x_1, +x_1).$$

The solution is the following:

$$\phi(x) = \begin{cases} \dfrac{V}{x_0}x - \dfrac{\langle P \rangle_1}{\varepsilon_0}x_1 \cdot \dfrac{x+x_0}{x_0}, & \text{in } (-x_0, -x_1) \\ \dfrac{1}{\varepsilon_0}\displaystyle\int_{-x_0}^{x} P\,\mathrm{d}x + \dfrac{V}{x_0}x - \dfrac{\langle P \rangle_1}{\varepsilon_0}x_1 \dfrac{x+x_0}{x_0}, & \text{in } (-x_1, +x_1) \\ \dfrac{V}{x_0}x - \dfrac{\langle P \rangle_1}{\varepsilon_0}x_1 \cdot \dfrac{x-x_0}{x_0}, & \text{in } (x_1, x_0) \end{cases} \tag{10}$$

and its derivative

$$\phi' = \begin{cases} \dfrac{V}{x_0} - \dfrac{\langle P \rangle_1}{\varepsilon_0}\dfrac{x_1}{x_0}, & \text{in } (-x_0, -x_1) \\ \dfrac{V}{x_0} - \dfrac{\langle P \rangle_1}{\varepsilon_0}\dfrac{x_1}{x_0} + \dfrac{1}{\varepsilon_0}P(x), & \text{in } (-x_1, +x_1) \\ \dfrac{V}{x_0} - \dfrac{\langle P \rangle_1}{\varepsilon_0}\dfrac{x_1}{x_0}, & \text{in } (x_1, x_0) \end{cases} \tag{11}$$

where

$$\langle P \rangle_1 = \frac{1}{2x_1}\int_{-x_1}^{x_1} P\,\mathrm{d}x.$$

The solution at the limit $x_1 \to x_0$ is

$$\phi_l = \begin{cases} -V & x \equiv -x_0 \\ \dfrac{1}{\varepsilon_0}\displaystyle\int_{x_0}^{x} P\,\mathrm{d}x + \dfrac{V}{x_0}x - \dfrac{\langle P \rangle}{\varepsilon_0}(x+x_0) & \text{in } (-x_0, x_0) \\ V & x \equiv x_0 \end{cases} \tag{12}$$

$$\phi_l' = \begin{cases} \dfrac{V}{x_0} - \dfrac{\langle P \rangle}{\varepsilon_0} & x \equiv -x_0 \\ \dfrac{V}{x_0} - \dfrac{\langle P \rangle}{\varepsilon_0} + \dfrac{P(x)}{\varepsilon_0} & \text{in } (-x_0, x_0) \\ \dfrac{V}{x} - \dfrac{\langle P \rangle}{\varepsilon} & x \equiv x_0. \end{cases} \tag{13}$$

Solution (13) coincides with (6) as long as we restrict ourselves to $(-x_0, +x_0)$. Notice that ϕ_l is continuous in $[-x_0, x_0]$ while ϕ_l' suffers a discontinuity at $\pm x_0$. It is worth remarking that the mean value $\langle \phi' \rangle$ is equal to V/x_0 and is independent of the polarization field just as in the classical case. In this latter case one usually assumes $P = -\varepsilon_0 \chi \phi'$, χ representing the electric susceptibility. Then expression (13) becomes:

$$\phi_l'|_{\text{class}} = \begin{cases} \varepsilon \dfrac{V}{x_0} & \text{in } \{-x_0, x_0\} \\ \dfrac{V}{x_0} & \text{in } (-x_0, x_0) \end{cases} \tag{13a}$$

where $\varepsilon = \chi + 1 \equiv$ the relative dielectric constant.

Bearing in mind that the electric charge (per unit surface) σ at the metal plate is given by the jump of the dielectric displacement, σ may be evaluated in two ways. One, which is based on formula (10), is by setting

$$\sigma = \lim_{x_1 \to x_0} \varepsilon_0[-\phi']_{x_0} \equiv \varepsilon_0 \phi_l'|_{x_0} \equiv \varepsilon_0 \frac{V}{x_0} - \langle P \rangle. \tag{14}$$

The second way, which is based on formula (6), is by setting

$$\sigma = [-\varepsilon_0 \phi' + P]_{x_0} = \varepsilon_0 \frac{V}{x_0} - \langle P \rangle. \tag{14a}$$

Notice that in (14) we are approaching x_0 "in the vacuum", while in (6) we are approaching x_0 "inside the dielectric". In the classical case we have $\langle P \rangle = -\varepsilon_0\ \chi(V/x_0)$, whence $\sigma = \varepsilon_0 \varepsilon (V/x_0)$ by formula (14) and the classical expression for σ is also recovered. In order to simplify the notations we shall define the following quantities:

$$[\phi']_{1,0} \equiv \lim_{x_1 \to x_0} [\phi']_{x_1}$$

$$[\phi']_0 \equiv [\phi']_{x_0}$$

$$[\phi_l']_0 \equiv [\phi_l']_{x_0}.$$

Then, with reference to expression (13) we will consider the following difference:

$$-\varepsilon_0 \{[\phi']_0 - [\phi']_{1,0}\}. \tag{15}$$

It is worth remembering that $-\varepsilon_0[\phi']_{x_1} \equiv P(x_1)$ represents the polarization charge (per unit surface) at the dielectric–vacuum interface, while $\varepsilon_0 \langle \phi_l' \rangle \equiv \varepsilon_0 (V/x_0)$ represents the electric charge at the metal plate in the absence of the dielectric.

Our conjecture is that expression (15) represents the extra charge at the metal–dielectric interface and that the quantity

$$-\frac{\varepsilon_0\{[\phi_l']_0 - [\phi']_{1,0}\}}{\varepsilon_0 \langle \phi' \rangle} \tag{16}$$

must be a constant. Then by posing

$$\frac{\langle P \rangle + P(x_0)}{\varepsilon_0 \dfrac{V}{x_0}} = \Sigma \tag{17}$$

and taking into account solution (13), the conjecture may be expressed as follows:

$$-\frac{[\phi']_0 + [\phi']_{1,0}}{\langle \phi_l \rangle} = 1 - \Sigma. \tag{18}$$

This means that the relative accumulation of electric charge at metal–dielectric interface with respect to the corresponding charge at the metal face in the absence of the dielectric has to be considered a feature of the interface contact of the two materials.

3. THE ROLE OF THE POLARIZATION GRADIENT

In order to specify completely the dielectric response we will refer to Mindlin's formulation and assume that Mindlin's equation holds inside the dielectric (Askar *et al.*, 1970; Maugin, 1988; Mindlin, 1969, 1972; Nelson 1979).

In the present case such an equation transforms into the following form

$$\alpha P''(x) - aP(x) - \phi'(x) = 0 \tag{19}$$

where a and α are constitutive coefficients. They are required to be strictly positive in order to preserve the positive definiteness of the polarization energy (Askar *et al.*, 1970; Maugin, 1988; Mindlin, 1972). To eqn (19) we must add eqns (1) for the first problem or eqns (7) for the second problem and the proper boundary conditions for each of them. Let us begin with the second problem. Since the dielectric is separated by the metal, we shall put

$$P'(x)|_{\pm x_1} = 0 \tag{20}$$

in accordance with the general theory (Mead, 1961, 1962; Mindlin, 1969).

In addition, the condition

$$P(x) = P(-x) \tag{21}$$

follows from the symmetry of the problem.

The solution of the problem is an easy task. P and ϕ' are constant fields and specifically:

$$P = -\frac{1}{a}\phi' = -\frac{1}{a}\frac{V}{x_0} \quad \text{in}\,(-x_1, x_1) \tag{22}$$

$$\phi' = \begin{cases} \dfrac{V}{x_0}\left(1+\dfrac{1}{\varepsilon_0 a}\dfrac{x_1}{x_0}\right) & \text{in}\,(-x_0, -x_1)\cup(x_1, x_0) \\[2ex] \dfrac{V}{x_0}\left\{1+\dfrac{1}{\varepsilon_0 a}\left(\dfrac{x_1}{x_0}-1\right)\right\} & \text{in}\,(-x_1, x_1). \end{cases} \tag{23}$$

By interpreting $1/\varepsilon_0 a$ as the electric susceptibility, solution (22) coincides with the classical one in the open interval $(-x_1, x_1)$. In order to choose the proper boundary conditions for the polarization and for the gradient of polarization at the metal–dielectric interface in the effective problem, we will refer to the limit solution of the associated problem, i.e. to the formulae (12) and (13).

For the effective problem condition (20) is no longer justified and we suggest it is substituted by condition (18). This condition in terms of gradient of polarization becomes as follows:

$$P(x_0)x_0 + \frac{\alpha}{a}P'(x_0) = \left(\frac{1}{a}+\Sigma\,\varepsilon_0\right)V \tag{24}$$

and furnishes the boundary condition for the global problem expressed by eqn (19) and by the second limit problem.

The problem is now completely formulated and the solution for P is the following:

$$P(x) = \varepsilon_0 \frac{V}{x_0} \left\{ \frac{\left(\Sigma + \frac{2}{\varepsilon_0 a}\right)\left(\frac{\cosh \mu x}{\cosh \mu x_0} + \frac{1}{\varepsilon_0 \mathrm{a}} \frac{\tanh \mu x_0}{\mu x_0}\right)}{1 + \left(1 + \frac{2}{\varepsilon_0 a}\right) \frac{\tanh \mu x_0}{\mu_x}} - \frac{1}{\varepsilon_0 a} \right\} \tag{25}$$

where $\mu = \frac{1}{\alpha}\left(a + \frac{1}{\varepsilon_0}\right)$.

A comparison with the classical linear case shows that expressions (16) and (17) become:

$$1 - \Sigma = \varepsilon + \chi \quad \text{and} \quad \Sigma = -2\chi \tag{26}$$

having introduced the dielectric constant ε and assumed $\chi \equiv 1/\varepsilon_0 a$. Substituting these values into formula (25) we recover the classical polarization field which is

$$P(x) = -\varepsilon_0 \chi \frac{V}{x_0}. \tag{27}$$

Solution (25) also predicts the possibility of a positive response from P depending on the value of Σ.† While this is unpredicted by the classical linear theory (Maugin, 1988; Mindlin, 1969; Trimarco, 1989; Truesdell and Toupin, 1960), it becomes reasonable as far as the internal structure dominates and the distribution of the electric charges inside the dielectric crystal is not markedly affected by the applied voltage.

4. THE CAPACITANCE

We will refer to the classical definition of the capacitance C_a (per unit surface) of a capacitor.

$$C_a = \frac{\sigma}{2V} \tag{28}$$

where σ is the electric charge given by formulae (14) and (14a). With reference to expressions (14) and (25) formula (28) becomes

$$C_a = \frac{\varepsilon_0 \varepsilon}{2x_0} \cdot \frac{1 + (1 - \Sigma) \frac{\tanh \mu x_0}{\mu x_0}}{1 + \left(1 + \frac{2}{\varepsilon_0 a}\right) \frac{\tanh \mu x_0}{\mu x_0}}. \tag{29}$$

Since C_a must be a positive quantity, according to its physical meaning, a first restriction follows from expression (29):

$$\Sigma \leqslant 2. \tag{30}$$

With reference to (29), C_a^{-1} as a function of x_0 shows an asymptotic behaviour as $x_0 \to \infty$; the asymptote has a unit slope and the following value as $x_0 \to 0$:

† We will see in the next section that the restrictions imposed on Σ are compatible with this possible response of P.

$$C_0^{-1} = \frac{2}{\mu\varepsilon_0\varepsilon}\left(\Sigma + \frac{2}{\varepsilon_0 a}\right). \tag{31}$$

This value is interpreted as the residual capacitance experimentally observed. With reference to formula (31), one more restriction on the possible values of Σ is

$$\Sigma \geqslant -\frac{2}{\varepsilon_0 a} \tag{32}$$

in order that expression (29) should not be negative, according to the physical meaning attributed to it. Taking into account conditions expressed by (30) and (32) there results

$$-\frac{2}{\varepsilon_0 a} \leqslant \Sigma \leqslant 2. \tag{33}$$

Notice that the classical form

$$C_a|_{\text{class}} = \frac{\varepsilon_0\varepsilon}{2x_0} \tag{34}$$

is easily recovered by putting $\Sigma = -2\chi$ and $\chi \equiv 1/\varepsilon_0 a$ into expression (29).

A comparison with Mindlin's result shows that it coincides with the one expressed by formula (30) provided that

$$-\frac{2}{\varepsilon_0 a} \leqslant \Sigma \leqslant -\frac{1}{\varepsilon_0 a}. \tag{35}$$

It is worth remarking that P results in being not negative under the restrictions imposed by (35) for Σ, with reference to formula (25). Positive values of $P(x_0)$ occur for $\Sigma \geqslant -1/\varepsilon_0 a$ and this fact may be interpreted as the occurrence of a "barrier" (Kittel, 1986).

We see from formula (31) that the residual capacitance C_0 decreases with Σ increasing from $-1/\varepsilon_0 a$ to 2. In addition the initial slope T_0 of the graph C_a^{-1} versus x_0, which is given by

$$T_0 = \frac{2(1+1/\varepsilon_0 a)}{2-\Sigma}, \tag{36}$$

increases to infinity as $\Sigma \to 2$ and the deviation of $C_a^{-1}(x_0)$ from its asymptote is quite negligible within distance much shorter than the ones usually established. This fact may suggest that the anomalous behaviour of the capacitance may occur for layers thinner than 30 Å.

There follows a table of values of C_0 depending on Σ (Table 1). In this table we refer to two hypothetical materials whose relative dielectric constants are $\varepsilon = 10$ and $\varepsilon = 20$, respectively. We are also reminded that $1/\mu \approx 2 \div 10$ Å (Askar *et al.*, 1970).

Table 1. Values of C_0 depending on Σ

$\varepsilon = 10$		$\varepsilon = 20$	
Σ	Co (μF/cm^2)	Σ	Co
−16	5	−38	10
−14	2.5	−34	3.3
−12	1.7	−30	2
−10	1.25	−22	1.1
−8	1	−20	1
−6	0.8	−12	0.7
−4	0.7	−4	0.55
0	0.53	0	0.55
1	0.51	1	0.49
2	0.5	2	0.48

5. COMMENTS

The capacitance resulting from the formula is very close to one found by Mindlin since the behaviour of C_a as a function of x_0 is just the same, though the constants are different. The constant Σ introduced here lies within a range larger than that found by Mindlin (1969, 1972) and predicts a possible counter-polarization against the electric field. We believe that Σ may be evaluated within the microscopic physics of semiconductors with specific reference to the binding energy (Kittel, 1986). This energy should be connected with the discontinuity of the charge at the metal–dielectric interface as given by expressions (15) and (18).

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