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***Abstract*- Charge distribution estimation at the surface and in the bulk of dielectrics is of great interest, both theoretically and experimentally, not only because it brings information on the storage and transport properties of the medium, but also because of various possible applications of thin dielectric layers e.g. in the field of miniaturized components. Scaling down the dimensions of these components to nanoscale level creates difficulties with the application of the commonly used diagnostic methods. With a resolution in the nanometer range, the atomic force microscopy (AFM) can be used to this field. By applying a voltage between the microscope tip and a sample, both conducting and insulating surfaces can be imaged. The AFM can be applied for charging of samples in contact mode or when a tip-to-sample spacing is introduced. The purpose of this work is to discuss about appropriate mechanisms for dielectric charging in AFM that are still controversial in the literature.**

Keywords: AFM, dielectrics, charging mechanisms.

I. INTRODUCTION

Charge distribution on the surface and in the bulk of dielectric materials is of great interest, both theoretically and experimentally, because of various possible applications. One can find a good review on mechanisms for insulator charging and charge detection methods in the recently published review paper of Rezende et al. [1]. As it is shown in Ref. 1, new diagnostic techniques are being developed, in the last two decades, to answer the increasing demand for characterization of miniaturized components used in microelectronics, telecommunications, electrophotography, electrets, etc. Scaling down the dimensions of these components to nanoscale level creates difficulties with the application of commonly used diagnostic methods. The well-known non-destructive diagnostic techniques for measurement of surface charge distribution in insulators like thermal-pulse or pressure pulse methods or probe-based methods have only achieved 10 μm of lateral resolution [2]. Recently developed diagnostic techniques, like Focused Laser Intensity Modulation Method [3], allow micrometer scale lateral resolution. Unfortunately, it remains inapplicable for charge detection in nanoscale dielectric layers.

Since the scanning force microscope (SFM) [4] was developed, it was used for studies of deposition and imaging of charges on insulator surfaces, by means of Kelvin force microscopy mode. By applying a voltage between the force microscope tip and a sample, both conducting and insulating surfaces can be imaged. An interesting review on the aspects of AFM and the probes used in various AFM applications has been recently published [5]. The atomic force microscope can be applied for charging of samples in both contact mode [6, 7, 9] or when a tip-to-sample spacing is introduced [8 – 13].

In the literature reporting measurements achieved in AFM, corona discharge is suggested as a mechanism of dielectric charging [8 – 11]. We first review this hypothesis. Then we will discuss the possibilities for sustaining gas discharges in small geometries. We will continue with a short discussion on field electron emission. The departure from Fowler-Nordheim theory due to the nanoscale dimensions is discussed next. The last section is about likely mechanisms of dielectric charging in AFM that we propose.

II. CORONA DISCHARGE

Corona discharge is a partial breakdown in gas. It occurs in highly inhomogeneous electric fields [14]. Several conditions are required for its ignition: (i) an asymmetric electrode configuration must be present; (ii) a high voltage must be applied to the smaller electrode; (iii) presence of some free electrons is needed; (iv) an avalanche must develop, leaving behind a space charge area. The total current in corona discharges is small, up to hundredths mA, depending on the applied voltage, electrode size and interelectrode distance. It is approximately the same for positive and negative corona discharges in air at atmospheric pressure, as the mobilities of positive and negative ions are close in air. This similarity in the current value of positive and negative corona discharges in air at atmospheric pressure coincides with the similar values of the measured surface potential when the dielectric charging with positive and negative charges is performed in AFM [8 – 11]. Perhaps it was a reason for the authors to use the analogy with conventional dielectric charging by corona discharge and to call the mechanism of dielectric charging in AFM corona discharge. Following the general trends of ignition of gas discharges and of ignition of corona discharge, in particular, we can state that the small tip-to-sample distance in the case of AFM dielectric charging does not allow an avalanche build up. Tip-to-sample distances of the order of 100 nm are not much larger than the characteristic size of the small electrode. The electric field can be strong in case of AFM, but not

enough strongly inhomogeneous as required for the ignition of corona discharge. A rough estimate of the electric field in AFM gives 10^8 V/m, if we consider a voltage of 20 V and an interelectrode distance of 200 nm (tip-to-sample distance + dielectric thickness). However, tip-to-sample distances of the order of 100 nm are even smaller than the electron mean free path in air at atmospheric pressure λ = 500 nm. It means that any electron created in the gap will immediately attain the anode without possibility of multiplication, i.e. without an avalanche build up. Corona discharge should thus be excluded from the likely mechanisms for dielectric charging in AFM, but the question of gas breakdown in very small gaps is to be revised.

III. DISCHARGE BREAKDOWN IN SMALL GAPS

The ignition and maintenance of gas discharges in small geometries (micrometer size) change drastically when the gap is in order of only few microns. Papers reporting on discharges in small gaps at high gas pressures (usually atmospheric pressure) [15 – 21] show that the breakdown threshold differs from the value estimated from the Paschen's curve when the interelectrode distance is below 5 μm. Actually, the deviation from Paschen's law occurs only for those experimental conditions for which the multiplication of electrons is very small. The departure from Paschen's curve was largely discussed first in the work of W. S. Boyle and P. Kisliuk [15]: when the multiplication is small, the high electric field is sufficient to draw field emission current from some small irregularities of the cathode. These electrons produce small number of ions, which by space charge effect increase the field on the cathode. This process is able to build up to a breakdown even with an extremely small ionization due to the reduced collisions in small gaps. The mechanisms in place for gas breakdown in very small gaps (≅ 2–3 μm or less, which tend to the typical tip-to-sample distances in AFM dielectric charging) are all connected and primarily depending on the high electric field. They largely differ from the standard mechanisms of ignition and maintenance of gas discharges: ionization in the volume and secondary electron emission. They should be found in the variety of mechanisms solely depending on the electric field. Usually the field dependent mechanisms demand a very high electric field. When dealing with AFM the electric field is of order of 10^8 V/m. The field emission effects, responsible for the departure from Paschen's curve at very small gaps and the theory that describes these effects, given by Fowler and Nordheim, are discussed next.

IV. FIELD ELECTRON EMISSION

Field electron emission is a process of electron emission from solid surfaces (metals, semiconductors and dielectrics) subject of submission to a high electric field, usually larger than 10^8 V/m. It is due to the fact that the high electric field renders the potential barrier at the interface between the emitting surface and the medium (whatever it is: vacuum, gas, solid) so thin (order of tens of Angstrom) that the electrons have chance to tunnel through it. The principles of field electron emission were established by R. H. Fowler and L. Nordheim in their theory based on quantum-mechanical description of tunneling of electrons [22]. In a strict sense, the Fowler-Nordheim theory applies for metals but it can be used for other materials if some additional assumptions that describe the electrode nature are made.

The field electron emission is measurable above a threshold field, which is determined by the work function of the emitting surface. Typical theoretically obtained values of the threshold field are in the range 10^8 V/m – 10^9 V/m. We consider here the current density according to the theory of E. L. Murphy and R. H. Good [24] that describes the combined effect of thermionic and field emission of electrons from metals. The expression (56) from Ref. 24 represents the Fowler-Nordheim expression for field electron emission current. It gives the zero-temperature approximation. For example, for a field of 2×10^9 V/m when the emitter is a nickel electrode (Φ = 5.15 eV [23], Φ is the work function), the emitted electron current density into vacuum is 2.6×10^{-3} A/m². For the same electric field, the current density is 2.9 A/m² in case of tungsten electrode (Φ = 4.55 eV), i.e. higher by 3 orders of magnitude. From these examples, one can notice the important role of the electrode material properties, in particular of the work function.

With the typical values of the work function of pure metals, which are of 4.0 eV or higher, the field emission effect is slightly temperature dependent as shown in Fig. 1 that represents the current density as a function of the electric field when the thermionic and field emissions act together [24]. This underlines one of the outstanding characteristic of the field emission from pure metals: its almost complete temperature independence up to 600 K. For the logarithmic scales of the electron current density emitted from pure metals, as used experimentally, the temperature effect on field emission currents can be disregarded. We will see later in our analysis that it is not the case when the material is characterized by a low work function (Φ < 2.0 eV), like for some materials used in the fabrication of AFM tips [5, 25, 26].

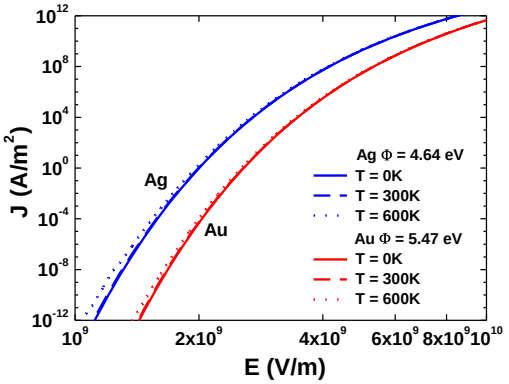

Fig. 1. Current density as a function of the electric field due to the thermionic and field emissions acting together. The current density is calculated according to expression (56) in Ref. 24 for Ag (Φ = 4.64 eV) and Au (Φ = 5.47 eV), for three different temperatures T = 0 K (solid line); 300 K (dash line) and 600 K (dot line).

As far as the geometry of the emitter is considered, the Fowler-Nordheim representation is verified for electron emitter, which is a sharply pointed wire, of apex radius of the order of μm [27]. It was found that the experimental plot $\ln I = f(1/V)$ is linear for a considerable range of currents below a critical value I_c. For currents higher than I_c, which corresponds to very high electric field intensities, a departure from linearity towards lower currents for a given voltage occurs. This departure is due to the space charge limited current (SCLC) [27]. However, we should bear in mind that the hypotheses of Fowler-Nordheim theory do not apply for all geometrical configurations and electric field distributions. The Fowler-Nordheim theory describes a planar cathode-anode gap with uniform electric field. It is assumed that the potential barrier is exact or rounded triangular barrier. This assumption is valid for potential barriers having width significantly smaller than the emitting surface, i.e. for bulk metals (dimensions larger than 100 nm). Under the above assumptions the free-electron model is correct and the WKB approximation is proper to estimate the barrier transmission coefficient, which prevents from solving the strict Schrödinger's equation. When using divergent field configuration, as the one in AFM, and emitters with radius of only few nanometers, a departure from the Fowler-Nordheim theory should be expected. In particular, it has been established that field emission current-densities from nanometer-size emitters can be several orders of magnitude higher than traditional field-emitting objects. The main reason could be that the traditional Fowler-Nordheim theory of field emission becomes doubtful and the strict treatment of the problem requires a revision of some of the hypotheses of the theory.

V. LIKELY MECHANISMS OF DIELECTRIC CHARGING IN AFM

Due to the peculiar features of field emission observed with nanometer size field emitters leading to higher current density than that obtained with traditional field emitters, charging of the dielectric surface in tip-to-sample mode in AFM through field electron emission can be envisaged as the operating mechanism. Quickly after reaching the dielectric surface the electrons will be trapped in electron acceptors levels (electron traps) that are due to the typical physical and chemical disorder existing at any dielectric surface [28 – 30].

To substantiate our hypothesis about the primary role of the field electron emission in the dielectric charging in AFM, we will present numerical results for the emitted electron current density from different materials used for the fabrication of AFM tips [5, 25, 26]. These results are obtained according to the theory of E. L. Murphy and R. H. Good (the expressions for the current density given in Ref. 24) that describes different type of electron emission: the thermionic emission, field electron emission and the intermediate region.

Figure 2 shows results for the current density as a function of the electric field in the case of thermionic and field emissions acting together at T = 300 K (solid lines) and in the zero-temperature approximation, i.e. when only the field emission proceeds (dash lines). The presented curves are for the materials listed in Table 1. We can notice from the data presented in Table 1 that the work function is strongly modified after surface treatment of the silicon emitters. The DLC coating leads to a decrease of the work function with more than a factor of 2. Doping of the DLC-coated Si-emitters with N atoms has an additional impact on the work function. With the increase of nitrogen concentration, the work function decreases due to a displacement of the Fermi level towards the conduction band. Because of the decrease of the work function of material, the current density increases by several decades for the same applied electric field. For typical values of the electric field in the AFM configuration (10^8 V/m) the measured currents can be quite significant. An estimate for 20 nm radius of apex of the AFM tip gives current values of about tens to hundreds of pA, which correspond to the current values measured in AFM experiment [13]. Results for the current density, presented in Fig. 2, clearly show the impact of temperature increase when the work function is below 2 eV, i.e. the role of thermionic emission being important even for a temperature T = 300 K. As expected this effect is more pronounced at low electric field ($E < 1.8 \times 10^8$ V/m) for all the cases considered here. The low limit of field emission decreases with the decrease of the work function and in the case of a work function as low as Φ = 0.47 eV, the thermionic and the field emission act together even for temperatures below T = 300 K. The high limit of field emission, which represents the applicability of Fowler-Nordheim formula, shifts towards higher electric fields with the increase of the work function and for Silicon (Φ = 4.3 eV) this limit at T = 300 K is $E = 1.0 \times 10^{10}$ V/m.

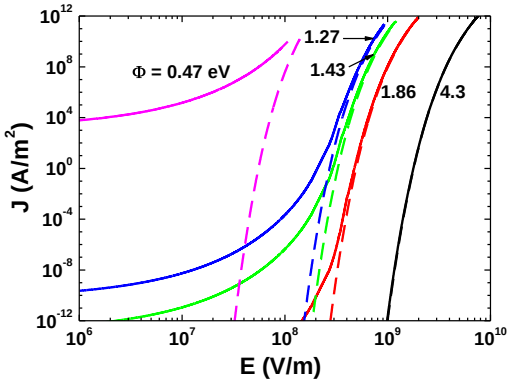

Fig. 2. Current density as a function of the electric field: (solid lines) the thermionic and field emissions acting simultaneously, T = 300 K; (dash lines) the zero-temperature approximation when only the field electron emission proceeds, for the materials studied in [25], cf. Table 1.

Table 1. Materials investigated for the use as AFM tip [25]. DLC= diamond-like carbon.

Material	N-doping for DLC	Φ (eV)
Silicon	--	4.30
DLC coated silicon	--	1.86
DLC coated silicon	5 sccm	1.43
DLC coated silicon	7 sccm	1.27
DLC coated silicon	10 sccm	0.47

Following the above presented results and their analysis one can suggest that the field electron emission acting simultaneously with the thermionic emission can be considered as the main mechanisms of dielectric charging in AFM. However, other mechanisms of charge generation should be considered as well. Because the electric field is high at the dielectric/metallic substrate interface (back side of the dielectric layer in the considered configuration), injection of positive charges can occur with a rate depending on the nature of the interface. This would lead to internal positive charging with a possible transportation of these charges towards the surface. Such injection process has been observed in dielectrics above field value of about 10^7 V/m, one order of magnitude lower than the average field considered here. As a result, the AFM measurement should "probe" the negative charge deposited by the tip minus the influence of the positive charge injected in the dielectric from the back electrode. Whether this occurs or not is beyond the scope of our current discussion but it certainly must be considered in future studies.

Nevertheless, there is still to explain the positive charging of the dielectric surface when the AFM tip is at a positive potential. This can be discussed taking into account that the tip electrode at a positive voltage would act as a collector for electrons extracted from the dielectric surface. In this scheme, the dielectric surface is the electron emitter and the tip is the collector, leading to a positive charging of the dielectric. This process can be envisaged taking into account the high field value and the nature of the dielectric surface layer characterized by surface states with lower ionization energy than the bulk material. This means the extracted electrons are not those from the dielectric valence band. Surface states have been evidenced experimentally and could provide adequate centres with a low ionization potential, without the need to remove valence band electron, a mechanism energetically more demanding.

VI. CONCLUSION

Measurements of dielectric charging using AFM tips with different work function but the same curvature radius are under way to substantiate our analysis both qualitatively and quantitatively.

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