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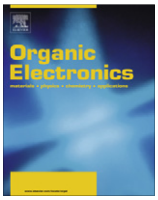

Tail states recombination limit of the open circuit voltage in bulk heterojunction organic solar cells

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ABSTRACT

An analytical theory is presented for bimolecular recombination through tail states and open circuit voltage in bulk heterojunction organic solar cells. It is developed rigorously using the hopping transport and the drift diffusion theory. Based on the proposed model, a variety of temperature, energy disorders of the material and illumination intensity dependencies of the open circuit voltage can be well described. Good agreement between the calculation and recent experimental data is found.

1. Introduction

Recombination processes in disordered hopping transport is of special current interest because it is one of the main factors limiting the conversion efficiency in organic solar cells. In the low mobility organic semiconductor, when the charge carrier hopping distance or the energy dissipation length of inelastic scattering is shorter than the Coulomb radius, the charge carrier bimolecular recombination is determined by the probability that the electrons and holes meet in space. This process is usually described by the Langevin theory [1], where the total rate of bimolecular recombination R_{rec} is proportional to the charge carrier mobility and carrier concentration by $R_{rec} = e(\mu_e + \mu_h)np/\varepsilon\varepsilon_0 = \beta np$, where μ_e and μ_h are the electron and hole motilities, respectively, $\varepsilon\varepsilon_0$ is the dielectric permittivity, n is the electron concentration, p is hole concentration, e is the elementary electronic charge, and $\beta = e(\mu_e + \mu_h)/\varepsilon\varepsilon_0$ is defined as the bimolecular recombination coefficient [2]. Relative analytical and numerical models for bimolecular recombination rate or recombination coefficient in the two dimensional lamellar structures of RRP3HT have been developed recently [2–4]. It was found that the recombination coefficient indeed depends on the total carrier concentration and the concentration dependence of the recombination rate should obey the power law as

$$R_{rec} \propto n^{\gamma} = n^{2.43}, \tag{1}$$

and thus β is weakly dependent on n as

$$\beta \propto n^{0.43}. \tag{2}$$

These results were verified by Monte Carlo simulation [2]. In regular organic solar cell, the power law $R_{rec} \propto n^{\gamma}$ has also been reported [5–10]. However, experimental data show that the parameter γ should be temperature dependent [11]. Moreover, charge transport in organic solar cells is typically described by multiple trapping or variable range hopping between the localized states [12–18], which should be taken into account as well in the recombination model.

In this paper, we will present an analytical bimolecular recombination coefficient model to include the concentration dependent mobility and the disorder of organic materials. Based on this recombination theory, a temperature, light intensity and disorder dependent open circuit voltage of bulk heterojunction organic solar cell is formulated.

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2. Model theory

In order to include the energetic disorder into a useful model that can describe the dependence of the recombination coefficient, we assume a density of states (DOS) distribution for both the donor highest occupied molecular orbital (HOMO) and the acceptor lowest unoccupied molecular orbital (LUMO) manifolds. Here the energy disorder for organic semiconductor is modeled by an exponential DOS with mean energy E_{LUMO} and width T_0 for the acceptor material (energy E_{HOMO} and width T_1 for the donor material) as [19,20]

$$g_n(E_n) = \frac{N_n}{k_B T_0} \exp\left(\frac{E_n - E_{LUMO}}{k_B T_0}\right) \quad (E < E_{LUMO}), \tag{3.a}$$

$$g_p(E_p) = \frac{N_p}{k_B T_1} \exp\left(\frac{-E_p + E_{HOMO}}{k_B T_1}\right) \quad (E > E_{HOMO}). \tag{3.b}$$

where N_n (N_p) denotes the total density of electron (hole) states. Considering the Fermi–Dirac function

$$f(E, E_F) = \frac{1}{1 + \exp\left(\frac{E - E_F}{k_B T}\right)}, \tag{4}$$

where k_B is the Boltzmann constant, the photo-generated total electrons n (holes p) can be calculated as

$$n = \int_{-\infty}^{\infty} dE_n g_n(E_n) f(E_n, E_{Fn}), \tag{5.a}$$

$$p = \int_{-\infty}^{\infty} dE_p g_p(E_p)\left[1 - f(E_p, E_{Fp})\right], \tag{5.b}$$

where E_{Fn} (E_{Fp}) is the position of the Fermi level for electrons (holes).

Connecting (3), (4) and (5), one can calculate the total electron (hole) concentration as

$$n = N_n \exp\left[\frac{E_{Fn} - E_{LUMO}}{k_B T_0}\right] \Gamma(1 - T/T_0)\Gamma(1 + T/T_0). \tag{6.a}$$

$$p = N_p \exp\left[\frac{E_{HOMO} - E_{Fp}}{k_B T_1}\right] \Gamma(1 - T/T_1)\Gamma(1 + T/T_1). \tag{6.b}$$

Here $\Gamma(z) = \int_0^\infty dy \exp(-y) y^{z-1}$. Under a percolation model, the mobility in the hopping system can be therefore calculated as [21,22] (see detailed derivations in the Appendix)

$$\mu = \frac{\sigma_0}{e}\left(\frac{\pi}{B_c \Gamma\left(1 - \frac{T}{T_0}\right)\Gamma\left(1 + \frac{T}{T_0}\right)}\left(\frac{T_0}{2\alpha T}\right)^3\right)^{\frac{T_0}{T}} \left(N_n \exp\left(\frac{E_{Fn} - E_{LUMO}}{k_B T_0}\right)\Gamma\left(1 - \frac{T}{T_0}\right)\Gamma\left(1 + \frac{T}{T_0}\right)\right)^{\frac{T_0}{T} - 1} = \mu_0(T) n^{\gamma}. \tag{7}$$

Here B_c is the critical number of the percolation system ($B_c \approx 2.8$ for three dimensional system [22]), $\gamma = T_0/T - 1$, α is the inverse localization length, σ_0 is conductivity prefactor in the percolation theory [10], and $\gamma = T_0/T - 1$. Considering that the mobility is dependent on the carrier concentration (therefore the position in (1) of [23]), the recombination coefficient model based on constant mobility has to be recalculated. Using the mobility developed above, we consider an area with radius r where there is one hole at $r = r'$ and electrons are around it, as described in [23]. The hole at the origin serves as a drain for incoming electrons. Electrons move within this area due to the radial electric fields $E(r)$ of the hole,

$$E(r) = \frac{e}{4\pi\varepsilon\varepsilon_0} \frac{1}{r^2}. \tag{8}$$

The radial electron current in this area, I, is related to the coordinated dependent electron concentration n by the following equations

$$\frac{I}{4\pi r^2} = -\mu_0(T) n E(r) - D\frac{\partial n}{\partial r}. \tag{9}$$

Here D is the diffusion coefficient ($D = \mu k_B T/e$ according to the Einstein relation). Substituting (7), (8) into (9), one can obtain

$$\frac{I}{4\pi r^2} = -\frac{e\mu_0(T) n^{\gamma+1}}{4\pi\varepsilon\varepsilon_0 r^2} - \frac{k_B T \mu_0(T)}{e(\gamma + 1)} \frac{\partial n^{\gamma+1}}{\partial r}. \tag{10}$$

The solution of (10) is

$$n^{\gamma+1} = \left[\frac{I}{k}\int_r^{r'} dx' \frac{1}{x'^2} \exp\left(-\frac{a}{r}\right) + C\right] \exp\left(\frac{a}{r}\right), \tag{11}$$

where $k = 4\pi\mu_0(T) k_B T/e(1 + \gamma)$ and $a = e^2(1 + \gamma)/4\pi\varepsilon\varepsilon_0 k_B T$. The boundary conditions for this problem are: $n(r') = 0$ (i.e., there is no electron), and $n(\infty) = n_0$ (the average bulk carrier concentration, in this manuscript, this value is set as the average experimental carrier concentration by charge extraction techniques). Then, (11) reduces to

$$I = ak\frac{n_0^{\gamma+1}}{1 - \exp\left(-\frac{a}{r'}\right)}. \tag{12}$$

Since the recombination coefficient, β, is the current per unit density, I/n_0 [23], we have

$$\beta = ak\frac{n_0^{\gamma}}{1 - \exp\left(-\frac{a}{r'}\right)}. \tag{13}$$

If we choose $r' = 0$, (13) becomes

$$\beta = \frac{\sigma_0}{\varepsilon\varepsilon_0}\left(\frac{\pi}{B_c \Gamma\left(1 - \frac{T}{T_0}\right)\Gamma\left(1 + \frac{T}{T_0}\right)}\left(\frac{T_0}{2\alpha T}\right)^3\right)^{\frac{T_0}{T}} n_0^{\frac{T_0}{T} - 1}. \tag{14}$$

Next we assume that the photogenerated carriers exceed the intrinsic carrier concentration and the bulk recombination is dominated over the outer interface recombination [24]. The Fermi level position can then be evaluated by requiring the system to satisfy the following conditions: first, the recombination rate must balance the photogeneration rate; second, the illuminated semiconductor must be space-charge neutral. The latter condition holds at open circuit, which reduces to

$$n = p = n_0, \tag{15}$$

Connecting of (14) and (15), we can calculate the recombination rate in the steady state as

$$R_{rec} = \beta n_0^2 = \vartheta(T) n_0^{\frac{T_0}{T}+1} = G(1-x)/L. \tag{16}$$

Here $\vartheta(T) = \frac{\sigma_0}{\varepsilon\varepsilon_0}\left(\frac{\pi}{B_c\Gamma\left(1-\frac{T}{T_0}\right)\Gamma\left(1+\frac{T}{T_0}\right)}\left(\frac{T_0}{2\alpha T}\right)^3\right)^{\frac{T_0}{T}}$, G is the absorbed photon flux, L is the absorption length and x stands for the ratio of geminate electron–hole pairs which recombine before charge separation. From (16), the average electron (hole) concentration in steady states reads as

$$n_0 = \left(\frac{G(1-x)}{\vartheta(T)L}\right)^{\frac{T}{T+T_0}}. \tag{17}$$

Submitting (17) into (6.a) and (6.b), we get

$$E_{Fn} = E_{LUMO} + k_B T_0 \times \ln\left\{\frac{1}{N_n\Gamma(1-T/T_0)\Gamma(1+T/T_0)}\left[\frac{G(1-x)}{\vartheta(T)L}\right]^{\frac{T}{T+T_0}}\right\}. \tag{18.a}$$

$$E_{Fp} = E_{HOMO} - k_B T_1 \times \ln\left\{\frac{1}{N_p\Gamma(1-T/T_1)\Gamma(1+T/T_1)}\left[\frac{G(1-x)}{\vartheta(T)L}\right]^{\frac{T}{T+T_0}}\right\}. \tag{18.b}$$

Finally, the open circuit voltage V_{oc} is determined by the Fermi levels as

$$qV_{oc} = E_{Fn} - E_{Fp}. \tag{19}$$

3. Results and discussion

From the expression (14), we can see that the recombination coefficient increases superlinearly with average carrier concentration. This is directly attributed to the increased mobility with the carrier concentration: higher concentration will increase the average energy, thus facilitating an activated hopping to the transport energy [25]. In Fig. 1, the recombination coefficient is plotted against the average carrier concentration under different temperatures. The dotted line is experimental data for the recombination coefficient in bulk heterojunction solar cells of a 1:1 blend of thick film of regioregular P3HT (Rieke Metals) and PCBM (Solenne b.v.) [11]. The theoretical calculations (solid lines) follow the model in (14), where we use σ_0, α and T_0 as the fitting parameters. The agreement between the calculation and experimental data is clearly observed. Here the input parameters are: $\varepsilon = 3$ (which is an approximation for most organic solids), $\sigma_0 = 12$ s/m, $T_0 = 512$ K, $\varepsilon_0 = 8.854 \times 10^{-12}$ K, and $\alpha^{-1} = 3.35$ nm. With these fitting parameters, we computed a mobility of 7.6×10^{-5}cm^2/Vs for $T = 300$ K and $n_0 = 5 \times 10^{22}$/m^3. In addition, the disorder parameter $T_0 = 512$ K, which corresponds to the density characteristic energy $E^{\mu} = k_B T_0 = 44.2$ meV [26], is consistent with the value observed for P3HT:PCBM system in [26].

From (18.a), (18.b), and (19), the linear relation between the open circuit voltage and the effective band gap E_g ($E_g = E_{LUMO} - E_{HOMO}$) is found as $qV_{oc} = E_g - \varDelta$, where the energy loss $\varDelta$ is strongly dependent on the energy disorder and the temperature. The temperature dependences of the open circuit voltage at different illumination intensities are shown in Fig. 2. The input parameters are $E_g = 1.7$ eV, $\alpha^{-1} = 1$ nm, $L = 100$ nm, $N_n = N_p = 1 \times 10^{22}$ cm^{-3}, $T_0 = T_1 = 500$ K, and $G = 7.5 \times 10^{16}$cm$^{-2}s^{-1}$ (for simplicity we assume $x = 0$ as in [24]), and the other parameters are the same as in Fig. 1. It is demonstrated that the open-circuit voltage decreases linearly with the temperature at a given G. The inset of Fig. 2 shows the open circuit voltage vs. the temperature at different energy disorders. We can see that a lower energy disorder (small T_0) implies an increment in V_{oc}. Fig. 3 compares the calculated and experimental data [27] of the light intensity dependent open circuit voltage at different temperatures in MMO-PPV/PCBM solar cell. The fitting parameters are $\alpha^{-1} = 3$ nm, $N_n = N_p = 1 \times 10^{21}$/cm^3, $T_0 = 500$ K,

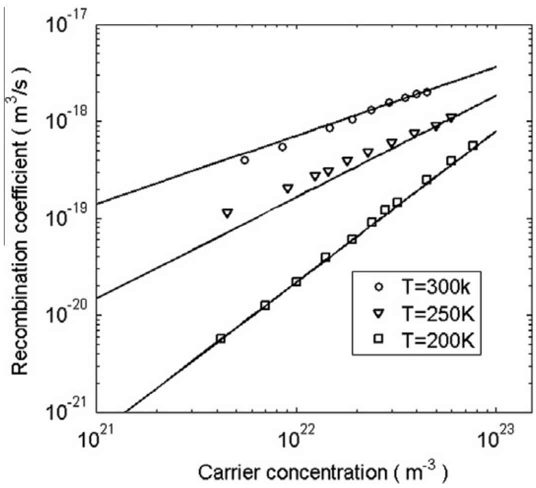

Fig. 1. Comparison between the calculation and experimental data for carrier concentration dependent recombination coefficient for different temperatures.

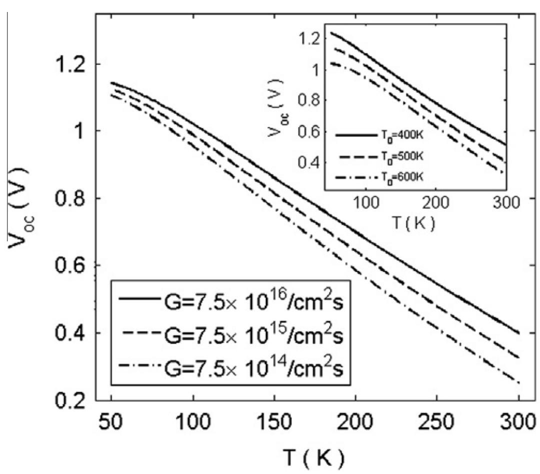

Fig. 2. Temperature dependences of the open circuit voltage at different illumination intensity. Inset: the temperature dependent open circuit voltage at different energy disorder of organic semiconductor.

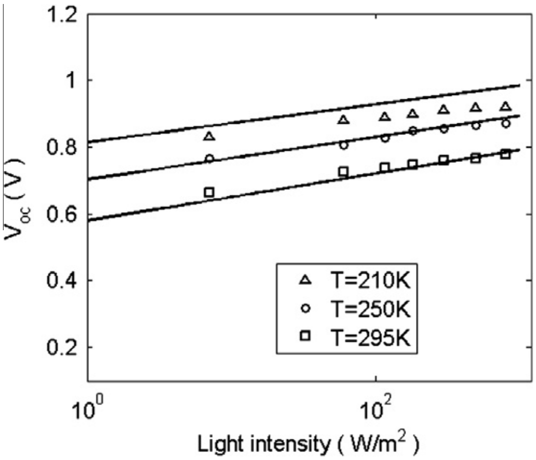

Fig. 3. The comparison between calculation and experimental data of light intensity dependent open circuit voltage at different temperature. The solid lines are theoretical calculations given by (19).

$T_1 = 400$ K, $E_g = 1.4$ eV, and $\sigma_0 = 4 \times 10^{10}$ s/m, and other parameters are the same as in [27]. Here we can see that the open circuit voltage is linearly dependent on $\log(G)$. Fig. 4 illustrates the temperature dependent open circuit voltage for large disorder parameters. The input parameters are $\alpha^{-1} = 3$ nm, $N_n = N_p = 1 \times 10^{21}/\text{cm}^3$, $T_0 = T_1$, $L = 100$ nm, $G = 7.5 \times 10^{16}\text{cm}^{-2}s^{-1}$, $E_g = 1.7$ eV, and $\sigma_0 = 4 \times 10^{10}$ s/m. It is nevertheless obvious that in the low temperature domain, the open circuit voltage increases with the temperature. At $T_{\max}$ of order 130 K, the open circuit voltage passes through the maximum and finally approaches $V_{oc} \propto T^{-1}$, which is quite different from the observation using small energy disorder parameters (Fig. 2). We also find that for a small value of α^{-1}, the open circuit voltages V_{oc} will increase with disorder parameter T_0, which can be seen from Fig. 5 ($\alpha^{-1} = 8$ Å and the other parameters are the same as in Fig. 4). This phenomena has been observed by experimental data for solar cell with DPM-12/P3HT blend [28], where lack of crystallization in the DPM-12 based device will lead to the change of DOS of the film, hence its high V_{oc} values.

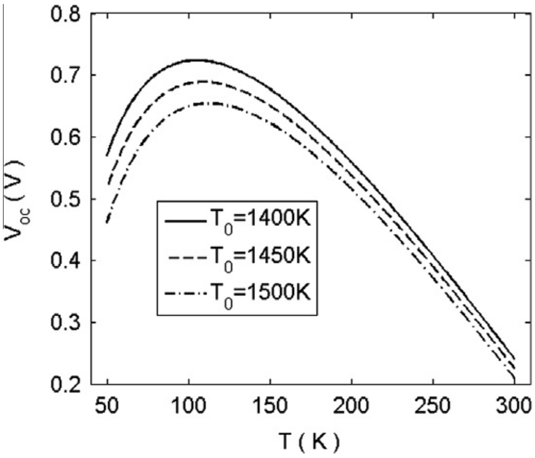

Fig. 4. Temperature dependences of the calculated open circuit voltage for large disorder parameters at $\alpha^{-1} = 3$ nm.

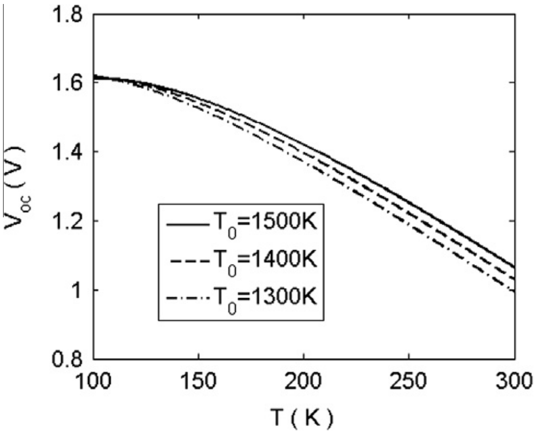

Fig. 5. Temperature dependences of the calculated open circuit voltage for small disorder parameters at $\alpha^{-1} = 8$ Å.

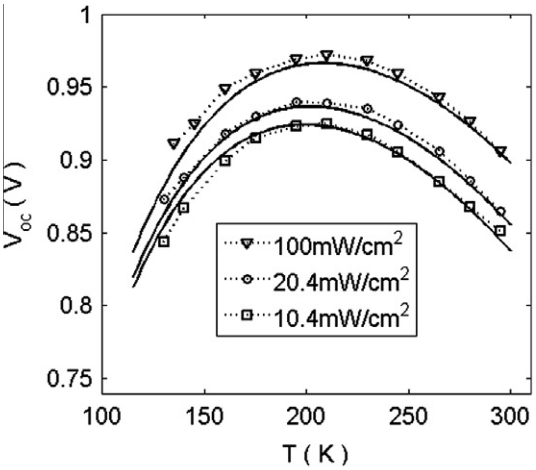

Fig. 6. The comparison between the calculation and experimental date of temperature dependent open circuit voltage. The solid lines are theoretical calculations given by (19).

The comparison between the theoretical and experimental data for the temperature dependent open circuit voltage V_{oc} of solar cell made using a blend of DPM-12 and OC1C10-PPV [29] is plotted in Fig. 6. The fitting parameters are $\alpha^{-1} = 3.9$ Å, $N_n = N_p = 3 \times 10^{22}/\text{cm}^3$, $T_0 = 3000$ K, $T_1 = 400$ K, $E_g = 1.4$ eV [30], and $\sigma_0 = 1.6 \times 10^9$ s/m. Good agreement is found here. We can see that, the deviation of $V_{oc}(T)$ from the linear dependence on T and the extremely high open circuit voltage V_{oc} of the DPM-12 based device can be explained by using a high energy disorder parameter T_0. The detailed physical mechanisms for the high open circuit voltage of the DPM based device is discussed in [31,32].

4. Conclusion

Bimolecular recombination processes and open circuit voltage model for organic solar cells are discussed. We show that, the recombination coefficient will increase with the carrier concentration superlinearly. The energetic disorder of organic semiconductors, the illumination intensity and temperature dependencies of the open circuit voltage can also be interpreted in the light of these theoretical insights. The calculated temperature and carrier concentration dependencies agree well with the observed open circuit voltage in organic bulk heterojunction solar cells.

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Appendix A

The charge transport is described by hopping between localized states according to the Miller–Abrehams equation

$$v = v_0 \begin{cases} exp\left(-2\alpha R_{ij} - \frac{E_j - E_i}{k_B T}\right) & E_j - E_i \geqslant 0 \\ exp(-2\alpha R_{ij}) & E_j - E_i \leqslant 0 \end{cases}, \tag{A1}$$

where v is the hopping rate, v_0 is the attempt-to-jump frequency, R_{ij} is the distance between sites i and j, α is the inverse localization length, E_i and E_j are the energy at the initial and the finial states, respectively. Under a percolation model, the system is treated as a random resistor network connecting each molecular site [21,22]. One may assign a conductance $G_{ij} = G_0 \exp[-s_{ij}]$ between site i and site j where

$$s_{ij} = 2\alpha R_{ij} + \frac{|E_i - E_{Fn}| + |E_j - E_{Fn}| + |E_i - E_j|}{2k_B T} \tag{A2}$$

The conductivity of the system could be expressed as $\sigma = \sigma_0 \exp(-S_c)$, where S_c is the exponent of the critical percolation conductance [21]. The onset of percolation is determined by calculating the critical average number of bonds per site

$$B(G = G_c) = B_c = \frac{N_B(S_{c'E_{Fn}})}{N_s(s_{c'Fn})}, \tag{A3}$$

where N_B and N_s are the density of bonds and density of sites in the percolation system, respectively. The density of bonds is given by [21]

$$N_B = 4\pi \int R_{ij}^2 g(E_i) g(E_j) \theta(s_c - s_{ij}) dE_i dE_j dR_{ij}, \tag{A4}$$

where θ is step function. The density of sites N_s that can form a bond is

$$N_s = \int g(E) \theta(s_c k_B T - |E - E_{Fn}|) dE_i dE_j dR_{ij}. \tag{A5}$$

Substituting (3) and (A2) into (A3), (A4), and (A5) we obtain the percolation criterion for our system

$$B_c = \pi \exp\left(\frac{E_{Fn} - E_{LUMO} + k_B T s_c}{k_B T_0}\right) \left(\frac{T_0}{2\alpha T}\right)^3. \tag{A6}$$

Combining the expression of the system conductivity with (16), the expression of the carrier mobility as a function of temperature and total carrier density eventually reads $\mu = \frac{\sigma_0}{e} \left(\frac{\pi}{B_c \Gamma\left(1-\frac{T}{T_0}\right) \Gamma\left(1+\frac{T}{T_0}\right)} \left(\frac{T_0}{2\alpha T}\right)^3 \right)^{\frac{T_0}{T}} \left(N_n \exp\left(\frac{E_{Fn}-E_{LUMO}}{k_B T_0}\right) \Gamma\left(1 - \frac{T}{T_0}\right) \Gamma\left(1 + \frac{T}{T_0}\right)\right)^{\frac{T_0}{T}-1} = \mu_0(T) n^{\gamma(T)}$.

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