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Temperature Dependence of the Drude-Lorentz Parameters of (TSeF)(TCNQ). W. BLUDAU, P. M. GRANT and P. E. SEIDEN, IBM Research Laboratory, San Jose, CA and Yorktown, NY--We report measurements and analysis of the polarized near-normal incidence reflectivity of (TSeF)(TCNQ) in the range 0.5 - 4.5 eV as a function of temperature between 300K and 4.2K. By using Drude-Lorentz analyses over the entire observed optical spectrum, we have obtained the temperature dependence of the plasma energy and lifetime of the Drude edge. Using a Mathiessen's rule type argument[1], we show that the temperature dependent part of the optical resistivity follows a power law similar to that of the dc resistivity. We discuss this temperature dependence in relation to electron-electron and electron-phonon scattering models.

[1] P. E. Seiden and D. Cabib, Phys. Rev. 13B, 1846 (1976).

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or the American Physical Society
erican Institute of Physics

A. LEE, T. M. RICE, AND F. WUDL, Bell Laboratories — X-ray scattering studies of tetrathiafalvalenium-thiocyanate [TTF(SCN)$_{.588}$] show a structural transition at $T_S \approx 350$ K in which the high-temperature tetragonal structure develops a monoclinic strain as the 1D stacks of TTF molecules slip along the c-axis. Furthermore at $T_o \approx 170$ K the development of charge-density waves (CDW) on the TTF stacks produces new superlattice Bragg peaks at reduced wave vector $\overline{q} = \overline{q}_\perp + 0.294\,\overline{c}^*$ where $\overline{q}_\perp$ is incommensurate and temperature dependent. Below T_S, the relative slip of adjacent TTF stacks (which is simply related to the deviation of the monoclinic angle from 90°) increases slowly until T_o, whereupon a rapid increase occurs. We consider a model in which the CDW and lattice distortion on neighboring TTF stacks are coupled through elastic forces in addition to the Coulomb interaction. The competition between these two forces can explain the temperature-dependent strain in the TTF lattice and the incommensurate transverse periodicity of the CDW.

HI 11 Optical Properties of the Semiconducting 'Metal-like' Compound $(NMe_3H)(I)(TCNQ)$. J.E. DEIS, D.B. TANNER, Ohio State University and A.J. EPSTEIN, J.S. MILLER, Xerox Webster Research Center--Room temperature polarized reflectance measurements of this 1-D organic conductor have been made for three mutually perpendicular directions over the frequency range of 50-28,000 cm^{-1} (0.006-3.5 eV). For two directions the reflectance is constant and low with the only structure being that resulting from intramolecular vibrations. For the third direction ($\underline{E}$ parallel stacking axis) the reflectance at low frequencies is nearly 50%, with structure associated with electron-molecular vibration coupling in the middle infrared, and a plasmon minimum at 0.46 eV. There is a strong reflection peak in the green. Kramers-Kronig analysis yields $\sigma_1(\omega)$ and $\varepsilon_1(\omega)$ for all three directions For the two perpendicular directions σ_1 is low for $\omega<1$ eV and rises for $\omega>1$ eV, $\varepsilon_1 \simeq 2$ and independent of ω. For $\underline{E}$ parallel stacking axis σ has a maximum in the ir and decreases at lower frequencies. ε extrapolates to $\sim$10 at low frequencies. The data are analyzed in terms of a semiconducting model with an energy gap of 1600 cm^{-1} (0.2 eV) consistent with the results of transport studies

HI 12 Pressure Dependence of Transport Properties in $(SNBr_{0.4})_x$ Crystals. W. D. GILL, J. F. KWAK, R. L. GREEN K. SEEGER* and G. B. STREET, IBM Research Laboratory, San Jose, CA 95193-- The pressure dependence of normal conductivity of $(SNBr_{0.4})_x$ crystals has been measured at 300K both // and $\perp$ to the b-axis for hydrostatic pressure up to 17 kbar. $\sigma_{//}$ increases by $\sim$1.3%/kbar in sharp contrast to the $\sim$40%/kbar increase observed in $(SN)_x$ crystals. The observed pressure dependence can be reasonably accounted for by lattice stiffening increasing the electron-phonon scattering lifetime implying that electron-electron scattering has been suppressed by bromine treatment of $(SN)_x$. Preliminary results indicate a monotonic decrease with pressure of the superconducting transition temperature T_c, an effect opposite to the pressure induced increase of T_c observed in $(SN)_x$.

*Permanent address: University of Vienna and the Ludwig Boltzmann Institut für Festkörperphysik, Vienna, Austria.

Supplementary Program

HI 13 Transport Properties of the Semiconducting 'Meta like' Compound $(NMe_3H)(I)(TCNQ)$. P.M. CHAIKIN, U.C.L.A., A.J. EPSTEIN and J.S. MILLER, Xerox Webster Research Cente We have measured the temperature, T, dependence of thermo electric power, S, and the conductivity of this anisotropi system along the highly conducting direction (b-axis) and perpendicular to it (a-axis). The results are consistent with a semiconductor-semiconductor transition along the b-axis at T = 150K. The thermopower along the a-axis is of opposite sign to that of the b-axis at room temperatur and is weakly temperature dependent even through the transition while dS/dT along the b-axis changes sign at the transition.

HI 14 Thermoelectric Properties as a Function of Band Filling: $(NMP)_x(Phen)_{1-x}(TCNQ)$, $0.5 \leq x \leq 1.0$. A.J. EPSTEIN and J.S. MILLER, Xerox Webster Research Center,, and P.M. CHAIKIN, U.C.L.A.--Substitution of neutral phenazine for the NMP cation which is of similar size, shape and polarizability, allows study of the properties of conducting TCNQ chains over a range from ~ 0.5 filled band (x = 1.0) to ~ 0.25 filled band (x = 0.5). We have measured the temperature dependence of the thermoelectric power, S, along the conductivity axis of single crystal samples of this continuous series. The resulting behavior varies continuously from that of (NMP)(TCNQ) (x = 1, S(295K) = 32 μV/°K) to that of $Qn\,(TCNQ)_2$ (x = 0.5, S(295K) = -65 μV/°K). The data show that (NMP)(TCNQ) and $Qn(TCNQ)_2$ are similar systems with different band fillings. The results are analyzed in terms of a model of strong on-site Coulomb interactions.

HI 15 Transport and Magnetic Properties of DTTTF-TCNQ (Dihydrothienotetrathiafulvalene tetracyanoquinodimethane),* Y. TOMKIEWICZ and E. M. ENGLER, IBM Research Center, Yorktown Heights, NY 10598 and J. ANDERSEN, Riso National Laboratory, Denmark.--Measurements on a new organic conductor α-DTTTF-TCNQ indicate that the transport and magnetic properties are decoupled: while the conductivity data show the existence of a metal-insulator transition at 110 K, the spin susceptibility does not show any transition. Such behavior has been previously explained either by the effect of disorder on a single stack in a system with complete charge-transfer or by incomplete charge transfer between magnetically different donor and acceptor stacks.

In the case of DTTTF-TCNQ, the detailed temperature dependence of the susceptibility over the temperature range 4<T<300 K combined with the relative temperature independence of the *g*-value precludes the possibility of incomplete charge-transfer being the source of the observed behavior. Disorder on the other hand can explain not only the magnetic behavior but also the unusually low ratio of the activation energy for conductivity to the transition temperature. *Submitted by B. D. SILVERMAN.

HI 16 Temperature Dependence of the Drude-Lorentz Parameters of (TSeF)(TCNQ). W. BLUDAU, P. M. GRANT and P. E. SEIDEN, IBM Research Laboratory, San Jose, CA and Yorktown, NY--We report measurements and analysis of the polarized near-normal incidence reflectivity of (TSeF) (TCNQ) in the range 0.5 - 4.5 eV as a function of temperature between 300K and 4.2K. By using Drude-Lorentz analyses over the entire observed optical spectrum, we have obtained the temperature dependence of the plasma energy and lifetime of the Drude edge. Using a Mathiessen's rule type argument[1], we show that the temperature dependent part of the optical resistivity follows a power law similar to that of the dc resistivity. We discuss this temperature dependence in relation to electron-electron and electron-phonon scattering models.

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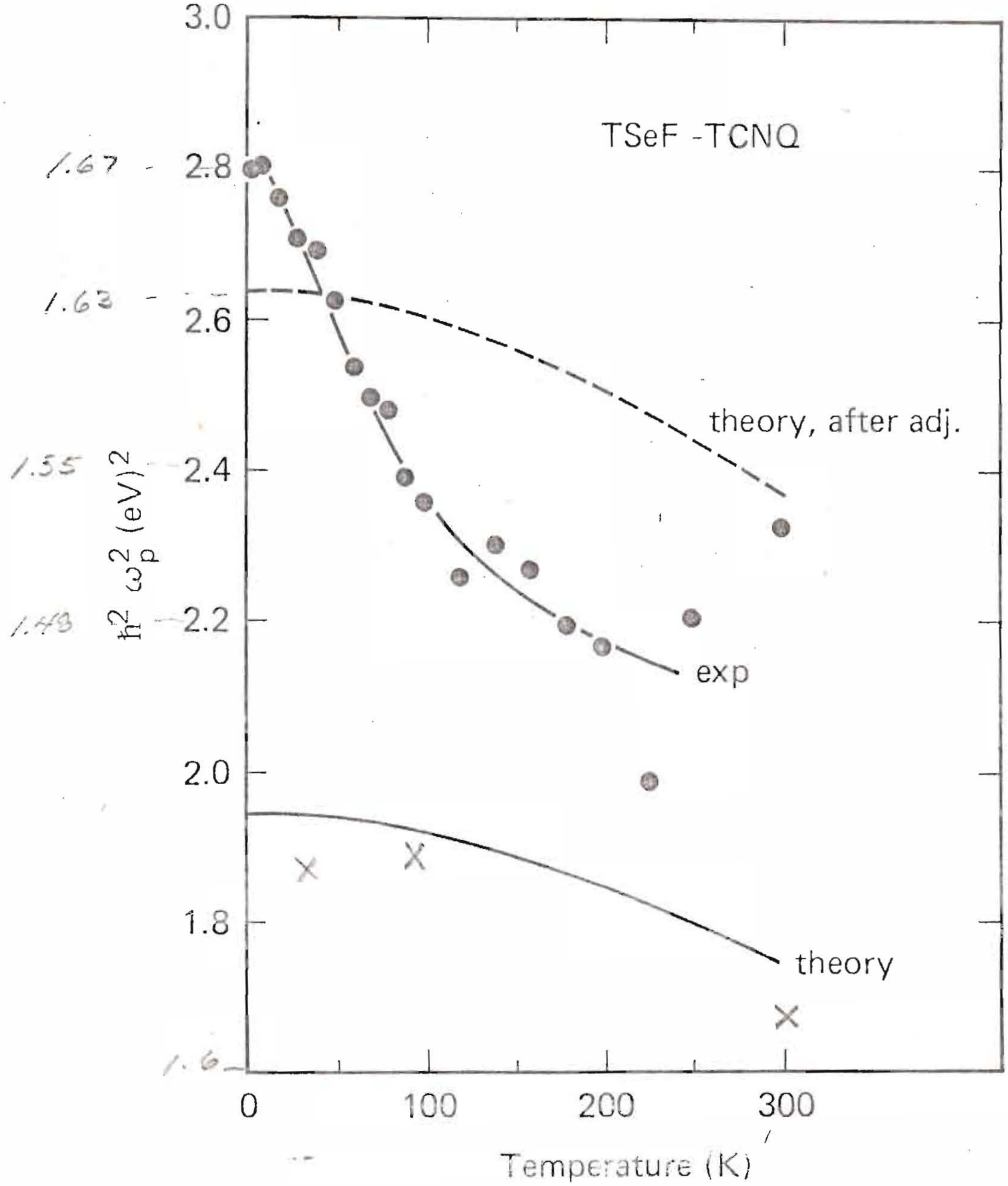
SESSION HJ: SUPERCONDUCTIVITY IN THIN FILMS, AMORPHOUS ALLOYS AND $(SN)_x$

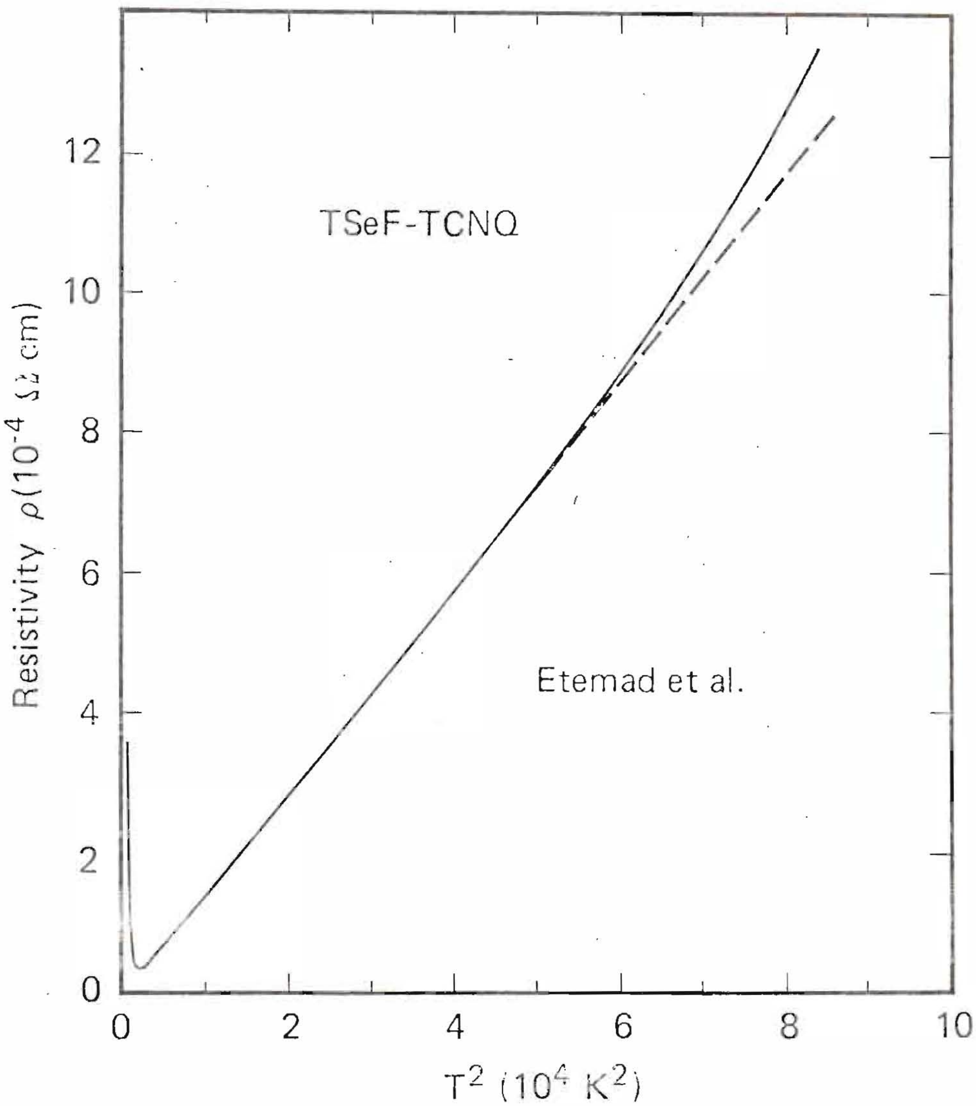
Wednesday afternoon, 29 March 1978
Arlington Room, Sheraton-Park at 2:00 P.M.
P. Chaudhari, presiding

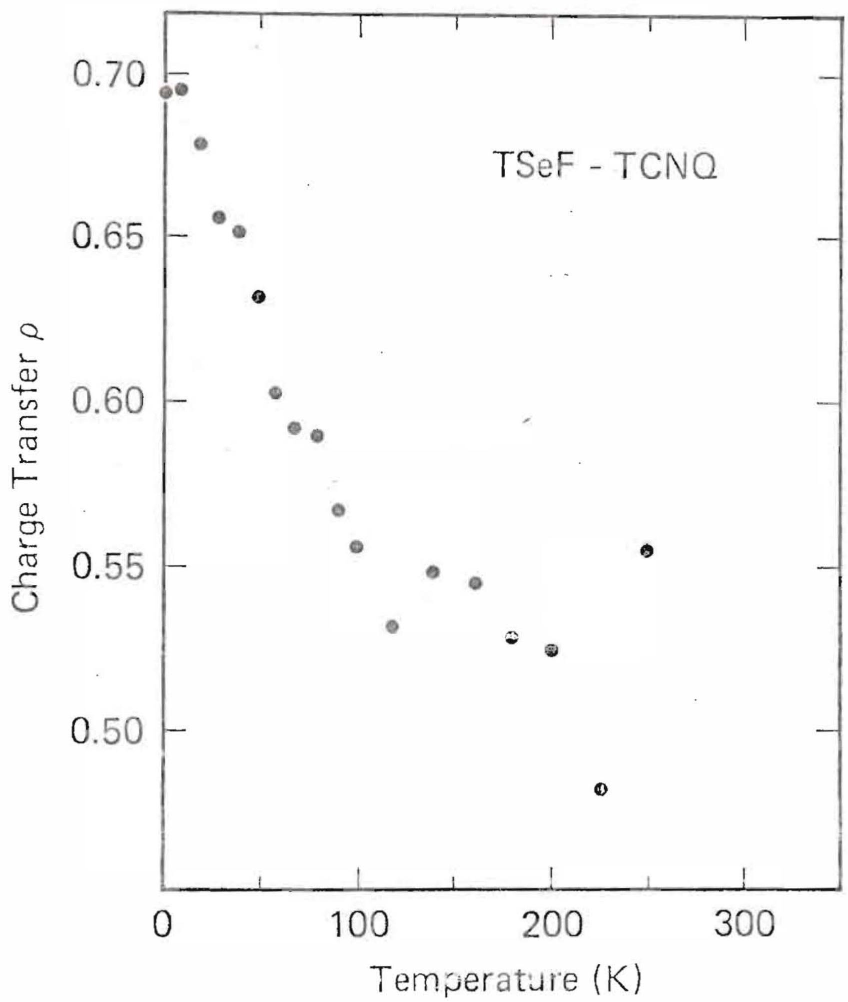
HJ 1 Normal State Resistance of the Superconducting Ternary Molybdenum Sulfides*. D. CHRISTOPHER MARTIN+, JOHN A. WOOLLAM, and SAMUEL A. ALTEROVITZ, NASA Lewis Res. Ctr., Cleveland, OH.--The resistance R(T) varies as T^2 over the range of temperature T from T_c to nearly 40K, in sintered, sputtered, and evaporated thin film $Cu_xMo_6S_8$, and in sputtered $PbMo_6S_8$ films. In addition, an inflection in R(T) is found in the range 50K to 70K, as well as a tendency towards saturation at high temperature. These results are strikingly similar to results found for A-15 superconductors. In sintered $PbMo_6S_8$ the resistance varies as T. At high temperature, for $Cu_xMo_6S_8$, abrupt changes in the slope of R(T) are found. These occur at temper-

$$\frac{1.67 - 1.48}{1.48} = 13\%$$

$$\frac{1.63 - 1.55}{1.55} = 5\%$$




TSeF-TCNQ
Etemad et al.
Resistivity ρ(10⁻⁴ Ω cm)
T² (10⁴ K²)
0
2
4
6
8
10
12



TSeF - TCNQ
Charge Transfer ρ
0.70
0.65
0.60
0.55
0.50
0
100
200
300
Temperature (K)

TEMPERATURE DEPENDENCE OF ω_p

ω_p can be calculated for a 2-band tight-binding model:

$$\hbar^2 \omega_p^2 = \frac{16\, e^2\, b^2}{V/n} \left(t_A + t_D \right) * \sin \rho \frac{\pi}{2}$$

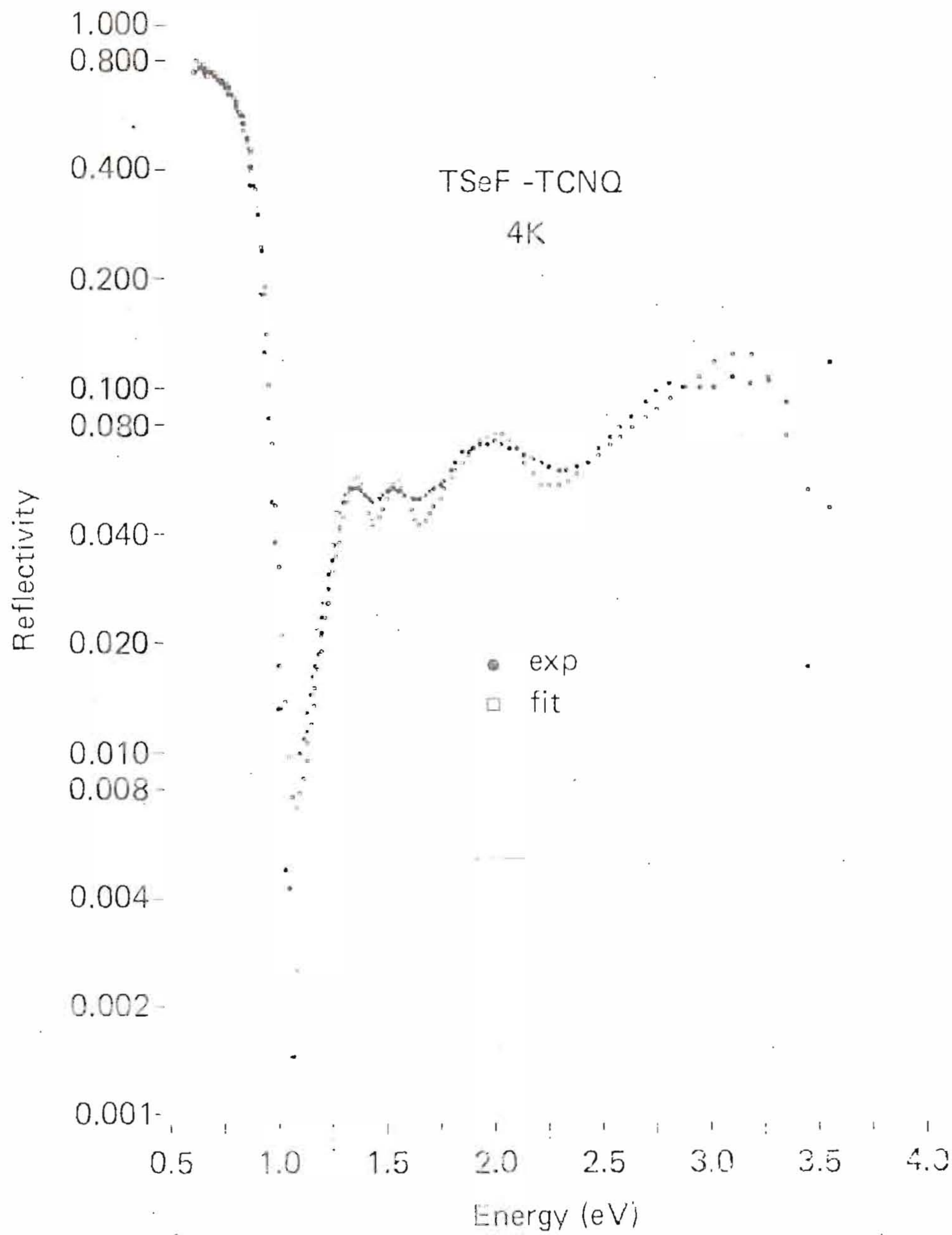
with

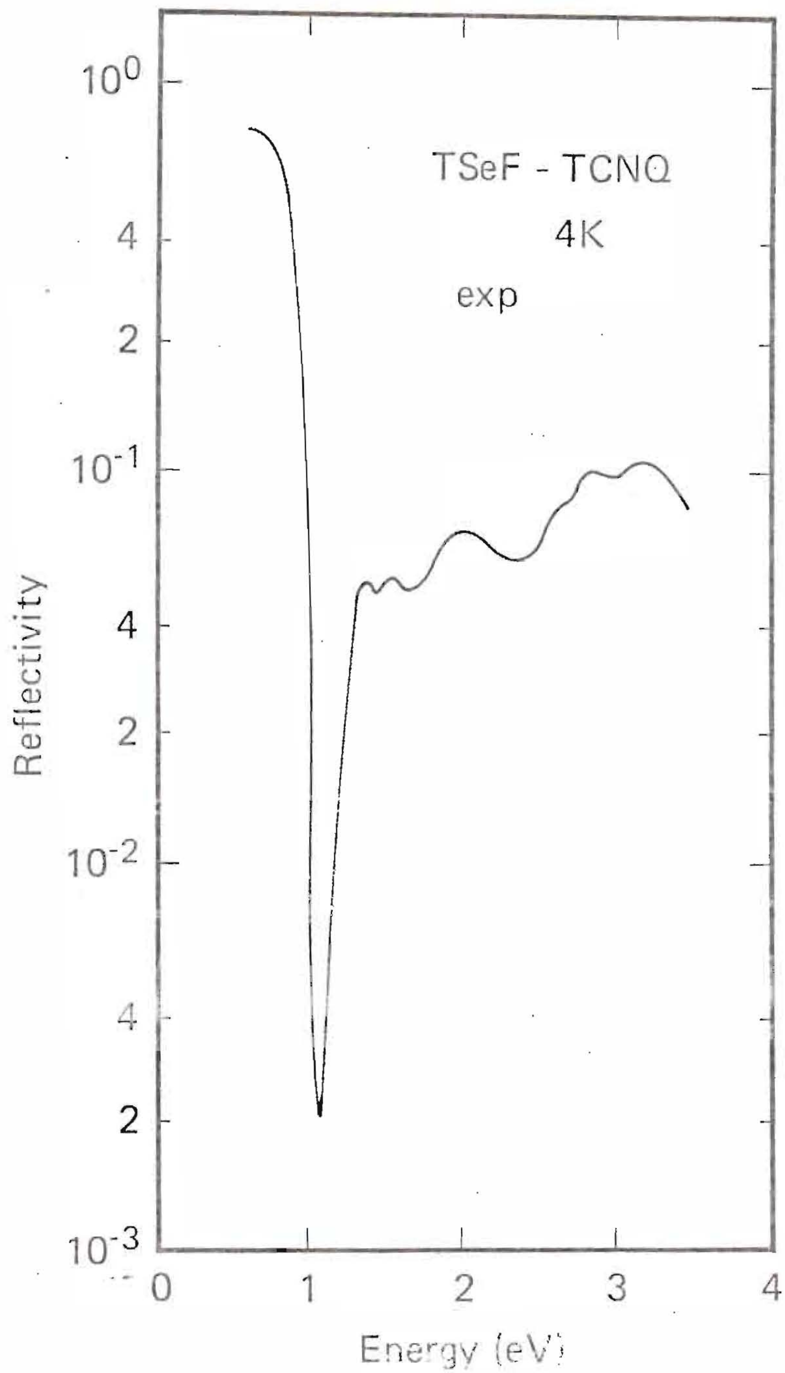
- b lattice parameter along the conducting axis
- V volume of the unit cell
- t_A , t_D transfer integral for acceptor and donor stack, resp.
- ρ transfer charge
- n number of bands with free carriers; (n=2)

Temperature dependence of

- b, V Schultz et al. : X-ray scattering
- t Herman: $t = t(b)$, $b = b(T)$

$$\rightarrow \quad \hbar^2 \omega_p^2 = f(T) * \sin \rho \frac{\pi}{2}$$


TSeF -TCNQ
4K
exp
fit
Reflectivity
1.000
0.800
0.400
0.200
0.100
0.080
0.040
0.020
0.010
0.008
0.004
0.002
0.001
0.5
1.0
1.5
2.0
2.5
3.0
3.5
4.0
Energy (eV)



TSeF - TCNQ
4K
exp
Reflectivity
10^0
4
2
10^{-1}
4
2
10^{-2}
4
2
10^{-3}
0
1
2
3
4
Energy (eV)

Measured: Reflectivity R as a function of energy

$$R = \frac{(n-1)^2 + k^2}{(n+1)^2 + k^2}$$

n, k optical constants

$n = n(\varepsilon)$
$k = k(\varepsilon)$

ε Dielectric function

Assumption: Reflectivity due to free carriers

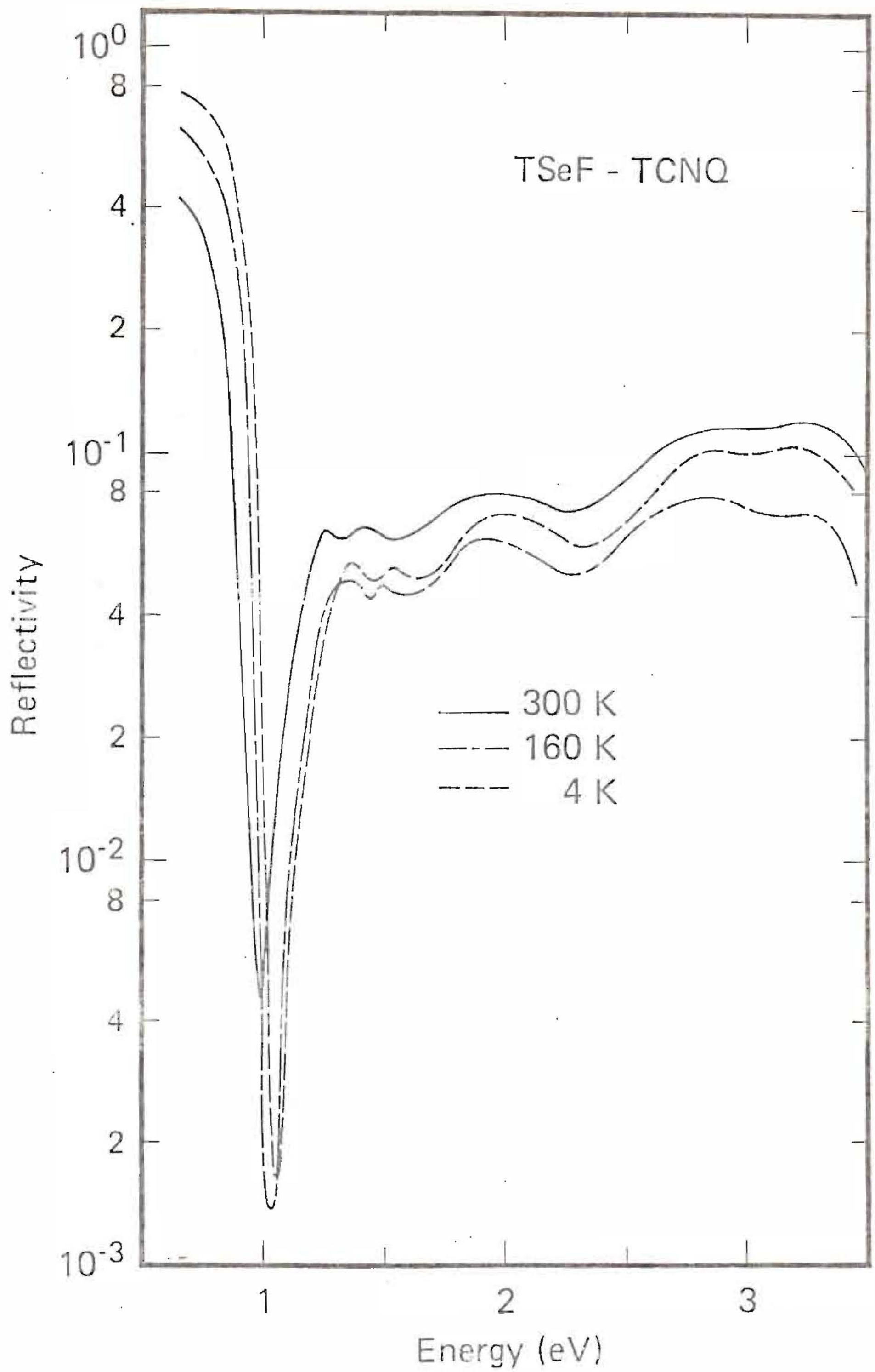
$\longrightarrow$ Drude - theory for ε

$$\varepsilon = \varepsilon_\infty - \frac{\omega_p^2}{\omega^2 + i\omega/\tau}$$

To approximate realistic ε_∞ : compose ε_∞ of a set of Lorentzians

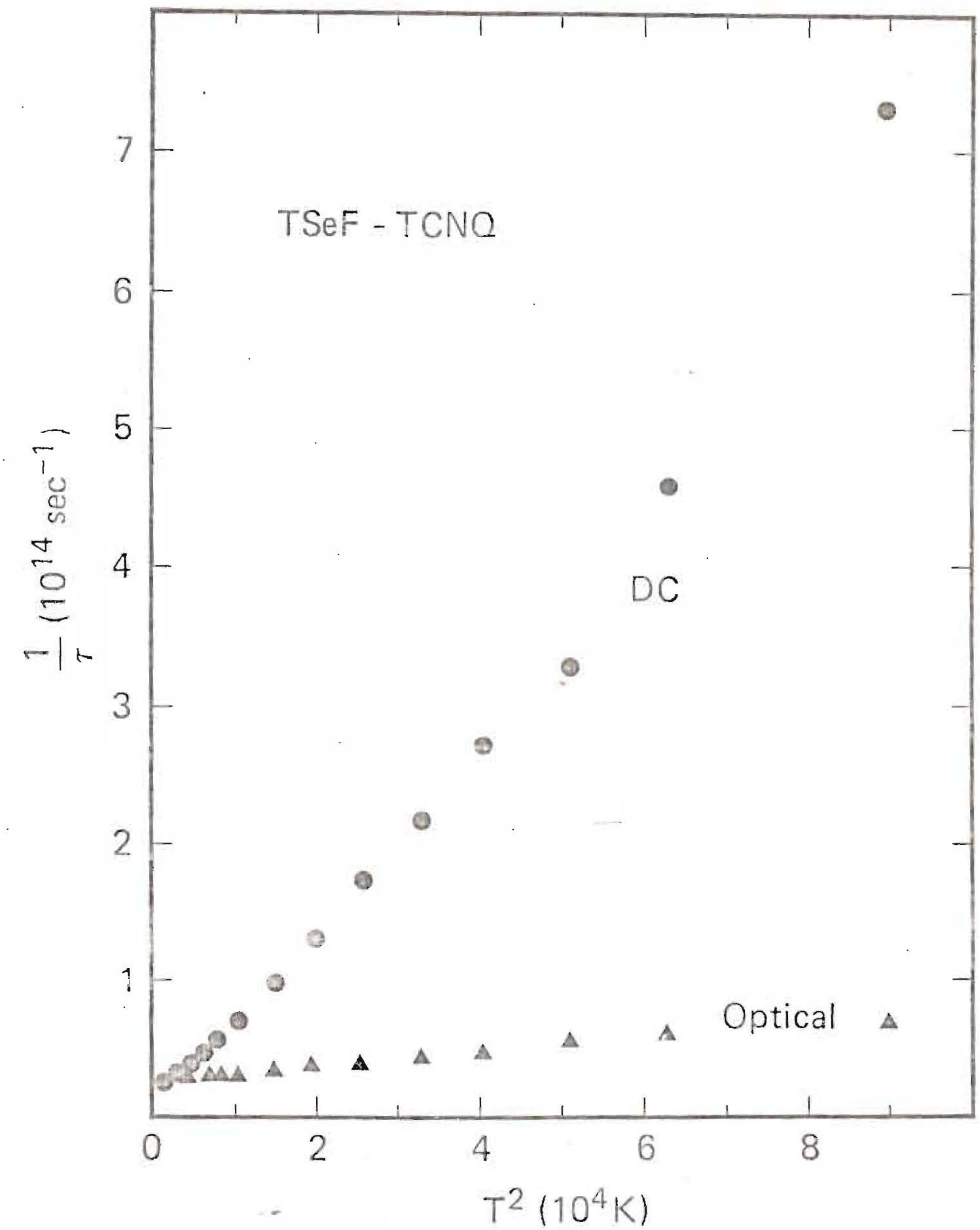
$$\varepsilon_\infty = 1 + \sum_j \frac{\nu_j f_j}{(\omega_{0j}^2 - \omega^2) + i\omega/\tau_j}$$

Note: We do not necessarily attribute any physical meaning
to the Lorentzians

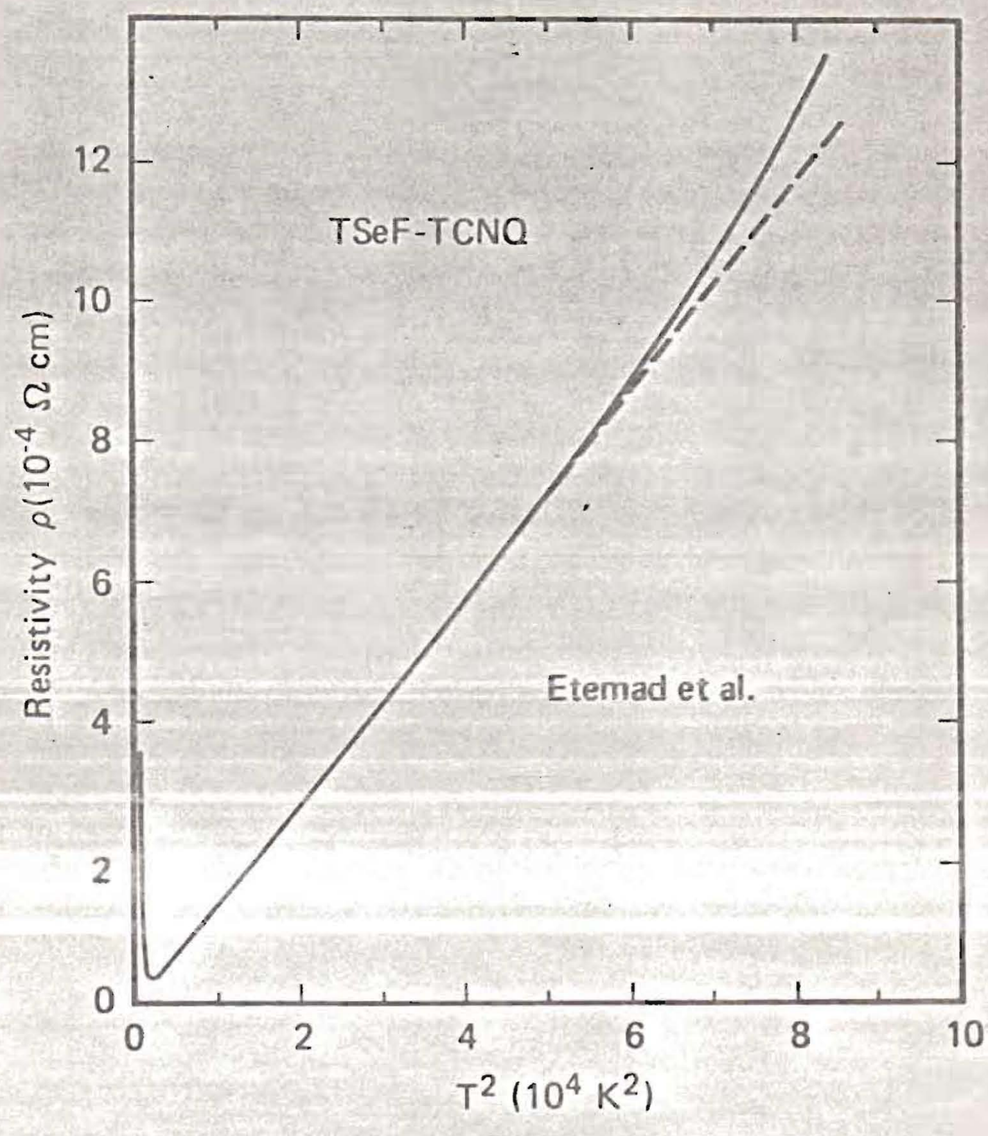


TSeF - TCNQ
300 K
160 K
4 K
Reflectivity
Energy (eV)
10^0
8
4
2
10^{-1}
8
4
2
10^{-2}
8
4
2
10^{-3}
1
2
3

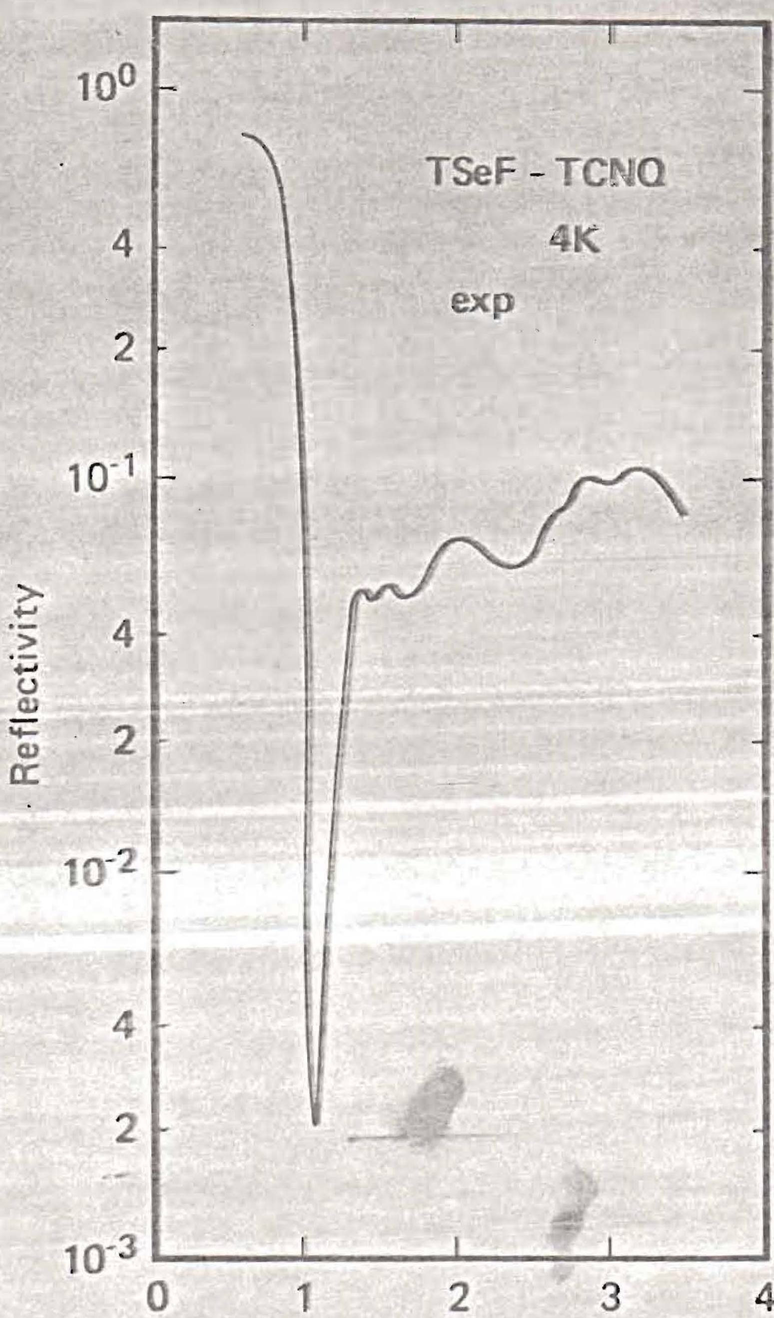
965 - IBM

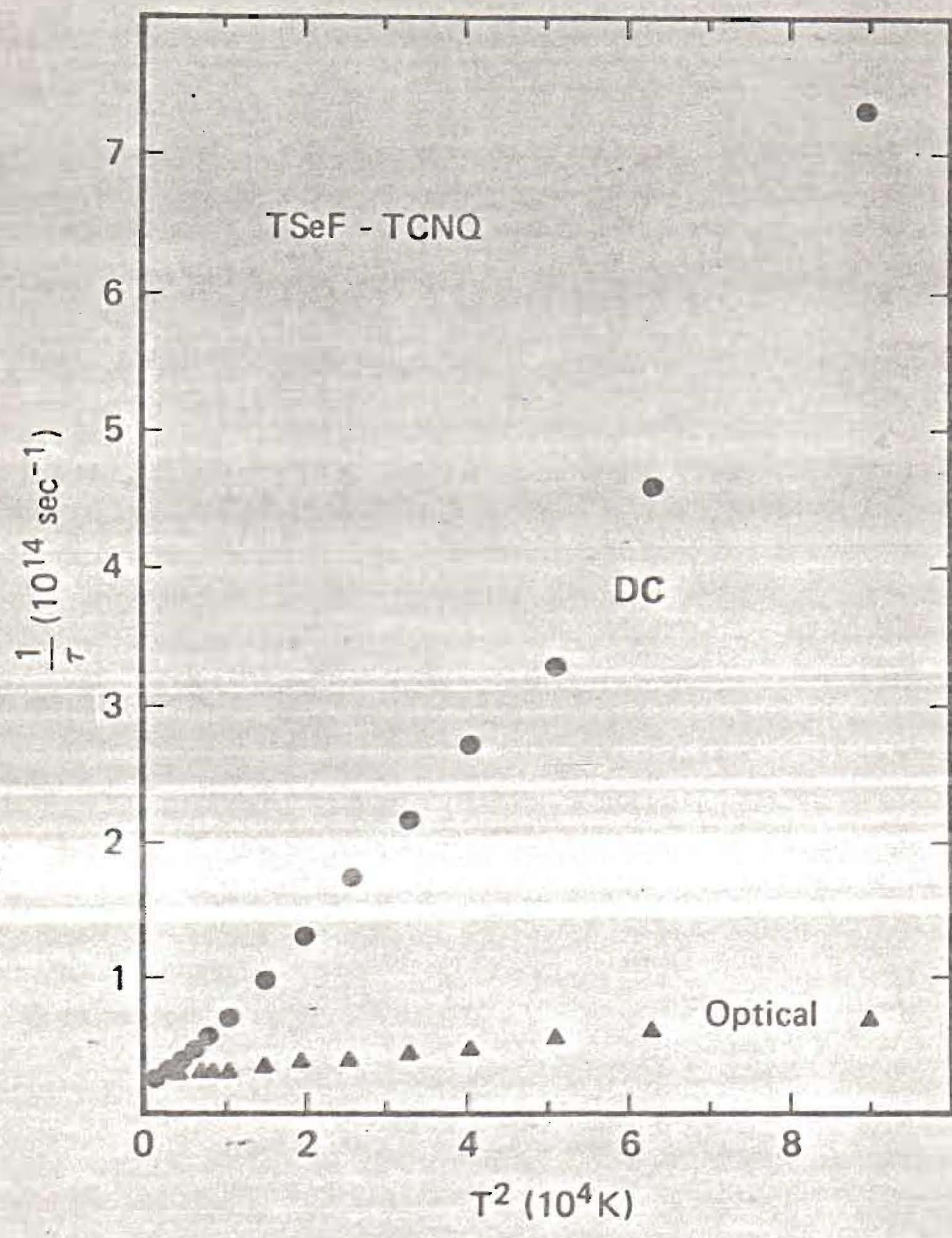


TSeF - TCNQ
DC
Optical
$\frac{1}{\tau}$ (10^{14} sec^{-1})
T^2 (10^4K)
0
2
4
6
8
1
2
3
4
5
6
7



TSeF-TCNQ
Etemad et al.
Resistivity ρ(10⁻⁴ Ω cm)
0
2
4
6
8
10
12
0
2
4
6
8
10
T² (10⁴ K²)


TSeF - TCNQ
4K
exp
Reflectivity
Energy (eV)
10^0
4
2
10^{-1}
4
2
10^{-2}
4
2
10^{-3}
0
1
2
3
4


TSeF - TCNQ
DC
Optical
$\frac{1}{\tau}$ (10^{14} sec^{-1})
T^2 (10^4K)
0
1
2
3
4
5
6
7
0
2
4
6
8

WHAT ABOUT $1/\tau\,(T)$?

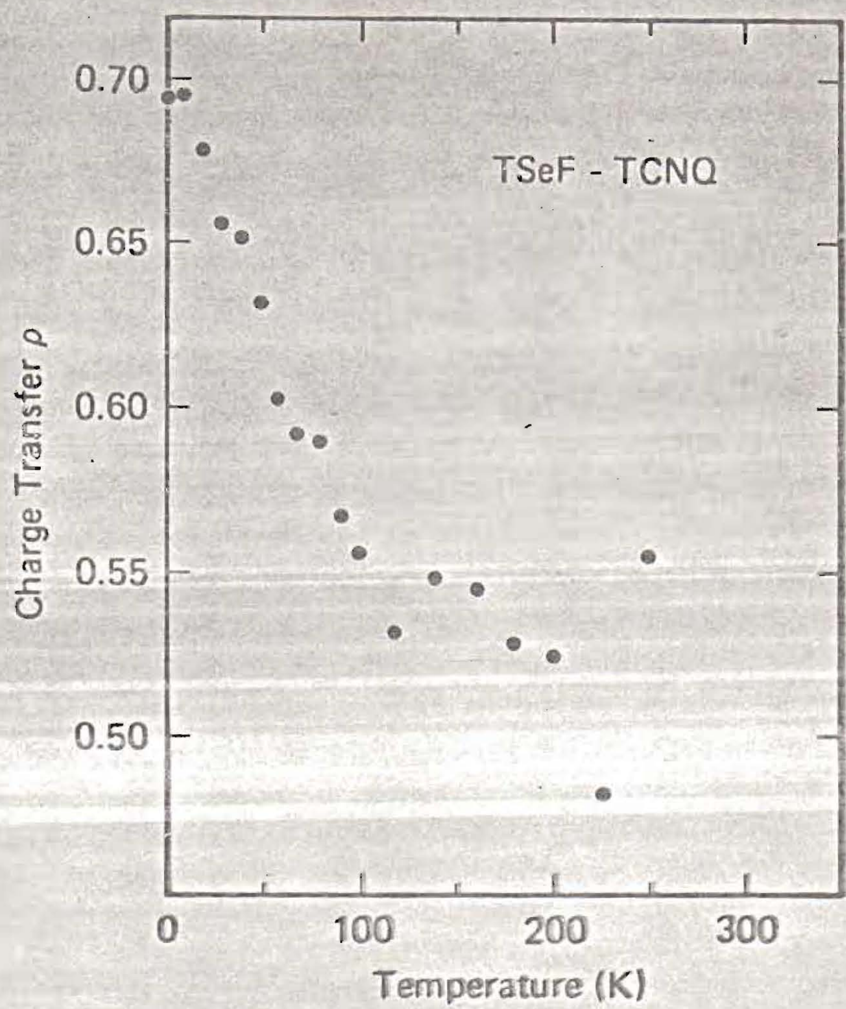
- $\tau^{-1}(T) = \tau_0^{-1} + \tau_{sp}^{-1}(T)$
  - τ_0^{-1} : OPTICAL PHONONS
    VIBRONIC STATES OF MOLECULES
    STATIC DISORDER
    SMALL HUBBARD GAPS
  - $\tau_{sp}^{-1}(T)$: ELECTRON - ELECTRON
    DYNAMIC DISORDER
    ELECTRON - LIBRON
    ELECTRON - PHONON

- ELECTRON - PHONON

$$\tau_{ep}^{-1}(T) = \left(\frac{2\pi}{\hbar}\right) \lambda \, (kT)$$

$$\lambda = \frac{N(E_F)\, I^2}{M \langle \omega^2 \rangle}$$

Nota Bene: $N(E_F)$, I^2 are T-dependent through simple volume effects


TSeF - TCNQ
Charge Transfer ρ
0.70
0.65
0.60
0.55
0.50
0
100
200
300
Temperature (K)