STRONGLY CORRELATED COLLECTIVE EXCITATIONS IN PLANAR TRANSDIMENSIONAL NANOSTRUCTURES

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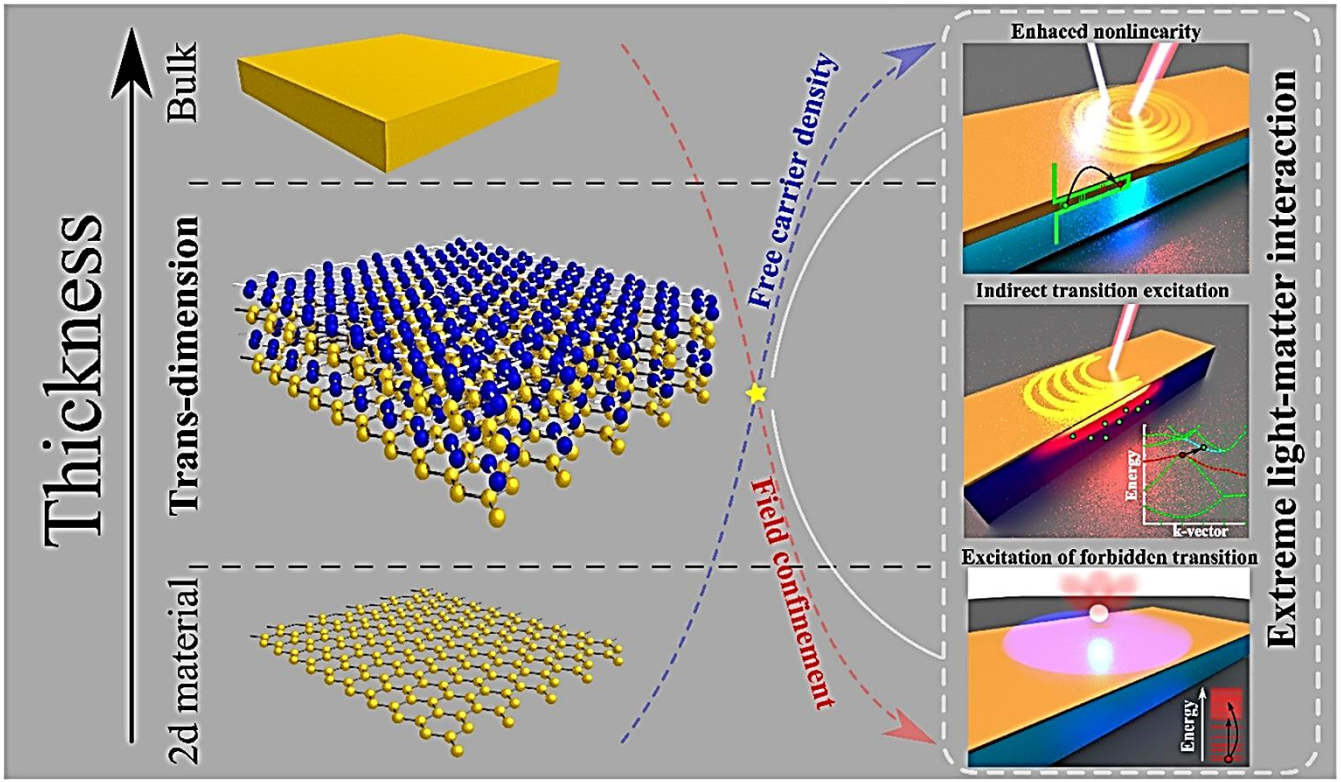
TRANSDIMENSIONAL QUANTUM MATERIALS

Ultrathin Material Films of Controlled Finite Number of Atomic Monolayers (finite thickness)



Schematics. *A.Boltasseva and V.M.Shalaev, ACS Photonics* 6, 1-3 (2019)

DOI: (10.1021/acsphotonics.8b01570)

WHY DO WE EVEN NEED QUANTUM ELECTRODYNAMICS ??

- **Depending on their material composition and thickness, metasurfaces can restructure the spectral and spatial distribution of both real and *vacuum* EM modes. While real modes can still be described semiclassically, the physical consequences of the *vacuum* EM mode restructuring can only be fully understood within the framework of medium-assisted quantum electrodynamics (QED)**
- **The standard (vacuum) QED field quantization scheme fails to work in presence of dispersing and absorbing media because the presence of absorption makes the operator Maxwell equations non-Hermitian. As a consequence, their solutions cannot be expanded in power orthogonal modes — strictly speaking — and the concept of modes itself becomes more subtle (or fragile…)**
- **EM field quantization (QED) is necessary for the correct description of the light-matter interaction scenarios with *virtual* (vacuum) photon excitations involved such as spontaneous emission, van der Waals interactions and such, which are mediated essentially by the *virtual* photon exchange**

The medium-assisted QED formalism (Schrödinger picture, Gaussian units):

geometry

$$\hat{E}_\alpha(\mathbf{r},\omega) = i\frac{4\pi\omega^2}{c^2}\int d\mathbf{r}' \sum_{\lambda=x,y,z} G_{\alpha\lambda}(\mathbf{r},\mathbf{r}',\omega)\hat{J}_\lambda(\mathbf{r}',\omega) + \text{h.c.}, \quad \hat{H}_\alpha(\mathbf{r},\omega) = -i\frac{c}{\omega}\sum_{\beta,\gamma=x,y,z}\varepsilon_{\alpha\beta\gamma}\nabla_\beta \hat{E}_\gamma(\mathbf{r},\omega)$$

medium

$$\hat{J}_\lambda(\mathbf{r},\omega) = \sqrt{\frac{\text{Im}\,\varepsilon_{\lambda\lambda}(\mathbf{r},\omega)}{\pi}}\,\hat{f}_\lambda(\mathbf{r},\omega), \quad [\hat{f}_\lambda(\mathbf{r},\omega), \hat{f}^\dagger_{\lambda'}(\mathbf{r}',\omega')] = \delta_{\lambda\lambda'}\,\delta(\mathbf{r}-\mathbf{r}')\,\delta(\omega-\omega')$$

$$[\hat{E}_x(\mathbf{r},t), \hat{H}_y(\mathbf{r}',t)] = i4\pi c\hbar\frac{\partial}{\partial z'}\delta(\mathbf{r}-\mathbf{r}'), \quad \&\,(x,y,z)\ \textit{cyclic permutations}$$

$$[\hat{E}_\alpha(\mathbf{r},t), \hat{H}_\alpha(\mathbf{r}',t)] = [\hat{E}_\alpha(\mathbf{r},t), \hat{H}_\alpha(\mathbf{r}',t)] = [\hat{E}_\alpha(\mathbf{r},t), \hat{H}_\alpha(\mathbf{r}',t)] = 0$$

W.Vogel & D.-G.Welsch, *Quantum Optics*, 3rd edn., Ch.10 (Wiley-VCH, 2006)

- **THERE IS STILL ONE MORE IMPORTANT INGREDIENT TO ADD IN: For thin films, the Coulomb field produced by the confined charges *outside* of their confinement region starts playing a perceptible role with the thickness reduction — a solely confinement related effect having nothing to do with the metasurface material composition.**

OUTLINE

- **Ultrathin Plasmonic Films of Finite Thickness: *Plasma Frequency Spatial Dispersion and Magneto-Optical Response***
- **Interlayer (Indirect) Excitons and Exciton Complexes in Layered Semiconductors: *Binding Energies within the Configuration Space Approach***
- **Organic Molecular Semiconductor Crystals with *Two* Isolated Frenkel Exciton States: *Frenkel-Charge-Transfer Exciton Intermixing***
- **Summary**

EFFECTS OF QUANTUM CONFINEMENT IN THE TRANSDIMENSIONAL REGIME

Keldysh-Rytova Potential

Coulomb interaction in thin semiconductor and semimetal films

L. V. Keldysh
P. N. Lebedev Physics Institute, USSR Academy of Sciences

(Submitted 28 April 1979)

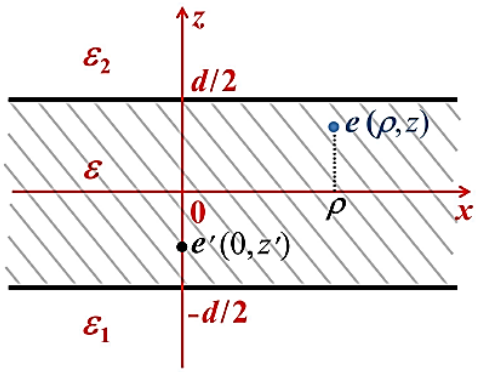
L.V.Keldysh, JETP Lett. 29, 658 (1980)
N.S.Rytova, Moscow State University Physics Bulletin 3, 30 (1967)

$$V(\vec{\rho}, z, z') = \frac{4\pi e e'}{\epsilon} \int \frac{d^2k}{(2\pi)^2} e^{i\vec{k}\vec{\rho}} \frac{\mathrm{ch}\left[|k|\left(\frac{d}{2} - z\right) + \eta_2\right] \mathrm{ch}\left[|k|\left(\frac{d}{2} + z'\right) + \eta_1\right]}{|k|\,\mathrm{sh}[|k|\,d + \eta_1 + \eta_2]}$$

$$V(\rho) \approx V_2(\rho) = \frac{2ee'}{(\varepsilon_1+\varepsilon_2)\rho}, \quad \text{if} \quad \frac{\rho}{d} \gg \frac{\varepsilon}{\varepsilon_1+\varepsilon_2} \gg 1$$

$$\eta_{1,2} = \frac{1}{2} \ln \frac{\epsilon + \epsilon_{1,2}}{\epsilon - \epsilon_{1,2}} .$$

$$V(\rho) \approx V_1(\rho) = \frac{2ee'}{\varepsilon d}\left[\ln\left(\frac{2\varepsilon}{\varepsilon_1+\varepsilon_2}\frac{d}{\rho}\right) - C\right], \quad \text{if} \quad \frac{\varepsilon}{\varepsilon_1+\varepsilon_2} \gg \frac{\rho}{d} \gg 1$$

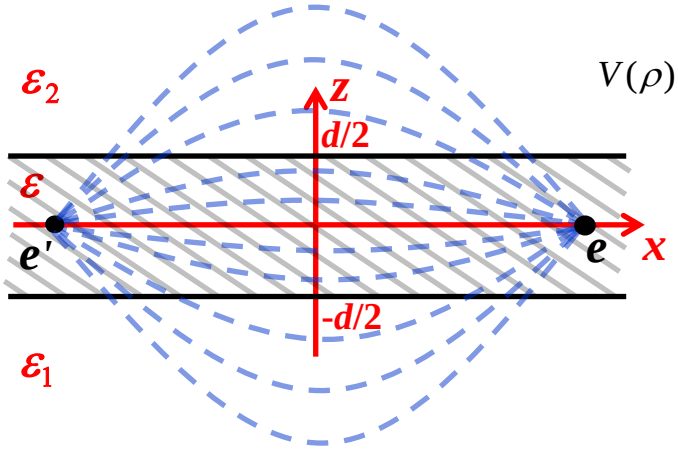


$$V(\vec{\rho}) = \frac{2ee'}{\epsilon d} \int_0^\infty \frac{J_0(t)\,dt}{t + \frac{\epsilon_1+\epsilon_2}{\epsilon}\frac{\rho}{d}}$$

$$d \ll \rho \quad = \quad \frac{\pi e e'}{\epsilon d}\left[\mathcal{H}_0\left(\frac{\epsilon_1+\epsilon_2}{\epsilon}\frac{\rho}{d}\right) - N_0\left(\frac{\epsilon_1+\epsilon_2}{\epsilon}\frac{\rho}{d}\right)\right],$$

PLASMA FREQUENCY SPATIAL DISPERSION AND NONLOCAL OPTICAL RESPONSE OF ULTRATHIN PLASMONIC FILMS

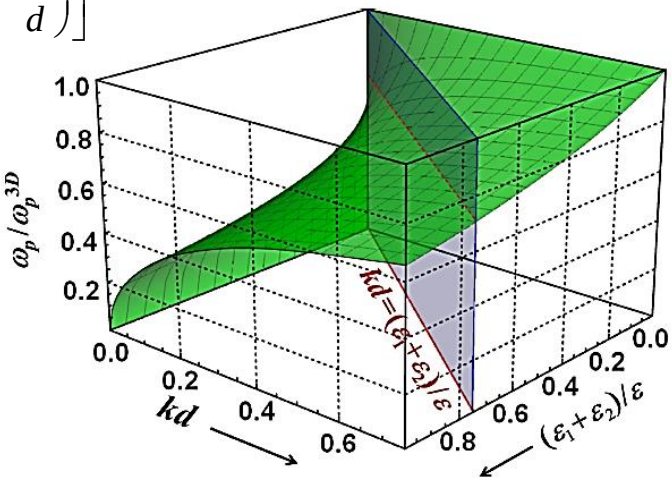
Momentum Space



$$V(\rho)=\frac{\pi ee'}{\varepsilon d}\left[H_0\left(\frac{\varepsilon_1+\varepsilon_2}{\varepsilon}\frac{\rho}{d}\right)-N_0\left(\frac{\varepsilon_1+\varepsilon_2}{\varepsilon}\frac{\rho}{d}\right)\right]$$

$$V(k)\neq\frac{4\pi ee'}{k^2}$$

$$V(k)=\frac{4\pi ee'}{k\left(kd+\dfrac{\varepsilon_1+\varepsilon_2}{\varepsilon}\right)}$$



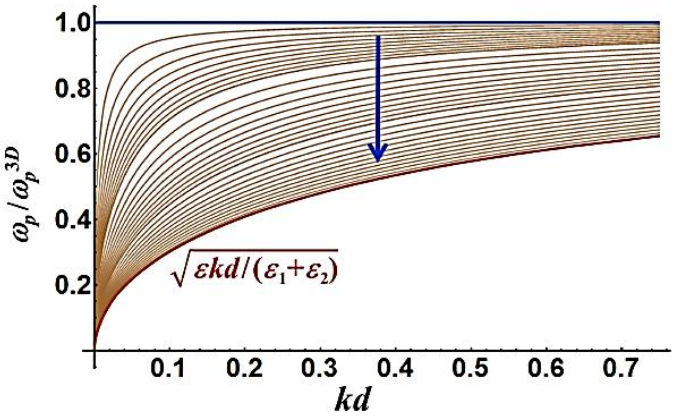
For thin enough plasmonic films, one has $N_{3D}d=N_{2D}$, so that Eq. (9) can be written as

$$\omega_p=\omega_p(k)=\frac{\omega_p^{3D}}{\sqrt{1+(\varepsilon_1+\varepsilon_2)/\varepsilon kd}}. \qquad (11)$$

If $(\varepsilon_1+\varepsilon_2)/\varepsilon kd\ll 1$ (relatively thick film), then $\omega_p=\omega_p^{3D}$ of Eq. (10), whereas one has

$$\omega_p=\omega_p^{2D}(k)=\sqrt{\frac{4\pi e^2N_{2D}k}{(\varepsilon_1+\varepsilon_2)m^*}} \qquad (12)$$

if $(\varepsilon_1+\varepsilon_2)/\varepsilon kd\gg 1$ (ultrathin film), which agrees precisely with the plasma frequency spatial dispersion of the 2D electron gas in air (see, e.g., Ref. [33]), but does show the explicit dependence on bottom (ε_1) and top (ε_2) surrounding materials.



$$\frac{\varepsilon(k,\omega)}{\varepsilon}=1-\frac{\omega_p^2}{\omega(\omega+i\gamma)}$$

Confinement-Induced Optical Response Nonlocality

I.V.Bondarev and V.M.Shalaev, Opt. Mater. Express 7, 3731 (2017)

PLASMA FREQUENCY SPATIAL DISPERSION AND NONLOCAL OPTICAL RESPONSE OF ULTRATHIN PLASMONIC FILMS

Coordinate Space

$$\frac{\varepsilon(k,\omega)}{\varepsilon}=1-\left(\frac{\omega_p^{3D}}{\sqrt{1+(\varepsilon_1+\varepsilon_2)/\varepsilon kd}}\right)^2\frac{1}{\omega(\omega+i\gamma)}$$

$$\Downarrow$$

$$(per\ unit\ area)$$

$$\Downarrow$$

$$\frac{\varepsilon(\boldsymbol{\rho},\boldsymbol{\rho}',\omega)}{\varepsilon}=\delta(|\boldsymbol{\rho}-\boldsymbol{\rho}'|)-\frac{1}{2\pi}\int_{2\pi/L}^{k_c}dk\,k\left(\frac{\omega_p^{3D}}{\sqrt{1+(\varepsilon_1+\varepsilon_2)/\varepsilon kd}}\right)^2\frac{e^{ik|\boldsymbol{\rho}-\boldsymbol{\rho}'|\cos\theta}}{\omega(\omega+i\gamma)}$$

$$\Downarrow \qquad\qquad \Downarrow$$

$$L\to\infty,\ (\varepsilon_1+\varepsilon_2)/\varepsilon kd<<1 \qquad\qquad L\to\infty,\ (\varepsilon_1+\varepsilon_2)/\varepsilon kd>>1,\ k_c|\boldsymbol{\rho}-\boldsymbol{\rho}'|>>1$$

$$\Downarrow \qquad\qquad \Downarrow$$

$$\frac{\varepsilon(\boldsymbol{\rho},\boldsymbol{\rho}',\omega)}{\varepsilon}=\left(1-\frac{\left(\omega_p^{3D}\right)^2}{\omega(\omega+i\gamma)}\right)\delta(|\boldsymbol{\rho}-\boldsymbol{\rho}'|)$$

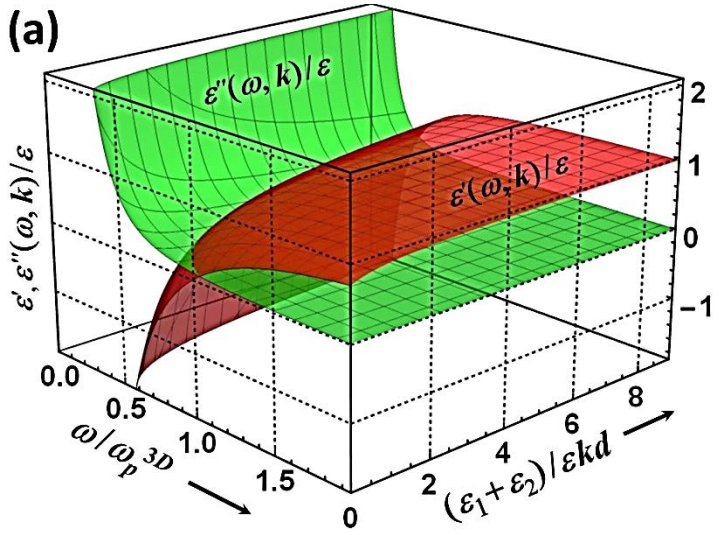
$$\frac{\varepsilon(\boldsymbol{\rho},\boldsymbol{\rho}',\omega)}{\varepsilon}\approx\left(1-\frac{k_cd}{\varepsilon_1+\varepsilon_2}\cdot\frac{\left(\omega_p^{3D}\right)^2}{\omega(\omega+i\gamma)}\right)\delta(|\boldsymbol{\rho}-\boldsymbol{\rho}'|)$$

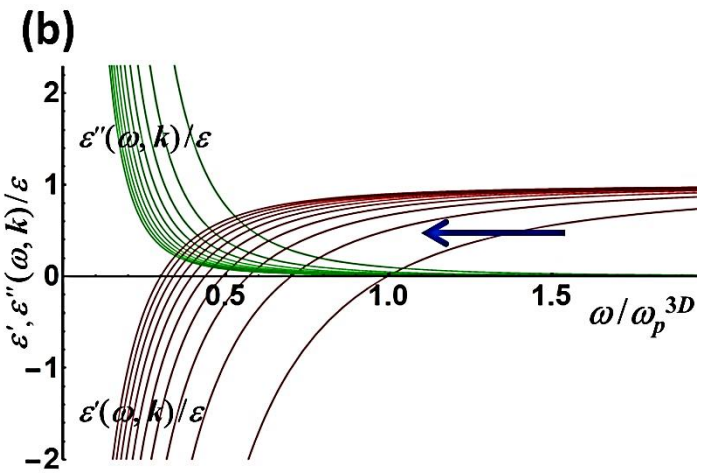
$$+\frac{k_cd}{\varepsilon_1+\varepsilon_2}\cdot\frac{\left(\omega_p^{3D}\right)^2}{\omega(\omega+i\gamma)}\cdot\frac{\cos\left(k_c|\boldsymbol{\rho}-\boldsymbol{\rho}'|+3\pi/4\right)}{\sqrt{2\pi^3k_c}\,|\boldsymbol{\rho}-\boldsymbol{\rho}'|^{5/2}}$$

Optical Response Nonlocality in coordinate space

I.V.Bondarev, H.Mousavi, & V.M.Shalaev, MRS Commun. 8, 1092 (2018)

PLASMA FREQUENCY SPATIAL DISPERSION AND NONLOCAL OPTICAL RESPONSE OF ULTRATHIN PLASMONIC FILMS

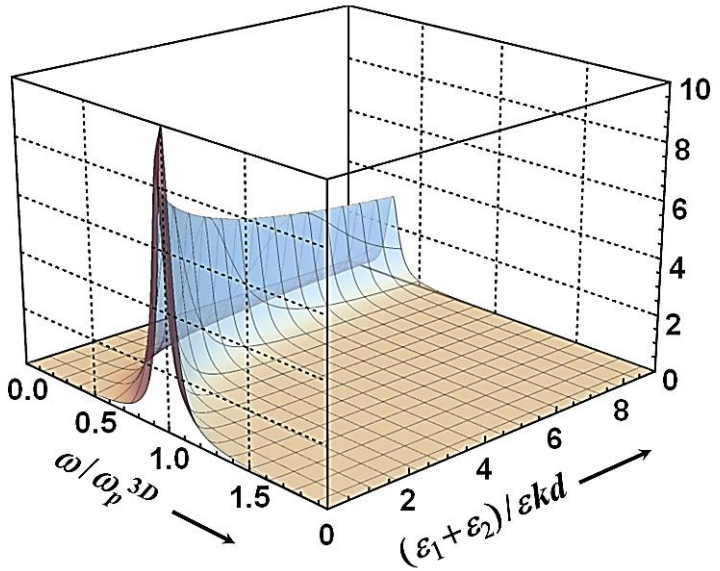




$$\frac{\varepsilon(k,\omega)}{\varepsilon} = 1 - \frac{\omega_p^2}{\omega(\omega + i\gamma)}$$

$$\gamma/\omega_p^{3D} = 0.1$$

$-\varepsilon\varepsilon''(\omega,k)/[\varepsilon'(\omega,k)^2+\varepsilon''(\omega,k)^2]$

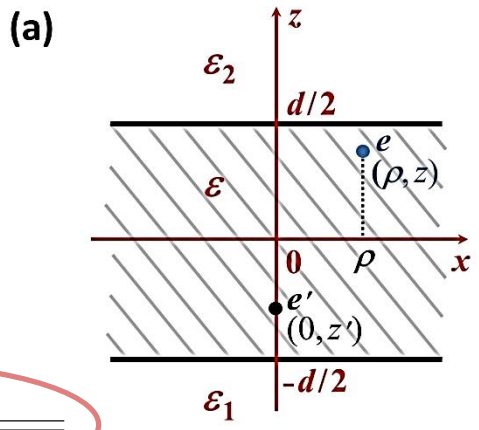


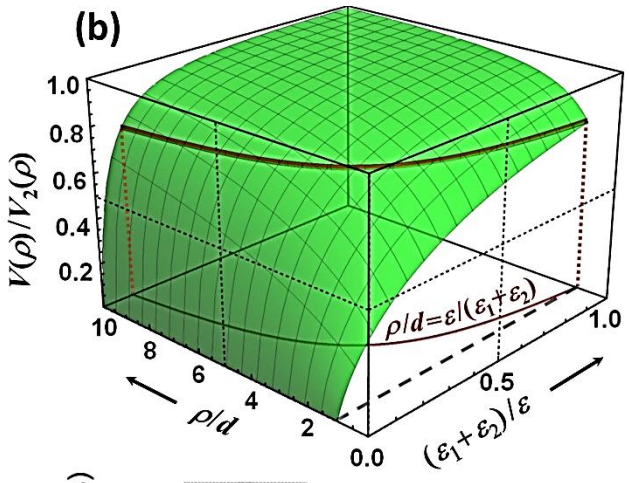
I.V.Bondarev and V.M.Shalaev, Opt. Mater. Expr. 7, 3731 (2017)

ULTRATHIN PLASMONIC FILMS OF FINITE THICKNESS. EFFECTS OF QUANTUM CONFINEMENT

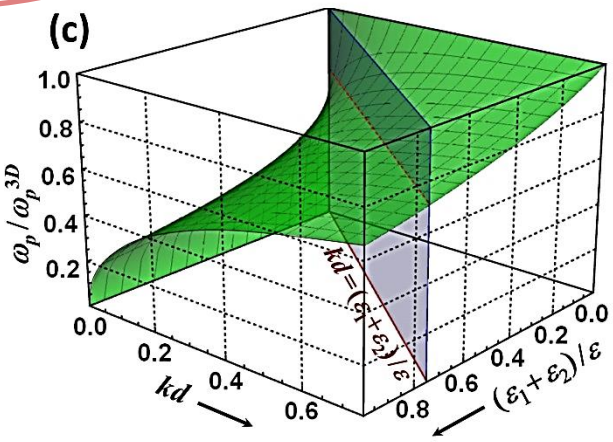
Comparison with Experiments

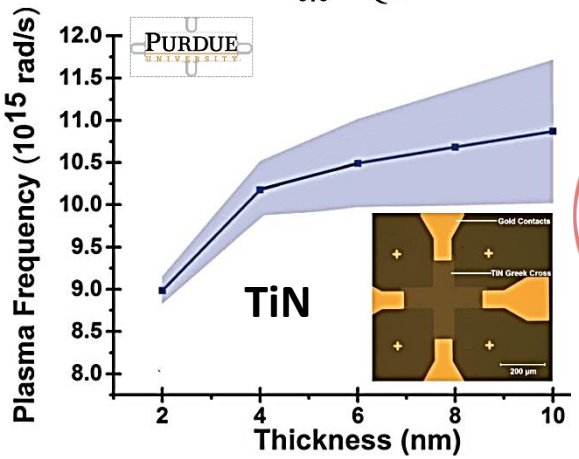
$$\omega_p = \omega_p(k) = \frac{\omega_p^{3D}}{\sqrt{1+(\varepsilon_1+\varepsilon_2)/\varepsilon kd}}$$





$$V_2(\rho) = \frac{2ee'}{(\varepsilon_1+\varepsilon_2)\rho}$$





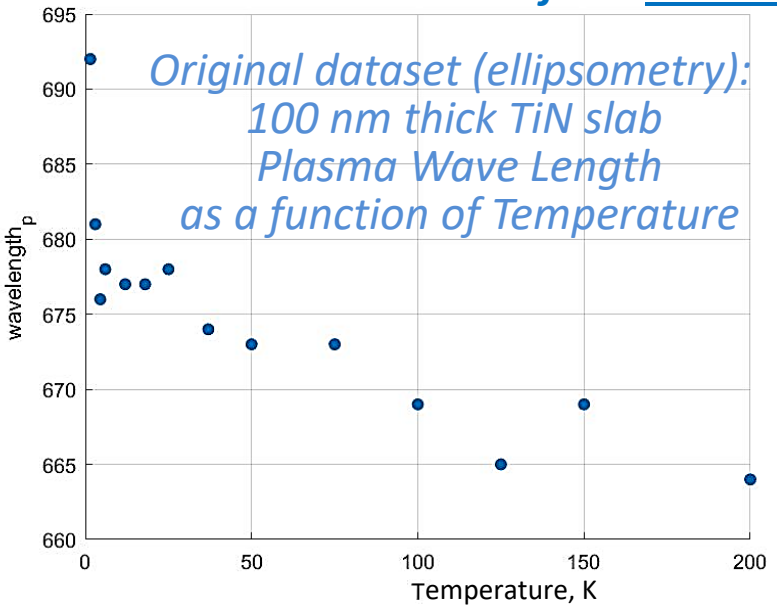
Same effect in topological insulator thin films:
T.P.Ginley & S.Law, Adv. Optical Mater. 1800113 (2018)

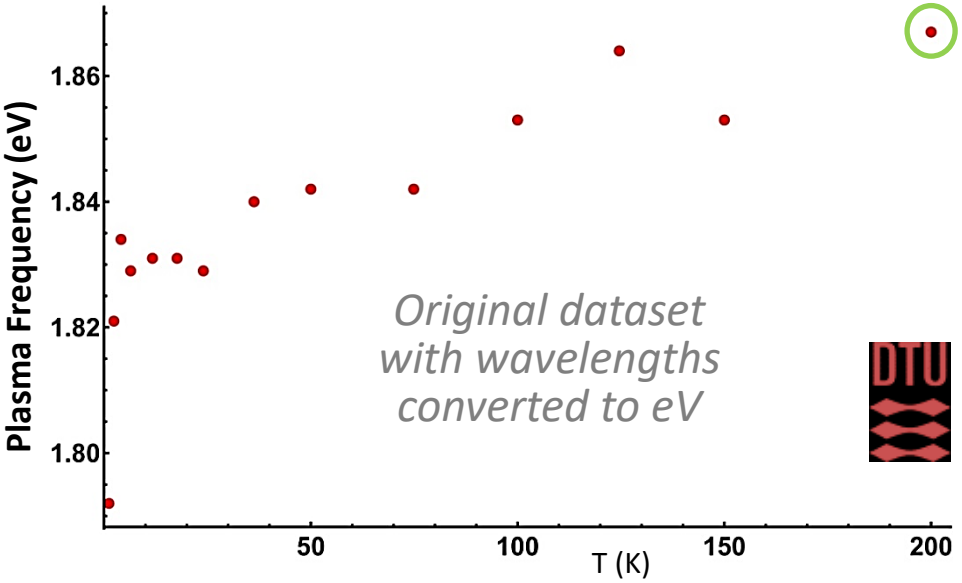
(a,b) Schematic of the confined thin film geometry (a) and the normalized Coulomb interaction potential (b) for ultrathin finite thickness plasmonic films. **(c)** Thin film plasma frequency normalized by the bulk plasma frequency derived theoretically in Ref.[1]. **(d)** Plasma frequency extracted from the ellipsometry measurements done on ultrathin TiN films of controlled variable thickness fabricated at Purdue University [2].

[1] I.V.Bondarev and V.M.Shalaev, Opt. Mater. Express 7, 3731 (2017); [2] D.Shah, *et al.*, Adv. Opt. Mater. 1700065 (2017)

[3] D.Shah, M.Yang, Z.Kudyshev, X.Xu, V.M.Shalaev, I.V.Bondarev, and A.Boltasseva, Nano Lett. 22, 4622 (2022)

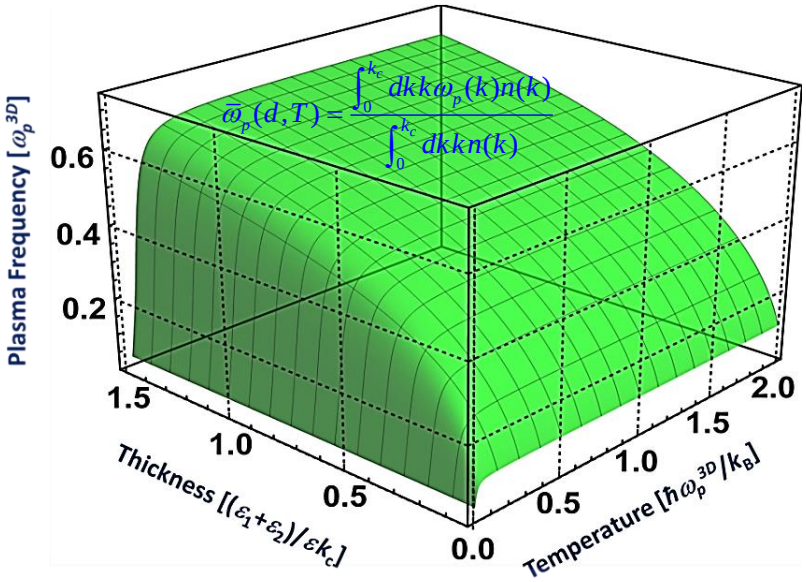
~~~ RAW DATA of the DTU Photonik group (Lavrinenko), Denmark ~~~

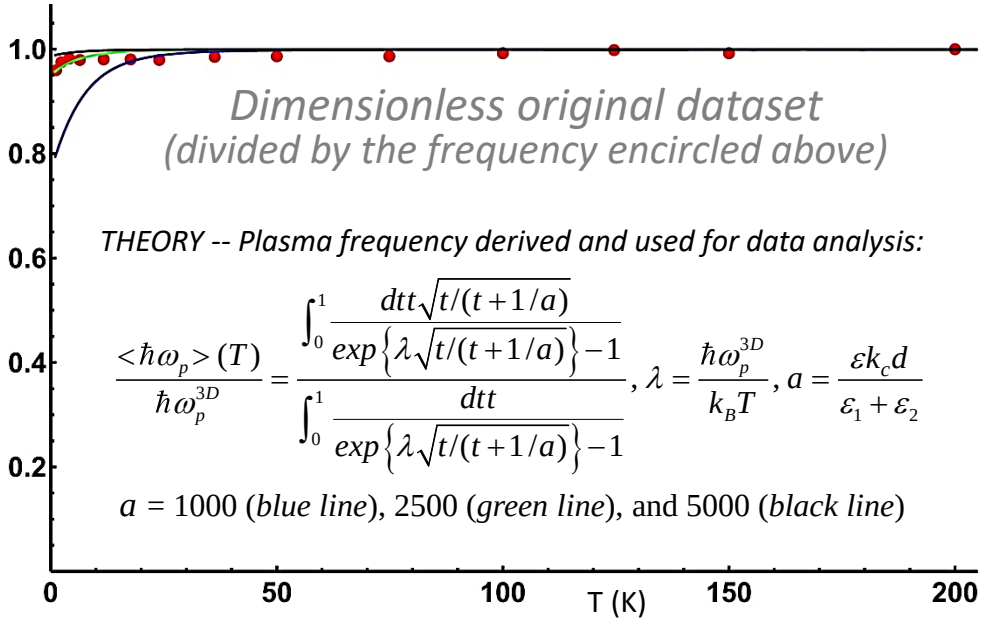




~~~ THEORETICAL ANALYSIS DONE @ NCCU (Bondarev), USA ~~~

THEORY -- General behavior of the plasma frequency as a function of temperature (T) and film thickness (d)





THEORY -- Plasma frequency derived and used for data analysis:

$$\frac{<\hbar\omega_p>(T)}{\hbar\omega_p^{3D}} = \frac{\int_0^1 \frac{dtt\sqrt{t/(t+1/a)}}{exp\left\{\lambda\sqrt{t/(t+1/a)}\right\}-1}}{\int_0^1 \frac{dtt}{exp\left\{\lambda\sqrt{t/(t+1/a)}\right\}-1}}, \lambda = \frac{\hbar\omega_p^{3D}}{k_B T}, a = \frac{\varepsilon k_c d}{\varepsilon_1+\varepsilon_2}$$

a = 1000 (*blue line*), 2500 (*green line*), and 5000 (*black line*)

L.Vertchenko, L.Leandro, E.Shkondin, O.Takayama, I.V.Bondarev, N.Akopian & A.V.Lavrinenko, Opt. Mater. Express 9, 2117 (2019)

PLASMA FREQUENCY SPATIAL DISPERSION & MAGNETO-OPTICAL RESPONSE OF ULTRATHIN PLASMONIC FILMS

L.D.Landau & E.M.Lifshitz, *Electrodynamics of Continuous Media*, 2nd edn., 1984

When spatial dispersion is present, the permittivity is a tensor, not a scalar, even in isotropic medium: a distinctive direction is generated by the wave vector.

$$\varepsilon_{ik}(\omega, \mathbf{k}) = \varepsilon_t(\omega, k)(\delta_{ik} - k_i k_k/k^2) + \varepsilon_l(\omega, k) k_i k_k/k^2,$$

where ε_t and ε_l depend only on the magnitude of the wave vector (and on ω).

$$\varepsilon_l(\omega, 0) = \varepsilon_t(\omega, 0) = \varepsilon(\omega).$$

PROBLEM 1. Find the relation between the functions $\varepsilon(\omega)$, $\mu(\omega)$ and the limiting values of $\varepsilon_l(\omega, k)$ and $\varepsilon_t(\omega, k)$ as $k \to 0$.

SOLUTION. We compare the expressions for the averaged microscopic current $\overline{\rho \mathbf{v}}$ in the forms (103.5) and (79.3). For a monochromatic field, we have in the first case

$$\overline{\rho v_i} = -i\omega[\varepsilon_{ik}(\omega, \mathbf{k}) - \delta_{ik}] E_k/4\pi,$$

and in the second case

$$\overline{\rho \mathbf{v}} = -i\omega[\varepsilon(\omega) - 1]\mathbf{E}/4\pi + ic[\mu(\omega) - 1]\mathbf{k} \times \mathbf{H}/4\pi.$$

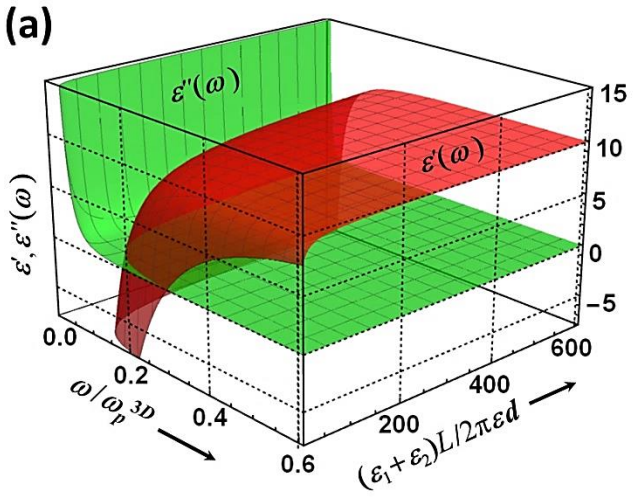
Substituting in the first $\varepsilon_{ik}(\omega, \mathbf{k})$ from (103.12), and in the second $\mathbf{H} = c\mathbf{k} \times \mathbf{E}/\omega\mu$ in accordance with Maxwell's equation, and equating the two expressions (for $k \to 0$), we find

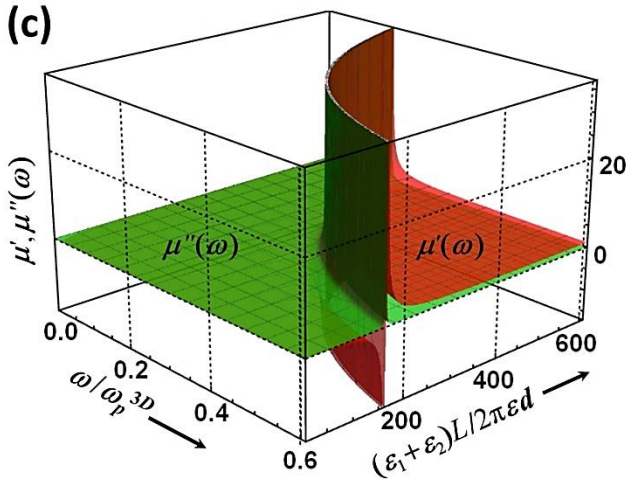
$$1 - \frac{1}{\mu(\omega)} = \frac{\omega^2}{c^2} \lim_{k \to 0} \frac{\varepsilon_t(\omega, k) - \varepsilon_l(\omega, k)}{k^2}$$

by comparing the terms in $\mathbf{k}(\mathbf{k} \cdot \mathbf{E})$. Together with (101.13), this gives the required relation

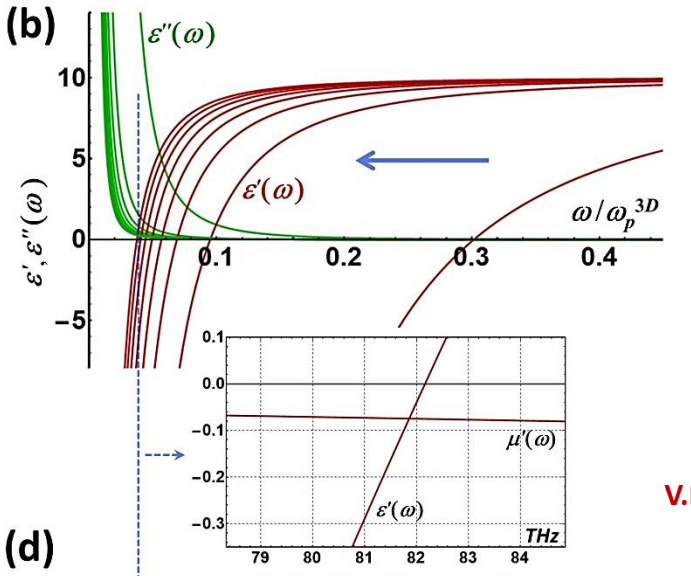
MAGNETO-OPTICAL RESPONSE OF TRANSDIMENSIONAL PLASMONIC FILMS

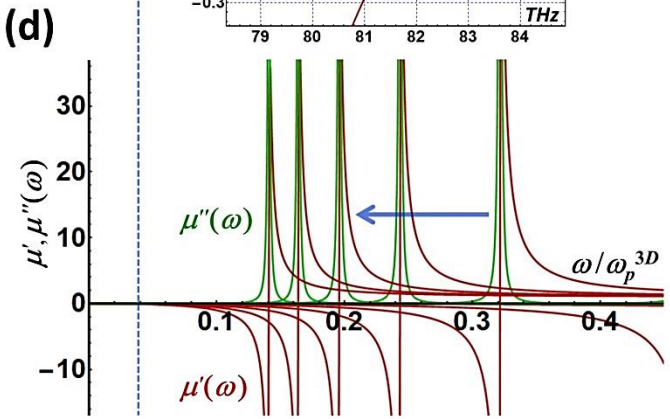
Possibility for Negative Refraction





$$\gamma/\omega_p^{3D} = 0.001, \quad L/\lambda_p^{3D} = 100$$





$$\gamma << \omega < \omega_p^{3D}\sqrt{\frac{2\pi\varepsilon d}{(\varepsilon_1+\varepsilon_2)L}}$$

$$\varepsilon'(\omega)|\mu(\omega)| + \mu'(\omega)|\varepsilon(\omega)| < 0$$

$$\Downarrow$$

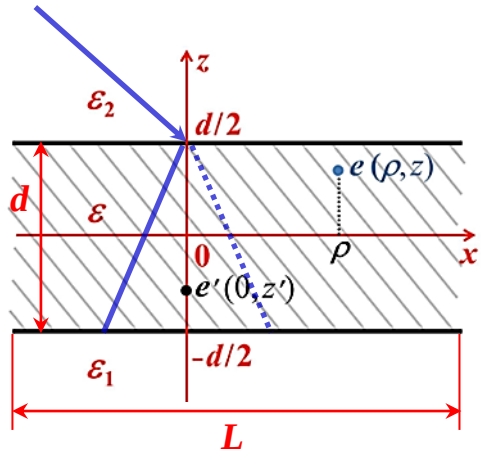
$$\varepsilon'(\omega) \,\&\, \mu'(\omega) < 0$$

$$\Updownarrow$$

Negative Refraction :

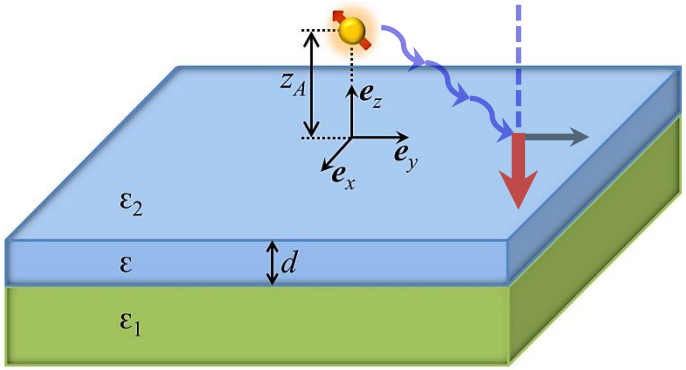
$$\sqrt{n(\omega)} = \sqrt{\varepsilon(\omega)\mu(\omega)} < 0$$

V.M.Shalaev, Nature Photonics 1, 41 (2007)



I.V.Bondarev, H.Mousavi & V.M.Shalaev, MRS Commun. 8, 1092 (2018)

QED EFFECT *example – Dipolar Spontaneous Decay*



$$\hat{E}_\alpha(\mathbf{r},\omega) = i\frac{4\pi\omega^2}{c^2}\int d\mathbf{r}' \sum_{\lambda=x,y,z} G_{\alpha\lambda}(\mathbf{r},\mathbf{r}',\omega)\hat{J}_\lambda(\mathbf{r}',\omega) + \text{h.c.},$$

$$\hat{J}_\lambda(\mathbf{r},\omega) = \sqrt{\text{Im}\,\varepsilon_{\lambda\lambda}(\mathbf{r},\omega)/\pi}\ \hat{f}_\lambda(\mathbf{r},\omega)$$

$$[\hat{f}_\lambda(\mathbf{r},\omega),\ \hat{f}^\dagger_{\lambda'}(\mathbf{r}',\omega')] = \delta_{\lambda\lambda'}\,\delta(\mathbf{r}-\mathbf{r}')\,\delta(\omega-\omega')$$

$$\frac{\varepsilon(k,\omega)}{\varepsilon} = 1 - \frac{\omega_p^2}{\omega(\omega+i\gamma)}$$

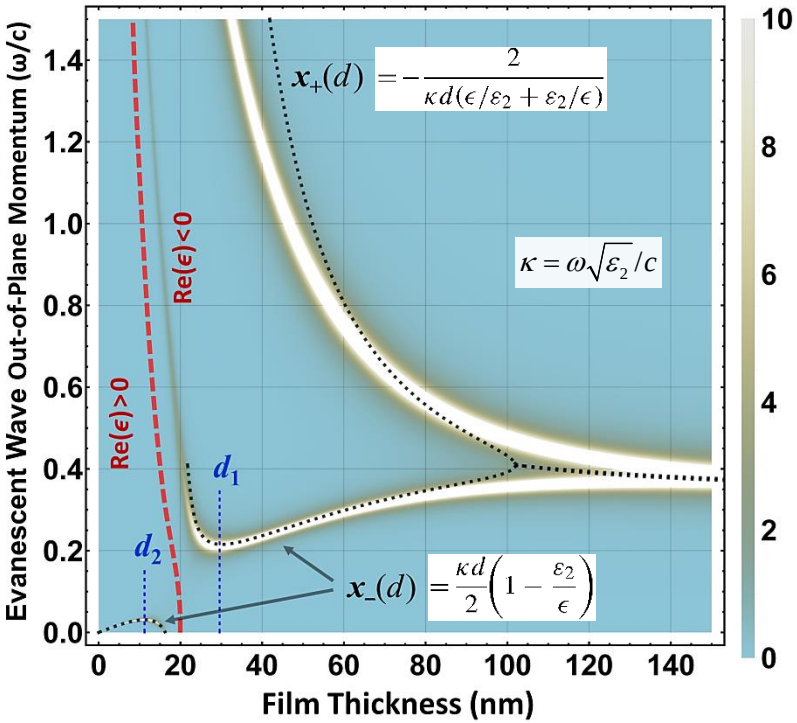
$$\omega_p(k) = \frac{\omega_p^{3D}}{\sqrt{1+(\varepsilon_1+\varepsilon_2)/\varepsilon kd}}$$

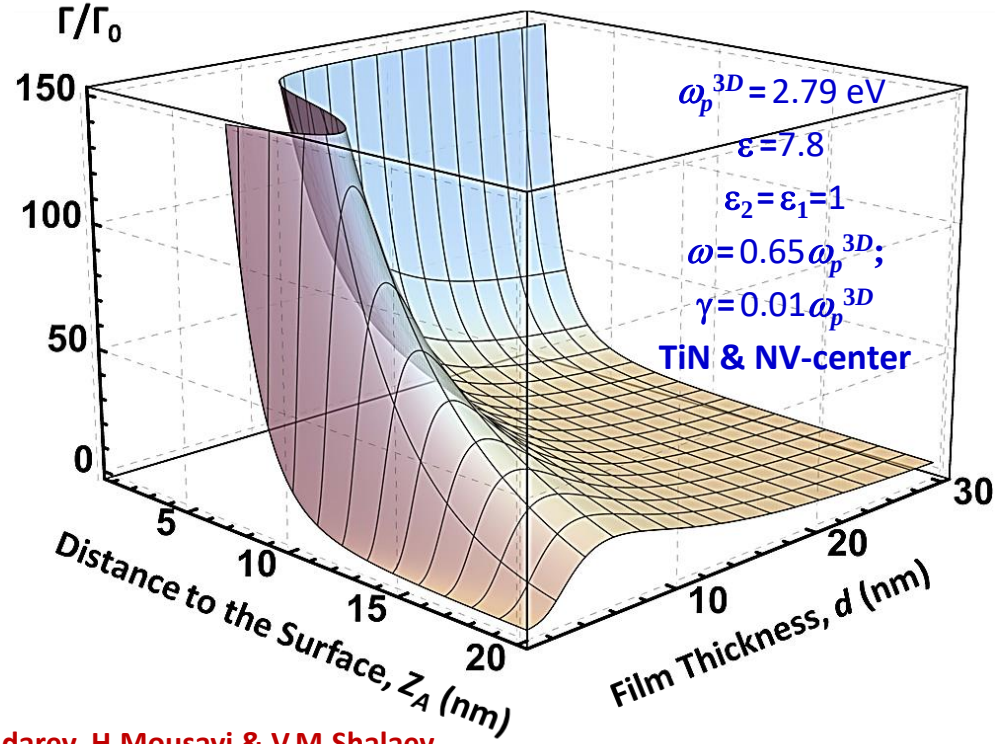
$$\Gamma(\mathbf{r}_A,\omega) = \frac{8\pi\omega^2}{\hbar c^2}\sum_{\alpha,\lambda=x,y,z}\left\langle \hat{d}_\alpha \hat{d}^\dagger_\lambda \right\rangle \text{Im}\,G_{\alpha\lambda}(\mathbf{r}_A,\mathbf{r}_A,\omega) \sim r_{s,p}^{prop} \ \&\ r_{s,p}^{evan}$$

$$\sum_{\beta=x,y,z}\left(\nabla\times\nabla\times - \omega^2\hat{\varepsilon}(\mathbf{r},\omega)/c^2\right)_{\alpha\beta} G_{\beta\lambda}(\mathbf{r},\mathbf{r}',\omega) = \delta_{\alpha\lambda}(\mathbf{r}-\mathbf{r}')$$

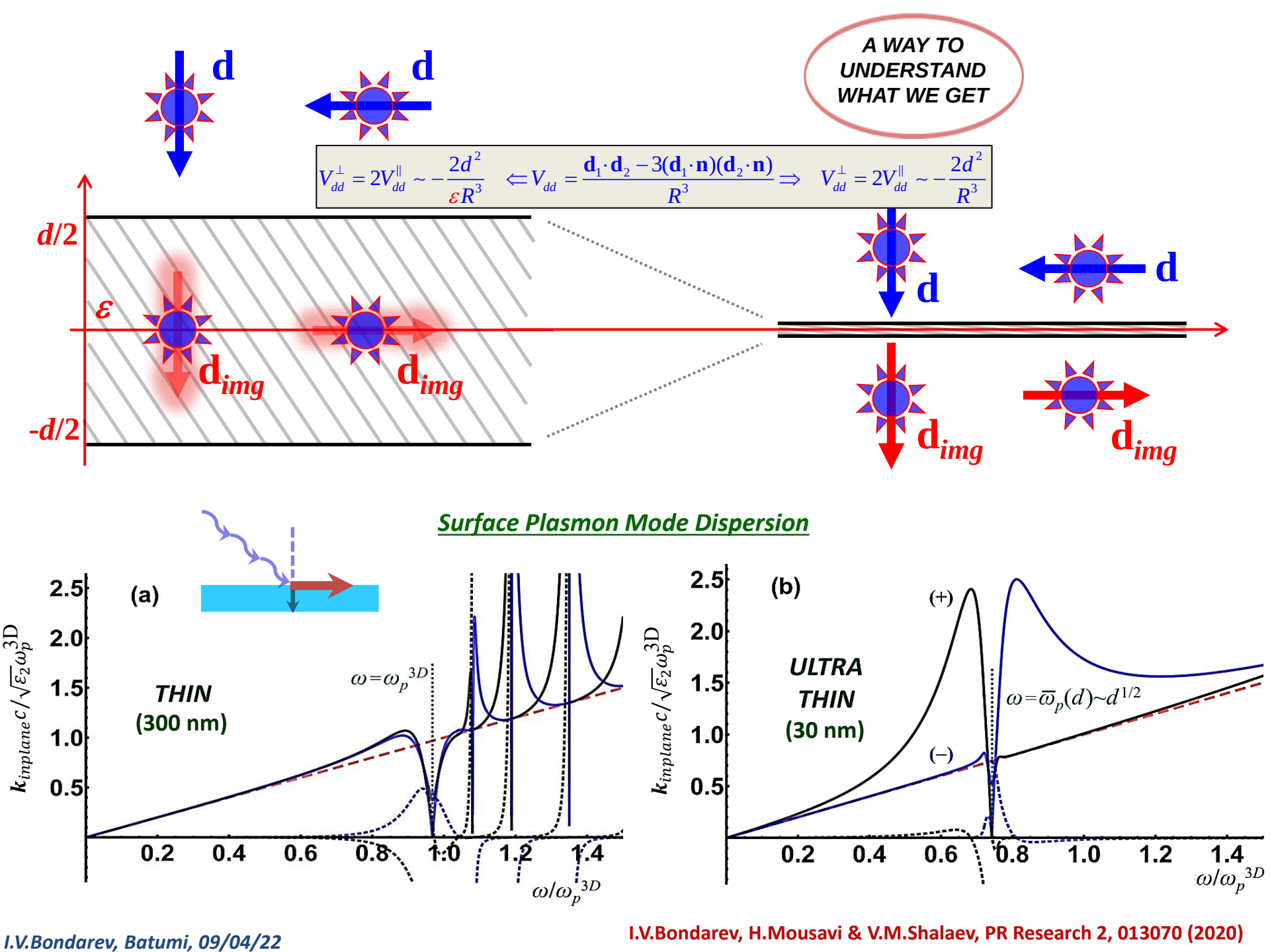
Green Tensor of Planar Multilayers:
M.S.Thomas, PRA 51, 2545 (1995)

$\text{Im}(r_p^{evan})$





I.V.Bondarev, H.Mousavi & V.M.Shalaev,
PR Research 2, 013070 (2020)



I.V.Bondarev, H.Mousavi & V.M.Shalaev, PR Research 2, 013070 (2020)

ARTICLES
PUBLISHED ONLINE: 18 APRIL 2016 | DOI: 10.1038/NPHOTON.2016.70
nature photonics

Cavity-enhanced light emission from electrically driven carbon nanotubes

Felix Pyatkov[1,2†], Valentin Fütterling[1†], Svetlana Khasminskaya[1], Benjamin S. Flavel[1], Frank Hennrich[1], Manfred M. Kappes[1,3], Ralph Krupke[1,2*] and Wolfram H. P. Pernice[1,4*]

nature COMMUNICATIONS
Received 9 Jun 2016 | Accepted 1 Sep 2016 | Published 10 Oct 2016 DOI: 10.1038/ncomms13078

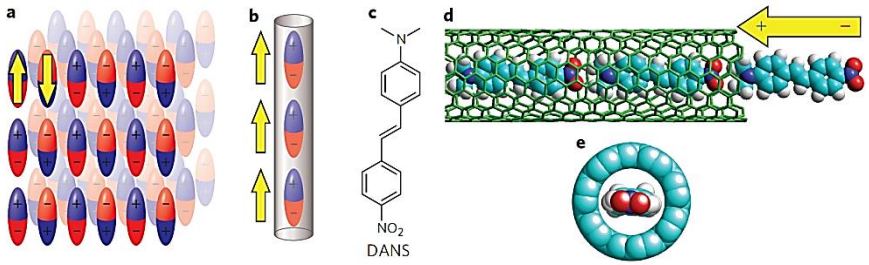
Near-infrared exciton-polaritons in strongly coupled single-walled carbon nanotube microcavities

Arko Graf[1,2], Laura Tropf[2], Yuriy Zakharko[1], Jana Zaumseil[1] & Malte C. Gather[2]

LETTERS
PUBLISHED ONLINE: 2 FEBRUARY 2015 | DOI: 10.1038/NNANO.2015.1
nature nanotechnology

Asymmetric dyes align inside carbon nanotubes to yield a large nonlinear optical response

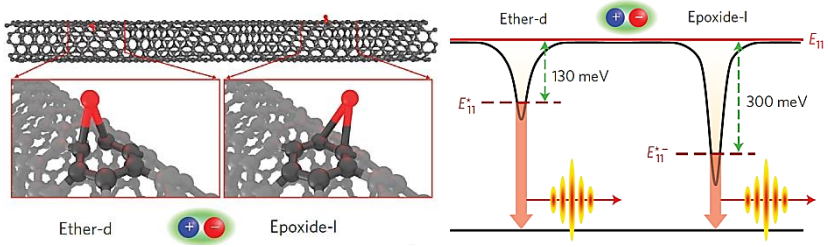
Sofie Cambré[1†], Jochen Campo[1†], Charlie Beirnaert[1], Christof Verlackt[1], Pegie Cool[2] and Wim Wenseleers[1*]



nature nanotechnology
LETTERS
PUBLISHED ONLINE: 13 JULY 2015 | DOI: 10.1038/NNANO.2015.136

Room-temperature single-photon generation from solitary dopants of carbon nanotubes

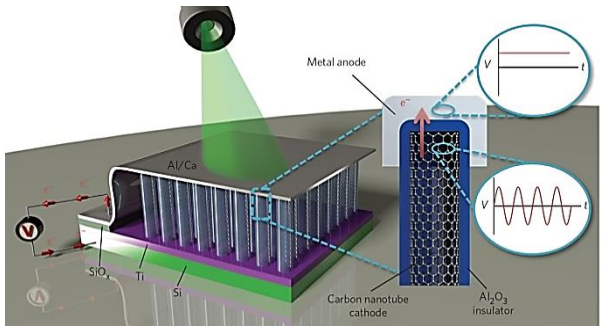
Xuedan Ma, Nicolai F. Hartmann, Jon K. S. Baldwin, Stephen K. Doorn* and Han Htoon*



nature nanotechnology
LETTERS
PUBLISHED ONLINE: 28 SEPTEMBER 2015 | DOI: 10.1038/NNANO.2015.220

A carbon nanotube optical rectenna

Asha Sharma[1,2†], Virendra Singh[1†], Thomas L. Bougher[1†] and Baratunde A. Cola[1,3*]



nature nanotechnology
ARTICLES
PUBLISHED ONLINE: 4 APRIL 2016 | DOI: 10.1038/NNANO.2016.44

Wafer-scale monodomain films of spontaneously aligned single-walled carbon nanotubes

Xiaowei He[1‡], Weilu Gao[1‡], Lijuan Xie[2], Bo Li[3], Qi Zhang[1], Sidong Lei[3], John M. Robinson[1†], Erik H. Hároz[4], Stephen K. Doorn[4], Weipeng Wang[3], Robert Vajtai[3], Pulickel M. Ajayan[3], W. Wade Adams[3], Robert H. Hauge[3,5] and Junichiro Kono[1,3,6*]

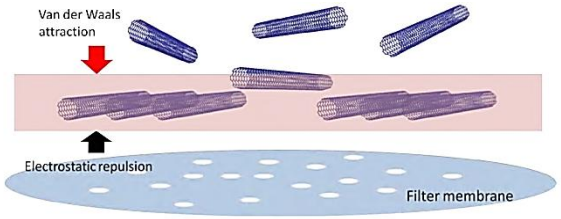
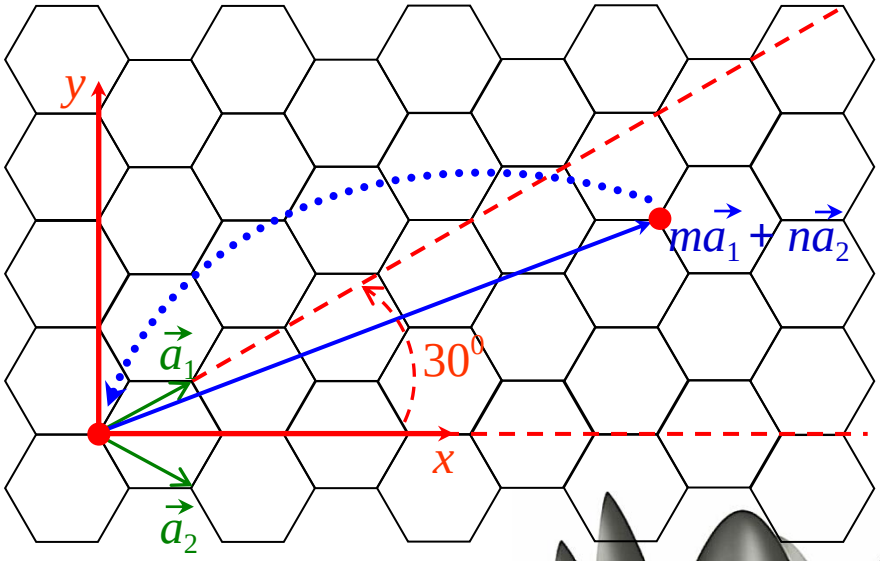


Figure S13. A schematic diagram shows the formation of CNT alignment in a confined region near the surface of the filter membrane.

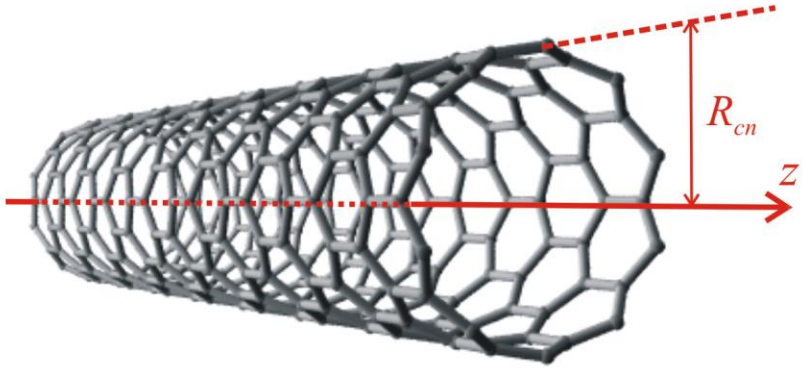
BASIC PHYSICAL PROPERTIES OF SINGLE-WALLED CARBON NANOTUBES

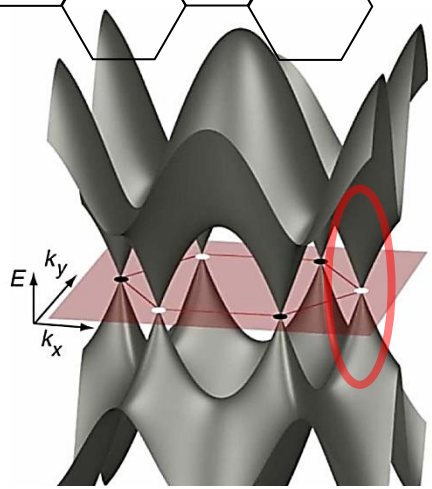
Classification

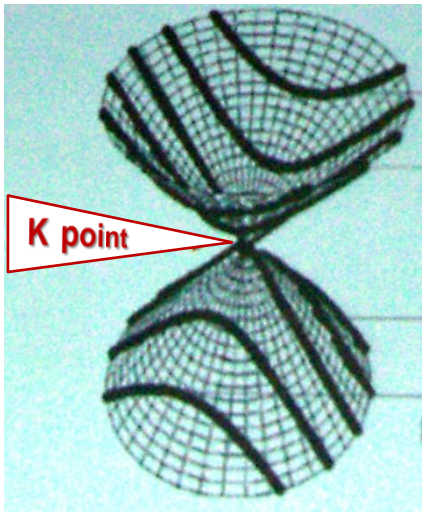
Graphene single sheet



Single-walled CN of (*m*,*n*) type

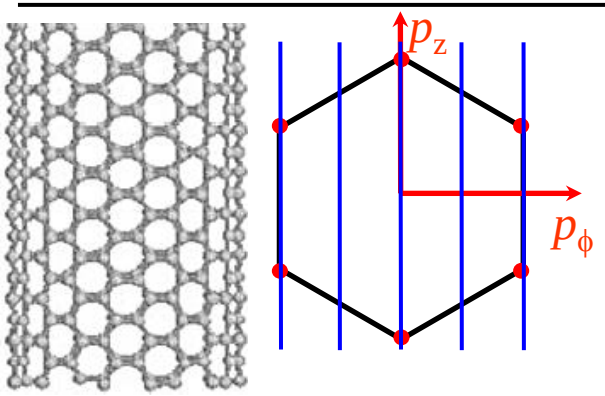




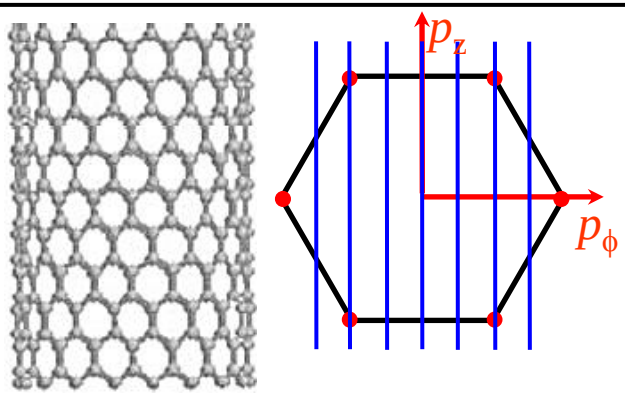


BASIC PHYSICAL PROPERTIES OF SINGLE-WALLED CNs

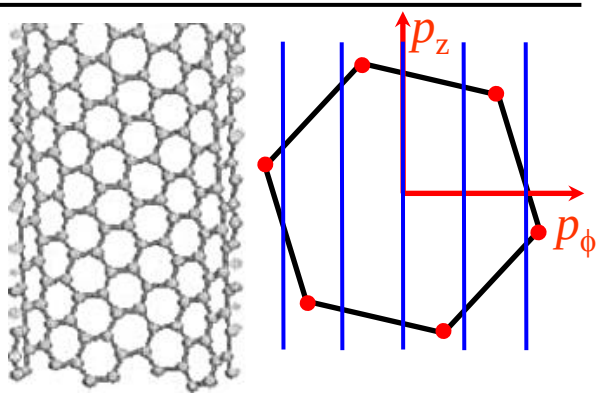
Brillouin zone structure and longitudinal conductivity



(m,m) – "Armchair": metallic for all m

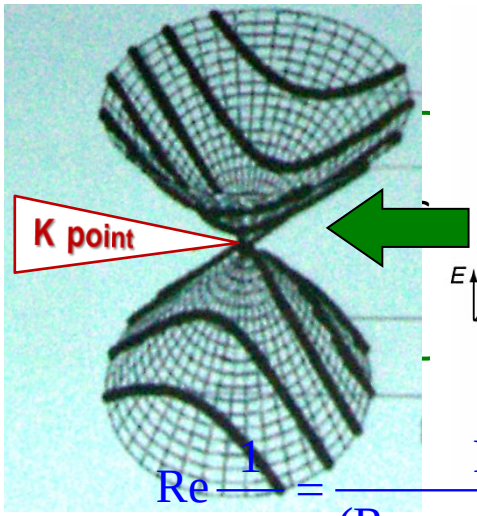


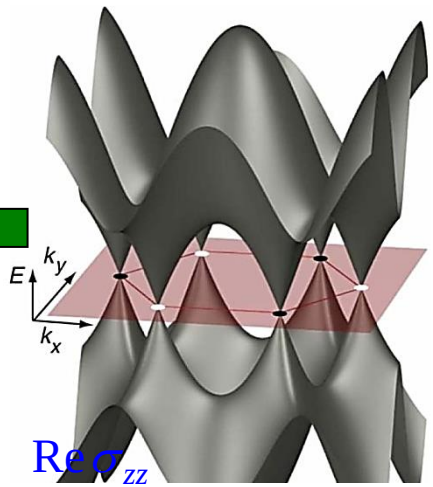
$(m,0)$ – "Zigzag": metallic for $m=3q$, semiconducting for $m\neq 3q$ ($q=1,2,3,\ldots$)



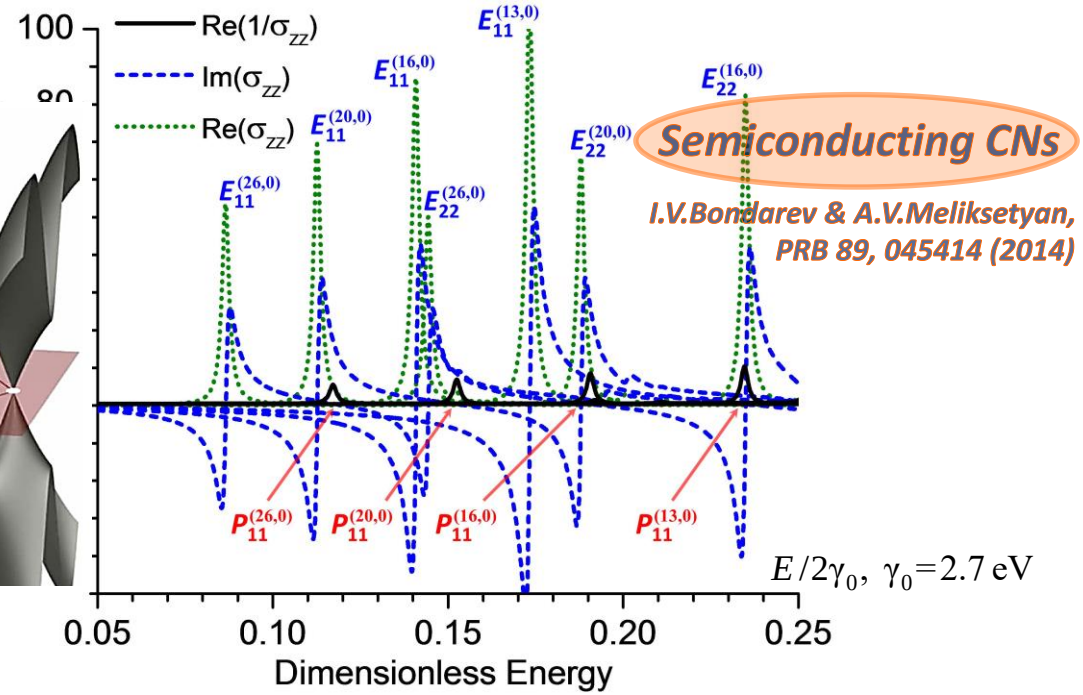
(m,n) – chiral CN: metallic or semi-conducting depending on the radius and chiral angle

$$p_\phi = \frac{\hbar s}{R_{cn}},\ s = 1,2,\ldots,m$$





$$\mathrm{Re}\frac{1}{\sigma_{zz}} = \frac{\mathrm{Re}\,\sigma_{zz}}{(\mathrm{Re}\,\sigma_{zz})^2 + (\mathrm{Im}\,\sigma_{zz})^2}$$

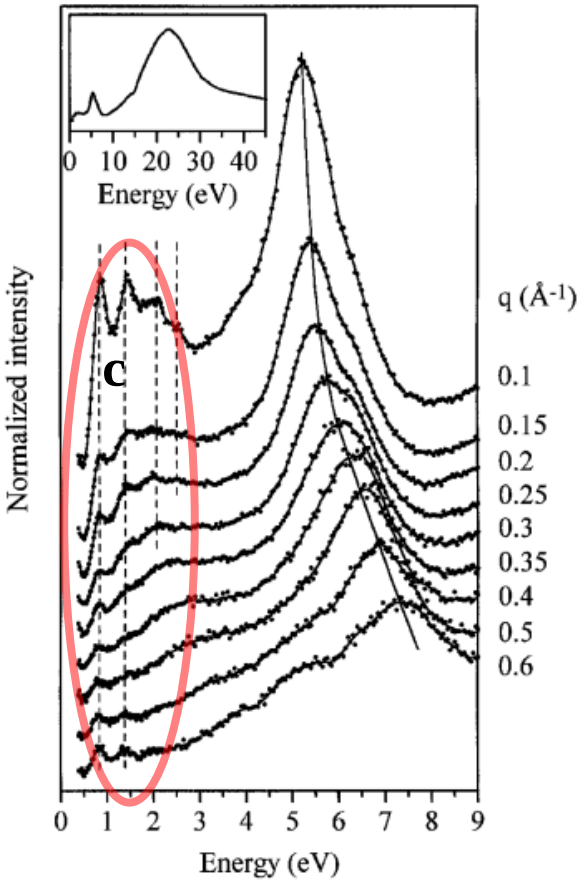


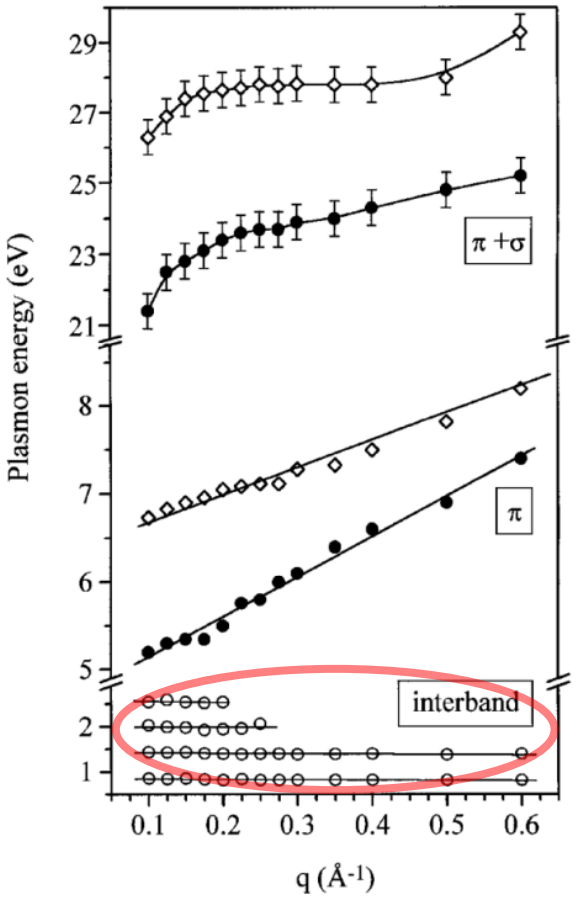
Semiconducting CNs

I.V.Bondarev & A.V.Meliksetyan, PRB 89, 045414 (2014)

EXPERIMENTAL ELECTRON ENERGY LOSS SPECTROSCOPY (EELS) SPECTRA OF SINGLE-WALLED CARBON NANOTUBES

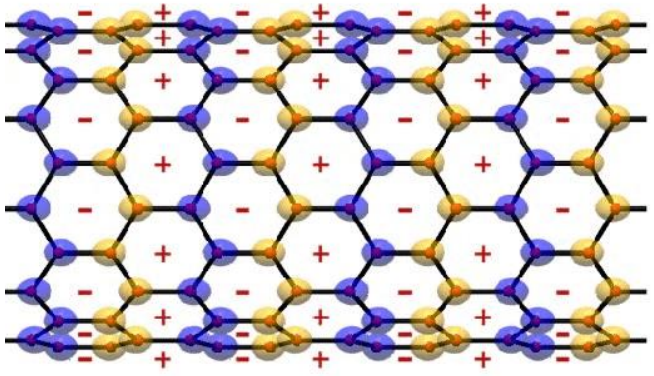
T.Pichler, M.Knupher, M.Golden, J.Fink, A.Rinzler, and R.Smalley, PRL 80, 4729 (1998)





Energy loss function:

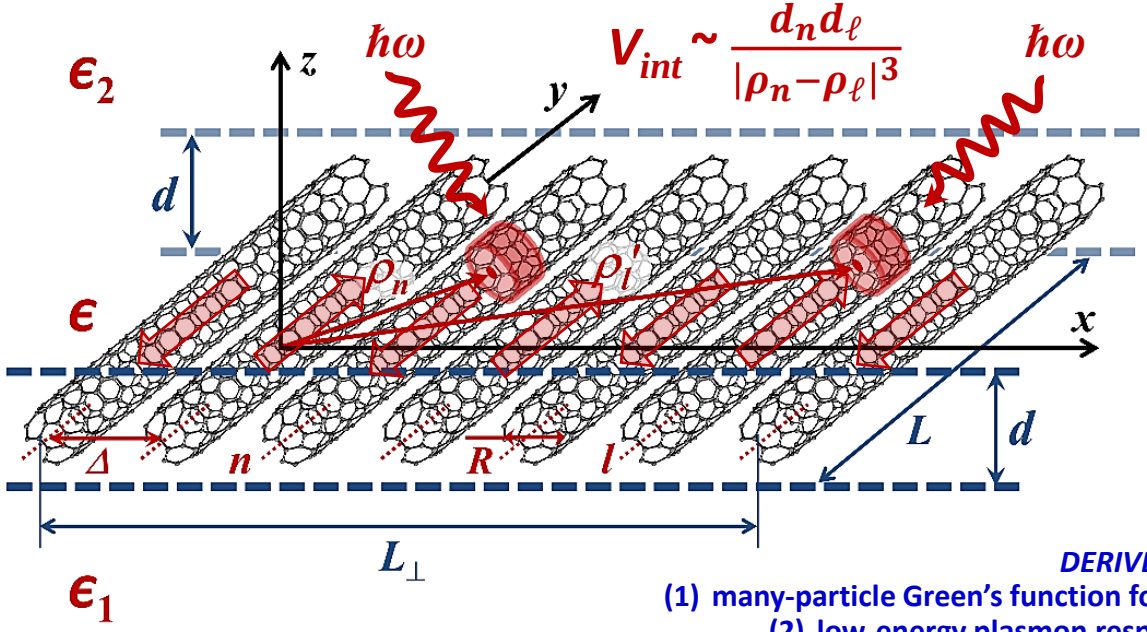
$$\sim -\mathrm{Im}\frac{1}{\varepsilon(\omega,q)} \sim \mathrm{Re}\frac{1}{\sigma(\omega,q)}$$



ULTRATHIN SWCN ARRAYS

Anisotropic Collective Optical Response

I.V. Bondarev and C.M. Adhikari, Physical Review Applied 15, 034001 (2021)



SWCNs cross-talk to each other through the exciton dipole-dipole coupling

***DERIVED using*:**

(1) many-particle Green's function formalism in Matsubara formulation

(2) low-energy plasmon response calculation technique

$$\frac{\varepsilon_{yy}(q,\omega)}{\epsilon} = 1 - \frac{2 f_{\rm CN}\,\sigma_{yy}(q,\omega)}{f_{\rm CN}\,\sigma_{yy}(q,\omega) + i\omega e^2 N_{\rm 2D} R/m^* \omega_p^2(q) d}$$

$$\varepsilon_{xx}(q,\omega) = \epsilon,$$

σ_{yy} is the isolated CN longitudinal conductivity

$f_{\rm CN}$ is the SWCN volume fraction in the dielectric layer

$$\omega_p(q) = \omega_p^{3D} \sqrt{\frac{2qRI_0(qR)K_0(qR)}{1 + (\varepsilon_1 + \varepsilon_2)/\varepsilon qd}}$$

$\omega_p^{3D} = \sqrt{4\pi e^2 N_{3D}/\varepsilon m^*}$ is the *effective* bulk plasma frequency ($N_{3D} = N_{2D}/d$ being the *volumetric* electron density)

(1) G. D. Mahan, Many-particle physics, NY 2000

(2) I.V.Bondarev, Opt. Mater. Express 9, 285 (2019)

ULTRATHIN SWCN ARRAYS

General Properties of Collective Excitations

I.V. Bondarev and C.M. Adhikari, Physical Review Applied 15, 034001 (2021)

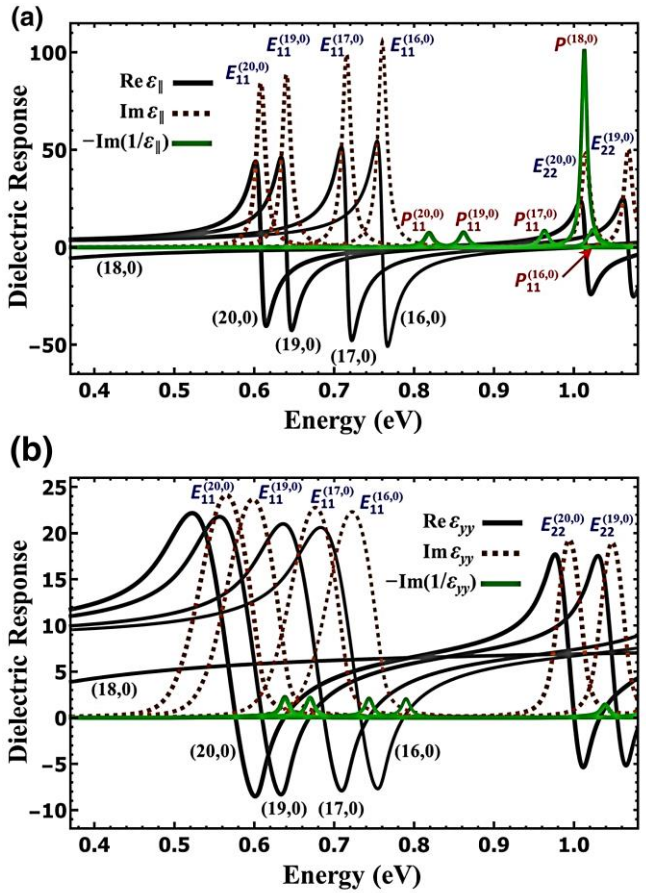


FIG. 4. (a) Individual dielectric responses along the CN axis (longitudinal) for the zigzag (16,0), (17,0), (18,0), (19,0), and (20,0) SWCNs *in vacuum*, obtained using the $\mathbf{k}\cdot\mathbf{p}$ method of the SWCN band-structure calculations [28]. All graphs are scaled down vertically by a factor of 10 for better visibility. (b) Respective room-temperature dielectric response functions along the CN alignment direction, calculated for $d = \Delta = 2R$.

$$\hbar\omega_s(q) = \sqrt{E_s^2(q) + 2E_s(q)V_{ss}(q)}$$

V_{int}

$s = 1,2,\ldots$ – 1st, 2nd exciton resonance of an individual SWCN

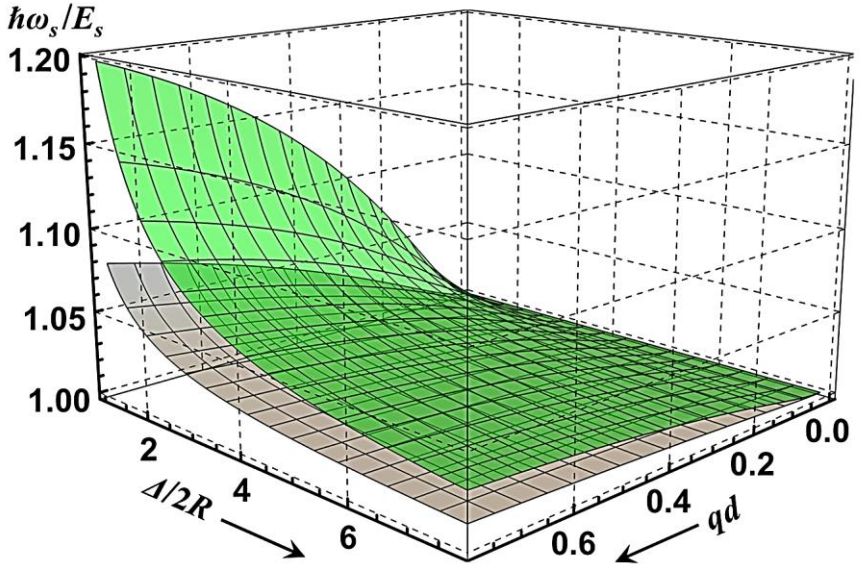
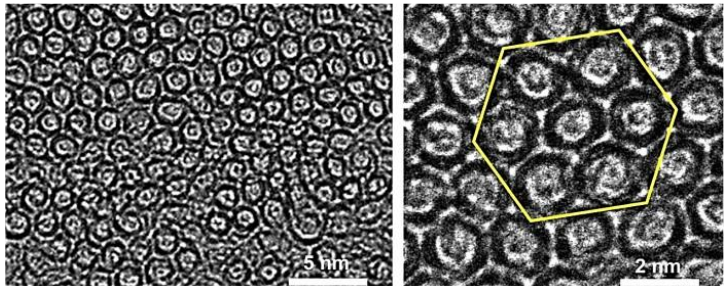


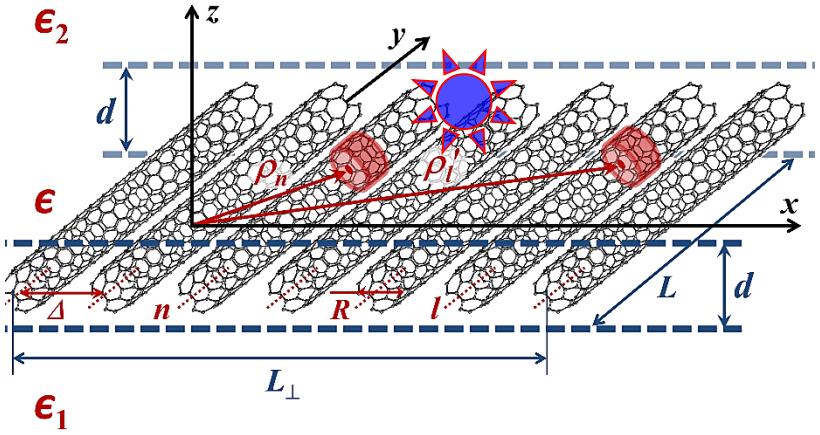
FIG. 2. Dispersion of collective excitations in the ultrathin SWCN arrays with $R/d = 0.5$ (green) and 0.25 (gray) relative to the isolated SWCN exciton dispersion as a function of array parameters qd and $\Delta/2R$.

EXPLORING EFFECTS OF ANISOTROPY IN SWCN FILMS

Finite-Thickness Effects in Ultrathin SWCN Plasmonic Films



Aligned Carbon Nanotube Films (cross-sectional view): Jun Kono group @ Rice; Abram Falk @ IBM; Jon Fan group @ Stanford; see, e.g., Nano Lett. 19, 3131 (2019)



$$\hat{\varepsilon}(q,\omega) = \begin{bmatrix} \varepsilon_{xx} & 0 \\ 0 & \varepsilon_{yy} \end{bmatrix} = \begin{bmatrix} \epsilon & 0 \\ 0 & \varepsilon_{yy}(q,\omega) \end{bmatrix}$$

$$\frac{\varepsilon_{yy}(q,\omega)}{\epsilon} = 1 - \frac{2 f_{\rm CN}\,\sigma_{yy}(q,\omega)}{f_{\rm CN}\,\sigma_{yy}(q,\omega) + i\omega e^2 N_{\rm 2D} R/m^* \omega_p^2(q) d}$$

σ_{yy} is the isolated CN longitudinal conductivity

$$\omega_p(q) = \omega_p^{3D}\sqrt{\frac{2qRI_0(qR)K_0(qR)}{1+(\varepsilon_1+\varepsilon_2)/\varepsilon qd}}$$

$\omega_p^{3D} = \sqrt{4\pi e^2 N_{3D}/\varepsilon m^*}$ is the *effective* bulk plasma frequency ($N_{3D} = N_{2D}/d$ being the *volumetric* electron density)

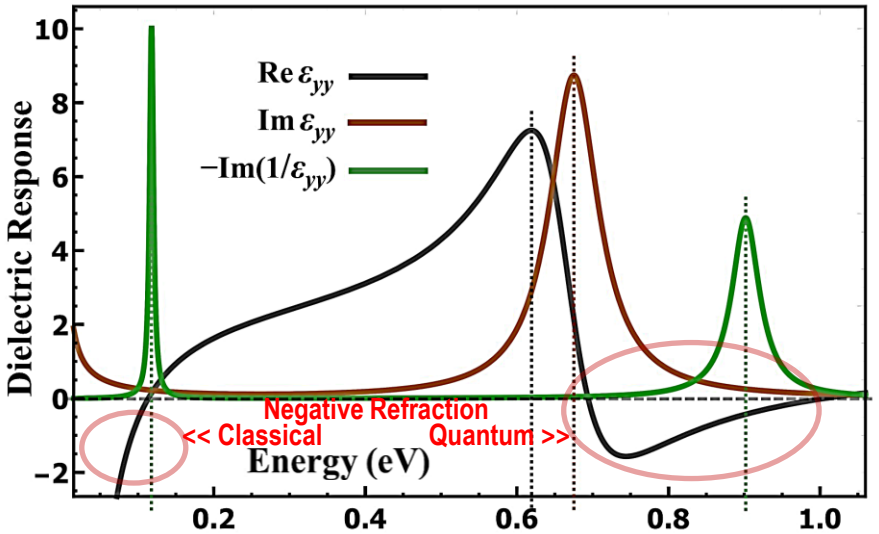


FIG. 5. The room-temperature (300 K) in-plane dielectric response functions along the CN alignment direction calculated for an ultrathin (∼10 nm) weakly inhomogeneous TD film of the MG-mixed (16,0), (17,0), (18,0), (19,0) and (20,0) SWCN

Thickness Controlled

Unidirectional

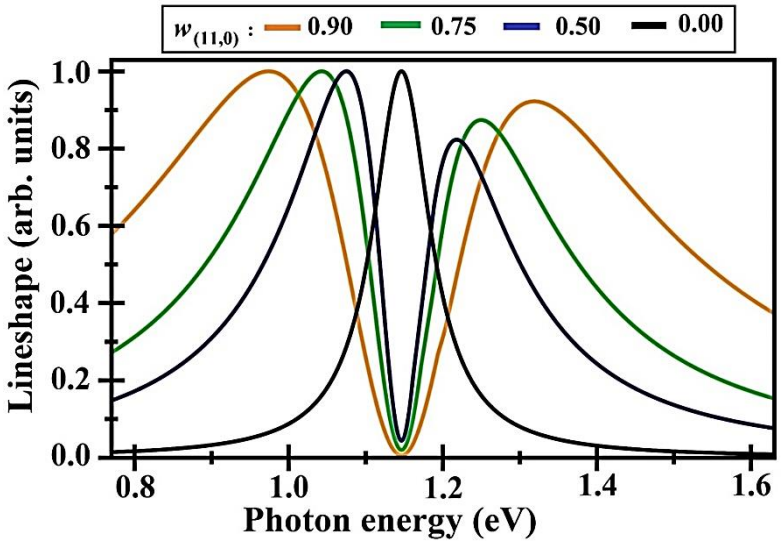
Spontaneous Emission Enhancement

I.V.Bondarev & C.M.Adhikari, PR Applied 15, 034001 (2021)
I.V.Bondarev, Opt. Mater. Express 9, 285 (2019)

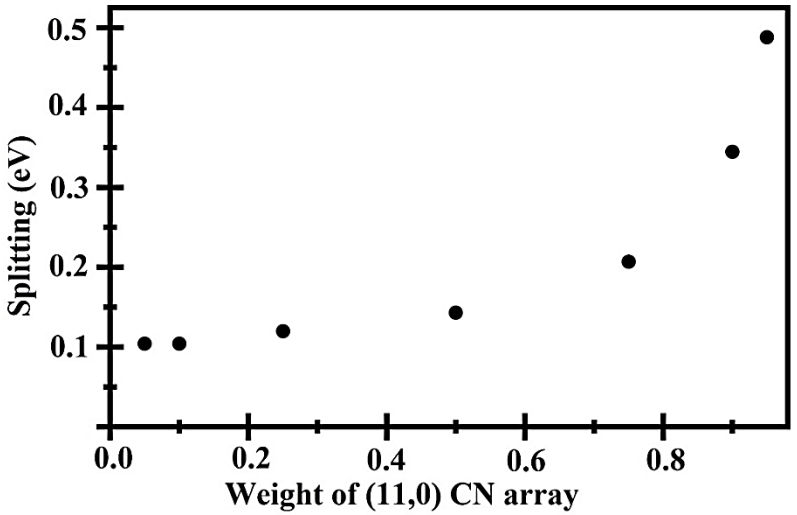
EXCITON-PLASMON COUPLING
in a Designed Mixture of SWCN Arrays

C.M. Adhikari and I.V. Bondarev, Journal of Applied Physics 129, 015301 (2021)

Lineshape profiles of the first exciton absorption resonance



Rabi-splitting as a function of weights of (11,0) CN Array



- All profiles exhibit the line-splitting (aka Rabi-splitting), which is a signature of the strong exciton-plasmon coupling
- The larger weight of the (11,0) CN array results in an increased broadening and a greater splitting of entire absorption profile
- The splitting quickly decreases with the reduction of the (11,0) CN array relative weight, to eventually turn into a single-peak resonance absorption profile for zero relative weight of the (11,0) CN array in the mixture
- The larger Rabi-splitting indicates the stronger exciton-plasmon coupling and the decreased light absorption

***Resonance Absorption Lineshape Profile*:**
I.V.Bondarev, Optics Express 23, 3971 (2015)

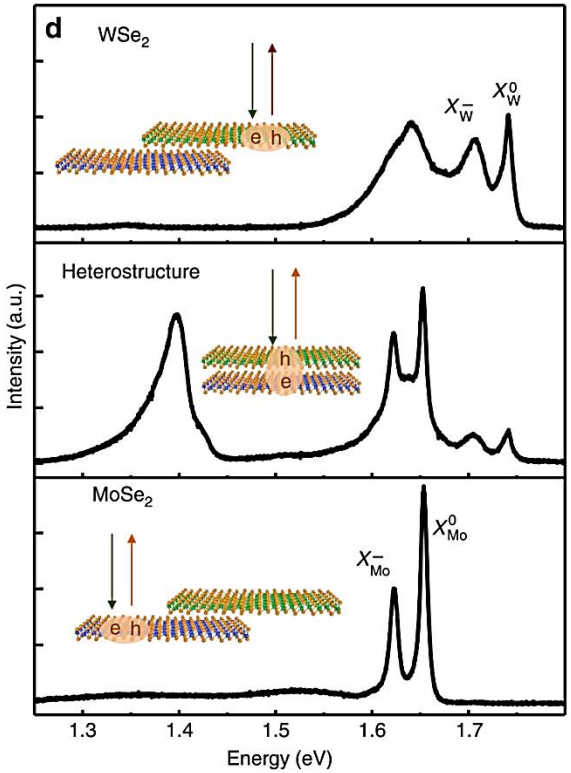
OUTLINE

- **Ultrathin Plasmonic Films of Finite Thickness: *Plasma Frequency Spatial Dispersion and Magneto-Optical Response***
- **Interlayer (Indirect) Excitons and Exciton Complexes in Layered Semiconductors: *Binding Energies within the Configuration Space Approach***
- **Organic Molecular Semiconductor Crystals with *Two* Isolated Frenkel Exciton States: *Frenkel-Charge-Transfer Exciton Intermixing***
- **Summary**

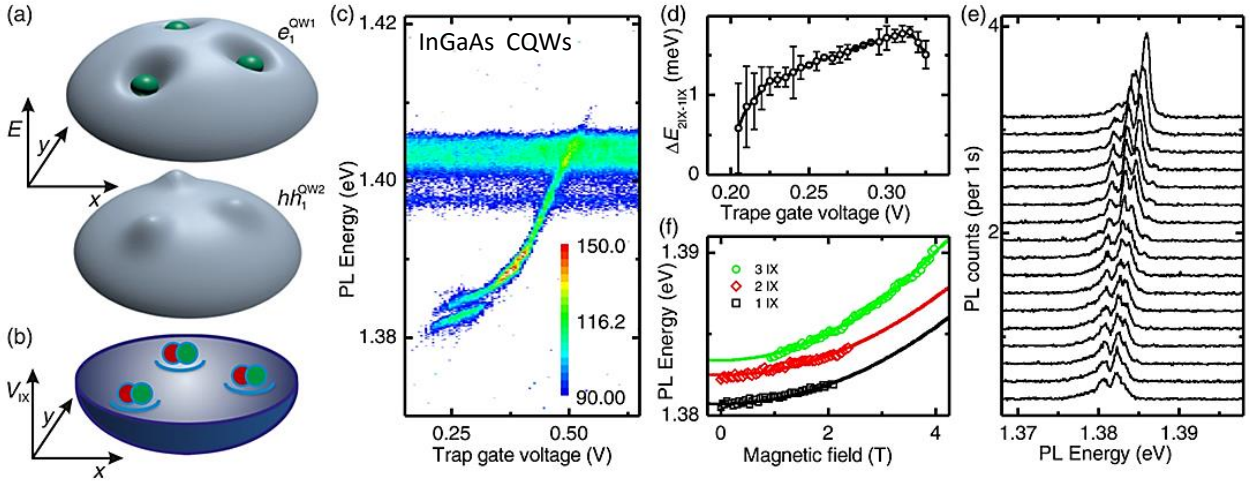
RECENT EXPERIMENTS

with Direct and Indirect Excitons in Coupled Quantum Wells (CQWs) and quasi-2D Layered van der Waals Heterostructures

P.Rivera, *et al*, Nature Commun. 6, 6242 (2015)



G.J.Schinner, et al, Phys. Rev. Lett. 110, 127403 (2013)



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(5) P.Rivera, J.R.Schaibley, A.M.Jones, J.S.Ross, S.Wu, G.Aivazian, P.Klement, K.Seyler, G.Clark, N.J.Ghimire, J.Yan, D.G.Mandrus, W.Yao, & X.Xu, Nature Commun. 6, 6242 (2015)
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(7) K.F.Mak & J.Shan, Nature Photonics 10, 216 (2016)
(8) Y.Y.Kuznetsova, C.J.Dorow, E.V.Calman, L.V.Butov, J.Wilkes, E.A.Muljarov, K.L.Campman, & A.C.Gossard, Phys. Rev. B 95, 125304 (2017)
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(11) J.I.A.Li, T.Taniguchi, K.Watanabe, J.Hone, & C.R.Dean, Nature Physics 13, 751 (2017)
(12) I.V.Bondarev & M.R.Vladimirova, Phys. Rev. B 97, 165419 (2018)
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(14) S.Ovesen, S.Brem, C.Linderälv, M.Kuisma, T.Korn, P.Erhart, M.Selig & E.Malic, Commun. Phys. (2019) 2:23
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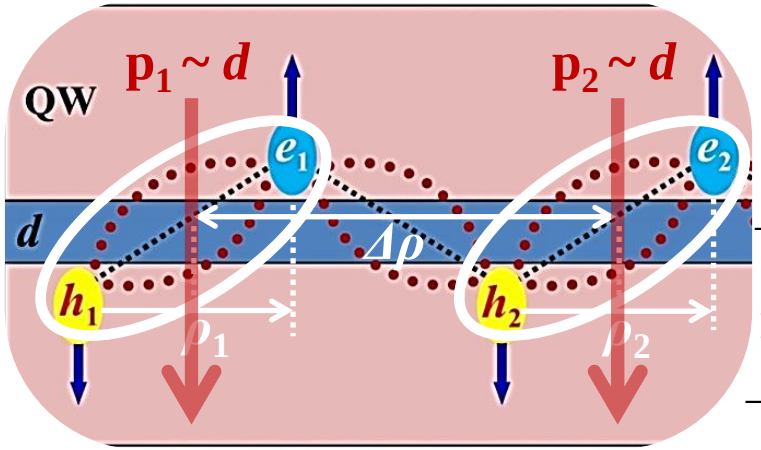
INTERLAYER (indirect) EXCITONS:

- ***Significant Binding Energies***
- ***Long Lifetimes***
- ***High-T BEC & Superfluidity***
- ***Wigner Crystallization***
- ***… more ??? …***

BIEXCITON AND TRION FORMED BY INDIRECT EXCITONS

Ground State Theory within the Configuration Space Approach (a.u.)

ADAPTED FROM: L.P.Gor'kov & L.P.Pitaevski, *The splitting energy of H_2 molecule therms*, Dokl. Akad. Nauk SSSR 151, 822 (1963) [English transl.: Soviet Phys.-Dokl. 8, 788 (1964)]



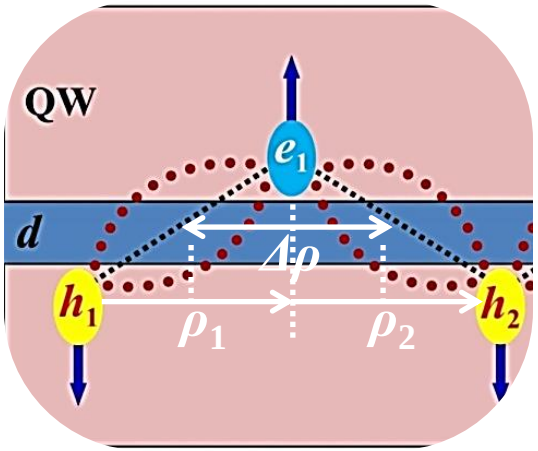
$$V_{p_1p_2} \sim d^2/\Delta\rho^3$$

$$\hat{H}(\rho_1,\rho_2,\Delta\rho,d) = -\frac{1}{\rho_1}\frac{\partial}{\partial\rho_1}\rho_1\frac{\partial}{\partial\rho_1} - \frac{1}{\rho_2}\frac{\partial}{\partial\rho_2}\rho_2\frac{\partial}{\partial\rho_2}$$

$$- \frac{1}{\sqrt{\rho_1^2+d^2}} - \frac{1}{\sqrt{\rho_2^2+d^2}} - \frac{1}{\sqrt{(\rho_1-\Delta\rho)^2+d^2}} - \frac{1}{\sqrt{(\rho_2+\Delta\rho)^2+d^2}}$$

Exact solution available: *R.P.Leavitt & J.W.Little, PRB 42, 11774 (1990)*

$$- \frac{2\ \ e_1\text{-}h_2}{\sqrt{[(\rho_1+\sigma\rho_2)/\lambda-\Delta\rho]^2+d^2}} - \frac{2\ \ e_2\text{-}h_1}{\sqrt{[(\sigma\rho_1+\rho_2)/\lambda+\Delta\rho]^2+d^2}}$$



$$+ \frac{2\ \ h_1\text{-}h_2}{|\sigma(\rho_1-\rho_2)/\lambda+\Delta\rho|} + \frac{2\ \ e_1\text{-}e_2}{|(\rho_1-\rho_2)/\lambda-\Delta\rho|}$$

positive trion ***negative trion***

$$\lambda = 1+\sigma; \quad \sigma = m_e/m_h \to 1$$

due to the mass reversal effect

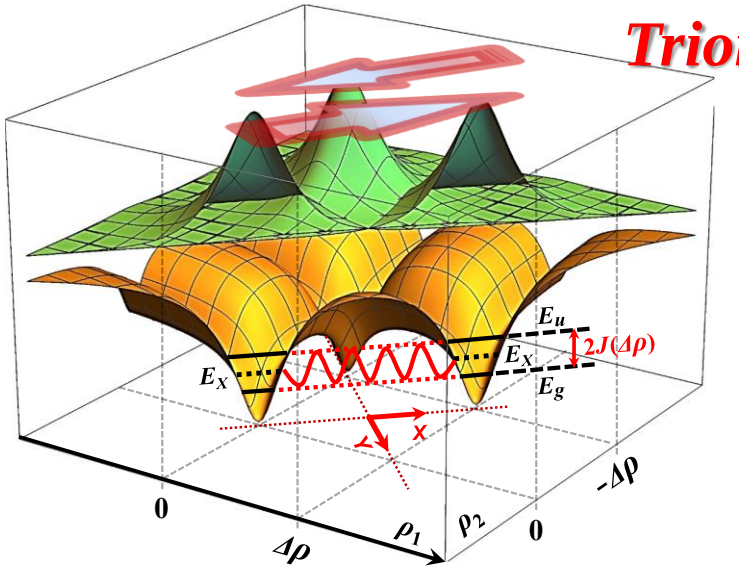
$$Ry^* = \frac{\hbar^2}{2\mu a_B^{*2}} = \frac{\mu(\text{in } m_0)}{\varepsilon^2}13.6\text{ eV};\ \ a_B^* = \frac{\varepsilon}{\mu}0.529\,\text{Å}$$

I.V.Bondarev & M.R.Vladimirova, PRB 97, 165419 (2018)

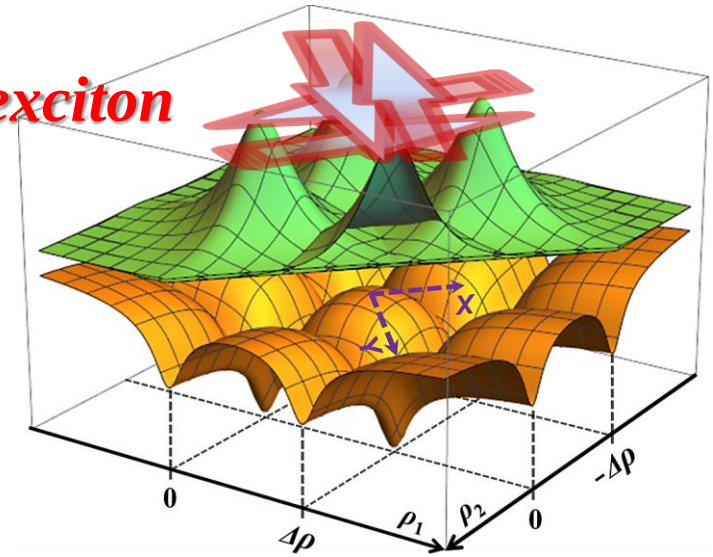
Tunnel Exchange Coupling Configuration for Two Ground-State Indirect Excitons to Form the Trion or Biexciton Complex

ADAPTED FROM: L.P.Gor'kov & L.P.Pitaevski, *The splitting energy of H_2 molecule therms*, Dokl. Akad. Nauk SSSR 151, 822 (1963) [English transl.: Soviet Phys.-Dokl. 8, 788 (1964)]

Trion



Biexciton



Trion Binding Energy

$$E_{X*} = E_g - 2E_X = -J_{X*}(\Delta\rho_0)$$

$$J_{X*}(\Delta\rho) = \int_{-\Delta\rho/\sqrt{2}}^{\Delta\rho/\sqrt{2}} \left| \psi_{X*}(x,y) \frac{\partial \psi_{X*}(x,y)}{\partial x} \right|_{x=0} dy$$

$$\psi_{X*}(x,y) = \psi_{IX}(\rho_1(x,y),d)\,\psi_{IX}(\rho_2(x,y),d)\,exp(-S_{X*}(x,y))$$

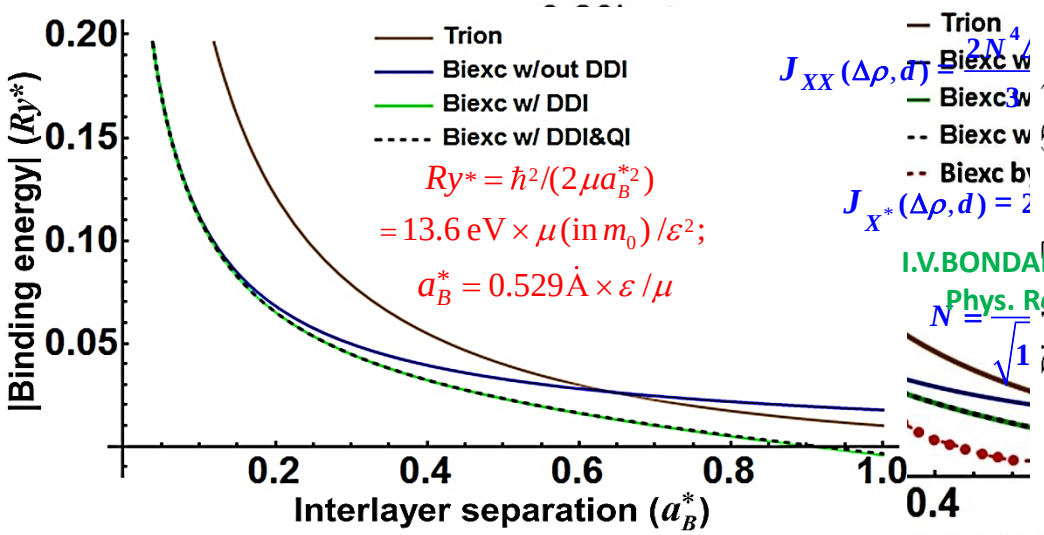
Biexciton Binding Energy

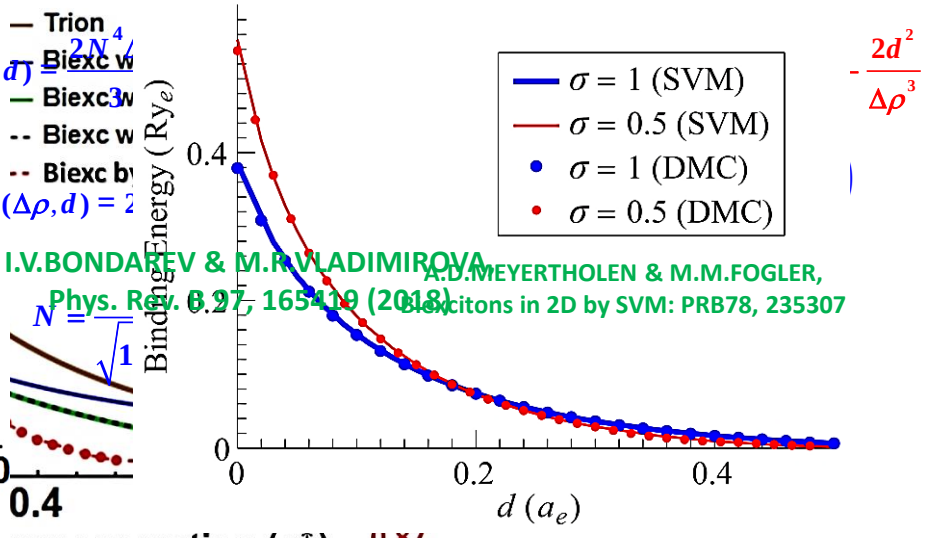
$$E_{XX} = E_g - 2E_X = -J_{XX}(\Delta\rho_0)$$

$$J_{XX}(\Delta\rho) = \frac{2}{3!} \int_{-\Delta\rho/\sqrt{2}}^{\Delta\rho/\sqrt{2}} \left| \psi_{XX}(x,y) \frac{\partial \psi_{XX}(x,y)}{\partial x} \right|_{x=0} dy$$

I.V.Bondarev & M.R.Vladimirova, PRB 97, 165419 (2018)
I.V.Bondarev, Mod. Phys. Lett. B 30, 1630006 (2016)
PRB 90, 245430 (2014); PRB 83, 153409 (2011)

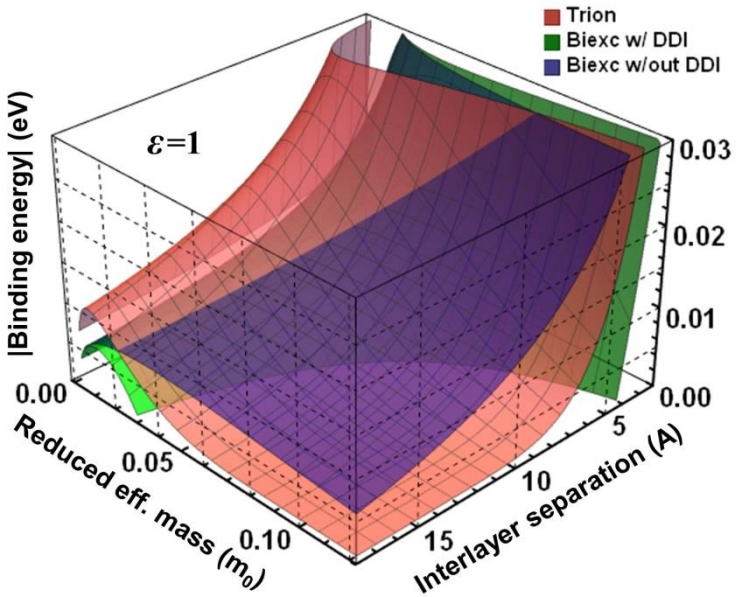
BIEXCITON AND TRION FORMED BY INDIRECT EXCITONS
Ground State Binding Energies Calculated

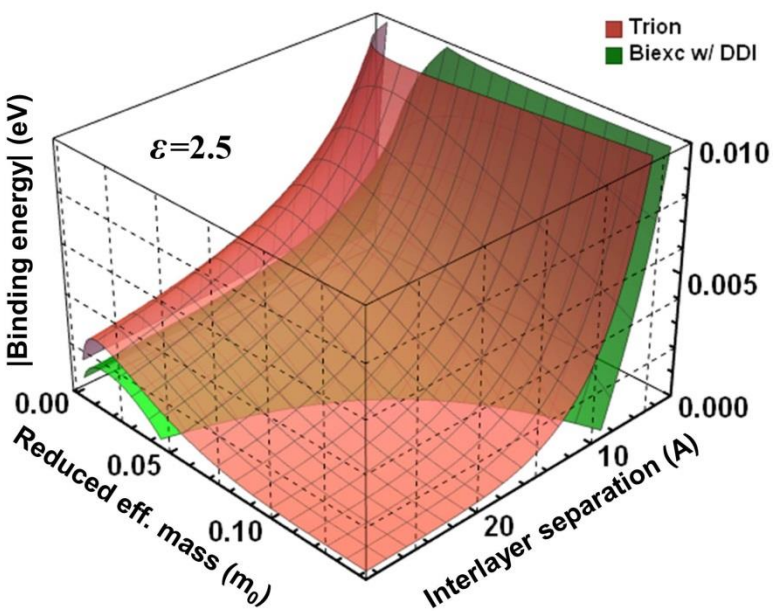




I.V.BONDAREV & M.R.VLADIMIROVA,
Phys. Rev. B 97, 165419 (2018)

A.D.MEYERTHOLEN & M.M.FOGLER,
Biexcitons in 2D by SVM: PRB78, 235307





Complexes of dipolar excitons in layered quasi-two-dimensional nanostructures

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[1]*Department of Mathematics & Physics, North Carolina Central University, Durham, North Carolina 27707, USA*
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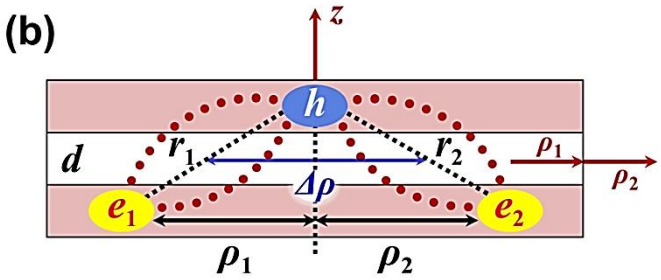
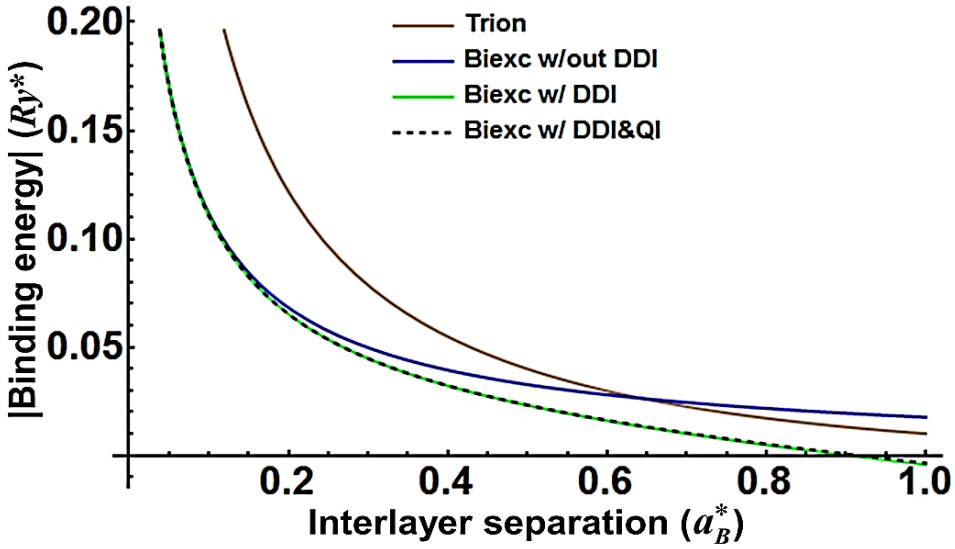
(Received 25 December 2017; revised manuscript received 19 March 2018; published 16 April 2018)

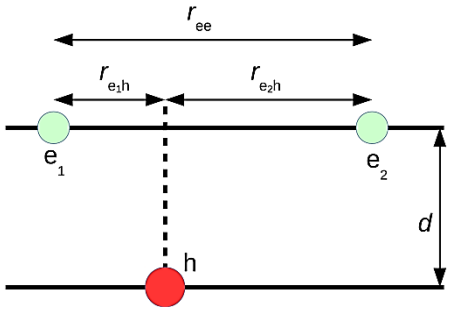


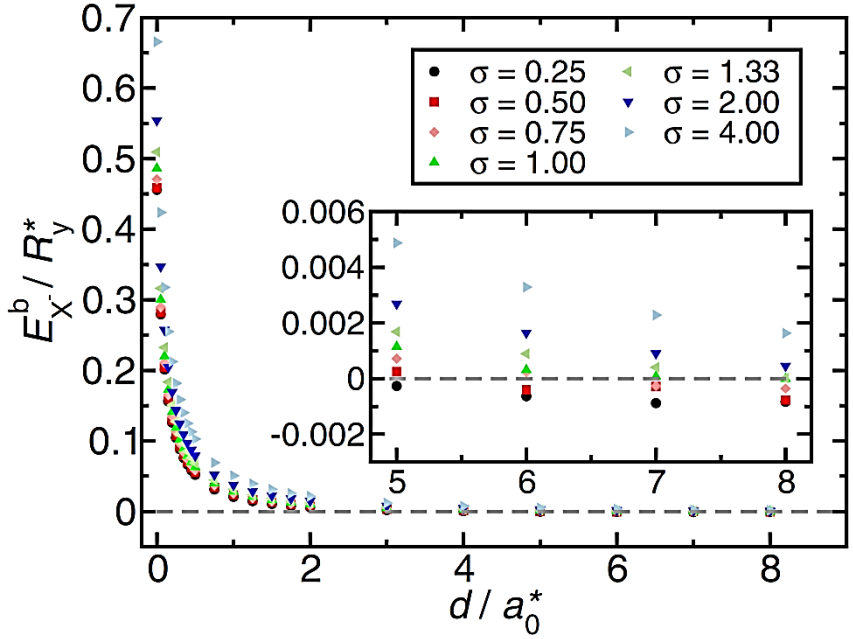


Stability of trions in coupled quantum wells modeled by two-dimensional bilayers

O. Witham,[1,2] R. J. Hunt,[2] and N. D. Drummond[2]
[1]*Institut für Theoretische Physik, Goethe-Universität Frankfurt, 60438 Frankfurt am Main, Germany*
[2]*Department of Physics, Lancaster University, Lancaster LA1 4YB, United Kingdom*

(Received 28 July 2017; published 20 February 2018)

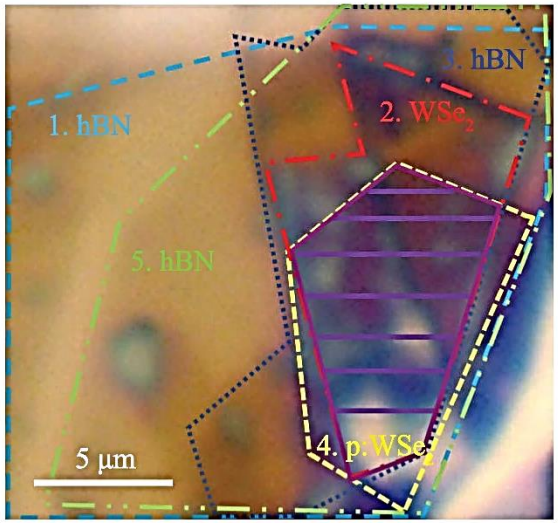


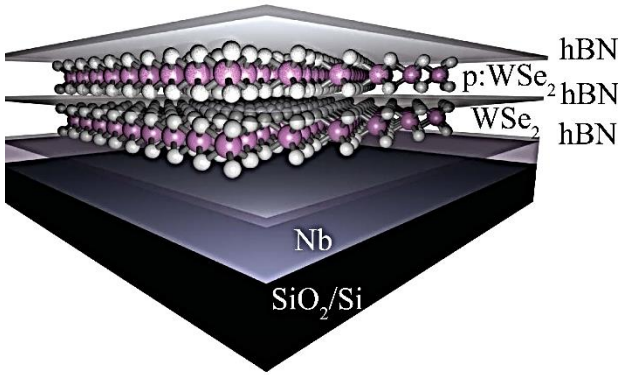


THE QUATERNION EXPERIMENT

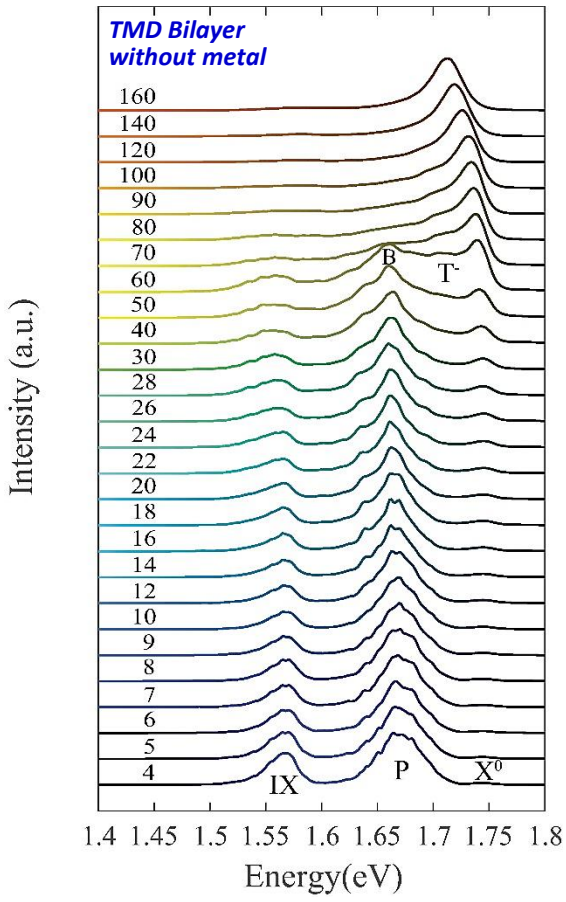
Photoluminescence Measurement Data (David Snoke group)

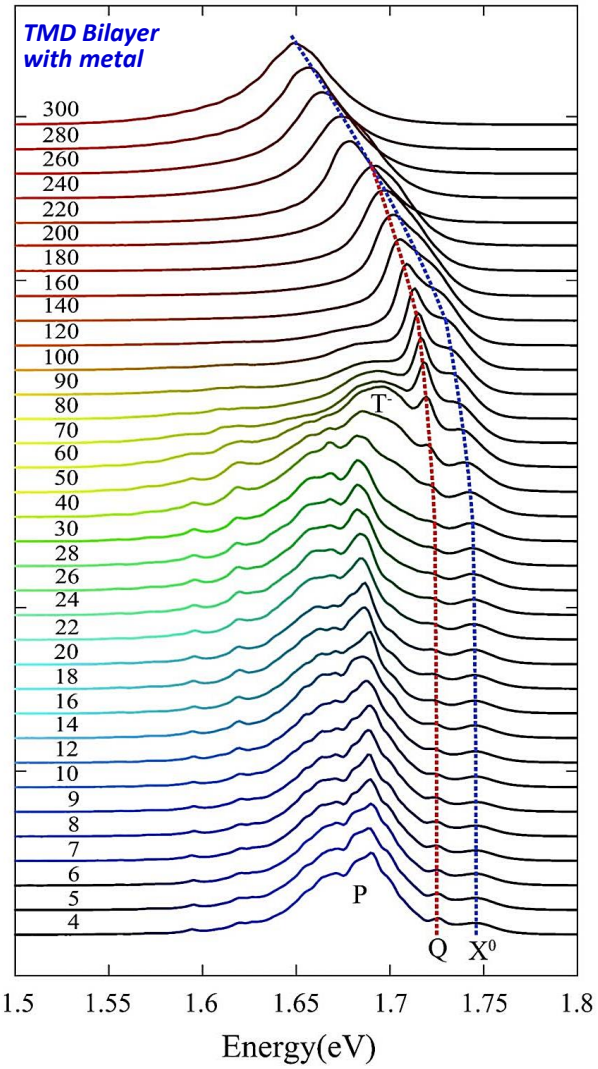
Z.Sun, J.Beaumariage, Q.Wan, H.Alnatah, N.Hougland, J.Chisholm, Q.Cao, K.Watanabe, T.Taniguchi, B.M.Hunt, I.V.Bondarev, and D.W.Snoke, Nano Letters 21, 7669 (2021)





University of Pittsburgh





INTRALAYER TRIONS ARE CHARGED "interlayer" EXCITONS

one obtains in the limit of d -> 0

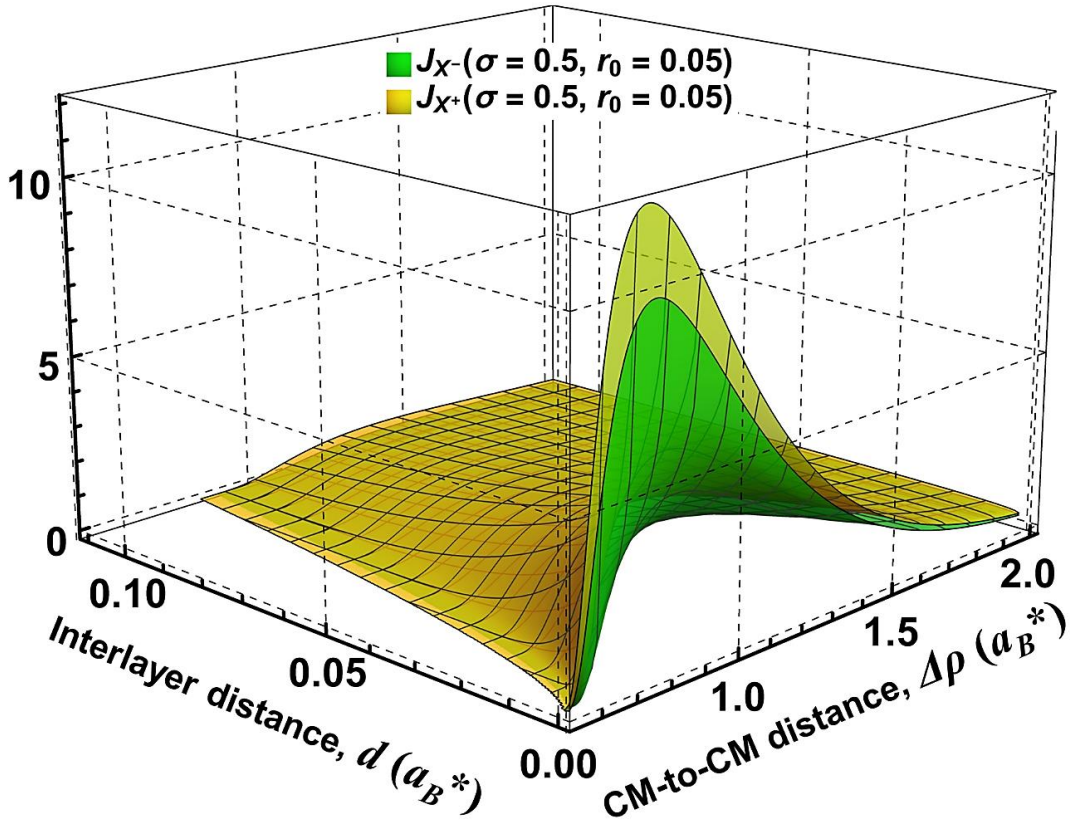
I.V. Z.Sun, J.Beaumariage, Q.Wan, H.Alnatah, N.Hougland, J.Chisholm, Q.Cao, K.Watanabe, T.Taniguchi, B.M.Hunt, I.V.Bondarev, and D.W.Snoke, Nano Letters 21, 7669 (2021)

$$J_{X^\pm}(\Delta\rho)=2^{10}e^{-4\Delta\rho}\Delta\rho\left[1+\frac{\Delta\rho}{4\left(r_0+\left\{\begin{smallmatrix}1\\ \sigma\end{smallmatrix}\right\}\Delta\rho/\lambda\right)(2\Delta\rho-1)}\right]\left(\frac{r_0+\left\{\begin{smallmatrix}1\\ \sigma\end{smallmatrix}\right\}\Delta\rho/\lambda}{r_0+\Delta\rho}\right)^{\frac{\lambda\Delta\rho}{\left\{\begin{smallmatrix}\sigma\\ 1\end{smallmatrix}\right\}(2\Delta\rho-1)}}$$

$$\lambda=1+m_e/m_h=1+\sigma$$

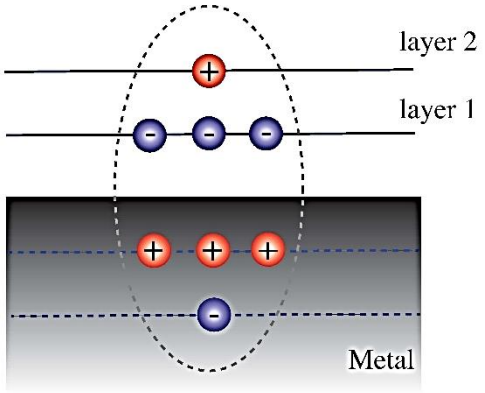
$$\Delta\rho_{X^\pm}=\frac{13-\left\{\begin{smallmatrix}\sigma\\ 1/\sigma\end{smallmatrix}\right\}}{8}-\left(3+2\left\{\begin{smallmatrix}\sigma\\ 1/\sigma\end{smallmatrix}\right\}\right)r_0$$

$$E_{X^\pm}(\sigma)\Big|_{d=0}=-J_{X^\pm}(\sigma,\Delta\rho=\Delta\rho_{X^\pm})$$



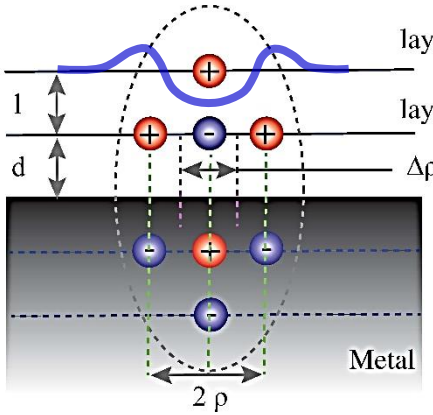
THE FORMATION OF QUATERNION
General Quasiclassical Theory

Z.Sun, J.Beaumariage, Q.Wan, H.Alnatah, N.Hougland, J.Chisholm, Q.Cao, K.Watanabe, T.Taniguchi, B.M.Hunt, I.V.Bondarev, and D.W.Snoke, Nano Letters 21, 7669 (2021)



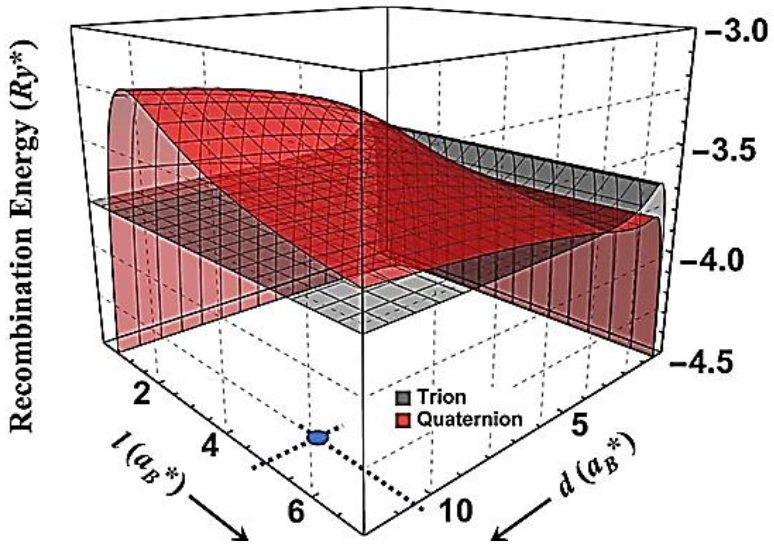
YUDSON MODEL >>>>>>>>>>

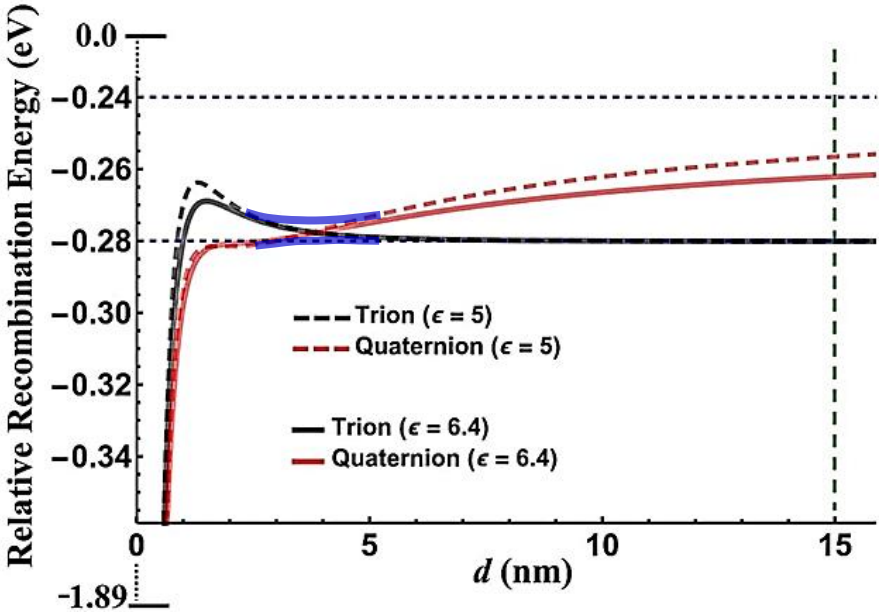
V.I.Yudson,
Phys. Rev. Lett. 77, 1564 (1996)



<<<<<<<<<< OUR MODEL

$$U(\rho,d,l)=U_0(\rho)+2\left(\frac{4}{\sqrt{(2d)^2+\rho^2}}-\frac{2}{\sqrt{(2d)^2+(2\rho)^2}}-\frac{3}{2d}+\frac{2}{\sqrt{l^2+\rho^2}}-\frac{1}{l}+\frac{2}{2d+l}-\frac{1}{2d+2l}-\frac{4}{\sqrt{(2d+l)^2+\rho^2}}\right)$$



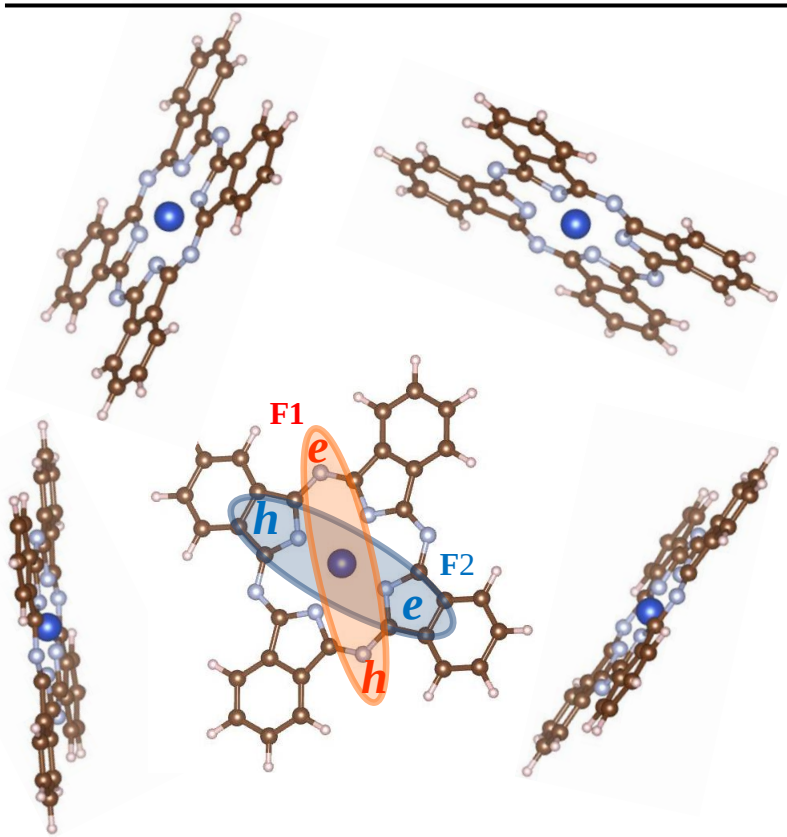


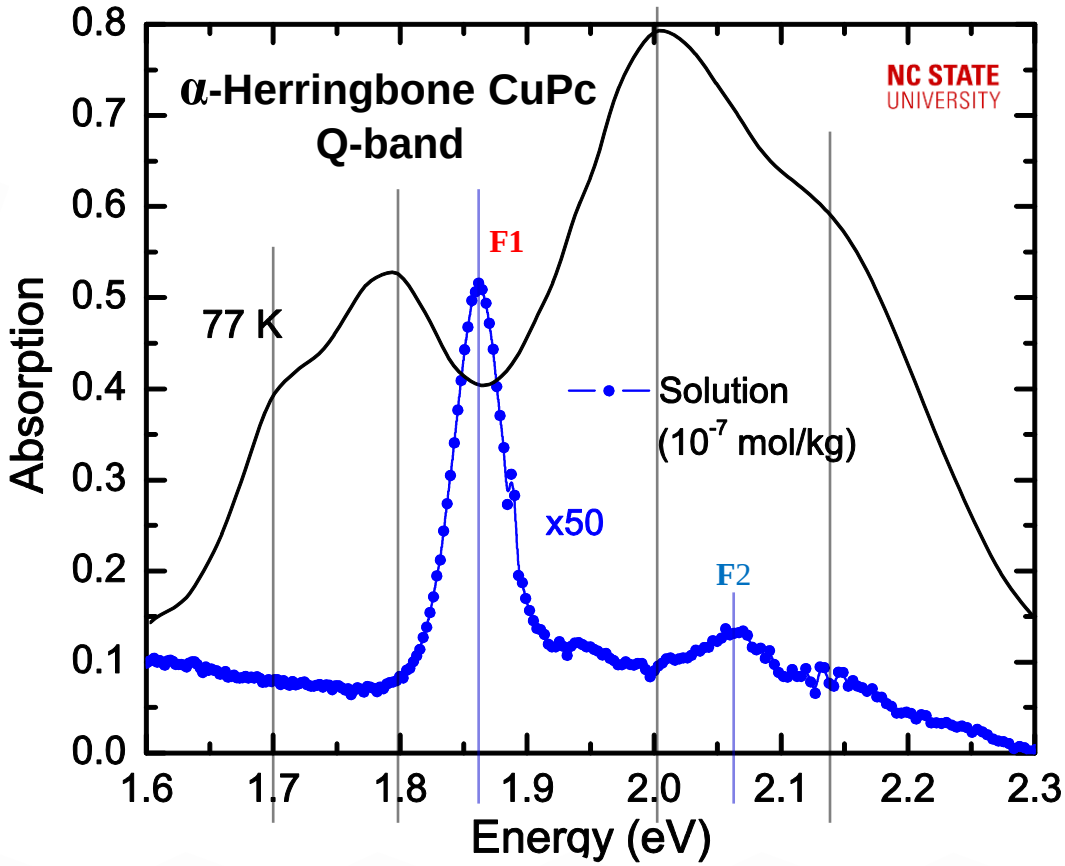
OUTLINE

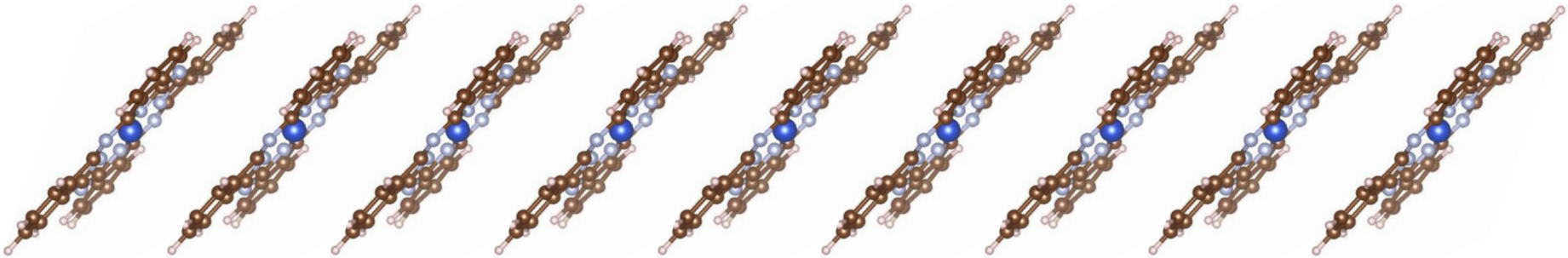
- **Ultrathin Plasmonic Films of Finite Thickness: *Plasma Frequency Spatial Dispersion and Magneto-Optical Response***
- **Interlayer (Indirect) Excitons and Exciton Complexes in Layered Semiconductors: *Binding Energies within the Configuration Space Approach***
- **Organic Molecular Semiconductor Crystals with *Two* Isolated Frenkel Exciton States: *Frenkel-Charge-Transfer Exciton Intermixing***
- **Summary**

Experimental Data I

Crystalline CuPc on a glass substrate vs CuPc in solution (NCSU data)



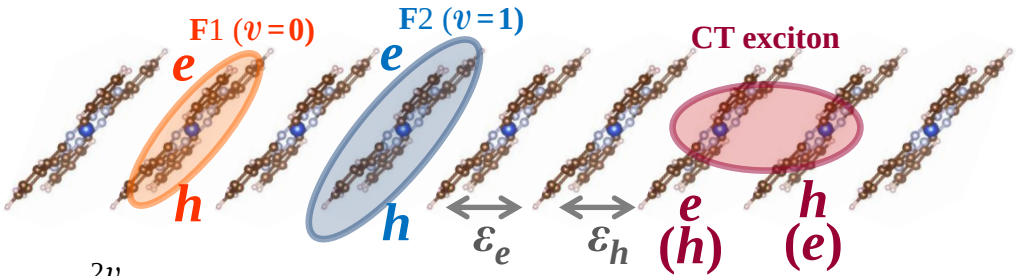




Quantum Theory of the Frenkel-CT Exciton Intermixing

Solving the Eigen-Value Problem for the Hamiltonian of the Two Frenkel Excitons Coupled Individually to the CT exciton

$$\hat{H}\{k\text{-}space\} = \sum_k \left(\hat{H}_k^F + \hat{H}_k^{FF} + \hat{H}_k^C + \hat{H}_k^{FC} \right),$$



$$\hat{H}_k^F = \sum_{v=0,1} \Delta_F^v B_{kv}^\dagger B_{kv}, \quad H_k^{FF} = \sum_{v=0,1} L_k^{vv'} B_{kv}^\dagger B_{kv'},$$

$$L_k^{vv'} \approx 2M s_v s_{v'} \cos k, \quad s_v = \left\langle \chi_n^{1v} \middle| \chi_n^{00} \right\rangle, \quad s_v^2 = F_{0v} = \frac{g^{2v}}{v!} e^{-g^2} \; (if \;\; g \approx 1, \; then \;\; s_0 \approx s_1 \approx 0.6 = s)$$

$$\hat{H}_k^C = \Delta_{CT} \left(\tilde{C}_{kg}^\dagger \tilde{C}_{kg} + \tilde{C}_{ku}^\dagger \tilde{C}_{ku} \right), \quad H_k^{FC} = \sum_{v=0,1} \sqrt{2} \varepsilon_k s_v B_{kv}^\dagger \tilde{C}_{kg} + h.c., \quad \varepsilon_k = \sqrt{\varepsilon_+^2 \cos^2 \frac{k}{2} + \varepsilon_-^2 \sin^2 \frac{k}{2}}, \quad \varepsilon_\pm = \varepsilon_e \pm \varepsilon_h$$

$$\xi_{v\mu}(k) = u_{v\mu}^*(k) B_{kv} + c_v^*(k) \tilde{C}_{kg}, \; v = 0,1; \; \mu = 1,2$$

M.Hoffmann, K.Schmidt, T.Fritz, T.Hasche, V.M.Agranovich, and K.Leo, Chem. Phys. 258, 73 (2000)

$$\hat{H} = \sum_{\mu=1,2} \sum_{v=0,1} \sum_k \hbar \omega_{v\mu}(k) \xi_{v\mu}^\dagger(k) \xi_{v\mu}(k) + E_0$$

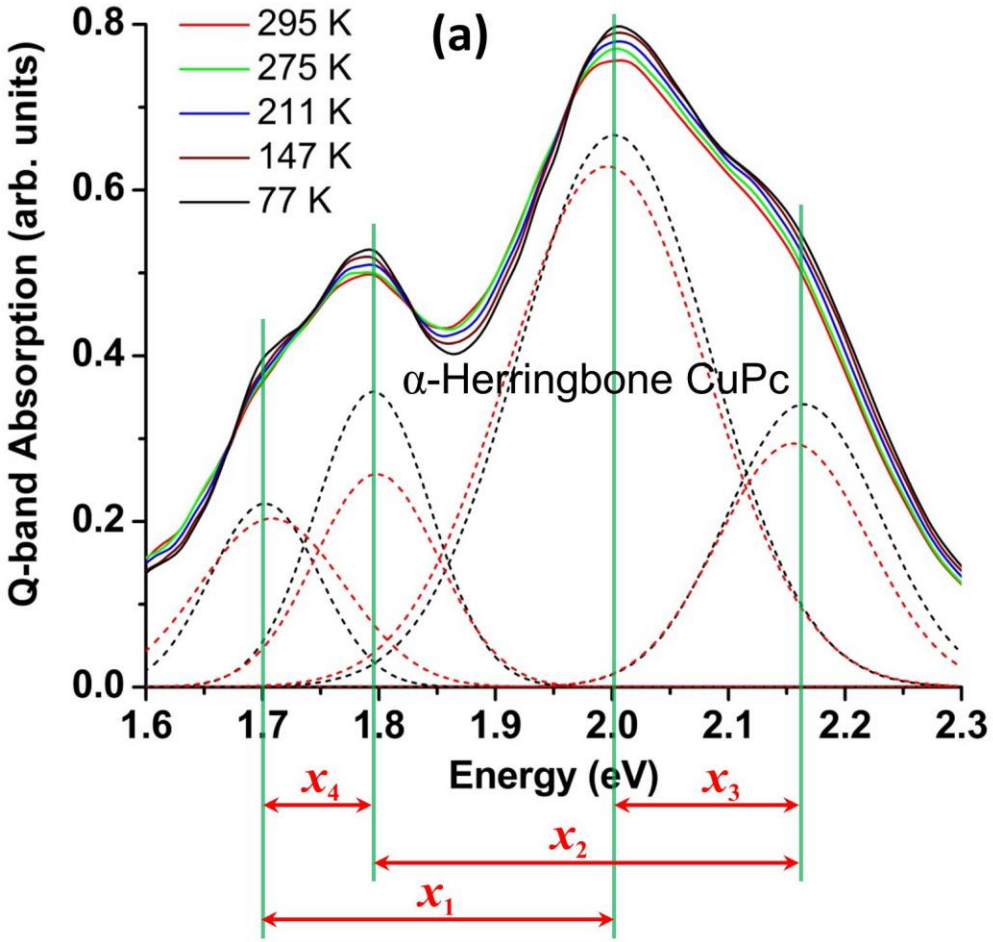
$$\hbar \omega_{v\mu}(k) = \hbar \omega_{v1,2}(k) = \hbar \omega_v^{(\pm)}(k) = \frac{1}{2} \left(D_k^{vv} + \Delta_{CT} \pm \sqrt{(D_k^{vv} - \Delta_{CT})^2 + 8(s_v \varepsilon_k)^2} \right),$$

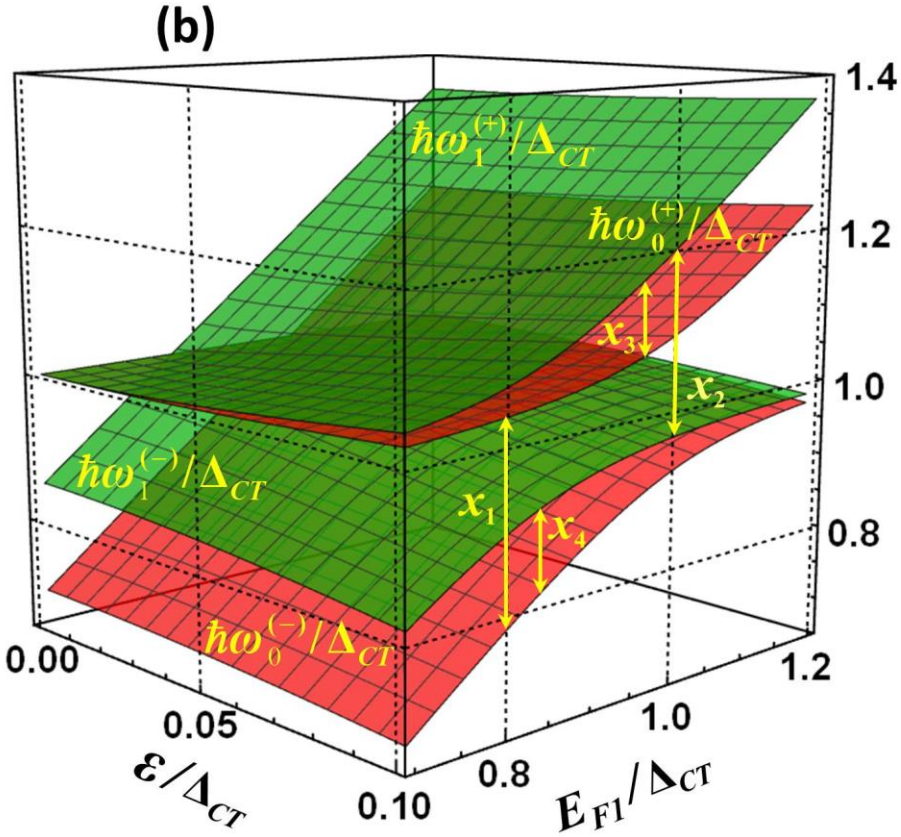
$$D_k^{vv'} = \Delta_F^v \delta_{vv'} + 2M s_v s_{v'} \cos k$$

$$E_{F1} = D_0^{00}, \quad E_{F2} = D_0^{11}, \quad E_v^{(\pm)} = \hbar \omega_v^{(\pm)}(0), \; v = 0,1$$

I.V.Bondarev, A.Popescu, R.A.Younts, B.Hoffman, T.McAfee, D.B.Dougherty, K.Gundogdu, and H.W.Ade, Appl. Phys. Lett. 109, 213302 (2016)

Comparison of the Theory with the Experiment





From the experimental graph:

$x_1 = 2.0 - 1.7 = 0.3$ eV,

$x_2 = 2.16 - 1.8 = 0.36$ eV,

$x_3 = 2.16 - 2.0 = 0.16$ eV,

$x_4 = 1.8 - 1.7 = 0.1$ eV $\Rightarrow$

From the k = 0 dispersion equation:

$$\Delta = E_{F2} - E_{F1} = x_3 + x_4; \quad x_1 + x_3 = x_2 + x_4$$

$$x_2^2 + x_1^2 = 2(E_{F1} - \Delta_{CT})^2 + \Delta[\Delta + 2(E_{F1} - \Delta_{CT})] + 16s^2\varepsilon^2$$

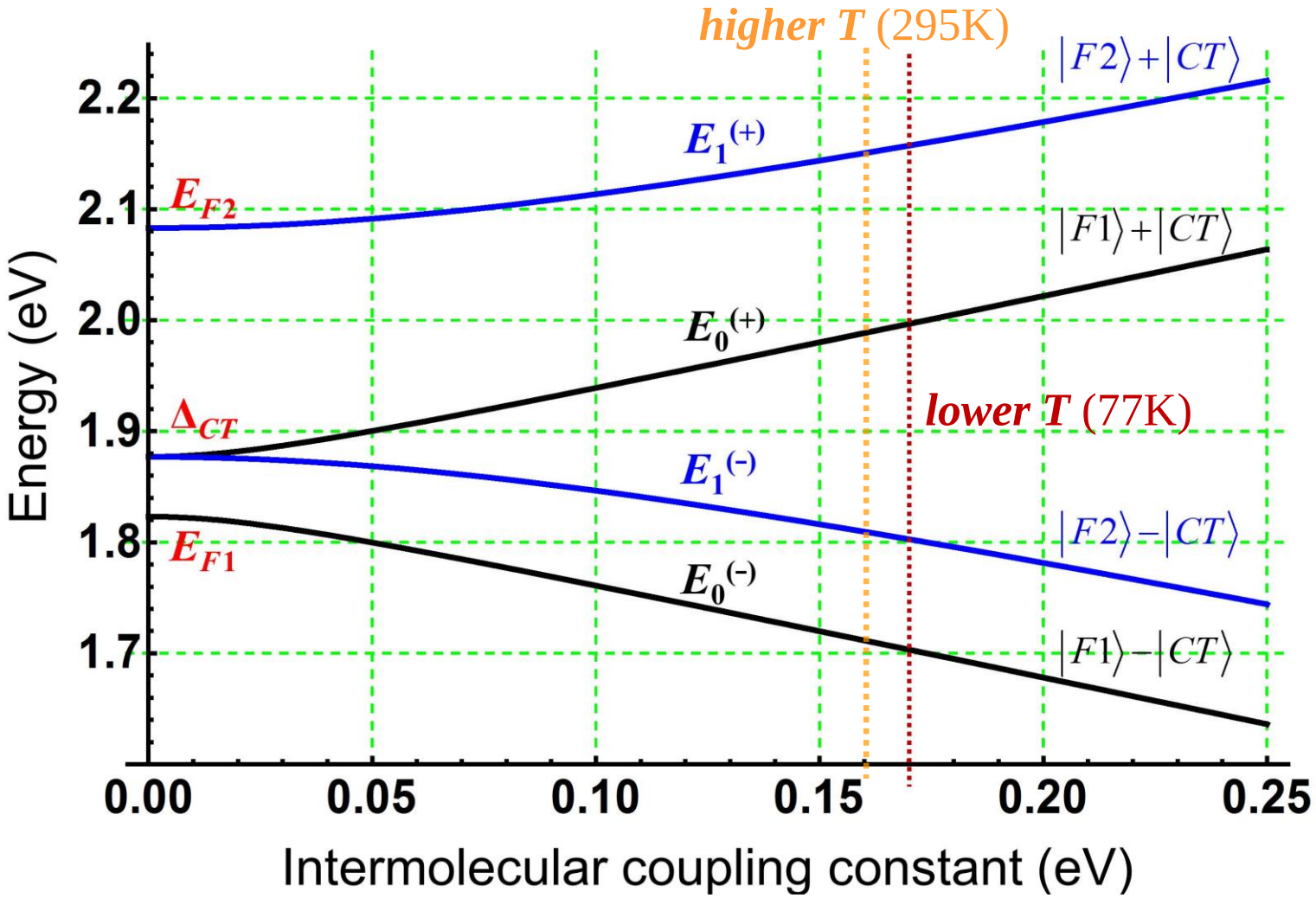
$$x_2^2 - x_1^2 = \Delta[\Delta + 2(E_{F1} - \Delta_{CT})]$$

$\Downarrow$

$E_{F1} = 1.82\,\text{eV}, E_{F2} = 2.08\,\text{eV}, \Delta_{CT} = 1.88\,\text{eV}, \varepsilon = 0.17\,\text{eV}$

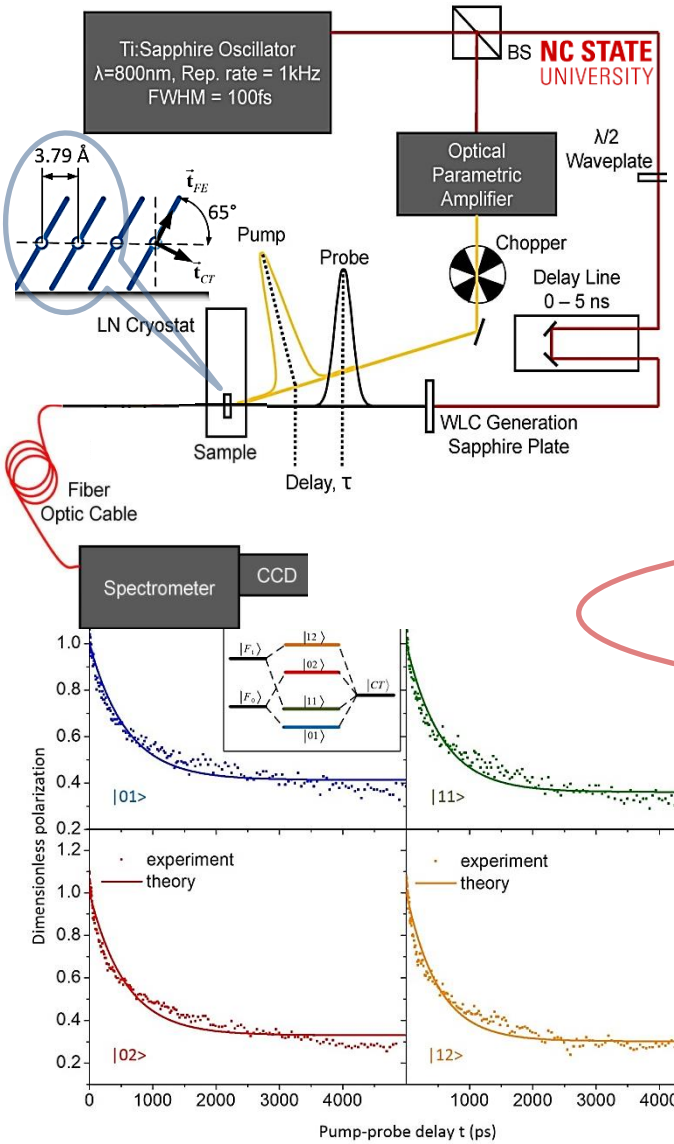
Calculated Frenkel-CT Exciton Splitting and Intermixing

as a function of the Intermolecular Coupling Constant

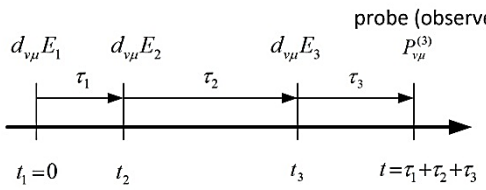


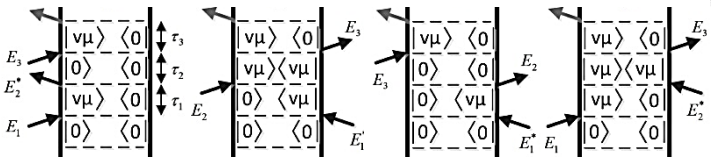
I.V.Bondarev, A.Popescu, R.A.Younts, B.Hoffman, T.McAfee, D.B.Dougherty, K.Gundogdu, and H.W.Ade, Appl. Phys. Lett. 109, 213302 (2016)

Monitoring Charge Separation Processes in Crystalline Organic Molecular Semiconductor Thin Films (CuPc)



$$P^{(3)}_{\nu\mu}(t) = \int_0^\infty d\tau_1 \int_0^\infty d\tau_2 \int_0^\infty d\tau_3 \, P_{\nu\mu}(\tau_3,\tau_2,\tau_1)\, E_1(t-\tau_3-\tau_2-\tau_1) E_2(t-\tau_3-\tau_2) E_3(t-\tau_3)$$





$$P_{\nu\mu}(t) = 8\frac{i}{\hbar^3} d^4_{\nu\mu} e^{-t/\tau_{\nu\mu}} \qquad 0 \le t \le \tau_2$$

$$\vec{P}_{\nu\mu}(t) = \langle \nu\mu | \hat{\vec{\mathcal{P}}}(t) | 0 \rangle \qquad \vec{P}_{\nu\mu}(t) = P^{\mathrm{FE}}_{\nu\mu}(t)\vec{\mathbf{t}}_{\mathrm{FE}} + P^{\mathrm{CT}}_{\nu\mu}(t)\vec{\mathbf{t}}_{\mathrm{CT}} \qquad \nu = 0,\ 1; \quad \mu = 1,\ 2$$

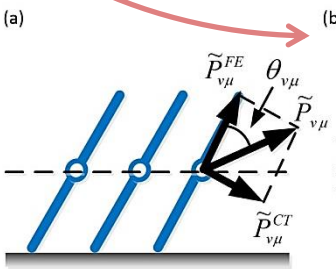
$$\tilde{P}^{\mathrm{FE}}_{\nu\mu}(t) = [1 - \tilde{P}^{\mathrm{FE}}_{\nu\mu}(\tau_2)] e^{-t/\tau_{\nu\mu}} + \tilde{P}^{\mathrm{FE}}_{\nu\mu}(\tau_2)$$

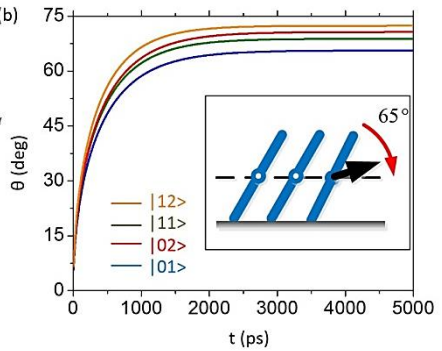
$$\theta_{\nu\mu}(t) = \tan^{-1}\left[\frac{\tilde{P}^{\mathrm{CT}}_{\nu\mu}(t)}{\tilde{P}^{\mathrm{FE}}_{\nu\mu}(t)}\right] = \tan^{-1}\frac{\sqrt{1-\tilde{P}^{\mathrm{FE}}_{\nu\mu}(t)^2}}{\tilde{P}^{\mathrm{FE}}_{\nu\mu}(t)}$$

$$\tilde{P}^{\mathrm{FE}}_{\nu\mu}(t) = P^{\mathrm{FE}}_{\nu\mu}(t)/P^{\mathrm{FE}}_{\nu\mu}(0)$$

$$\tilde{P}^{\mathrm{CT}}_{\nu\mu}(t) = P^{\mathrm{CT}}_{\nu\mu}(t)/P^{\mathrm{FE}}_{\nu\mu}(0)$$

A.Popescu, R.A.Younts, B.Hoffman, T.McAfee, D.B.Dougherty, H.W.Ade, K.Gundogdu, and I.V.Bondarev, Nano Letters 17, 6056 (2017)





Frenkel (single-molecule) exciton states spread over time towards CT (inter-molecular) states to turn the overall polarization along the 1D crystal axis – ***We monitor the process of the charge separation in real time !!!***

SUMMARY

- **TOPIC I: Transdimensional Effects in Ultrathin Metallic Films of Finite Thickness**
  - **Plasma frequency spatial dispersion to result in nonlocal/anisotropic optical response**
  - **Negative refraction, resonance magnetic response, and novel QED effects – largely enhanced/unidirectional spontaneous emission due to ENZ mode degeneracy lifting**
  - ***FUTURE: Quantum nanooptoplasmonics, gyrotropic hyperbolic metasurfaces, …***

- **TOPIC II: Interlayer (Indirect) Excitons and Exciton Complexes in Layered Semiconductors**
  - **The configuration space method to evaluate the ground-state binding energies of the neutral and charged exciton complexes (biexciton and trion)**
  - **Quasiclassical theory developed to confirm the experimental spectroscopic evidence for Quaternion *e-h* states (doubly-charged boson) in bilayer systems with metal layers**
  - ***FUTURE: Non-linear optics, spin-optronics, BEC & unconventional superconductivity, …***

- **TOPIC III: Organic Molecular Semiconductor Crystals with Two Frenkel Exciton States**
  - **Frenkel-Charge-Transfer intermixing theory to explain optical spectroscopy data**
  - ***FUTURE: Mechanically stable organic semiconductors for advanced optoelectronics***

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THANK YOU !!