Dimensional quantization effects in the thermodynamics of conductive filaments

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Abstract
We consider the physical effects of dimensional quantization in conductive filaments that underlie operations of some modern electronic devices. We show that, as a result of quantization, a sufficiently thin filament acquires a positive charge. Several applications of this finding include the host material polarization, the stability of filament constrictions, the equilibrium filament radius, polarity in device switching, and quantization of conductance.

Keywords: nanofilament, RRAM, phase change memory, thermodynamics, dimensional quantization

1. Introduction

The role of conductive filaments (CFs) is critically important for functionality of such electronic devices as phase change memory (PCM) [1], resistive random access memory (RRAM) [2, 3], and related threshold switches; CFs are responsible for the breakdown phenomena in gate dielectrics and some other structures. It is customary to describe CFs as thin metallic cylinders with appropriate material parameters. That approach requires modifications for modern devices where CF radii \( R \) fall in the nanoscale domain, thus bringing up the phenomenon of dimensional quantization.

More specifically, the available numerical values estimated experimentally are: \( R \sim 2.3–5\, \text{ nm} \) [4], \( R \sim 1.3–2.5\, \text{ nm} \) [5], \( R \sim 0.5–1.5\, \text{ nm} \) [6], \( R \sim 5–10\, \text{ nm} \) [7], \( R \sim 3–8\, \text{ nm} \) [8], \( R \sim 5\, \text{ nm} \) [9]. Using \( \Delta = 2h^2/mR^2 \) as the energy scale for dimensional quantization (see equation (2) below) with \( R = 1\, \text{ nm} \) and the true electron mass \( m = m_0 \), one gets \( \Delta \sim 0.1\, \text{ eV} \), rather substantial on the thermal energy scale \( kT \sim 0.025\, \text{ eV} \) at room \( T \). \( \Delta \) can be much greater or smaller for the broad ranges of effective masses [10, 11] \( m/m_0 \sim 0.1–1 \) and \( R \sim 0.5–5\, \text{ nm} \).

Here we consider mostly the effects of dimensional quantization in CF thermodynamics. We show that such effects are responsible for CF equilibrium radius, nucleation barrier, and the instability of CF constrictions; in addition, we briefly revisit the concept of quantum point contacts (QPC) developed for constricted CFs [12–15]. Schematic representations of the types of objects dealt with here are illustrated in figure 1.

Note that the concept of dimensional quantization assumes long enough electron mean free path \( l \) exceeding the system linear dimensions [16], here, \( l \gg R \) (quantization along CF is not required). \( l \) can be estimated from the available data for mobility, \( \mu \sim 10–50\, \text{ cm}^2\, \text{V}^{-1}\, \text{s}^{-1} \) for various amorphous semiconductors [16–20]. Using [16] \( \mu \approx eL/(2\pi kTm) \) with \( \mu \sim 10–50\, \text{ cm}^2\, \text{V}^{-1}\, \text{s}^{-1} \) and \( m/m_0 \sim 0.1–1 \) where \( e \) is the electron charge, yields a broad range of \( l \sim 0.1–10\, \text{ nm} \). The required condition \( l \gg R \) can hold for a significant part of that range. (In the meantime, it is conceivable that the dimensional quantization will be suppressed for large radius CFs in materials with heavy effective masses and high degrees of disorder.)

Here, we will separately discuss the uniform (section 2) and constricted (section 3) CFs. Section 4 will present some applications of our findings. Conclusions are given in section 5.

2. Uniform filament

2.1. Density of states

Consider a uniform CF as a long (\( L \gg R \)) metal cylinder embedded in a dielectric and connected to the electrodes. As described in cylindrical coordinates, the 2D quantization in the radial direction produces discrete energy levels \( E_n \),
radius $R$, Fermi energy; $E_{FR}$ with $m_n$ Figure 2. Nanotechnology and without dimensional quantization.

The density of states in the CF $g(E)$ is calculated by summation of the 1D sub-band contributions $g_1(E - E_n) \Theta(E - E_n)$ for all the energy levels from 1 to $E_n = E$ and dividing the result by the filament volume $\pi R^2 L$. Here, $\Theta(x) = [1 \text{ when } x > 0; 0 \text{ when } x < 0]$ and $g_1(E) = (L/\hbar \pi) \sqrt{m/2E}$. Replacing the summation with integration yields the envelope density of states,

$$
g(E) = \frac{1}{\pi R^2} \sum_{n=1}^{E/\Delta} \Theta(E - E_n) \frac{m}{\sqrt{2 \pi \hbar}} \sqrt{E - E_n}$$

$$\approx \int_{E/\Delta}^{E/\Delta} \Theta(E - E_n) \sqrt{2 \pi \hbar}\sqrt{m} dn \frac{m}{\sqrt{2(E - E_n)}} = \frac{\sqrt{2} m^{3/2} \sqrt{E - \Delta}}{\pi^2 \hbar^3} \tag{3}$$

The latter result neglects $g_1$ spikes illustrated in figure 2 compared to the smooth envelope obtained by integration of many $g(E)$ tails. That approximation is justified when a spike value $g_1(E - E_n)$ corresponding to a thermal energy $E - E_n \sim kT$, is smaller than that of the envelope in equation (3), which reduces to the inequality,

$$kT \gg \pi \frac{\Delta^2}{16 E - \Delta} \tag{4}$$

Since $E - \Delta \approx E_F$, it is well obeyed for practical temperatures, say $T \gtrsim 300$ K.

Equation (3) naturally predicts the standard 3D density of states $g(E) \propto \sqrt{E}$ when the CF radius is large, i.e. $\Delta \to 0$. For finite $\Delta$, it follows from equation (3) that dimensional quantization shifts the standard 3D energy spectrum up by energy $\Delta$, as illustrated in figure 2 (right). That shift makes electron energy levels in a band $\Delta$, and, in particular the Fermi level in CF, $E_{FR}$ be raised in energy above the system Fermi energy $E_{F0}$. Such levels will thus lose their electrons to the lower energy states outside the CF, rendering it with a net positive charge (confined to its surface layer as always for metals).

2.2. Filament charging
The above positive charge induces an electric potential self-consistently shifting the electron energy levels in CF down, which eliminates the difference $\Delta$ between the CF $E_{FR}$ and the system $E_{F0}$ Fermi energies. That energy shift can be described in the terms of band bending caused by the above potential as illustrated in figure 3. The confinement related charging here is conceptually similar to the much earlier considered surface effects [23].

Denoting by $\Lambda$ the CF’s linear charge density, the electric field and potential outside CF become respectively

$$\mathcal{E} = \frac{2\Lambda}{\kappa r} \quad \text{and} \quad \phi = (2\Lambda/\kappa) \ln(R/r) \tag{5}$$

at distances $r$ smaller than the screening length, $r_s$, where $\kappa$ is the dielectric permittivity. We approximately account for the screening by cutting off the electric potential perturbation at $r = r_s$, which provides the downward electron energy shift $(2\Lambda e/\kappa) \ln (r_s/R)$. Self-consistently setting the latter equal $\Delta$...
Figure 3. Top: the energy band cross section of CF and its surrounding host. The difference $\Delta = E_{FR} - E_{F0}$ is balanced by the band bending shown for the conduction band edge $E_c$ induced by CF charging. Bottom: a real space filament cross section with surface positive charges.

yields,

$$\Lambda = \frac{\kappa \Delta}{2e \ln(r_s/R)}.$$  \hspace{1cm} (6)

The screening radius $r_s \sim L \sim 10$ nm is determined by distance $L$ between the metal electrodes due to their electron redistribution. The redistribution is dictated by the condition of tangential component of the electric field vanishing in the presence of charged CF [24]. That screening mechanism does not depend on material properties and should not be confused with the Debye screening. Of course, a degree of the dielectric host polarization remains (as discussed in section 4.1 below) providing some additional screening, but it will not have any significant effect on $r_s$.

It is instructive to estimate the ratio of additional charge caused by quantization over the total electron charge in a neutral CF. That ratio is given by

$$\frac{\Lambda}{n_0 \pi R^2} = \frac{a_B}{R} \left[ \frac{1}{n_0 R^3 \pi \ln(r_s/R)} \right]$$

with $a_B = \frac{\hbar^2 \kappa}{me^2}$, \hspace{1cm} (7)

where $n_0$ is the intrinsic electron concentration in CF and $a_B$ is the effective Bohr radius. Our numerical estimates utilize the parameters characteristic of CFs in HfO$_2$ based RRAM: $R \sim 1$ nm [5, 9], the effective electron mass [10] $m \sim (0.1-1)n_0$, and $\kappa \sim 25$. Also, we assume $n_0 \sim 10^{21}$ cm$^{-3}$ corresponding to the typical moderate metal conductivity. With these parameters, equation (6) yields the number of electrons in a single CF, $\Lambda L/e \sim 5-50$ and $n_0 \pi R^2 \sim 3$. Two conclusions can be made: (1) the relative change of electron concentration in CF due to dimensional quantization may be not small, $\Delta Q/Q \sim 2-20$ for a 1 nm radius CF, and (2) it is likely that at least several electrons are present within the CF, so charge quantization is not important.

One additional condition of CF charging is that its related decrease in electronic energy,

$$F_\Delta = g(E_F)(\Delta^2/2)\pi R^2L$$ \hspace{1cm} (8)

exceeds the electric field energy

$$F_e = \int \frac{\kappa E^2}{8\pi} d^3r = \frac{\Delta^2 \kappa L}{4e^2 \ln(r_s/R)},$$ \hspace{1cm} (9)

where we have used $\Lambda$ from equation (6) and treated the logarithm in the integrand as a constant. The resulting inequality,

$$2\pi g(E_F)(\kappa/\kappa)R^2 \ln(r_s/R) > 1$$ \hspace{1cm} (10)

obeys for all practical parameters.

Note that CFs can be charged regardless of dimensional quantization due to the difference $\delta E_F$ between CF and the host material Fermi energies. That additional linear charge density, $\Lambda'$, related to the CF electric capacitance [25] is given by equation (6) with $\delta E_F$ instead of $\kappa \Delta$. While typically insignificant in high-$\kappa$ materials, $\Lambda'$ can be easily accounted for by redefining $\Lambda \rightarrow \Lambda + \Lambda'$.

3. Constricted filament

If the CF radius $R$ varies along the CF $z$-axis forming a hourglass or truncated cone shape, an analytical description is possible when the dependence $R(z)$ is gradual, $|dR/dz| \ll 1$. In that case, the adiabatic approximation can be employed [26] making the transversal quantization $z$-dependent, $\Delta(z) = 2h^2/mR^2(z)$. The longitudinal (along $z$) dynamic is then described by the Schrödinger equation with the effective potential $\Delta(z)$ forming an approximately parabolic barrier at the constriction, which provides settings for QPC [26] and is illustrated in figure 4 (left). The negative curvature of QPC parabolas can be estimated as

$$K_{QPC} = -8h^2/(mR^2L^2)$$ \hspace{1cm} (11)

assuming that $R$ decreases by $\sim 50\%$ over length $L/2$.

In the same approximation, CF charging becomes $z$-dependent through equation (6) with $\Delta = \Delta(z)$, hence, a greater positive charge density around constriction. Along such non-uniformly positively charged CF, electron energy will decrease towards the constriction. It is illustrated in figure 4 (right) with positive curvature parabolas. Using
standard electrostatics and $\Lambda$ from equation (6), the curvature can be estimated as

$$K_{\text{CHARGE}} = 4\kappa h^2/[\ln R^2 L^2 \ln(r_i/R)].$$

(12)

It follows that $K_{\text{QPC}}$ and $K_{\text{CHARGE}}$ are more or less comparable in absolute value. However, their mutual cancelation would be a sheer coincidence: either negative or positive curvature will dominate.

We conclude that linear charge density increasing towards the constriction is a characteristic feature of non-uniform CF. While the analysis here was carried out in the adiabatic approximation, $dR/dz \ll 1$, its results should remain valid qualitatively even when the latter inequality is not strong.

4. Applications

Here we consider several applications of the above theory, though it is not intended to provide any complete explanation of RRAM or other device functionality (for which we refer to our recent work [25]). Our consideration below is limited solely to possible effects of dimensional quantization and its related CF charging.

4.1. CF field, polarization, and instability

For a 1 nm radius CF, the lateral field strength at its surface is estimated as $\mathcal{E} = 2\Lambda/(\kappa R) \sim 1$–10 MV cm$^{-1}$ given the above $\Lambda L/e \sim 5$–50. That strong electric field can lead to certain effects through (i) the surrounding material polarization and (ii) significant increase of the system free energy.

(i) Far enough from the electrodes, the radial field of CF can polarize the host by aligning ferroelectric domains (e.g. in doped HfO$_2$ films [27]) or moving charged defects (say, charged vacancies in HfO$_2$). Such a polarization will affect the nonvolatile memory resistance by contributing to the electric potential around CF and altering its charge $\Lambda$ (see figure 3). That effect is described as the decrease $\delta \mu$ of CF screening radius, which, through equation (6), translates into the relative decrease in resistance estimated as $\delta r_S/\bar{r}_S$. The polarization can be changed by external bias [25] or Joule heat in writing cycles. Assuming the initial polarization is partially destroyed, our analysis then predicts gradual increase in resistance in the course of endurance cycling, consistent with some observations [28, 29].

(ii) According to equation (9), the field strength and energy increase, respectively, as $R^{-2}$ and $R^{-4}$ with filament thinning. That points at the fundamental instability of CF constrictions (where $R$ is a minimum), tending to break or heal in order to decrease the energy. In other words, the CF neck charge creates the highest electric field energy conducive of eliminating that neck.

4.2. Equilibrium radius

While the field energy of equation (9) favors CF radius increase, its surface energy $2\pi RL\sigma$ has the opposite trend, where $\sigma$ is the interfacial energy. The sum of these two contributions in CF free energy $F$ has a minimum, $dF/dR = 0$, that determines the equilibrium radius $R_0$ of CF through the relation,

$$R_0 = \left[ \frac{2h^4\kappa}{\sigma e^2 m^2 \ln(r_i/R_0)} \right]^{1/5}.$$  

(13)

Given a relatively small exponent of 1/5 on the right hand side, $R_0$ under the logarithm can be approximated by any ‘reasonable’ value of the order of 1–10 nm. With that in mind, calculations using the typical $[30] \sigma \sim 100$ dyn cm$^{-1}$ and other relevant parameters mentioned above yields $R_0 \sim 1$ nm, which is consistent with the smallest of the observed CF radii listed in section 1 above. Also, due to the small exponent of 1/5, the latter estimate is not very sensitive to the material parameters; hence, $R_0$ not varying significantly between different materials. Therefore, $R_0$ can serve as an estimate of the ultimate size limit of filamentary RRAM devices: thinner CFs would increase electric field energy enough to overcome the surface tension and expand the radius. Along the same lines, $R_0$ can be interpreted as an estimate for the ultimate minimum radius of CF constrictions.

Note that $F_\Delta$ from equation (8) does not contribute to the above free energy because of the cancelation with the increase of electron energies illustrated in figures 2 (right) and 3. Also, taking into account the contribution from the chemical potential differentials $\delta \mu$ between the CF and host bulk material does not have a significant effect on the estimate in equation (13) for the typical $\delta \mu$ estimated in [25].

4.3. Field induced nucleation

Consider the nucleation of metallic needle-shaped particles in the electric field of high strength $\mathcal{E}_0$. The nucleus free energy is given by,

$$F = F_\mathcal{E} + 2\pi RL\sigma - L^3 \kappa \mathcal{E}_0^2,$$

(14)

where $F_\mathcal{E}$ is given by equation (9) and the last term represents the polarization electrostatic energy gain written here to the accuracy of insignificant multipliers [31, 32]. The term $F_\mathcal{E}$ distinguishes our analysis from the preceding work. The critical nucleus dimensions $R_0$ and $L_0$ are obtained from $dF/dR = dF/dL = 0$. It is straightforward to see that $R_0$ is the same as in equation (13), while $L_0$ and the nucleation barrier are a factor of $\sqrt{2}$ smaller than that of preceding theory. We conclude that through CF charging the dimensional quantization will exponentially increase the field induced nucleation rate.

4.4. Bias polarity effects

A new feature in field induced nucleation arises when we add the term, $E_0 AL^2/2$, for the energy of a linear charge $\Lambda$ in the external field. Because the sign of $\Lambda$ is predetermined, that term will make CF nucleation dependent on the field polarity. It was observed indeed that PCM operations are bias polarity dependent [33–35], although other explanations are possible, such as the Thomson effect or polarity dependent elemental segregation [35].
We note another polarity effect for the bias induced CF charge. It was shown [25] that many RRAM related observations can be explained by accounting for CF charging and the related radial electric field in response to an external bias. In particular, it was shown that when SET and RESET voltages across the device are determined by the condition that the atomic motility becomes respectively frozen and unfrozen, then the absolute values of those voltages will be equal. (While beyond the scope of this paper, we note parenthetically that the latter equality takes place only for bipolar, but not the unipolar RRAM where SET and RESET processes are physically different.) Here, we add that taking into account \( \triangle \) from equation (6) will increase or decrease the absolute value of CF charge corresponding to a given external bias depending on bias polarity; CF thinness will aggravate the asymmetry. That can result in somewhat unequal absolute values of SET and RESET voltages for bipolar RRAM devices. Furthermore, based on the discussion in section 4.1, one can expect lower SET and RESET voltages for constricted CFs compared to the uniform CFs of the same average radius.

### 4.5. Quantum conductance

In the QPC model [26], the CF has a constriction; hence \( R = R(\zeta) \) as is depicted in figure 1 and \( \triangle(z) = 2k^2/mR^2(z) \) adiabatically depends on the CF’s longitudinal coordinate \( z \). The transverse quantization is approximated by \( E_n = n\Delta(0) - \alpha z^2 \) presenting a set of parabolic barriers in figure 4(left) where \( \Delta(0) \) and \( \alpha \) are two constants. When the electric bias \( V = |E_F1 - E_F2|/e \) between the contacts with Fermi levels \( E_F1 \) and \( E_F2 \) changes, so does the number of conductive channels \( N_{\text{ch}} = [eV/\Delta(0)] \) where \( [X] \) means the greatest integer in \( X \). Assuming ballistic electron transport, the CF’s conductance is given by \( G_0N_{\text{ch}}t \) where \( G_0 \equiv 2e^2/\pi\hbar \approx (12.9 \text{ k}^2 \text{f}^{-1}) \) is the quantum conductance and \( t \) is the barrier transparency due to tunneling or activation. Recent work [12–15] attributed some RRAM observations to QPC model.

When the positive curvature dominates, the conductance quantization takes place as long as the transport remains ballistic. Indeed, in spite of the energy modulation, the product of electron velocity \( v = \sqrt{2E/m} \) times \( g(E) \propto 1/\sqrt{E} \) is energy independent, as required by the derivation [36, 37]. If the transport is diffusive inside the classically allowed region of CF in figure 4 (right), then two triangular barriers by the channel edges exhibit additive QPCs [36, 37].

The triangular barrier transparency \( t \) is due to thermally assisted tunneling [38]. Omitting the details, our analysis yields such a CF conductance

\[
G = G_0N_{\text{ch}} \exp\left[-\frac{\Delta}{kT} + \frac{\hbar^2k^2L^2}{96m^2(\alpha kT)^3}\right]
\]  

(15)

exponentially decreasing with CF length and increasing with its radius; non-Ohmicity can be introduced with non-ideal contacts. Equation (15) predicts the stepwise changes in conductance and shows that quantum conductance can be explained when size dependent quantization takes place even without the constriction feature.

### 4.6. Variability

Dimensional quantization will contribute to variability of filamentary nano-devices, with amorphous structure. Equation (1) predicts that random deviations from a perfect circular cross section will create variability in the quantization energy \( \Delta \) and its related effects. As an example, the number of conductive channels and the observed steps in conductance of equation (15) will vary between different samples.

### 5. Conclusions

In conclusion, we have shown that dimensional quantization results in charging of conductive nano-filaments. The linear charge density is inversely proportional to the square of filament radius. It is a maximum around the filament constriction (if any). The filament charging can exhibit itself in several effects of practical importance, such as strong polarization of the surrounding material, thermodynamic instability of filament constrictions, the notion of equilibrium filament radius and ultimate minimum size, polarity effects in field induced nucleation, and quantization of conductance without the feature of constriction.

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