Space charge in submicron cavities by quantum electrodynamics?

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Abstract: Space charge has been related to the aging of polyethylene (PE) under high electrical DC fields. For fields > 10-20 kV/mm, the amorphous phase is deformed causing nanovoids to grow to submicron (< 1 μm) cavities by the chain scission of PE molecules. Once the cavities are formed, space charge is generally thought produced by partial discharge. But the mean free path (MFP) of air at or below atmospheric pressure is larger than submicron dimensions, and therefore an alternative mechanism is suggested. Nicely suited are mechanisms that rely on quantum electrodynamics (QED) and take advantage of the inherent nature of submicron cavities to have an electromagnetic (EM) resonance beyond the vacuum ultraviolet (VUV). In QED, the infrared (IR) radiation from atoms in the penetration depth of the resonant VUV radiation standing across the QED cavities is suppressed. Since the suppressed IR radiation can only be conserved by an equivalent gain of EM energy at the resonant frequency of the QED cavity, the suppressed IR radiation is induced to undergo spontaneous frequency up-conversion to the VUV, the process called cavity QED induced EM radiation. Subsequently, the VUV irradiation of impurities in the PE produces electrons leaving positive charged ions that form the space charge.

Introduction

Space charge in PE has been known [1] as one of the key phenomenon in electrical breakdown and conduction mechanisms in dielectrics generally, space charge is dispersed in bulk PE by microscopic cavities. Submicron < 1 μm cavities present after PE extrusion under field induced strain grow [2] over the life of the insulator to 1-5 μm cavities at concentration of about 10^6 / mm^3. Indeed, the density of voids is proposed [3] as a measure of life in PE insulators.

Space charge is thought produced in the cavities by partial discharge and charge injection from the electrodes.

Partial discharges require collisions of air molecules that depend on their mean free path. At atmospheric pressure, the mean free path of air is greater than about 0.1 μm. But for submicron cavities, there are only ten air molecules available, and therefore partial discharge is usually [4] dismissed as the source of space charge.

Absent partial discharge, either another mechanism is operating or charge injection is required to supply space charge to the submicron cavities. Regardless, space charges in cavities have directly been linked to aging of PE.

> "It is our contention that space charges are a consequence of aging, i.e., charges are injected only when physical defects (such as microcavities) have been formed by the field-induced strain…space charges are related to the formation of submicrocavities, and therefore, are a consequence, not a cause of high field aging." [5]

But charge injection has difficulty explaining why absent a field the residual charge ~ 0.25 C / m^3 is measured [5] which is fairly typical of the residual charges always detected in PE. Another mechanism is operating to supply space charge.

Purpose

The purpose of this paper is to propose that space charge in PE is produced in submicron cavities by cavity QED induced EM radiation.

Theory

Cavity QED induced EM radiation follows from the most basic laws of physics – that low frequency EM radiation is suppressed in a high frequency QED cavity - the generality of the law applicable to QED cavities in both the solid and liquid state. The submicron QED cavity is depicted in Fig. 1.

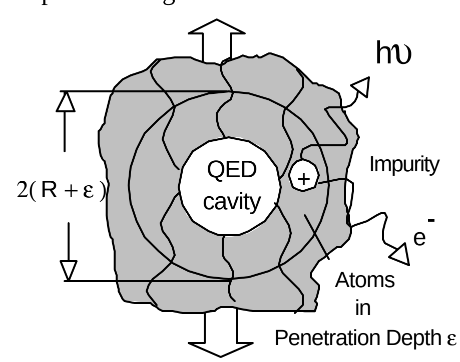

Figure 1 Submicron Cavity and Impurity

Typically, IR radiation from QED cavity surfaces at ambient temperature is suppressed from atoms within the penetration depth of the resonant VUV radiation standing across the QED cavities. But in a QED cavity the EM energy loss from the suppression of IR radiation can only be conserved by an equivalent gain at its resonant frequency, and therefore metal impurity atoms in the QED cavity surface are spontaneously excited by VUV radiation to liberate electrons while forming positive space charge.

In cavity QED induced EM radiation, standing EM waves gain energy from the suppression of IR radiation from atoms on the cavity surface. The EM wave has wavelength λ and includes the depth ε necessary to assure absorptive (or reflective) boundary conditions.

$$\lambda = 4(R + \varepsilon) \qquad (1)$$

The Beer-Lambert law gives the depth ε by the absorption coefficient α of the cavity wall. The EM radiation intensity I at depth ε is related to the intensity I_o at the QED cavity surface by,

$$I / I_o = \exp(-\alpha\varepsilon) \qquad (2)$$

For $\alpha\varepsilon = 5.15$, over 99 % of intensity I_o is absorbed.

At ambient temperature, the thermal kT energy of atoms in depth ε is emitted as EM radiation in the IR given [6] by the harmonic oscillator in Fig. 2.

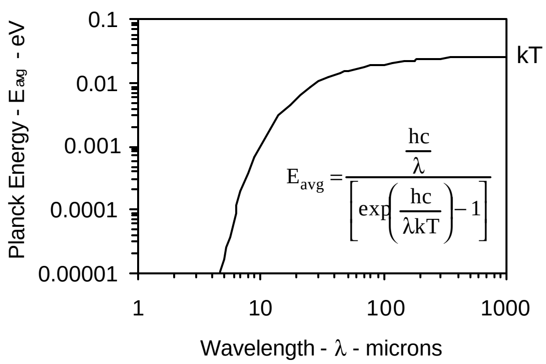

Figure 2 – Harmonic oscillator at 300 K. In the inset, λ is wavelength, T is temperature, h is Planck's constant, k is Boltzmann's constant, and c is the speed of light.

Since the EM resonance of the QED cavity is considered in the VUV, the full thermal kT energy of the atoms in the depth ε is suppressed. The total IR energy U_{IR} suppressed,

$$U_{IR} = \frac{4\pi}{3} \Psi \left[(R+\varepsilon)^3 - R^3 \right] \qquad (3)$$

where, $\Psi \sim N_{dof}$ x ½ kT / Δ^3, and Δ is the cubical spacing between atoms at solid density. N_{dof} is the number of degrees of freedom of the CH_2 groups in the PE molecules. Typically, $N_{dof} = 6$.

Suppressed IR is a loss of EM energy that in a QED cavity may only be conserved by an equivalent gain in Planck energy at its resonant frequency. For QED cavities having EM resonance in the VUV, the conservation of EM energy is expressed by,

$$N_{VUV} E_{VUV} = U_{IR} \qquad (4)$$

where, N_{VUV} is the number of photons having average Planck energy E_{VUV} over the depth ε,

$$E_{VUV} = \frac{hc}{4(R + 0.5\varepsilon)} \qquad (5)$$

Combining, the number N_{VUV} of photons in the penetration depth ε,

$$N_{VUV} = \frac{8}{3}\pi \frac{kT}{hc} N_{dof} (R + 0.5\varepsilon) \left[\frac{(R+\varepsilon)^3 - R^3}{\Delta^3} \right] \qquad (6)$$

Computation of N_{VUV} depends on the absorption coefficient α of PE, but is unknown. For the purpose of illustration, the α for water is selected to represent PE. Regardless, water has the optimum absorption for liquids and may be considered an upper bound for PE. The absorption α for water [7] is shown in Fig. 3.

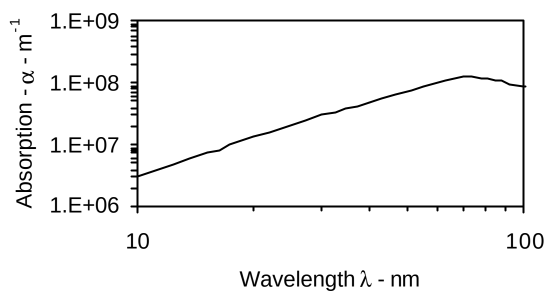

Figure 3 –Absorption coefficient α for water

From (6) and (7), the Planck energy E_{VUV} and number N_{VUV} of VUV photons produced for $\alpha\varepsilon = 5.15$ at T = 300 K is shown in Fig. 4. Maximum $E_{VUV} \sim 8.3$ eV and minimum $N_{VUV} \sim 2\text{x}10^5$ occurs at R ~ 18 nm.

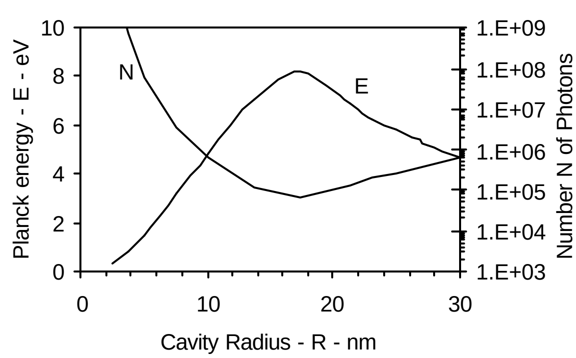

Figure 4 –Planck Energy and Number of Photons

The space charge Q in the dielectric is not injected charge from the electrodes, but rather produced from the VUV irradiation of metal impurities in the PE,

$$Q = YN_{VUV} \tag{7}$$

where, Y is the yield [8] of cations / VUV photon.

Application

The submicron QED cavity under field induced strain may form by: (1) the chain reaction initiated by the scission of a single macromolecule, and (2) QED radiation of a nanovoid nucleated in the intermolecular space during PE processing shown in Fig. 5.

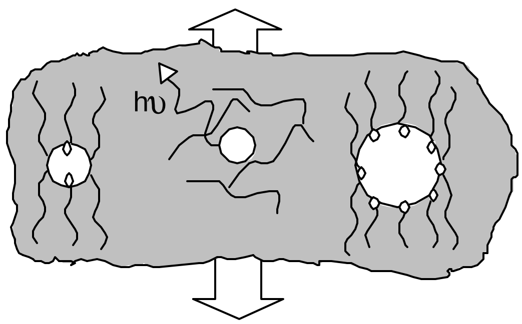

Figure 5 Formation of Submicron Cavities

Cavity formation by chain reaction

The scission of a single macromolecule is shown in Fig. 5(a). Scission forms [9] the highly reactive primary end radical ($-CH_2$ - C^*H_2). The primary end radical then scissions its neighbors by forming the internal radicals ($-CH_2$ - C^*H_2 $-CH_2-$) that in turn scission their neighbors, and so forth in a chain reaction to form the submicron QED cavity shown in Fig. 5(c).

The radius R* of the QED cavity formed is related to the number N_b of macromolecules having broken bonds in a close packed configuration,

$$R^* = \sqrt{\frac{N_b S}{\pi}} \tag{8}$$

where, S is the area of the macromolecule. For PE, both [9,10] give $S \sim 1.824 \times 10^{-19}$ m^2. Experimental data [9] gives $N_b \sim 1600$, and therefore R* ~ 10 nm.

But submicron cavities formed by chain reaction initiated from the scission of a single macromolecule under field induced strain is questionable because fields in the 10 to 50 kV/mm range [5] lack the energy to scission PE backbones having strength of about 1 – 3 eV. Another mechanism is forms the submicron cavity.

In fact, it is more likely that the submicron cavities grow from nanovoids nucleated in the intermolecular space during PE extrusion.

Cavity formation by QED radiation

The nucleation of a nanovoid R < 1 nm in the intermolecular space is shown in Fig. 5(b). Since bonds need not break for growth in the intermolecular space, field induced strain can readily grow the void to R ~ 4 nm. From Fig. 4, the QED cavity produces $N_P \sim 10^9$ photons having Planck energy ~ 1 eV to spontaneously scission N_S molecules as shown in Fig. 5(c).

The quantum yield ϕ_S of LDPE scission [11] in sunlight having Planck energies 1 – 3 eV is $\phi_S \sim 1 \times 10^{-6}$. For $N_P \sim 1 \times 10^9$ photons, the number N_S of scissions is, $N_S = \phi_S N_P \sim 1000$ that is a low estimate of ~ 1600 scissions measured per cavity (Table III of [9]). Alternatively, photolysis by ~ 1 eV N_P photons scission $N_S \sim 1000$ macromolecules is comparable to the chain reaction scission of $N_b \sim 1600$ macromolecules initiated by the scission of a single molecule. The radius R* of the QED cavity formed is related to the number N_S of scissions in a close packed configuration,

$$R^* = \sqrt{\frac{N_s S}{\pi}} \tag{9}$$

Space charge production

Whether formed by chain reaction or QED radiation, the QED cavity continues to grow $R > R^*$ under field induced strain until the EM radiation in the VUV is produced capable of exciting metal impurities in the PE. Space charge is produced by the cation formed by the VUV radiation hυ interacting with the metal impurity.

Indeed, Fig. 4 shows QED cavities of radius R ~ 18 nm produce ~ 2×10^5 VUV photons having Planck energies of ~ 8 eV. For metallic impurities in the PE, electron yields Y [8] are of order 10^{-4} / VUV photon at ~ 8 eV. Thus, the space charge $Q \sim YN_{VUV} \sim 20$ positive charged impurities are produced in each of the QED cavities throughout the bulk PE.

Discussion

Primary end radicals and number of cavities

In PE, the concentration of submicron cavities is almost the same as that of free radicals, but is smaller than the number of scissioned macromolecules. (Table III of [9]). In field induced strain scission, it only takes one primary end radical to initiate the chain reaction of N_b broken bonds that forms the submicron QED cavity, the number of QED cavities is the same as the number of primary radicals; whereas, in photolysis, the number N_S of macromolecules spontaneously scissioned with QED radiation does not depend on the chain reaction initiated by the primary end radical.

But under field induced strain, the nucleation of a void in the intermolecular space is favored to the scission of a macromolecule, and therefore photolysis is the likely mechanism by which cavities grow in PE.

Negative resistance

Space charge in PE insulators is a phenomenon of field-induced strain. At fields <10 kV/mm, the current density is negligible. The current density increases with field to ~ 80 - 100 kV/mm, but thereafter decreases reaching a minimum at about 150 V/mm. Further increases in field > 150 V/mm increase the current density. At the same time, the space charge saturates at about 150 V/mm. (Fig. 1 and 11 of [5]). The phenomenon has been observed for some time and is referred to as "negative resistance."

"Negative resistance" and saturation of space charge is explained in [5] by the argument there are no more defects created and that charge injection from the electrodes reaches a constant state. But this is difficult to understand because as the space charge is saturating the current increases after reaching the minimum. The difficulty here is that the source of space charge and current density are both thought to originate by charge injection from the electrodes.

In contrast, space charge produced by cavity QED induced EM radiation is independent of charge injection from the electrodes. But space charge is only produced over a range of QED cavities having EM resonant frequencies coincident with the excitation frequency of the impurities in the PE. Further cavity growth produces lower EM resonant frequencies that do not excite the impurities in PE, and therefore space charge is no longer produced consistent with the data.

Number of space charges and cavities

Space charge in a PE insulator is explained [5] by assuming each QED cavity can accommodate one charge; whereas, the number of radicals is shown [9] to correspond to the number of cavities. This means [5] and [9] are consistent with each other if the radical produces one charge. But this is unlikely.

By QED radiation, the number of space charges in a QED cavity depends on the yield of the impurity and is not limited to an arbitrary value of one charge.

Conclusions

Cavity formation initiated by the scission of single macromolecule under field induced strain is less likely than the nucleation of a nanovoid in the intermolecular space and subsequent growth by QED radiation. However, optical absorption and scission yield data for PE is lacking to confirm this conclusion.

References

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