Optical processes and excitons

- Dielectric function and reflectance
- Kramers-Kronig relations
- Excitons
    - Frenkel exciton
    - Mott-Wannier exciton
- Raman spectroscopy

Dept of Phys

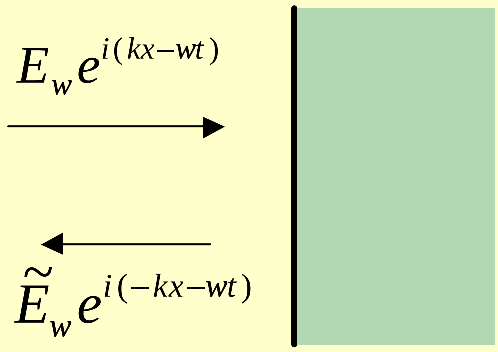
M.C. Chang

Dielectric function, reflectivity r, and reflectance R

Response of a crystal to an EM field is characterized by ε(k,ω), (k≈0 compared to G/2)

Experimentalists prefer to measure reflectivity r

(normal incidence)



$$r(w) \equiv \frac{E'_w}{E_w}$$

If $$\sqrt{e(w)} = n(w)$$

then (Prob.3) $$r(w) = \frac{n-1}{n+1} \equiv \sqrt{R(w)}e^{iq(w)}$$

It is easier to measure R than to measure θ

∴ measure R(ω) for ∀ω → θ(ω) (with the help of KK relations)

→ n(ω)

→ ε(ω)

Kramers-Kronig relations (1926)

examples of response function:

$$j_w = s(w)E_w \qquad \text{Ohm's law}$$

$$P_w = c(w)E_w \qquad \text{polarization}$$

$$\tilde{E}_w = r(w)E_w \qquad \text{reflective EM wave}$$

KK relation connects real part of the response function with the imaginary part

Example: Response of charged (independent) oscillators

For the j-th oscillator (atom or molecule with bound charges),

$$m_j\left(\frac{d^2}{dt^2} + h_i\frac{d}{dt} + w_j^2\right)x_j = F_{jw}e^{-iwt}, \quad F_{jw} = -Z_j eE_w$$

$$x_j(t) = x_{jw}e^{-iwt} \qquad \text{steady state}$$

$$p_{jw} = -Z_j ex_{jw} = a_j(w)E_w \qquad \text{polarization}$$

$$\Rightarrow \quad a_j(w) = \frac{Z_j^2 e^2}{m_j\left(w_j^2 - w^2 - ih_j w\right)}$$

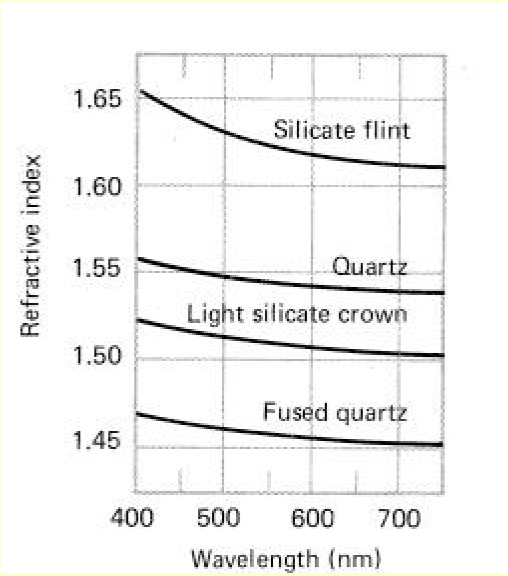
Figs from http://www.physics.uc.edu/~sitko/LightColor/10-Optics1/optics1.htm

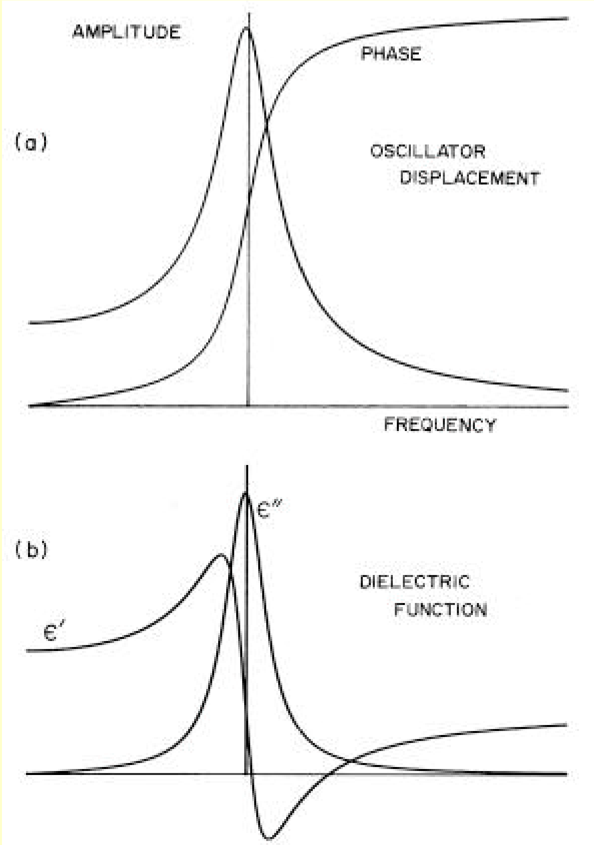
Collection of oscillators

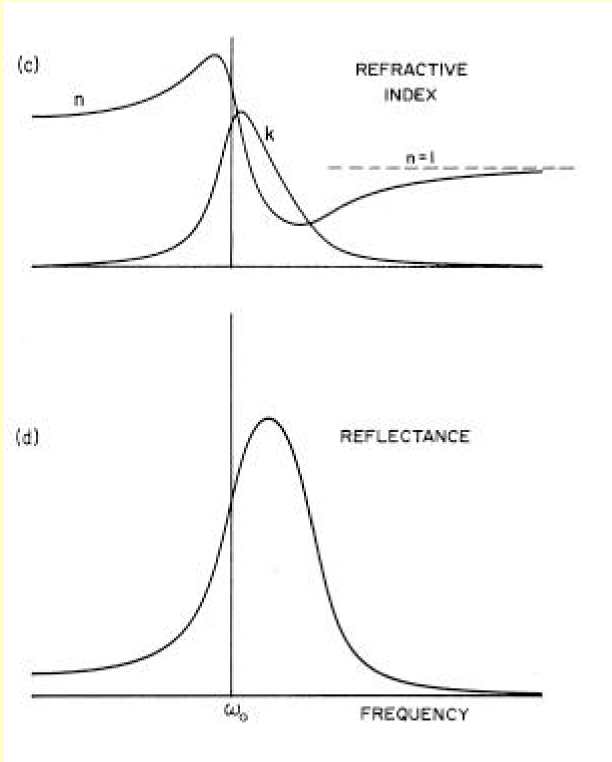
$$P_\omega = \frac{1}{V}\sum_j -Z_j e x_{j\omega} = \chi(\omega) E_\omega \quad \Rightarrow \quad \chi(\omega) = \frac{1}{V}\sum_j \frac{Z_j^2 e^2}{m_j\left(\omega_j^2 - \omega^2 - i\eta_j \omega\right)}, \quad (1)$$

for identical oscillators $\rightarrow \dfrac{nZ^2 e^2 / m}{\omega_0^2 - \omega^2 - i\eta\omega}$

Different refractions for blue/red (normal)







Some properties of χ(ω):

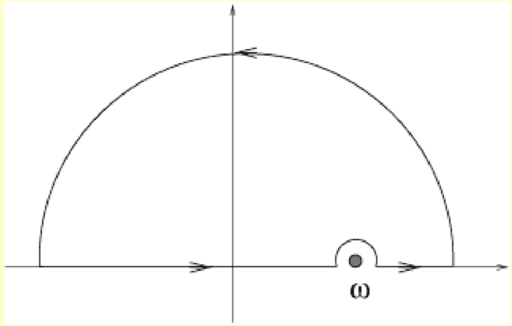
(1) χ(ω) has no pole above (including) x-axis.

(2) $\int d\omega \, \dfrac{\chi(\omega)}{\omega} = 0$ along (upper) infinite semi-circle

(3) χ’(ω) is even in ω, χ’’(ω) is odd in ω.

From (1), (2), we have $a(w) = \frac{1}{pi} P \int_{-\infty}^{\infty} \frac{a(s)}{s-w} ds$

(α can be χ, or σ, or...)



$$\Rightarrow \quad a'(w) = \frac{1}{p} P \int_{-\infty}^{\infty} \frac{a''(s)}{s-w} ds$$

$$= \frac{1}{pi} P\left(\int_{0}^{\infty} \frac{a''(s)}{s-w} ds + \int_{0}^{\infty} \frac{a''(s)}{s+w} ds \right)$$ property (3) used

Also,

$$a'(w) = \frac{2}{p} \int_{0}^{\infty} \frac{s a''(s)}{s^2 - w^2} ds \quad (2)$$

$$a''(w) = -\frac{2w}{p} \int_{0}^{\infty} \frac{a'(s)}{s^2 - w^2} ds \quad (3)$$

Offers a compatibility check for experimentalists

A few sum rules:

$$\frac{2}{p} \int_{0}^{\infty} \frac{a''(s)}{s} ds = a'(0)$$

An example of "fluctuation-dissipation" relation

From (1) and (2), ω>>1 (Prob. 2):

$$\frac{2}{p} \int_{0}^{\infty} s a''(s) ds = \frac{1}{V} \sum_j f_j, \qquad f_j = \frac{Z_j^2 e^2}{m}$$

sum rule for oscillator strengths

From (3), ω>>1 (Prob. 4):

$$\frac{2}{p} \int_{0}^{\infty} a'(s) ds = \lim_{w \to \infty} w a''(w), \quad (4)$$

and many more…, e.g., (4)+(5) (next page):

$$\int_{0}^{\infty} s'(s) ds = \frac{1}{8} w_p^2$$

By Ferrel and Glover (1958)

Apply the KK relation to $\tilde{E}_w = r(w) E_w$

$$\ln r(w) = \ln R^{1/2}(w) + iq(w)$$

then $$q(w) = -\frac{w}{p}\int_0^\infty \frac{\ln R(s)}{s^2 - w^2} ds$$

$$= -\frac{1}{2p}\int_0^\infty \ln\left|\frac{s+w}{s-w}\right| \frac{d\ln R}{ds} ds$$

- constant R(s) doesn't contribute
- s>>ω, s<<ω don't contribute

Conductivity of free electron gas (ω_j=0, with little damping)

$$c(w) = \frac{1}{V}\sum_j \frac{e^2}{m\left(w_j^2 - w^2 - ih_j w\right)}$$

$$\Rightarrow c(w) = -\frac{ne^2}{mw}\lim_{h\to 0}\frac{1}{w+ih} = -\frac{ne^2}{mw}\left[\frac{1}{w} - ipd(w)\right]$$

(KK relations can be checked easily in this case)

$$e(w) = 1 + 4pc(w) = 1 - \frac{w_p^2}{w^2}\left[1 - ipwd(w)\right]$$

Recall that (chap 10) $$e = 1 + \frac{4pis}{w}$$

$$\therefore \quad s = (e-1)\frac{w}{4pi} = \frac{ne^2}{m}\left[pd(w) + \frac{i}{w}\right], \quad (5)$$

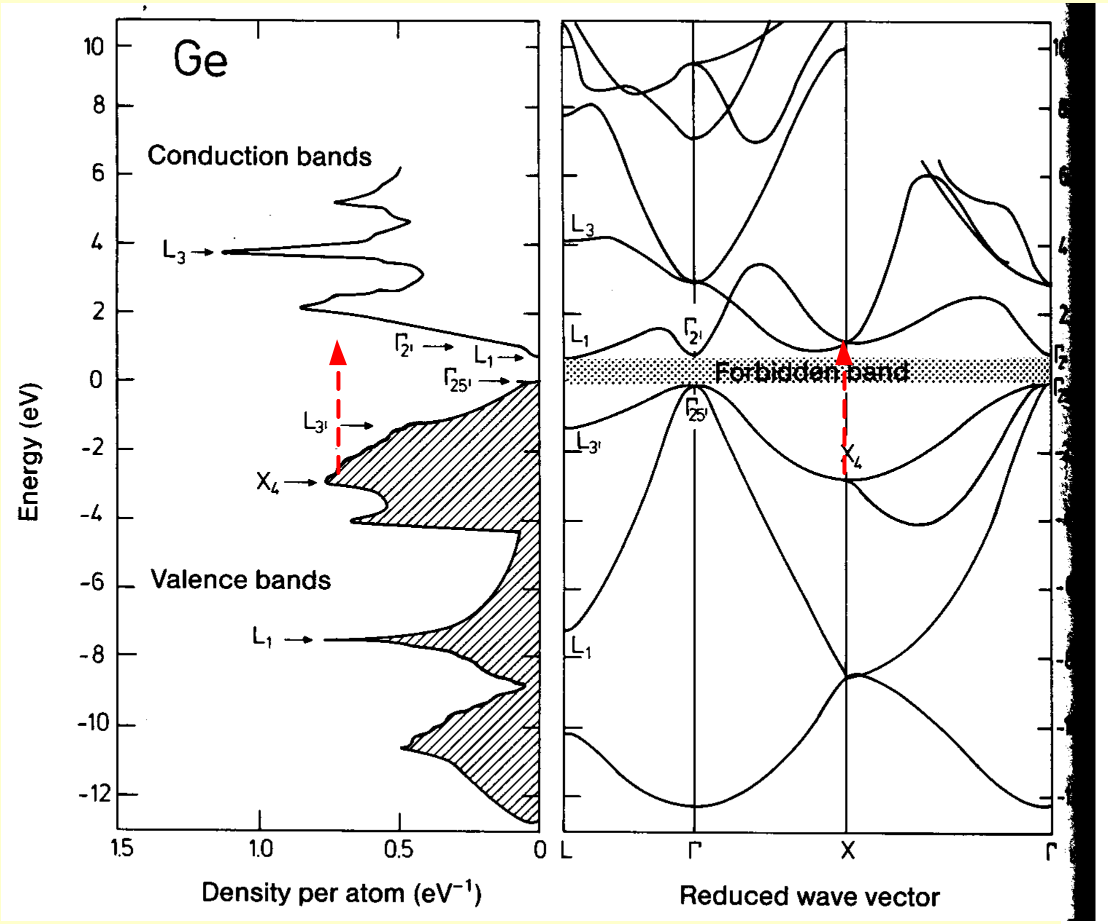
Real part of DC conductivity diverges (compare with $ne^2\tau/m$)

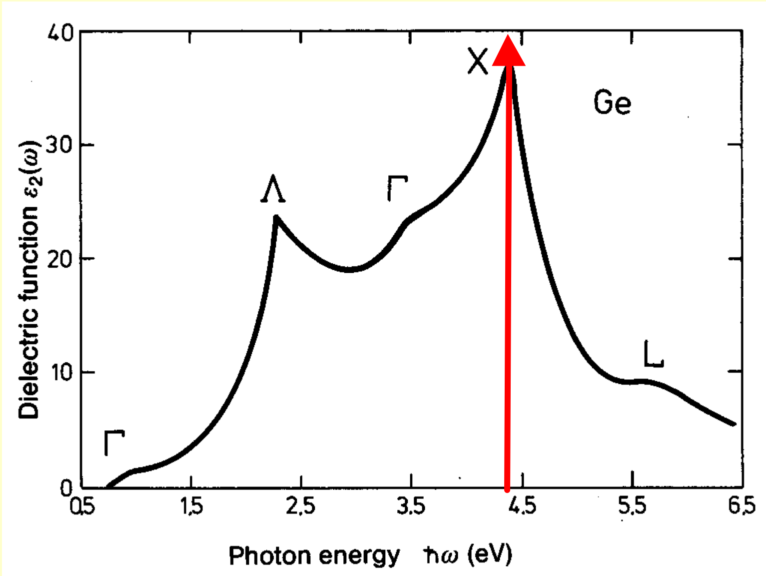
Dielectric function and the semiconductor energy gap (prob.5)

band gap absorption (due to nonzero ε'') can be very roughly approximated by

$$\varepsilon''(\omega) = \frac{\omega_p^2}{\omega}\pi\delta(\omega) \Rightarrow \frac{\omega_p^2}{2\omega}\pi\delta(\omega-\omega_g)$$

$$\Rightarrow \varepsilon'(\omega) = 1 - \frac{\omega_p^2}{\omega^2-\omega_g^2} \quad \therefore \quad \varepsilon'(0) = 1 + \frac{\omega_p^2}{\omega_g^2}$$



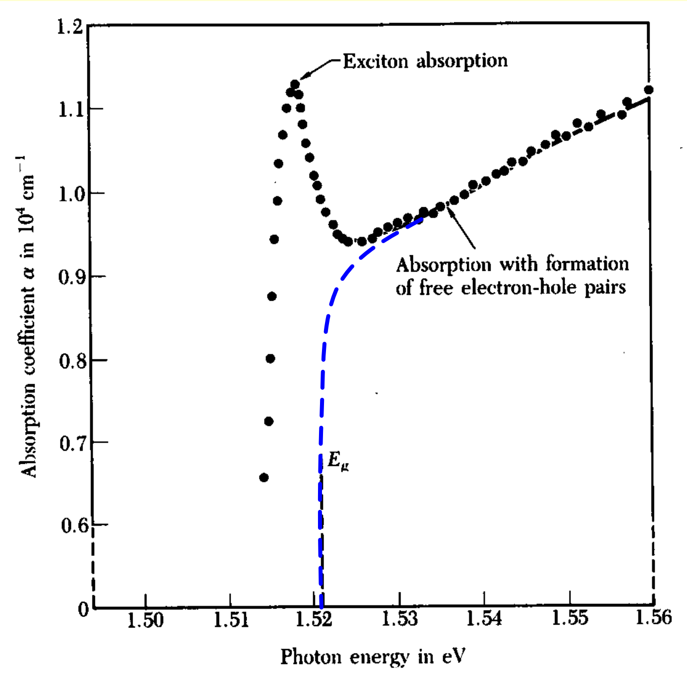


	Si	Ge	GaAs	InP	GaP
ε'	12.0	16.0	10.9	9.6	9.1
Eg (theo)	4.8	4.3	5.2	5.2	5.75
Ex (exp't)	4.44	4.49	5.11	5.05	5.21

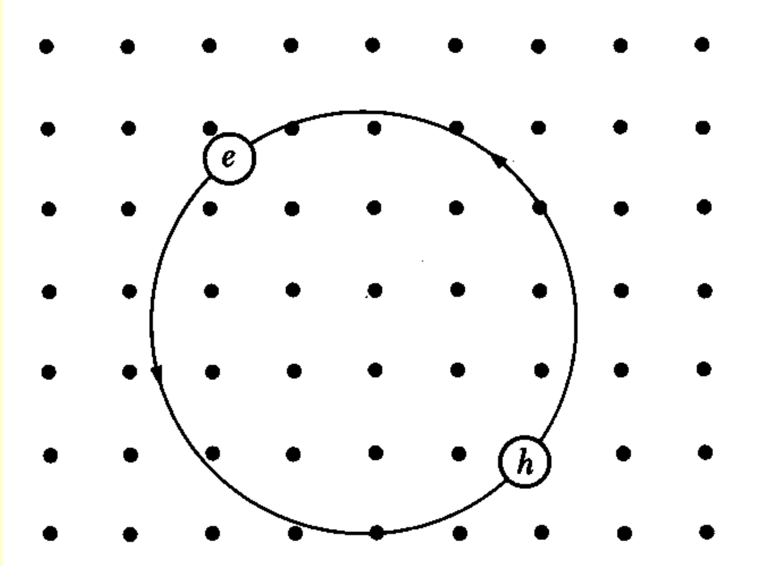
(ref: Cardona and Yu)

Figs from Ibach and Luth

Interband absorption for GaAs at 21 K



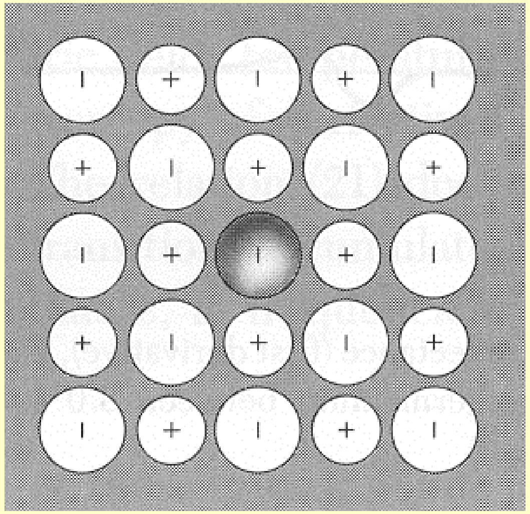
Excitons (e-h pairs) in insulators



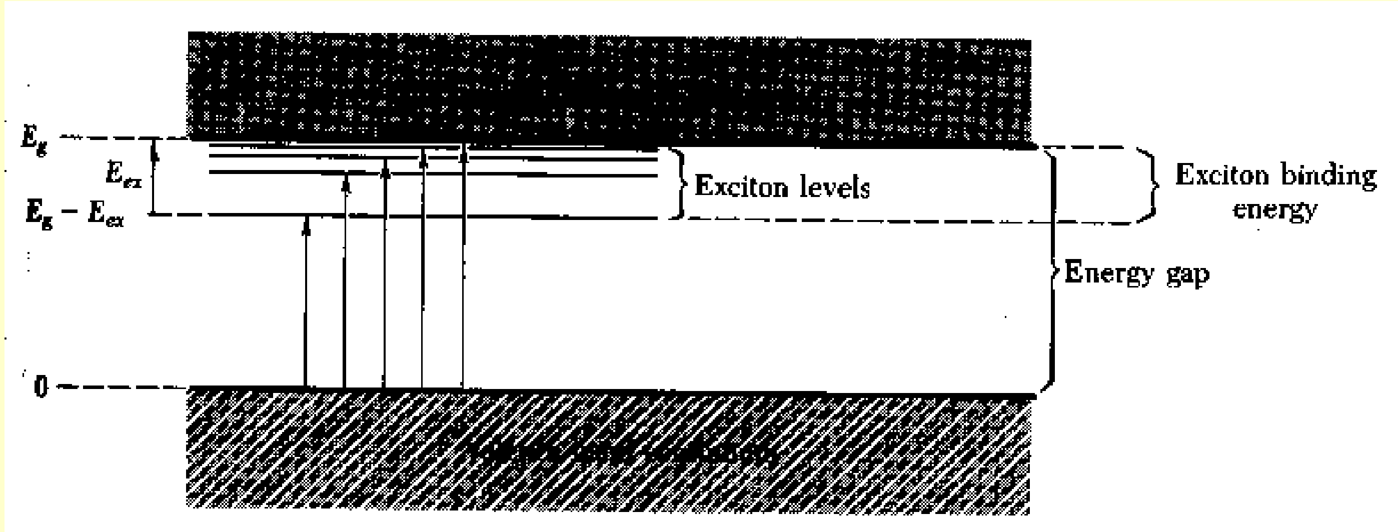
Tight bound Frenkel exciton:

(excited state of a single atom)

- alkali halide crystals
- crystalline inert gases

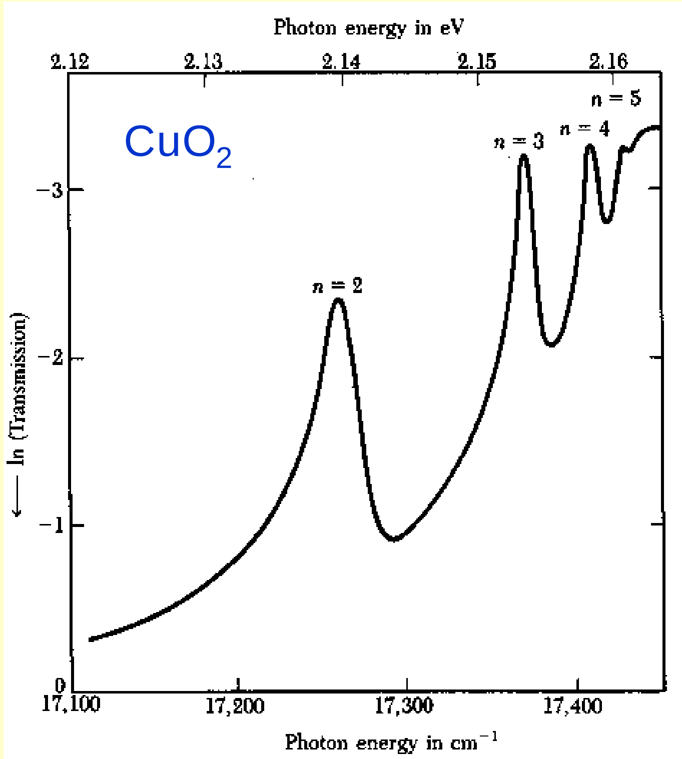


Weakly bound Mott-Wannier excitons

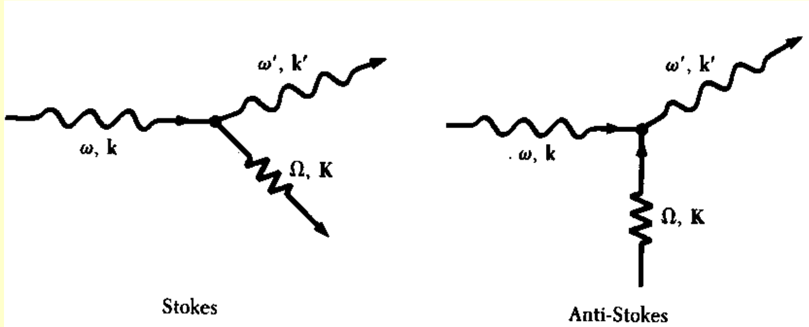


Similar to the impurity states in doped semiconductor:

$$E_n = E_g - \frac{me^4}{2\hbar^2 e^2 n^2}, \quad \frac{1}{m} = \frac{1}{m_e} + \frac{1}{m_h}$$



Raman effect in crystals



$$I(w-\Omega) \propto \left|\langle n_k+1|u|n_k\rangle\right|^2 \propto n_k+1$$

$$I(w+\Omega) \propto \left|\langle n_k-1|u|n_k\rangle\right|^2 \propto n_k$$

$$\frac{I(w+\Omega)}{I(w-\Omega)} = \frac{n_k}{n_k+1} = e^{-\hbar\Omega/k_B T}$$

1930