Initial investigation into the effect of blending multi-walled carbon nanotubes (MWNT) with CdTe type (II) quantum dots to produce Light Emitting Diodes

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ABSTRACT

We studied the high-bias conductance versus voltage G-V characteristics of a multi-walled carbon nanotube (MWNT) mixture with type (II) CdTe quantum dots and observed a sudden conductance increase when the bias exceeds a certain threshold voltage (V_{TH}). Both MWNT and single walled carbon nanotubes (SWNT) are severely entangled and possess extremely low solubility in any solvent. Due to the strong interaction among the carbon nanotubes (CNT), it is not easy to obtain a fine and stable dispersion of CNTs. All these observations are explained by the annealing of the MWNT-electrode contact, which takes place at V_{TH}. The conductance increase in G-V characteristics appears only at a positive or negative bias, depending on the type of specimen used. This asymmetry is attributed to the ballistic or quasi-ballistic electron transport within MWNTs. Recently, conductance as high as (460-490) G_o or Poncharal *et al* results of about 10^3G_o have been reported. The aim of this work was to improve the intensity of light emitted by the mixture of Quantum dots (QDs) and Multi-walled carbon nanotubes. However, due to the severe aggregation of the MWNT, the results suffered and therefore the analysis was directed towards the transport of MWNT instead. Further work will include adding dispersants to the above mixture in order to achieve the desired aim of the enhancement of the light emission. On the other hand, dispersion of CNTs can be achieved by simply using various surfactants which adsorb on the surface of CNTs and can effectively minimize the strong attraction among CNTs by electrostatic repulsion or steric hindrance.

Since the discovery of carbon nanotubes CNTs (1,2)they have been the subject of intensive research.Carbon nanotubes(CNTs) which is a new 1D carbon material ,the CNTs are related to three other carbon forms,the two-dimensional graphite,the three-dimensional diamond,and the spherical,soccer ball like fullerenes.Depending on their production,single-walled carbon nanotubes(SWNTs) and multiwalled carbon nanotubes(MWCNTs)are distinguished.Among the realistic applications are field emission devices(3),transparent conductive films(4)and biosensors(5).However,owing to their macromolecular structure and their affinity to agglomeration,carbon nanotubes are insoluble or poorly dispersible in organic solvent.This is a serious problem that obstructs their incorporation in advanced composite materials and this drawback can be alleviated through chemical modification or functionallization of carbon nanotubes surfaces(6-9).Another ideal candidate material for the next generation of electronics is the quantum dot.Recent advancements in nanotechnology(9-13) have provided the possibility of controlling materials synthesis at the molecular level.Since both morphology and chemical composition can now be manipulated,leading to radically new material properties due to a combination of quantum confinement and surface-to-volume ratio effects .One of the main consequences of reducing the size of semiconductors down to nanometer dimensions is to increase the energy band gap leading to visible luminescence.As compared to quantum dots,carbon nanotubes have been found to be one-dimensional conduction materials with high charge mobilities(14).There fore,combining the optical properties of quantum dots with the long range charge transport characteristics of carbon nanotubes may generate advanced optoelectronic functionalities(15).In separate work on swnt composite(16) the performance of type(II) CdTe light emitting diodes demonstrated an improvement by doping with carboxylic functionalised single walled carbon nanotubes.On the contrary due to aggregation compositing MWNT with type(II) CdTe quantum dot produced moderate levels of emitted light Fig(1) ,this is due to the dominant behaviour of the MWNT.Carbon nanotubes(CNTs)can be metallic or semiconducting depending on their atomic structure(17),with a band gap that decreases as the diameter increases.CNTs have a strong covalent structure which means that they have a smaller space for scattering therefore more chance to exhibit ballistic conduction.This proposal was first confirmed by frank et al(18),ponchared et al (19) expanded this experiment and showed that the conductance of the CNTs increased linearly with voltage.Transport experiments(20,21) suggest that carbon nanotubes are remarkably good conductors with long electron mean free paths(22,23).Berger et al produced mean free path=200μ for MWNT.

Fig(2) shows the current-voltage characteristics of a composite of CdTe type(II) and MWNT different percentage.It can be seen that when the voltage is within a certain ± range the I-V plot is linear,which indicates that the MWCNT is metallic.The slope of each I-V,each G-V curve and the voltage range as well as the low-bias conductance vary for different samples.If the applied voltage is outside the linear range,optical phonons scattering becomes dominant which results in the current saturation(25).The resulting energy consumption will eventually leads to the destruction of the ballistic transport as the current is observed at higher voltages in the samples.

The I-V characteristics for these nanotube mixtures are nearly symmetric with respect to the voltage polarity.This behaviour may be further analyzed by considering the conductance characteristics. FIG s(G-V) shows a typical (G-V) curve which is linear and nearly symmetric with respect to the bias polarity at low biases.These are in good agreement with previously reported results (26,27,28,29).The slope of each G-V curve as well as the low-voltage conductance vary for different samples. Current saturation sometimes was observed in our G-V measurements in agreement with kerr et al and Collins et al (31),and at higher biases,the G-V characteristics start to deviate from their linear behaviour at low biases and the current starts to decrease.fig(2) shows an example of G-V characteristics for biases from 0.2 to 10 V.It can be seen that conductance deviates upward from its linear bias dependce.The bias voltage at which the G-V curve becomes nonlinear is ~ 3.0 V in fig(3);however,it is sample dependent and ranges from (-10) to10( ).On a further examination of the G-V curve we noticed a rapid increase with the bias.This rapid increase in conductance appears only Fig(4) during the forward bias sweep but it can appear in (+ve) or (-ve) side of the curve.The rapid increases in G-V characteristics,which take place during bias increase only,correspond to an irreversible modification of the contact geometry induced by the local contact heating.This was reported by two research groups (29,32).Therefore,the results shown in this work demonstrate that .the contact heating effects are not limited in liquid-metal electrodes but are present in our MWNT-Al/Ca contacts as well.Another interesting feature is the occurance of an unusal dependence on the bias polarity of the conductance increments Fig(5).Occasionally the conductance jumps are observed only at either positive or negative bias.(with a slight asymmetry some times observed which causes the conductance to be slightly lower for the negatively biased direction compared to the positively biased one).It is known that when the length of a conductor is smaller than the electron mean free path the electron transport is ballistic.Li et al (33) provided a clear evidence that MWNT is a ballistic conductor and suggested that the mean free path ℓ can reach the length of the MWNT,10-30μm in comparison to previous experiments which use several micrometers (18,34).Using the linear dependent relation between ℓ and r (r is the radius of the SWCNT)derived by Jiang et al(35) gives a universal expression of ℓ for all metallic nanotubes

$$\ell = 2 (\sqrt{3}) \pi V_{\circ}^{2} r \ / (2\sigma_{\varepsilon}^{2} + 9\sigma^{2}) \quad \ldots\ldots\ldots\ldots\ldots(1)$$

Where σ_ε andσ_v are variances of the on-site energy ε and the nearest-neighbor tight-binding parameter V_0 .r is the radius of the SWNT.From the equation,ℓ is proportional to r.This linear dependent relationship between ℓ and r holds for each metallic component shell of a MWCNT. From eq(1) the mean free path of our MWCNT should also be one order greater than several micrometers.Adding to this theorietical analysis the work of Ando and Nakanishi that the absence of backscattering is responsible for the long mean free path in carbon nanotubes(36).Berger et al(34) obtained a mean free path greater than 30 μm at room temperature.It is within this range (10-30 μm our MWCNT) that we can assume that the mean free path be reached by our MWCNT .As it was mentioned earlier that most of the time irreversible conductance jumps(9) appear on both aides of the G-V curve but occasionally,the conductance jumps are observed only at positive or negative biases.Since this behaviour is not expected from conventional contact heating effects.Our result is similar to Berger et al(29).,and M.Tsutsui et al (35) which can be observed only when the electrode used is an anode .Berger et al (29) explained their finding by assuming the ballistic electron transport through MWCNTs.When MWCNTs are ballistic conductors,,the contact heating is much effective at an anode contact where hot electrons are injected and dissipate their energies.The same explaination can be applied to our asymmetric G-V characteristics.A theoretical study (38) showed that the voltage drop take place mainly at nanotube-contact interfaces.In our case,MWCNTs have two contacts with the electrodes,so either one of which can become an anode depending on the voltage polarity.However,when one of the two MWCNT-electrode contacts happens to remain “frozen” or well-annealed,due to some unknown reasons,the contact annealing,or the irreversible conductance increment is observed only at such a bias polarity that makes the other contact an anode.Conversely,the observed polarity-asymmetry in the G-V curves can provide us supportive evidence for the ballistic electron transport in MWCNTs.

In summary,we studied the high-bias conductance versus voltage G-V characteristics of a multi-walled carbon nanotube(MWCNT) mixture with type (II)CdTe quantum dots and observed a sudden conductance increase when the bias exceeds a certain threshold voltage V_{tH}.Both MWCNT and single walled carbon nanotubes (SWNT) are severely entangled and possess extremely low solubility in any solvent.Due to the strong interaction among the carbon nanotubes (CNT),it is not easy to obtain a fine and stable dispersion of CNTs.All these observations are explained by the annealing of the MWCNT-electrode contact,which takes place at V_{tH} .The conductance increase in G-V characteristics appears only at a positive or negative bias,depending on the type of specimen used.This asymmetry is attributed to the ballistic or quasi-ballistic electron transport within MWCNTs.Recently,conductance as high as (460-490)G_0 have been reported.The aim of this work was to improve the intensity of light emitted by the mixture of quantum dots(QDs) and Multi-walled carbon nanotubes.However,due to the severe aggregation of the MWCNTs ,the results suffered and therefore the analysis was directed towards the transport of MWCNT instead.Further work will include adding adding dispersants to the above mixture in order to achieve the desired aim of the enhancement of the light emission.On the other hand,dispersion of CNTs can be achieved by simply using various surfactants which adsorb on the surface of CNTs and can effectively minize the strong attraction among CNTs by electrostatic repulsion or steric hindrance.

$G=I/V$ $Go=2(e^2)/h=(13k\Omega)^{-1}$ Relative Conductance(G/Go)=(G/.0000771938)

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Captions

Figure- 1 Absorbance spectrum of Type-II CdTe/CdS/ZnS core/shell/shell Quantumdots as a drop cast film.

Figure 2 - Photoluminescence,, spectra of CdTe /CdS/ZnS as a dropcast film (solid line) and in solution (dashed line).

Figure-3Current-voltage-Luminance Plot of ITO/ CdTe/CdS/ZnS core/shell/shell quantum dots/Al.

Figure-4Current-voltage-Luminance Plot of ITO/ CdTe/CdS/ZnS core/shell/shell quantum dots/Ca-Al .

Figure-5 Current-voltage-light plot of ITO/ CdTe/CdS/ZnS core/shell/shell quantum dots Spin coated /Ca-Al.

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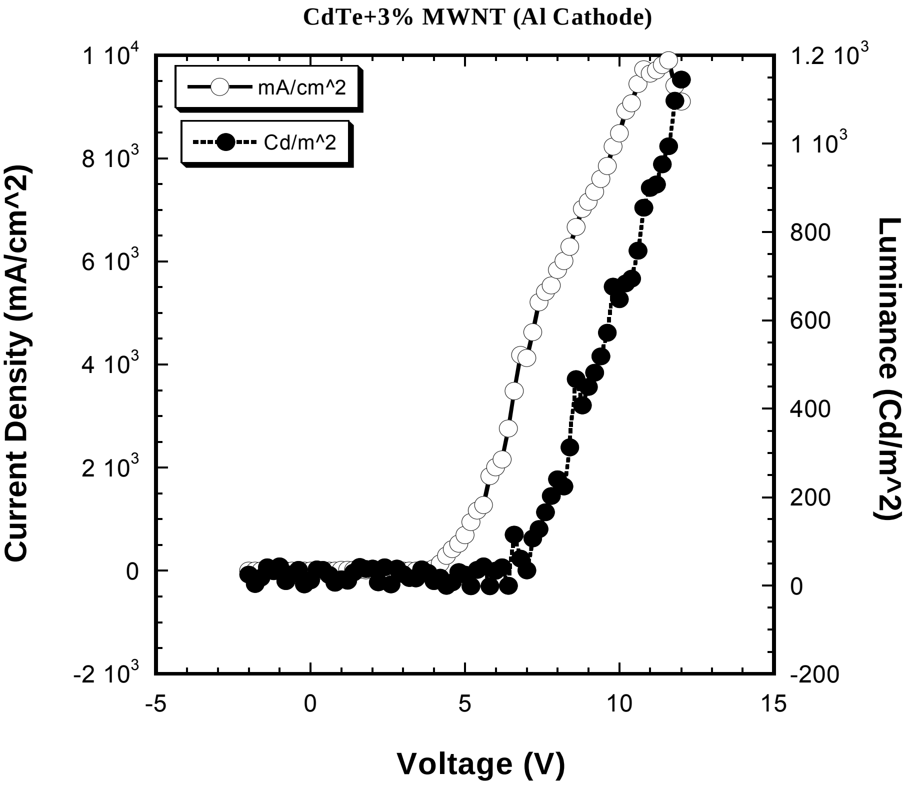
Figure-6 A plot of Current-voltage of ITO/ CdTe/CdS/ZnS core/shell/shell/Al **.**

Figure-6 inset Semi-logarithmic plot of Current-voltage of ITO/ CdTeCdS/ZnS core/shell/shell/Al.

Figures

Figure 1



Fi gure 2

Figure2

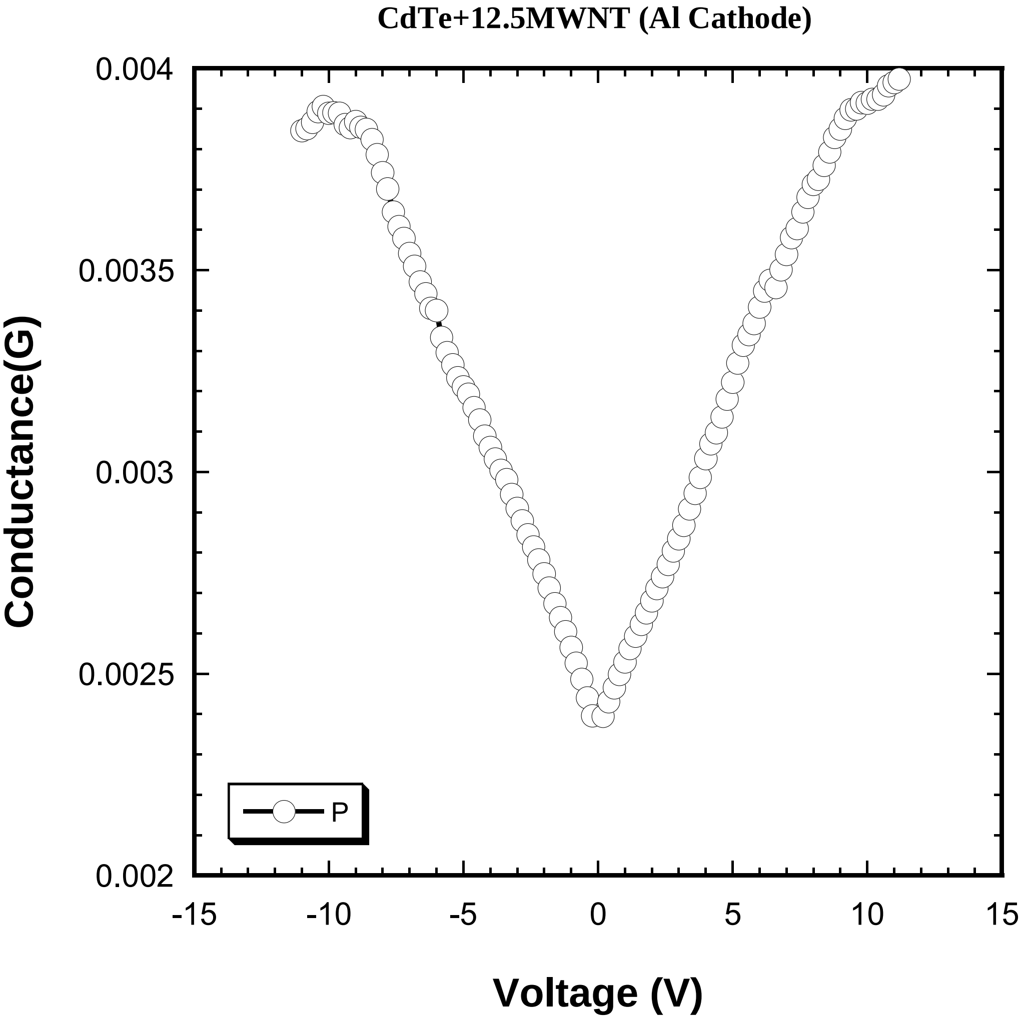
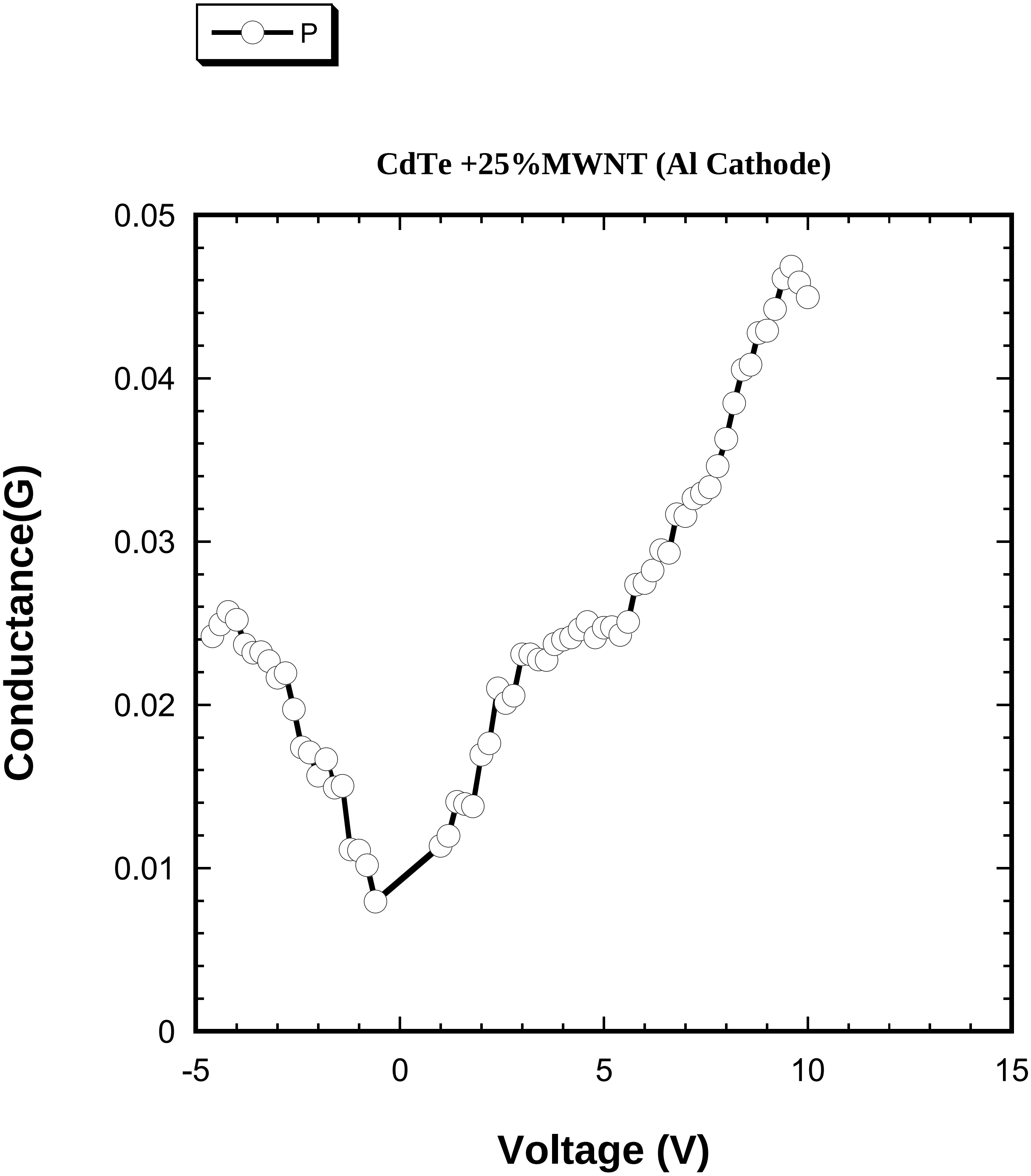

CdTe+12.5MWNT (Al Cathode)
Conductance(G)
0.004
0.0035
0.003
0.0025
0.002
-15
-10
-5
0
5
10
15
Voltage (V)
P

Figure3 and Figure 4



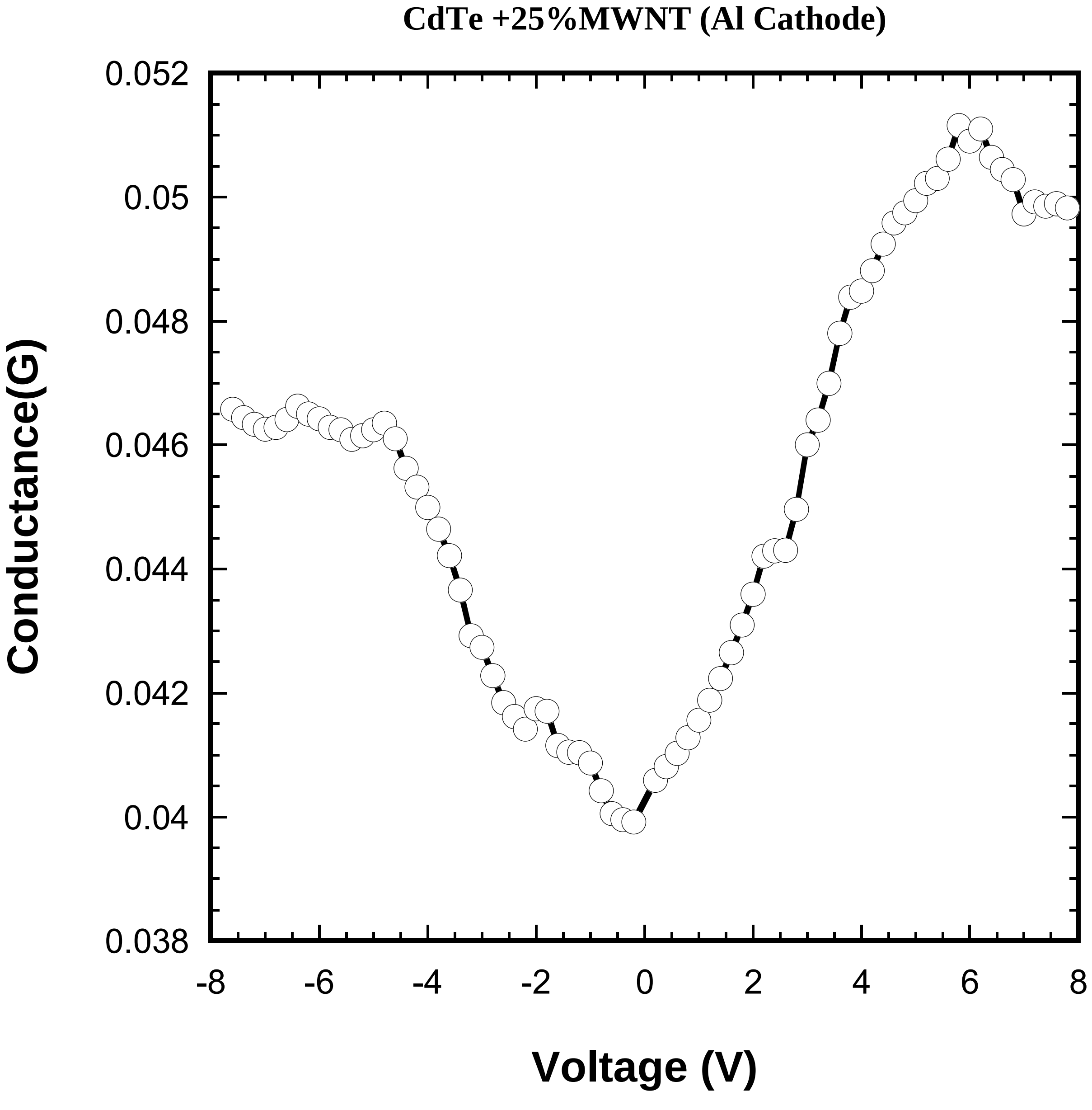

CdTe +25%MWNT (Al Cathode)
Conductance(G)
0.052
0.05
0.048
0.046
0.044
0.042
0.04
0.038
-8
-6
-4
-2
0
2
4
6
8
Voltage (V)

Figure 5a1

CdTe +12.5%MWNT (Ca Cathode)

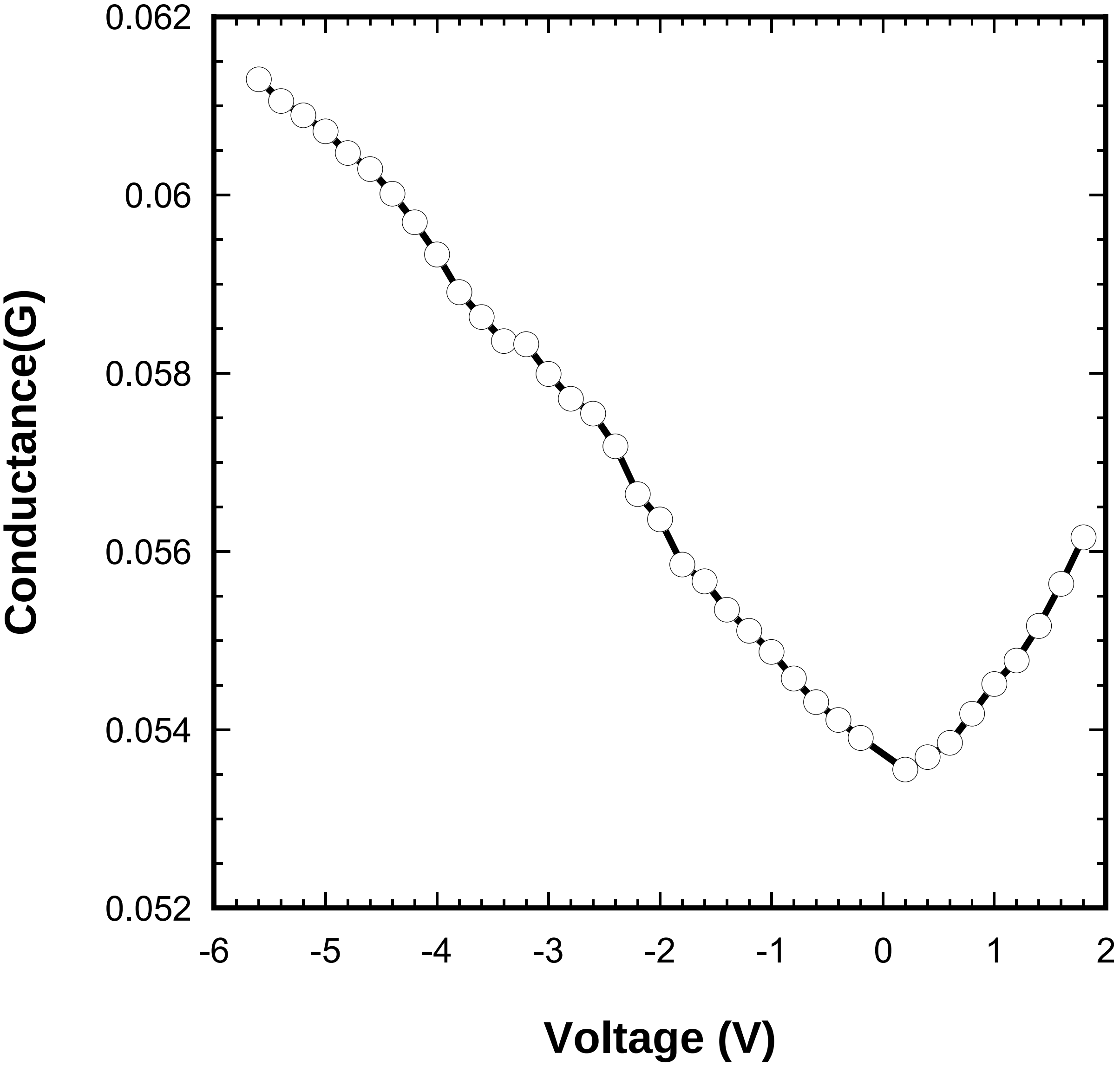


Figure 5a2

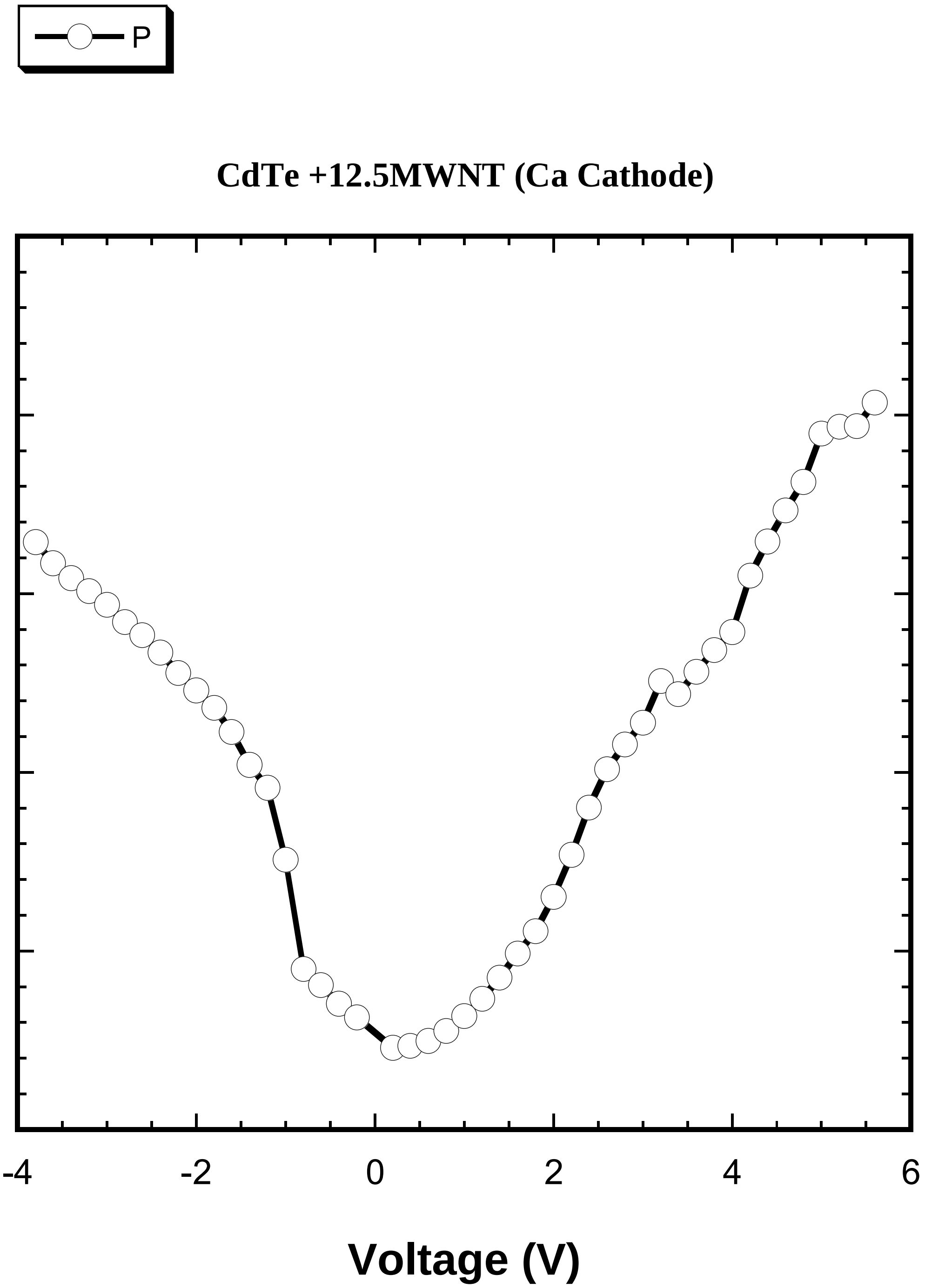

P
CdTe +12.5MWNT (Ca Cathode)
0.08
0.075
0.07
0.065
0.06
0.055
Conductance(G)
-4
-2
0
2
4
6
Voltage (V)

Figure 5b1

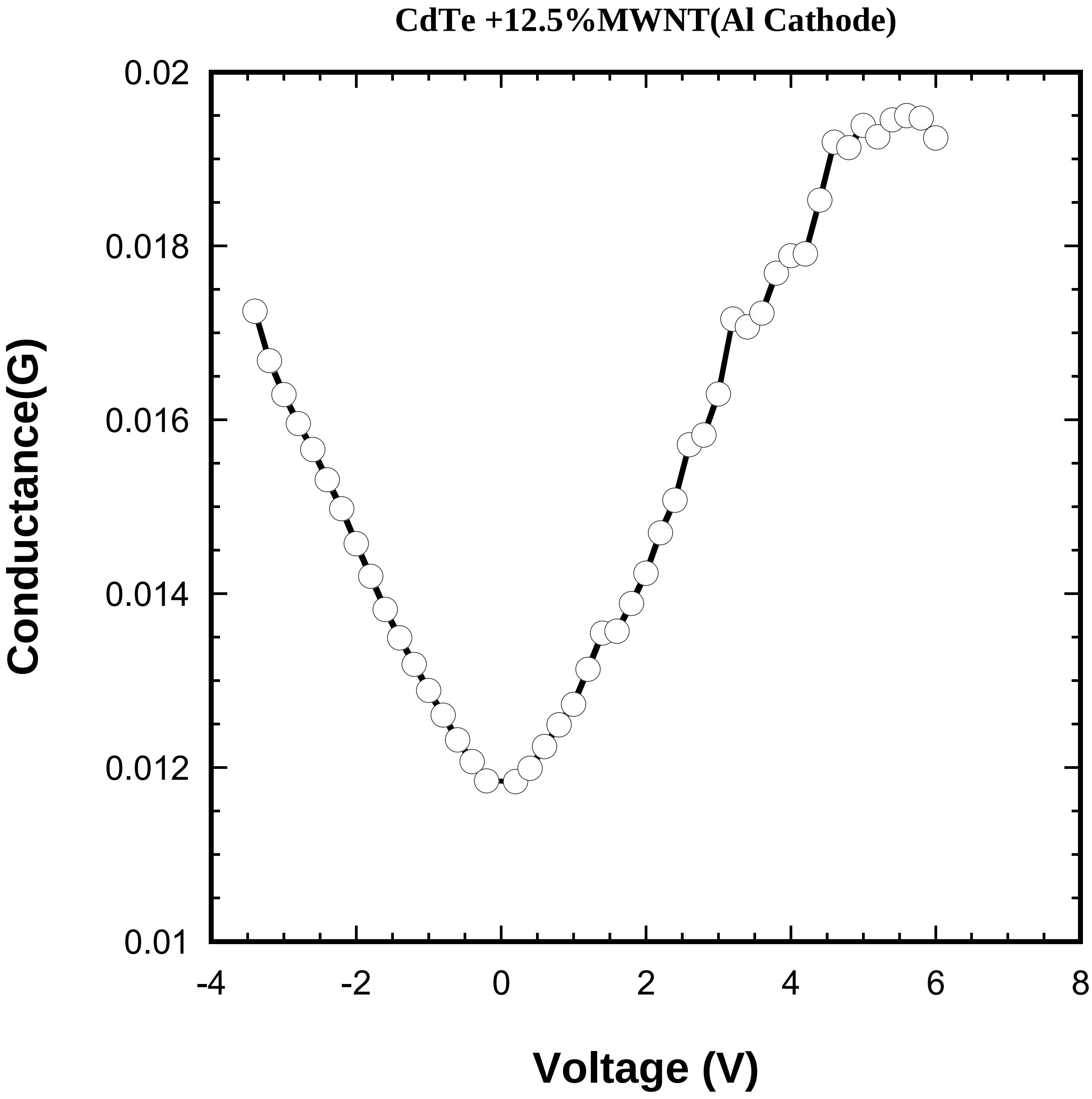


Figure 5b2

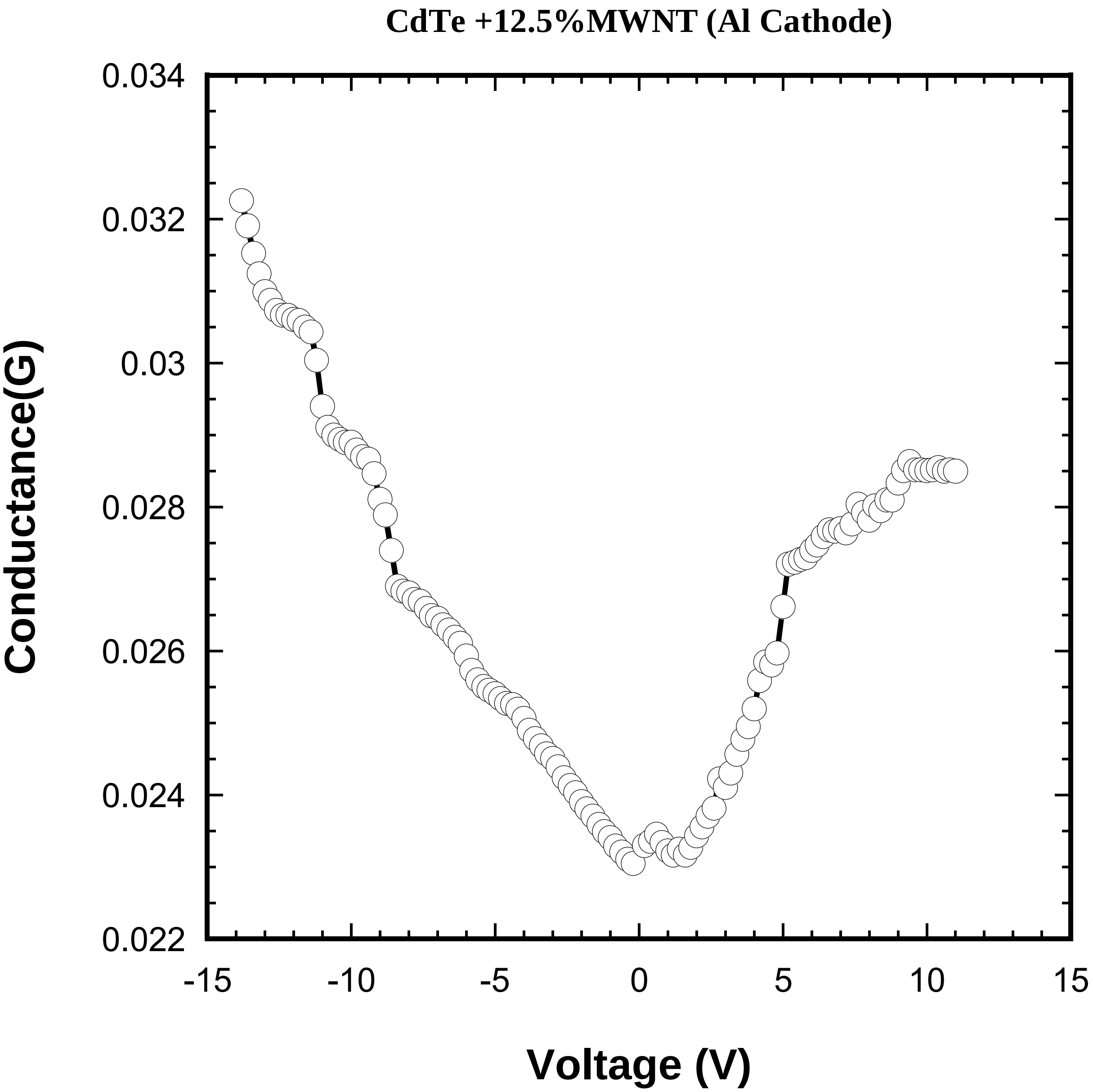

CdTe +12.5%MWNT (Al Cathode)
Conductance(G)
0.034
0.032
0.03
0.028
0.026
0.024
0.022
-15
-10
-5
0
5
10
15
Voltage (V)

Figure 5c1

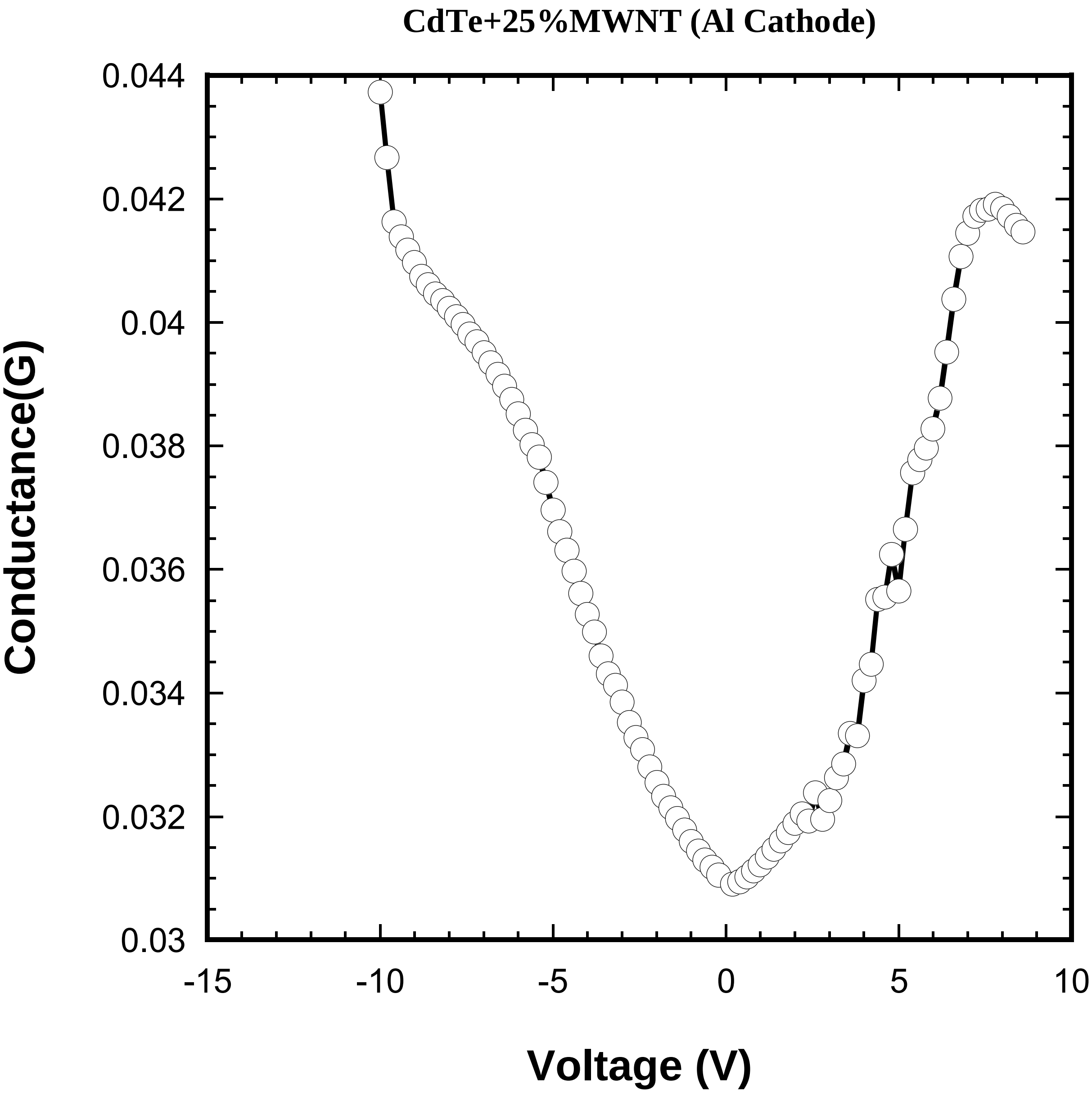

CdTe+25%MWNT (Al Cathode)
Conductance(G)
0.044
0.042
0.04
0.038
0.036
0.034
0.032
0.03
-15
-10
-5
0
5
10
Voltage (V)

Figure 5c2

CdTe +25%MWNT (Al Cathode)