# Thermally activated field assisted carrier generation and transport in $N,N'$-di-[(1-naphthalenyl)-$N,N'$-diphenyl]-(1,1′ biphenyl)-4,4′-diamine doped with 2,3,5,6-tetrafluoro-7,7′,8,8′-tetracyanoquinodimethane

Gayatri Chauhan,[1] Ritu Srivastava,[1,a)] Virendra Kumar Rai,[1] Arunandan Kumar,[1] S. S. Bawa,[1] P. C. Srivastava,[2] and M. N. Kamalasanan[1]
[1]*Center for Organic Electronics, National Physical Laboratory (Council of Scientific and Industrial Research), Dr. K. S. Krishnan Road, New Delhi 110012, India*
[2]*Department of Physics, Banaras Hindu University, Varanasi-221005, India*

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Current density-voltage ($J$-$V$) characteristics of $N,N'$-di-[(1-naphthalenyl)-$N,N'$-diphenyl]-(1.1′ biphenyl)-4,4′-diamine ($\alpha$-NPD) doped with 2,3,5,6-tetrafluoro-7,7′,8,8′-tetracyanoquinodimethane have been studied as a function of doping concentration (0–0.8 wt %) and temperature (105–300 K). The current density was found to increase with increase in doping concentration. In the doped samples as field increases above $3.3\times10^4$ V/cm the current abruptly starts increasing at a higher rate, which is ascribed as due to increased free charge carrier generation in the bulk. The enhanced free charge carrier generation is due to field assisted thermal dissociation of donor-acceptor pairs (Poole–Frenkel process) as well as charge injection at the interface. The released carriers increase the charge carrier density which brings the Fermi level near the highest occupied molecular orbital level of the $\alpha$-NPD and reduces the space charge region near the interface favoring the tunneling of charge carrier across the interface, which is enough to support Ohmic conduction. The carrier generation has been found to be a thermally activated process. At higher fields (i.e., above $1.52\times10^5$ V/cm) the nonlinear $J$-$V$ characteristics have been explained as due to field dependent mobility of holes. © *2008 American Institute of Physics*.
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## I. INTRODUCTION

Very efficient optoelectronic devices have been fabricated by doped inorganic semiconductors. However, there are very few attempts to dope organic semiconductor material for optoelectronic applications. It is known that the conductivity of organic materials (extended $\pi$-electron systems) can be influenced by doping with donor or acceptor molecules. There are some reports on the doping of organic semiconductor using various dopants.[1–3] Doping of an organic semiconductor by charged moieties increases the concentration of charge carriers (lifts up the Fermi level) and increases energetic disorder.[4,5] Although the increase in charge carriers facilitates conductivity, yet the disorder suppresses the carrier hopping rate and decreases carrier mobility. The understanding of the fundamental aspects of doping and its effect on charge transport would enable one to develop devices with better performance.

There are some earlier attempts to understand the fundamental effects of doping in organic semiconductors. Yang and Heeger[6] introduced doped polyaniline as hole transport layer for polymer light-emitting diodes (LEDs) and Ganzorig and Fujihara[7] doped a spin-coatable form of N,N'Bis(3methylphenyl)-N,N'-diphenylbenzidine (TPD) with $SbCl_5$ as a hole transport layer. Another widely used hole injection and transport layer in bilayer polymer LEDs is poly(3,4-ethylenedioxythiophene) doped with poly(4-styrenesulfonic acid).[8] The studies on 2,3,5,6-tetrafluoro-7,7′,8,8′-tetracyanoquinodimethane ($F_4$-TCNQ) doped ZnPc were extensively carried out using ultraviolet photoelectron spectroscopy (UPS). It is found that the doping results in electron transfer from highest occupied molecular orbital (HOMO) of ZnPc to the lowest unoccupied molecular orbital (LUMO) of $F_4$-TCNQ. This electron transfer was energetically favorable ($\mathrm{ZnPc_{HOMO}}=5.28$ eV and $\mathrm{F_4\text{-}TCNQ_{LUMO}}=5.24$ eV) and was facilitated by narrowing of space charge region near the Au electrode which assisted in the injection of the charge carriers by which the current increased by six to seven orders of magnitude.[9] Later Gao and Kahn[10] studied the doping properties of another hole transport material $N,N'$-di-[(1-naphthalenyl)-$N,N'$-diphenyl]-(1.1′ biphenyl)-4,4′-diamine ($\alpha\text{-NPD}_{\mathrm{HOMO}}=5.52$ eV) with $F_4$-TCNQ. Even though electron transfer from $\alpha$-NPD to $F_4$-TCNQ is slightly unfavorable, the authors have assumed that the $F_4$-TCNQ dispersed in $\alpha$-NPD will have the slightly different polarization energy which will compensate the difference in energy levels. They have studied the Au/$\alpha$-NPD and Au/$\alpha$-NPD doped with $F_4$-TCNQ interface using direct and inverse photo electron spectroscopy, contact potential difference measurement, and current voltage ($I$-$V$) measurement. They have found enhanced current in $I$-$V$ measurement and this enhancement in the current has been attributed to reduction in potential barrier due to Richardson–Schottky (RS) thermionic emission from the electrode to the $\alpha$-NPD. Also they found the evidence of charge transfer from $\alpha$-NPD to $F_4$-TCNQ. Wan *et al.*[11] stud-

[a)]Electronic mail: ritu@mail.nplindia.ernet.in.

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ied the effect of surface contamination of Au electrode on $\alpha$-NPD and showed that even a small ambient exposure of Au electrode prior to $\alpha$-NPD deposition results in a considerable modification of the interface property due to the exchange of charge from $\alpha$-NPD to the induced density of interface states (IDISs).

In the present paper we have studied the $\alpha$-NPD doped with $F_4$-TCNQ hole only devices at various dopant concentrations and at various temperatures. The variation in the current with temperature has shown some interesting results revealing that the doping has switched the conduction process from injection limited to bulk limited. The free carrier generation is a thermally activated process and the carrier mobility is a field dependent parameter.

## II. EXPERIMENTS

Indium-tin oxide (ITO) (thickness of ~120 nm) coated glass substrate with a sheet resistance 20 Ω/□ (Vin Karola, USA) was patterned and cleaned sequentially using deionized water, acetone, trichloroethylene, and isopropyl alcohol for 20 min each using an ultrasonic bath. Prior to film deposition, the surface of ITO substrate was treated with oxygen plasma for 5 min to enhance its work function. Pure and doped $\alpha$-NPD films were deposited under a high vacuum ($10^{-5}$–$10^{-6}$ torr). The doped films were deposited by coevaporation process. The thicknesses of these films were measured *in situ* by a quartz crystal thickness monitor. The typical thicknesses of the different layers of pure and $F_4$-TCNQ (Acros Organics, Belgium) doped $\alpha$-NPD (Sigma Aldrich) were ~300 nm. The doping concentration was restricted up to 0.8% (by weight) since beyond this level, films show pealing off tendency from the substrate. There after gold (Au) electrode (~600 nm) was vacuum deposited on the top through a shadow mask resulting into a hole only device, as both the ITO and Au have nearly a similar work function (ITO=4.8 eV and Au=5.1 eV, respectively). The device configurations were ITO/$\alpha$-NPD/Au and ITO/$F_4$-TCNQ doped $\alpha$-NPD/Au having a pixel size of $5\times5$ mm$^2$. *J-V* measurements were performed in vacuum from low temperature to room temperature using a Keithley 2400 programable digital source meter. Although ITO and Au have nearly a similar work function, the injection properties for holes from ITO Fermi level to the HOMO of $\alpha$-NPD (i.e., ITO is positively biased and Au is negatively biased) and that of Au Fermi level into HOMO of $\alpha$-NPD (i.e., Au is positively biased and ITO is negatively biased) are different (figure is not shown here). The former shows the temperature and field dependent transport properties, whereas the latter shows high injection barrier and low current. Here we studied the hole only devices where ITO is positively biased and Au is negatively biased.

## III. RESULT AND DISCUSSION

Figure 1(a) shows the *J-V* characteristics of pure $\alpha$-NPD (~300 nm) in the configuration ITO/$\alpha$-NPD (300 nm)/Au at temperatures of 300, 260, 235, 195, 165, and 120 K. It is seen from Fig. 1(a) that the *J-V* characteristics of pure $\alpha$-NPD show nonlinear rise in current with increase in voltage and a nearly temperature independent *J-V* characteristic typical of tunneling[12,13] of carriers from the Fermi level of ITO to HOMO of $\alpha$-NPD. The estimated Fermi levels of ITO and $\alpha$-NPD from UPS studies are 4.8 and 5.52 eV, respectively. However, UPS measurements by Hill and Kahn[14] showed that the formation of surface dipoles at ITO/$\alpha$-NPD interface increases the injection barrier to about 1.2 eV. The variation in $J$ with temperature is too low to account for the mechanism based on RS-type charge injection. Further the value of $J$ is too low to account for space charge limited conduction considering the high charge carrier mobility ($6.1\times10^{-4}$ cm$^2$/V s) reported for holes in $\alpha$-NPD. A Fowler–Nordheim (FN) tunneling mechanism is usually used to explain the injection process in case of such large injection barriers. In the case of FN tunneling the current density ($J$) depends on the electric field ($F$) by the following equation:

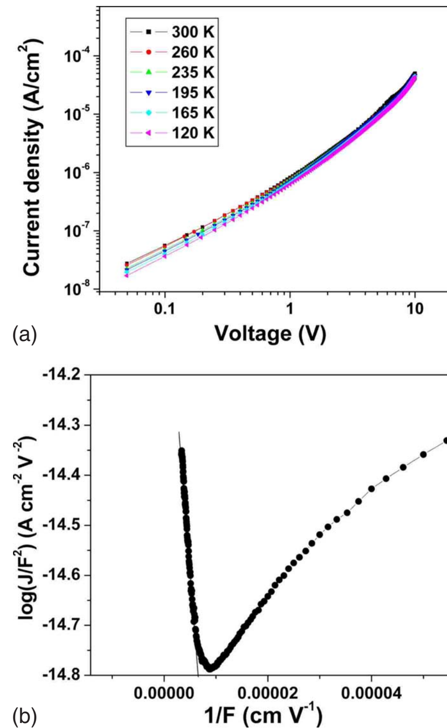


FIG. 1. (Color online) (a) log($j$)-log($v$) characteristics of pure $\alpha$-NPD at different temperatures (300, 260, 235, 195, and 150 K). (b) log($J/F^2$) vs $1/F$ plot of pure $\alpha$-NPD at 300 K.

$$J = AF^2 \exp\left(\frac{-k}{F}\right). \tag{1}$$

Equation (1) can be rewritten as

$$\ln\left(\frac{J}{F^2}\right) = \ln A - k\left(\frac{1}{F}\right), \tag{2}$$

where

$$A = \frac{q^3}{8\pi h \phi_b},$$

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$$k = 8\pi \frac{\sqrt{2m^*}\phi_b^{3/2}}{3qh}. \quad (3)$$

Here $\phi_b$ is the barrier height, $m^*$ is the effective mass of holes in $\alpha$-NPD, $k$ is a barrier shape dependent parameter, $q$ is the electronic charge, and $h$ is the Planck's constant.

It can be seen from Eq. (2) that for the current to be FN limited $\log(J/F^2)$ versus $1/F$ should show a linear behavior especially at higher electric fields. The value of $\phi_b$ can be calculated from the slope of the linear portion of $\log(J/F^2)$ versus $1/F$ plot existing at the interface between ITO Fermi level and HOMO of $\alpha$-NPD during charge injection. Figure 1(b) shows the plot of $\log(J/F^2)$ versus $1/F$ showing a straight line from $F=3.08\times10^5$ V/cm to $F=1.52\times10^5$ V/cm and the value of $\phi_b$ evaluated from the slope of linear part has been found to be ~0.87 eV.

When a $p$-type organic semiconductor ($\alpha$-NPD) is doped with a strong acceptor such as $F_4$-TCNQ, electrons will be transferred from the HOMO of the host to the LUMO of the dopant and all the dopants will be ionized if the energy of the HOMO is greater than the LUMO energy of the dopant. Even if this condition is not fulfilled, still all the dopants can be ionized since the energy deficit "Δ" for charge transfer from HOMO of the host to LUMO of the dopant can be compensated by the Coulomb interaction between ionized dopants and released charge carriers localized in nearby hopping sites. This implies that carrier can be released from dopants even if their LUMOs are well above the HOMO of the host material.[5] The doping process of $\alpha$-NPD (HOMO =5.52 eV) with $F_4$-TCNQ (LUMO=5.24 eV) is an endothermic process (i.e., LUMO of the dopant is slightly above the HOMO of the host $\alpha$-NPD) with a deficit in energy of 0.28 eV.[10] The charge transfer is facilitated by the Coulomb interaction energy between the donor and acceptor thereby binding the pair with a binding energy of 0.28 eV. At low temperatures and low electric fields, very few the donor and acceptors are ionized and the charge carriers are not available for the transport process. However, at higher electric fields and higher temperatures, these pairs are ionized by a Poole–Frenkel (PF) effect in which the binding energy of bound carriers is reduced in the presence of an applied electric field. The dissociation will increase the hole concentration in $\alpha$-NPD and will bring the Fermi level near the HOMO level of the $\alpha$-NPD. This process is likely to bend the HOMO levels near the metal-semiconductor interface and reduce the width of the space charge region near the interface favoring the increased tunneling of charge carriers into the semiconductors and making the contact Ohmic for hole injection. Similar results have been reported for $F_4$-TCNQ doped ZnPc by Blochwitz *et al.*[15] The increased charge injection can also be due to trapping of charge carrier at the IDIS.

Figure 2(a) shows the $J$-$V$ characteristics at room temperature (300 K) of pure $\alpha$-NPD and $F_4$-TCNQ doped $\alpha$-NPD films (~300 nm). Curves a, b, and c correspond to $F_4$-TCNQ doping concentrations of 0.0, 0.4, and 0.8 wt %, respectively. At low voltages unlike undoped samples which show FN tunneling, the $J$-$V$ characteristics of doped samples show Ohmic current ($J\propto V$). Further, there is an increase in

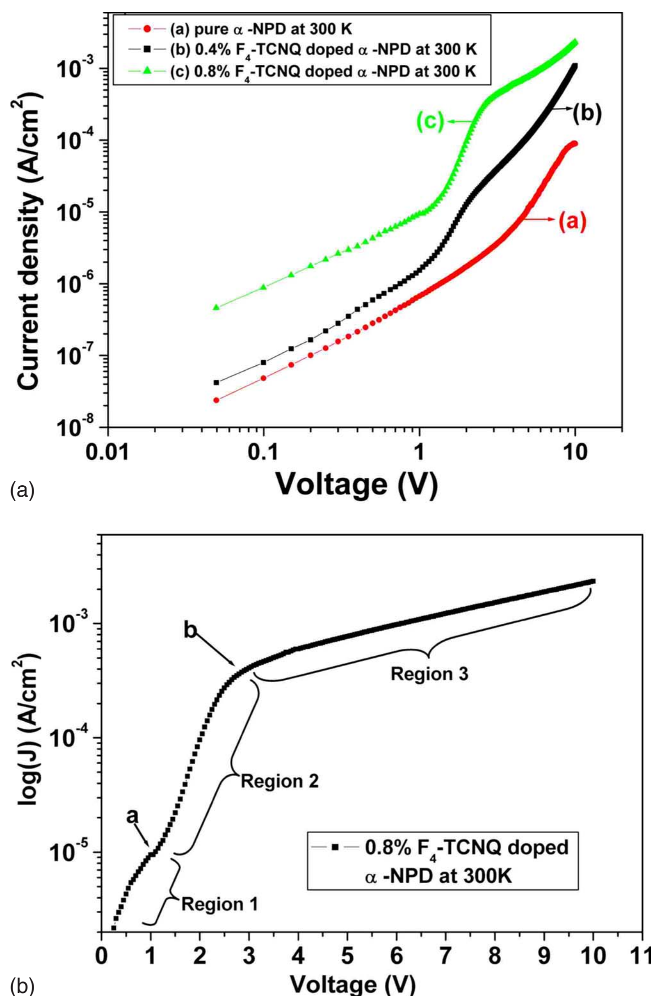


FIG. 2. (Color online) (a) log($j$)-log($v$) characteristics of (a) pure $\alpha$-NPD, (b) 0.4% $F_4$-TCNQ doped $\alpha$-NPD, and (c) 0.8% $F_4$-TCNQ doped $\alpha$-NPD at 300 K. (b) log($j$) vs $V$ plot of 0.8% $F_4$-TCNQ doped $\alpha$-NPD. Points a and b show the sharp change in the $J$-$V$ curve at ~1 and 3 V.

the current density upon increased doping concentration. The current density at low voltages is not thermally activated as there is very little change in current density upon varying the temperature (see the low voltage part in Fig. 3). This shows that some of the donor-acceptor pairs remain ionized even at low temperatures and low electric fields. One of the possible explanation is that the large electric field due to the image

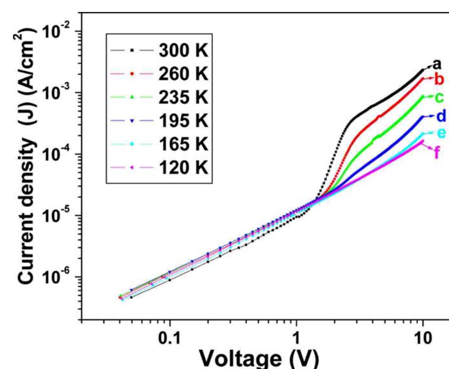


FIG. 3. (Color online) log($j$)-log($v$) characteristics of 0.8% $F_4$-TCNQ doped $\alpha$-NPD at different temperatures, i.e., 300, 260, 235, 195, 165, and 120 K (curves a, b, c, d, e, and f).

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force near the ITO/$\alpha$-NPD interface dissociates the electron hole pair and ionizes the donor completely and the resulting holes get trapped in the IDIS, thereby reducing the interface barrier enabling an Ohmic contact.

It is clearly seen from Fig. 2(a) that the current varies nonlinearly and the nonlinearity aspect becomes more explicit at higher doping levels (0.4 and 0.8 wt %). In fact a very interesting phenomenon is observed in curves "b" and "c" in Fig. 2(a), where the $J$-$V$ behavior at intermediate fields shows an enhanced nonlinearity than the corresponding undoped film [curve "a" in Fig. 2(a)] obtained in the same field range. This has been explicitly shown in Fig. 2(b), where the $J$-$V$ characteristics at room temperature of 0.8 wt % doped $\alpha$-NPD show a distinct behavior between 0 and 1 V (region 1), 1 and 3 V (region 2), and 3 and 10 V (region 3). To further understand the effect of doping, we have studied the $J$-$V$ characteristics of $\alpha$-NPD as a function of temperature at highest doping concentration of ~0.8%. Curves a, b, c, d, e, f, and g in Fig. 3 show such characteristics at temperatures of 300, 260, 235, 195, 165, and 120 K, respectively. It is seen from Fig. 3 that enhancement in nonlinearity is not only observed at room temperature but also at a relatively low temperature (~300, 260, 235, and 195 K) for higher doping (0.8 wt %) case. At low temperatures and low electric fields the sample shows nearly temperature independent and Ohmic $J$-$V$ characteristics, indicating that the charge transport is not thermally activated and the contacts are Ohmic to support bulk limited conduction, and there is a low concentration of charge carriers present in the sample. As the temperature and electric field increase, field induced dissociation of donor-acceptor pairs takes place effecting the release of charge carriers (PF effect). The sudden increase in current density between 1 and 3 V in Fig. 3 is due to this process. The equation for current density is

$$J = nq\mu F \tag{4}$$

and the current density due to barrier lowering in PF effect is

$$J = J_0 \exp\left[\frac{-(\phi_{\mathrm{PF}} - \beta_{\mathrm{PF}}\sqrt{F})}{kT}\right], \tag{5}$$

where $J_0$= low field current density and

$$\beta_{\mathrm{PF}} = \sqrt{\frac{q^3}{\pi\varepsilon\varepsilon_0}},$$

where $q$ is the unit electronic charge, $\varepsilon_0$ is the permittivity of the free space, $\varepsilon$ is the dielectric constant of the material, $F$ is the electric field, $k$ is the Boltzmann constant, $n_0$ is the carrier density when all the donors are ionized, $\beta_{\mathrm{PF}}$ is the PF field lowering constant, and $\phi_{\mathrm{PF}}$ is the binding energy.

The theoretical value of $\beta_{\mathrm{PF}}$ calculated (using $\varepsilon=3$) is $4.4\times10^{-5}$ eV m$^{1/2}$ V$^{-1/2}$. The experimental values obtained from the slope of the graph log($J$) versus $\sqrt{F}$ [Fig. 4(a)] are $4.2\times10^{-5}$ eV m$^{1/2}$ V$^{-1/2}$. This value is very close to the theoretically calculated value. This suggests that the dominant conduction mechanism is the PF type. The field assisted thermally released carriers may bring the Fermi level very near to the HOMO of $\alpha$-NPD, which is likely to bend the HOMO level near the interface and reduce the thickness of

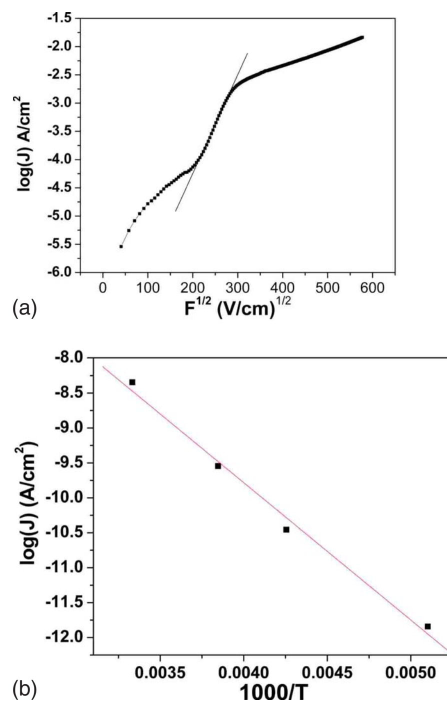


FIG. 4. (Color online) (a) log($j$) vs $\sqrt{F}$ plot of 0.8% $F_4$-TCNQ doped $\alpha$-NPD indicating the linear dependence in the range of 1–2.5 V. (b) log($j$) vs 1000/$T$ plot of 0.8% $F_4$-TCNQ doped $\alpha$-NPD at $V$=2.45 V.

the space charge layer assisting the tunneling of charge carriers across the interface from the bulk. Further, the charge carriers may get trapped at the IDIS and reduce the interface barrier and enhance the charge injection which further supports the Ohmic conduction.

The linear dependence of log($J$) with square root of electric field shown in Fig. 4(a) is an evidence of this effect. When the current density is saturated, i.e., $n=n_0$, the value of $\phi_{\mathrm{PF}}$ becomes equal to $\beta\sqrt{F}$. From this the value of $\phi_{\mathrm{PF}}$ calculated as ~0.12 eV [shown in Fig. 4(b)] is very near to the energy deficit reported for the charge transfer from the donor to the acceptor (0.28 eV). The difference between the energy estimated from the present study and that obtained from the difference in energy value estimated from UPS studies may be due to polarization effect as suggested by Gao and Kahn.[10]

At about 3 V the dissociation of donor-acceptor pairs becomes complete and the charge carriers saturate. The saturated carrier density should be a constant equal to the total number of ionized dopants assuming that all dopants are ionized. This value comes out to be ~$2\times10^{19}$. Since the saturated current density ($J_s$) for different temperatures is different, we presume that the charge carrier immediately after dissociation gets retrapped in shallow traps with a trap energy $E_T$.

In such a case the carrier density $n$ will have Arrhenius temperature dependence

$$n = n_t \exp(-E_T/kT). \tag{6}$$

It has been seen in Fig. 4(b) that the plot of log($J$) versus 1000/$T$ is indeed a straight line. The value of $E_T$ calculated from Fig. 4(b) is 0.29 eV.

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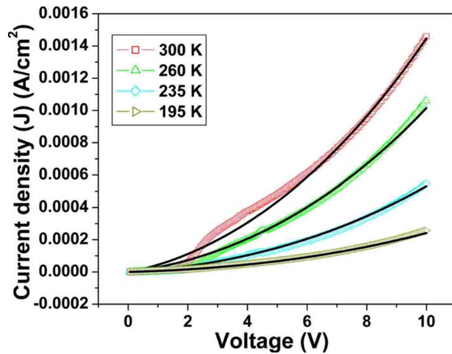


FIG. 5. (Color online) Theoretical fitted *J-V* characteristics of 0.8% $F_4$-TCNQ doped $\alpha$-NPD at temperatures 300, 260, 235, and 195 K calculated from Eq. (8). The open symbols represent the experimental values and the solid lines represent the theoretical fit.

The current density ($J$) is found to increase slowly with further increase in voltage. This has been explained by invoking the field dependent charge carrier mobility. The field dependent mobility is given by the equation

$$\mu = \mu_0 \exp \beta\sqrt{F}. \tag{7}$$

Using Eqs. (4) and (7), we can write

$$J = nqF\mu_0 \exp \beta\sqrt{F}, \tag{8}$$

where $n$ is the free charge carrier density, $q$ is the electronic charge, $F$ is the applied field, $\mu_0$ is the zero field mobility, and $\beta$ is the field lowering constant. Figure 5 shows the *J-V* characteristics of the 0.8 wt % doped $\alpha$-NPD in the temperature range of 300–195 K, which has been compared with the $J$ values calculated using Eq. (8). In Fig. 5 the open symbol represents the experimental curves, while the solid black line corresponds to the theoretically generated curves at corresponding temperature using Eq. (8). As shown in Fig. 5 the experimental curves in the temperature range of 300–195 K agree very well with the field enhanced mobility model with the fitting parameter $n = 2 \times 10^{19}$. The values of other parameters at different temperatures have been given in Table I. From Fig. 5 we can say that in the higher field region, i.e., above $1.52 \times 10^5$ V/cm, the conduction is dominated by field enhanced mobility, which is also applicable at low temperatures. The low value of charge carrier mobility obtained in this case compared to the reported value of $6.1 \times 10^{-4}$ cm$^2$/V s for pure $\alpha$-NPD (Ref. 16) is due to the retrapping of the charge carriers by shallow traps.

TABLE I. Values of $\mu_0$ and $\beta$ at different temperatures for 0.8% $F_4$-TCNQ doped $\alpha$-NPD.

| | Temperature | | | |
|---|---|---|---|---|
| Parameter | 300 K | 260 K | 235 K | 195 K |
| $\mu_0$ (cm$^2$/V s) | $2.15 \times 10^{-5}$ | $1.55 \times 10^{-5}$ | $4.5 \times 10^{-6}$ | $1.9 \times 10^{-6}$ |
| $\beta$ (cm/V)$^{1/2}$ | $3.2 \times 10^{-3}$ | $3.8 \times 10^{-3}$ | $4.2 \times 10^{-3}$ | $4.4 \times 10^{-3}$ |

## IV. CONCLUSION

We have measured the *J-V* characteristics of pure and $F_4$-TCNQ doped $\alpha$-NPD at different doping levels and temperatures. The study reveals that (1) charge injection from ITO electrode to pure $\alpha$-NPD is FN injection limited. (2) Doping of $\alpha$-NPD with $F_4$-TCNQ lowers the barrier making the contact Ohmic. (3) As the electric field increases field induced dissociation of donor-acceptor pairs takes place, increasing the carrier concentration inside the sample. (4) These released carriers get retrapped in shallow traps, making the carrier concentration thermally activated and reducing the charge carrier mobility compared to undoped $\alpha$-NPD. (5) Charge conduction at higher fields is controlled by field enhanced carrier mobility.

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