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# Current voltage relation of amorphous materials based *pn* diodes—the effect of degeneracy in organic polymers/molecules

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A general representation of the current in an amorphous semiconductor *pn* diode is developed. This expression is applied to examples of density of states functions (exponential, Gaussian, and Gaussian with exponential tail) commonly found in conjugated molecules and other amorphous materials. We find that the ideality factor could be voltage dependent and that its functional form is closely related to the shape of the density of states. © *2009 American Institute of Physics*. [doi:10.1063/1.3245283]

## I. INTRODUCTION AND BACKGROUND

Recently, it has become possible to realize high quality *pn* or *pin* diodes[1] from organic materials due to successful introduction of *n*- and *p*-type doping.[2,3] While there is significant progress in the quality of these devices, few reports exist regarding the expected effect of using organic materials rather than inorganic semiconductors. More specifically, it is now established that organic semiconductors belong to the family of amorphous semiconductors in which the most common density of states (DOS) functions are either exponential, Gaussian, or the combination of the two.[4] Here we derive expressions for the *I*-*V* characteristics of *pn* diodes comprised of organic semiconductors. The motivation to reexamine the *I*-*V* characteristics of such diodes is largely due to the degeneracy of the organic semiconductors in which the Einstein relation is charge density dependent and this dependence is a function of the DOS. To account for this one has to use the concept of generalized Einstein relation:[4,5]

$$\frac{D(n)}{\mu(n)} = \frac{n}{q\dfrac{\partial n}{\partial E_F}} \triangleq \frac{kT}{q}\eta(n). \tag{1}$$

Using the generalized Einstein relation to derive the *I*-*V* relations in a *pn* diode, a slightly different approach to the one found in most textbooks allows us to derive a more general solution. However, before embarking on the derivation of the diode characteristics we first remind ourselves the basic equations governing the diffusion current of excess minority carriers (as *n* in *p* side). The one dimensional continuity equation states:

$$\frac{\partial \hat{n}}{\partial t} + \frac{dF(x)}{dx} + \frac{\hat{n}}{\tau} = 0, \tag{2}$$

where according to Fick's law,

$$F(x) = -D\frac{d\hat{n}}{dx}, \tag{3}$$

where $\tau$ is the electron lifetime (assumed to be constant). In Eqs. (2) and (3) we relied on the notation that the carrier density ($n$) is composed of the equilibrium carrier density ($\bar{n}$) and of the excess carrier density ($\hat{n}$) such that $n=\bar{n}+\hat{n}$. After substituting $F(x)$ one arrives at

$$\frac{\partial \hat{n}}{\partial t} = \frac{d}{dx}\left(D\frac{d\hat{n}}{dx}\right) - \frac{\hat{n}}{\tau}. \tag{4}$$

Assuming steady state ($\partial\hat{n}/\partial t=0$) leads to

$$\frac{d}{dx}\left(D\frac{d\hat{n}}{dx}\right) = \frac{\hat{n}}{\tau}, \tag{5}$$

and solving Eq. (5) we arrive at a generic expression describing diffusion currents,

$$\left(D\frac{d\hat{n}}{dx}\right)^2 = 2\int_{\infty}^{x} \frac{D}{\tau} \times \hat{n} \times \frac{d\hat{n}}{dx}dx + C. \tag{6}$$

## II. GENERAL EXPRESSIONS FOR THE *PN* DIODE CURRENT

As mentioned above, most of the textbook literature assumes the semiconductor to be nondegenerate and thus the derivation that follows from Eq. (6) relies on simplification procedures that assume Boltzmann statistics being strictly valid. To treat degenerate, and in particular amorphous, materials we first derive general and implicit expressions that will later be used to derive the diode current for a given DOS function. In standard semiconductor device textbooks it is common to describe diode characteristics under two extreme structural configurations defined with respect to the carriers' diffusion length. In one case, denoted "long diode," the distance between the contact and the junction is significantly longer than the diffusion length of the minority carriers. In such a configuration the contact has no effect on the current at the junction. The other case, denoted "short diode," is for a very short distance between the contact and the junction, resulting in no significant charge recombination between the junction and the contact. In the case of amorphous organic

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semiconductors the layer's thickness is typically around 100 nm, which may seem short but due to the low mobility of organic semiconductors this may prove to be rather long. We were able to find estimates for diffusion length in intrinsic hole transporting layer[6] of up to 20 $\mu$m but we were not able to find experimental evidence for diffusion length in doped layers, as in a *pn* diode. If one assumes that Langevin recombination holds at the high doping levels then the diffusion length of minority carriers would be well below 100 nm making the diode a long one. Having the above in mind and considering that the analysis presented here could also be applied to other materials systems we will consider both long and short diodes.

### A. Long diode

If the distance between the contact and the junction is much longer than the diffusion length of the minority carriers one can neglect the diffusion current at the contact interface. Mathematically this is equivalent to setting the position of the contact at infinity ($x=\infty$) and at that boundary the following condition holds: $Dd\hat{n}/dx|_{x=\infty}=0$. This boundary condition implies that $C$ in Eq. (6) equals zero. Next we perform change of variables, $\hat{n}=\hat{n}(x)$ and find

$$\left(D\frac{d\hat{n}}{dx}\right)^2\Bigg|_{x=x_p} = 2\times\int_0^{\hat{n}(x_p)}\frac{D}{\tau}\times\hat{n}\times d\hat{n}, \tag{7}$$

and finally

$$J_e = \left. qD\frac{d\hat{n}}{dx}\right|_{x=x_p} = q\sqrt{\frac{2}{\tau}}\times\sqrt{\int_0^{\hat{n}(x_p)} n\times D(n)dn}. \tag{8}$$

Similar expression can be derived for the hole current and thus the diode current is $J=J_e+J_h$.

By rearranging Eq. (8) we can derive a simplified expression for the current,

$$J_e = q\sqrt{\frac{2}{\tau}} \times\sqrt{\underbrace{\left(\frac{\int_0^{\hat{n}(x_p)} n\cdot D(n)dn}{\int_0^{\hat{n}(x_p)} D(n)dn}\right)}_{E(n)}\times\underbrace{\left(\frac{\int_0^{\hat{n}(x_p)} D(n)dn}{\int_0^{\hat{n}(x_p)} dn}\right)}_{E(D)}\times\hat{n}(x_p)}. \tag{9}$$

We note that in the square root the first expression is the expectation value of the carrier density weighed by the diffusion coefficient, the second is the expectation value of the diffusion coefficient, and the third is the excess charge density at the edge of the junction of the $P$ side,

$$J_e = q\sqrt{\frac{2\langle D\rangle}{\tau}}\sqrt{\langle\hat{n}\rangle\times\hat{n}(x_p)}. \tag{10}$$

To show that Eq. (10) reduces to the standard diode equation one assumes $D$ to be independent of the charge density (nondegenerate material),

$$J_e = q\sqrt{\frac{2\langle D\rangle}{\tau}}\sqrt{\langle\hat{n}\rangle\times\hat{n}(x_p)} = q\sqrt{\frac{2D}{\tau}}\sqrt{\frac{\hat{n}(x_p)}{2}\times\hat{n}(x_p)} = q\sqrt{\frac{D}{\tau}}\hat{n}(x_p), \tag{11}$$

and using the Boltzmann approximation that holds for nondegenerate materials,

$$J_e = q\sqrt{\frac{D}{\tau}}\times\bar{n}_p(e^{qV_a/kT}-1), \tag{12}$$

where $V_A$ is the applied voltage. Equation (12) is the text book diode equation.[7]

### B. Short diode

For the short diode one assumes that the recombination in the layer is negligible such that Eq. (5) turns into

$$\frac{d}{dx}\left(D\frac{d\hat{n}}{dx}\right) = \frac{\hat{n}}{\tau}\simeq 0, \tag{13}$$

and hence

$$\left(D(n)\frac{d\hat{n}}{dx}\right) = \text{const} = J_e/q. \tag{14}$$

Integrating Eq. (14) between the contact ($x=0$) and the junction ($x=x_p$) and performing the same change of variables, $\hat{n}=\hat{n}(x)$, we find that the expression for the electron current in a short diode is

$$J_e = \frac{q}{x_p}\int_{n_0}^{n(x_p)} D(n)d\hat{n}. \tag{15}$$

Again, similar expression can be derived for the hole current and thus the diode current is $J=J_e+J_h$.

## III. ANALYSIS OF SPECIFIC DENSITY OF STATES FUNCTIONS

As an example for the general formulas developed above we apply this analysis to three types of DOS functions, which are commonly used to describe amorphous materials: (a) exponential DOS, (b) Gaussian DOS, and (c) Gaussian with an exponential tail. The first one is solved analytically and the latter two are treated numerically. In all cases, both sides of the diode are assumed to have the same type of DOS. Alternatively, if the junction is comprised of materials with different types of DOS, the derivation remains valid if the current in the diode is unipolar.

### A. Exponential DOS

An exponential DOS takes the form of

$$\text{DOS}(E) = \frac{N_t}{kT_0}\exp\left(\frac{E}{kT_0}\right)\times H(-E), \tag{16}$$

where $N_t$ is the total DOS, $T_0$ is the characteristic temperature of the exponential DOS distribution, and $H(-E)$ is the Heaviside function that take the value of 1 for $E\leq 0$ and zero elsewhere. As such an abrupt change in the DOS at $E=0$ is

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not physical we limit ourselves to cases where $T_0 > 300$ k such that the position of the carriers in the DOS would be low enough to justify ignoring the "real" DOS around $E=0$. It has been shown that for such DOS the mobility and the Einstein relation enhancement factor can be written as[8,9]

$$\mu_e \propto Cn^{\beta}; \quad \eta = \frac{T_0}{T}, \tag{17}$$

where $\mu_e$ is the mobility and $C$ is a material dependent parameter. Percolations theory predicts $\beta = T_0/T - 1$ and using the mean medium approximation[10,11] (at $T=300$ and $T_0 \geq 300$) we got: $\beta = 1 - 4.0624 \times \exp(-T_0/300/0.68049)$.

For the voltage dependence we would also need the charge density ($n$) dependence on the Fermi level ($E_F$):[9]

$$n = G\exp\left(q\frac{E_F}{kT_0}\right) = \hat{n} + \bar{n}, \tag{18}$$

where $G$ is only temperature dependent.

#### 1. Long diode

For infinitely long diode case we start with Eq. (8) and insert Eq. (17) into it:

$$\begin{aligned} J_{e\_\exp} &= q\sqrt{\frac{2}{\tau}} \times \sqrt{\int_0^{\hat{n}(x_p)} n \times \frac{kT_{0e}}{q} Cn^{\beta} dn} \\ &= q\sqrt{\frac{2CkT_{0e}}{\tau q}} \times \sqrt{\int_0^{\hat{n}(x_p)} n^{(\beta+1)} dn}. \end{aligned} \tag{19}$$

Solving Eq. (19),

$$\begin{aligned} J_{e\_\exp} &= q\sqrt{\frac{2CkT_{0e}}{\tau q}\frac{1}{\beta+2}} \times \hat{n}(x_p)^{(\beta+2/2)} \\ &= q\sqrt{\frac{2CkT_{0e}}{\tau q}\frac{1}{\beta+2}} \times [n(x_p) - \bar{n}]^{(\beta+2/2)}. \end{aligned} \tag{20}$$

To express the current in terms of applied voltage we first write it as a function of the Fermi level by inserting Eqs. (18) and (20),

$$\begin{aligned} J_{e\_\exp} = q\sqrt{\frac{2CkT_{0e}}{\tau q}} \times \sqrt{\frac{1}{\beta+2}G^{(\beta+2)}}\Bigg[\exp\left(q\frac{E_F}{kT_{0e}}\right) \\ - \exp\left(q\frac{\bar{E}_F}{kT_{0e}}\right)\Bigg]^{(\beta+2/2)} \end{aligned} \tag{21}$$

where $\bar{E}_F$ is the position of the Fermi level within the DOS at equilibrium. Using $V_A = E_F - \bar{E}_F$ and rearranging the terms we arrive at the expression for the electron current,

$$J_{e\_\exp} \propto \left[\exp\left(q\frac{V_A}{kT_{0e}}\right) - 1\right]^{(\beta+2/2)}. \tag{22}$$

Similarly for holes,

$$J_{h\_\exp} \propto \left[\exp\left(q\frac{V_A}{kT_{0h}}\right) - 1\right]^{(\beta+2/2)}.$$

If $T_{0e} = T_{0h} = T_0$ then the ideality factor of $J = J_{e\_\exp} + J_{h\_\exp}$, at high enough voltage, is $n_f = 2\eta/\beta+2$, which is similar to the one derived in Ref. 8 under the assumption that D/$\mu$ is varying slowly, compared to the charge density, across the junction [which strictly holds for exponential DOS, see Eq. (17)]. Assuming $T_0 = 450$ we find $n_f = 1.2$ for both the percolation and Mean Medium Approximation (MMA) models.

#### 2. Short diode

For the short diode case we insert Eqs. (17) and (15),

$$\begin{aligned} J_e &= \frac{q}{x_p}\int_{n_0}^{n(x_p)} C\frac{kT_{0e}}{q}n^{\beta}d\hat{n} \\ &= \frac{q}{x_p}C\frac{kT_{0e}}{q(\beta+1)}(n(x_p)^{(\beta+1)} - n_0^{(\beta+1)}), \end{aligned} \tag{23}$$

$$\begin{aligned} J_e = \frac{q}{x_p}C\frac{kT_{0e}}{q(\beta+1)}G^{(\beta+1)}\Bigg[\exp\left(q\frac{E_F}{kT} \times \frac{\beta+1}{T_0/T}\right) \\ - \exp\left(q\frac{E_{F0}}{kT} \times \frac{\beta+1}{T_0/T}\right)\Bigg], \end{aligned} \tag{24}$$

$$J_e \propto \left[\exp\left(q\frac{V_A}{kT} \times \frac{\beta+1}{T_0/T}\right) - 1\right]. \tag{25}$$

Namely, for the short diode case the characteristics are ideal and the ideality factor is $n_f = \eta/\beta+1$. Assuming $T_0 = 450$ we find $n_f = 1$ and $n_f = 0.97$ for the percolation and MMA models, respectively.

### B. Gaussian DOS

For Gaussian DOS the situation is less trivial as the values for $D(n)$, $\mu(n)$, and $n(E_F)$ are computed numerically.[8,10–12] Example for such computation, using the discrete mean medium approximation,[11] is shown in Fig. 1 and we will use these values to compute the diode characteristics below.

Since the entire computation is numeric in nature we would also derive the ideality factor numerically using

$$n_f = \frac{q}{kT}J\Big/\left(\frac{\partial J}{\partial V_A}\right). \tag{26}$$

#### 1. Long diode

Using the values in Fig. 1 and the numerical relation between the charge density and the Fermi level (not shown) we computed the current [Eq. (8)] as a function of the Fermi level. The calculation was done for two DOS widths of $\sigma = 4$ kT and $\sigma = 5$ kT ($T=300$). Figure 2(b) shows the computed currents, in semilog-$y$, where the centers of the Gaussians were taken as $E=0$. We note that the current tends to level off at the point where the Fermi level reaches the center of the Gaussian and half of the available states are filled. Translating the Fermi level values to applied bias is not trivial and hence we chose to do so by defining that at a bias equal to the built in potential ($V = V_{\mathrm{bi}}$) the current should be negligibly small ($\sim 10^{-10}$ in the current case). The results of this translation are shown in Fig. 2(a). We note that the slope

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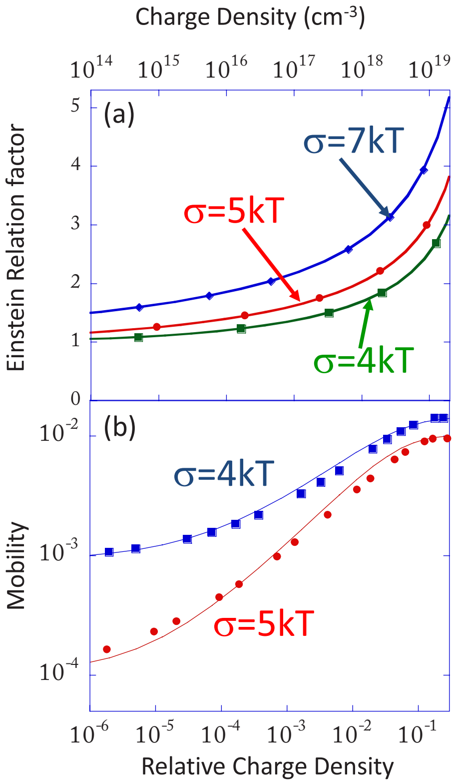

FIG. 1. (Color online) Einstein relation enhancement factor (a) and the charge carrier mobility (b) as a function of the charge density. The calculation was performed for several Gaussian width $\sigma$ as denoted on the graph.

for the DOS of $\sigma=4$ kT (round symbols) is steeper than that of the DOS of $\sigma=5$ kT (square symbols) and that both are not of single exponent.

To derive the ideality factor we apply Eq. (26) to the results shown in Fig. 2 and present them in Fig. 3. Figure 3(a) shows the computed ideality factor as a function of the Fermi-level position relative to the center of the DOS for the two Gaussian DOS functions characterized by $\sigma=5$ kT (square symbols) and $\sigma=4$ kT (round symbols). We observe that the ideality factor is an increasing function of the Fermi level or bias and that it is higher for the wider DOS. Figure 3(b) shows that same data but as a function of current since in practice it would probably be easier to compare different devices by normalizing their currents and not their Fermi levels.

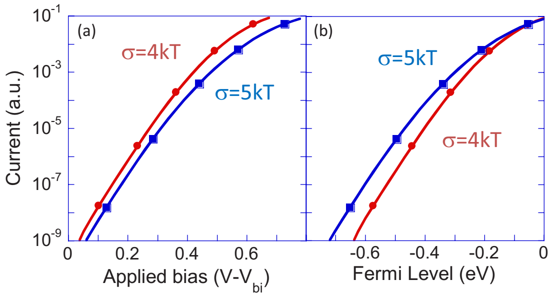

FIG. 2. (Color online) Current as a function of applied bias (a) and of the position of the Fermi level relative to the DOS center (b). The calculations were performed for DOS width of $\sigma=4$ kT (round symbols) and $\sigma=5$ kT (square symbols).

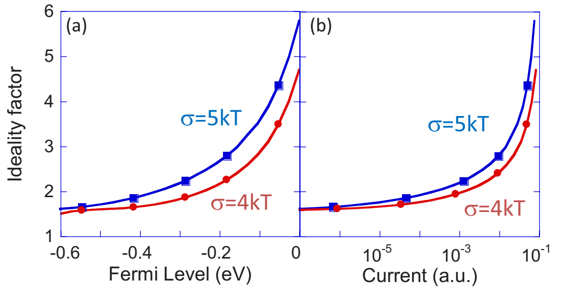

FIG. 3. (Color online) (a) The computed ideality factor as a function of the Fermi-level position relative to the center of the DOS for two Gaussian DOS function characterized by $\sigma=5$ kT (square symbols) and $\sigma=4$ kT (round symbols). (b) Same data as in (a) but presented as a function of the current flowing through the diode.

### *2. Short diode*

Using the values in Fig. 1 and the numerical relation between the charge density and the Fermi level (not shown) we computed the current of the short diode [Eq. (15)] as a function of the Fermi level [Fig. 4(b)]. The calculation was done for two DOS widths of $\sigma=4$ kT and $\sigma=5$ kT ($T=300$). Figure 4(a) shows the characteristics as a function of applied bias using the same normalization (translation) procedure described for Fig. 2.

To derive the ideality factor we apply Eq. (26) to the results shown in Fig. 4 and present them in Fig. 5. Figure 5(a) shows the computed ideality factor as a function of the Fermi-level position relative to the center of the DOS for the short (square symbols) and long (round symbols) diodes. We observe that the ideality factor is an increasing function of the Fermi level or bias and that it is only slightly higher for the long diode. As before, Fig. 5(b) shows that same data but as a function of current since in practice it would probably be easier to compare different devices by normalizing their currents and not their Fermi levels.

## C. Gaussian DOS and exponential tail

Tal *et al.*[4] measured DOS of $N,N^{\mathrm{I}}$-diphenyl-$N,N^{\mathrm{I}}$-bis(1-naphthyl)-1,10-biphenyl–4,4$^{\mathrm{II}}$-diamine ($\alpha$-NPD) to be made of Gaussian and exponential tail with the functional form of

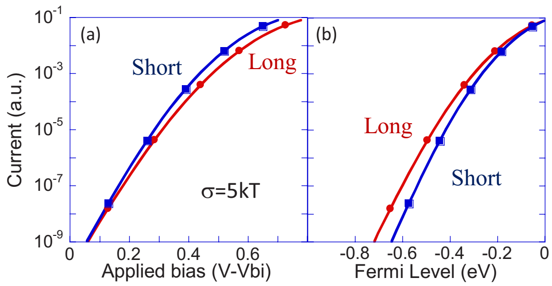

FIG. 4. (Color online) Current as a function of applied bias (a) and of the position of the Fermi level relative to the DOS center (b). The square symbols are the short diode characteristics and the round symbols are for the long diode case. The calculations were performed for DOS width of $\sigma=5$ kT $T=300$.

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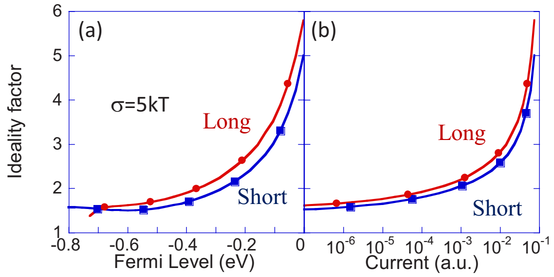

FIG. 5. (Color online) (a) The computed ideality factor as a function of the Fermi-level position relative to the center of the DOS for short (square symbols) and long (round symbols) diodes. (b) Same data as in (a) but presented as a function of the current flowing through the diode. The Gaussian DOS function is characterized by $\sigma=5\ \mathrm{kT}$.

$$\mathrm{DOS} = 10^{21}\frac{1}{\sqrt{2\pi}\sigma}\exp\left(-\frac{1}{2}\left[\frac{E}{\sigma}\right]^{2}\right) + \frac{4.9\times 10^{18}}{kT_0/q}\exp\left(\frac{E}{kT_0/q}\right), \tag{27}$$

where $\sigma/\mathrm{kT}=4$ ($T=300$ k) and $T_0=1350$ (note that unlike Ref. 4 we define here the center of the Gaussian as $E=0$). Using this expression we calculated the mobility[11] and Einstein relation.[5] The results of this calculation are presented in Fig. 6 where the full line denotes the mobility and the dashed line denotes the Einstein enhancement factor ($\eta$). We note that similarly shaped curves were measured in Ref. 13.

Using the same procedure as in the previous sections we computed the current and ideality factors for the complex DOS [Eq. (27)] and the results are presented in Figs. 7 and 8, respectively.

## IV. DISCUSSION

We have developed a general procedure and expressions to deduce the ideality factor in *pn* diodes made of materials that may be degenerate and/or have a density dependent mobility. The expressions are general such that they account also for nondegenerate regime; however, any semiconductor exhibiting Gaussian or exponential DOS would be degenerate under practical device operating conditions. Having emphasized the role of the Einstein relation we should also mention that at high electric fields the Einstein relation may also be enhanced[14] probably due to carrier heating[11] related effects. The analysis presented above involves the current flowing outside the junction area where the electric field would be minimal and, hence, we can justify not including the high electric field effects in our model.

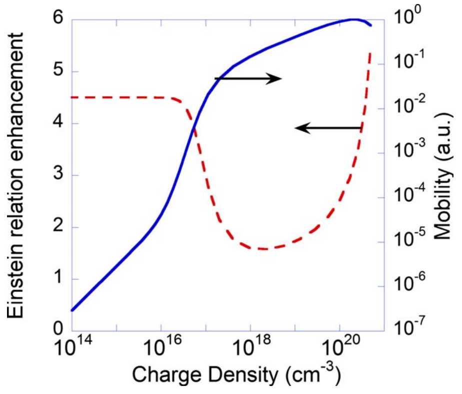

FIG. 6. (Color online) Calculation of the Einstein relation enhancement factor $\eta$ and the mobility as a function of charge density for a DOS having the function form as in Eq. (27).

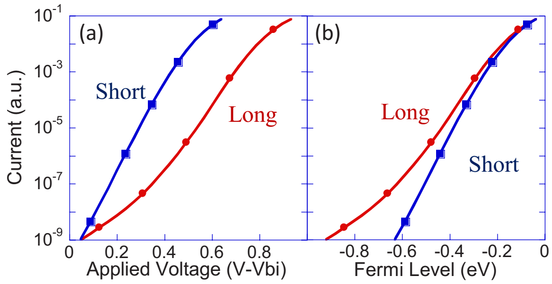

FIG. 7. (Color online) Current as a function of applied bias (a) and of the position of the Fermi level relative to the DOS center (b). The square symbols are the short diode characteristics and the round symbols are for the long diode case. The calculations were performed for DOS function described by Eq. (27) and using the data of Fig. 6.

For the exponential DOS, which is most frequently used in the context of Field Effect Transistors (FETs), we were able to derive analytic expressions for the ideality factor of the long ($n_f=2\eta/\beta+2$) and short ($n_f=\eta/\beta+1$) diodes. We note that for no density dependence ($\eta=1$, $\beta=0$) both converge to the ideal case of $n_f=1$. These expressions show that the degeneracy and enhancement of the Einstein relation ($\eta$) would make the *I-V* less steep or increase the value of $n_f$ and that the power dependence of the mobility on charge density ($\beta$) would make the *I-V* steeper or lower the value of $n_f$.

For the exponential DOS we modeled the transport using both percolation and MMA models. Using the percolation model one finds that a short diode based on a material with exponential DOS would display an ideal diode behavior regardless of the characteristic temperature of the exponential DOS. For the MMA model we find essentially the same result up to $T_0=600$ k and above this value it predicts a slight increase in the ideality factor.

For the Gaussian DOS the solution had to be numeric and we found that it is impossible to strictly define an ideality factor for a *pn* diode based on such a material (see Fig. 5). The analysis was performed for a relatively common disorder parameter ($\sigma=5\ \mathrm{kT}$).[15] We found that for such a DOS

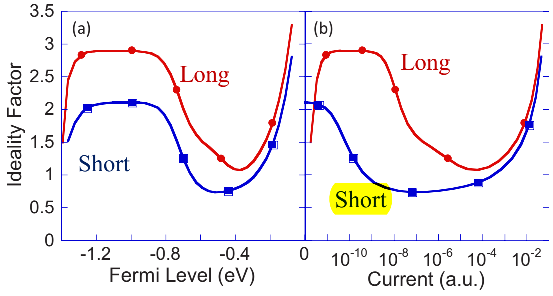

FIG. 8. (Color online) (a) The computed ideality factor as a function of the Fermi-level position relative to the center of the DOS for short (square symbols) and long (round symbols) diodes. (b) Same data as in (a) but presented as a function of the current flowing through the diode. The calculations were performed for DOS function described by Eq. (27) and using the data of Fig. 7.

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the slope of the current (on a semilog-$y$ scale) is bias dependent and that the apparent ideality factor tends to increase with bias. At relatively low currents the ideality factor assumes a value of $n_f=1.5$ but at the higher current regime it is increasing and may reach values as high as $n_f=5$. One can rationalize this by examining Fig. 1 where we note that the Einstein enhancement factor, $\eta$, goes up as a function of the charge density (or quasi-Fermi level). This increase is due to the decrease in the slope of the DOS which in Eq. (1) it appears in the denominator as $\partial n/\partial E_F$. At the very high injection regime the mobility starts to saturate and the reduction in $\beta$ also contributes to the enhancement of the ideality factor. However, we suspect that experimentally it may be difficult to probe the very high $n_f$ regime in a diode configuration. The reasoning is since it occurs where the current becomes significantly high and other effects (as serial resistance) may start playing a role. By comparing short and long diodes we found that there is not much difference in the ideality factor derived for the two cases.

Finally, we analyzed the recently described realistic DOS measured in a FET device configuration.[4,13] Such a DOS is the combination of Gaussian and exponential tail, which was reported to exhibit a nonmonotonic functional form of the Einstein relation as a function of charge density. Again, this can be attributed to the slope of the DOS, which in this case is changing nonmonotonically due to the transition from exponential to Gaussian shape. Evaluating numerically the characteristics of a diode based on such material revealed that the nonmonotonic behavior is also found for the ideality factor, $n_f$, as a function of current. Unlike the pure Gaussian or pure exponential cases the complex DOS case shows significant difference in the ideality factor between short and long diodes (Fig. 8). This difference is largely observed at the low current regime and is large enough such that one might be able to observe such differences experimentally.

## V. CONCLUSIONS

Through the analysis of $pn$ diodes made of materials having different DOS' shape we were able to show that the $I$-$V$ characteristics are highly dependent on the DOS. The moderate rise in the DOS as a function of energy results in a less steep rise of the current giving rise to the enhancement of the diode's ideality factor. In a Gaussian type DOS, where the total DOS is finite, the current tends to saturate at higher bias giving rise to farther enhancement of the ideality factor. Namely, careful measurements of the diode $I$-$V$ curve and its derivation using Eq. (26) should reveal the type or shape of the DOS of the material inside the operating device. Finally, our results indicate that making the diode a short diode improves the ideality factor and drives it toward 1. While this may be technologically challenging it could provide another route for improving amorphous $pn$ diodes characteristics.

## ACKNOWLEDGMENTS

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