THE UNIVERSITY OF CHICAGO

arXiv: 1912.02822

Kavli Institute
for Cosmological Physics
at The University of Chicago

New Constraints on Dark Matter from Organic Targets: What Organic Chemistry can do for Direct Detection

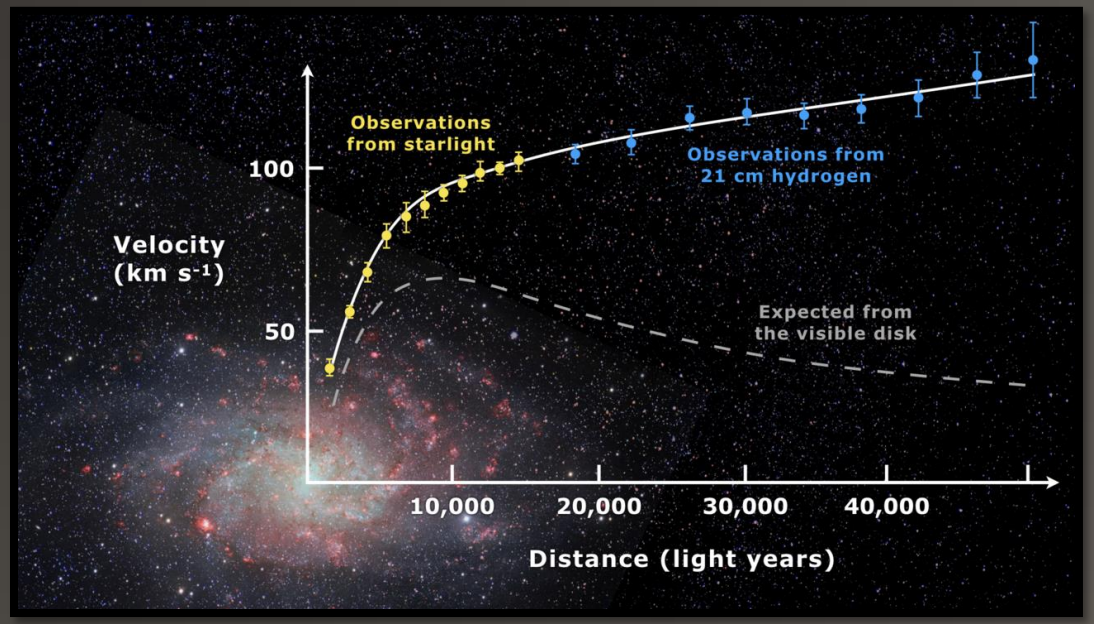
CARLOS BLANCO

Based on work with J. Collar, Y. Kahn, & B. Lillard

arXiv: 1912.02822

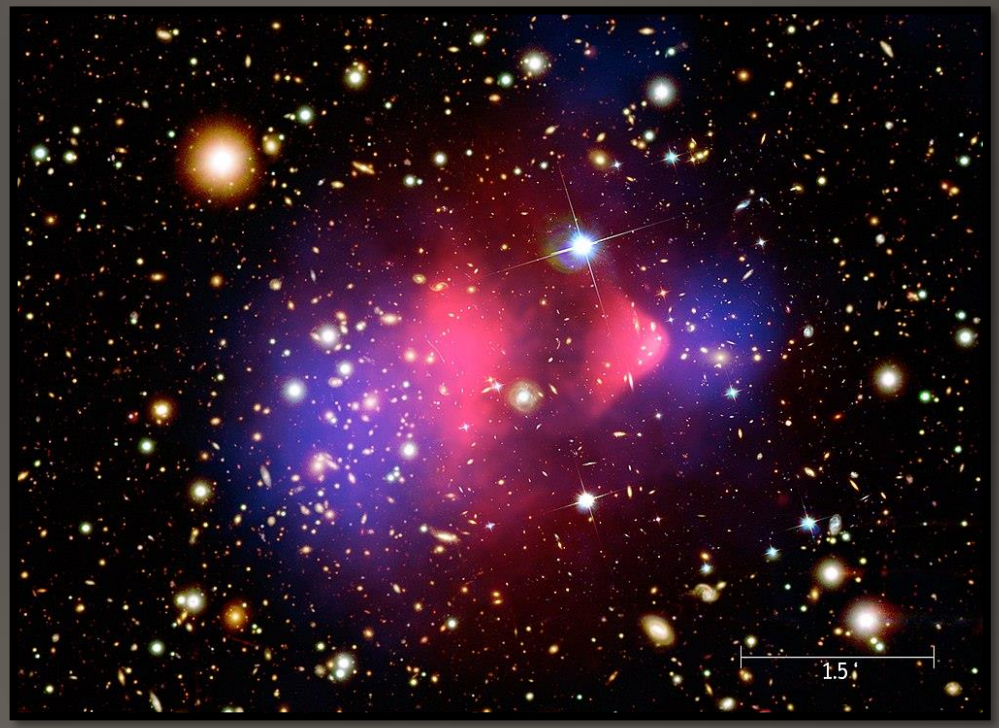
Evidence for Dark Matter

Galactic rotation curves



Solid: Observed
Dashed: Expected from visible disk

Bullet cluster



Blue: Mass from grav lensing
Pink: Baryonic mass from x-rays

X-ray: NASA/CXC/CfA/M.Markevitch et al.;
Optical: NASA/STScI; Magellan/U.Arizona/D.Clowe et al.;
Lensing Map: NASA/STScI; ESO WFI; Magellan/U.Arizona/D.Clowe et al.

Corbelli & Salucci, MNRAS 1999
Bosma, Astronomical Journal, 1989

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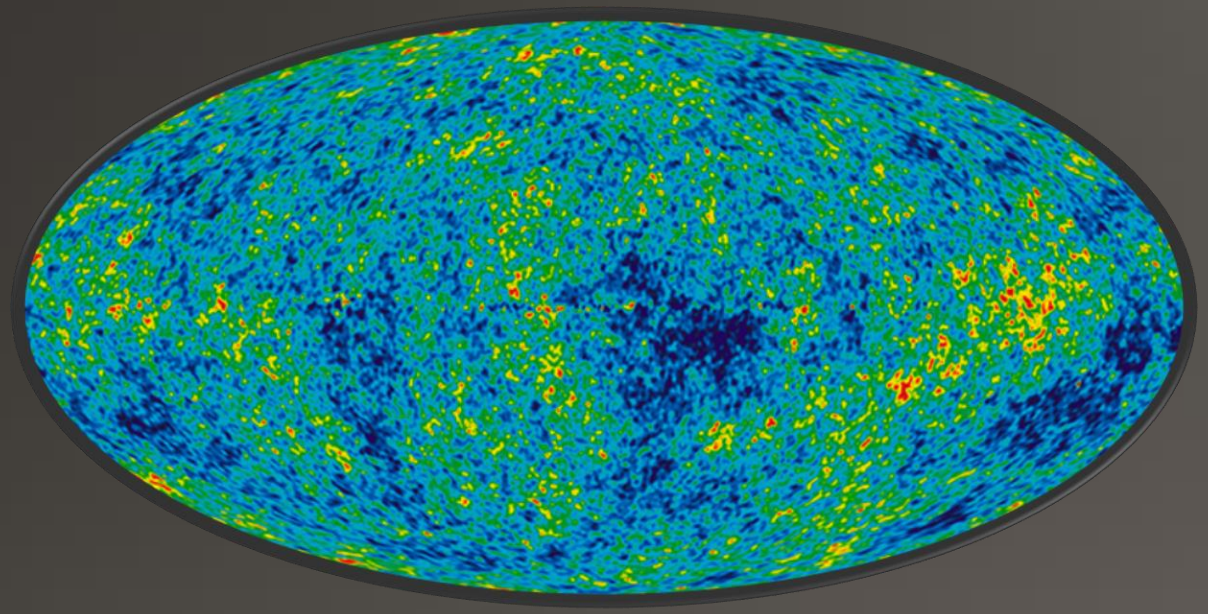
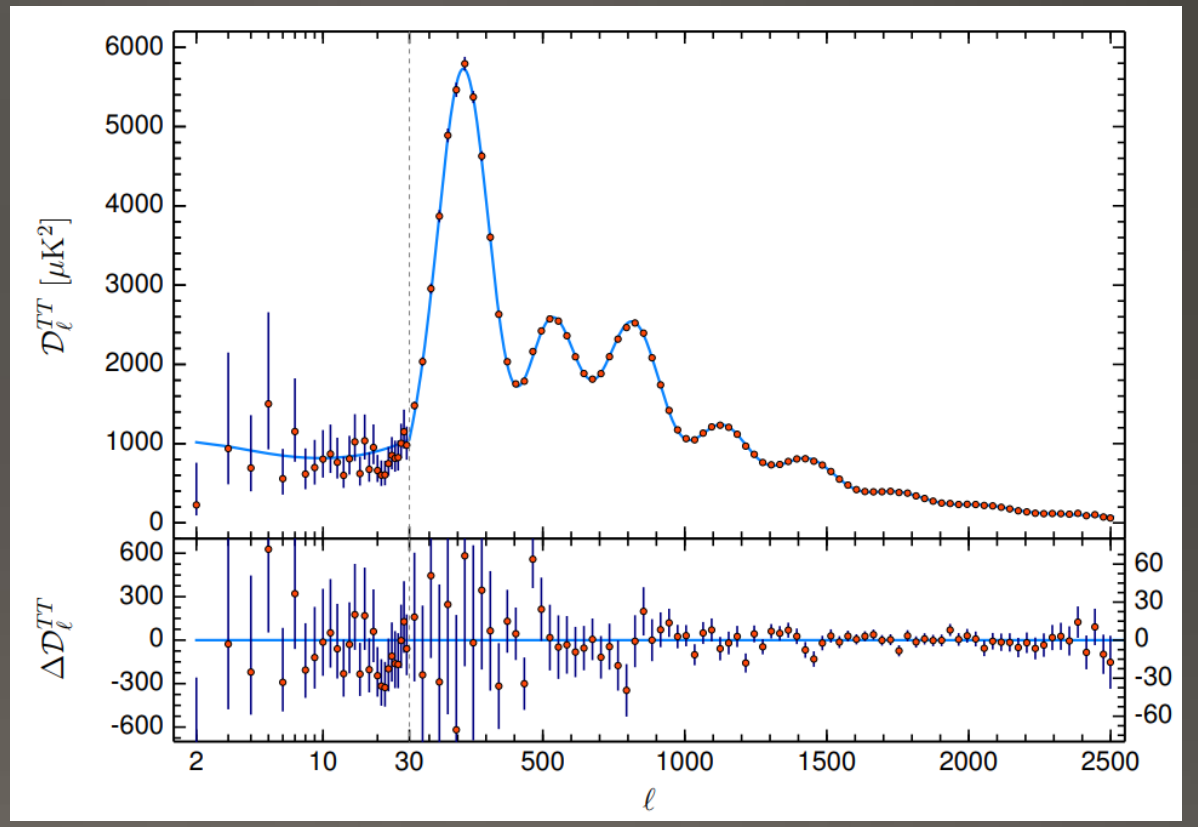
Evidence for Dark Matter



$$\Omega_c = 0.1200 \pm 0.0012$$



Planck results: A&A 2019

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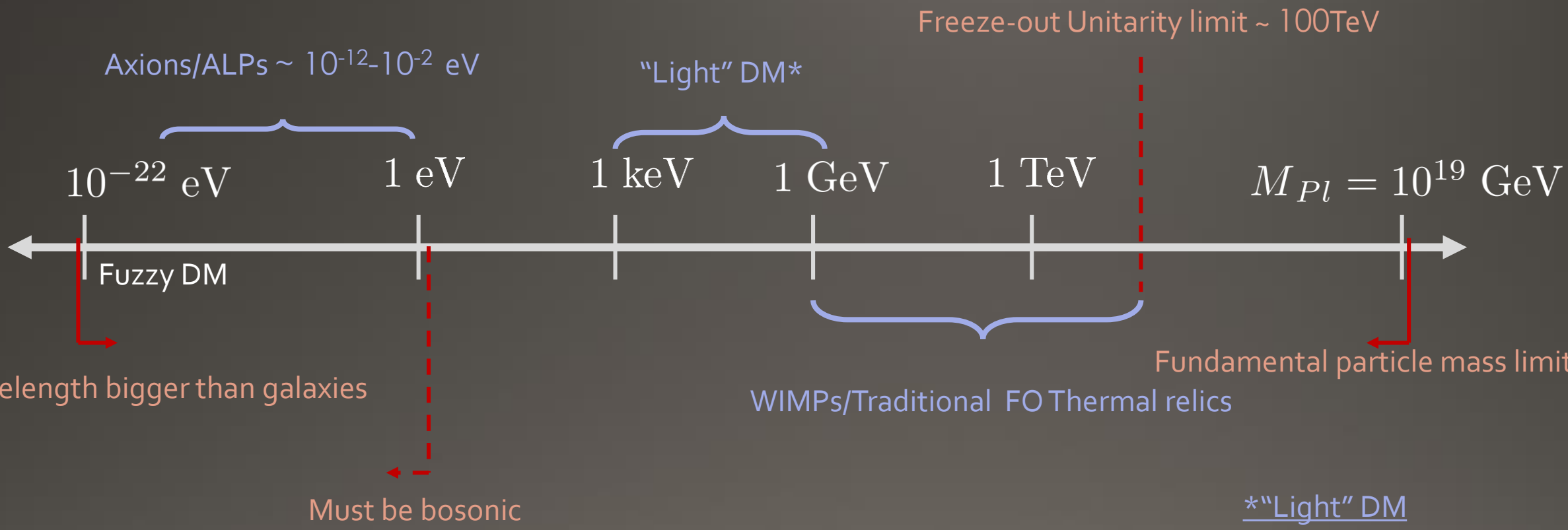
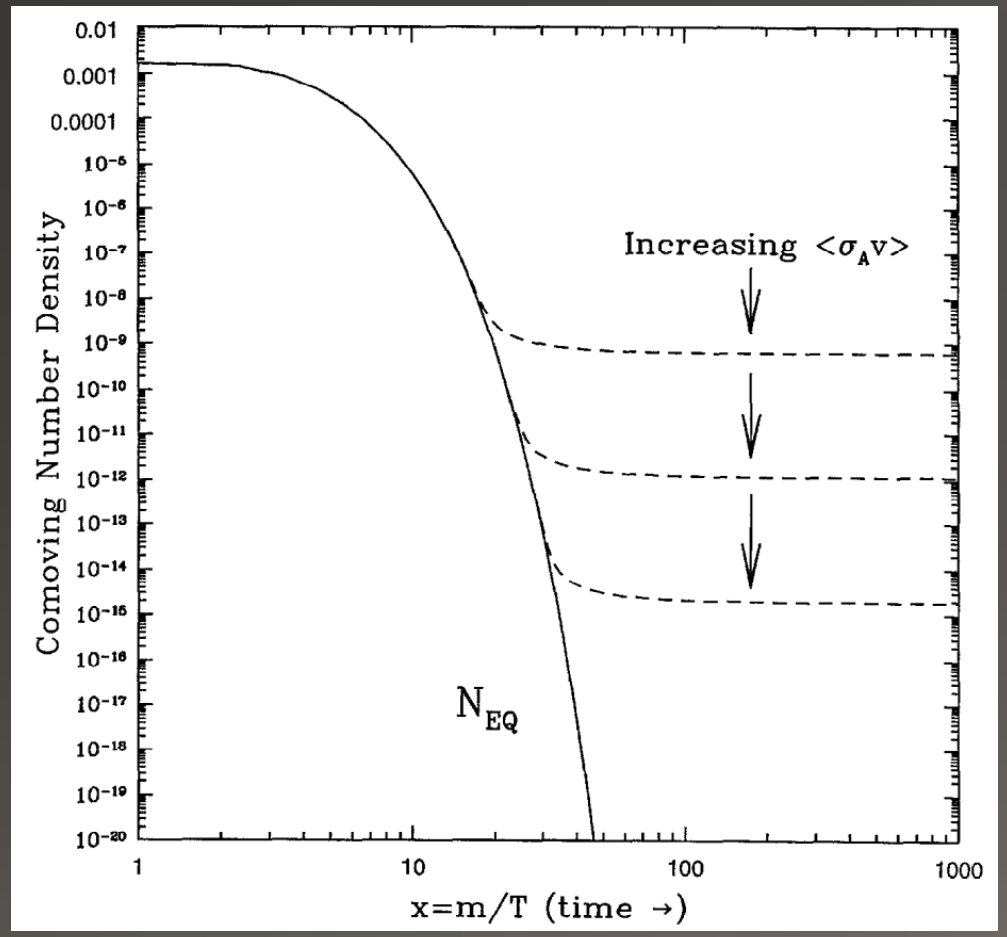
Dark Matter Mass Range



(NOT TO SCALE)

*"Light" DM
- Assymetric DM
- Hidden sector
- ELDER/SIMPs
- Freeze-In
- ...etc

WIMPs: Thermal Freeze-Out



Griest et. al: Phys Rep. 1996

$$\Omega_c h^2 \sim \frac{1}{\langle \sigma v \rangle}$$

Known from Planck

Predicts:

$$\langle \sigma v \rangle_{FO} \approx 3 \times 10^{-26}\ \mathrm{cm}^3 \mathrm{s}^{-1}$$

Weak scale! For masses between 1 GeV and 100 TeV

Subject to constraints from :

- CMB (recombination)
- Fermi-LAT (Dwarf spheroidal galaxy observations)
- BBN
- LEP, LHC, AMS,...etc

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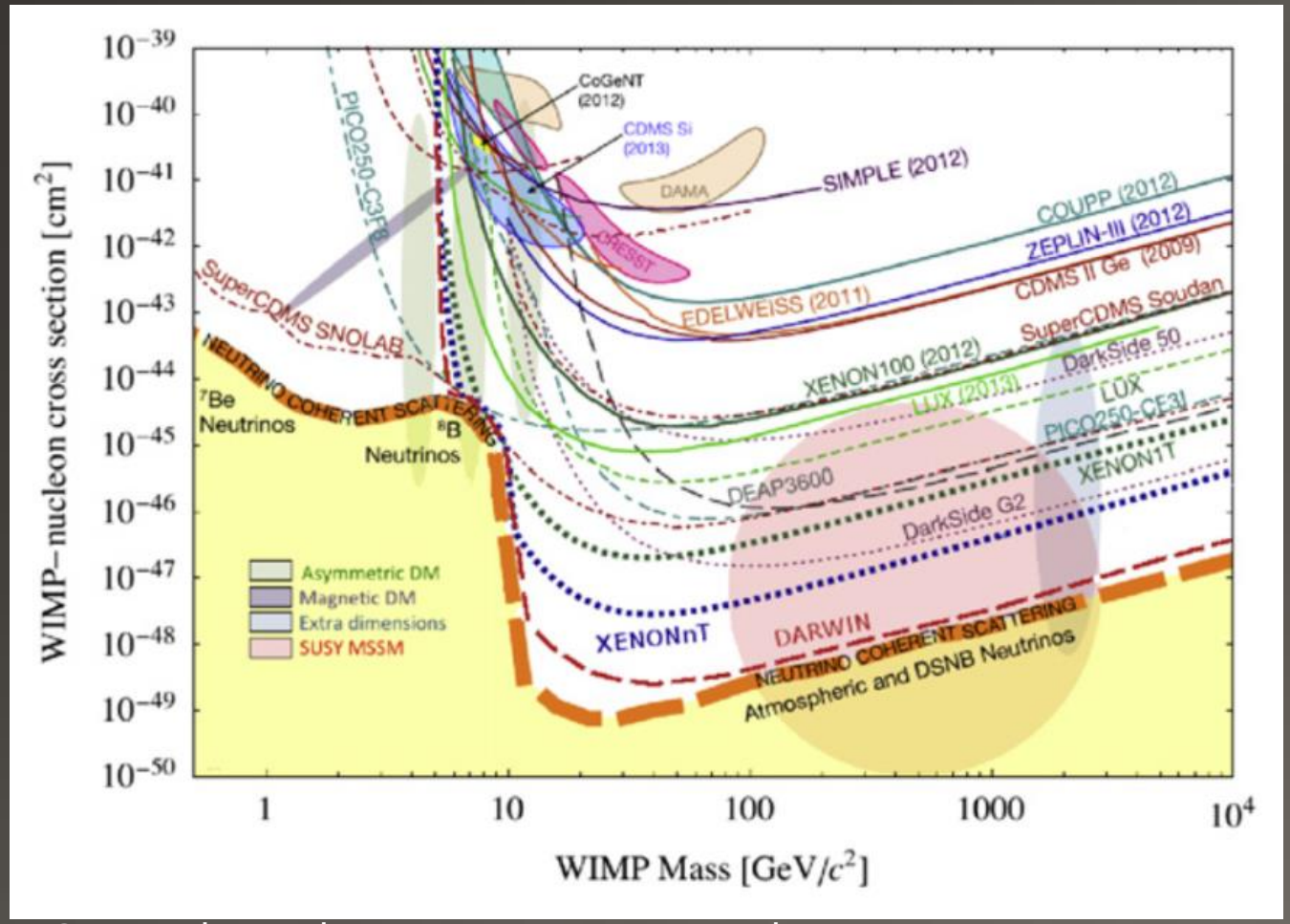
Current Direct Detection



Spin-independent cross section to nucleons

- Noble gasses: Very effective at probing masses above 1 GeV
- Will encounter irreducible neutrino floor from
- Linear at high masses due to number density
- Loss of sensitivity due to kinematics at low masses

L. Baudis Phys D. U. 2014

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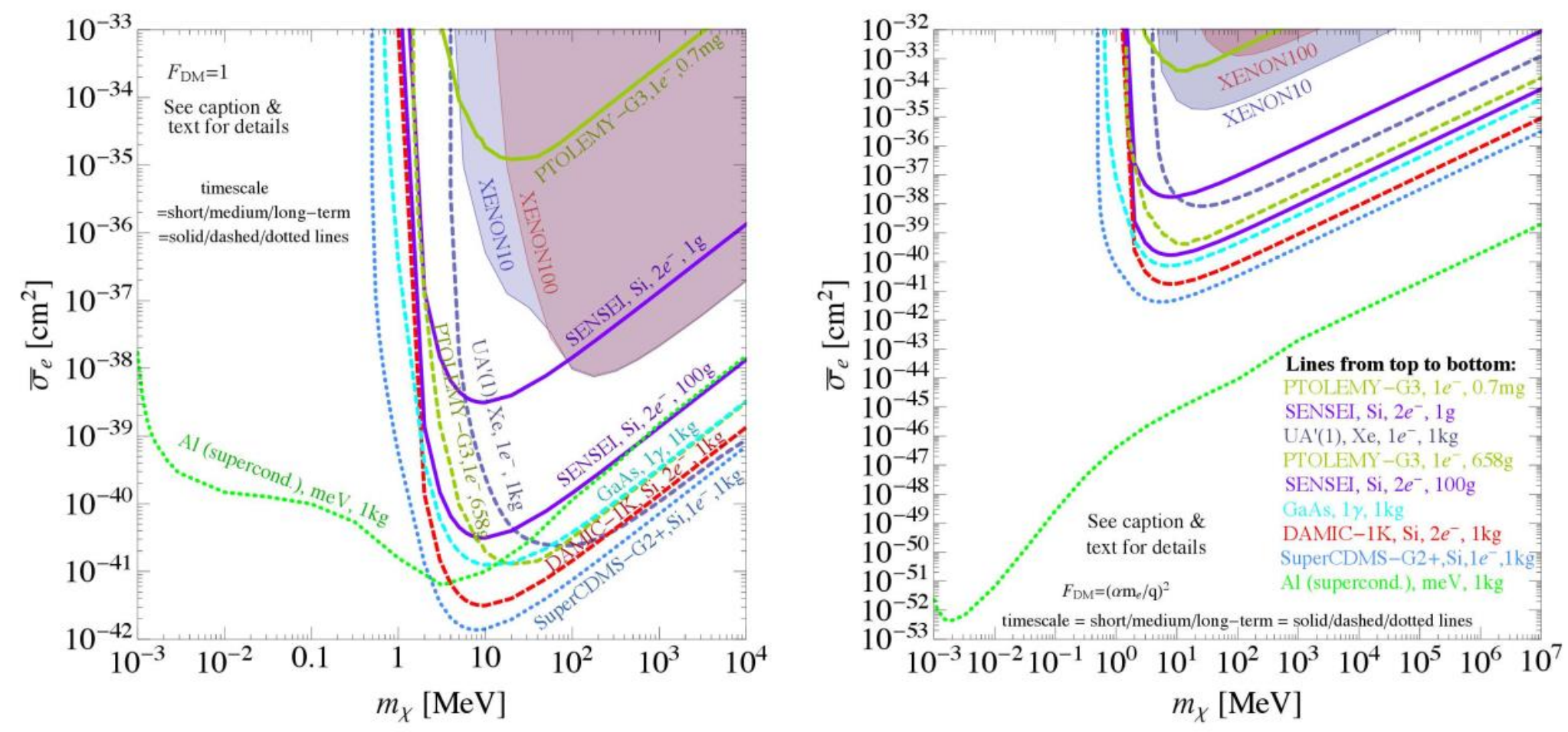
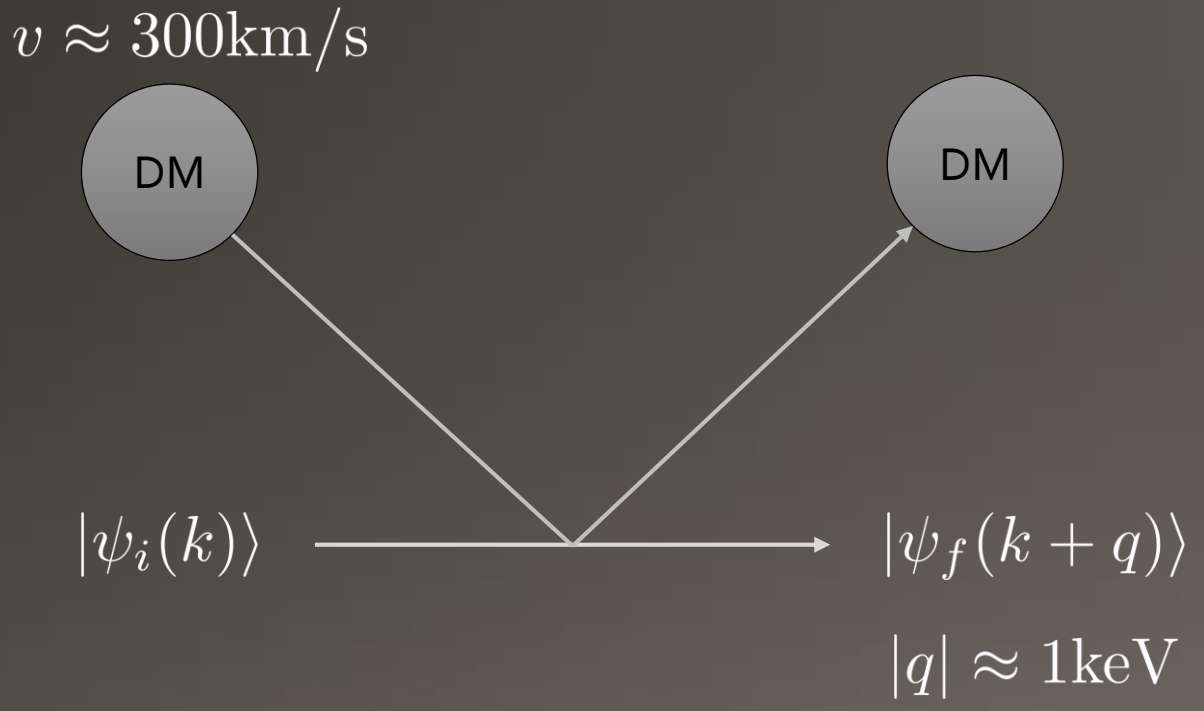
Sub-GeV Dark Matter (e^- scattering)



Cosmic Visions, ArXiv: 1707.04591

Light DM-electron scattering



Kinematic Requirement

$$\Delta E = \vec{v}_\chi \cdot \vec{q} - \frac{q^2}{2\mu_{\chi \mathrm{m}}}$$

$$\Delta E \sim \mathcal{O}(\text{few eV})$$

What has such transition energies?

- Semiconductor bandgaps
- Inorganic crystal scintillators
- Organic liquid scintillators

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DM-Electron Interaction Rate

Photon Rate

$$R = \xi \frac{2N_B \rho_\chi \bar{\sigma}_e}{8\pi m_\chi \mu_{\chi e}^2} \int \frac{d^3\mathbf{q}}{q} \eta\left(v_{\min}\right) F_{\mathrm{DM}}^2(q) \sum_i \left|f_i(\mathbf{q})\right|^2$$

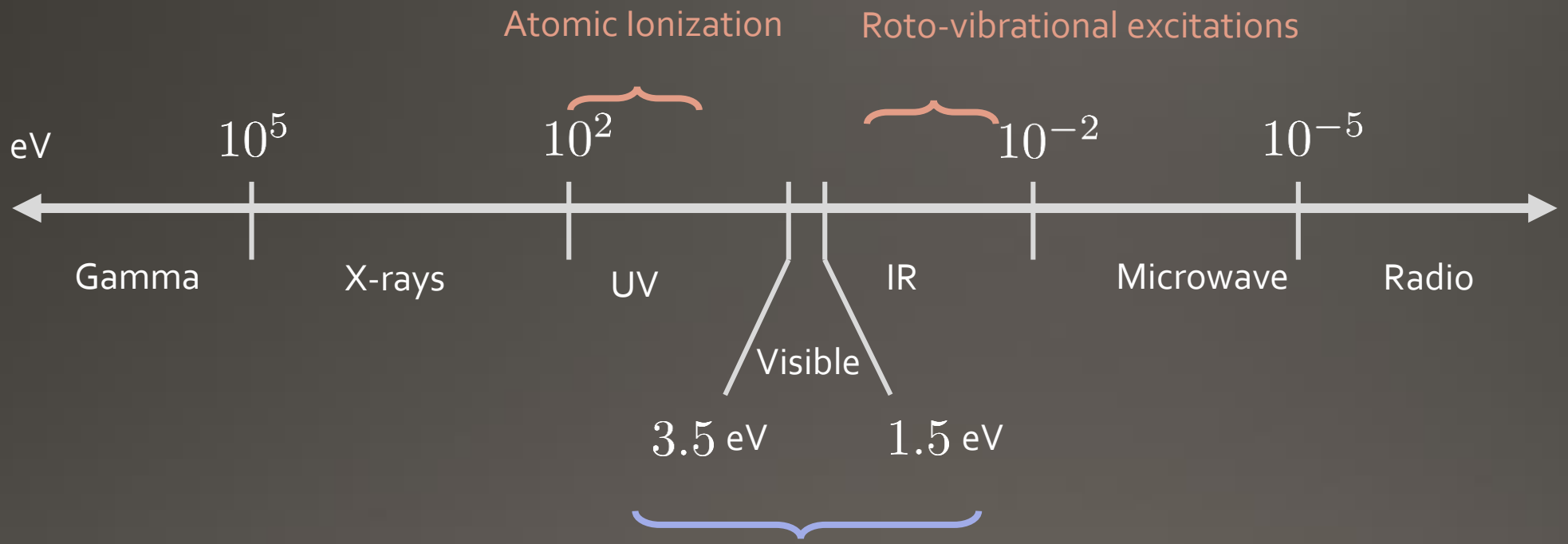
Sensitive when R > ~0.1Hz

ρ_χ :DM density ξ :detector efficiency

Molecular form factor

$$f_i(\mathbf{q}) = \int d^3\mathbf{p} \tilde{\psi}_n(\mathbf{p}) \tilde{\psi}^\star_{n+i}(\mathbf{p}+\mathbf{q})$$

Low Energy Transitions



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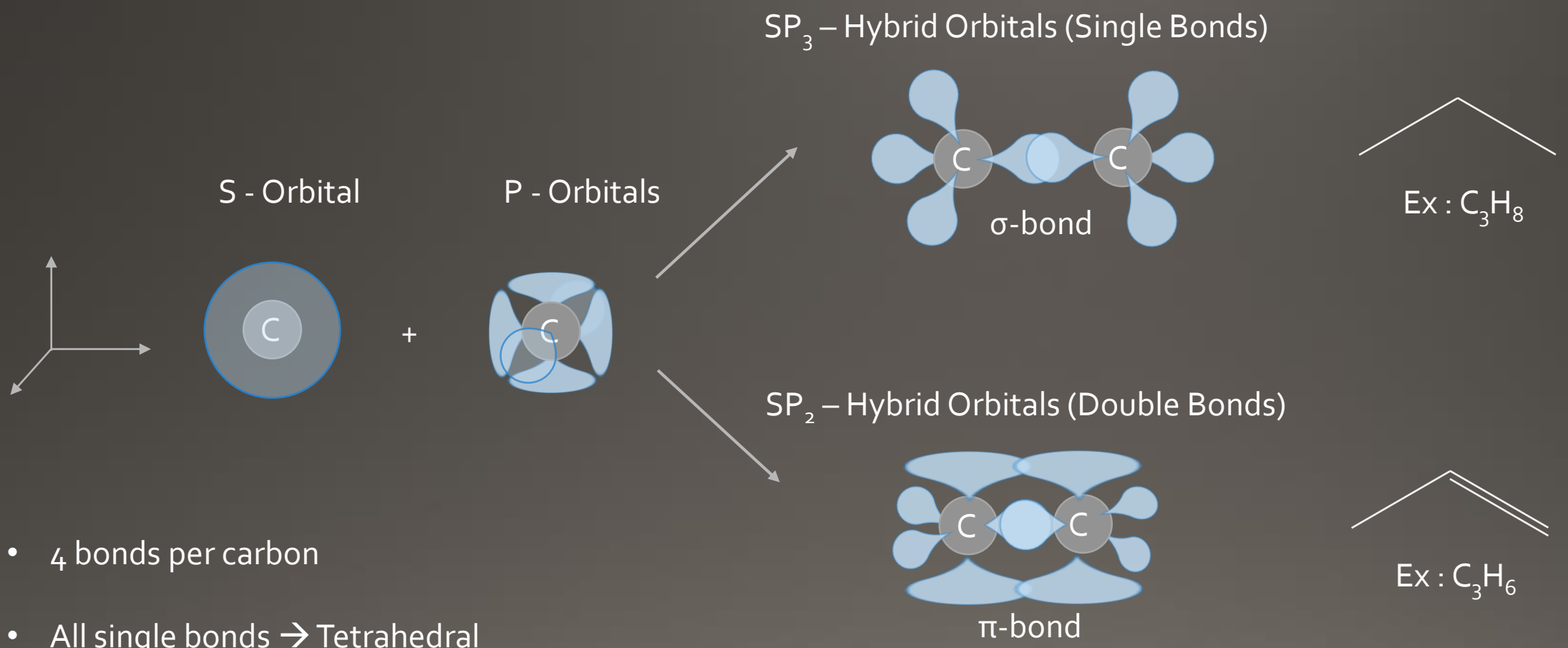
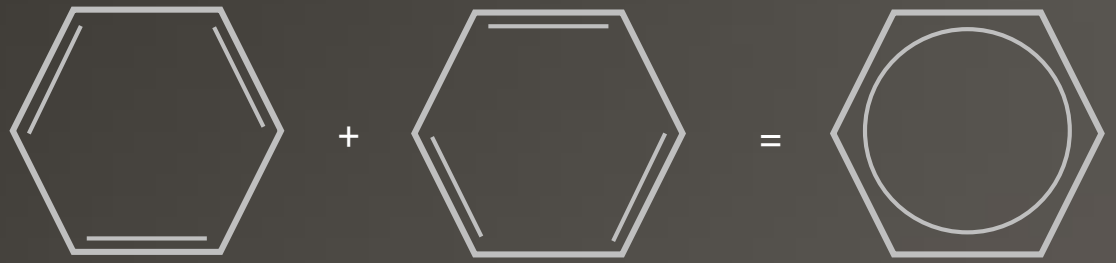
Organic Chemistry for Physicists: Orbitals



- 4 bonds per carbon
- All single bonds → Tetrahedral
- Double bond → Planar

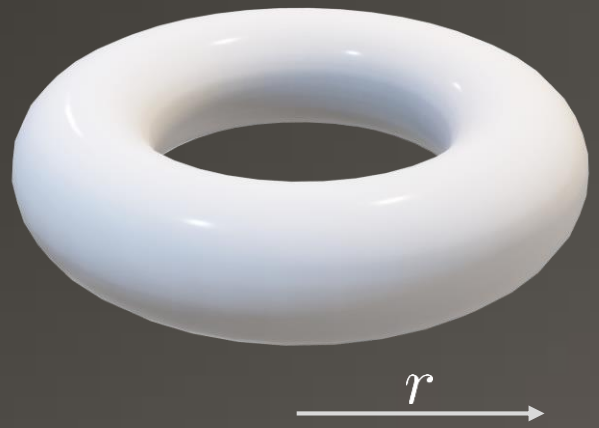
Organic Chemistry for Physicists: Benzene



- Two Kekule resonance forms
- Combined give an "aromatic" ring of delocalized charge
- π-electrons free to move from atom to atom
- σ-electrons still well localized in SP_2 orbital

Free electron model

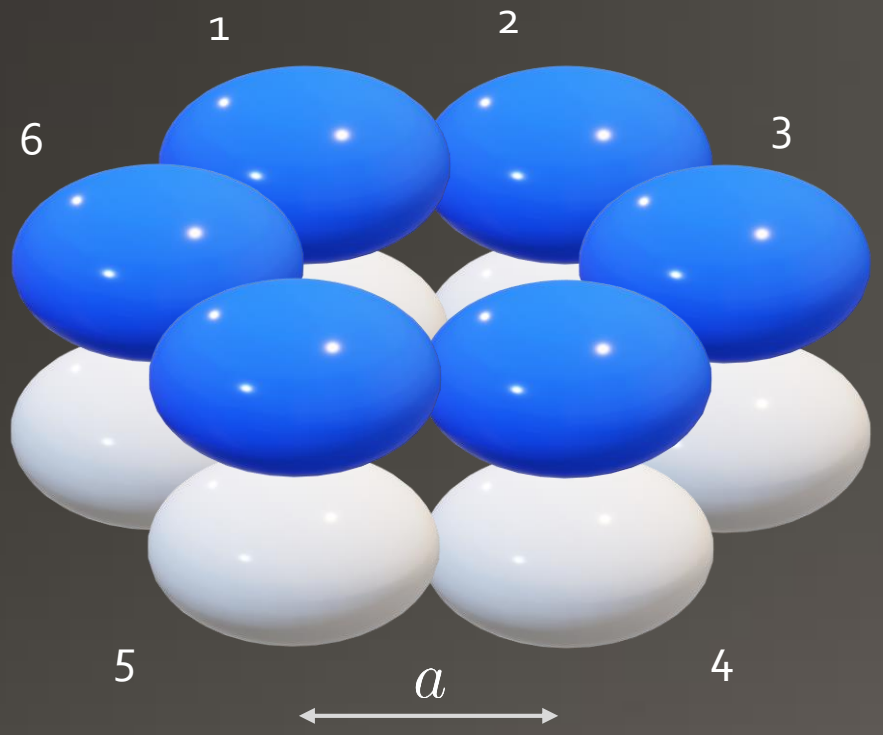
1D ring of charge



$$\Psi_n = (\tfrac{2}{r})^{1/2} \sin \frac{\pi n x}{r} \qquad E_n = \frac{h^2 n^2}{8 m r^2}$$

- Let the π -electron travel on a one dimensional ring
- Well known solutions
- Quadratic spacing, two electrons per state
- No degenerate energy states
- Radius becomes an empirical parameter
- Not great for anything except benzene's first excitation

Linear Combination of Atomic Orbitals



Take linear combination of Slater atomic orbitals

$$\phi_{2p_z}(\mathbf{r}) = \mathcal{N} a_0^{-3/2} \frac{r\cos\theta}{a_0} \exp\left(\frac{-Z_{\text{eff}} r}{2a_0}\right)$$

One centered at each nuclei

$$\Psi_i = \sum_{i=1}^{6} c_i \phi_{2P_z}(\mathbf{r} - \mathbf{R}_i)$$

Solutions come from diagonalizing the Hamiltonian matrix

$$\sum_{i=1}^{6} \left[(H_{ji} - E_i \delta_{ji})\, c_i\right] = 0, \text{ for } j = 1, 2, \ldots, 6$$

$$H_{ji} = \langle \Psi_j | \mathcal{H} | \Psi_i \rangle$$

Energies depend only on on-site, H_{ii}, energy and nearest neighbor, H_{ij}, resonance integrals

$$\alpha \equiv H_{ii} \qquad \beta \equiv H_{ij}$$

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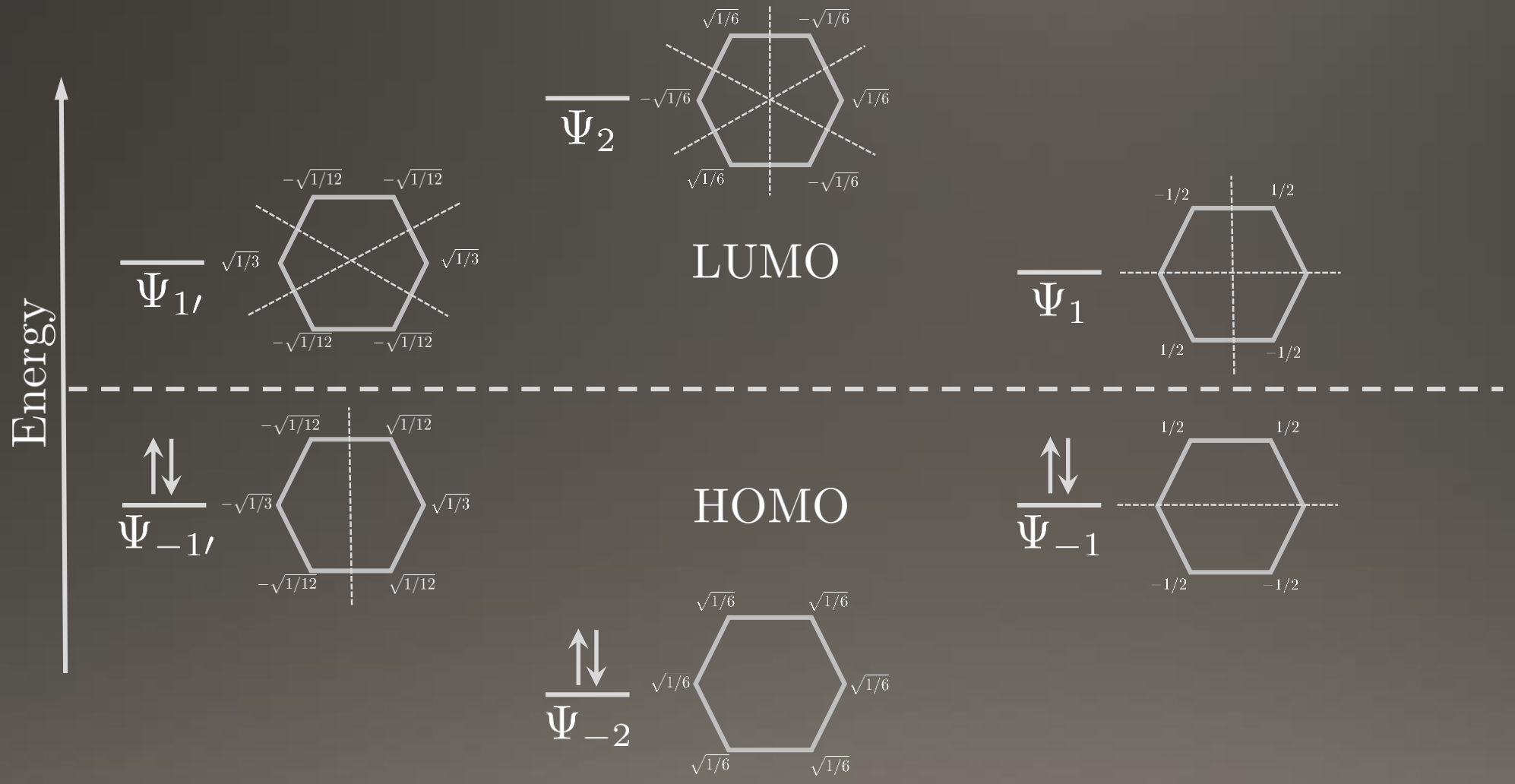
LCAO: Benzene Hückel solutions



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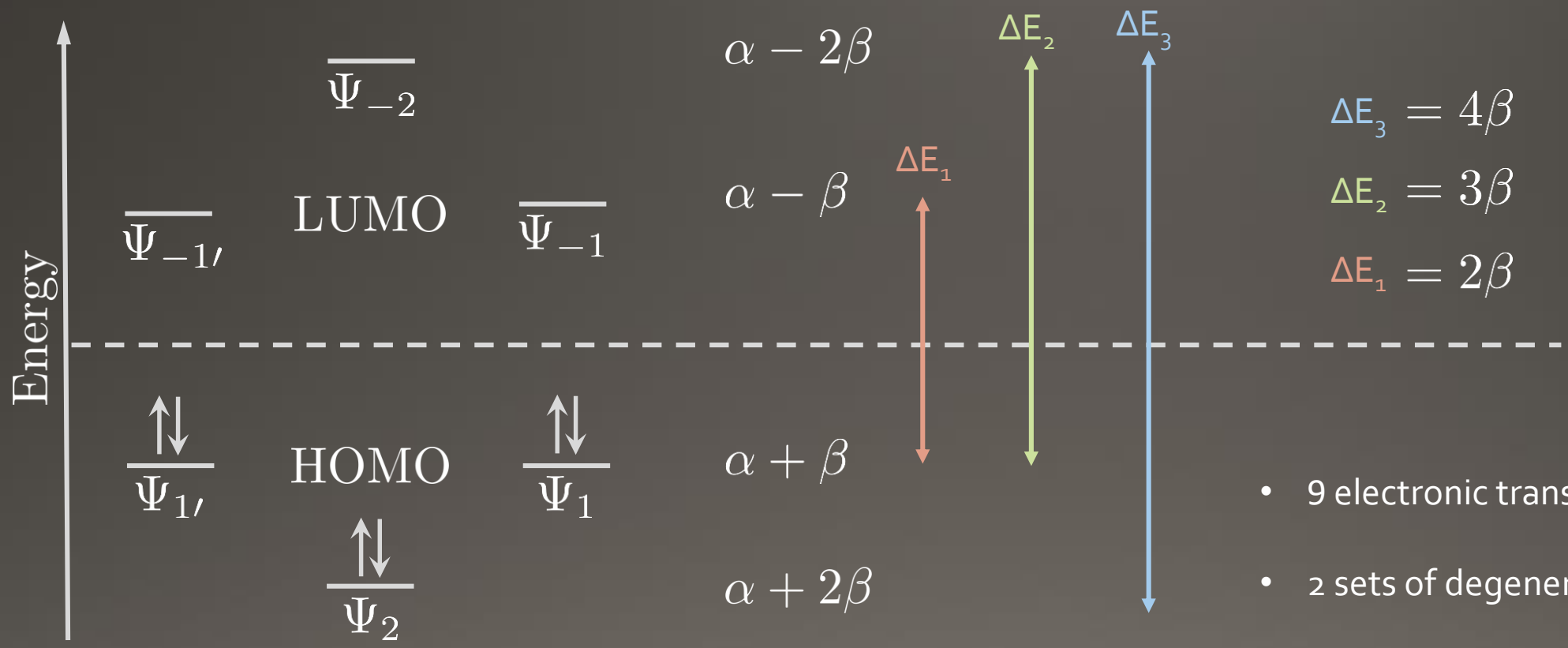
Hückel Transitions



$\Delta E_3 = 4\beta$

$\Delta E_2 = 3\beta$

$\Delta E_1 = 2\beta$

- 9 electronic transitions
- 2 sets of degenerate states
- 3 excitation energies

Anti-symmetry & Slater Determinants

Electrons need to live in anti-symmetric states! (exclusion principle)

$$\Psi\left(\mathbf{x}_1, \mathbf{x}_2, \ldots, \mathbf{x}_N\right) = \frac{1}{\sqrt{N!}} \begin{vmatrix} \chi_1\left(\mathbf{x}_1\right) & \chi_2\left(\mathbf{x}_1\right) & \cdots & \chi_N\left(\mathbf{x}_1\right) \\ \chi_1\left(\mathbf{x}_2\right) & \chi_2\left(\mathbf{x}_2\right) & \cdots & \chi_N\left(\mathbf{x}_2\right) \\ \vdots & \vdots & \ddots & \vdots \\ \chi_1\left(\mathbf{x}_N\right) & \chi_2\left(\mathbf{x}_N\right) & \cdots & \chi_N\left(\mathbf{x}_N\right) \end{vmatrix}$$

$$\equiv |\chi_1, \chi_2, \cdots, \chi_N\rangle$$

$$\equiv |1, 2, \ldots, N\rangle$$

Example, a simple 2 particle state:

$$\Psi\left(\mathbf{x}_1, \mathbf{x}_2\right) = \frac{1}{\sqrt{2}}\left\{\chi_1\left(\mathbf{x}_1\right)\chi_2\left(\mathbf{x}_2\right) - \chi_1\left(\mathbf{x}_2\right)\chi_2\left(\mathbf{x}_1\right)\right\}$$

$$= \frac{1}{\sqrt{2}} \begin{vmatrix} \chi_1\left(\mathbf{x}_1\right) & \chi_2\left(\mathbf{x}_1\right) \\ \chi_1\left(\mathbf{x}_2\right) & \chi_2\left(\mathbf{x}_2\right) \end{vmatrix}$$

Pariser, Pople, & Parr Method

$$\mathcal{H}_{ppp} = \mathcal{H}_{core} + \sum_{ij} \frac{e^2}{r_{ij}}$$

Hückel "core" Hamiltonian

Electron repulsion term

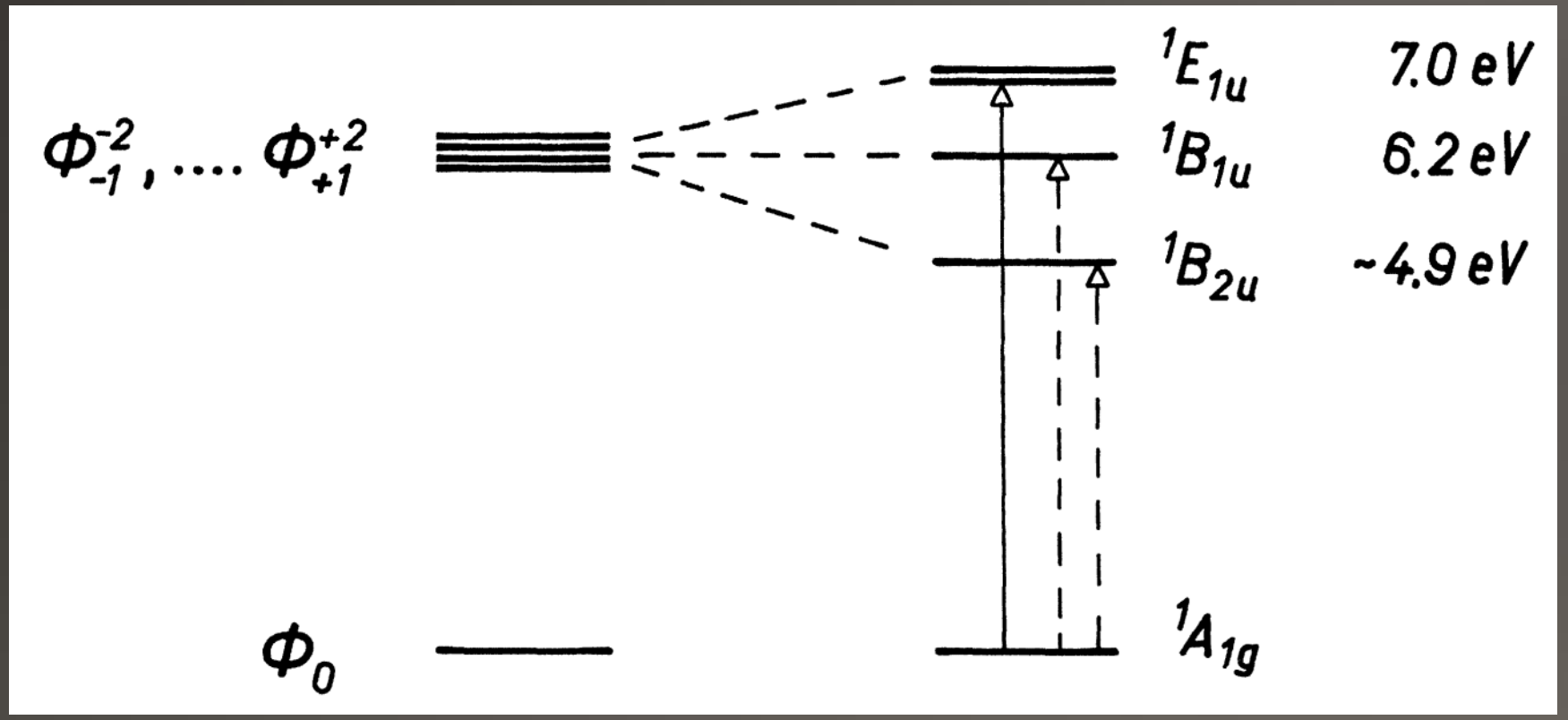
1st singlet excitations

$$\Psi_1^{s0} = 1/\sqrt{2}\,(\Psi_1^{-1} + \Psi_{1'}^{-1'})$$
$$\Psi_2^{s0} = 1/\sqrt{-2}\,(\Psi_1^{-1} - \Psi_{1'}^{-1'})$$
$$\Psi_3^{s0} = 1/\sqrt{2}\,(\Psi_1^{-1'} + \Psi_{1'}^{-1})$$
$$\Psi_4^{s0} = 1/\sqrt{-2}\,(\Psi_1^{-1'} - \Psi_{1'}^{-1})$$

$$\Psi_G = |\psi_2\overline{\psi_2}\psi_1\overline{\psi_1}\psi_{1'}\overline{\psi_{1'}}|$$

$$\Psi_i^j = \frac{1}{\sqrt{2}}(|\psi_1\overline{\psi_1}...\psi_i\overline{\psi_j}...\psi_N\overline{\psi_N}| - |\psi_1\overline{\psi_1}...\psi_j\overline{\psi_i}...\psi_N\overline{\psi_N}|)$$

PPP Transitions



Wagniere, 1976

Hückel excitations

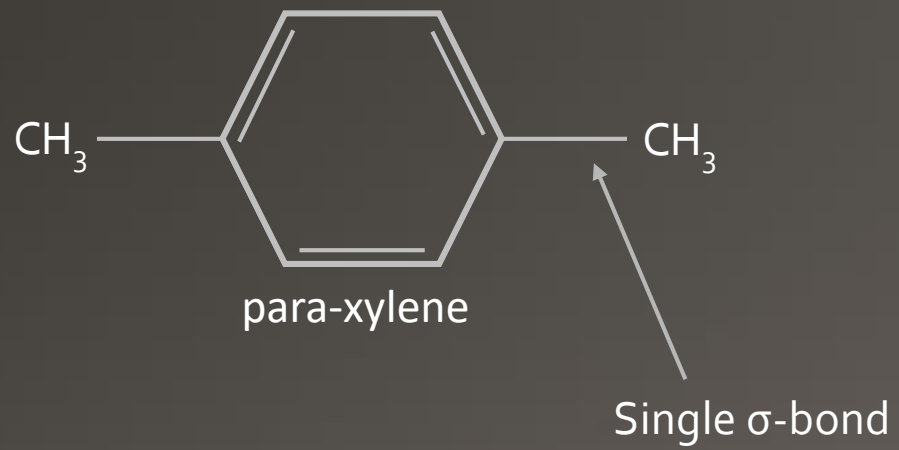
PPP excitations

1st singlet excitations

- Transform as symmetry subgroups of the symmetry group of Benzene
- Splitting due to electron repulsion, "Configurational Interactions"

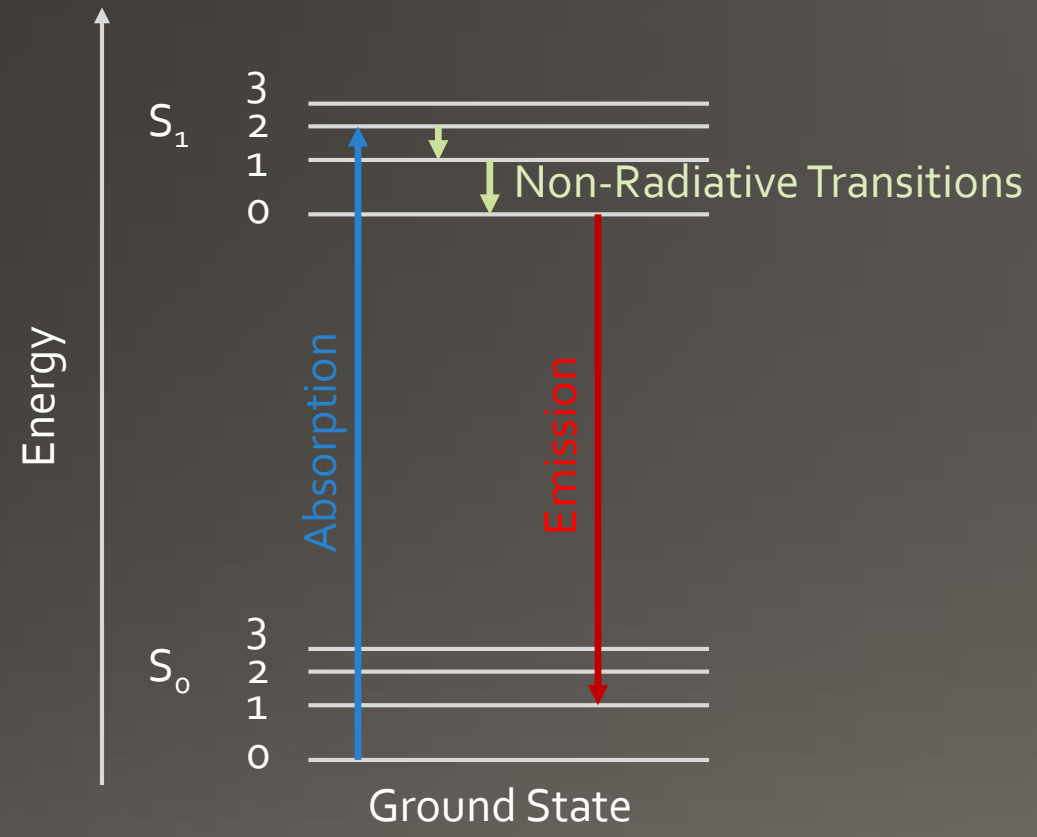
Extension to p-xylene (EJ-301)

EJ-301
Solvent: para-xylene
Fluor: 5% by mass



- Absorption spectra of p-xylene is well described by slightly perturbed aromatic peaks
- Expected since methyl groups don't affect LCAO at leading order
- Produce perturbation at ~5% level in energy,
  - Benzene HOMO/LUMO gap = 4.9 eV
  - p-xylene HOMO/LUMO gap = 4.7 eV

Fluorescence: Quick review

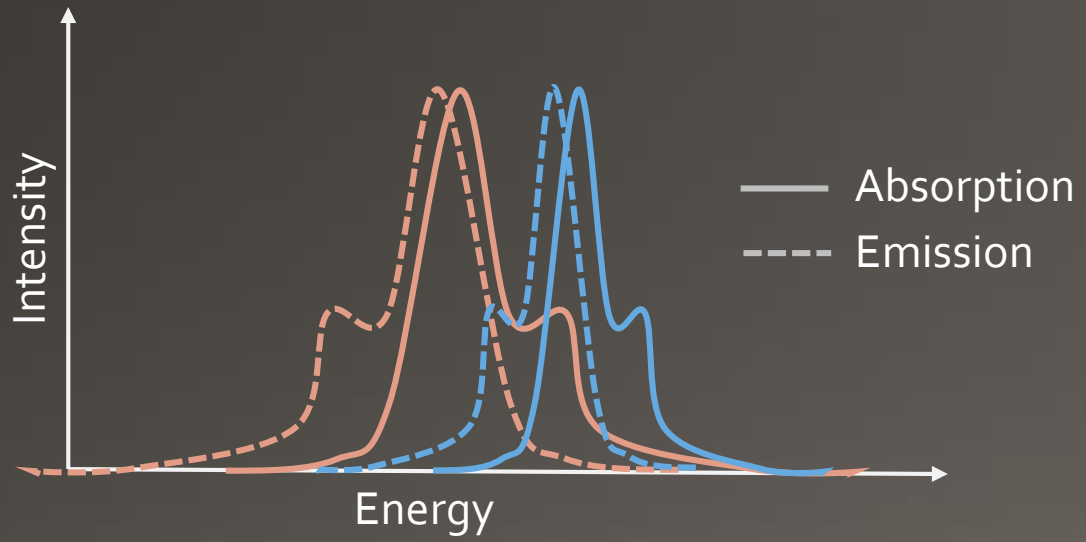


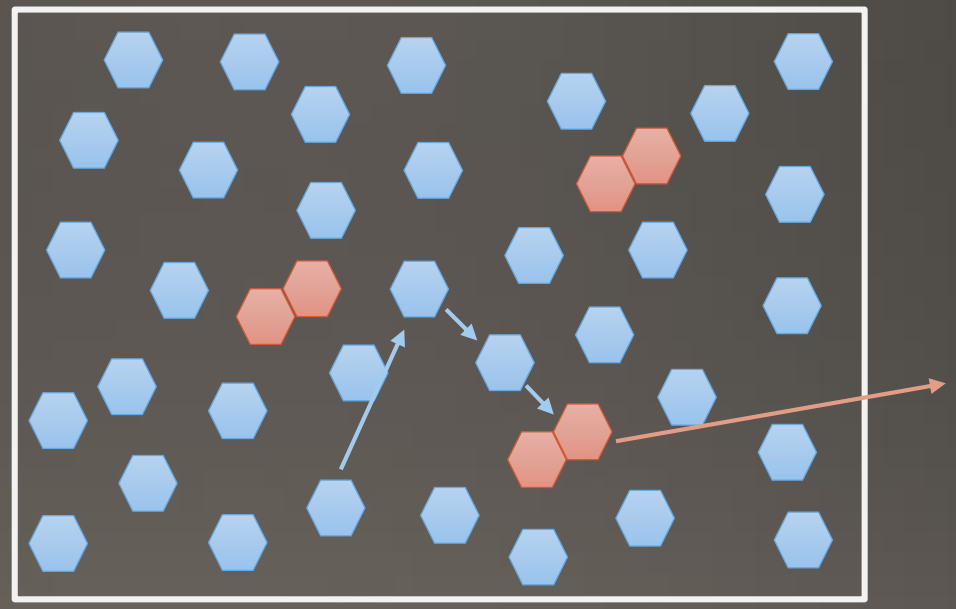
- Absorbtion: Blue photon (E_A) promotes electronic transition: $S_0 \rightarrow S_1$ $\Delta J \neq 0$
- Non-Radiative Transition: Internal conversion or vibrational deexcitation
- Emission: Red Photon (E_E) emitted as electronic state relaxes back to S_0
- $E_E < E_A$

Fluorescence: Binary Scintillators

Solvent: Primary target starts the signal

Solute: Dilute fluor gets the signal out of the bulk





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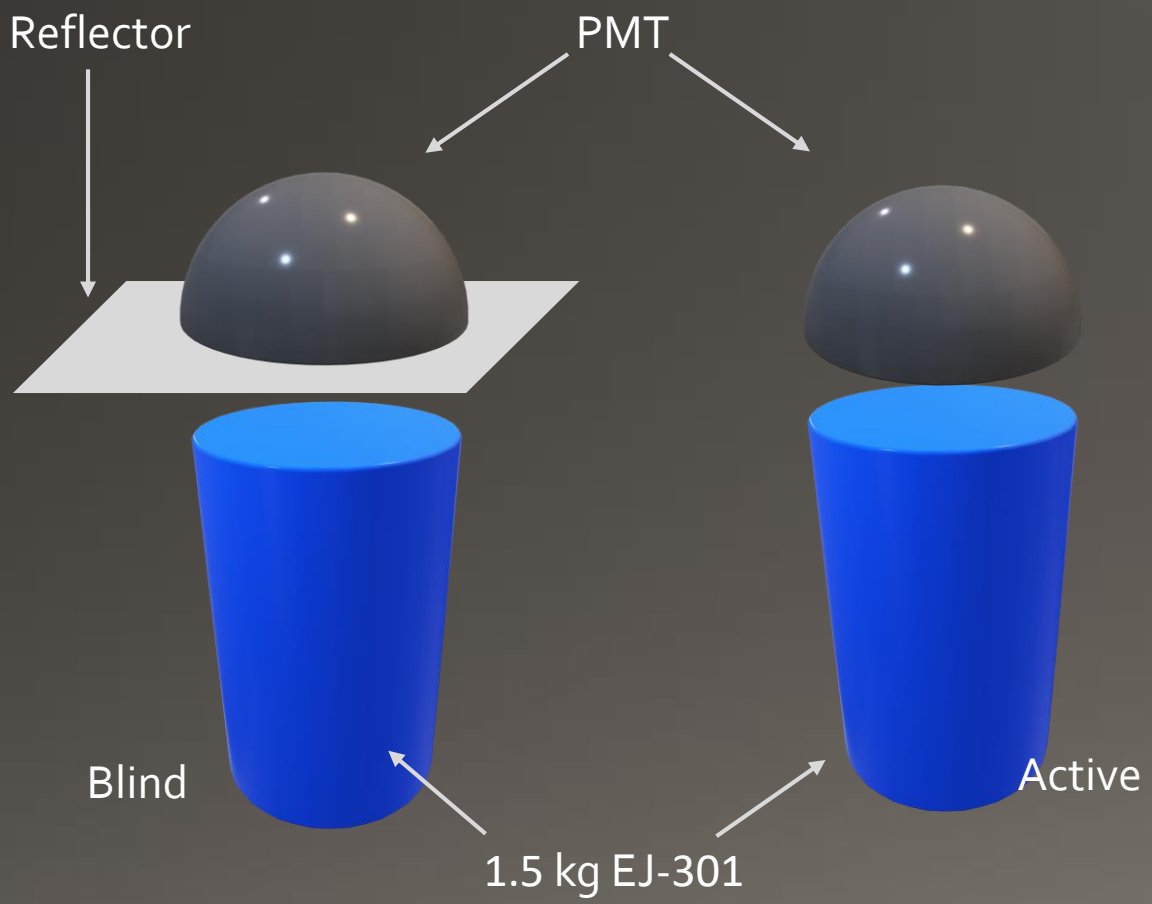
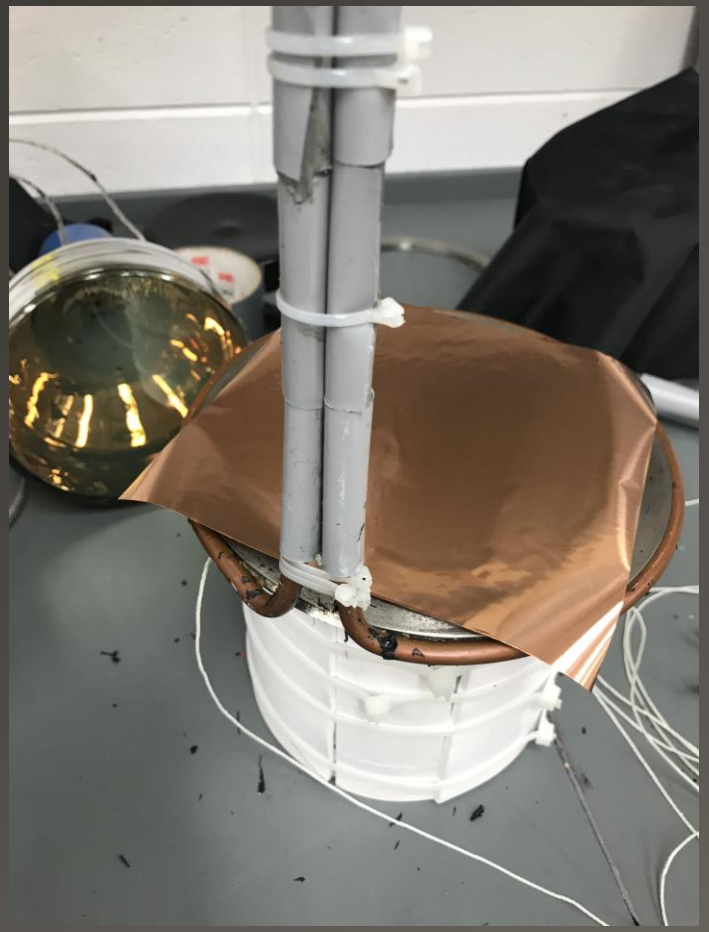
Experiment



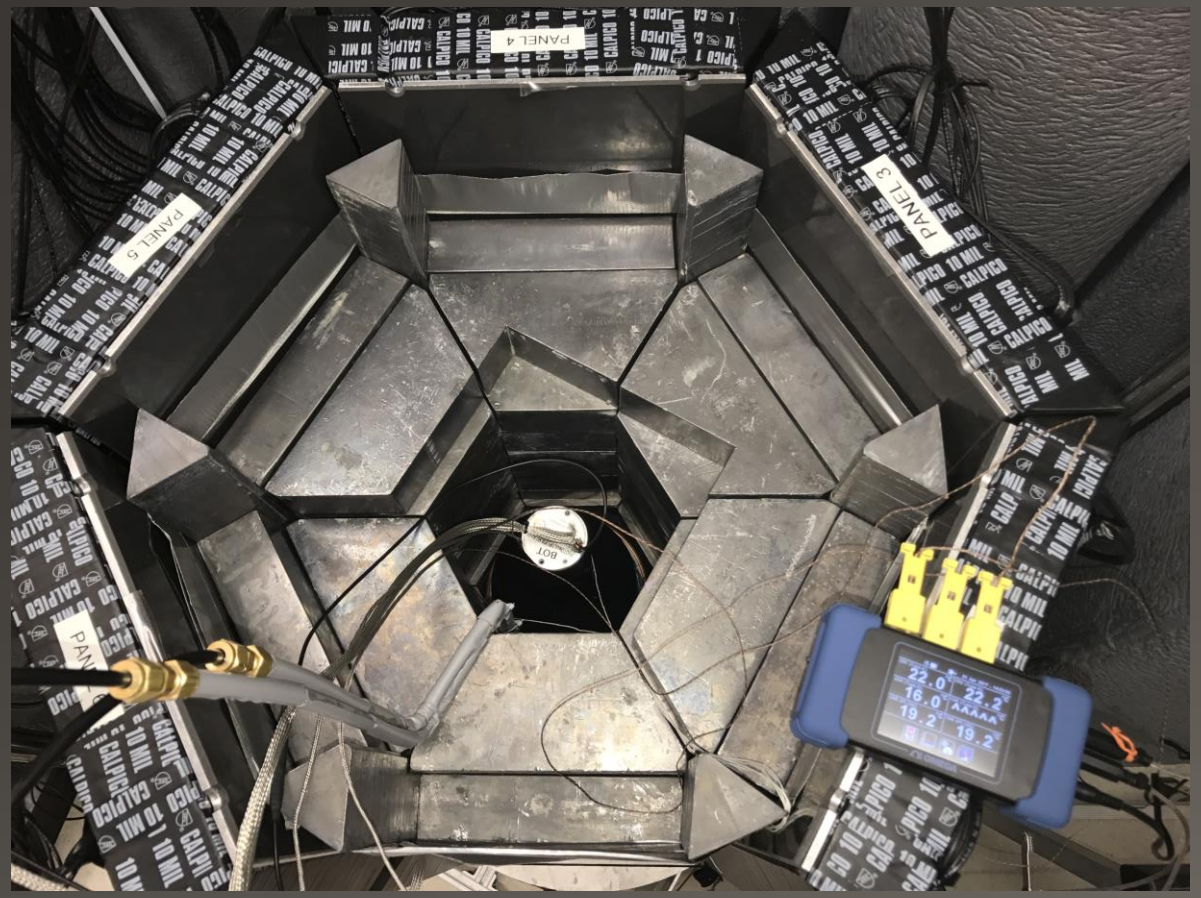
- 1.5 kg of EJ-301
- 0.1° Temperature control
- Need only about a week in each mode
- Record PMT rate in two modes,
  - Blind: Reflector shielding from the scintillator
  - Active: PMT observing the scintillator

arXiv: 1912.02822

Experimental Setup



Experimental Setup



Interaction Rate

Photon Rate

$$R = \xi \frac{2N_B \rho_\chi \bar{\sigma}_e}{8\pi m_\chi \mu_{\chi e}^2} \int \frac{d^3\mathbf{q}}{q} \eta\left(v_{\min}\right) F_{\mathrm{DM}}^2(q) \sum_i \left|f_i(\mathbf{q})\right|^2$$

Sensitive when R > ~0.1Hz

ρ_χ :DM density ξ :detector efficiency

Molecular form factor

$$f_i(\mathbf{q}) = \int d^3\mathbf{p} \tilde{\psi}_n(\mathbf{p}) \tilde{\psi}^\star_{n+i}(\mathbf{p}+\mathbf{q})$$

$$\xi \approx (0.77)(0.28) = 0.166$$

PMT efficiency EJ-301 quantum efficiency

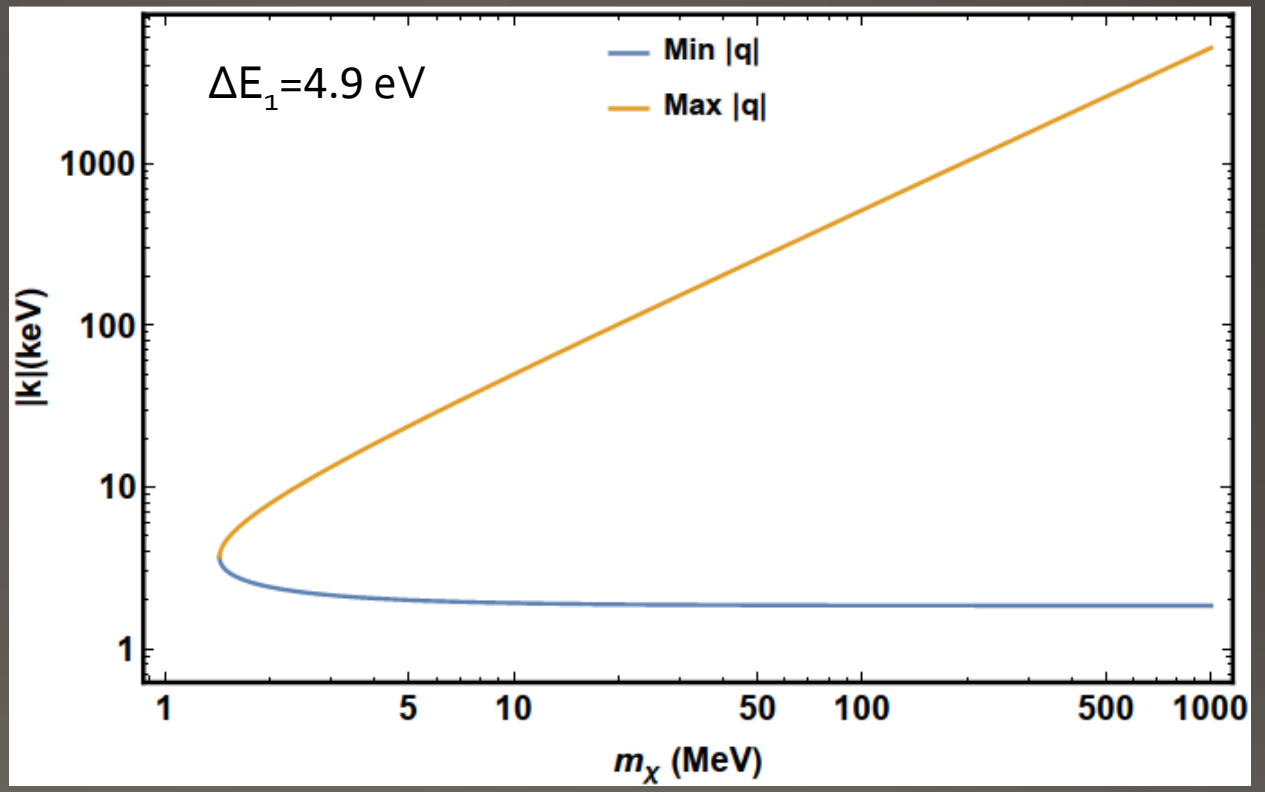
SAO Fourier transform

$$\widetilde{\Psi}(\mathbf{k}) = \left(\sum_{i=1}^{6} c_i^\psi e^{-i\mathbf{k}\cdot\mathbf{R}_i}\right) \widetilde{\phi}(\mathbf{k}) \equiv \mathcal{B}_\psi(\mathbf{k}) \widetilde{\phi}(\mathbf{k})$$

$\mathcal{B}_\psi$:k_x & k_y dependent phases

Kinematically Allowed Momenta

- Only has support on a range of momenta.
  - If q is too large, the DM would have escaped
  - If q is too small, there's not enough energy to excite the transition



$$\eta\left(v_{\min}\right)=\int\frac{4\pi v^{2}dv}{v}g_{\chi}(v)\Theta\left(v-v_{\min}\right)$$

$$v_{\min}(\mathbf{q})=\frac{\Delta E_{ij}}{q}+\frac{q}{2m_{\chi}}$$

$$g_{\chi}(v)=\frac{1}{K}\exp\left(-\frac{|\mathbf{v}+\mathbf{v_E}|^{2}}{v_{0}^{2}}\right)\Theta\left(v_{\rm esc}-|\mathbf{v}+\mathbf{v_E}|\right)$$

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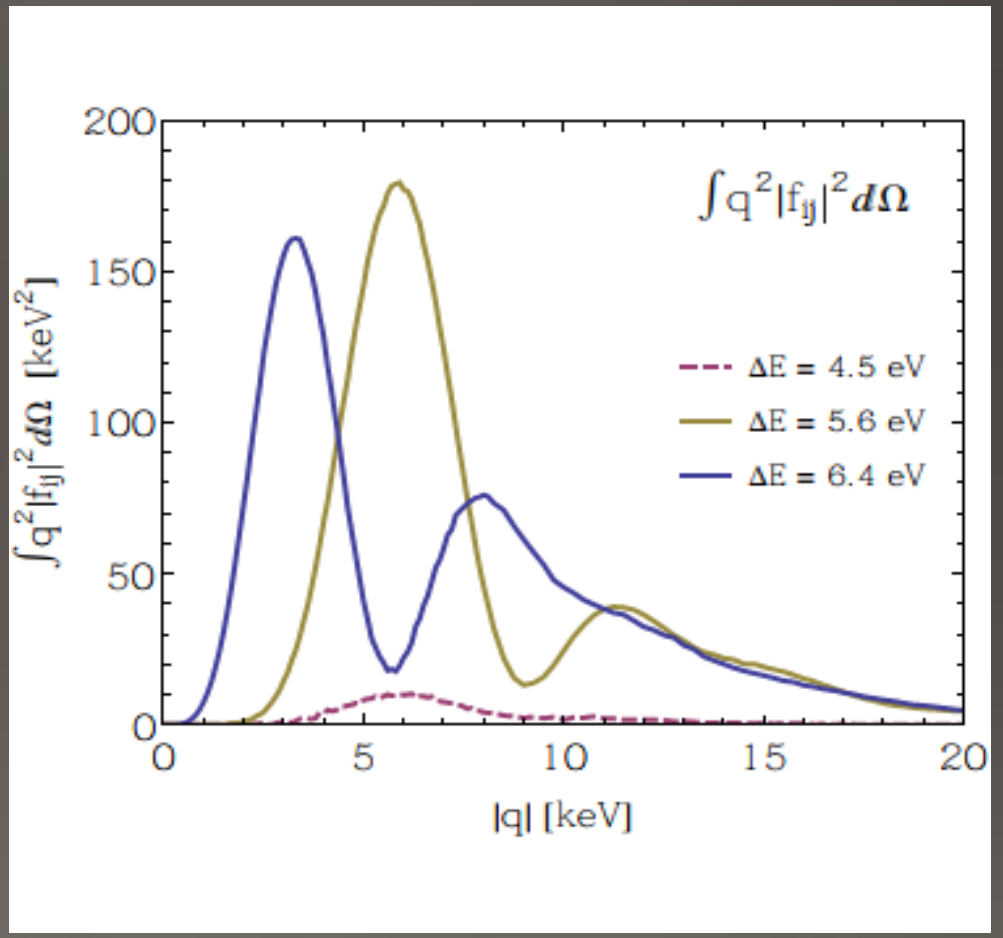
Molecular Form Factor

3 Dimensional integral in k-space

$$f_i(\mathbf{q}) = \int d^3\mathbf{p}\tilde{\psi}_n(\mathbf{p})\tilde{\psi}^{\star}_{n+i}(\mathbf{p}+\mathbf{q})$$

$$= \langle\psi_f(k+q)|e^{iq\cdot x}|\psi_i(k)\rangle$$

$$\widetilde{\Psi}(\mathbf{k}) = \left(\sum_{i=1}^{6} c_i^{\psi} e^{-i\mathbf{k}\cdot\mathbf{R}_i}\right)\widetilde{\phi}(\mathbf{k}) \equiv \mathcal{B}_{\psi}(\mathbf{k})\widetilde{\phi}(\mathbf{k})$$



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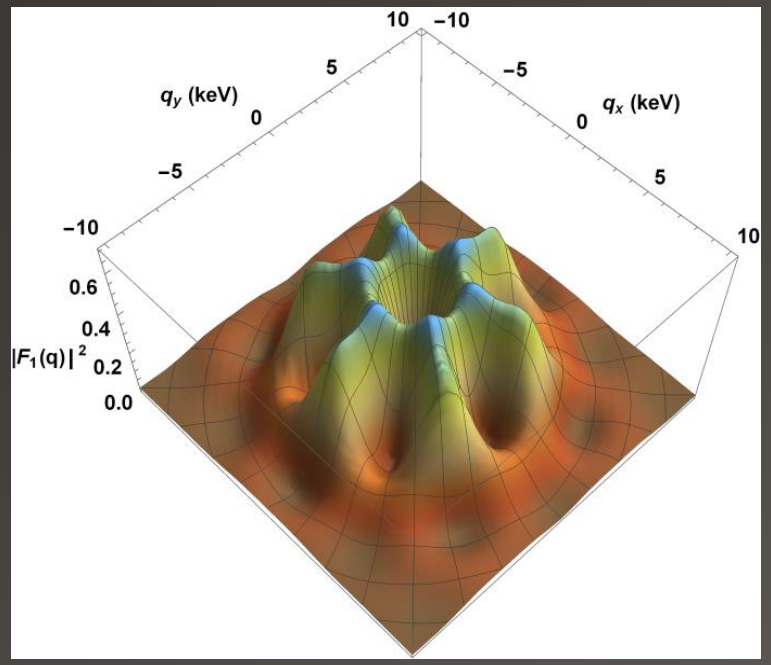
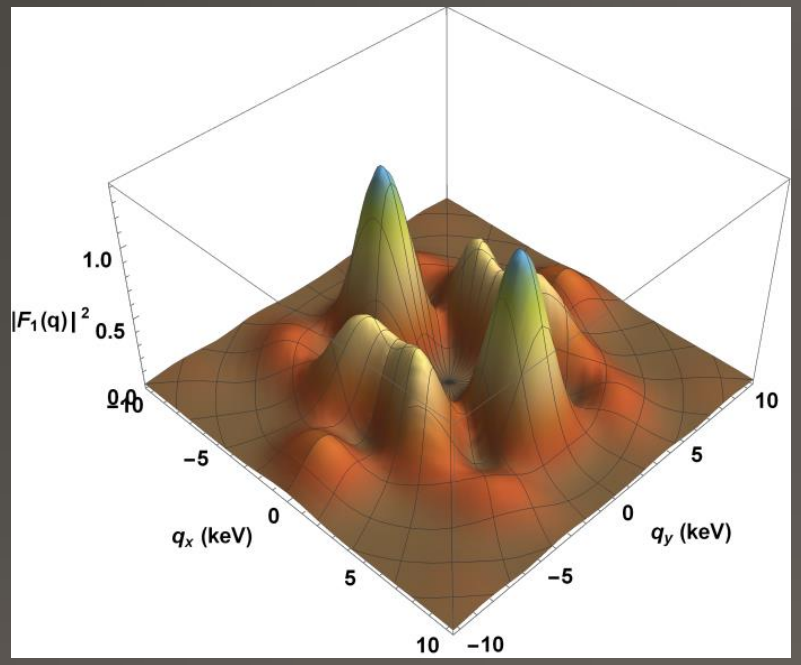
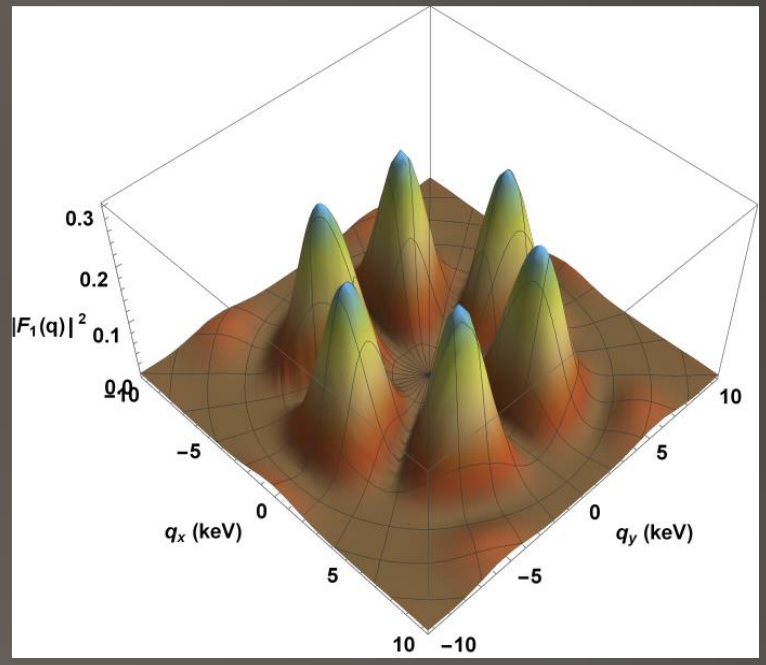
Form Factors Illustrated



$|F_1(q)|^2$
θ =90°



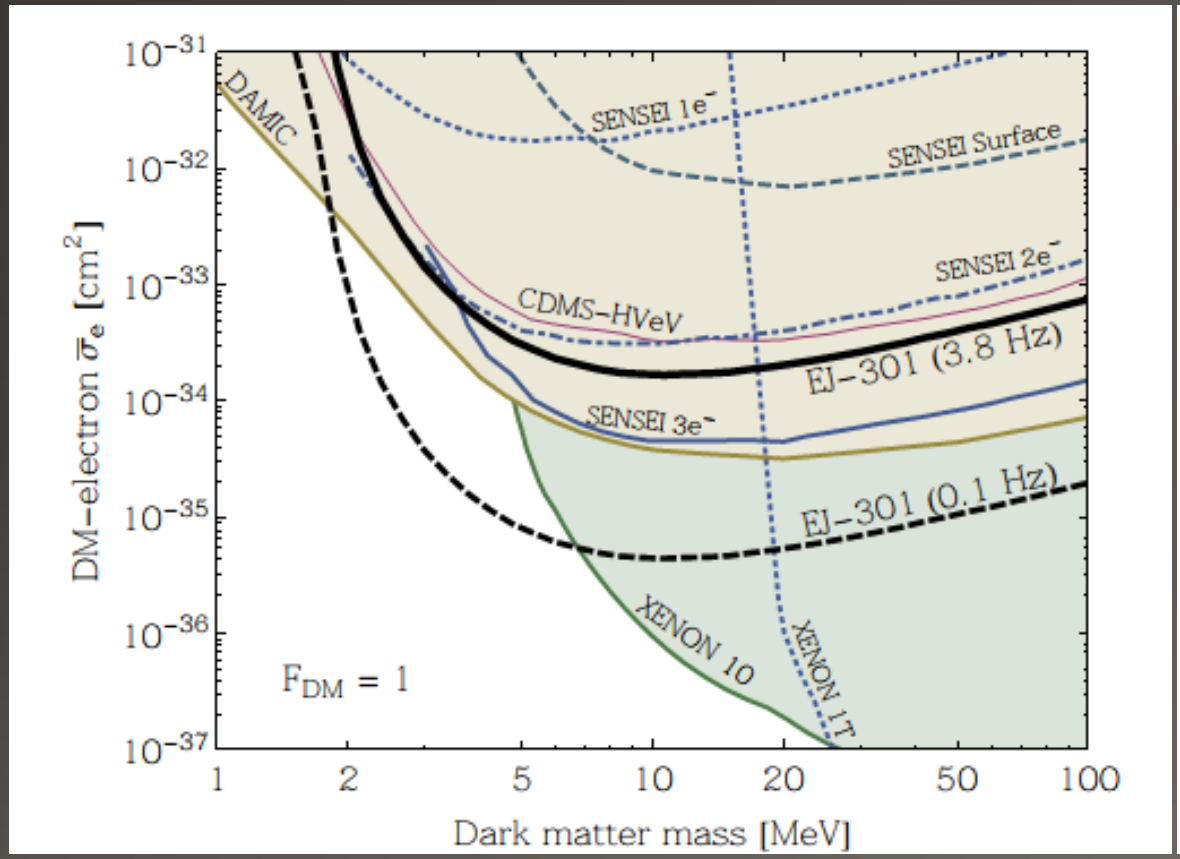
$|F_2(q)|^2$
θ =90°

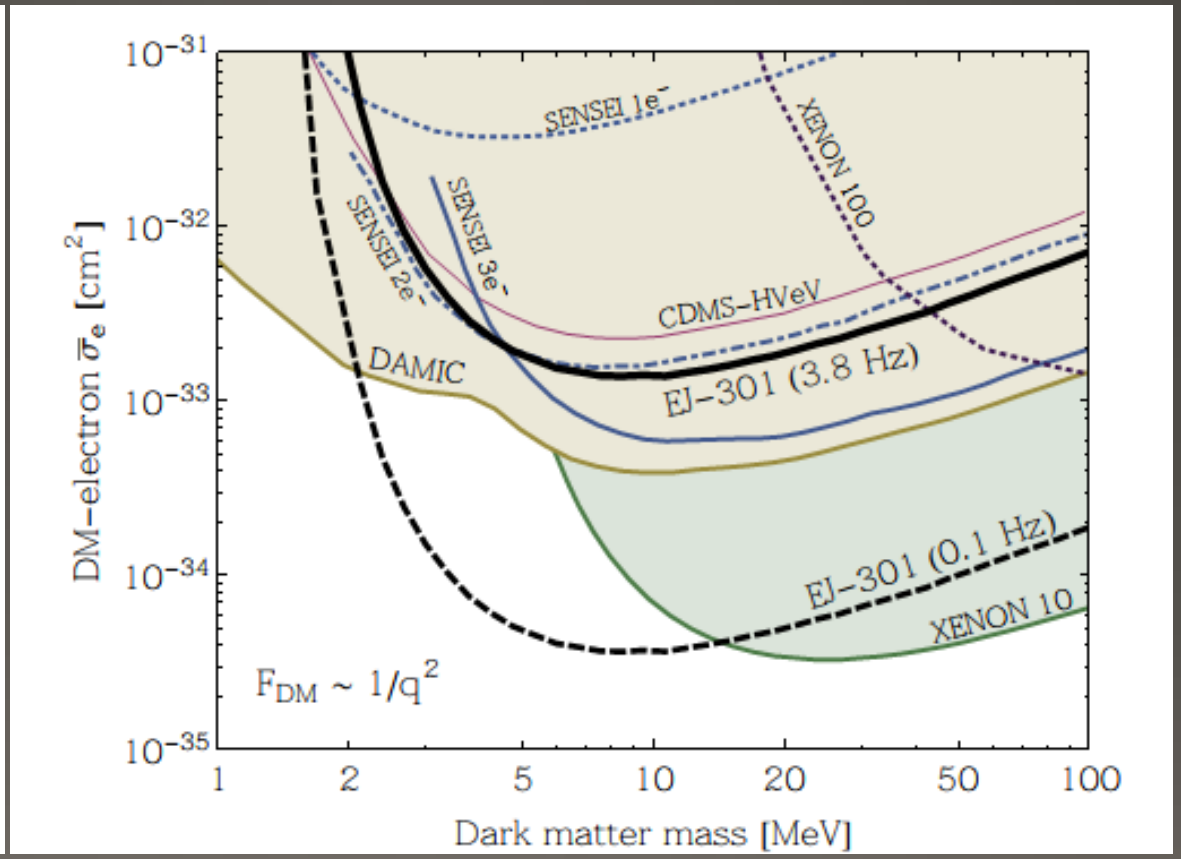


$|F_3(q)|^2$
θ =90°

arXiv: 1912.02822

Predicted Sensitivity





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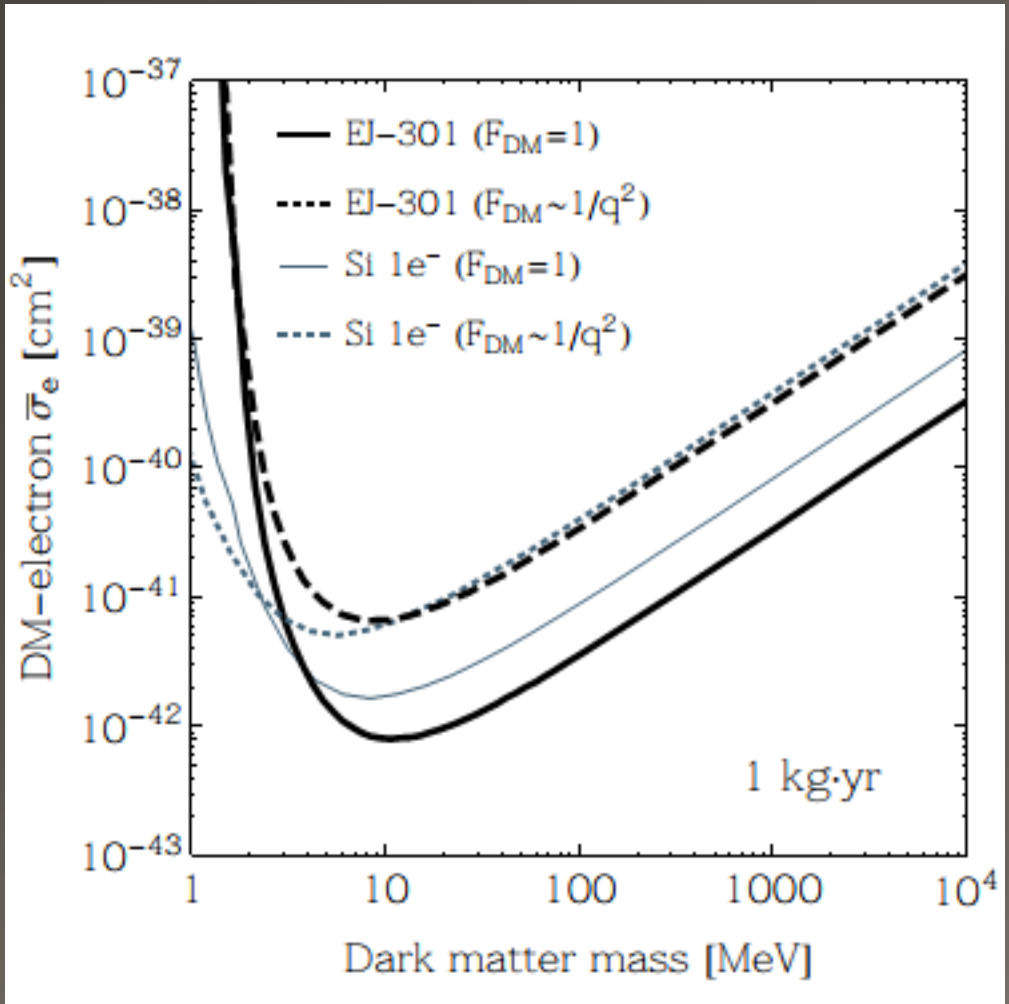
Sensitivity Comparison: Silicon



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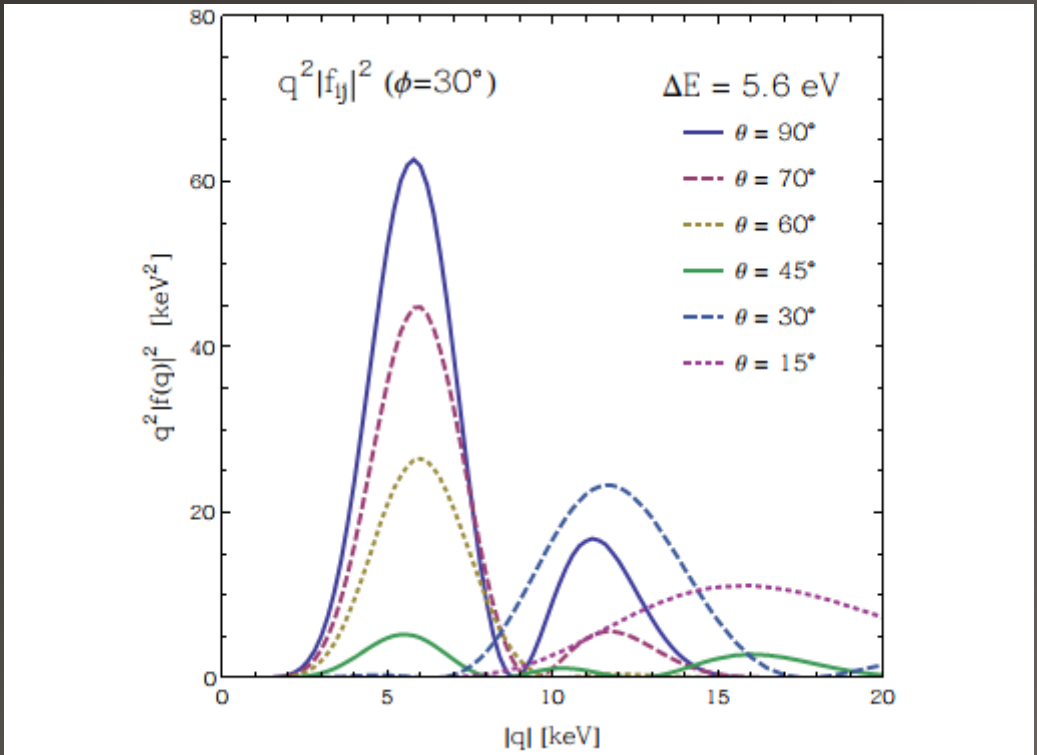
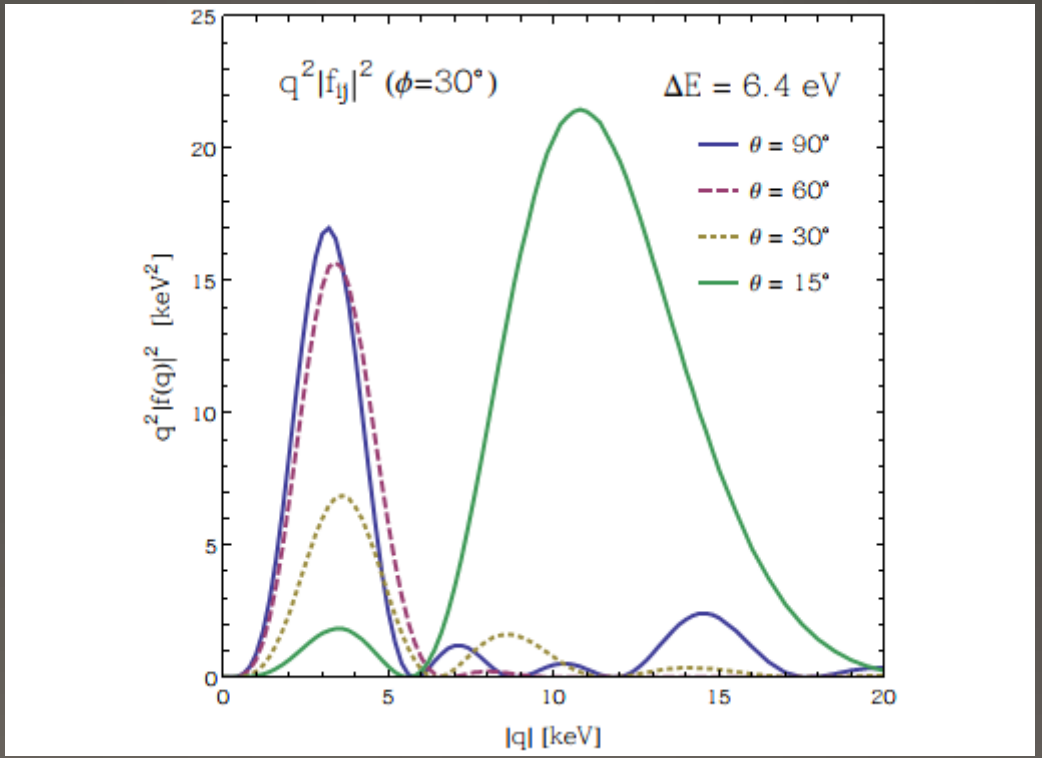
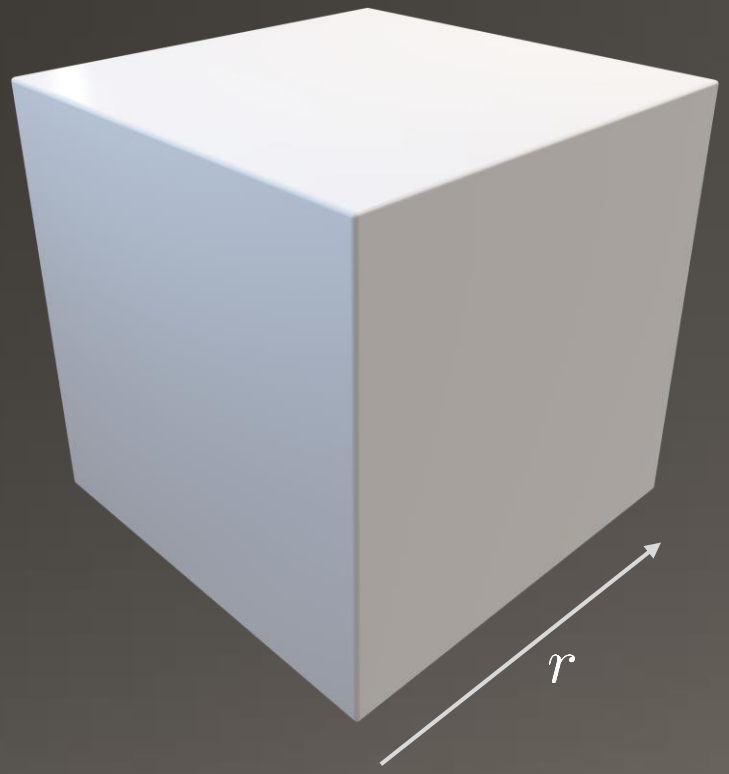
Directional Detection





arXiv: 1912.02822

Future Targets: e⁻s in Bigger Boxes



$$E_n \sim \frac{n^2\hbar^2\pi^2}{2mr^2}$$

$$E_1 \sim 0.1\mathrm{eV}\left(\frac{10^{-7}\mathrm{m}}{r}\right)^2$$

Need a box that is nano- to micro-scale!

i.e. Need systems with tens to thousands of atoms

arXiv: 1912.02822

Future Targets: Larger Polyacenes

Naphthalene

$E_1 = 3.9\text{eV}$

Phenanthrene

$E_1 = 3.5\text{eV}$

Natural extension, add more rings!

- Anthracene is known to be one of the best organic scintillators
- More complex ring structures are possible
- Long poly-ene bridges form the backbone of many dyes
- Quantum yield near unity
- Asymmetric ground state!

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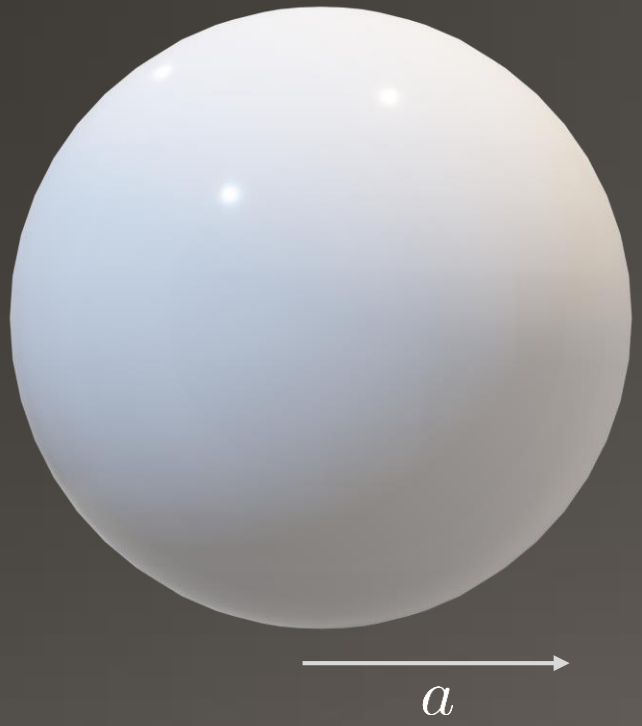
Future Targets: Quantum Dots

Semiconducting nano-spheres



$$E_{\text{confinement}} = \frac{\hbar^2\pi^2}{2a^2}\left(\frac{1}{m_e}+\frac{1}{m_h}\right) = \frac{\hbar^2\pi^2}{2\mu a^2}$$

$$E_{\text{exciton}} = -\frac{1}{\epsilon_r^2}\frac{\mu}{m_e}R_y = -R_y^*$$

$$E = E_{\text{bandgap}} + E_{\text{confifenent}} + E_{\text{exciton}}$$

$$= E_{\text{bandgap}} + \frac{\hbar^2\pi^2}{2\mu a^2} - R_y^*$$

ϵ_r Dielectric constant

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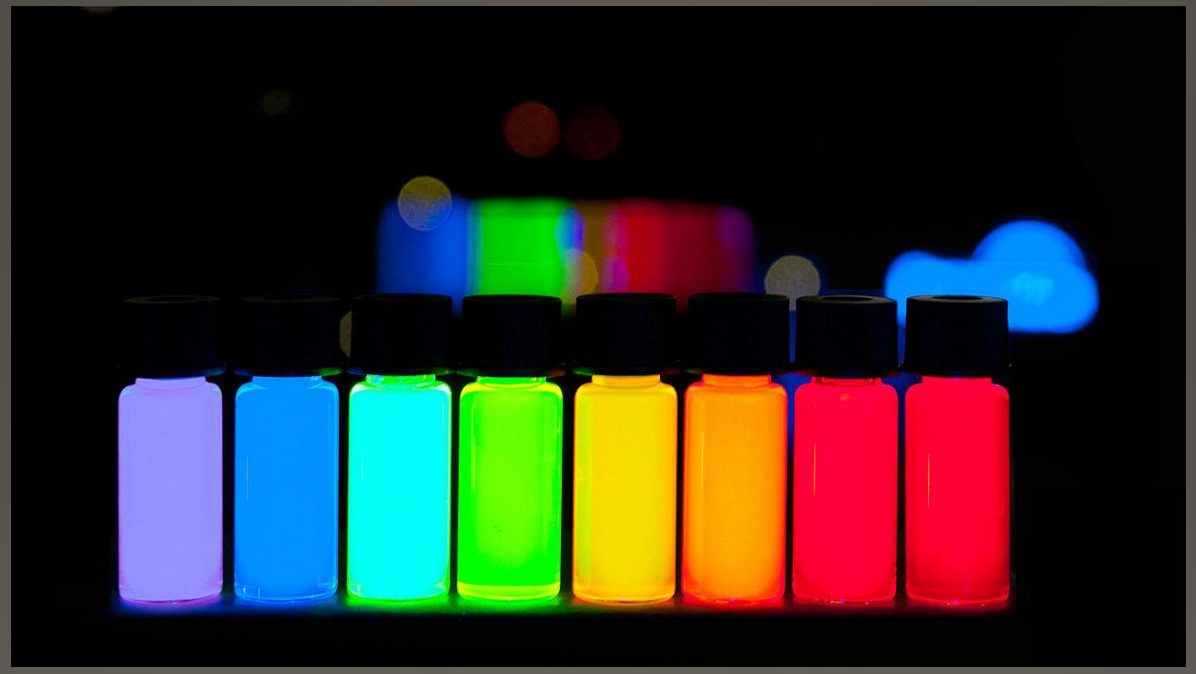
Future Targets: Quantum Dots

$a \sim 1 - 10$ nm

PlasmaChem ZnCdSeS C.C. 3.0



Scalable, Cheap, tunable!

E.x. 60% by mass scintillators

arXiv: 1912.02822

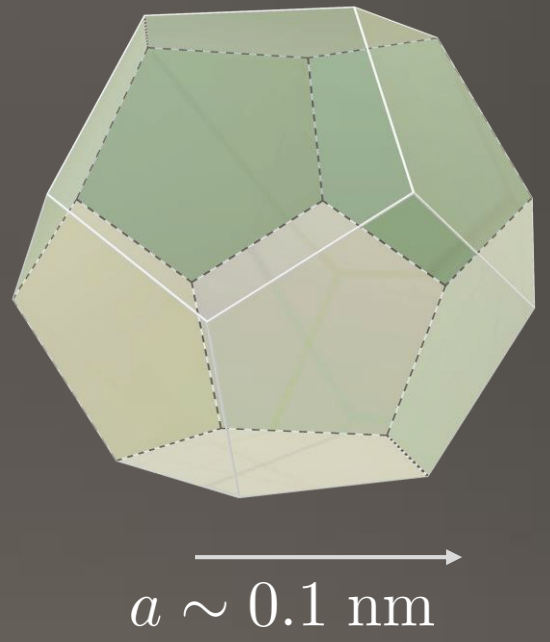
Future Targets: Solvated Electrons

LiNH3 CC 3.0
https://www.youtube.com/watch?v=Qx-gVTaRAq4

Purple color comes from eV-scale electronic transition at low conc.

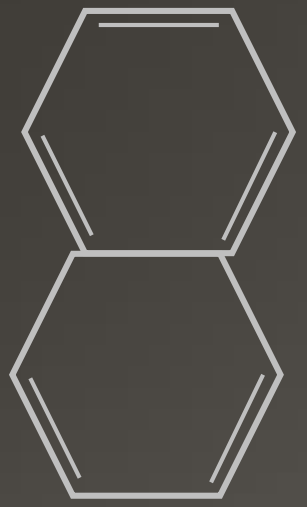
Metallic color comes from quasi-conduction band at high conc.

Solvation shell encapsulates a quasi-free electron in alkali metal solutions



Electron acts like a particle in a cavity with electronic properties which are concentration dependent.

Future Considerations: Nuclear Scattering



Nuclear recoils can induce electronic excitations

- Molecular analog to Migdal effect
- Can be used to problem DM-Nucleon cross section
- Simple molecules and solvation shell are particularly well-suited
- Again, for aromatics, quantum yield near unity!

Future Prospects

- Explore scintillators with lower thresholds
- Crystal organic scintillators for directionality
- Expand formalism to more complicated molecules
- Expand formalism to include molecular deformations

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Summary

- Covalent molecules have electronic transitions with ΔE~ few eV
- Well characterized excitation spectra
- Experiment limited by threshold not statistics
- Short duration experiments
- Comparatively inexpensive
- Formalism being developed for DM interactions w/ covalent molecules
- Table-top experiments probing “high” energy BSM physics.