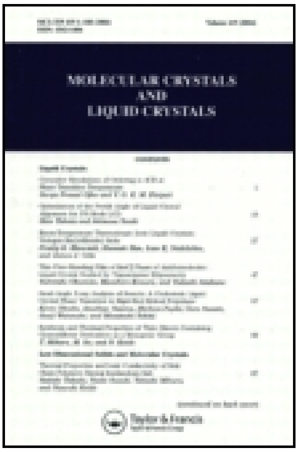

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Does the Temperature Dependence of the Charge Carrier Mobility in Disordered Organic Semiconductors at Large Carrier Concentrations Obey the Meyer-Neldel Compensation Law?

I. I. Fishchuk [a] , A. K. Kadashchuk [b c] , J. Genoe [b] , V. N. Poroshin [c] & H. Bässler [d]

[a] Institute for Nuclear Research, National Academy of Sciences of Ukraine, Kyiv, Ukraine

[b] IMEC, Kapeldreef, Heverlee-Leuven, Belgium

[c] Institute of Physics, National Academy of Sciences of Ukraine, Kyiv, Ukraine

[d] Philipps-Universität Marburg, Hans-Meerwein-Strasse., Marburg, Germany

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Does the Temperature Dependence of the Charge Carrier Mobility in Disordered Organic Semiconductors at Large Carrier Concentrations Obey the Meyer–Neldel Compensation Law?

I. I. FISHCHUK,[1] A. K. KADASHCHUK,[2,3] J. GENOE,[2]
V. N. POROSHIN,[3] AND H. BÄSSLER[4]

[1]Institute for Nuclear Research, National Academy of Sciences of Ukraine, Kyiv, Ukraine
[2]IMEC, Kapeldreef, Heverlee-Leuven, Belgium
[3]Institute of Physics, National Academy of Sciences of Ukraine, Kyiv, Ukraine
[4]Philipps-Universität Marburg, Hans-Meerwein-Strasse., Marburg, Germany

The temperature-activated charge transport in disordered organic semiconductors at large carrier concentrations has been thoroughly considered, by using a recent analytical model [Phys.Rev.B **76***, 045210 (2007)] assuming a Gaussian density-of-states (DOS) distribution and Miller–Abrahams jump rates. We demonstrate that the apparent Meyer–Neldel compensation rule is recovered with regard for the temperature dependences of the charge carrier mobility upon varying the carrier concentration, but not for varying the DOS distribution width. We show that this phenomenon is entirely due to the evolution of the occupational DOS distribution as a function of the state filling. Predictions of the model are in a quantitative agreement with available experimental results.*

Keywords Charge carrier mobility; Gaussian density-of-states distribution; Mayer–Neldel rule; organic field-effect transistors; organic semiconductors

Introduction

Many modern electronic solid state devices such as field-effect transistors (FETs), light emitting diodes (LEDs), and solar cells or processes like the electrophotographic process employed in todays's photocopiers are based upon non-crystalline materials such as organic semiconductors [1–3] or amorphous silicon. In order to improve their operation, it is essential to understand how charge carriers in amorphous solids are transported. In crystalline materials, electronic transport bands exist. In contrast, the random structure present in amorphous solids implies that the electronic states are localized and energetically distributed, so that the transport

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Address correspondence to A. K. Kadashchuk, Institute of Physics, NAS of Ukraine, Prospect Nauky 46, 03028 Kyiv, Ukraine. E-mail: kadah@iop.kiev.ua occurs via the incoherent hopping in this density of states (DOS) [4–6]. During the device operation, only a part of the DOS is populated by charges. The shape of the DOS and its occupied fraction determine the transport properties of a material. There are two limiting cases.

(i) An amorphous solid is intrinsically an insulator, as is the case for many organic π-conjugated materials. Their electronic transport is due to extrinsic charges that are either injected from the electrodes or are created by photogeneration. In these materials, the density of occupied transport states (n) is much lower that the total density of states (N), i.e., $n \ll N$. Therefore, each charge carrier explores an essentially empty DOS. If the DOS is a Gaussian with variance σ, the (almost infinitely diluted) ensemble of non-interacting carriers at not too low temperatures has been shown to form an occupational density of states distribution (ODOS) [4] of the same Gaussian shape yet displaced by an energy $\varepsilon_0 = -\sigma^2/k_B T$ below the center of the DOS. In this case, the Fermi level ε_F is below the thermal quasiequilibrium level ε_0 and, as a consequence, is irrelevant for the charge hopping process. Charge transport requires jumps from temporarily filled ODOS-states to empty DOS-states, and the temperature dependence of the charge mobility follows the $\ln(\mu) \propto T^{-2}$ law [4,6] and does not depend on the carrier concentration.

(ii) The other extreme case is realized in low-bandgap disordered inorganic semiconductors, such as amorphous silicon. In this case, a part of the DOS is filled, so that a Fermi level is established, and the Fermi–Dirac distribution determines the ODOS. Here, for not too low temperatures, the charge carrier mobility features the simple thermal activation dependence $\ln(\mu) \propto T^{-1}$ with virtually constant activation energy reflecting the temperature independent position of the Fermi level ε_F with respect to the mobility edge [7], although the activation energy does depend on the charge carrier concentration reflecting a concentration shift of the Fermi level.

An intermediate situation is established, when a sizeable fraction of the available hopping states is occupied in an intrinsically insulating material. This is realized, for example, in an organic FET, in which the transport is confined to a very thin transport channel, so that n/N can reach values of 10^{-2} or higher [8]. Measurements of the charge carrier mobility in organic materials at large carrier concentrations have revealed an Arrhenius-type $\ln(\mu) \propto T^{-1}$ dependence that extrapolates to an unusually high prefactor mobility of $30 \ldots 40\,\mathrm{cm}^2/\mathrm{Vs}$ [9] which exceeds values determined via the time-of-flight technique by $2 \ldots 3$ orders of magnitude [4,6]. Interestingly, the Arrhenius-type $\mu(T)$ dependences measured at different gate voltages and, concomitantly, the charge carrier densities and on different FET structures intersect at a given finite T_0, by suggesting that the Meyer–Neldel rule (MNR) [10] is obeyed.

The MNR is an empirical relation originally derived from chemical kinetics [10]. It describes the fact that the enthalpy and the entropy of a chemical reaction are proportional. More generally, it states that, in a thermally activated rate process, an increase of the activation energy is partially compensated by an increase of the prefactor, so that

$$R(T) = R_{00} \exp(E_a/E_{MN}) \exp(-E_a/kT), \tag{1}$$

where E_{MN} is called the "Meyer–Neldel energy". There are numerous examples, notably, in semiconductor physics, in which the MNR is fulfilled. In context of the multiple trapping dominating the charge transport, it was associated with the presence of the exponential distribution of traps [11–15]. Widenhorn et al. [16] argued that the MNR arises for an arbitrary system, where both the intrinsic rate process and a process involving impurities contribute, while Yelon and Movaghar [17] suggested that the MNR is obeyed when a thermally activated process requires a large number of individual excitations which increase the entropy. Recently, Emin [18] advanced an adiabatic polaron hopping model that considers the carrier-induced softening of the atoms' vibrations at the electron motion and showed that this results in the MNR. It neglects the inherent energetic disorder which has been shown to be of key importance for the charge transport in amorphous organic materials used for OFETs, even when polaron effects in some materials are essential [6,19]. Obviously, there is no common explanation for the MNR effect, and there might be no universal explanation at all [16].

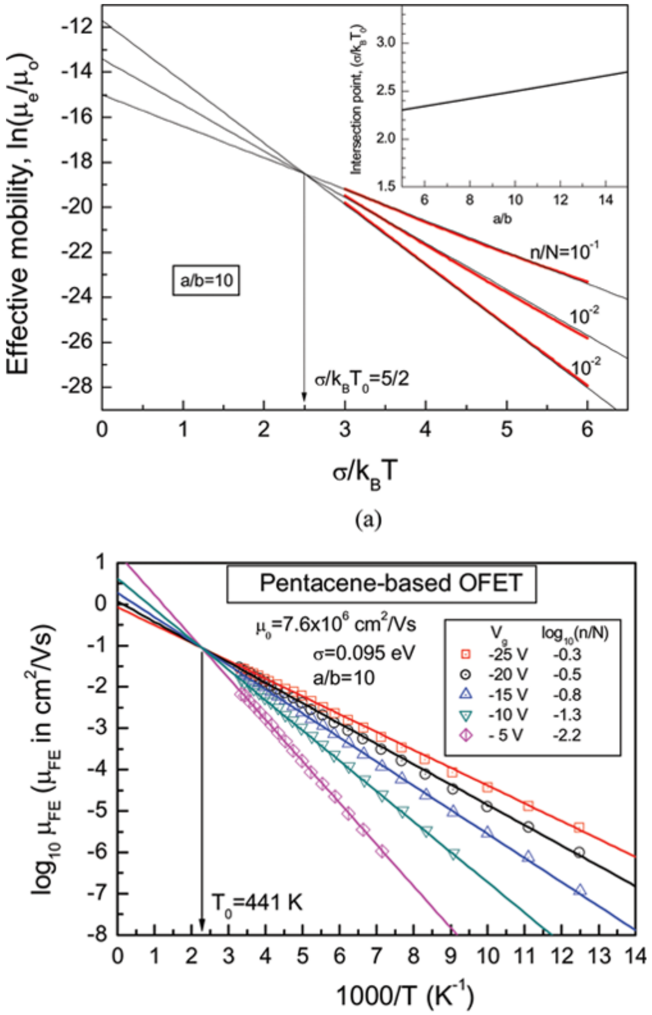
Discussion

In this work, we have applied the recently suggested theory we draw heavily upon the effective medium formalism [20], which is based on a Gaussian DOS distribution and Miller–Abrahams jump rates, to consider thoroughly the temperature dependence of the charge mobility in disordered organic solids at large carrier concentrations. In particular, we were aimed to rationalize the physics behind the experimentally observed MNR effect and related phenomena in the framework of a hopping-transport model with accounting for the evolution of the ODOS distribution as a function of the carrier concentration in a Gaussian-type inherent disordered system. We will show that the establishment of the MNR is a characteristic signature of the hopping transport in a random system with a variable concentration of carriers *irrespective of their polaronic character*. Considering that the electron-phonon coupling is weak in the relevant materials, e.g., polyphenylene- and polythiophene-type conjugated polymers and C_{60}, and, consequently, the charge transport is controlled by disorder rather than by polaron effects [4–6], we use Miller–Abrahams-type jump rates and the concept of effective transport energy [6]. The essential system parameters are the variance σ of the Gaussian DOS, $g(\varepsilon)$, the ratio of densities of occupied and total transport states n/N, and the ratio a/b of the intersite distance (a) and the localization radius (b) of a charged site. For an arbitrary charge concentration, the ODOS distribution is given by

$$P(\varepsilon) = \frac{g(\varepsilon)f(\varepsilon, \varepsilon_F)}{\int_{-\infty}^{\infty} d\varepsilon \; g(\varepsilon)f(\varepsilon, \varepsilon_F)}, \tag{2}$$

where $f(\varepsilon, \varepsilon_F)$ is given by the Fermi–Dirac statistics, and ε_F is the Fermi level obtained from $n = \int_{-\infty}^{\infty} d\varepsilon \; g(\varepsilon)f(\varepsilon, \varepsilon_F)$. For details of the calculations, see Ref. [20].

Figure 1*a* (thick curves) shows the calculated temperature dependences of the effective mobility plotted on a $\ln(\mu_e/\mu_0)$ *vs.* σ/k_BT scale for different charge-carrier concentrations, assuming $a/b = 10$. The calculations are restricted to a temperature regime defined by $\sigma/k_BT \geq 3$. The Arrhenius-type of the $\mu(T)$ dependence indicates that the ODOS is virtually temperature-independent, quite in contrast of the case where $n/N \to 0$. A remarkable result is that if one would – hypothetically – extend



(a)

(b)

Figure 1. (a) Temperature dependence of effective charge mobility $\ln(\mu_e/\mu_0)$ vs. σ/k_BT calculated by Eqs. (25, 42, 45) from Ref. [20] (thick red curves) for different carrier concentrations and $a/b = 10$. Thin lines show the extrapolation to $T \to \infty$ by Eq. (4). The inset shows the intersection point σ/k_BT_0 vs. the a/b ratio; (b) Temperature dependence of the FET mobility in solution-processed pentacene measured at different V_g (symbols) [21] and results of their fitting with Eq. (4) (solid lines). The MNR isokinetic temperature T_0 is indicated by arrow. The estimated material parameters and the effective carrier concentrations $\log(n/N)$ for corresponding gate voltages are shown in the figure.

the above calculations to higher temperatures, then the asymptotes (thin lines) would intersect at a finite temperature $T_0 \cong 2\sigma/5k_B$.

Note that the effective medium approach [20] does not allow one to present the results in a closed analytic form. However, the calculated results including their (hypothetical) extension toward infinite T can be parametrized in terms of an approximate analytical equation for the charge carrier mobility μ_e as a function of σ/k_BT, a/b, and n/N,

$$\mu_e = \mu_0 \exp\left[-2\frac{a}{b} + \frac{1}{2}\left(\frac{a}{b} - 7\right)\right] \exp \left\{-\left[1 - \frac{3}{40}\frac{a}{b} - \frac{1}{1'5}\frac{a}{b}\log_{10}\left(\frac{n}{N}\right)\right]\left(\frac{\sigma}{k_BT} - y_0\right)\right\}, \tag{3}$$

where $y_0 = (1/5)[21/2 + (1/5)(a/b)]$. It is valid for $8 < a/b < 12$ and for $10^{-3} \leq n/N \leq 10^{-1}$, which is relevant for the FET operation. The inset in (Fig. 1*a*) proves that the isokinetic temperature T_0, at which the $\ln(\mu) \propto T^{-1}$ graphs intersect, depends rather weakly on a/b. Rewriting Eq. (3) yields

$$\mu_e = \mu_0 \exp\left[-2\frac{a}{b} + \frac{1}{2}\left(\frac{a}{b} - 7\right)\right] \exp\left[-E_a\left(\frac{1}{k_BT} - \frac{1}{k_BT_0}\right)\right], \tag{4}$$

where

$$E_a = \left[1 - \frac{1}{40}\frac{a}{b} - \frac{1}{15}\frac{a}{b}\log_{10}\left(\frac{n}{N}\right)\right]\sigma; \; T_0 = \frac{E_{MN}}{k_B} = \frac{\sigma}{k_By_0}. \tag{5}$$

It turns out that Eq. (4) can be extended to lower carrier concentrations, $10^{-5} \leq n/N < 10^{-3}$, if E_a is substituted by $E_a^* = 0.85E_a$. Equation (4) is nothing else than the conventional Meyer–Neldel relation [cf. Equation (1)] that has been verified by experiments on several FET devices [21–23]. An example is the data for the hole mobility in a FET based on pentacene (see Fig. 1*b*). Assuming $a/b = 10$, $T_0 = 441$ K translates into $\sigma = 0.095$ eV, which is a value typical of a disordered organic material derived from time-of-flight studies of the charge transport [4]. Note, however, that the experimental data cover only the $T < T_0$ regime, i.e., there is no experimental proof that the extrapolation on $\mu(T)$ toward $T > T_0$ is justified. In fact, we argue that this extrapolation is basically unwarranted. Another example is the T-dependence of the hole mobility in a FET with poly-3-hexylthiophene (P3HT) [21,22] (not shown here). Data bear our MNR-behavior with an isokinetic temperature $T_0 = 460$ K that translates into $\sigma = 0.099$ eV. This is in excellent agreement with the value of 0.098 eV derived before from fitting the temperature-dependent current-voltage (J-V) characteristics measured on this material [24]. The fact that, for $T < T_0$, the activation energy E_a [see Eq. (5)] decreases at increasing the gate voltage is an indication that the ODOS moves closer to the center of DOS upon increasing n/N.

Interestingly, the structure of Eq. (4) predicts that if one extends a family of $\ln(\mu)$ vs. T^{-1} graphs calculated for $T < 400$ K at the variable E_a, i.e., by changing the active transistor layer, yet a constant carrier concentration as $T \rightarrow \infty$, they would intersect at $T \rightarrow \infty$ rather than at a finite T_0 as the MNR would imply (see Fig. 2). This is in disagreement with the conventional MNR that predicts a correlation between the prefactor rate and the activation energy regardless of how the change of E_a is accomplished, i.e., by either changing the DOS distribution width itself or changing the degree of state filling.

In Figure 2, we present the experimental data for $\ln(\mu) \propto T^{-1}$ taken at a constant (moderately low) concentration of holes in diodes that operate in the space charge

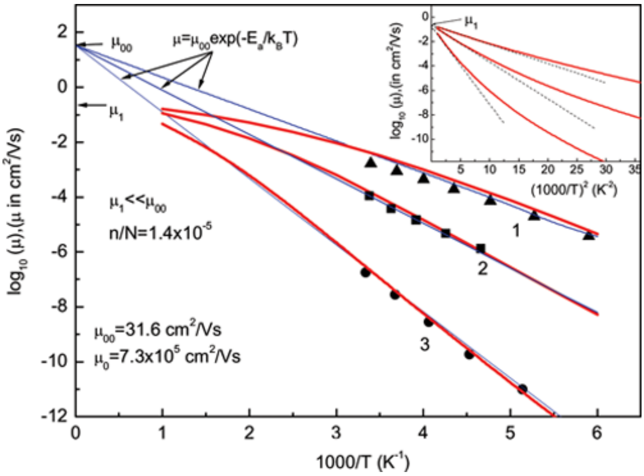


Figure 2. Arrhenius plot of temperature dependence for the charge mobility measured in (1) PCBM, (2) region-regular (rr)-P3HT, and (3) OC_1C_{11}-PPV films (symbols) [9]. Fitting the experimental data by the present theory (Eqs. 25, 42, and 45 from Ref. [20]) (thick red curves) and extrapolated (see text) by Eq. (4) (thin curves). E_a and σ-values are derived via data fitting using Eq. (5) and are listed in Table 1. Insert shows the same calculated temperature dependences plotted in $\log(\mu) \propto T^{-2}$ scale to get an intersect at μ_1.

limited transport regime and are based on different organic semiconductors, namely, poly-3-hexylthiophene (P3HT), poly[2-methoxy-5-(3′,7′-dimethyloctyloxy)-*p*-phenylene vinylene] (OC_1C_{10}-PPV), and [6,6]-phenyl-C_{61}-butyric acid methyl ester (PCBM) [9]. If one would extrapolate those data to $T \to \infty$, one would – and this is the essential message of Craciun's et al. work [9] – ends up with a $T \to \infty$ mobility of $\approx 30 \ldots 40\,\mathrm{cm}^2/\mathrm{Vs}$. However, the theory advanced above indicates that this extrapolation is illegitimate as it is illegitimate in the "MNR" case shown in (Fig. 1). The reason is that, at higher temperatures, the ODOS is no longer determined by the T-independent carrier concentration but by the T-dependent lift toward the transport energy and, concomitantly, toward the center of the DOS distribution. Thick curves in (Fig. 2) show the calculated temperature dependence of the mobility. At high temperatures, as soon as ε_0 becomes larger than ε_F, $\mu(T)$ approaches the conventional $\ln(\mu) \propto T^{-2}$ dependence, featuring a $T \to \infty$ prefactor mobility μ_1 of about $0.16\,\mathrm{cm}^2/\mathrm{Vs}$ that is, indeed, consistent with data derived from time-of-flight studies but inconsistent with the conclusion in Ref. [9] predicting unjustifiably a large prefactor mobility of $\approx 30\,\mathrm{cm}^2/\mathrm{Vs}$. It should be mentioned that the physical meaning of the prefactor mobility is the mobility in a material devoid of disorder like as molecular crystals where the room-temperature charge mobility typically ranges from 0.1 to $1\,\mathrm{cm}^2/\mathrm{Vs}$. It depends on the a/b ratio. Therefore, a similar prefactor observed in different materials implies that this ratio is rather similar for them, although there are no physical reason for the existence of one universal prefactor mobility for organic semiconducting materials. The data analysis based upon the present work [see. Eq. (5)] allows us to extract the disorder parameter σ (see Table 1). The agreement with σ-values determined before from the measured temperature dependences of the hole mobility in these materials is gratifying.

The straightforward conclusion is that the apparent observation of the MNR is related to the charge accumulation in a FET at a variable gate voltage and has nothing to do with a compensation between the prefactor and the activation energy in a thermally activated kinetic process. Since, in the current theory, (i) the polaronic character of a charge carrier plays no role, and (ii) the input prefactor in the expression for the jump rate is invariant, the apparent MNR behavior must solely be due to the distribution of charges within the DOS upon increasing their concentration. The present theory allows one to calculate how the shape of the ODOS evolves as a function of the charge concentration (Fig. 3). For very small carrier concentrations, $n/N = 10^{-7}$, the ODOS is a Gaussian with the same width as that of the DOS itself yet displaced by $\varepsilon_0 = -\sigma^2/k_BT$. Note that the majority of the activated jumps start from the upper portion of the ODOS rather than from its center. When the DOS is loaded by charge carriers, the shift of the ODOS distribution toward the DOS center is accompanied by a *gradual shrinking* (narrowing) with increase in the carrier concentration (see Fig. 3). Therefore, activated jumps start, on average, from somewhat lower (occupied) states implying that the mobility increases less as a function of the carrier concentration comparing to what one would expect if the ODOS retains its shape. This explains why ln $\mu(T^{-1})$ graphs intersect at a finite temperature T_0 at variable n/N, while, at a constant (moderately low) n/N, they intersect as $T \to \infty$. We should emphasize that this effect is inherent just in a Gaussian-shaped DOS but not for an exponential DOS distribution. In the latter case, the ODOS distribution, as follows from the calculations (not shown here), always retains its shape independent of the carrier concentration. This circumstance has another important implication showing that approximating a Gaussian DOS by an exponential distribution [28], where the thermal quasiequilibrium level ε_0 is never established, is unjustified for organic solids and potentially cannot predict correctly the temperature dependence of the mobility.

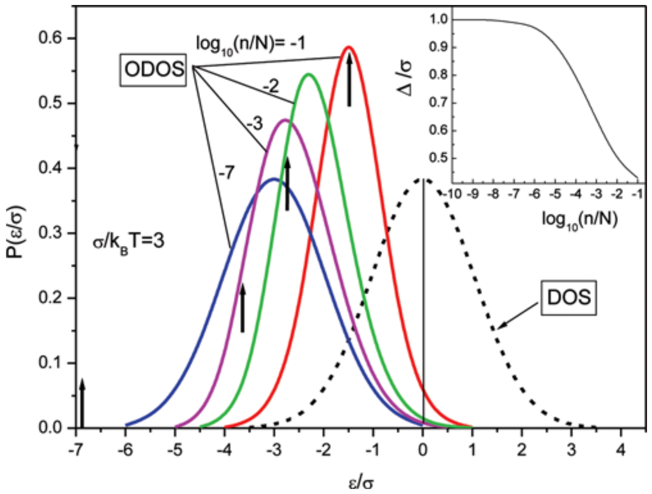


Figure 3. Normalized ODOS distribution in a disordered organic semiconductor with Gaussian DOS at different carrier concentrations n/N calculated by Eqs. (2) for $\sigma/k_BT = 3$. The DOS distribution is shown by dotted curve for reference. Arrows indicate position of calculated Fermi levels for the considered carrier concentrations. Inset shows calculated width of the ODOS, Δ, as a function of carrier concentration.

Figure 3 shows how the energetic distribution of charge carriers migrating within a Gaussian DOS evolves as a function of the charge carrier concentration. For $n/N \rightarrow 0$, the ODOS is virtually empty. It is temperature-dependent and describes which energy level is visited by an individual carrier in the course of its stochastic motion. In this case, the Fermi level is well below the ODOS center. Upon the progressive state filling due to the carrier injection into a FET, the counter-charges being located within the gate electrode, the Fermi level lifts gradually and is located within the ODOS [20,24,27]. Its leading edge shrinks, yet it is still much broader than that in a classic semiconductor. This controls the charge transport as a function of both the carrier concentration and the temperature. A detailed treatment of this phenomenon will be presented in a subsequent paper.

Conclusions

In summary, the principal results of this study are the following: (i) the observed MNR behavior for the FET charge mobility in organic disordered solids is demonstrated to be a direct consequence of the functional dependence of the state filling in a Gaussian-type hopping system, and the polaron formation needs not necessarily be involved to rationalize this phenomenon; (ii) the narrowing of the occupational DOS profile upon increasing the carrier concentration occurs for materials with a Gaussian DOS distribution (but not for an exponential DOS) and gives rise to the MNR effect; (iii) extrapolation of the $\ln(\mu) \propto T^{-1}$ data to $T \rightarrow \infty$ suggested in the previous studies is unwarranted and yields unjustifiably large the mobility prefactor; (iv) the obtained compact analytical relations can be readily used for the estimation of important material parameters and the effective carrier concentration from experimentally accessible $\mu(T)$ data.

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