Linear and non-linear optical response of semiconductors

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Synopsis

1. Motivation : Exotic optical effects.
2. Formalism : Optical properties at microscopic level.
3. Examples : Superlattice.
4. Summary

Optics

What is Optics?

Optics can be defined as interaction of light with matter.

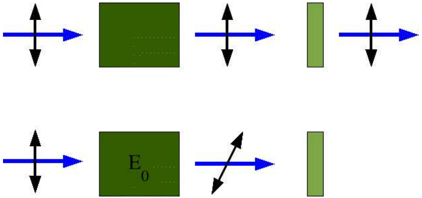
$$\vec{E} \cdot \vec{r} - \frac{\vec{v}}{c} \times \vec{B}$$

Motivation : Some exciting optical effects

Pockel's effect or Linear electro optic effect : 1883

Shutters

Switches

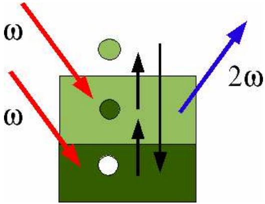


Motivation : Some exciting optical effects

Second Harmonic Generation : 1961

Blue lasers

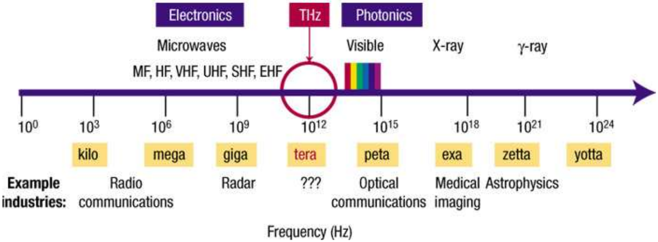
Data storage



Motivation : Some exciting optical effects

Optical rectification : 1964

THz production
Imaging



Formalism:

$$H|n\rangle = \epsilon_n|n\rangle$$

$$\rho = |\phi\rangle\langle\phi| = \sum_n |n\rangle\langle n|$$

$$\langle\vec{P}\rangle = Tr(\rho\vec{P})$$

Liouville equation:

$$i\frac{\delta\rho}{\delta t} = [H,\rho]$$

$$H = H_0 + H_1$$

$$\rho = \rho^0 + \rho^1 + \rho^2 + ...$$

$$i\frac{\delta}{\delta t}\left(\rho^0 + \rho^1 + \rho^2\right) = \left[H_0 + H_1, \rho^0 + \rho^1 + \rho^2\right]$$

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Linear optics

First order terms:

$$i\frac{\delta\rho^1}{\delta t} = \left[H_0, \rho^1\right] + \left[H_1, \rho^0\right]$$

$$i\frac{\delta}{\delta t}\left(\langle v|\rho^1|c\rangle\right) = \langle v|\left[H_0, \rho^1\right] + \left[H_1, \rho^0\right]|c\rangle$$

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Approximations:

1. Only the electronic system reacts.
2. Dipole approximation:

$$H_1 = \vec{E} \cdot \vec{r} - \left[\frac{\vec{v}}{c} \times \vec{B} \right]$$

3. Time dependence of perturbation, induced potential and hence density matrix:

$$\vec{E} = \vec{E}_0 \exp(-i\omega t + \eta t)$$

4. Steady state:

$$\langle \vec{P} \rangle = \langle \vec{P}^{(1)} \rangle + \langle \vec{P}^{(2)} \rangle + \dots$$

$$\vec{P}_i = \sum_j \chi^{(1)}_{ij}(\omega)\vec{E}_j(\omega) + \sum_{jk} \chi^{(2)}_{ijk}(\omega, 2\omega)\vec{E}_j(\omega)\vec{E}_k(\omega) + \dots$$

$$\langle \vec{P}^{(n)} \rangle = Tr(\rho^n \vec{P}^{(n)})$$

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$$i\frac{\delta \rho^1_{vc}}{\delta t} = (\epsilon_v - \epsilon_c)\rho^1_{vc} + (f_v - f_c)(H_1)_{vc}$$

First order susceptibility

$$\chi^{(1)}_{ij}(\omega) = \sum_{vc} \frac{(f_c - f_v)\mathbf{r}^i_{vc}\mathbf{r}^j_{cv}}{\epsilon_c - \epsilon_v - \omega + i\eta}$$

$$\vec{r}_{vc} = \frac{-\langle v|\vec{\nabla}|c\rangle}{\epsilon_{vc}}$$

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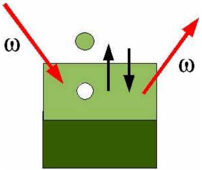
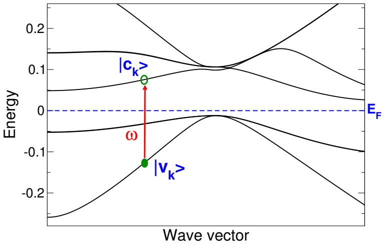
Schematic LO

Linear response of the material

- $y = ax$ and if $x = b\cos(\omega t)$ then $y = ab\cos(\omega t)$





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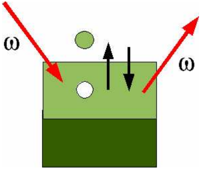
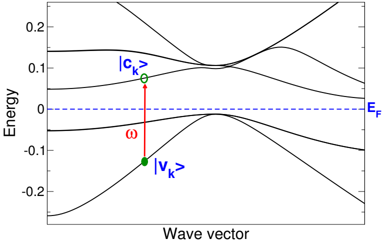
Linear response of the material

- $\chi^{(1)}_{ij}(\omega) = \sum_{vc} \frac{(f_c - f_v)\mathbf{r}^i_{vc}\mathbf{r}^j_{cv}}{\epsilon_c - \epsilon_v - \omega + i\eta}$





Nonlinear optics

Second order term:

$$\rho^2_{vc} = \frac{\left[H_1, \rho^1\right]_{vc}}{(\epsilon_v - \epsilon_c) - \omega_1 - \omega_2 + i\eta}$$

$$\chi^{(2)}_{ijk}(2\omega, \omega) = \frac{1}{\Omega} \sum_{vcl\mathbf{k}}' W_{\mathbf{k}} \left\{ \frac{2\mathbf{r}^i_{vc}\{\mathbf{r}^j_{cl}\mathbf{r}^k_{lv}\}}{(\omega_{lv} - \omega_{cl})(\omega_{cv} - 2\omega)} - \frac{1}{(\omega_{cv} - \omega)} \left[\frac{\mathbf{r}^k_{lc}\{\mathbf{r}^i_{cv}\mathbf{r}^j_{vl}\}}{(\omega_{vl} - \omega_{cv})} \frac{\mathbf{r}^j_{vl}\{\mathbf{r}^k_{lc}\mathbf{r}^i_{cv}\}}{(\omega_{lc} - \omega_{cv})} \right] \right\} +$$

Nonlinear optics

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$$\begin{aligned}\chi^{(2)}_{ijk}(2\omega,\omega) &= \frac{1}{\Omega}\sum_{vcl\mathbf{k}}{}' W_{\mathbf{k}} \left\{ \frac{2\mathbf{r}^i_{vc}\{\mathbf{r}^j_{cl}\mathbf{r}^k_{lv}\}}{(\omega_{lv}-\omega_{cl})(\omega_{cv}-2\omega)} \right. \\ &- \left. \frac{1}{(\omega_{cv}-\omega)}\left[\frac{\mathbf{r}^k_{lc}\{\mathbf{r}^i_{cv}\mathbf{r}^j_{vl}\}}{(\omega_{vl}-\omega_{cv})} \frac{\mathbf{r}^j_{vl}\{\mathbf{r}^k_{lc}\mathbf{r}^i_{cv}\}}{(\omega_{lc}-\omega_{cv})}\right]\right\} +\end{aligned}$$

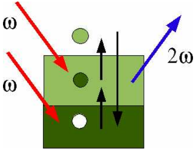
Nonlinear optics

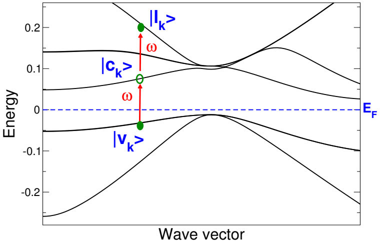
$$
\begin{aligned}
+ \quad & \frac{1}{\Omega}\sum_{\mathbf{k}} W_{\mathbf{k}} \left\{ \sum_{vcl}{}^{\prime} \frac{\omega_{cv}^{-2}}{(\omega_{cv}-\omega)} \right. \\
& \left[\omega_{lv} \mathbf{r}^{j}_{vl} \{ \mathbf{r}^{k}_{lc} \mathbf{r}^{i}_{cv} \} - \omega_{cl} \mathbf{r}^{k}_{lc} \{ \mathbf{r}^{i}_{cv} \mathbf{r}^{j}_{vl} \} \right] + \\
& \left. \sum_{vc}{}^{\prime} \frac{\mathbf{r}^{i}_{vc} \{ \mathbf{r}^{j}_{cl} \mathbf{r}^{k}_{lv} \}}{\omega_{cv}^{2}(\omega_{cv}-2\omega)} \left[-8i + 2 \sum_{l}{}^{\prime} (\omega_{cl}-\omega_{lv}) \right] \right\} \\
+ \quad & \frac{1}{2\Omega}\sum_{\mathbf{k}} W_{\mathbf{k}} \left\{ \sum_{vcl} \frac{1}{\omega_{cv}^{2}(\omega_{cv}-\omega)} \right. \\
& \left. \left[\omega_{vl} \mathbf{r}^{i}_{lc} \{ \mathbf{r}^{j}_{cv} \mathbf{r}^{k}_{vl} \} - \omega_{lc} \mathbf{r}^{i}_{vl} \{ \mathbf{r}^{j}_{lc} \mathbf{r}^{k}_{cv} \} \right] - i \sum_{vc} \frac{\mathbf{r}^{i}_{vc} \{ \mathbf{r}^{j}_{cv} \mathbf{\Delta}^{k}_{cv} \}}{\omega_{mn}^{2}(\omega_{mn}-\omega)} \right\}
\end{aligned}
$$

Schematic NLO

Nonlinear response

$y = ax + cx^2$ and if $x = b\cos(\omega t)$ then $y = ab\cos(\omega t) + \frac{1}{2}bc\left[\cos(2\omega t) + 1\right]$
since $\cos^2(\theta) = \frac{1}{2}\left[\cos(2\theta) + 1\right]$





Some exciting optical effects

Optical effects in terms of microscopic quantities.

1. Second harmonic generation:

$$\chi^{(2)} \; \vec{E}_\omega^2 \; \cos(2\omega t)$$

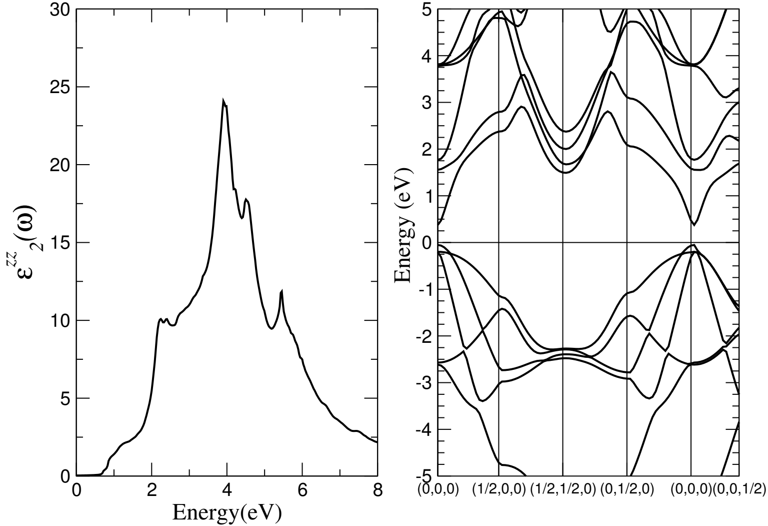
2. Pockel's effect:

$$\chi^{(2)} \, \vec{E}_0 \; \vec{E}_\omega \; \cos(\omega t)$$

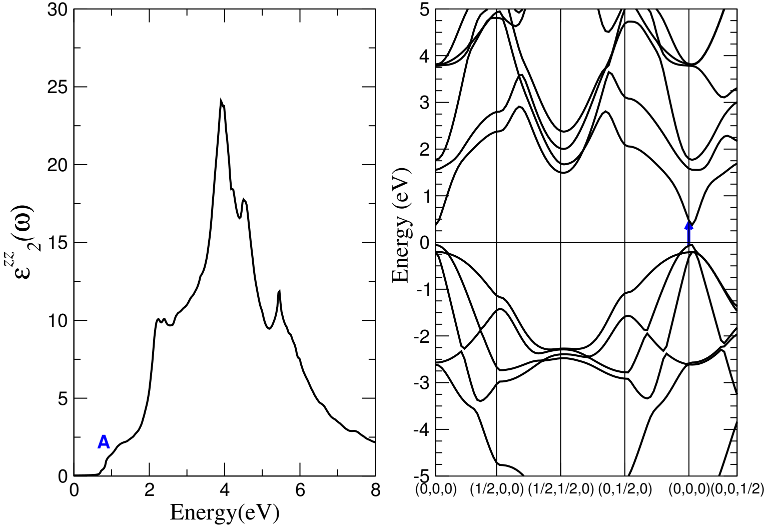
3. Optical rectification:

$$\chi^{(2)} \, \vec{E}_\omega^2$$

LO: dielectric function for InP/GaP(110)



Feature identification from bandstructure: InP/GaP(110)



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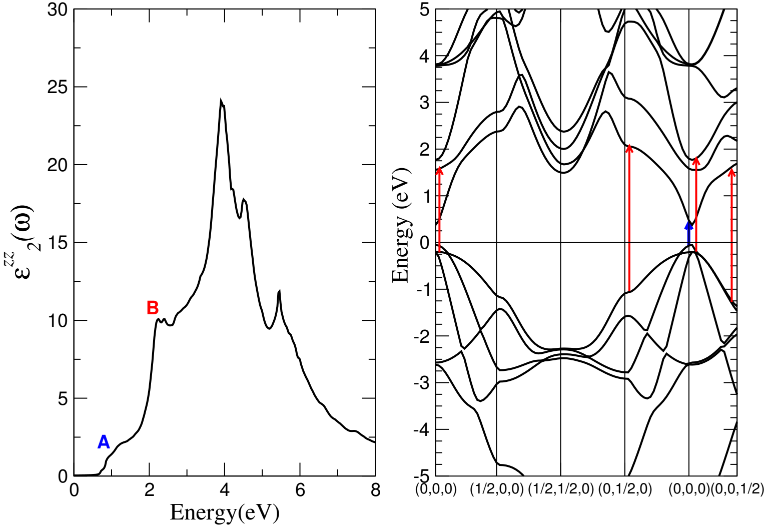

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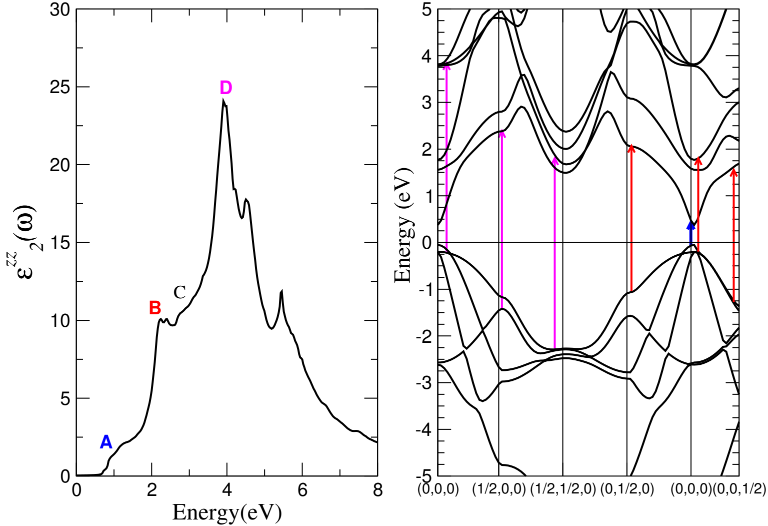

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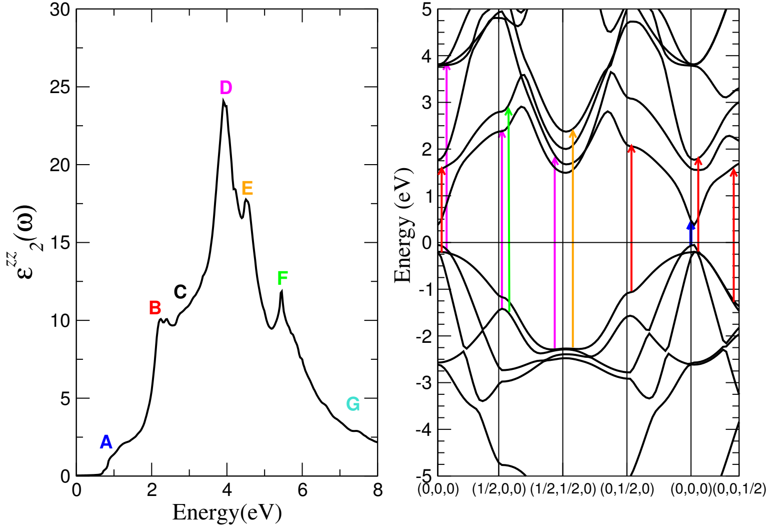
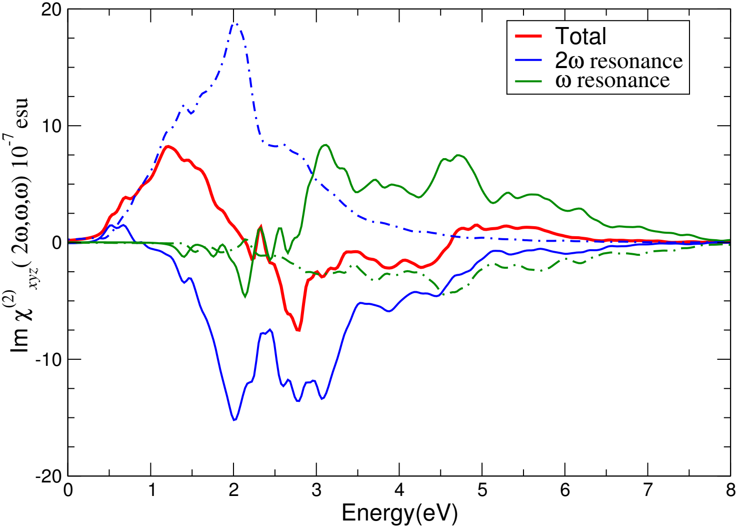


NLO: Second harmonic generation by InP/GaP(110)



Symmetry sensitivity of NLO

Non-linear optics for centro-symmetric system

$$\vec{P}_i^{(2)}(\omega) = \chi_{ijk}^{(2)}(2\omega, \omega)\vec{E}_j(\omega)\vec{E}_k(\omega)$$

$$-\vec{P}_i^{(2)}(\omega) = \chi_{ijk}^{(2)}(2\omega, \omega)(-\vec{E}_j(\omega))(-\vec{E}_k(\omega))$$

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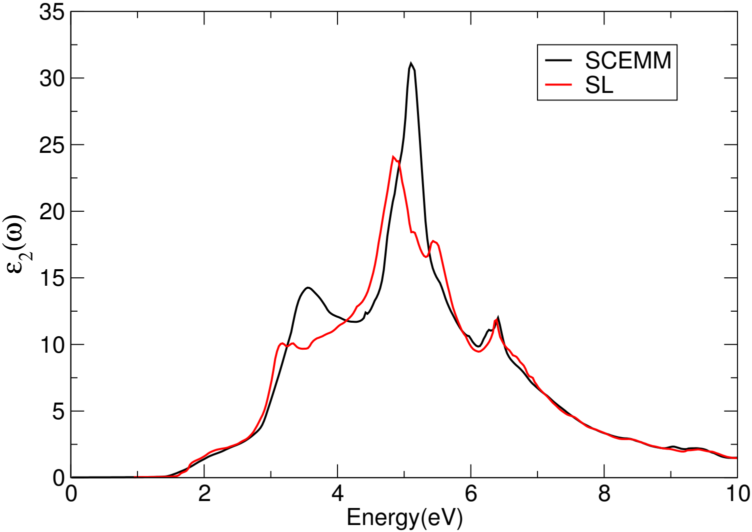
Symmetry sensitivity of NLO: SHG by InP/GaP(110)



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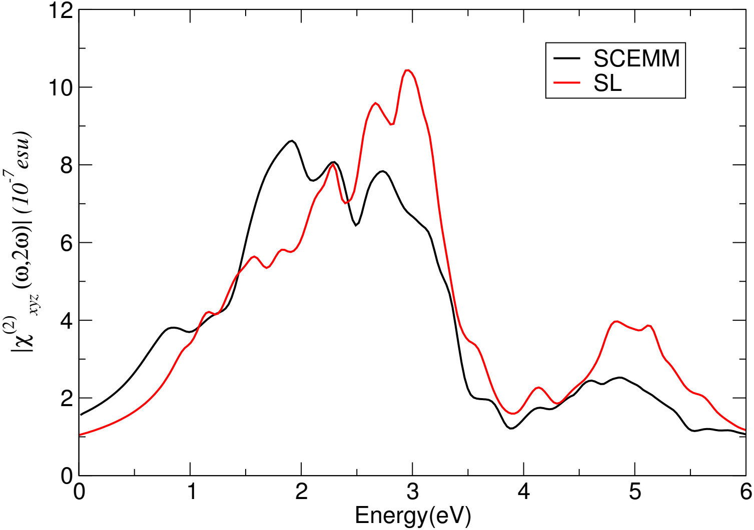

Non inclusions

Some more approximations:

1. Single particle KS spectrum
2. No excitonic effects

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Summary

The talk established

1. Link between microscopic properties of material and exotic optical effects.
2. Sensitivity of SHG to interface / surface.
3. Optics is an excellent tool for material characterization

References

Material for further reading

1. Optical Properties of Solids by F. Wooten
2. The Principles of Nonlinear Optics by Y. R. Shen
3. S. Sharma and C. Ambrosch-Draxl, Physica Scripta T109 128 (2004) or at /cond-mat/0305016
4. S. Sharma et al., Phys. Rev. B 68 014111 (2003)
5. S. Sharma et al., Phys. Rev. B 67 165332 (2003)

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Visit: UCSB