

## Probing Quantum Transport by switching Anchor Positions

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**Abstract:** A novel approach towards probing quantum transport through the molecular junction comprising of benzene molecule terminated by thiol endgroups stringed to two gold electrical contacts was suggested by switching the anchor-thiol endgroup positions from meta to para and their corresponding stress and conductance behaviour was computed. Similar molecular junction was formed from anthracene and its charge transport characteristics-Current-Voltage, Conductance-Voltage, Differential conductance- Voltage response were examined at room temperature. The effect of variation of the bond angle on stress and conductance of benzenedithiol (BDT) and anthracenedithiol (ADT) molecules with respect to gold electrodes was anatomized using Extended Huckel Theory (EHT) based semi-empirical model. The simulated results revealed that the para position exhibited higher conduction on account of constructive interference in comparison to meta position whereas latter exhibited higher stress because of destructive interference that led to realignment of  $\pi$ -cloud and hence more stress on the system under observation. This behaviour was demonstrated in both the molecules. The charge transport properties of ADT were elucidated once by considering the endgroup anchor at para position and then at meta position and we concluded that the manipulating these geometric parameters led to quantum interference which affected the overall electron transport and provided us with an insight about the phenomena at nanometre-scale, which could be helpful towards fabricating molecular devices based on these molecules.

**Keywords:** Molecular Engineering, Molecular Electronics, Anchor Group, Tunneling, Quantisation, Discreteness, Quantum Interference.

### 1. INTRODUCTION

The on-going miniaturisation of the silicon electronic technology will reach its physical limit in the foreseeable future [1], hence alternative technologies are becoming very attractive that can allow us to further reduce the size of electronic components up to nanolevel. Kuhn described a visionary idea to design electronics from molecular structures known as "Molecular Engineering" [2], now widely termed as "Molecular Electronics" [3, 4]. Compared with the traditional semiconductor (SC) technology, this new approach changed both the operating principles and the materials used to fabricate the devices [5]. The concept of molecular junctions has been recognized as a subset of the more general area of molecular electronics [6]. Molecular junctions reported till date vary significantly in size, number of molecules, anchor groups, temperature and the nature of the electrical contacts [7], but they all share one core principle i.e. when a molecule or collection of parallel molecules is oriented between two electrical contacts, their molecular properties affect or control electron flow through the junction as shown in figure 1A). In atomic sized contacts, the classical ohm's law ( $V=I.R$ ) is not applicable since we are now dealing with quantum regime, where current flows as a transmission process

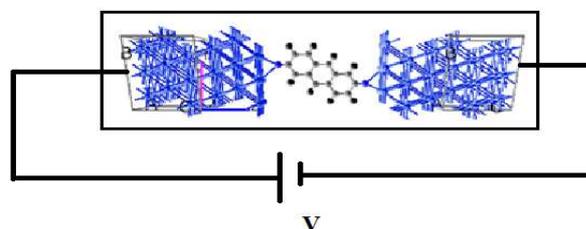


Figure 1A): Molecular junction comprising electrodes on both sides and in between is the scattering region comprising the molecule (Anthracene).

and described by Landauer formula[8]:

$$G_0 = 2e^2/h = 77.5 \mu S \quad (1)$$

where  $e$  is the electronic charge and  $h$  is the Planck's constant. The term "molecular wire" encourages description of electron transport through molecules in term of metallic ("Ohmic") conductivity. In fact, none of the molecular bridges studied to date show Ohmic behavior: electron transfer through "molecular wires" occurs by electron tunneling, and organic wires are "conducting" in the same sense that polyethylene is conducting. The "molecular wires" facilitate electron transport relative to vacuum, but are not similar to metals either in the magnitude of their conductivity or in the mechanism of this conduction. The use of

molecular properties to make electronic devices was first envisaged by Aviram and Ratner [9] in a theoretical paper in 1974. Because there were no technologies that could establish electrical contacts across individual molecules, experimental investigations of the fundamental processes involved in electron transfer through molecules have focused on liquid-phase systems [10]. Only in the late 90s has the combination of nanotechnology, scanning probe microscopies, and methods to form electrically functional connections to metal surfaces triggered the fabrication of metal–molecule(s)–metal junctions, and opened the door to experimental “molecular electronics”. Different type of junctions have been used to sandwich molecules (several, a few, or individual molecules) between two metal surfaces, and to measure their electrical properties [11]. Junctions that include organic molecules of modest structural complexity have showed properties that suggested that it may be possible to build devices that mimic the function of electronic components (conductors, transistors, rectifiers, logic gates). When electric current flows through a single molecule, the conductance is mainly decided by the quantized electronic structure of the molecule. The presence of accessible charge states near the electrode's Fermi level can help electron transport through a molecule. A wide variety of current–voltage characteristics can be created by structural changes in the molecule. Since molecules can exhibit a wide range of molecular orbital patterns, there should be a corresponding wide range of  $I$ - $V$  transfer functions. Control over the electronic and optical properties of semiconductors (SCs) is a major goal in science and technology. Semi conductor nanoparticles provide us such an opportunity, due to the three-dimensional size quantization they exhibit, which modifies their electronic properties like band gap with particle size [12]. The transport properties in molecular junctions are dominated by some effects of quantum origin, such as: tunnelling, quantization of molecular energy levels and discreteness of electron charge and spin. However, recently it was pointed out that also quantum interference effects can lead to substantial variation in the conductance of molecule-scale devices [13]. Interference reveals the wave nature of the electrons passing from the source to drain through the molecule. Recently, the effect of quantum interference was observed for photoinduced electron transfer across linearly conjugated and cross-conjugated acyclic bridges[14] but to the best of our knowledge, there are only two observations of the influence of cross-conjugation in tunnelling junctions, both of which concluding only that meta contacts lead to less current passing through a single-molecule junction compared with para contacts[15-17]. Another study observed a decrease in the conductance of single-molecule junctions that are photochemically switched between linearly conjugated and cross-conjugated states;

however, the authors did not ascribe this effect to cross-conjugation or quantum interference[18]. The properties of the contacts between the molecule and the leads are also important and they strongly affect the overall conductance of a single molecule device. In this paper, we have presented the view of terminating molecule with different endgroup positions- meta(1,3) and para(1,4) positions as shown in figure 1B). the phase difference between the two paths around the ring is 0 in para configuration whereas  $\pi$  in meta configuration. The variation of interference conditions is achieved by changing the connection between the molecule and electrodes.

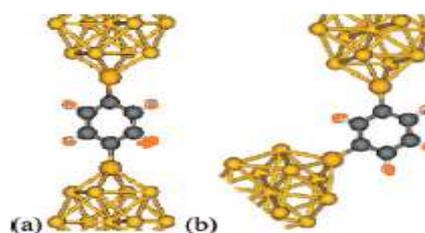


Figure 1B): a) Para and b) Meta geometries

## 2. Modelling and simulation

The molecular devices, we selected for our study comprised of a) Benzene molecule b) Anthracene molecule both terminated by thiol group and stringed to two semi-infinite gold electrodes having miller indices of [111] in two different positions- Meta and Para as shown in figure 2 and 3. In both cases, the molecule under consideration faced different interference conditions and so we observed distinct changes in transport characteristics. We first performed geometric optimization using Guassian-03 till the force and the stress converges to 0.1 eV/Å<sup>0</sup> and the further transport calculations were carried out using the Atomistix Toolkit (ATK) software 12.2.0, which is based on the non-equilibrium Green's functions (NEGF) that employs Transiesta-C. Device Extended Huckel with cerda parameters were employed for tight binding approach with mesh cut-off set to be 100 Hartrees as shown in figure 4. BDT has been extensively used to test quantum transport because the conjugated  $\pi$  ring should offer delocalized electronic states beneficial to transport and the sulphur atoms at both sides of electrodes form strong coordination bonds to the gold electrodes. Bias voltage 1V was applied on the both left-right metallic electrodes and the electric current flowing through the device was obtained from the transmission function T as [19]:

$$I(V) = 2e/h \cdot \int T(E) [f(E - \mu_s) - f(E - \mu_D)] dE \quad (2)$$

Where  $f$  denotes Fermi distribution functions for with chemical potentials.

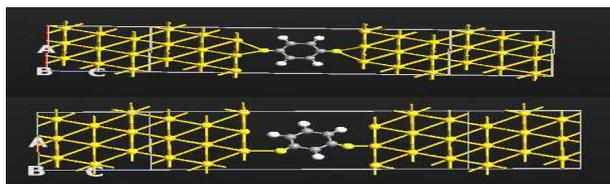


Figure 2: Benzenedithiol (BDT) stringed between two gold electrodes in: a) Para b) Meta position.

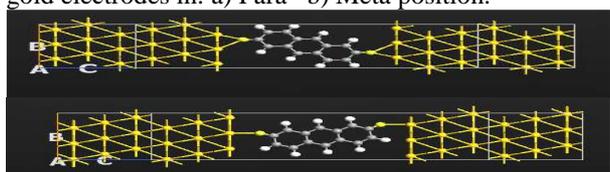


Figure 3: Anthracenedithiol (ADT) stringed between two gold electrodes in: a) Para b) Meta position.

$$\mu_{S/D} = E_F + - e V / 2 \quad (3)$$

where 'S' and 'D' referred to as the source and drain, respectively. The conductance is given by:

$$G = \frac{1}{2} G_0 [T(\mu_S) + T(\mu_D)] \quad (4)$$

### 3. Results and discussions

After the initial geometry optimization as the force and stress on the molecule under observation approached  $0.1 \text{ eV/\text{Å}^0}$ , the transport metrics of current and conductance were measured separately for the following different variations as discussed above in figures 2 and 3.

#### Stress

The stress was measured by constraining endgroup i.e. thiol being placed in both para as well as meta positions of Benzene molecule firstly and then Anthracene molecule coupled to Gold electrodes and the results obtained were different due to the varied quantum interferences on account of localization of charges resulted by linear and cross conjugation respectively. Para positions are linearly conjugated, whereas the meta position is cross-conjugated. Linearly conjugated molecules must always have an even-length backbone, an even length backbone with a single odd-length side chain must be a radical, and an odd-length backbone with a single odd length side chain is a cross-conjugated molecule. Although the meta position is in the same  $\pi$  circuit and one carbon atom away from either the para positions, there is no resonance structure that places the lone pair of a substituent on the meta position of the molecule rings.

Stress in energy per unit element i.e.  $\text{eV/\text{Å}^3}$  is given by the derivative of the total energy, E with respect to a strain  $\epsilon_{\alpha\beta}$  [19]:

$$\sigma_{\alpha\beta} = \frac{1}{V} \frac{dE}{d\epsilon_{\alpha\beta}} \quad (5)$$

In this equation, V is the volume of the system and the strain tensor displaces positions by [19]

$$R_i^j = \sum_{\alpha\beta} (\delta_{\alpha\beta} + \epsilon_{\alpha\beta}) R_{i\alpha}^j \quad (6)$$

The values of stress of benzenedithiol in para and meta positions with varying bonding angle are plotted in figure 4. We found that the values of stress increases with increasing bonding angle for both cases but when we compared the stress of para and meta, we observed that stress of meta benzenedithiol is more as compared to that of para benzenedithiol as bond angle is increased from 0 to 30 degrees whereas for 40 degrees, meta showed decrease in stress and again increased for 50 degrees. Similarly, we computed values of stress of anthracenedithiol for para and meta positions shown in figure 5. We here again observed similar behaviour that stress of meta position is higher than para anthracenedithiol but here, meta and para showed continuous rise in stress. The values of stress of para and meta ADT at 40 degrees were reported as  $-0.294 \text{ eV/\text{Å}^3}$  and  $-0.278 \text{ eV/\text{Å}^3}$  respectively. The stress reported for bond angle 0 degree by meta BDT was  $-0.31 \text{ (eV/\text{Å}^3)}$  whereas para BDT was  $-0.33 \text{ (eV/\text{Å}^3)}$  and that of meta ADT was  $-0.37 \text{ (eV/\text{Å}^3)}$  whereas para ADT was  $-0.39 \text{ (eV/\text{Å}^3)}$ . Stress for meta device was found to be higher than that of para in both epochs. This increase in stress by altering thiol group positions from para to meta was because of the quantization of molecular energy levels and the distribution of the electronic charges within the system. The alteration of endgroups caused the varied quantum interference because of which there will be a definite effect on conduction which was observed then.

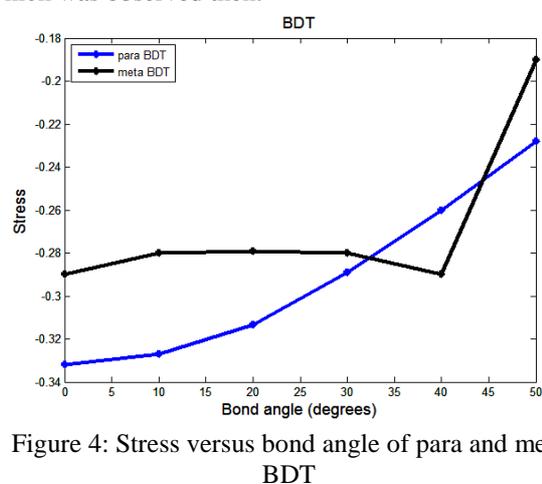


Figure 4: Stress versus bond angle of para and meta BDT

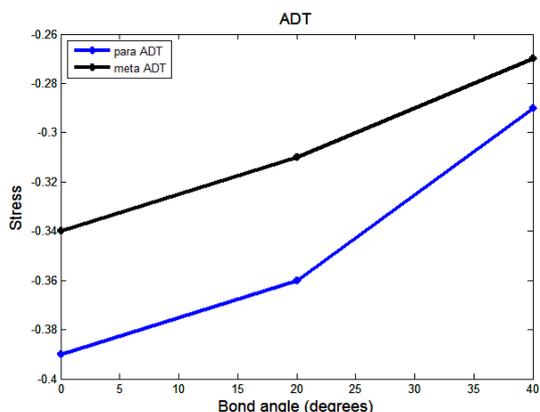


Figure 5: Stress versus bond angle of para and meta ADT

The conductance values computed at different bonding angles for both para and meta positions are shown in table 3.

### Conductance

The conductance of both benzenedithiol and anthracenedithiol in para and meta positions coupled to Gold electrodes though was measured under bias voltage 1V. The observed values of the conductance took into consideration quantum interference, whether constructive or destructive. Quantum interference in coherent transport through single molecular rings provides a mechanism to control the current in molecular electronics. It is a direct consequence of the topology of the  $\pi$  conjugated system.

From table 1 and figure 6, we observed that the conductance of para benzenedithiol was greater than meta benzenedithiol. Hence, we can say that para configuration exhibited constructive quantum interference whereas meta counterpart exhibited destructive quantum interference.

The electron wave travel through all available paths and upon arrival at the point of interest, they are superimposed. When both paths are of equal length both waves will arrive with the same phase, thus interfering constructively. On the other hand, when one wave has a phase shift of  $\pi$ , destructive interference will occur resulting in zero transmission. This is the reason why conductance of para configuration is more than that of meta counterpart. Similar simulation was performed with Anthracenedithiol (ADT) where we again observed that para ADT showed higher conductance than that of meta ADT figure 7. The conductance of para ADT was found to be 0.54 times the quantized conductance whereas that of meta counterpart was 0.34 times quantized conductance. This behaviour of increasing low bias conductance arises from the shift of the HOMO towards resonance with the Fermi level in symmetrical configuration which is para whereas in the asymmetric or meta case, positive bias shifts the

HOMO level away from the Fermi level, effectively postponing current onset and hence lower conductance. Destructive quantum interference or we can say that the transmission is suppressed by lowering the overlap between regions of the  $\pi$  system, both resonant and off-resonant transport are disrupted across the energy range. But, one interesting observation was that in BDT, conductance of both para and meta BDT increases with increase in bond angle whereas we noticed opposite behaviour in ADT where conductance showed

Table 1: Conductance versus bond angle for para and meta BDT

Bonding angle (degrees)	Para Conductance ( $\mu$ S)	Meta Conductance ( $\mu$ S)
0	19.66	0.8174
20	21.20	0.800
40	28.6778	1.46

decrease with increasing bond angle. A change in the configuration of connection between the molecule and two electrodes results in variation of the interference conditions and obvious changes in the transmission function. Green's function represents the orbital overlap across the molecule divided by the energy distance of the Fermi level from a molecular resonance. In the following equation our simplified representation assumes that the central molecule and the contacts share the commutative property as:

$$[\sum_i H_{molecule}] = 0 \quad (7)$$

$$G_{\phi_L, \phi_R}(E) = \sum_{\mu} \frac{\langle \phi_L | \mu \rangle \langle \mu | \phi_R \rangle}{(E - E_{\mu} - \Sigma_{\mu})} \quad (8)$$

In this case the conductance  $G(E)$  is equal to the overlap of the molecular orbitals and the terminal atomic orbitals on the left and right electrodes divided by the energy difference between  $E$  and a molecular resonance  $E_{\mu}$  and the self-energy  $\Sigma_{\mu}$ . Increasing conductance with changing bond angle can be related with the overlapping and increasing gap between orbitals as in BDT and ADT respectively. With increase in bond angle, BDT showed increasing conductance whereas ADT showed fall in conductance. The overlapping or partial merging of orbitals in BDT decides the strength of bonding; greater the overlap,

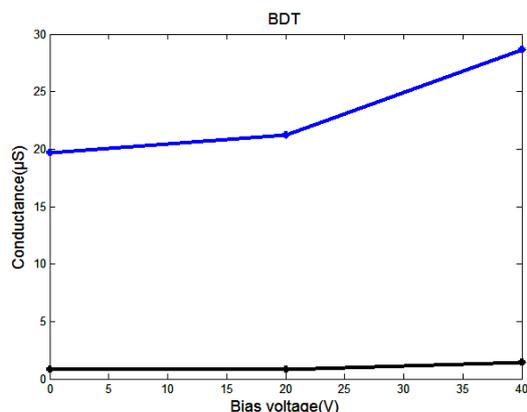


Figure 6: Conductance versus bond angle of para(blue) and meta(black)BDT

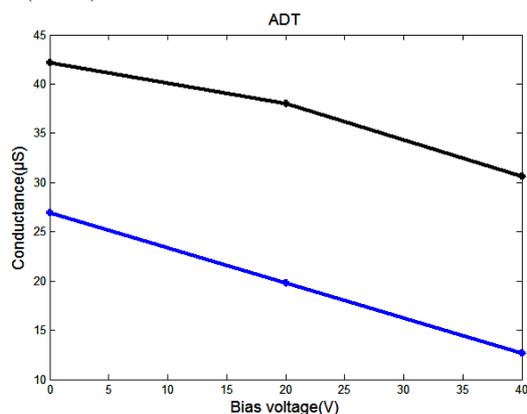


Figure 7: Conductance versus bond angle of para(black) and meta (blue)ADT

Table 2: Conductance versus bond angle for para and meta ADT

Bond angle (degrees)	Para Conductance (μS)	Meta Conductance (μS)
0	42.2	26.91
20	38.1	19.83
40	30.6	12.73

stronger is the bonding and hence more is the conductance whereas in case of ADT, increasing bond angle disturbed the pi-electron cloud such that the orbitals instead of merging showed large gap causing poor bonding and hence reducing conductance. Hence, we can say that the conductance can be proliferated by increasing or decreasing the bond angle of conjugated molecules. We observed that 40 degrees is the optimised bond angle for BDT whereas for ADT, it was observed to be 0 degrees. For BDT, we can generalise directly proportional relation of conductance and bond angle whereas inversely proportional relation for ADT. We also observed the different charge transport characteristics for para and meta anthracenedithiol and

the simulated values of current and conductance for bias voltage varying from -2 to 2V.

Table 3: Current and conductance versus Bias voltage of Para ADT

Bias voltage (v)	Current (nA)	Conductance (nS)
-2	-45498	462.68
-1.6	-44531	15120
1.6	56632	33469
2	59073	25041

Table 4: Current and conductance versus Bias voltage of Meta ADT

Bias voltage (v)	Current (nA)	Conductance (nS)
-2	-1607	39.7
-1.6	-1483	1099
1.6	28073	3108
2	31859	3470

As the bias voltage was increased from -2V to positive bias variation, we observed increase in current and hence conductance for both meta and para ADT. The current- voltage characteristics showed metallic (ohmic) conductivity when the Fermi level of electrodes comes in resonance with the energy of HOMO-LUMO molecular orbitals[20]. The computed values of current and conductance were reported in tables 3 and 4 where para ADT exhibited 25041nS conductance whereas that of meta ADT was 3470nS at highest bias voltage 2V taken into consideration. V-I, Conductance- Voltage and differential conductance-voltage characteristics revealed higher values of para configuration than that of meta with bias voltage variations from -2 to 2V. Figures 11 and 12 show the I-V and differential conductance versus voltage curves for meta and para anthracenedithiol (ADT). We observed that I-V curves in meta and para ADT showed similar linear curves where current increased with increase in bias voltage whereas dI/dV characteristics of both configurations of ADT were observed to be contrasting. The response of differential conductance with bias voltage of meta and para ADT was again found to be similar where we found increase during negative bias and fall during positive bias region. The magnitude of the current flowing through the device is very sensitive on the manner of attachment between the molecule and metal surfaces. Large values of the current and dI/dV are predicted for device of para configuration, while reduction of the current and dI/dV by orders of magnitude is expected for device of meta. Such reduction is caused by destructive interference taking place in meta configuration and constructive interference in para configuration. When upper and lower routes in meta configuration yield phase difference  $\pi$  within HOMO-LUMO gap, partial waves

interfere destructively. This is the reason why  $T(E)$  shows minimum and hence lower values of current, conductance and differential conductance in meta ADT.  $T(E)$  shows strong minimum when the partial waves have equal weight.

### Transmission spectra

For different values of bias voltages from -2 to 2V, the effect on transmission spectrum was also analysed for both meta and para ADT and shown in figures 8 and 9 respectively. The coupling between the electrodes and the molecule leads to overlapping of the hybridized orbitals and a change in HOMO-LUMO gaps. The stronger the coupling, more the orbitals are broadened[20]. In order to know that which bias voltages supports strongest peaks or least HOMO-LUMO gaps, the transmission spectrum is plotted against bias voltages with energy ranging from -5eV to 5eV and the energy zero parameter is adjusted as average Fermi level. The peaks were shifted and broadened by the fact of the coupling with the electrodes just like discrete energy levels of the molecule. A change in the configuration of connection between anthracene and two electrodes resulted in shifts in the resonance peaks and in reduction of their height. Well-separated energy levels gave rise to distinct peaks in the spectrum, while molecular levels close in energy can overlap and eventually interfere causing reduction of resonance peaks due to destructive interference in meta ADT. The transmission spectra of meta and para ADT shown clearly demonstrated more transmission peaks and hence high conduction in para ADT whereas that of meta ADT showed more flatness, hence lesser transmissions and lower conduction.

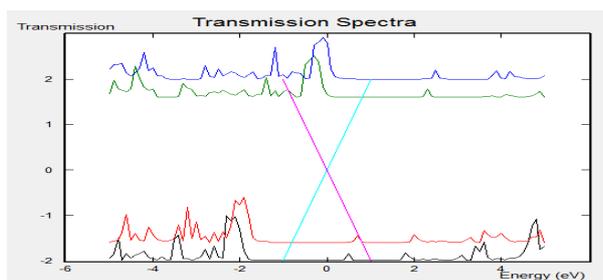


Figure 8: Transmission Spectrum of meta ADT

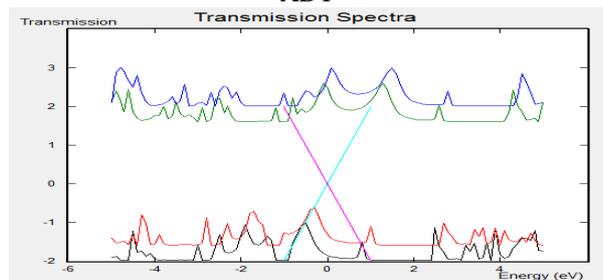


Figure 9: Transmission Spectrum of para ADT

### Stress versus conductance

From the values of stress and conductance computed for para and meta benzene dithiol, we found the higher conductance of para (symmetrical) configuration whereas lower value for meta (asymmetrical) configuration. Here, we took the two parameters of stress and conductance and derived their relation for both meta and para BDT. The conductance firstly rises to a point which is the most optimised point of  $-0.291\text{eV}/\text{Ang}^{**3}$  beyond which the conductance showed falling slope with increase in stress. Here the relation between stress and conductance for meta BDT can be generalised as:

$$\text{Conductance} \propto 1/\text{Stress} \quad (9)$$

Conductance-Stress relation for para BDT can be expressed as:

$$\text{Conductance} \propto \text{Stress} \quad (10)$$

If we compare the optimised stresses of both para and meta BDT, we found the stress of meta BDT and para BDT ( $-0.291\text{eV}/\text{Ang}^{**3}$  and  $-0.26\text{eV}/\text{Ang}^{**3}$  respectively) clearly indicated compressive and expanding nature of stress and hence lower conductance (destructive interference) in meta BDT and higher conductance (constructive interference) in para BDT.

## 4. CONCLUSION

BDT and ADT based molecular junctions were studied using Extended Huckel Theory where the stress, current, conductance and transmission spectra were computed by geometrical alternations i.e. by switching endgroup positions from meta to para and concluded that the para position exhibited higher conduction than its meta counterpart. The stress versus bond angle for meta and para concluded higher stress of meta device than that of para device because of varied quantum interference. Constructive quantum interference was observed in para device and destructive interference in meta device because of which high conduction and more transmissions were observed in para ADT. Hence, we can say that there is a striking variation in conductance of molecular devices with the alternations in the geometry (here anchor positions). Since, the stress- conductance relation showed constructive and destructive quantum interference in para and meta geometries respectively, we conclude that such devices can be a boon in deploying piezoelectric technology via molecular devices. The effective destructive quantum interference in meta configuration can be used to control electric current through single molecular devices whereas constructive quantum interference can be related to ON/OFF ratio. By careful design of molecular structure, quantum interference effects can be exploited

to make single molecules perform binary logic functions, such as NOR, AND, XOR operations or even more complex digital circuits can be made such as half adders. We firmly believe that the model suggested in this research paper can be applied for other aromatic molecules and the results can be made reproducible within the sufficient level of accuracy.

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