

# Charge carrier transport in organic semiconductors

K. C. Kao and W. Hwang, Electrical Transport in Solids, (Pergamon, New York, 1981), p.159

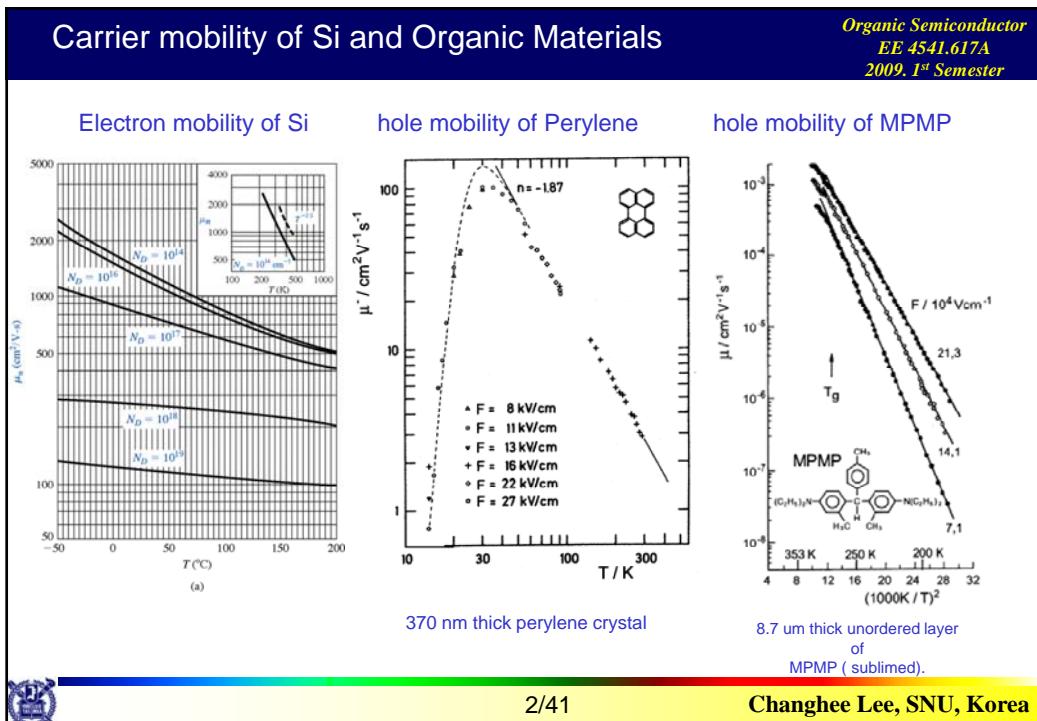
2009. 5.12.

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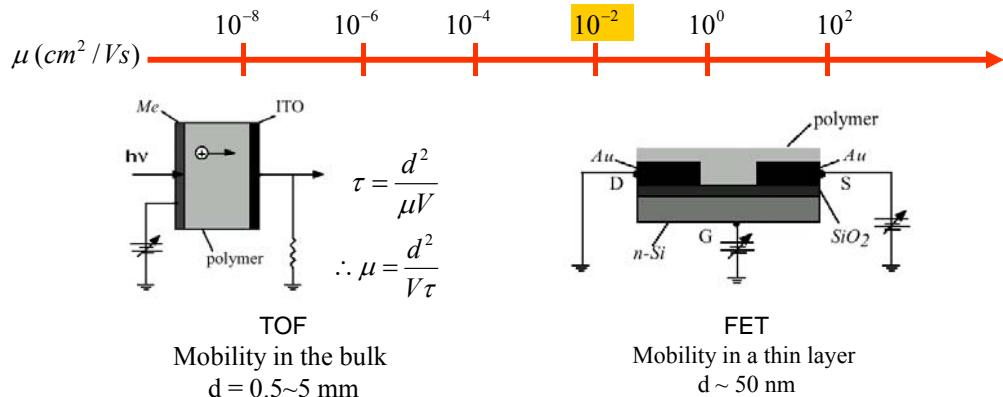


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## Mobility Measurement Techniques

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EE 4541.617A  
2009, 1<sup>st</sup> Semester



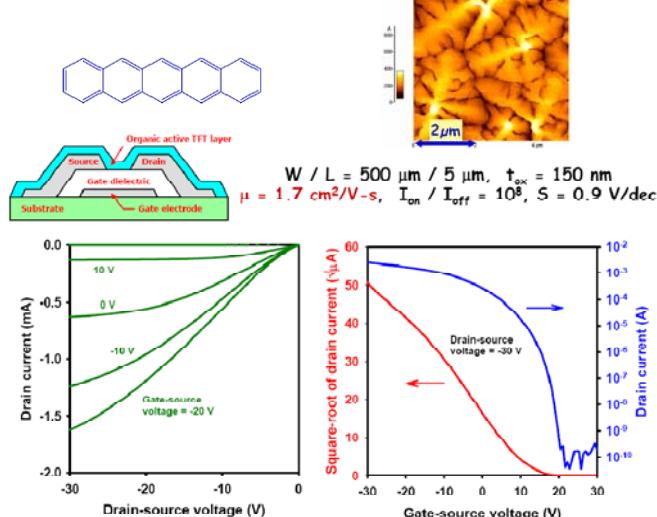
### Other mobility measurement techniques

- Dark injection in the space-charge limited current regime
  - I-V characteristics of space charge limited current
  - Transient EL
  - SHG measurement [T. Manaka, E. Lim, R. Tamura, M. Iwamoto, *Nature Photon.* 1, 581–584 (2007).]
- .....



## Mobility Measurement Techniques: Organic TFT

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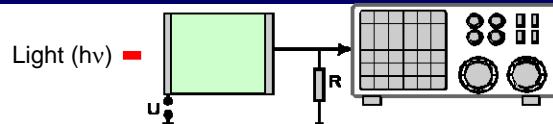


T. N. Jackson, Penn State U.



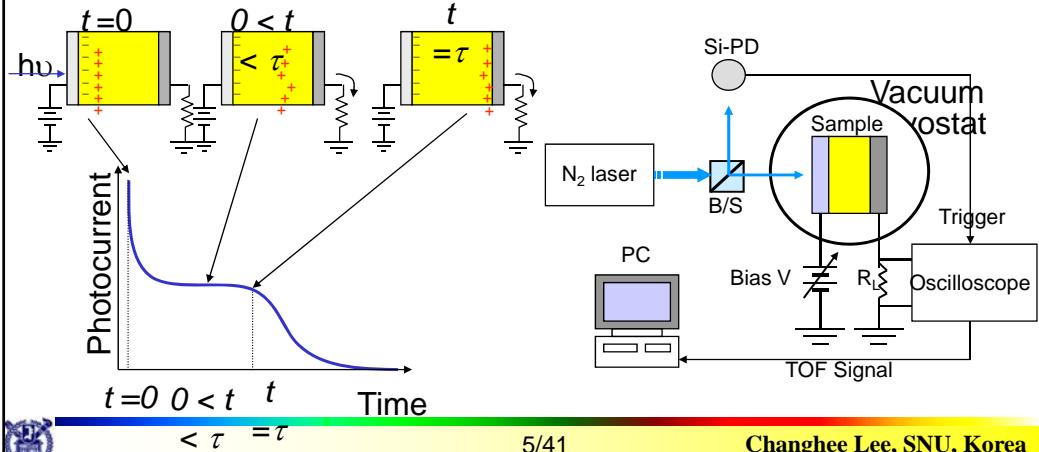
## Mobility measurement techniques: TOF-PC

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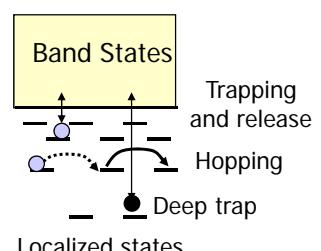
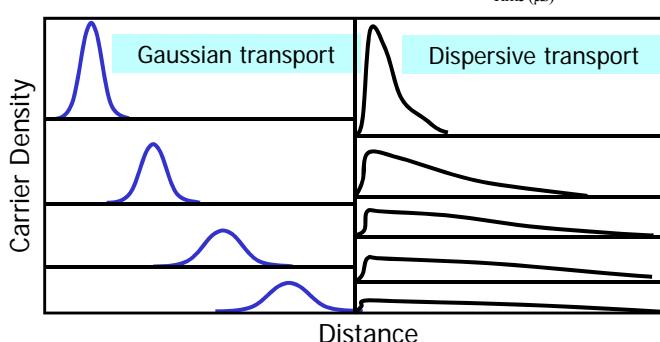
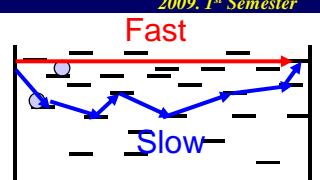
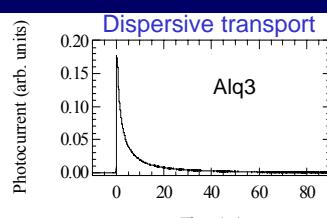
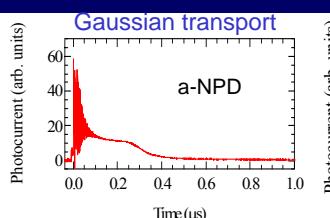
$$\mu = \frac{d^2}{V\tau}$$

### Time-of-flight photoconductivity



## Transport of charge carriers in organic materials

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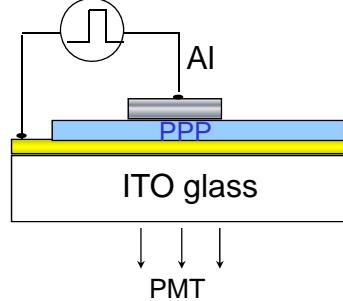


Ref.) Harvey Scher, Michael F. Shlesinger and John T. Bendler, Physics today, Jan. p.26 (1991)



## Mobility Measurement Techniques: Transient EL

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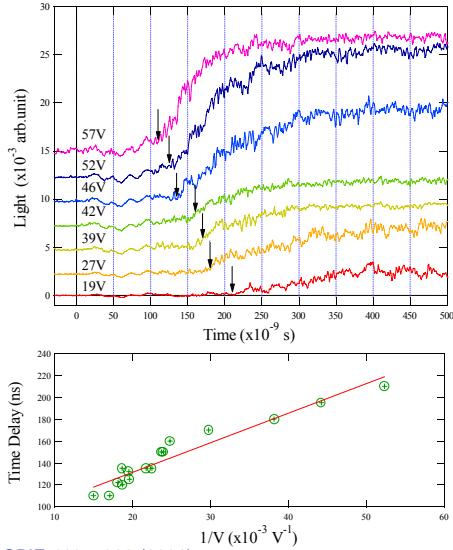
Mobility from the delay time

$$\tau = d^2 / \mu V$$

Hole mobility

in the vacuum-deposited PPP:

$$\text{ITO/PPP/Al: } \mu = 4.5 \times 10^{-6} \text{ cm}^2/\text{Vs}$$



G. W. Kang, C. H. Lee, W. J. Song, and C. Seoul, SPIE 4105, 362 (2001).

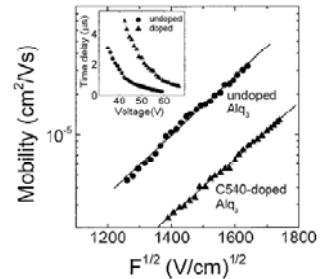
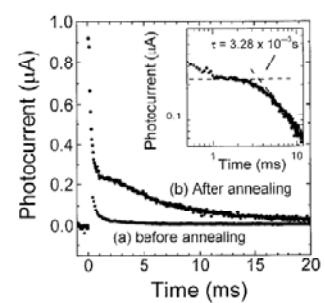
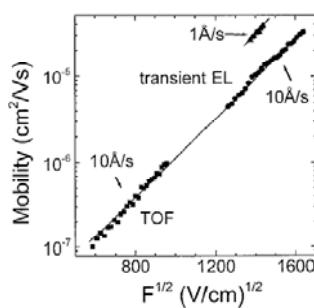
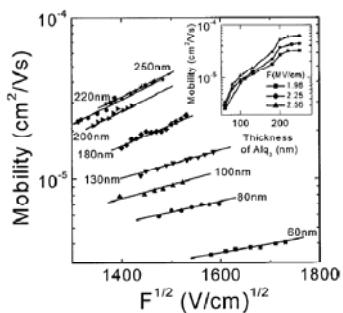
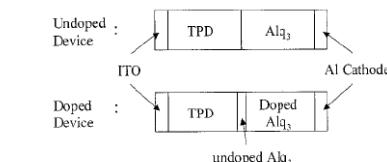


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## Transient EL Mobility vs TOF-PC mobility in Alq<sub>3</sub>

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For a thin layer of Alq<sub>3</sub> ( $d < 180$  nm), it is found that  $t_d$  is affected by both the charging effect and carrier transit time through the Alq<sub>3</sub> layer. For a thicker layer of Alq<sub>3</sub> ( $d > 200$  nm),  $t_d$  approaches the intrinsic electron transit time through Alq<sub>3</sub>.

S. C. Tse, H. H. Fong, and S. K. So, J. Appl. Phys. 94, 2033 (2003)



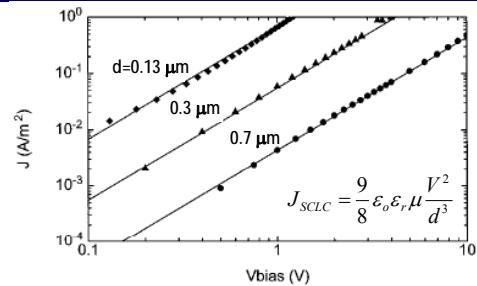
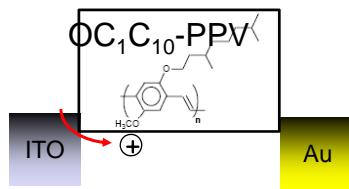
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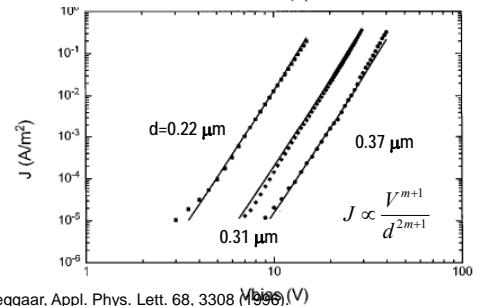
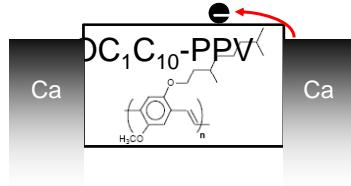
## Space-charge-limited current

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Hole only device



Electron only device



Poly(dialkoxy-p-phenylene vinylene) (OC<sub>x</sub>C<sub>10</sub>-PPV)

P. W. M. Blom, M. J. M. de Jong, and J. J. M. Vleggaar, Appl. Phys. Lett. 68, 3308 (1996).



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## Transient SCLC

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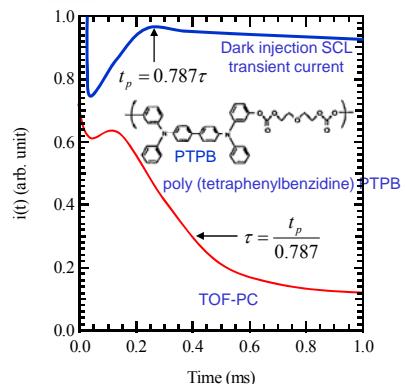
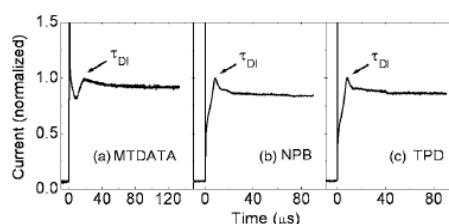
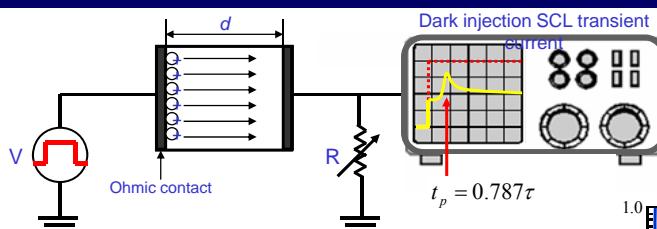


FIG. 5. Room temperature DI signals for MTDATA, NPB, and TPD under applied field strengths of 0.10, 0.09, and 0.09 MV/cm, respectively. The film thicknesses for MTDATA, NPB, and TPD were 0.76, 4.11, and 5.13 μm, respectively.

S.C. Tse, S.W. Tsang, and S.K. So, J. Appl. Phys. 100, 063708 (2006).

M. Abkowitz, J. S. Facci, and M. Stolka, Appl. Phys. Lett. 63, 1892 (1993).



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## Mobility Measurement Techniques: dark charge injection

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In a monopolar and single-layer configuration, the carrier transit time is shorter than in the absence of space-charge effects due to the enhancement of the electric field at the leading edge of the carrier packet.

$$t_{tr} = 0.786 \frac{d}{\mu E}$$

The transient current overshoots its steady-state value by a factor of 1.21 and starts at 0.44 times the steady-state value.

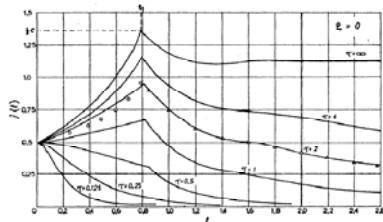
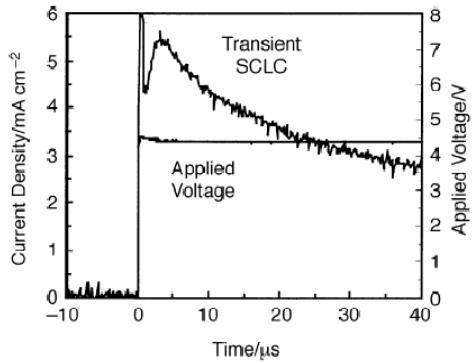


FIG. 5. The time dependence of the SCLC density for insulating crystals characterized by various trapping times, and  $\theta_0 = 0$ .

A. Many and G. Rakavy, Phys. Rev. 126, 1980 (1962)

ITO/300nm m-MTDATA/Ag with ITO biased positively.



M. Stossel et al., Phys. Chem. Chem. Phys. 1, 1791 (1999)



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## Carrier injection efficiency

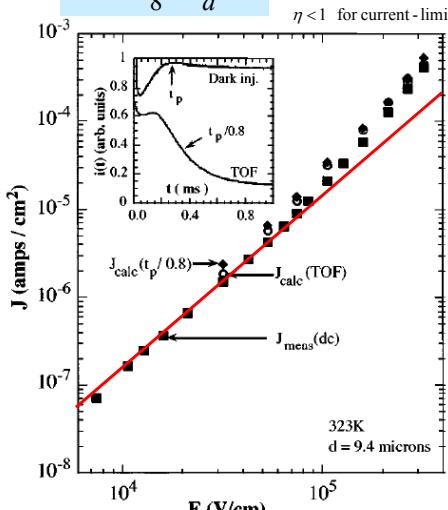
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$$J_{SCLC} = \frac{9 \epsilon_0 \epsilon_r \mu V^2}{8 d^3}$$

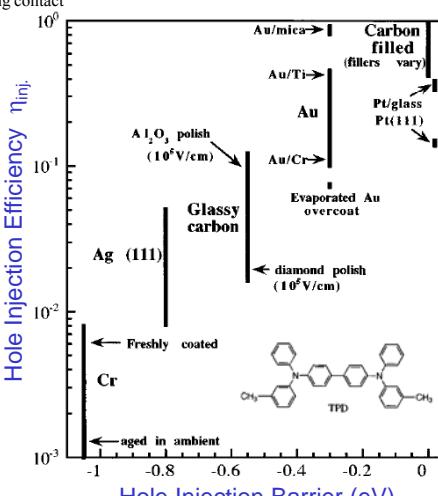
Injection efficiency:  $\eta = \frac{\text{injected current}}{\text{SCLC}}$

$\eta = 1$  for ohmic contact

$\eta < 1$  for current-limiting contact



M. Abkowitz, J. S. Facci, J. Rehm, J. Appl. Phys. 1998, 83, 2670.

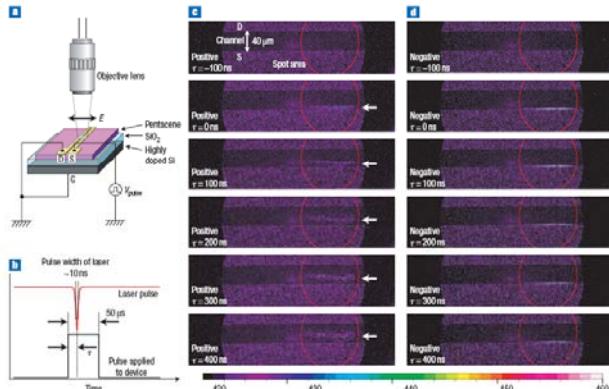


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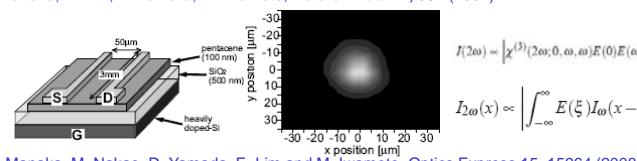
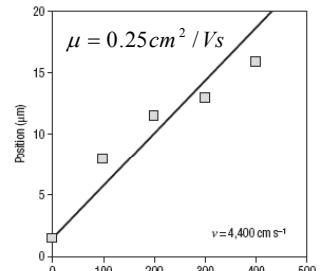
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## Mobility Measurement Techniques: TRM-SHG

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T. Manaka, E. Lim, R. Tamura, M. Iwamoto, *Nature Photon.* 1, 581 (2007).



T. Manaka, M. Nakao, D. Yamada, E. Lim and M. Iwamoto, *Optics Express* 15, 15964 (2008).

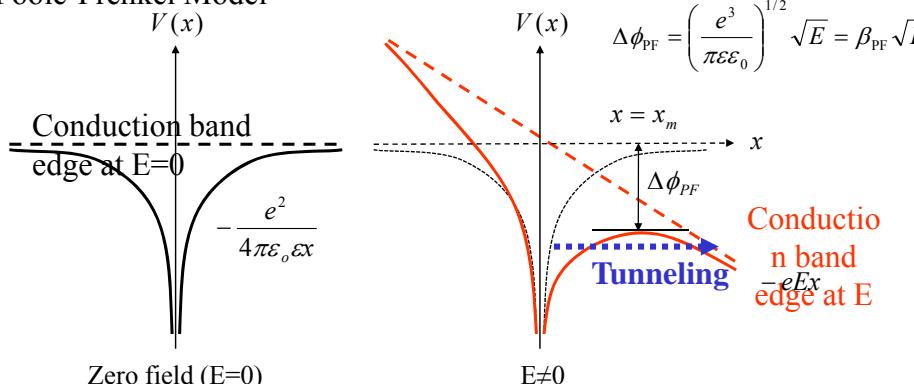
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## Charge Transport in Disordered Organic Solids

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### (1) Poole-Frenkel Model



$$\mu(F, T) = \mu_{PF} \exp\left(-\frac{\Delta E}{k_B T}\right) \exp\left(\frac{\beta_{PF} \sqrt{E}}{k_B T}\right)$$

$$\beta_{PF} = \sqrt{\frac{e^3}{\pi \epsilon \epsilon_0}}$$

$\Delta E$  : Activation energy at  $\mu_{PF}$   $E=0$   
 $\beta_{PF}$  : PF Mobility



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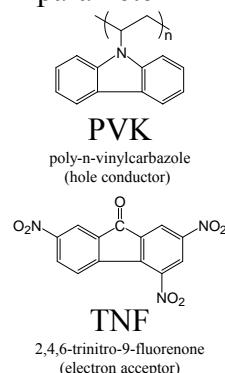
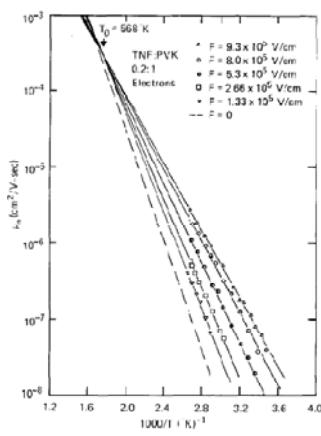
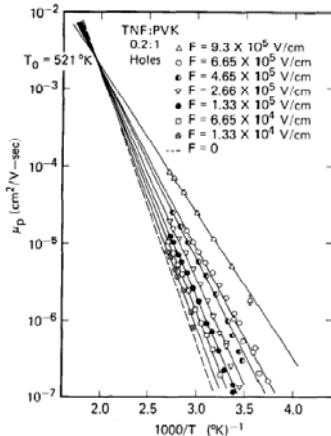
: PF constant  
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## Charge Transport in Disordered Organic Solids

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2009. 1<sup>st</sup> Semester

### (2) Gill's modified Poole-Frenkel model

$$\mu(F, T) = \mu_{PF} \exp\left(-\frac{\Delta E}{k_B T_{eff}}\right) \exp\left(\frac{\beta_{PF} \sqrt{E}}{k_B T_{eff}}\right) \frac{1}{T_{eff}} = \frac{1}{T} - \frac{1}{T_0} \quad T_0 : \text{Empirical parameter}$$



W. D. Gill, J. Appl. Phys. **43**, 5033 (1972).



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### (3) Bassler's Gaussian Disorder Model

- The energy of each site is distributed in accordance with the Gaussian distribution
- Energies of adjacent sites are uncorrelated and motion between sites is Markovian (no phase memory)
- The transition rates for phonon-assisted tunneling (Miller and Abrahams):

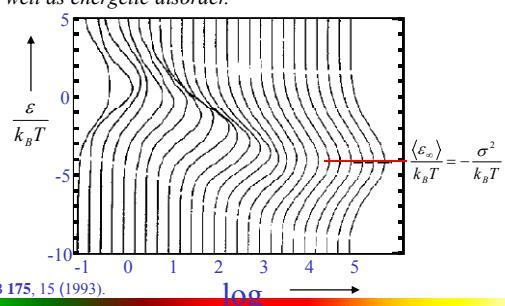
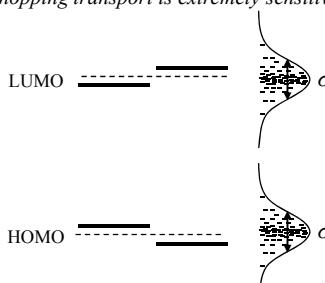
$$W_{ij} = v_{ph} \exp(-2\alpha R_{ij}) \begin{cases} \exp\left(-\frac{\epsilon_i - \epsilon_j}{kT}\right), & \epsilon_i > \epsilon_j \\ 1, & \epsilon_i > \epsilon_j \end{cases}$$

A. Miller, E. Abrahams, Phys. Rev. **120** (1960) 745.



$\alpha$  = inverse localization length,  $R_{ij}$  = distance between the localized states,  $\epsilon_i$  = energy at the state i.

- Since the hopping rates are strongly dependent on both the positions and the energies of the localized states, *hopping transport is extremely sensitive to structural as well as energetic disorder*.



H. Bassler, Phys. Status Solidi B **175**, 15 (1993).

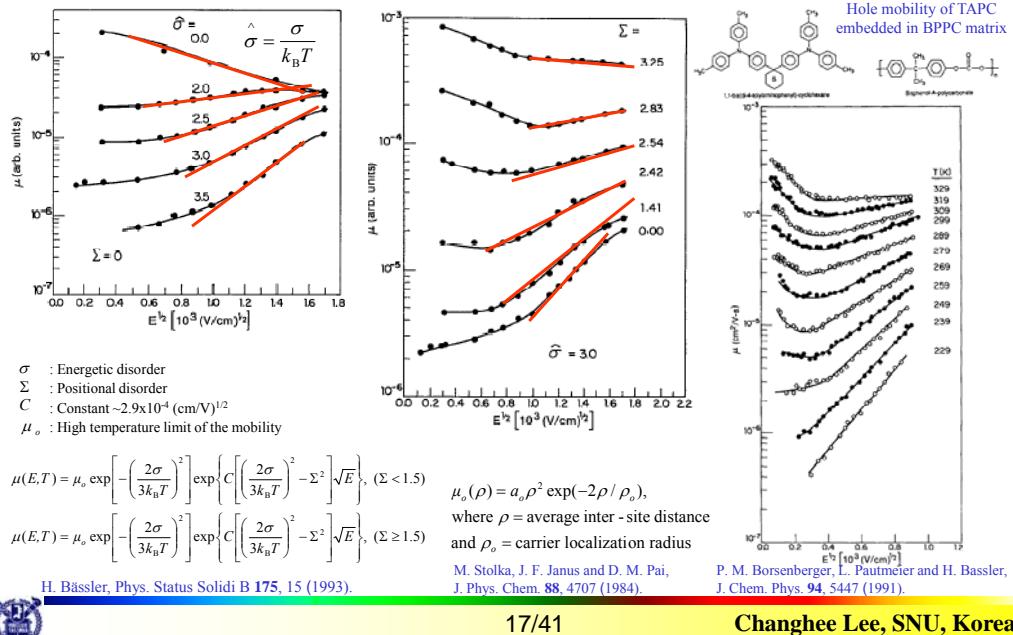


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## Charge Transport in Disordered Organic Solids

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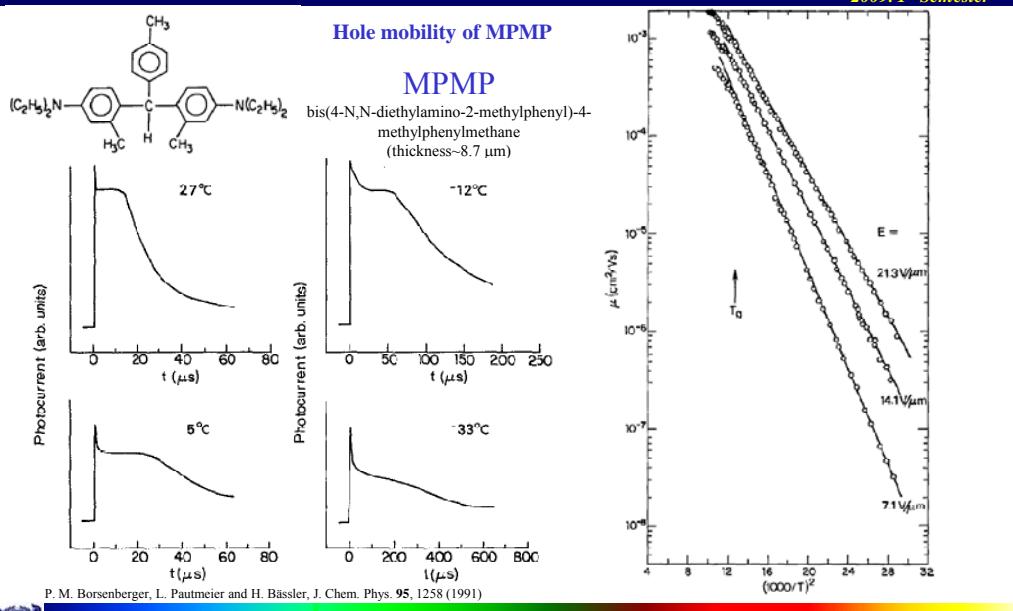


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## Charge Transport in Disordered Organic Solids

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# Disorder parameters

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## Disorder parameters

- $\sigma$ : The width of the DOS. Random distribution of both permanent and van der Waals dipoles lead to local fluctuations in electric potential → increase  $\sigma$  by an amount proportional to the square root of the dipole concentration and to the strength of the dipole moment. → reduce the carrier mobility. The smaller dipolar interaction is better for the carrier transport.
- $\Sigma$ : The degree of positional disorder. Amorphous morphology of molecular solids or doped polymers lead to the variation in the intermolecular distances.

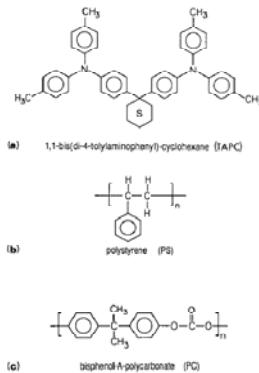


FIG. 1. Molecular structures of compounds used in this study.

P. M. Borsenberger and H. Bässler, J. Chem. Phys. 95, 5327 (1991).

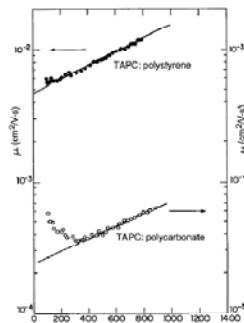


FIG. 2. Logarithm of the mobility vs  $E^{1/2}$  for TAPC doped polystyrene and TAPC doped polycarbonate measured at 295 K.

$\mu_{\text{TAPC doped polystyrene}} \gg \mu_{\text{TAPC doped polycarbonate}}$

## Dipole moment:

- TAPC [1,1-bis(di-4-tolylaminophenyl)cyclohexane] = 1.0 D
- PC (bisphenol-A-polycarbonate) = 1.0 D
- PS (polystyrene) = 0.1 D
- Larger dipolar interaction increases both  $\sigma$  and  $\Sigma$ .
- The elimination of random dipolar fields due to static dipole moments of PC reduces both  $\sigma$  and  $\Sigma$  and thereby increases the mobility.

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# Disorder parameters

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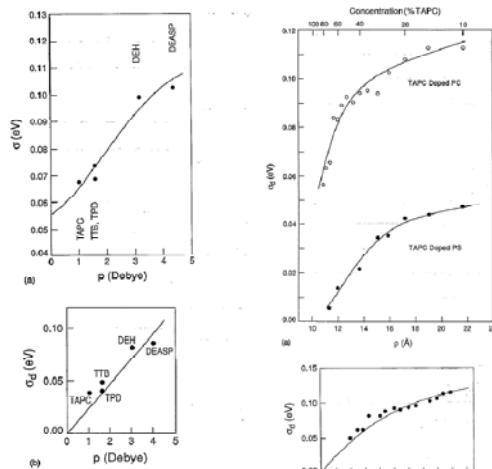


FIG. 3. (a) Experimental data for the total width of the DOS  $\sigma$  (eV) as a function of the dipole moment  $p$  (Debye) (from Ref. 9). The data points are DEH, DEAS, TAPC, TPD, DEH, and DEASP. (b) The dipolar contribution  $\sigma_d$  derived from the data of (a) assuming  $\sigma_{\text{dip}} = 0.055$  eV. The full line is a fit based upon Eq. (2) with  $a = 6 \text{ \AA}$  and  $\sigma = 3.5$ .

A. Dieckmann, H. Bässler and P. M. Borsenberger, J. Chem. Phys. 99, 8136 (1993).

the total Gaussian width of the DOS is

$$\sigma = (\sigma_d^2 + \sigma_{vdW}^2)^{1/2}.$$

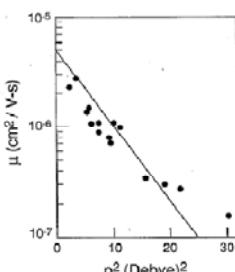


FIG. 4. Hole mobilities measured by Suguchi and Nishizawa (Ref. 8) at room temperature for a series of polar transport molecules dispersed in a poly(carbonate) host in a 1:1 ratio. The full line is the prediction of the current calculation for  $a = 6 \text{ \AA}$  and  $\epsilon = 3.5$ .

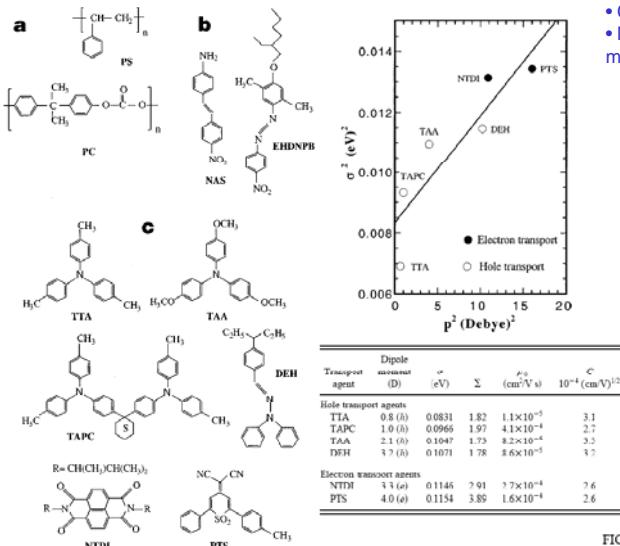
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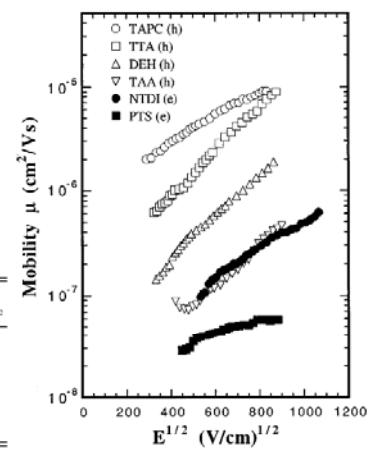
## Effect of dipolar molecules on carrier mobilities

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A. Goonesekera and S. Ducharme, J. Appl. Phys. 85, 6506 (1999)

- Conduction through the dopant.
- Mobility is strongly affected by the dipole moment of the dopant for holes and electrons



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## Temperature dependence of the hole mobility of PPVs

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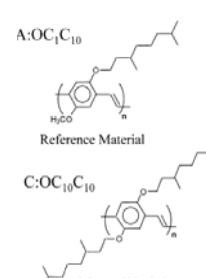
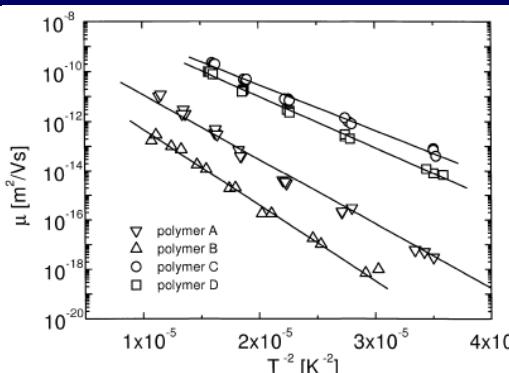


Table 1  
Parameters  $\mu_\infty$ ,  $\sigma$  (energetic disorder bandwidth) and  $a$  (site-spacing) describing the temperature dependence of the zero-field mobility  $\mu_0$  and field activation factor  $\gamma$  for the different polymers studied

Sample	$\mu_\infty$ ( $\text{m}^2/\text{V s}$ )	$\sigma$ (meV)	$a$ (nm)
A	$5.1 \times 10^{-9}$	112	1.2
B	$4.0 \times 10^{-10}$	121	1.7
C	$1.6 \times 10^{-7}$	93	1.1
D	$1.5 \times 10^{-7}$	99	1.2

P.W.M. Blom, M.C.J.M. Vissenberg, Materials Science and Engineering 27, 53-94 (2000)



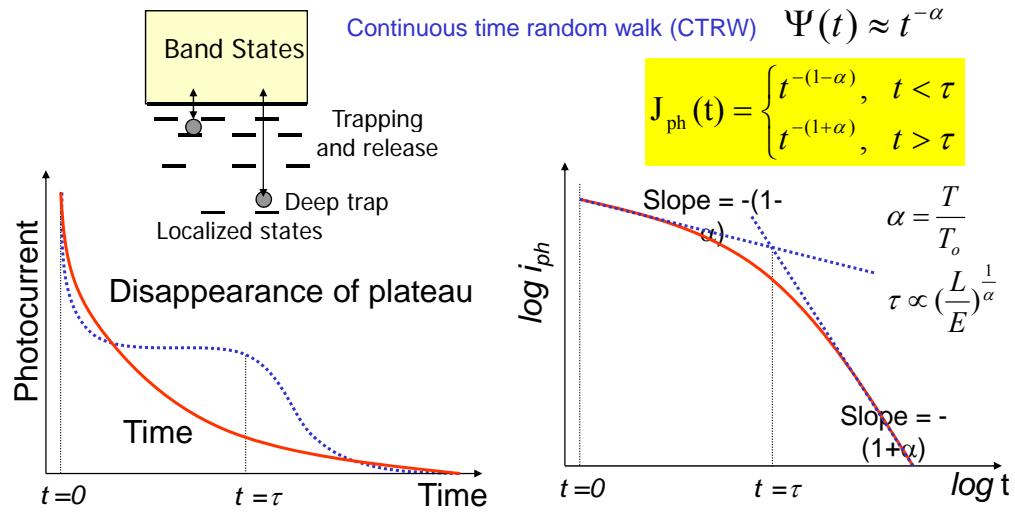
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### (4) Scher-Montroll dispersive transport model



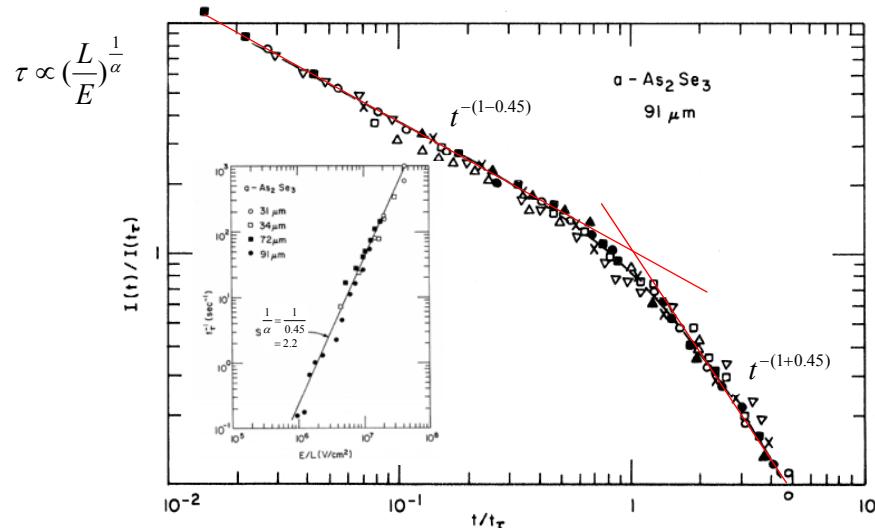
Scher and Montroll, Phys. Rev. B 12, 2455 (1975)

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## Scher-Montroll model of dispersive transport

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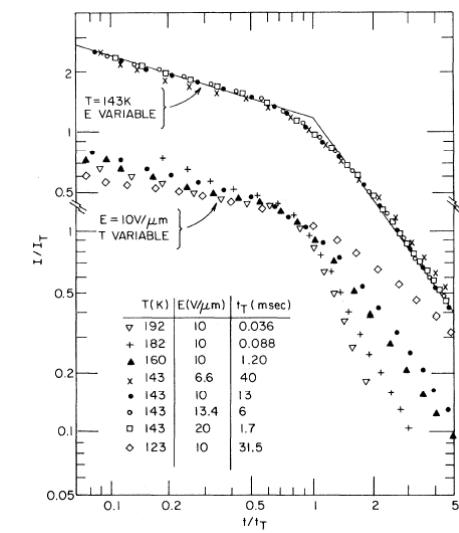
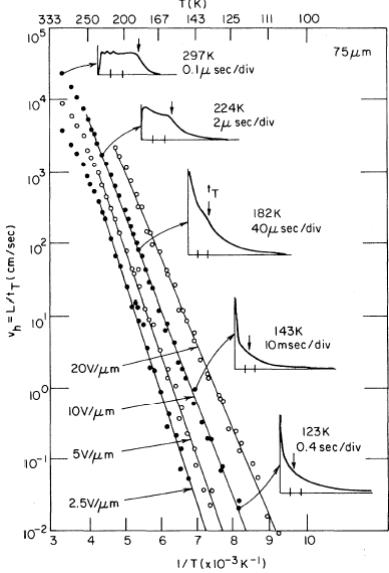
H. Scher and E. W. Montroll, Phys. Rev. B 12, 2455 (1975)

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## Dispersive Transport in $\alpha$ -Selenium

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G. Pfister, Phys. Rev. Lett. 36, 271 (1976)



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## Space-charge-limited current in organic semiconductors

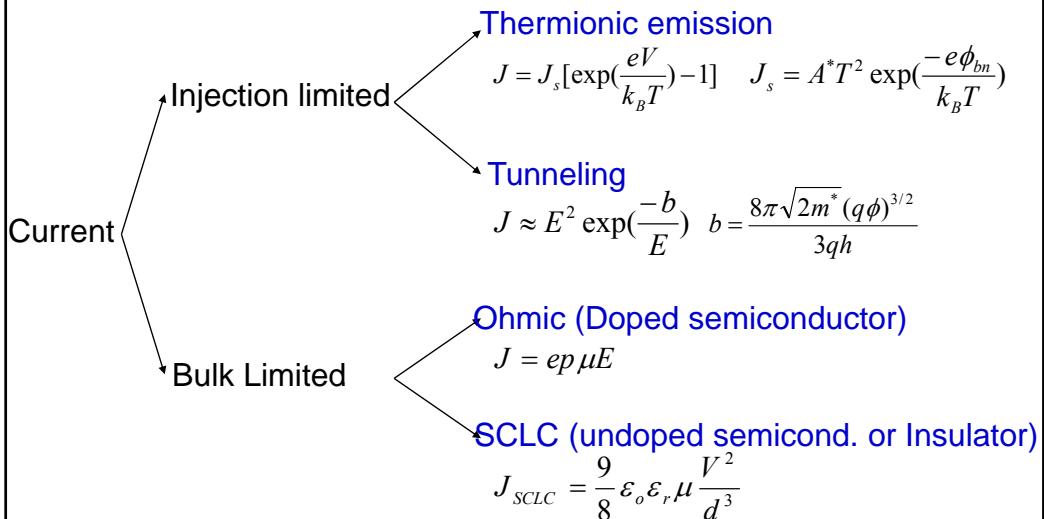


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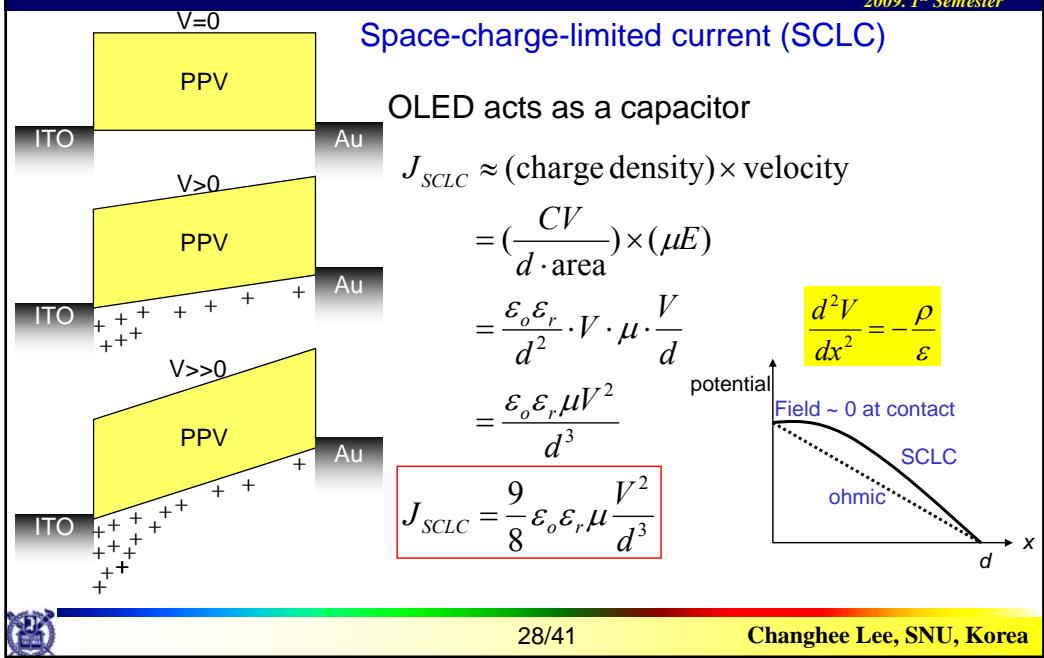
## Charge –voltage characteristics of organic semiconductors

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## Space-charge-limited current (SCLC)

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## Ohmic Contact (1)

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Poisson equation :  $\frac{dF}{dx} = \frac{qn}{\epsilon}$ .

Current flow equation :  $J = qn\mu F - qD \frac{dn}{dx} = 0$ ,

since there is no external applied field.

$$\therefore \frac{dn}{n} = \frac{\mu}{D} F dx = \frac{q}{k_B T} F dx,$$

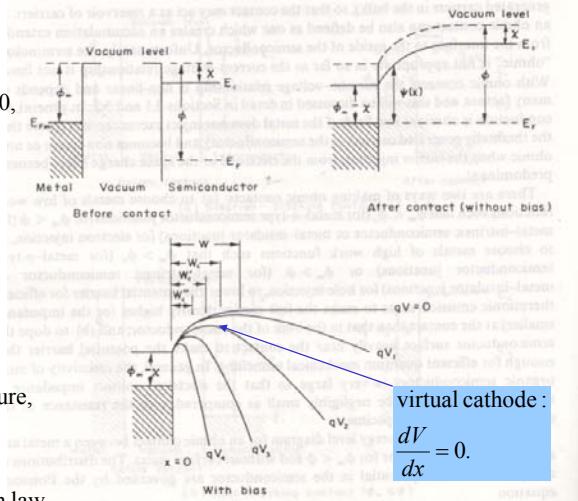
where the Einstein relation  $\frac{D}{\mu} = \frac{k_B T}{q}$  is used.

$$\therefore \log \frac{n}{n_s} = \frac{q}{k_B T} \int_0^x F dx.$$

From the boundary condition in the right figure,

$$\psi(x) - (\varphi_m - \chi) = -q \int_0^x F dx.$$

$$\therefore n = n_s \exp\left[-\frac{\psi(x) - (\varphi_m - \chi)}{k_B T}\right], \text{ i.e., Boltzmann law.}$$



virtual cathode :  
$$\frac{dV}{dx} = 0.$$



## Ohmic Contact (2)

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$$\frac{d^2\psi}{dx^2} = -q \frac{dF}{dx} = -\frac{q^2 n}{\epsilon} = -\frac{q^2 n_s}{\epsilon} \exp\left[-\frac{\psi(x) - (\varphi_m - \chi)}{k_B T}\right].$$

Integrating both sides after multiplying  $2(\frac{d\psi}{dx})$

and using the boundary condition  $\frac{d\psi}{dx} = 0$  when  $\psi = \varphi - \chi$ ,

$$(\frac{d\psi}{dx})^2 = \frac{2q^2 n_s k_B T}{\epsilon} \left\{ \exp\left[-\frac{\psi(x) - (\varphi_m - \chi)}{k_B T}\right] - \exp\left[-\frac{(\varphi - \varphi_m)}{k_B T}\right] \right\}.$$

Integrating this equation gives

the width of the accumulation region

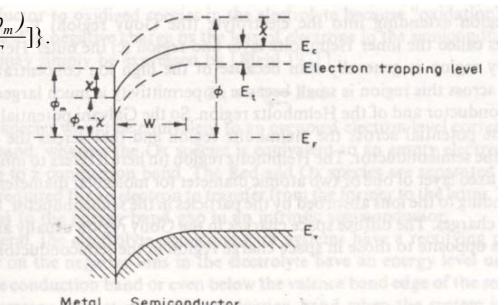
$$W = \left( \frac{2\epsilon k_B T}{q^2 N_c} \right)^{1/2} \exp\left(\frac{\varphi - \chi}{2k_B T}\right) \left[ \frac{\pi}{2} - \sin^{-1}\left\{ \exp\left(-\frac{\varphi - \varphi_m}{2k_B T}\right) \right\} \right].$$

If  $\varphi - \varphi_m > 4k_B T$ ,

$$W \approx \frac{\pi}{2} \left( \frac{2\epsilon k_B T}{q^2 N_c} \right)^{1/2} \exp\left(\frac{\varphi - \chi}{2k_B T}\right).$$

For semiconductors containing shallow traps confined in a discrete energy level  $E_t$ ,

$$W \approx \frac{\pi}{2} \left( \frac{2\epsilon k_B T}{q^2 N_t} \right)^{1/2} \exp\left(\frac{\varphi - \chi - E_t}{2k_B T}\right).$$



## Space-charge-limited current

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Distribution function of trap density

$$h(E, x) = N_t(E)S(x).$$

$$\text{Poisson equation : } \frac{dF(x)}{dx} = \frac{\rho}{\epsilon} = \frac{q[p(x) + p_t(x)]}{\epsilon}.$$

Current flow equation :  $J = q\mu_p p(x)F(x)$ .

$$p_t(x) = \int_{E_l}^{E_u} h(E, x) f_p(E) dE, \quad p(x) = N_v \exp \left[ -\frac{E_F}{k_B T} \right] J.$$

For  $p_t(x) = 0$ ,

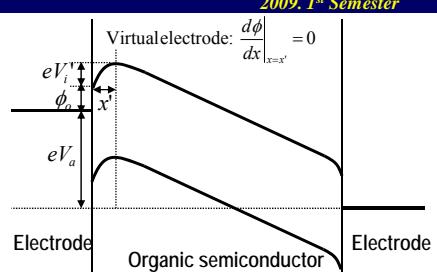
$$2F(x) \frac{dF(x)}{dx} = \frac{d[F(x)]^2}{dx} = \frac{2J}{\epsilon\mu_p}. \quad \therefore [F(x)]^2 - [F(x=0)]^2 = \frac{2J}{\epsilon\mu_p} x.$$

$$\therefore F(x) = \sqrt{\frac{2J}{\epsilon\mu_p} x}.$$

The distance  $W_a$  between the actual electrode surface and the virtual anode ( $-dV/dx=F=0$ ) is so small that we can assume  $F(x=W_a \rightarrow 0)=0$  for simplicity.

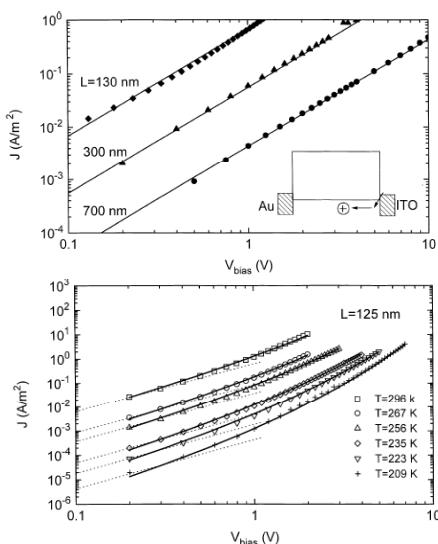
Boundary condition,  $V = \int_0^d F(x) dx$

$$J = \frac{9}{8} \epsilon \mu_p \frac{V^2}{d^3}. \quad \text{Mott - Gurney law. (no trap)}$$



## Space-charge-limited current

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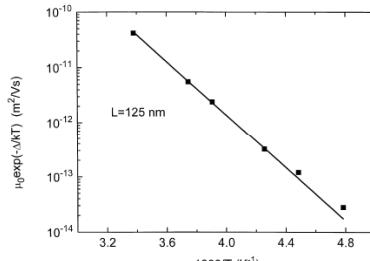


ITO/PPV/Au hole-only devices

$$J_{SCLC} = \frac{9}{8} \epsilon_o \epsilon_r \mu \frac{V^2}{d^3}$$

Fit using  $\mu = 0.5 \times 10^{-6} \text{ cm}^2/\text{Vs}$  &  $\epsilon_r = 3$

$$\mu(F, T) = \mu_0 \exp \left( -\frac{\Delta E - \beta_{PF} \sqrt{F}}{k_B T_{eff}} \right)$$



P.W.M. Blom, M.C.J.M. Vissenberg, Materials Science and Engineering 27, 53-94 (2000)



## Space-charge-limited current

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At low field, ohm's law holds if the density of thermally generated free carriers  $p_o$  inside the specimen is predominant such that

$$qp_o\mu_p \frac{V}{d} \gg \frac{9}{8}\epsilon\mu_p \frac{V^2}{d^3}.$$

The onset of the departure from Ohm's law or the onset of the SCL conduction takes place when  $V_\Omega = \frac{8}{9} \frac{qp_o d^2}{\epsilon}$ .

By rearranging this equation we have

$$\frac{d^2}{\mu_p V_\Omega} = \frac{9}{8} \frac{\epsilon}{qp_o \mu_p} = \frac{9}{8} \frac{\epsilon}{\sigma_o} \text{ or } \tau_t \approx \tau_d.$$

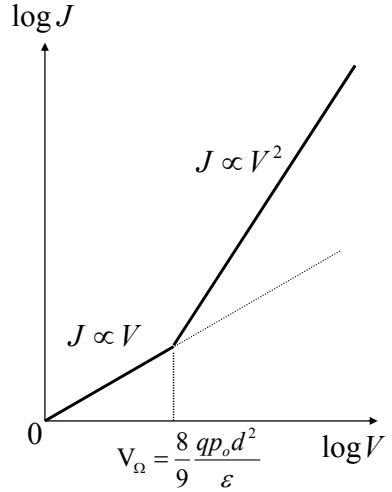
Therefore, the transition from the ohmic to the SCL regime

takes place when the carrier transit time  $\tau_t = \frac{d^2}{\mu_p V_\Omega}$  at  $V_\Omega$

is approximately equal to the dielectric relaxation time  $\tau_d = \frac{\epsilon}{\sigma_o}$ .

At  $\tau_t > \tau_d$ , ohmic conduction is predominant.

At  $\tau_t < \tau_d$ , SCLC conduction is predominant.



K. C. Kao and W. Hwang, Electrical Transport in Solids, (Pergamon, New York, 1981), p.159



## Space-charge-limited current

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dielectric relaxation time  $\tau_d = \frac{\epsilon}{\sigma_o}$ .

Continuity equation  $q \frac{\partial(p + p_o)}{\partial t} = -\nabla \cdot J$ ,

where current is given by  $J = q(p + p_o)\mu_p F - qD_p \nabla(p + p_o)$ .

$$\therefore \frac{\partial p}{\partial t} = -\left(\frac{qp\mu_p}{\epsilon}\right)p + D_p \nabla^2 p.$$

If  $p < p_o$  and p spreads uniformly over the specimen in a time

comparable to the dielectric relaxation time  $\tau_d = \frac{\epsilon}{\sigma_o}$ , the 2nd term can be ignored.

$$\therefore \frac{\partial p}{\partial t} \approx -\left(\frac{qp_o\mu_p}{\epsilon}\right)p. \therefore p(t) = p(t=0) \exp(-t/\tau_d).$$

$\tau_d$  is a measure of the time required for the carrier to re-establish equilibrium.



## Space-charge-limited current

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Traps confined in single or multiple discrete energy levels

$$h(E, x) = N_t \delta(E - E_t) S(x)$$

$$\text{Trapped charge density } p_t(x) \approx \frac{N_t S(x)}{1 + N_t \theta_t / p(x)},$$

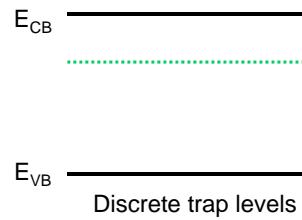
$$\text{where } \theta_t = \frac{g_p N_v}{N_t} e^{-\frac{E_t}{kT}}.$$

$$J_{SCLC} = \frac{9}{8} \varepsilon_0 \varepsilon_r \mu \theta_t \frac{V^2}{d_{eff}^3}.$$

Ignoring non-uniform spatial distribution of traps

$$\theta_t = \frac{p}{p + p_t}, \quad d_{eff} = d$$

$$J_{SCLC} = \frac{9}{8} \varepsilon_0 \varepsilon_r \mu \theta_t \frac{V^2}{d^3}.$$



## Space-charge-limited current

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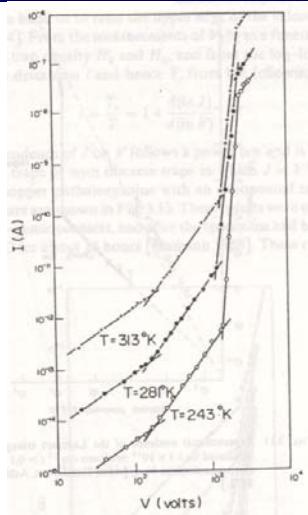


FIG. 3.10. The current-voltage characteristics of naphthalene single crystals as functions of temperature. [After Campos 1972.]

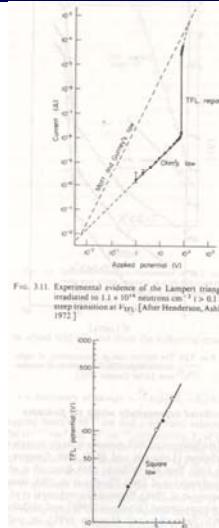


FIG. 3.11. Experimental evidence of the Lampert triangle in silicon irradiated to  $1.1 \times 10^{14}$  neutrons  $\text{cm}^{-2}$  ( $> 0.1$  MeV) with a temperature at  $T_{TFL} = 100^\circ\text{C}$ . [After Henderson, Ashley, and Shen 1972.]

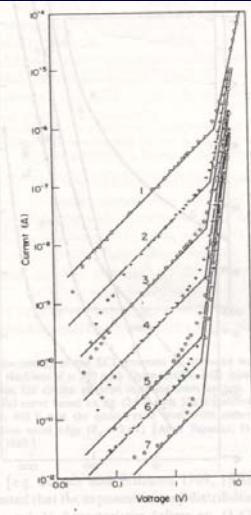


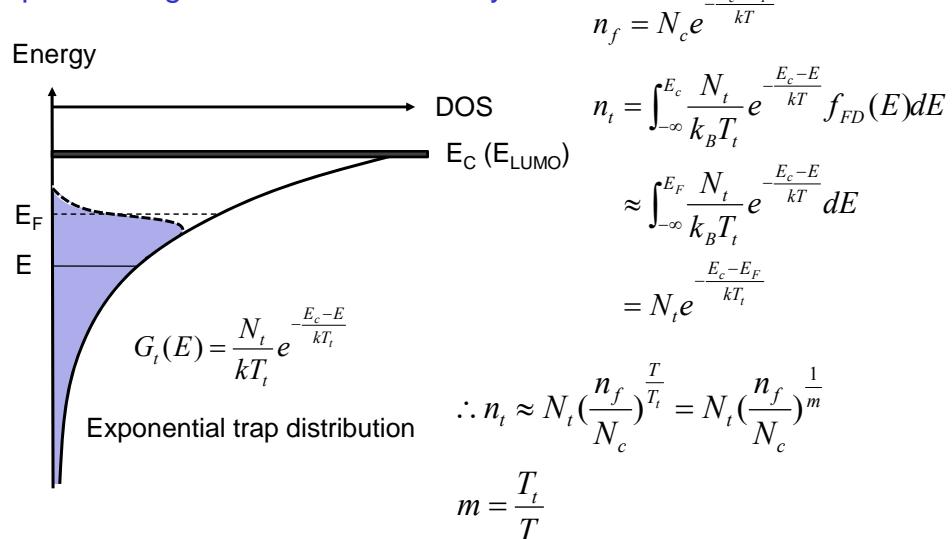
FIG. 3.12. The TFL threshold voltage (V<sub>TFL</sub>) for neutron-irradiated silicon as a function of specimen thickness. [After Henderson, Ashley, and Shen 1972.]



## Space-charge-limited current

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### Space-charge-limited current: analytic solution



## Space-charge-limited current

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Poisson equation :  $\frac{dF(x)}{dx} = \frac{q[n_f(x) + n_t(x)]}{\epsilon_0 \epsilon} \approx \frac{qn_t(x)}{\epsilon_0 \epsilon} \approx \frac{qN_t}{\epsilon_0 \epsilon} \left(\frac{n_f}{N_c}\right)^{1/m}.$

Current flow equation :  $J = q\mu n_f(x)F(x).$

$$F^{1/m} \frac{dF(x)}{dx} = \frac{N_t}{\epsilon_0 \epsilon} \left(\frac{J}{q\mu N_c}\right)^{1/m}.$$

$$\therefore \frac{m}{m+1} F^{\frac{m+1}{m}} = \frac{N_t}{\epsilon_0 \epsilon} \left(\frac{J}{q\mu N_c}\right)^{1/m} x.$$

$$\therefore F(x) = \left(\frac{m+1}{m} \frac{N_t}{\epsilon_0 \epsilon}\right)^{\frac{m}{m+1}} \left(\frac{J}{q\mu N_c}\right)^{\frac{1}{m+1}}.$$

Boundary condition,  $V = \int_0^d F(x) dx.$

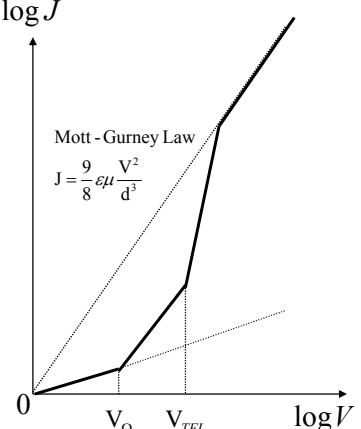
$$J = q^{1-m} \mu N_c \left(\frac{2m+1}{m+1}\right)^{m+1} \left(\frac{m}{m+1} \frac{\epsilon_0 \epsilon}{N_t}\right)^m \frac{V^{m+1}}{d^{2m+1}}, \quad m = \frac{T_t}{T}.$$

Ohmic  $\Rightarrow$  Trapped SCLC

$$V_\Omega = \frac{qd^2 N_t}{\epsilon_0 \epsilon_r} \left(\frac{p_o}{N_v}\right)^{\frac{1}{m}} \left(\frac{m+1}{m}\right) \left(\frac{m+1}{2m+1}\right)^{\frac{m+1}{m}}.$$

Trapped SCLC  $\Rightarrow$  Trap-free SCLC

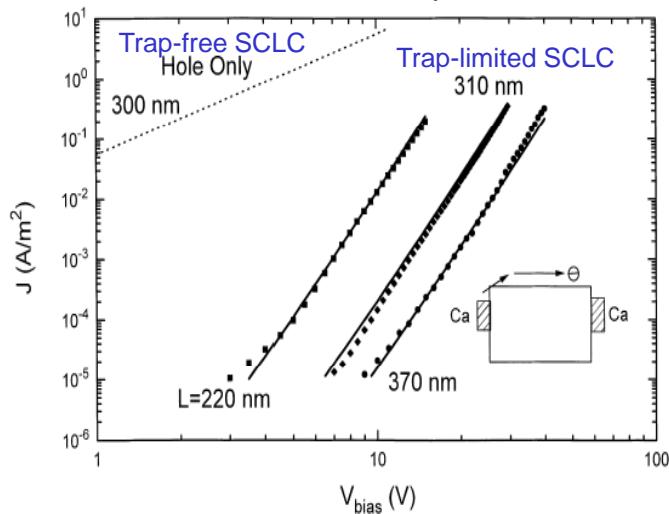
$$V_{TFL} = \frac{qd^2}{\epsilon_0 \epsilon_r} \left[\frac{9}{8} \frac{N_t^m}{N_v} \left(\frac{m+1}{m}\right)^m \left(\frac{m+1}{2m+1}\right)^{\frac{1}{m-1}}\right].$$



## Space-charge-limited current

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Ca/PPV/Ca electron-only devices



P.W.M. Blom, M.C.J.M. Vissenberg, Materials Science and Engineering 27, 53-94 (2000)

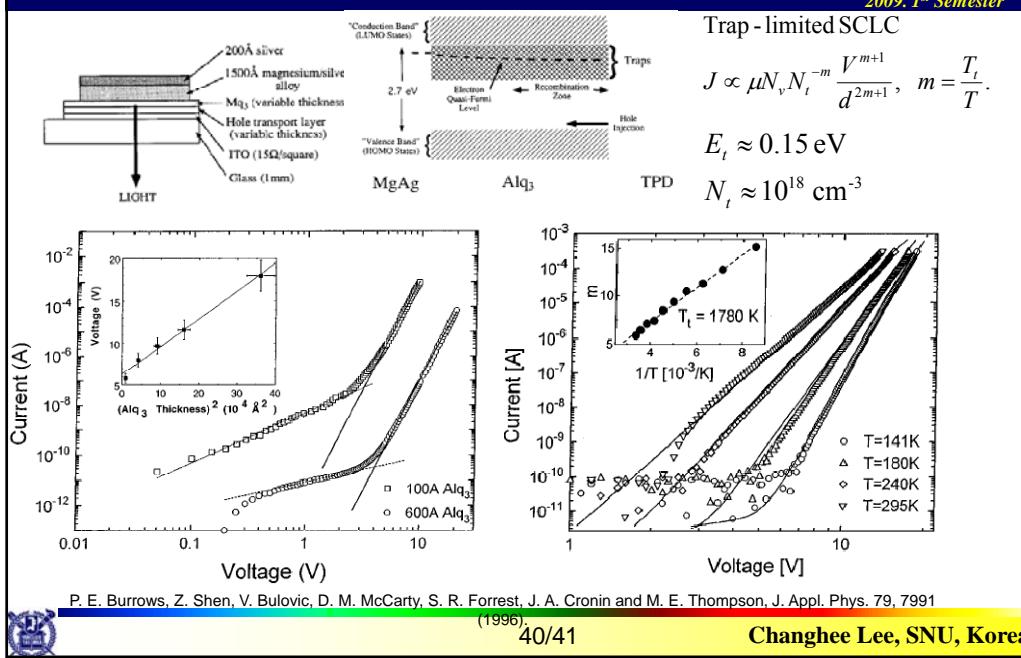


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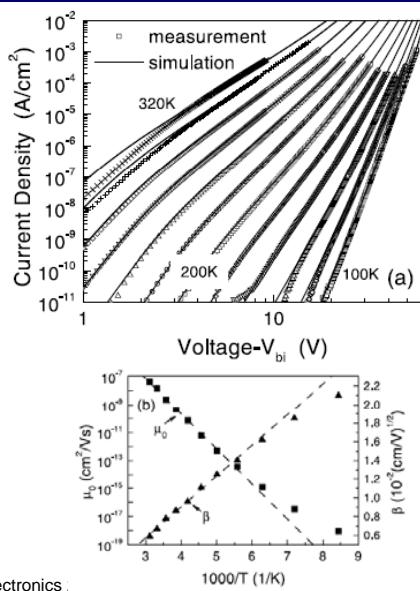
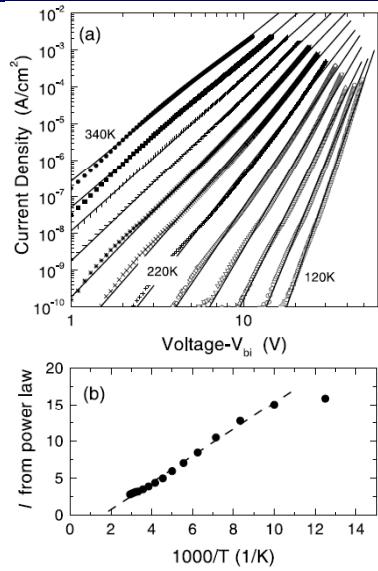
## SCL current with an exponential trap distribution

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# Trap-limited SCL current with field-dependent $\mu$

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