Photovoltaic effect for narrow-gap Mott Insulators

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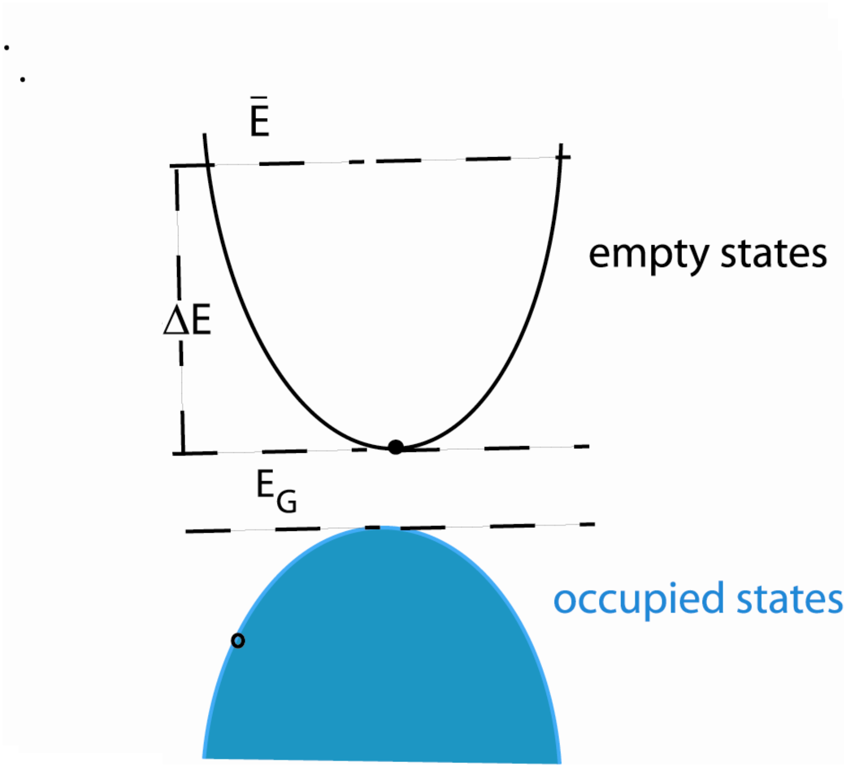
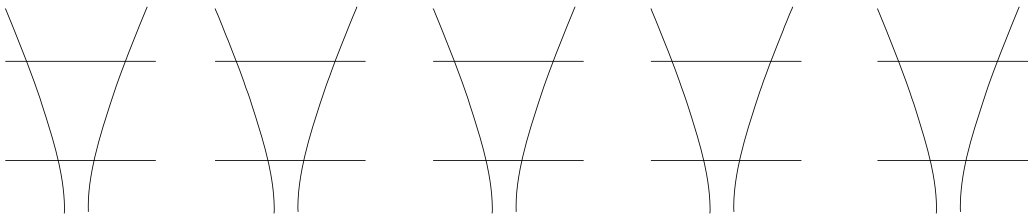
What is wrong with a band semiconductor



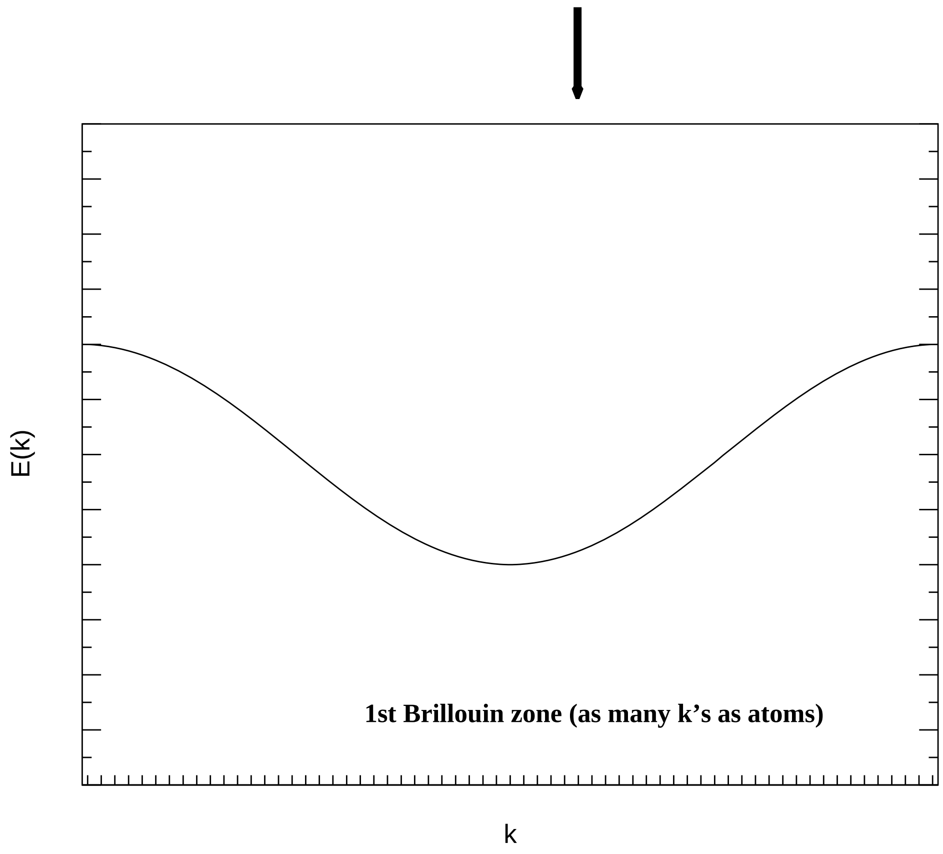
- The average energy range of the incident photon is 0.5 eV-3.5 eV.
- If the gap is chosen to be small, the excess energy ΔE beyond the gap of the higher energy photons is wasted into heat via phonon emission or electron-phonon scattering and within 10^{-12} secs the created electrons or holes relax in their band edges.
- If the gap is chosen to be large, it cuts-off most of the solar photons.
- A stack of cascaded multiple p-n junction with various gaps maybe an expensive way to increase efficiency.

What is a band insulator versus a Mott insulator

N atoms. Each having 2 energy levels.



From each atomic level we have N atomic orbitals giving rise to N Block states

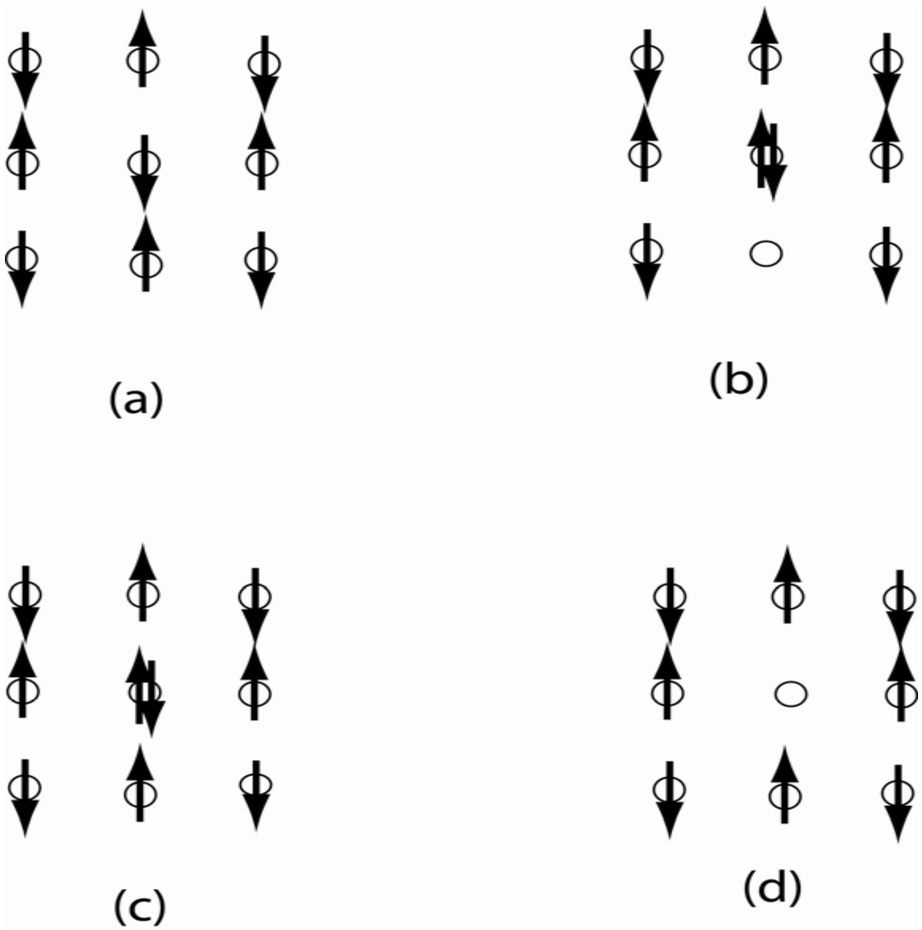


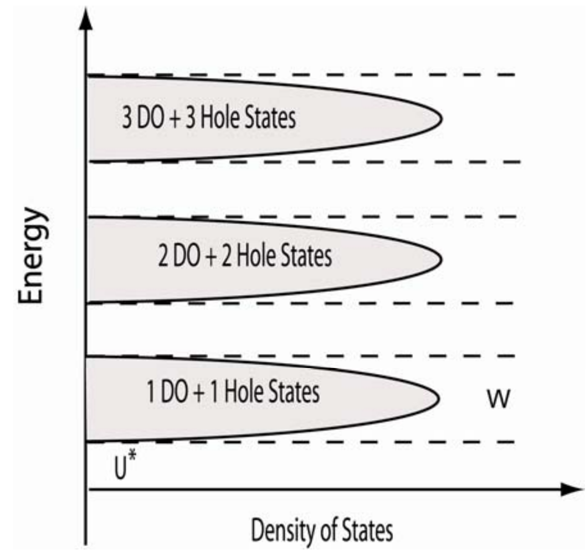
If we consider spin, there are 2N states.

a) one electron per atomic orbital, leads to a half-filled band (metal).
b) two electrons per atomic orbital, lead to a filled band (insulator).

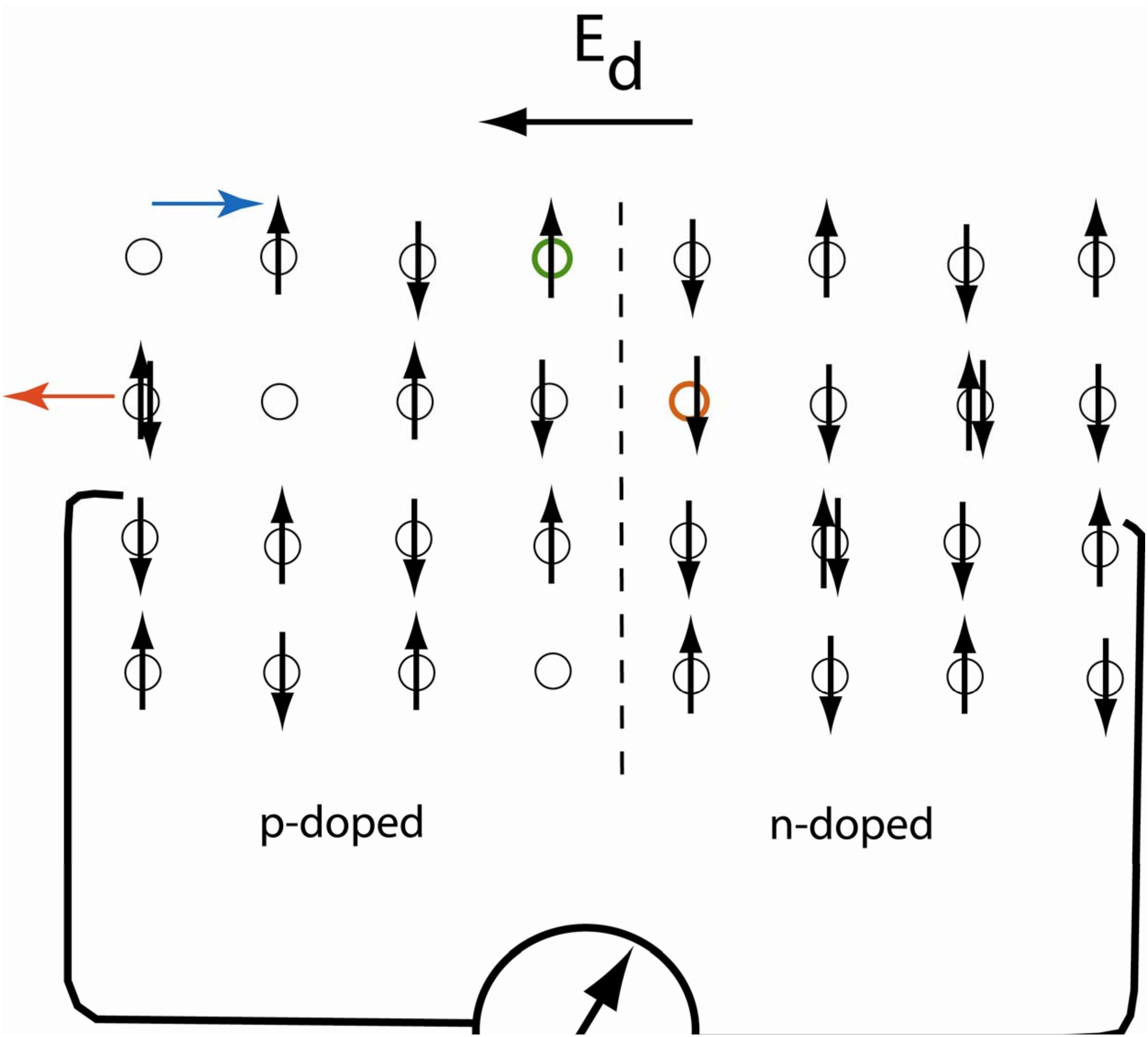
In a Mott insulator, one electron per atomic orbital leads to an insulator.

The Mott insulator



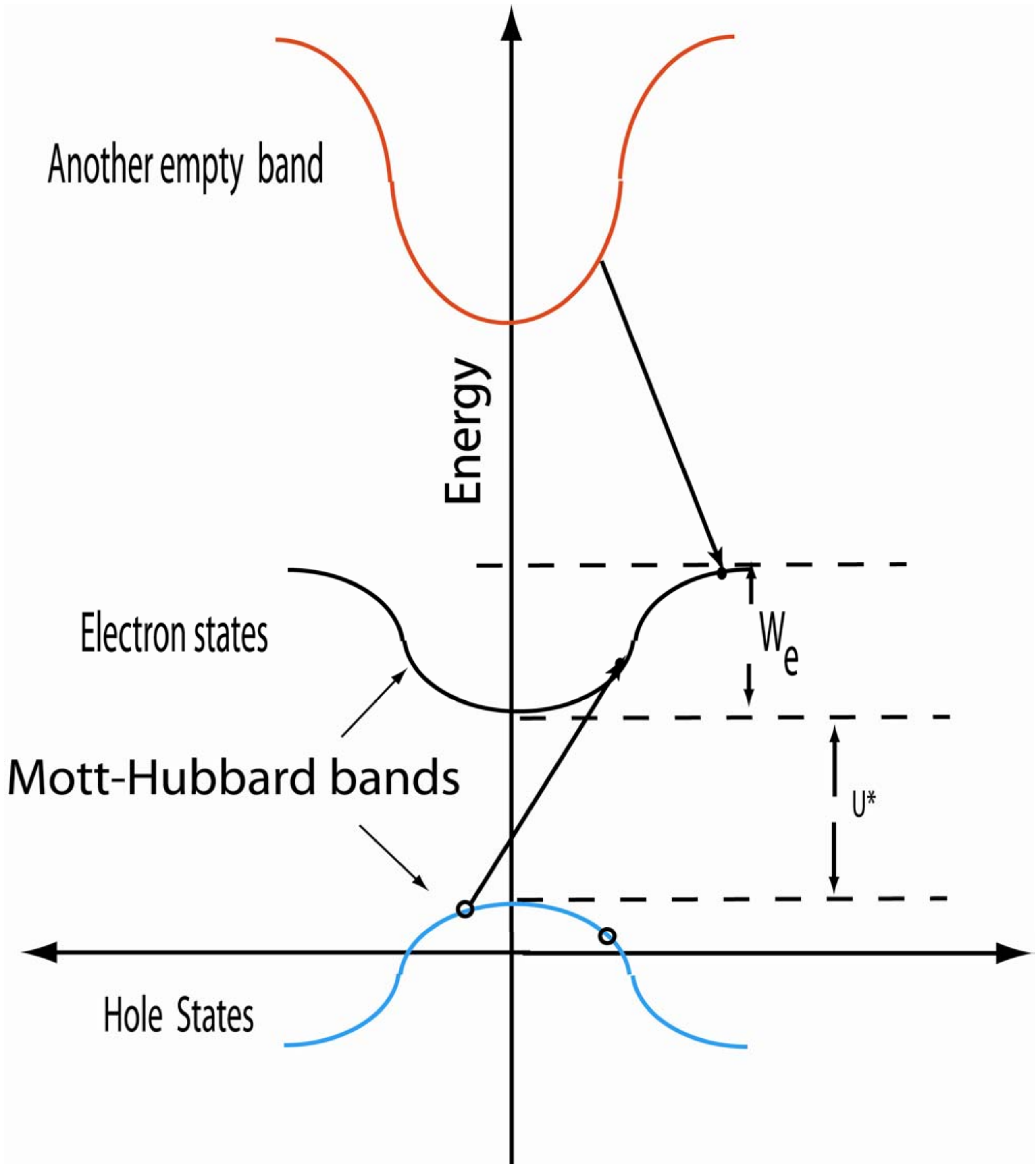


$$\hat{H} = -t \sum_{<ij>\sigma} c_{i\sigma}^{\dagger} c_{j\sigma} + U \sum_{i} \hat{n}_{i\uparrow} \hat{n}_{i\downarrow}$$

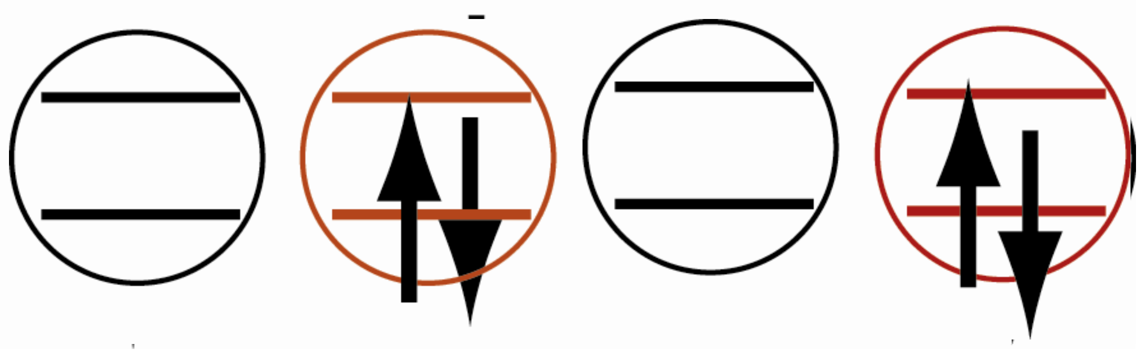


E_d
p-doped
n-doped

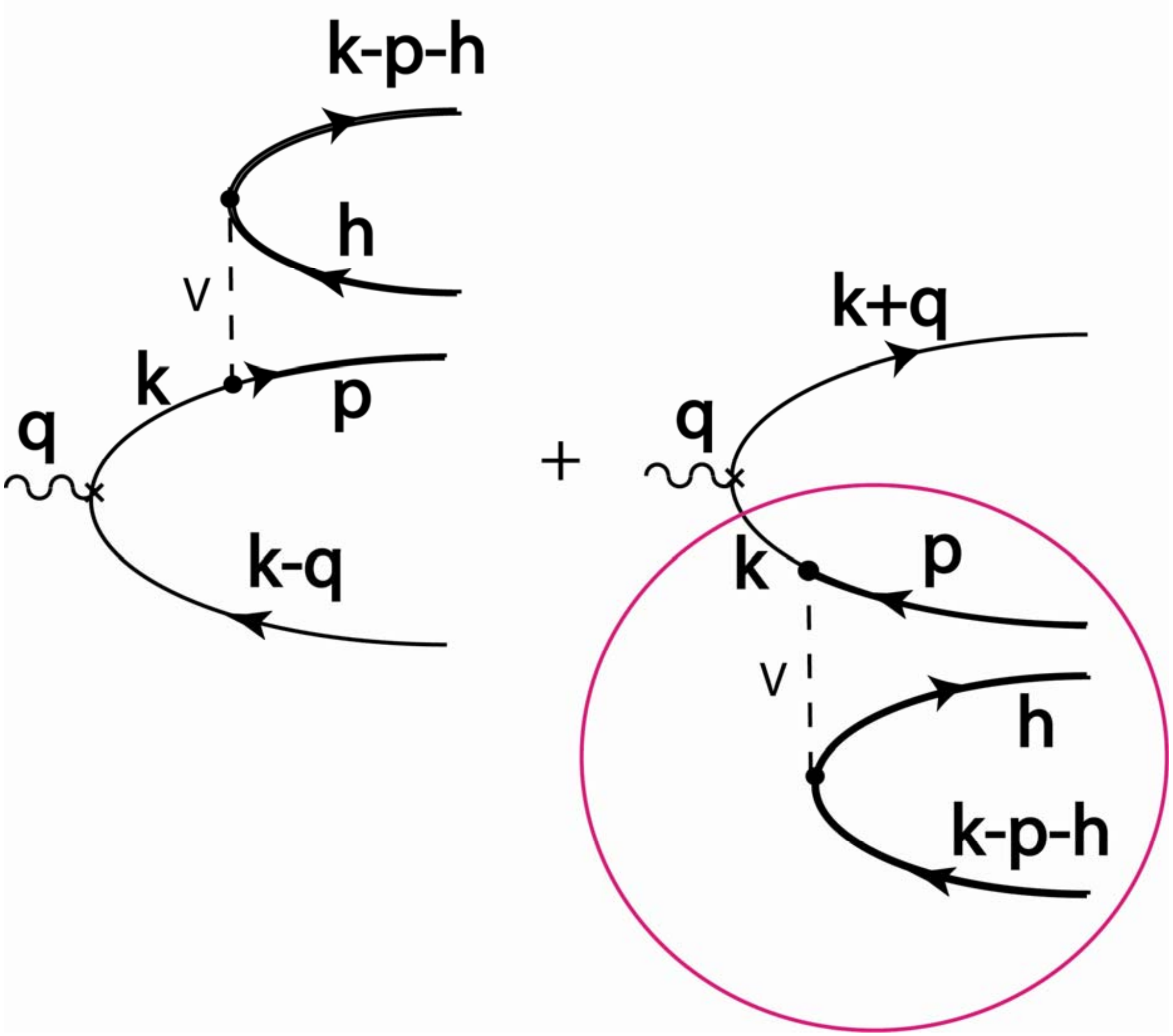
Photovoltaic effect in narrow-band narrow-gap Mott Insulator



Another empty band
Energy
Electron states
W
e
Mott-Hubbard bands
U*
Hole States



- As a perturbative process



Fermi's golden rule

$$\Gamma_{\mathbf{k}}(\epsilon) = \frac{2\pi}{\hbar} \sum_{\mathbf{p},\mathbf{h}} |M|^2 \delta(\epsilon - E_{\mathbf{k}-\mathbf{p}+\mathbf{h}} - E_{\mathbf{p}} + E_{\mathbf{h}}),$$
$$\epsilon \equiv E - U^*.$$

M is the coupling of the initially excited state $|\mathbf{k}\ \nu\rangle$ with the 2qp-1qh state $\alpha_{\mathbf{h}}\alpha^\dagger_{\mathbf{p}}\alpha^\dagger_{\mathbf{k}-\mathbf{p}-\mathbf{h}}|0\rangle$,

$$M = \frac{V}{N} Z_{\mathbf{h}} Z_{\mathbf{p}} Z_{\mathbf{k}-\mathbf{p}+\mathbf{h}},$$
$$V \equiv \langle \mathbf{k}\ \nu, \mathbf{h}|\hat{V}|\mathbf{p}, \mathbf{k}-\mathbf{p}+\mathbf{h}\rangle,$$
$$Z_p = \langle 0|\alpha_p|\mathbf{p}\ \mu\rangle.$$

Here the state $|\mathbf{k}\nu\rangle$ is the high energy band where the electron or hole is initially excited and $|\mathbf{p}\rangle$ is the band which becomes the Mott band after including the effects of the Hubbard interaction.

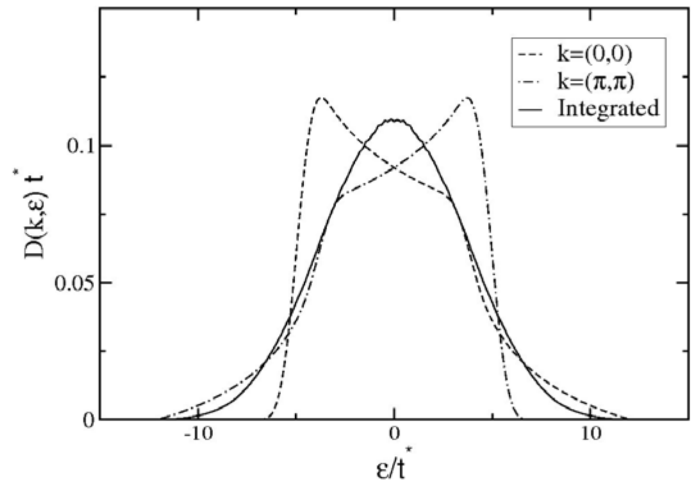
It will be further approximated as

$$\Gamma_{\mathbf{k}}(\epsilon) \simeq \frac{2\pi}{\hbar} V^2 \bar{Z}^6 D(\mathbf{k}, \epsilon),$$
$$D(\mathbf{k}, \epsilon) = \frac{1}{N^2} \sum_{\mathbf{p},\mathbf{h}} \delta(\epsilon - E_{\mathbf{k}-\mathbf{p}-\mathbf{h}} - E_{\mathbf{p}} + E_{\mathbf{h}}).$$

$\bar{Z}$ is the Z averaged over the Brillouin zone. Similarly

$$\bar{\Gamma}(\epsilon) \simeq \frac{2\pi}{\hbar} V^2 \bar{Z}^6 \Delta(\epsilon),$$
$$\Delta(\epsilon) = \frac{1}{N} \sum_{\mathbf{k}} D(\mathbf{k}, \epsilon).$$

$$E_k = -2t^* \sum_{\mu=1}^{d} \cos(k_\mu a_\mu)$$



$$\Delta \sim 1/W$$

The value of Z is calculated using the strong U limit of Hubbard i.e, the t-J model

Take V=U=1 eV, t=1/4

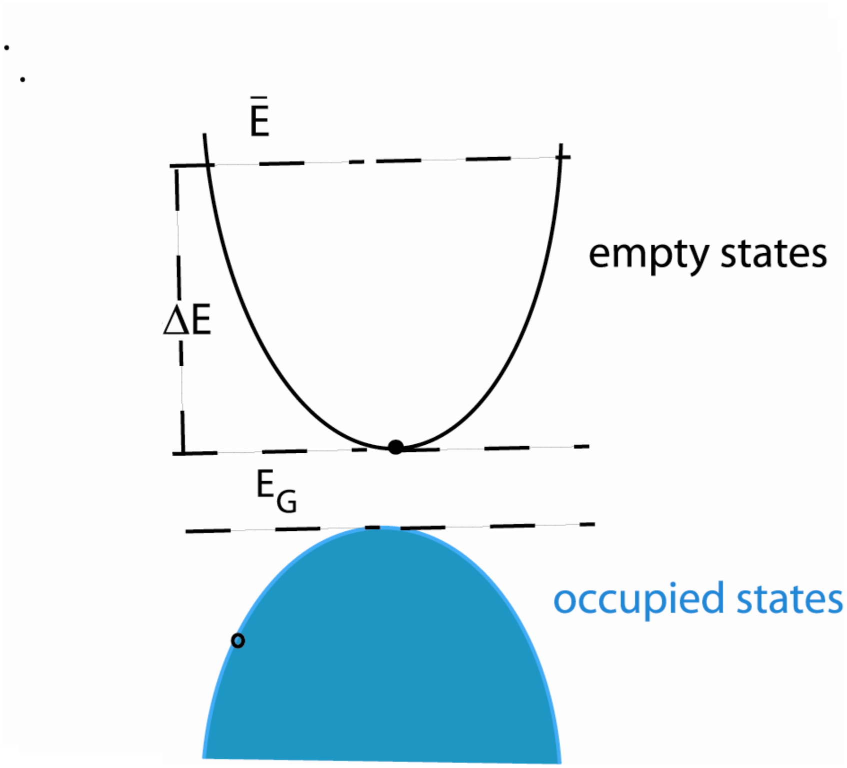
For these value

J/t=4t/U=1 and W=0.96 t, Z=0.53

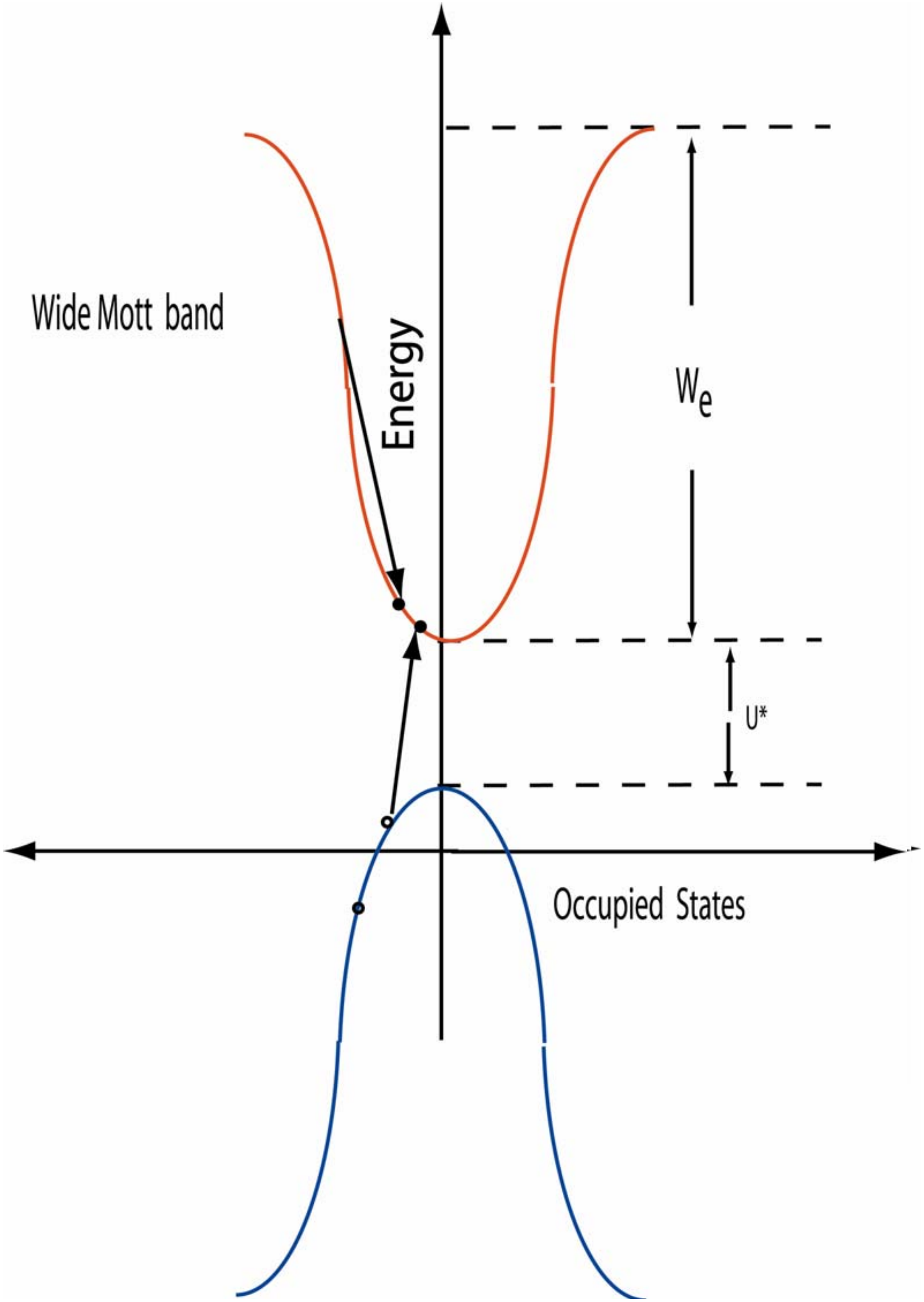
i.e. W=0.24 eV Γ=0.9x10^{15} sec^{-1}

which is much larger than decay via phonon emission or phonon scattering.

What is wrong with a band semiconductor



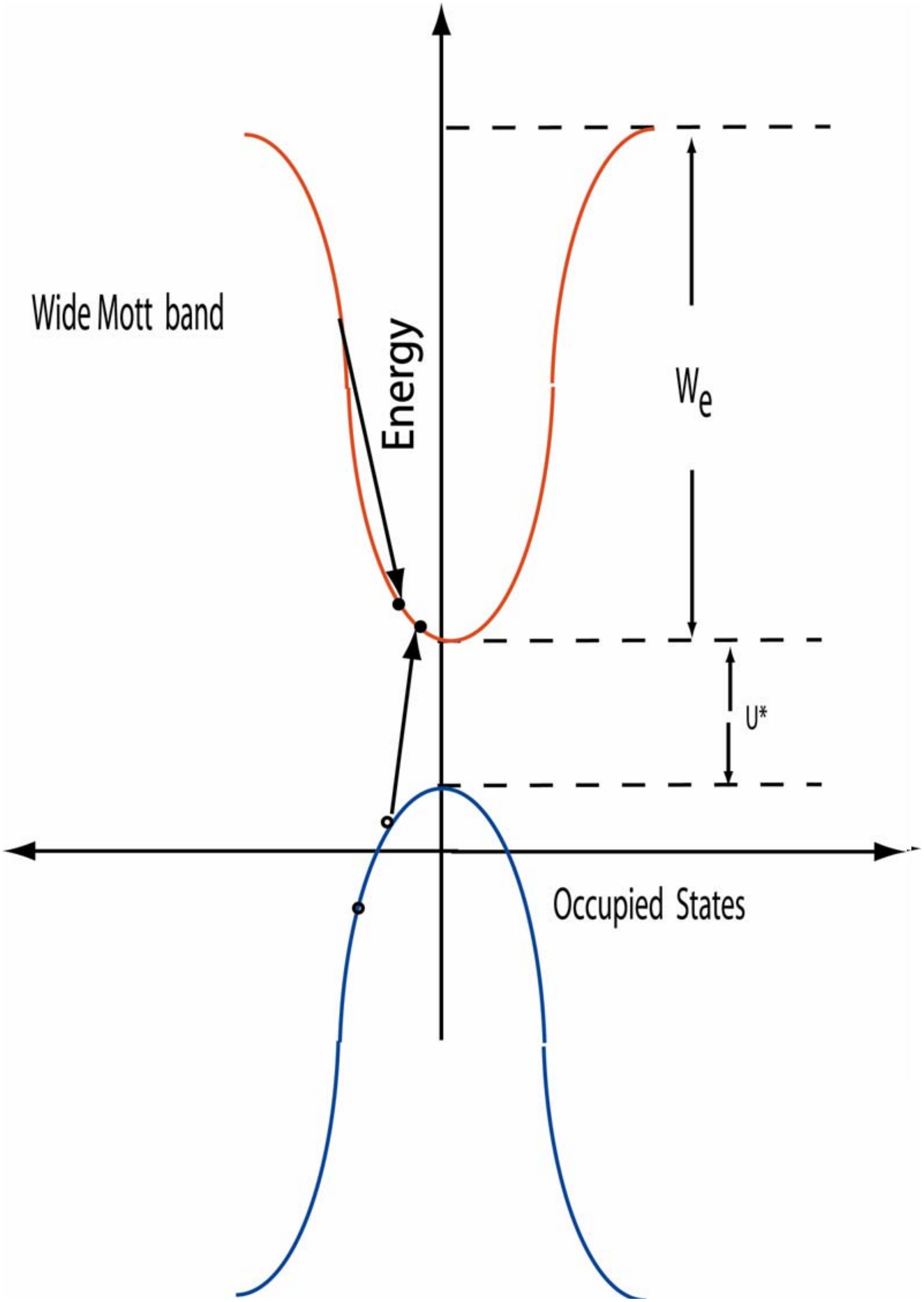
The excess energy ΔE beyond the gap of the higher energy photons is wasted into heat via phonon emission or electron-phonon scattering and within 10^{-12} secs the created electrons or holes relax in their band edges.



This process takes only 10^{-14}-10^{-15} secs.

So, before the phonons find enough time to take away the excess energy, it is converted to multiple doublon/hole pairs

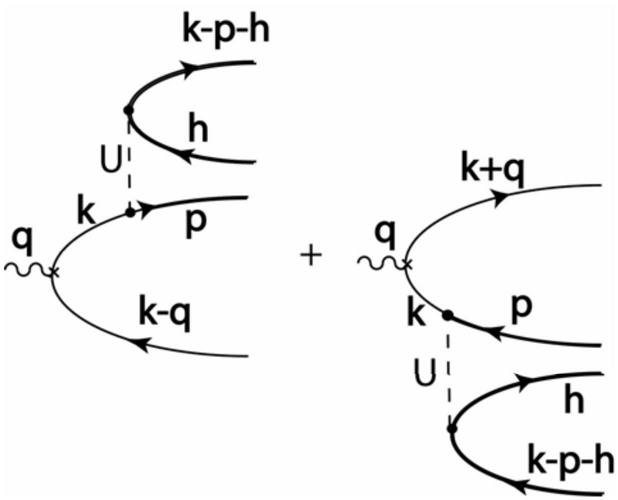
Photovoltaic effect in wide-band narrow-gap Mott Insulators



Narrow-gap Wide-band
Mott insulators.

When the energy E of the photo-excited electron is greater than nU*, the electron can decay into n+1 electrons plus n holes by means of the on-site Coulomb interaction U.

Narrow-gap Wide-band Mott insulators.



Narrow-gap wide-band case: $W >> U^*$

For any value of W we can tune U to get a small gap U^* and $U > W/2$.

For $W = 8$ eV and $U^* = 0.5$ eV we need a $U > 4$ eV.

The decay rate can be approximated by

$$\hbar\Gamma \sim 2\pi\frac{U^2}{W}\bar{Z}^6$$

For $W = 8$ eV and $U > 4$ eV, we obtain (using $\bar{Z} \sim 0.5$)

$$\Gamma \sim 0.5 \times 10^{15} sec^{-1}$$

Issues regarding efficiency

$$J_{absorbed} = \frac{\Omega_s}{4\pi^3\hbar^3c^2}\int_{U^*}^{\infty} d\epsilon \frac{\epsilon^3}{\exp(\frac{\epsilon}{k_B T}) - 1}$$

T=5760 K the average temperature on Sun's surface, Ω_s=Solid angle by which we see the Sun.

Impact Ionization and Auger recombination

$$D_1 \leftrightarrow D_1' + D_2 + H, \quad H_1 \leftrightarrow H_1' + H_2 + D$$

Carrier-carrier scattering

$$D_1 + D_2 \leftrightarrow D_1' + D_2',$$

$$H_1 + H_2 \leftrightarrow H_1' + H_2',$$

$$D + H \leftrightarrow D' + H'$$

Photon absorption and recombination (photo-luminescence)

$$D + H \leftrightarrow \gamma.$$

Equilibrium state

$$J_{emitted} = J_{absorbed},$$
$$J_{emitted} = \frac{\Omega_e}{4\pi^3\hbar^3c^2}\int_{U^*}^{\infty} d\epsilon \frac{\epsilon^3}{\exp(\frac{\epsilon-\mu_\gamma}{k_B T_e})-1}.$$

T_e is the cell's temperature, $\mu_\gamma=0$ is the chemical potential of the D-H system in equilibrium with photons

Since all the processes involving only electronic degrees of freedom are much faster than the relaxation processes involving phonons

$$D \leftrightarrow D' + \Gamma,\ H \leftrightarrow H' + \Gamma, D + H \leftrightarrow \Gamma$$

The maximum efficiency under these conditions can be calculated as the maximum of the following quantity η as a function of the voltage V and the Mott-Hubbard gap *U**

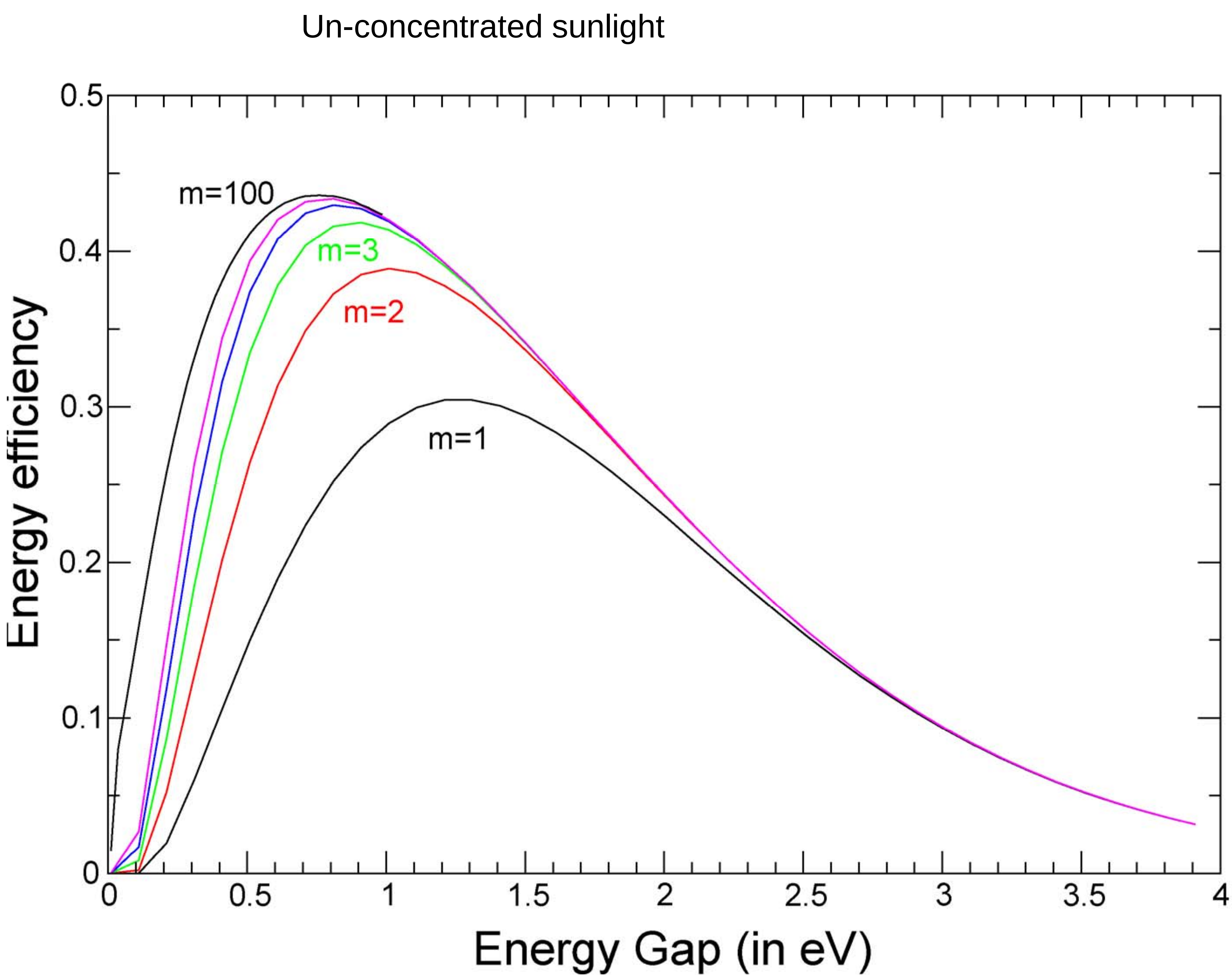
$$\eta(V,U^*) = qV\frac{g(U^*) - \xi\ r(V,U^*)}{P_{in}},$$

$$P_{in} = \int_0^\infty d\epsilon \frac{\epsilon^3}{\exp(\epsilon/k_B T_s)-1}$$

$$g(U^*) = \int_{U^*}^\infty d\epsilon \frac{m(\epsilon)\epsilon^2}{\exp(\epsilon/k_B T_e)-1},$$

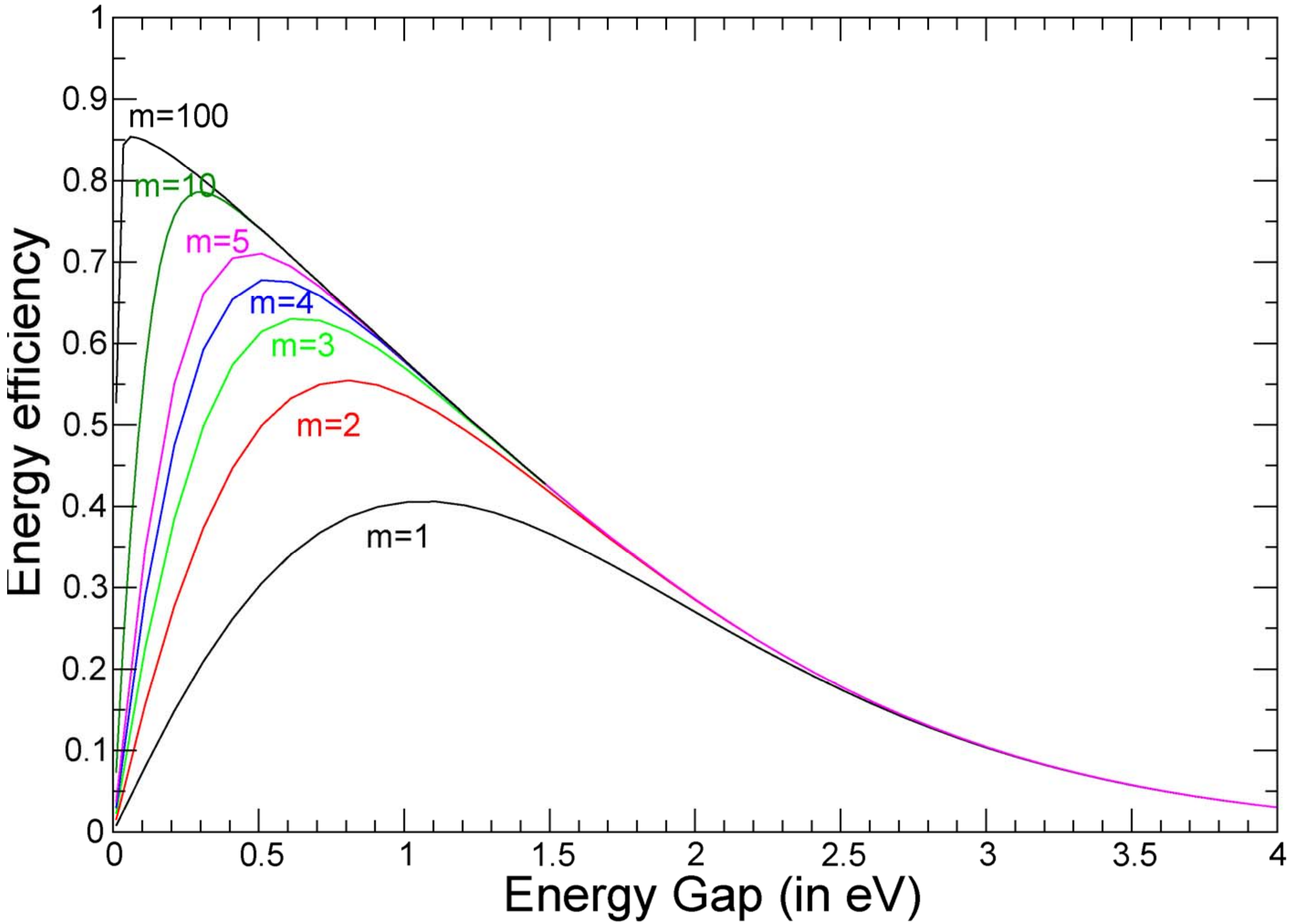
$$r(V,U^*) = \int_{U^*}^\infty d\epsilon \frac{m(\epsilon)\epsilon^2}{\exp((\epsilon - qV)/k_B T_e) -}$$

$m(\varepsilon)$ is the smaller of the following (a) integer part of ε/U^*, and (b) a maximum allowed value M_{max} ,and $\xi=\pi/\Omega_s$ or 1 for fully concentrated sunlight.



Un-concentrated sunlight
m=100
m=3
m=2
m=1
Energy efficiency
Energy Gap (in eV)
0
0.1
0.2
0.3
0.4
0.5
0.5
1
1.5
2
2.5
3
3.5
4

Fully concentrated sunlight



Photovoltaic effect on Perovskite heterojunctions

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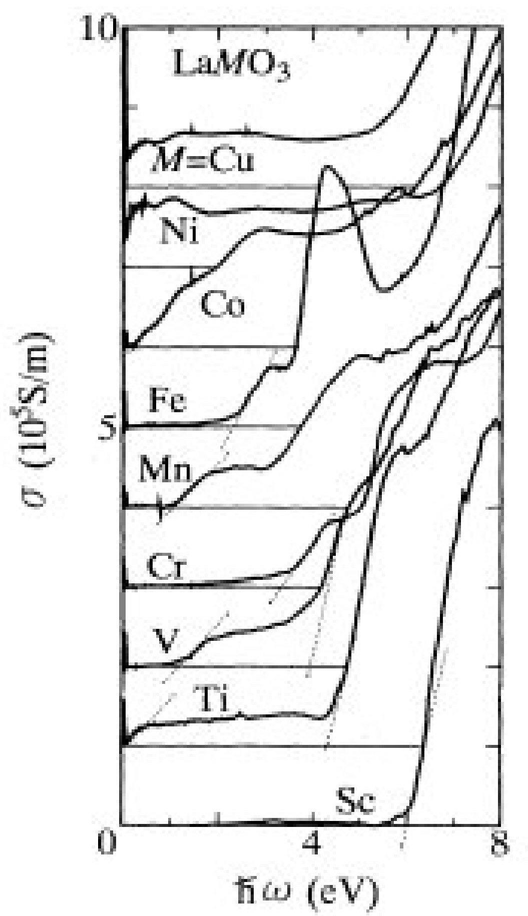
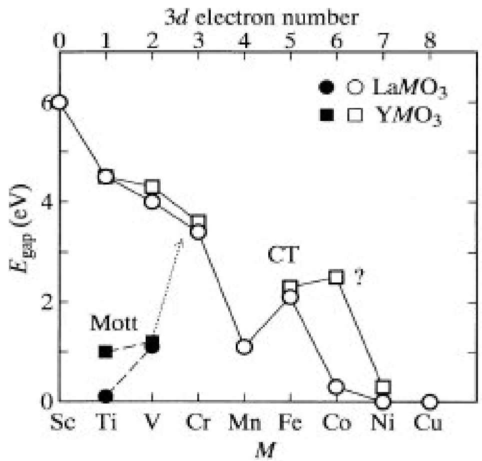
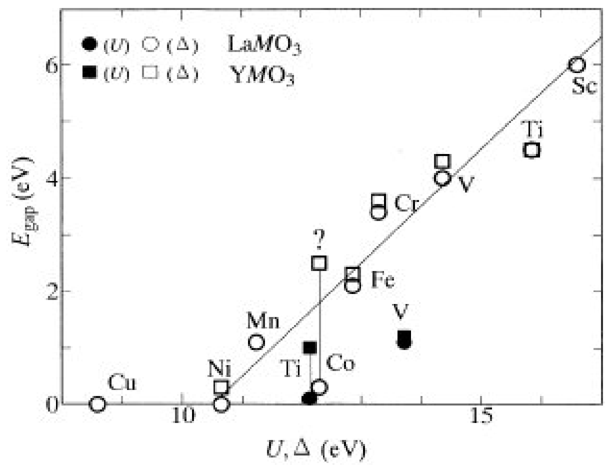


Optical conductivity
and optical gaps in some
perovskite materials

Gaps in Perovskites

17 008 T. ARIMA, Y. TOKURA, AND J. B. TORRANCE 48





We are focusing our attention to the following class of materials

VO_2

$LaVO_3$ and $SrVO_3$

$LaTiO_3$ and $YTiO_3$

We would like to study optical properties of these materials
Using hybrid functionals and many-body perturbation theory.

Maitri Warusawithana of NHMFL using MBE is growing
epitaxially $LaVO_3$ films on $SrTiO_3$

We plan to carry out photoconductivity studies of films of such materials.

Fast optical pump and probe techniques should probe the decay products
of photo-excited electron/hole pairs.

Density functional theory studies of films of the above class of materials.

Conclusions

- When a solar photon excites an electron-hole pair above the Mott-Hubbard gap in a narrow-gap narrow-band Mott insulator the photo-excited electron or hole which temporarily lands on a different high energy band can decay quickly into n+1 quasi-electrons plus n quasi-holes from the Mott band or n+1 quasi-holes plus n quasi-electrons. The time scale for this energy conversion is shorter than the one required for electron/hole relaxation through phonons or spin wave excitations.
- In the case of narrow-gap wide-band Mott insulators the excess energy of the photo-excited electron (or hole) is converted into (n+1)-quasi-electrons plus n quasi-holes (or n+1 quasi-holes plus n quasi-electrons) also within short time scales before the electron or hole loses its excess energy via phonon or other excitations.
- Therefore, a solar cell made using a narrow gap Mott insulator for the illuminated side of the p-n junction should lead to an efficient solar cell through the production of a high current.
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