Scintillating Developments in the Directional Detection of Sub-GeV DM

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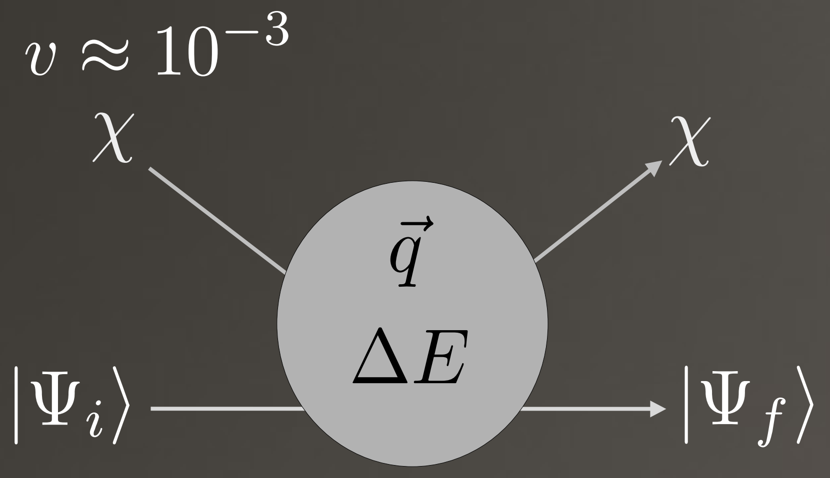
Stockholm University

Oskar Klein centre

Direct detection

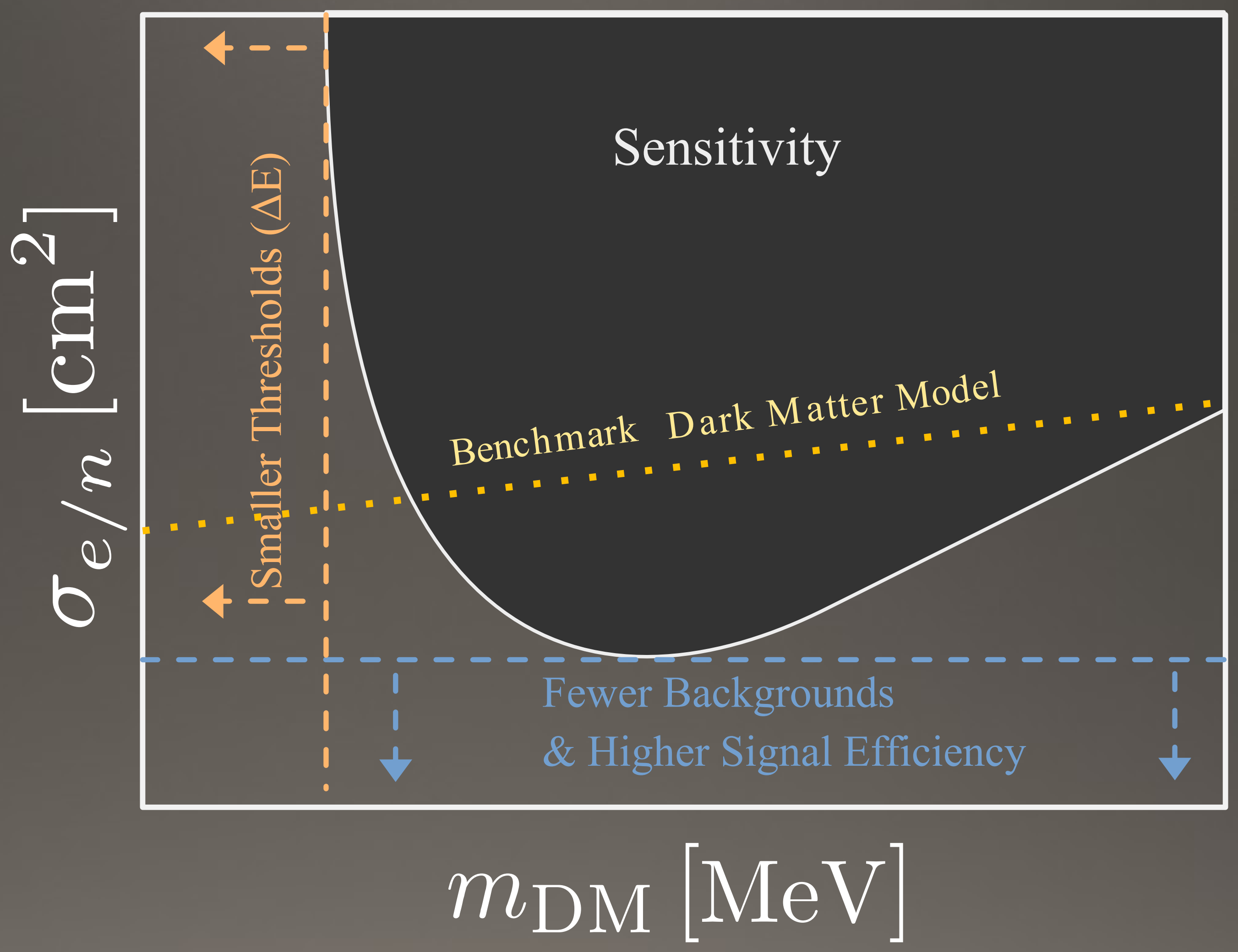


Energy Budget

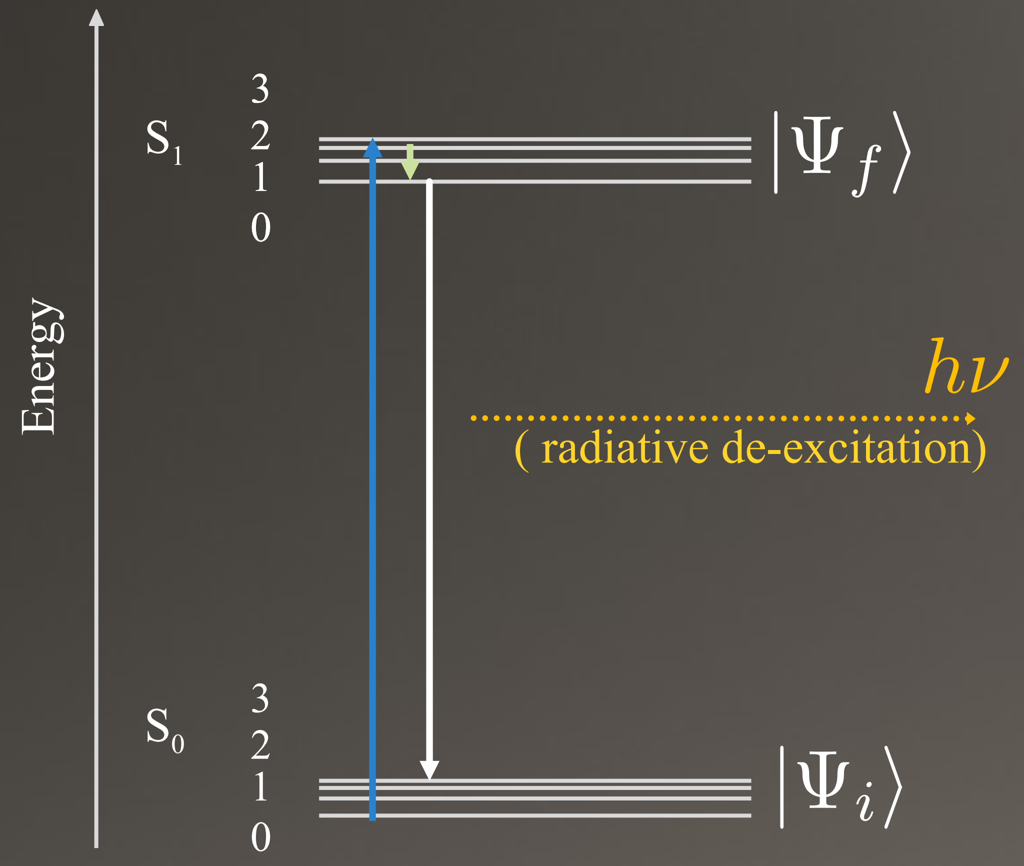
$$\Delta E \sim v^2 m_\chi \approx \mathcal{O}(\mathrm{eV}) \left(\frac{\mathrm{m}_\chi}{1\,\mathrm{MeV}}\right)$$

Momentum Budget

$$q \sim v m_\chi \approx \mathcal{O}(\mathrm{keV}) \left(\frac{\mathrm{m}_\chi}{1\,\mathrm{MeV}}\right)$$



Luminescent Detectors



Fluorescence

(Singlet transitions)

Energy Scale

$$\Delta E \approx \mathcal{O}(\mathrm{eV})$$

Momentum Scale

$$q \sim \alpha m_\chi \approx \mathcal{O}(\mathrm{keV})$$

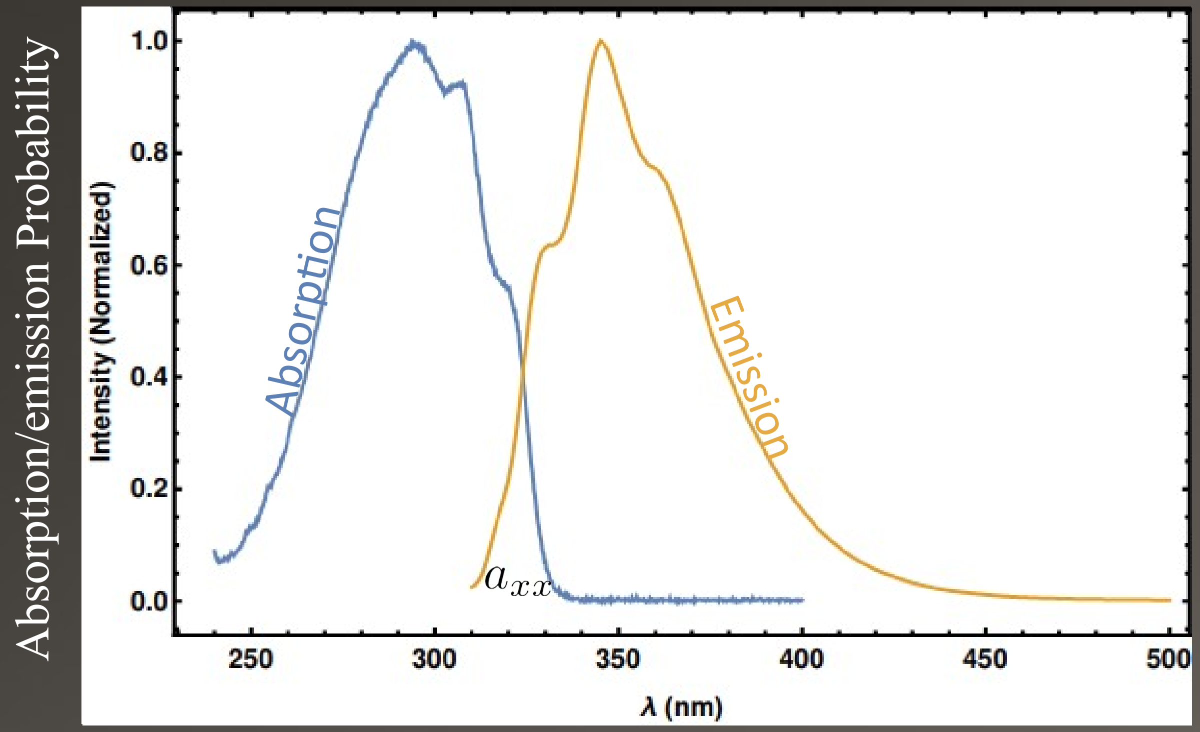
Disambiguation*

Scintillation: Initial O(keV) recoil produces *many* ionized/excited states. These quickly deexcite, producing photons.

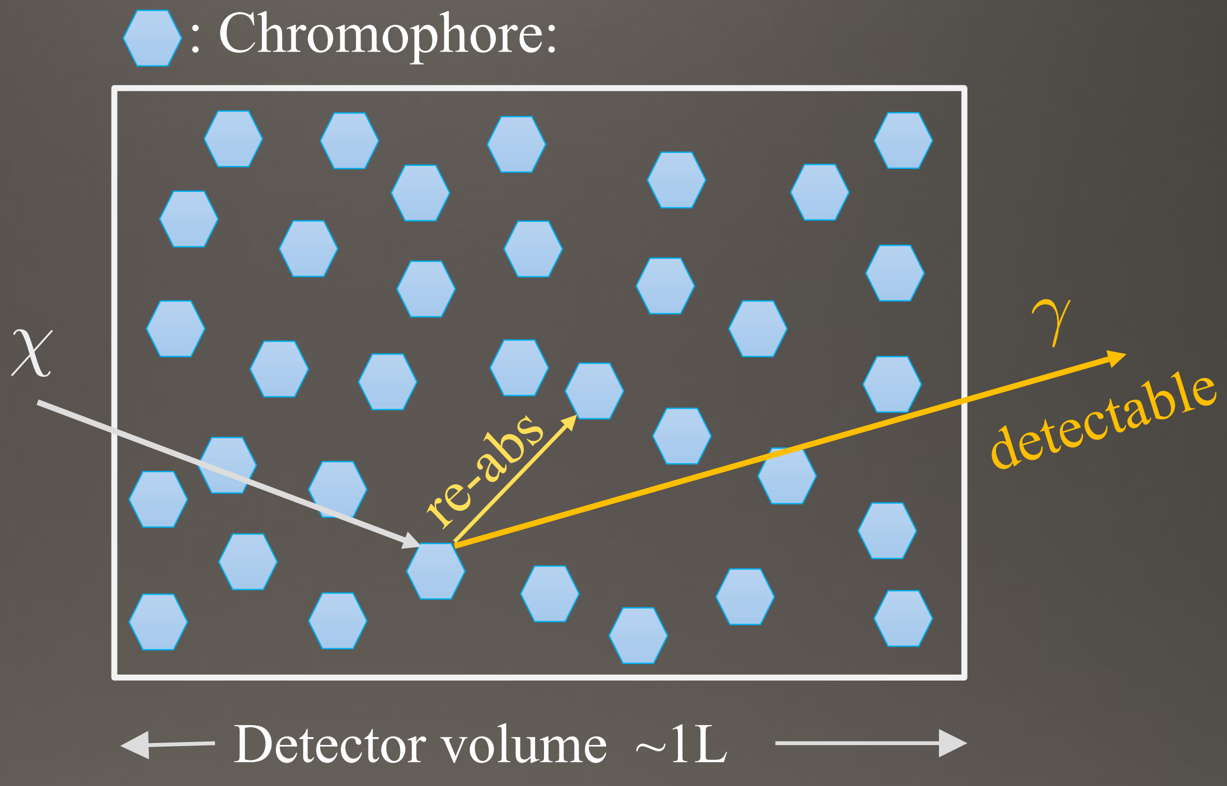
Fluorescence: Initial O(eV) recoil produces *single* excited state, which deexcites, producing single photon.

Fluorescence with DM

fluorescence spectra



Decreasing energy (E) →

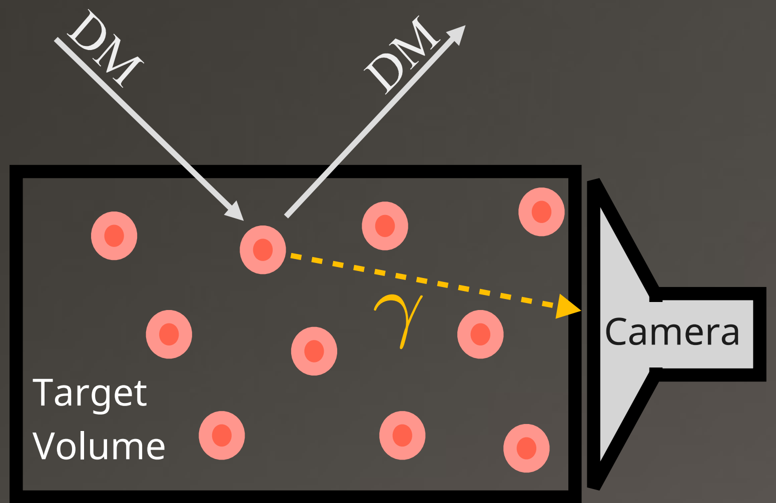


Probability for the photon to free stream

$$\Phi_{\mathrm{FB}} \sim (1 - a_{xx})$$ e.g. molecular crystals: $\Phi_{\mathrm{FB}} \approx 65\%$

Overview: new materials for Direct Detection

Recoil-induced *fluorescence* (radiative deexcitation)

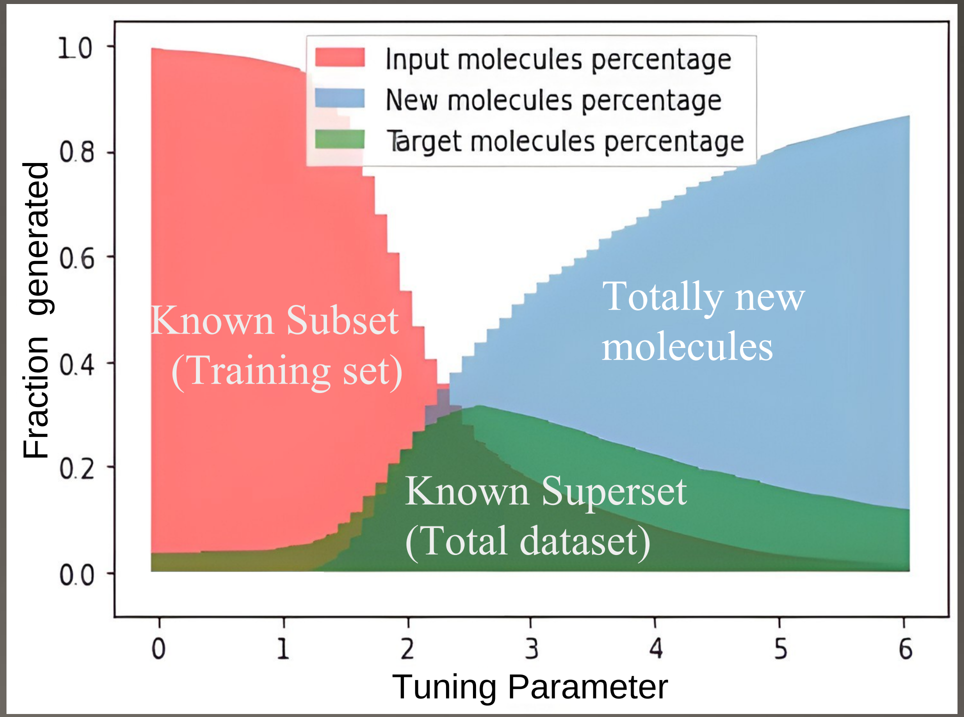


:unit that de-excites by emitting a photon

Could be:

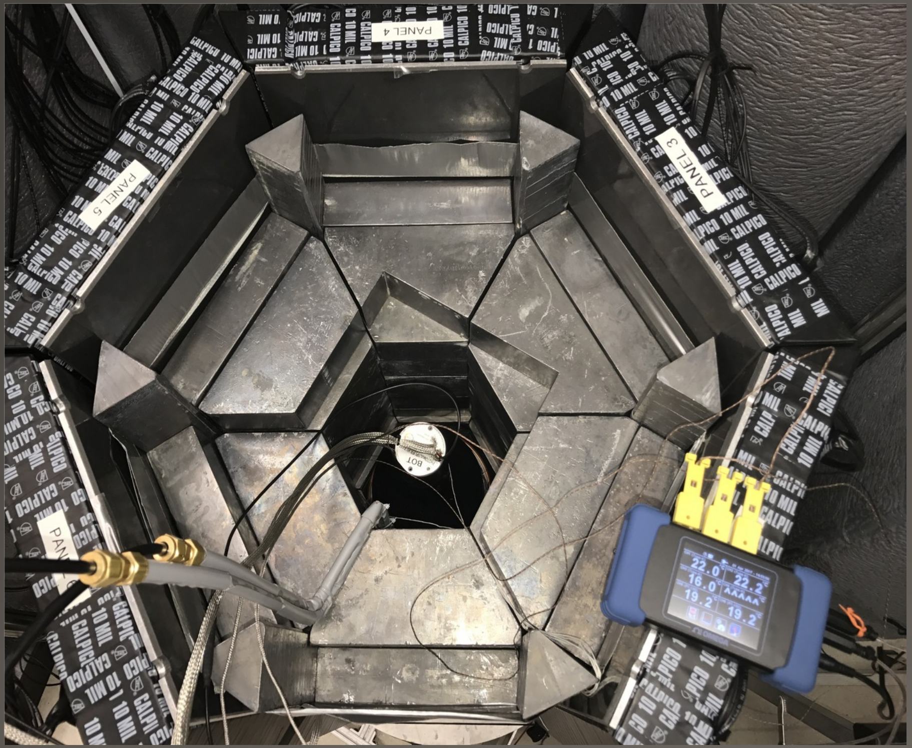
- Nanostructures (quantum dots)
- Molecules in ordered crystal
- Hybrid material (QDs in Molecular matrix)

ML to explore vast data of material space



Generative ML models will identify novel materials that maximize signals

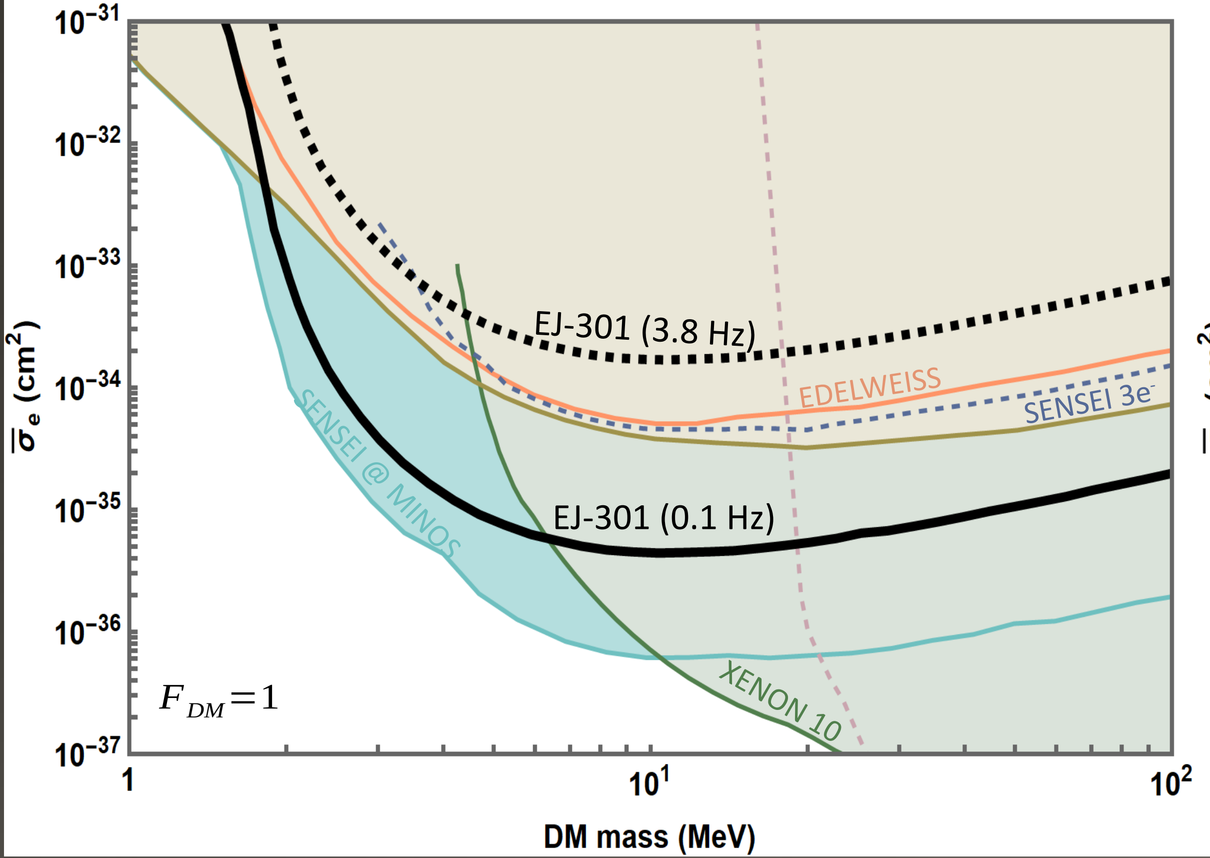
First Experimental Setup

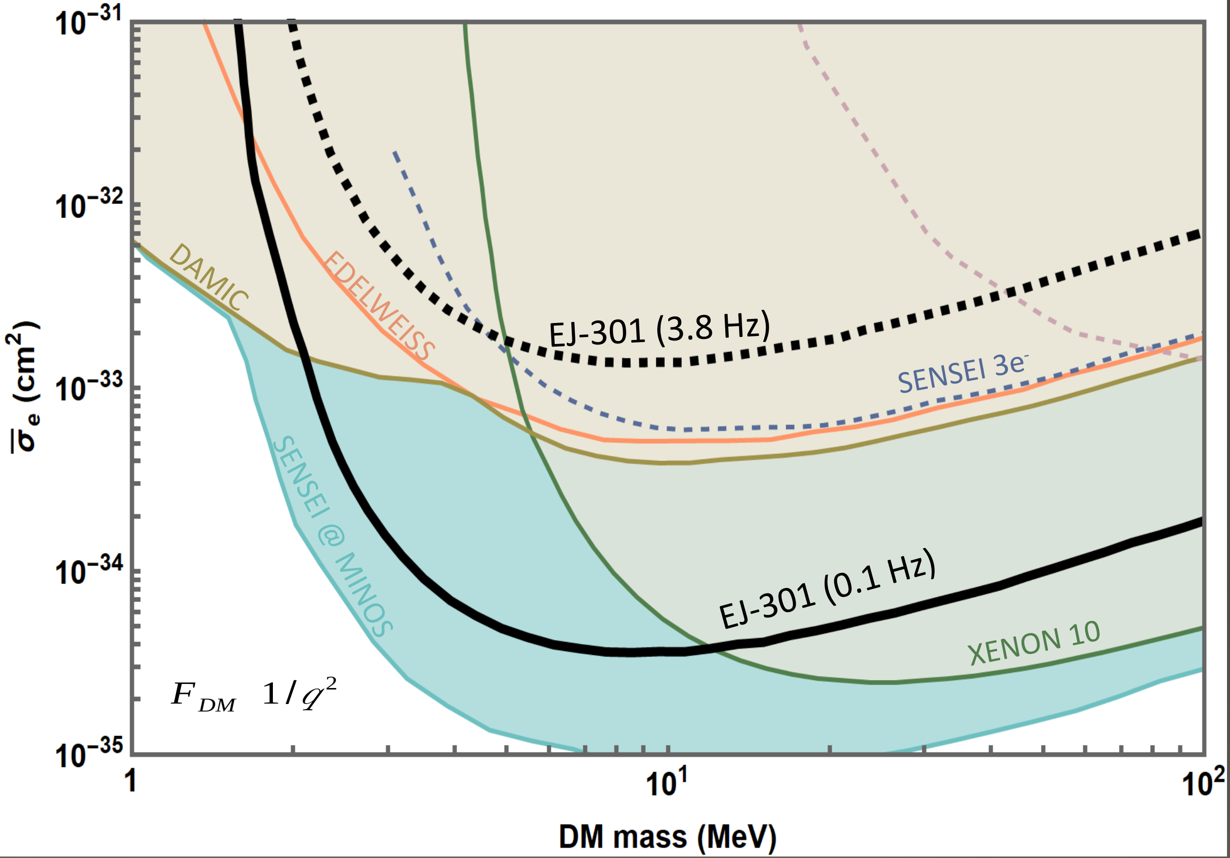


[CB, Collar, Kahn, Lillard: 1912.02822]

Results: EJ-301

(Contact interaction) (Long-range interaction)

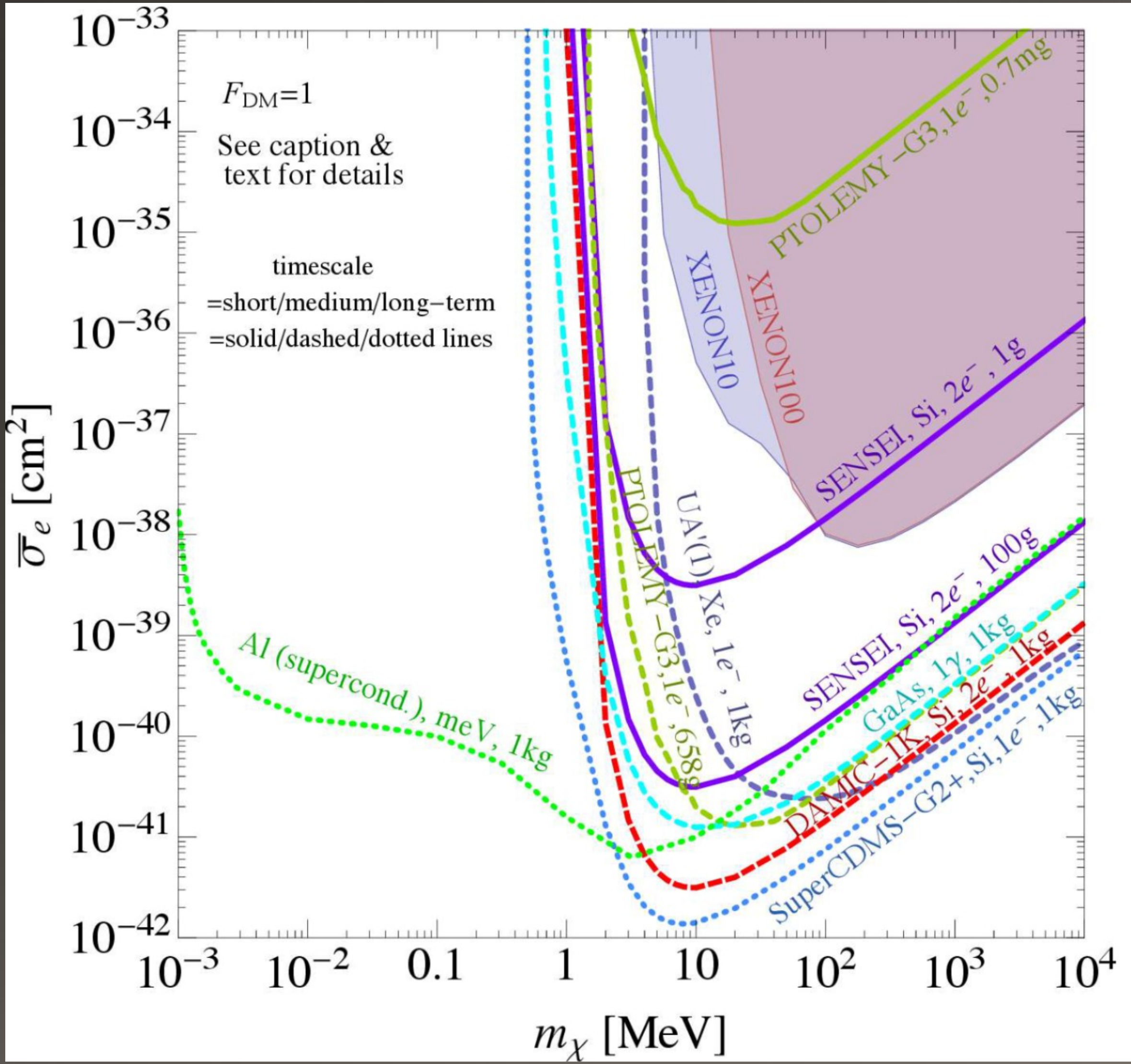




[CB, Collar, Kahn, Lillard: 1912.02822]

About 6 months from theory development to results!

The Field in Context



[US Cosmic Visions: Community Report: 1707.04591]

Many materials are proposed to probe the sub-GeV Space

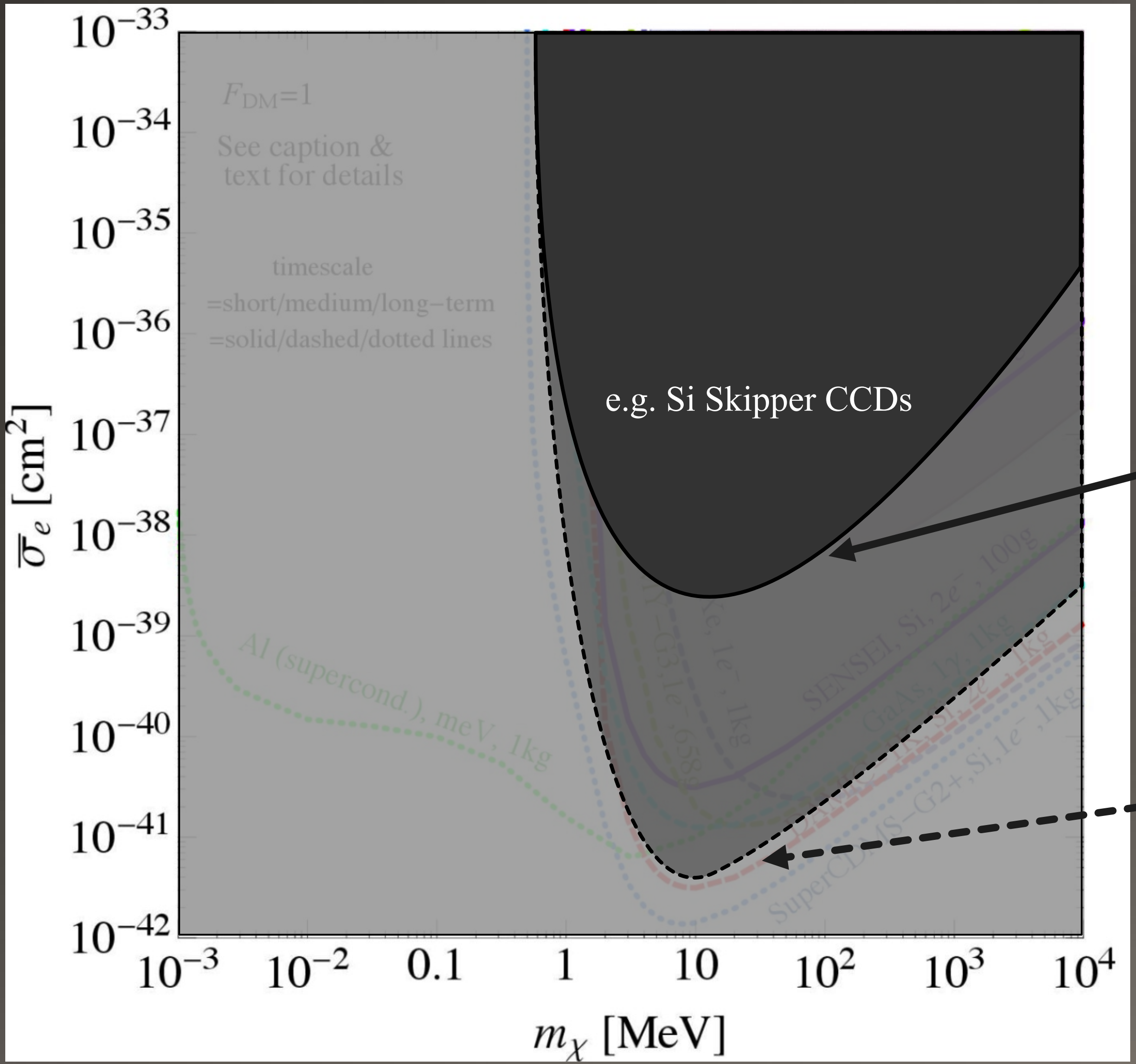
In 2017:

Short term (2 years)

Medium term (2-5 years)

Long term

Outlook and Potential Reach



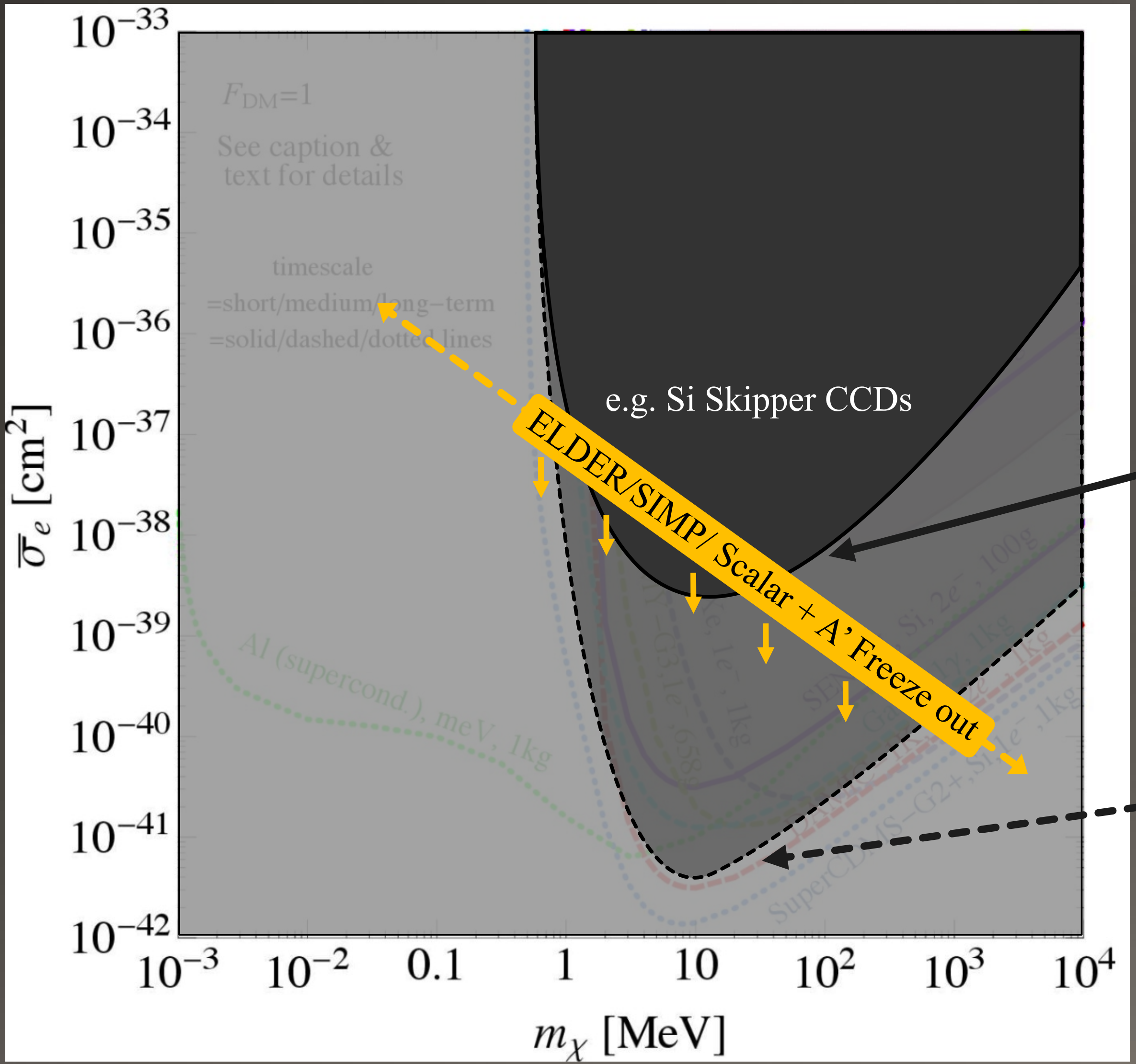
[US Cosmic Visions: Community Report: 1707.04591]

Present day:

Experimental results

Zero-background projection

Outlook and Potential Reach



[US Cosmic Visions: Community Report: 1707.04591]

Present day:

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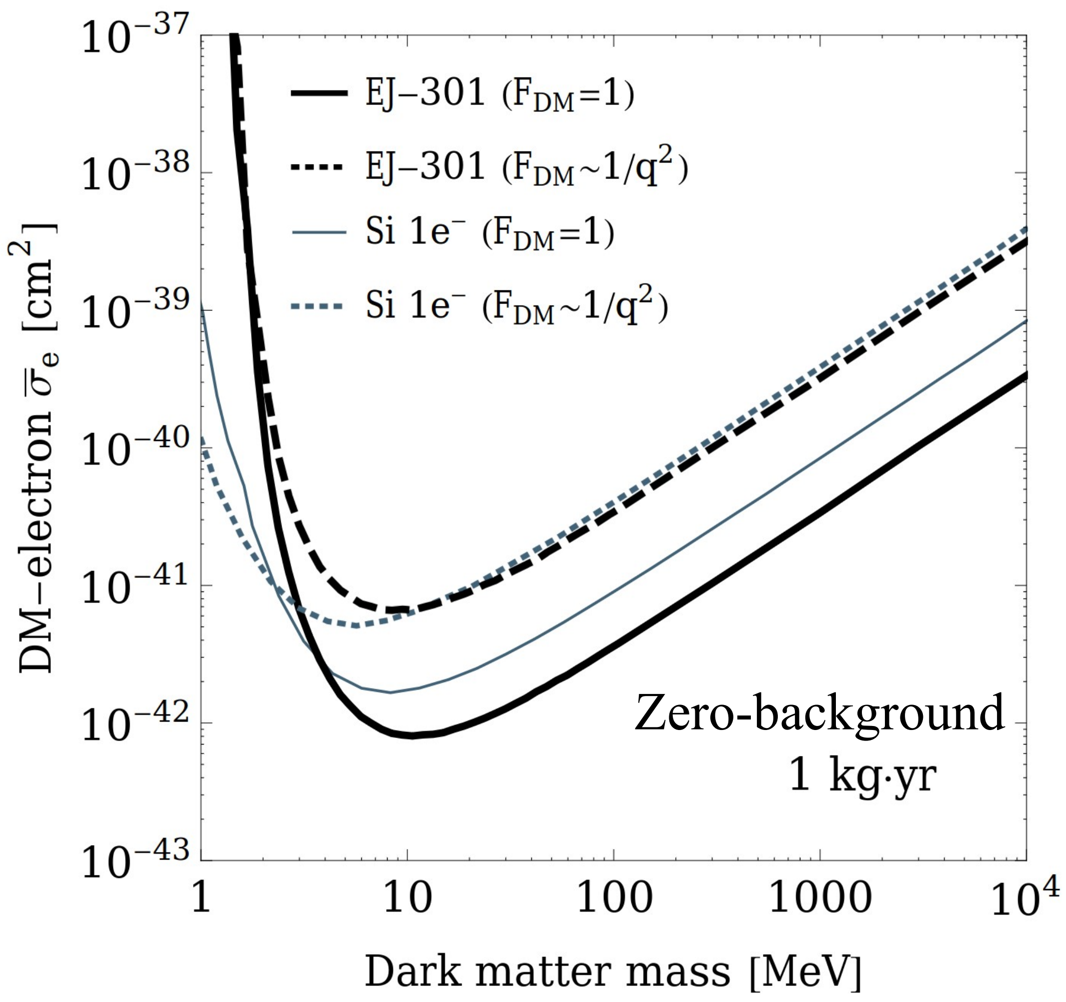
Zero-background projection

Outlook and Potential Reach

[**CB**, Collar, Kahn, Lillard: 1912.02822]



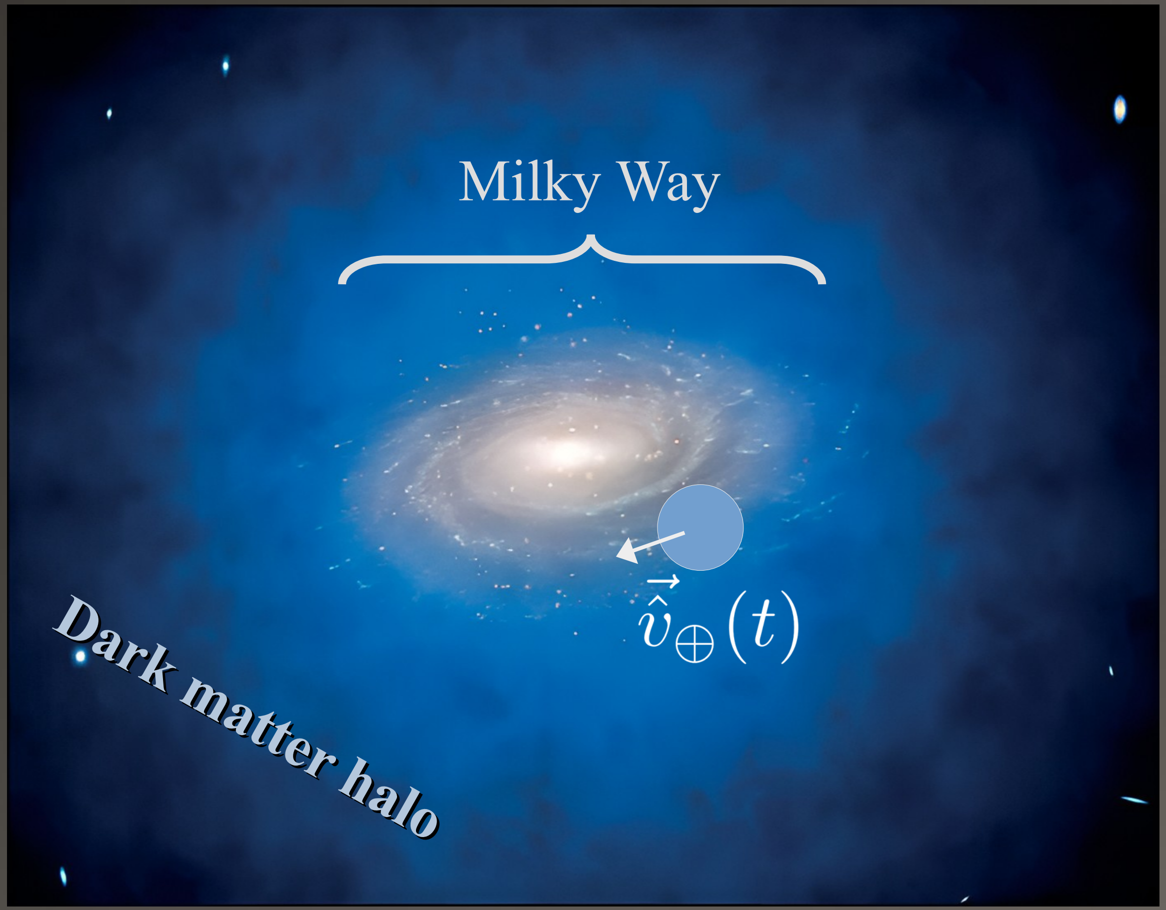
The obstacle

Backgrounds

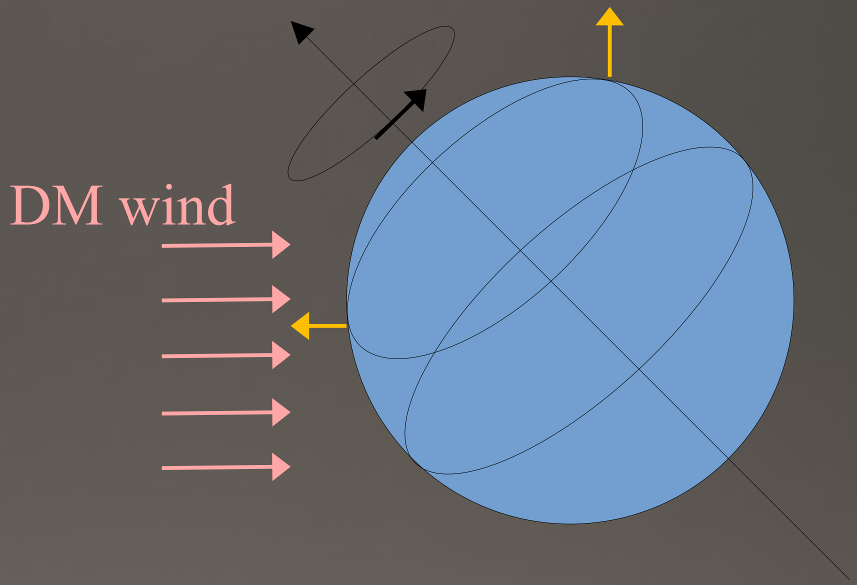
4 orders of magnitude of potential reach awaiting

Pound (kg) for Pound (kg) molecules produce about as much signal as e.g. Si.

Directional Detection

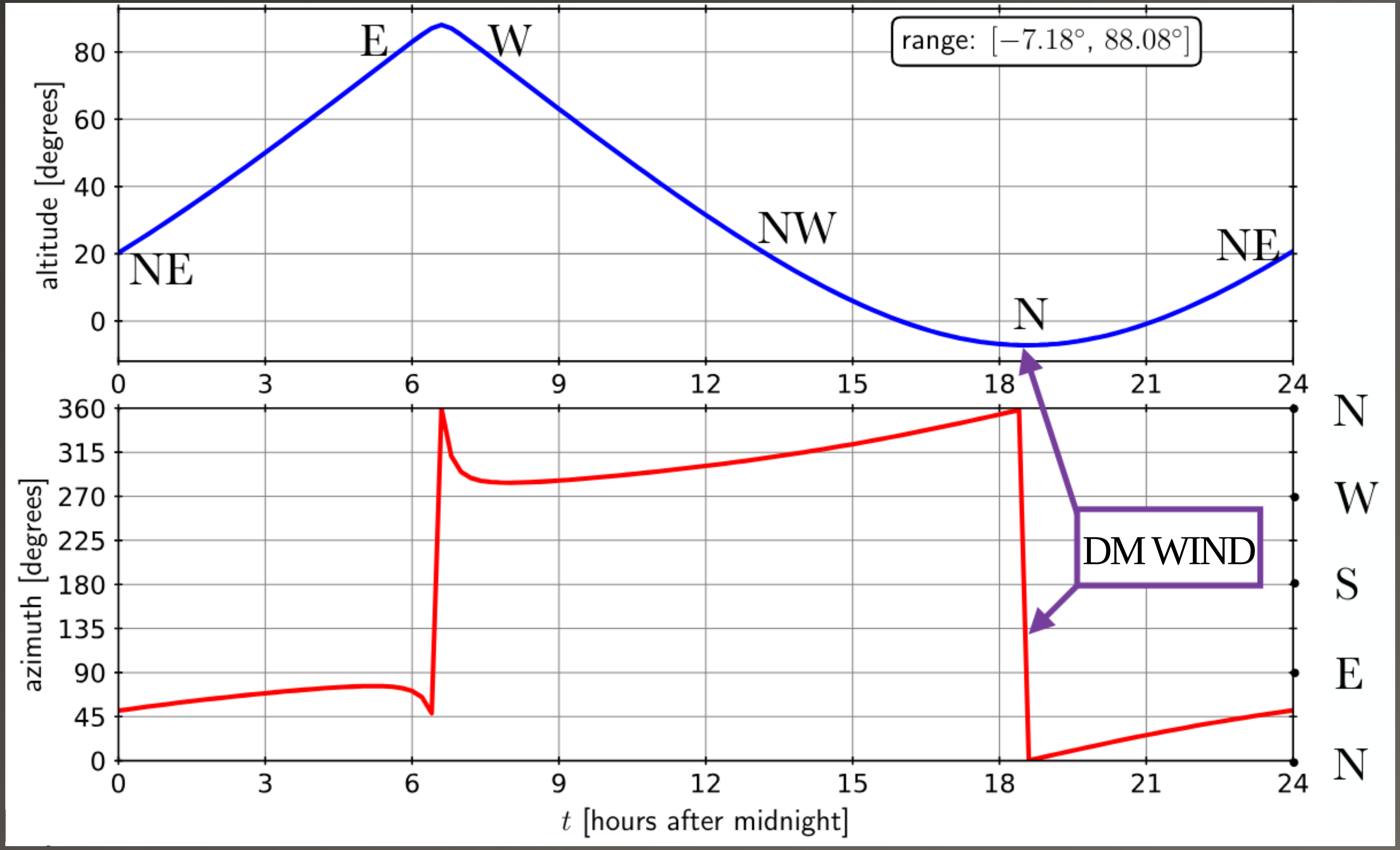


Effective dark matter "wind" from relative motion

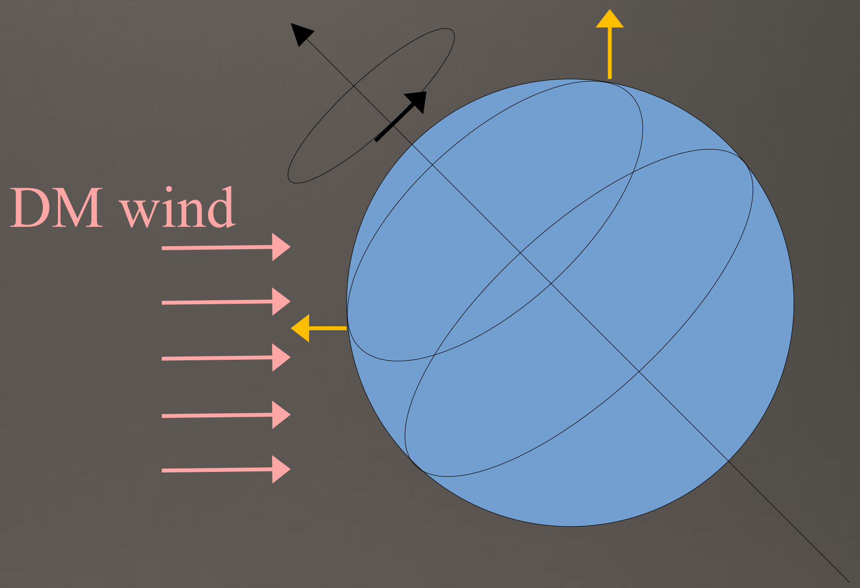


Change in relative orientation between detector and dark matter wind leads to ***daily*** modulation

Directional Detection



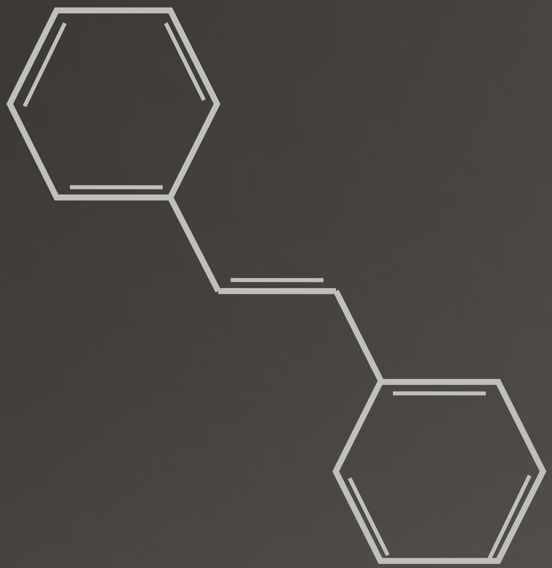
Credit: Benjamin Lillard



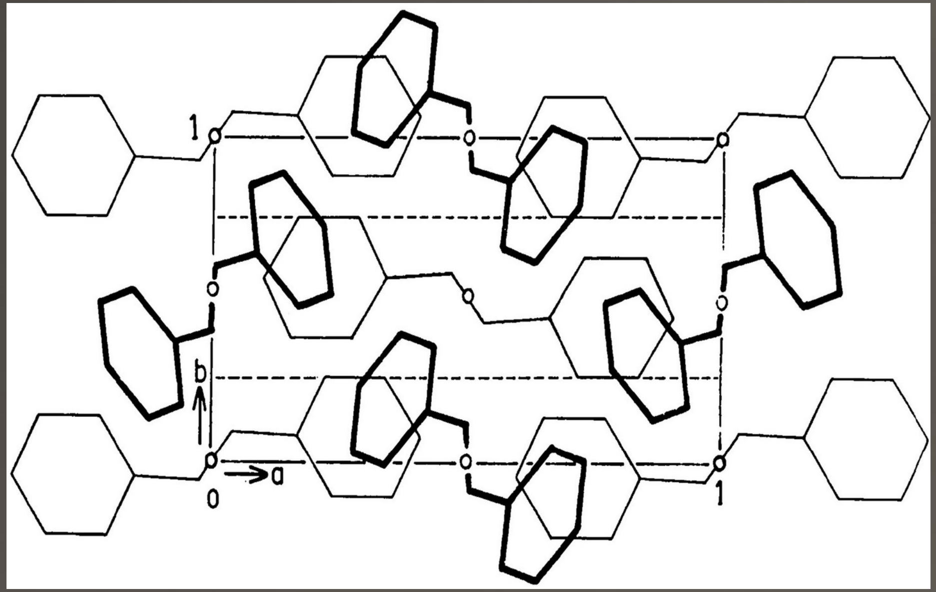
DM wind sweeps out a cone on the sky, with opening angle:

- $\theta_N = 47.6^\circ$ (today)
- $\theta_N \sim 42^\circ$ (average)

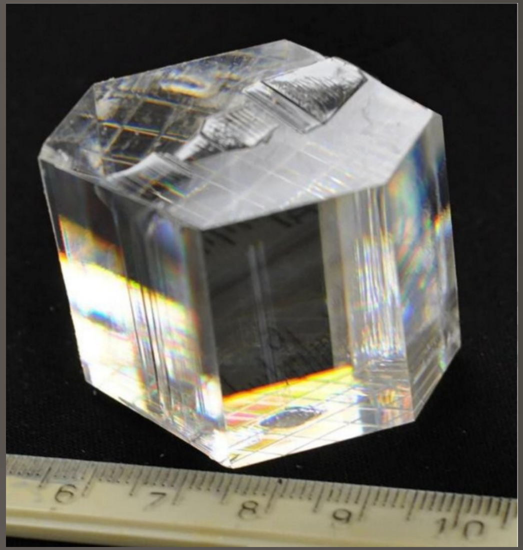
First Guess: Trans-Stilbene



De-localized and planar network of double bonds



Molecular planes oriented in crystal lattice

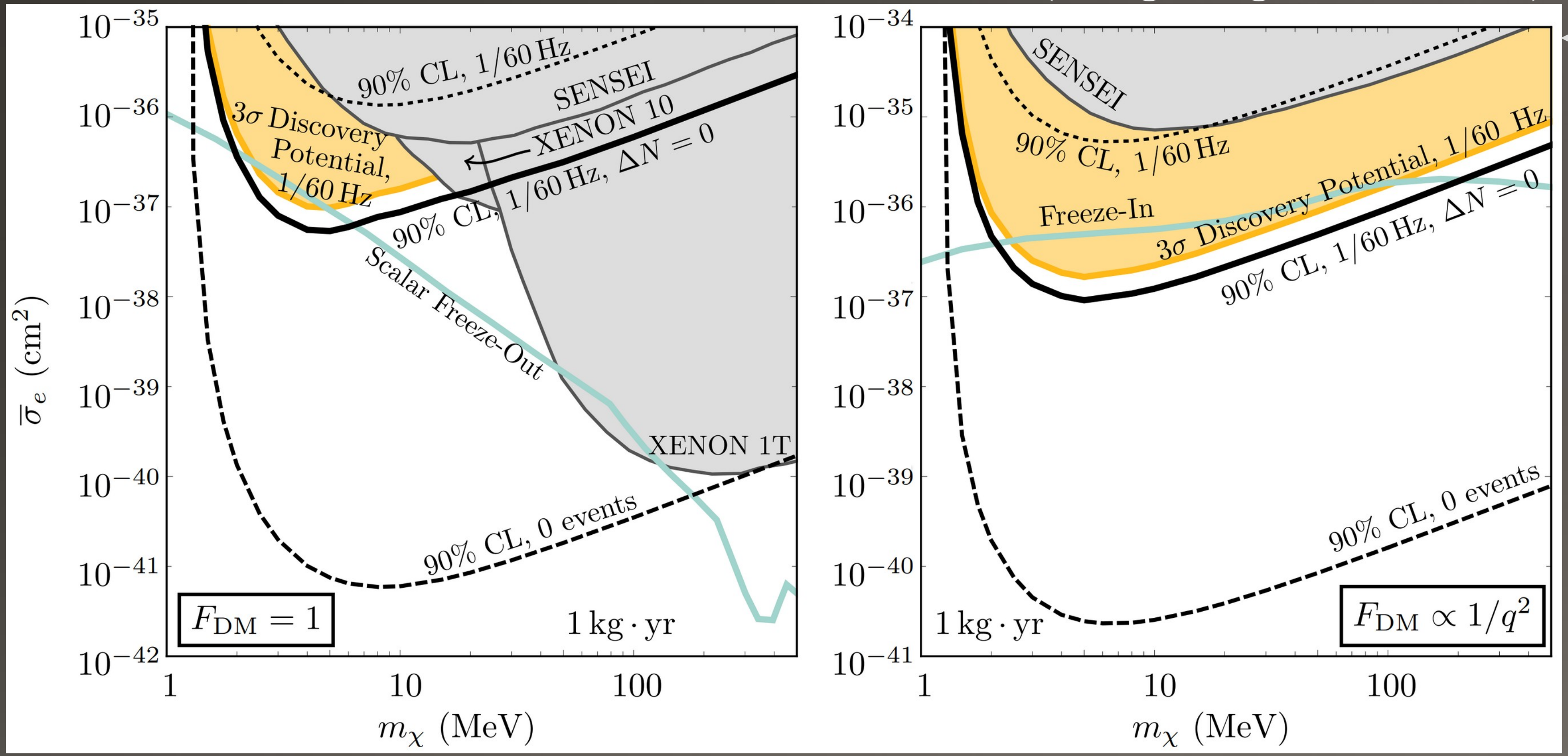


Carman, et.al. '18 (J. of Crystal Growth)

Large optical-quality crystals

Sensitivity & Reach

(Contact interaction) (Long-range interaction)



[CB, Kahn, Lillard, McDermott: 2103.08601]

Assuming realistic backgrounds

Exclusion w/o modulation

Discovery potential w/ mod.

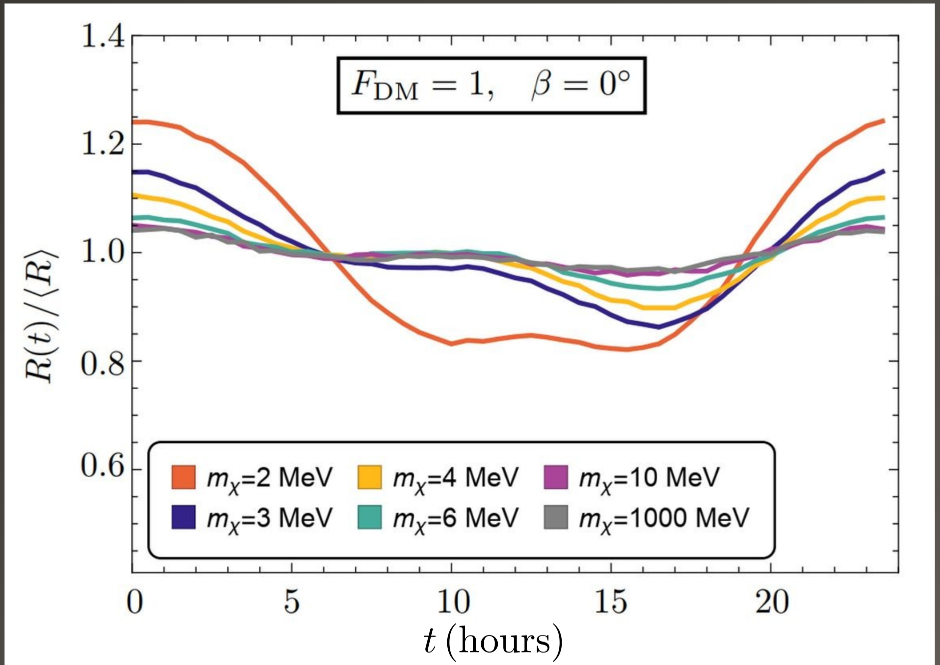
Exclusion w/ modulation

Modulating signals improve sensitivity by about two orders of magnitude and provide the potential for discovery

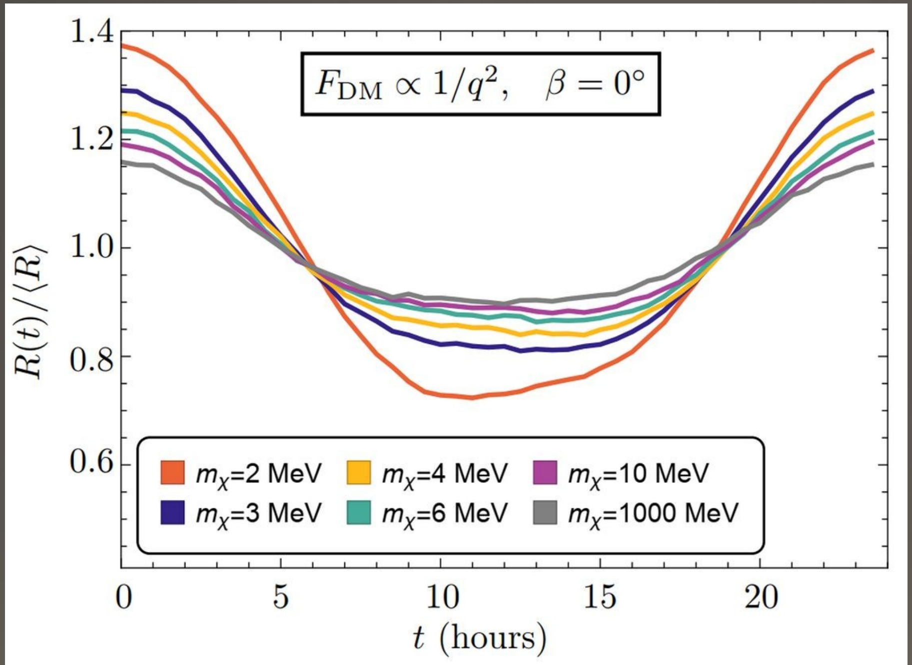
*1kg of t-stilbene can probably be found within a few blocks of this room

Daily Modulation

(Contact interaction)



(Long-range interaction)



Predicted rate changes by up to 70% throughout the day.

That's a verifiable signal!

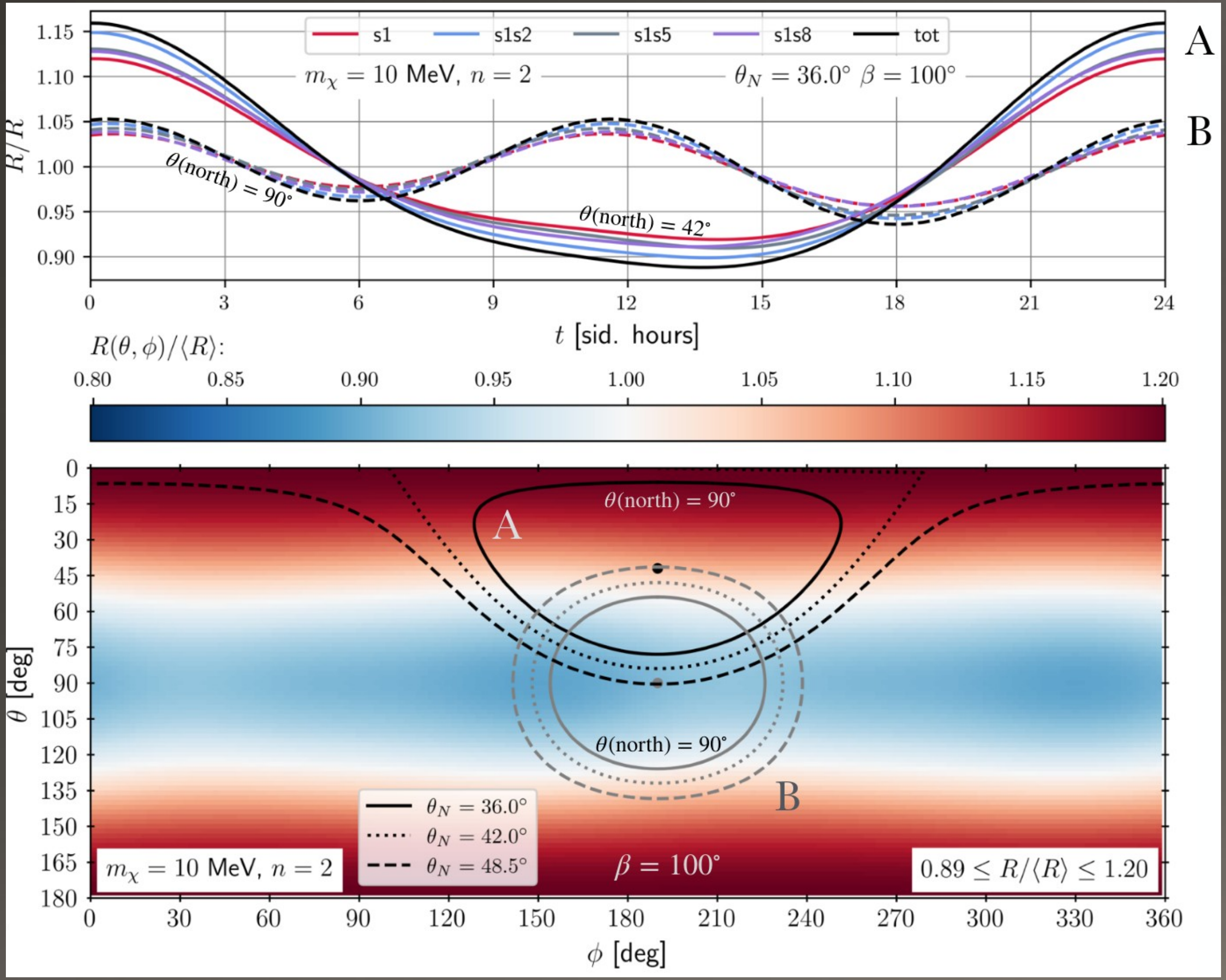
[CB, Kahn, Lillard, McDermott: 2103.08601]

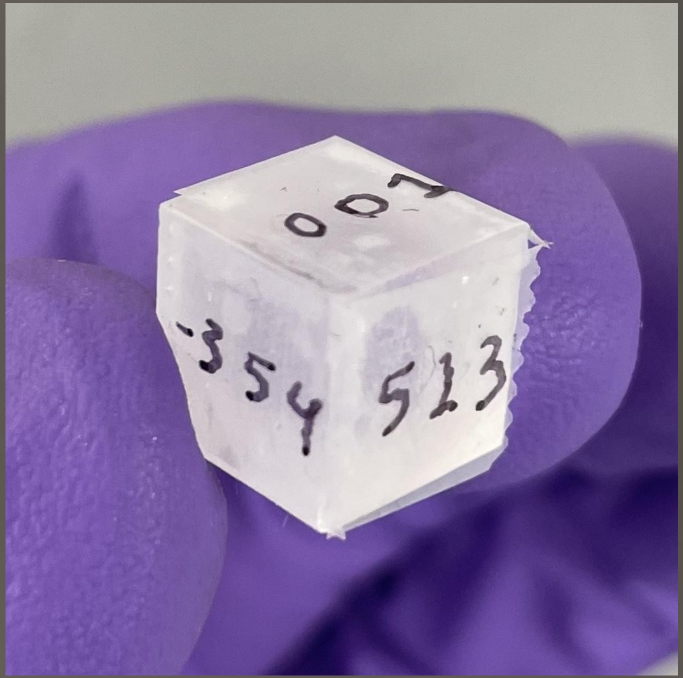
Modulation amplitude remains as high as 10% even at the highest masses. This is due to the fundamental anisotropy of the molecular form factor.

$$f_{\rm RMS} = [5\% - 25\%]$$

$$\boxed{N_\sigma \sim f_{\rm RMS}\sqrt{N_{\rm events}}}$$

Looking for Daily Modulation





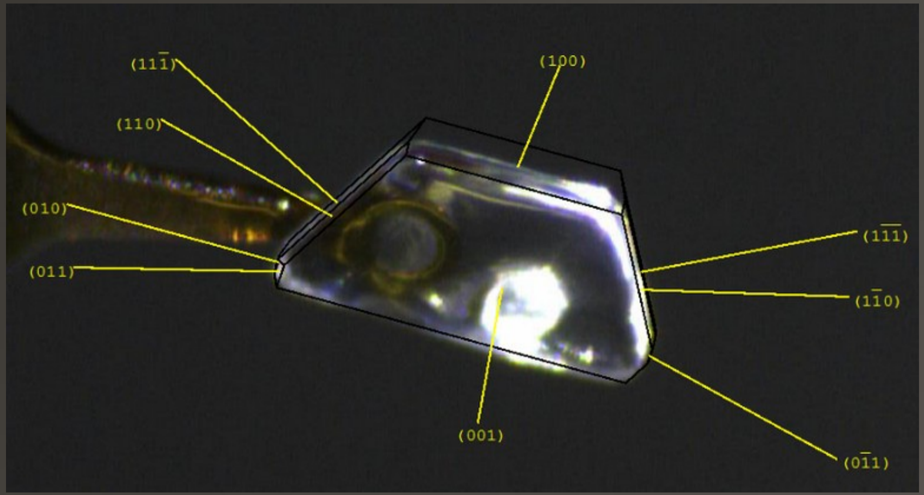
Different trajectories through rotation angles give distinct waveforms and phases.

So we can use the same crystal in many orientations or many different crystals!

Credit: Benjamin Lillard - https://github.com/blillard/vsdm

Characterizing t-stilbene

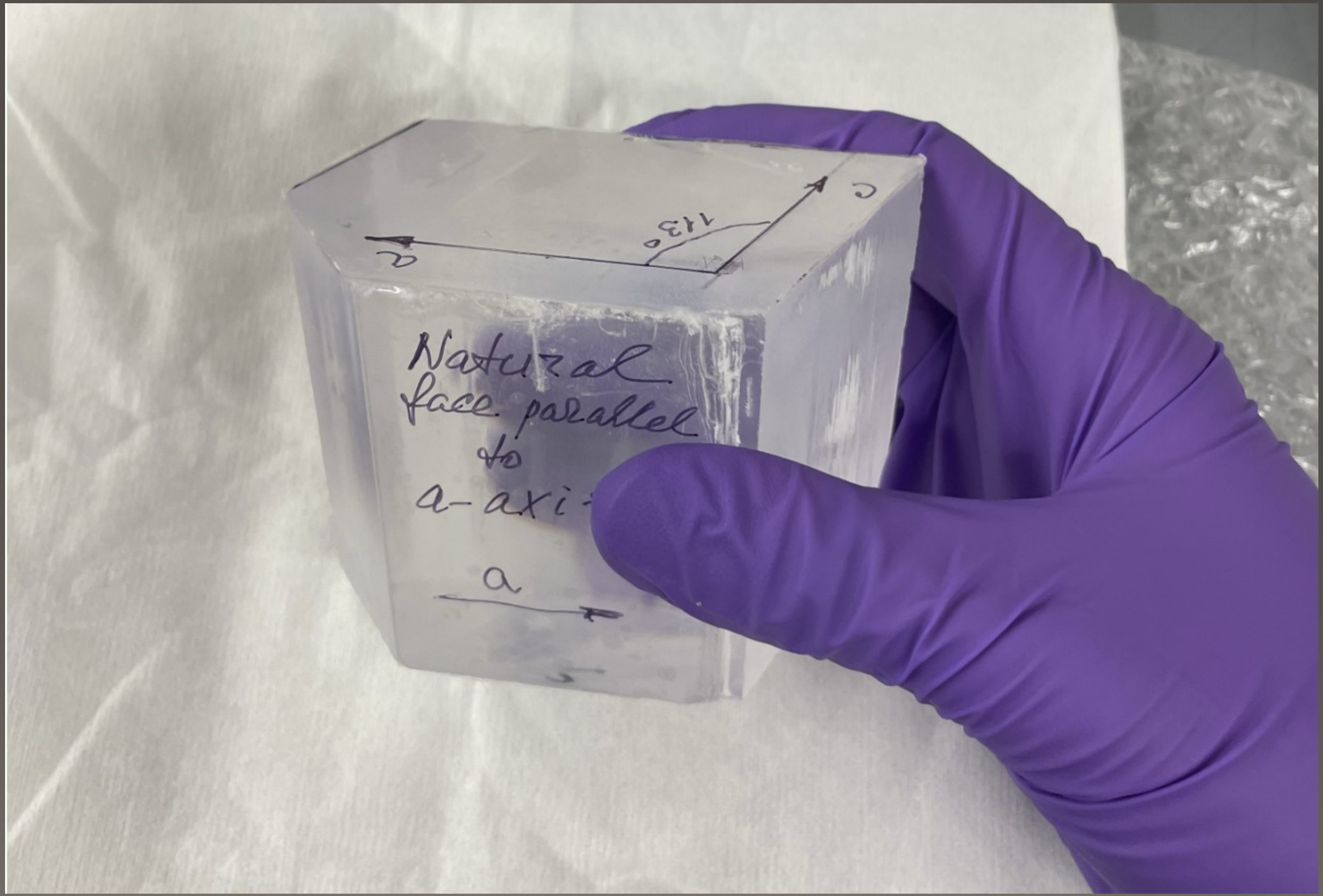
Calibrate at mg-scale



Credit: Dane Johnson
Freedman Group (MIT)

Natalia Zaitseva (LLNL)

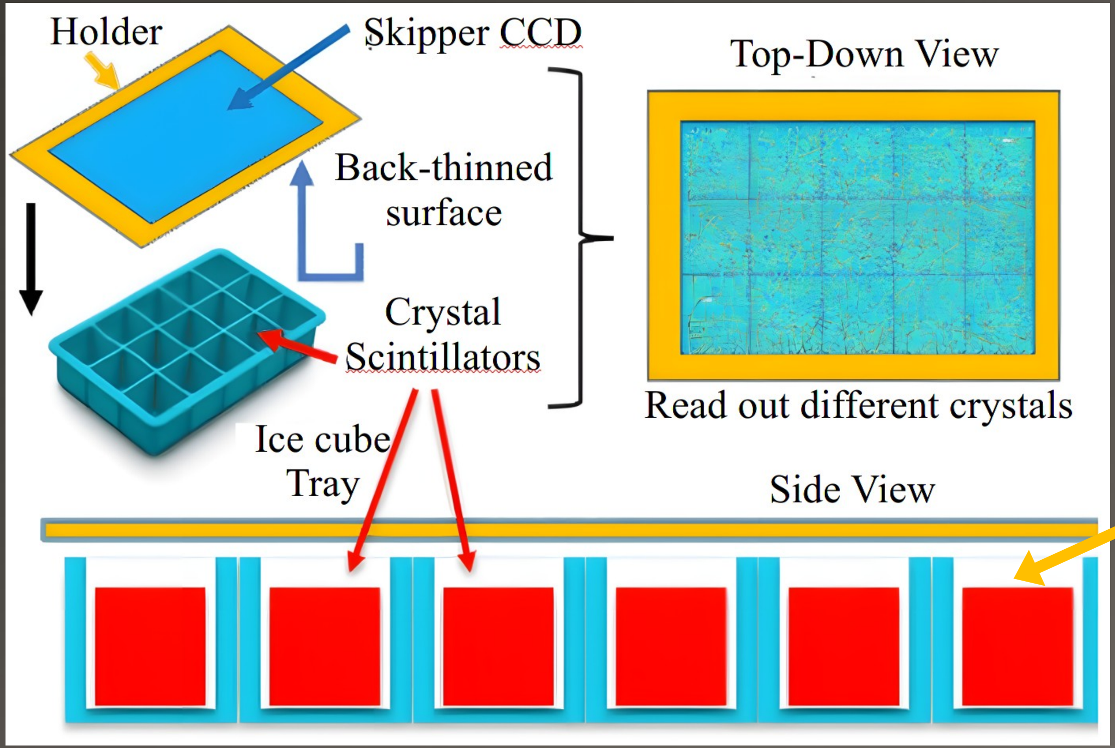
Deploy up to kg-scale

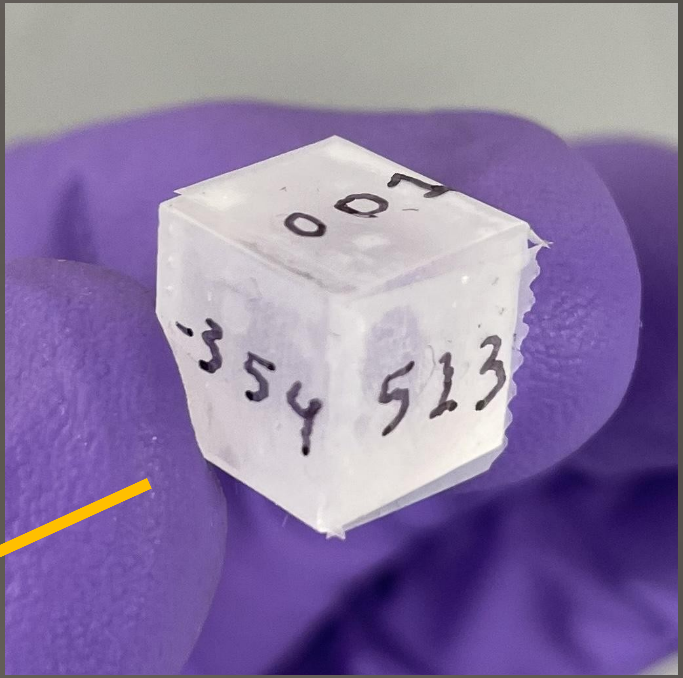


Experimental Deployment

DIANA*

Daily Modulation in an Intrinsically Anisotropic Array



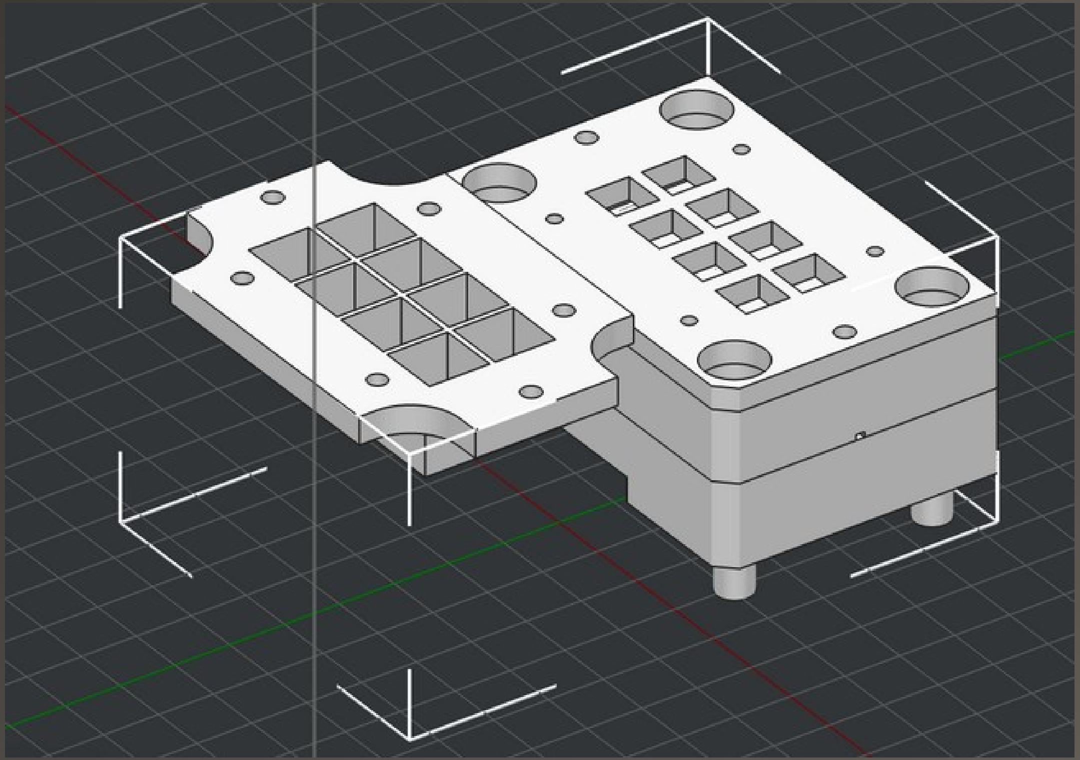


Many crystal samples can be read out by a single skipper CCD.

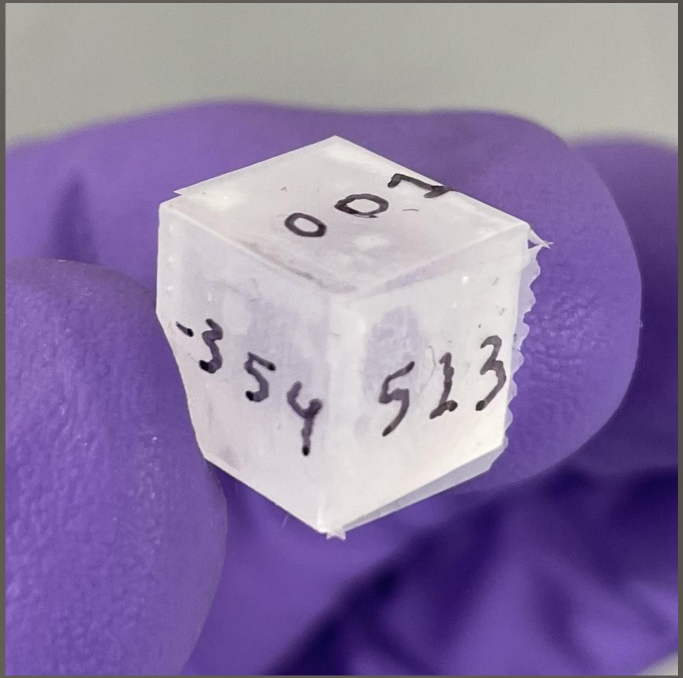
Collaboration: FermiLab, U. Toronto, MIT, UIUC, U. Oregon, Penn State

Experimental Status

Credit: Nora Hoch (MIT) & Dan Baxter (FermiLab)



CAD: Crystal holder

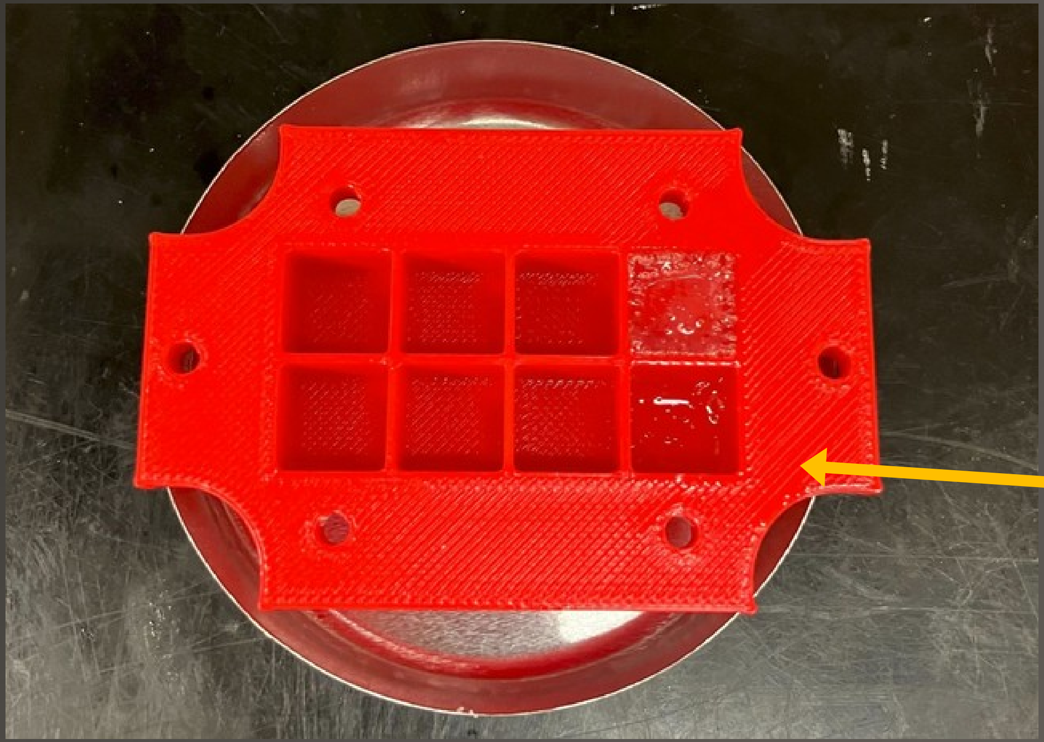


Prototyping is underway at FermiLab.

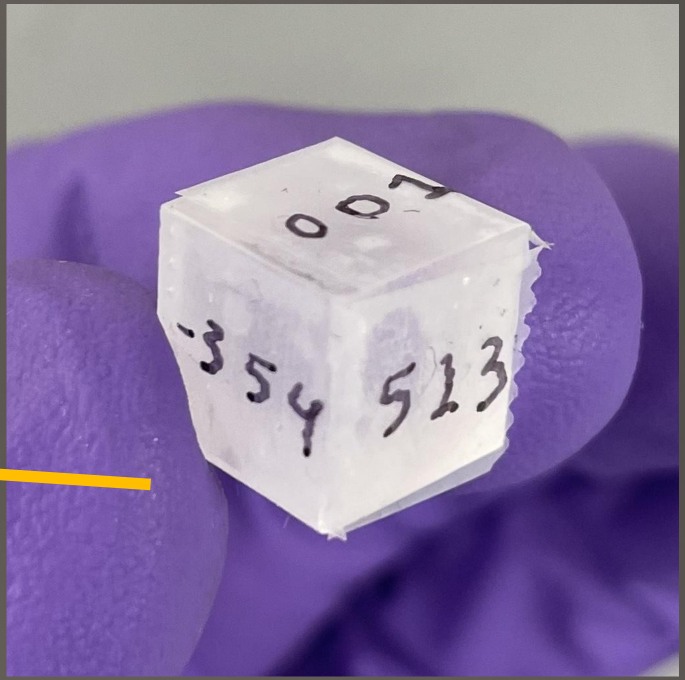
Collaboration: FermiLab, U. Toronto, MIT, UIUC, U. Oregon, Penn State

Experimental Status

Credit: Nora Hoch (MIT) & Dan Baxter (FermiLab)



Printed Prototype



3D printed crystal sample holder to test mounting and orientations.

Collaboration: FermiLab, U. Toronto, MIT, UIUC, U. Oregon, Penn State

Astro-Skipper CCDs as Readout

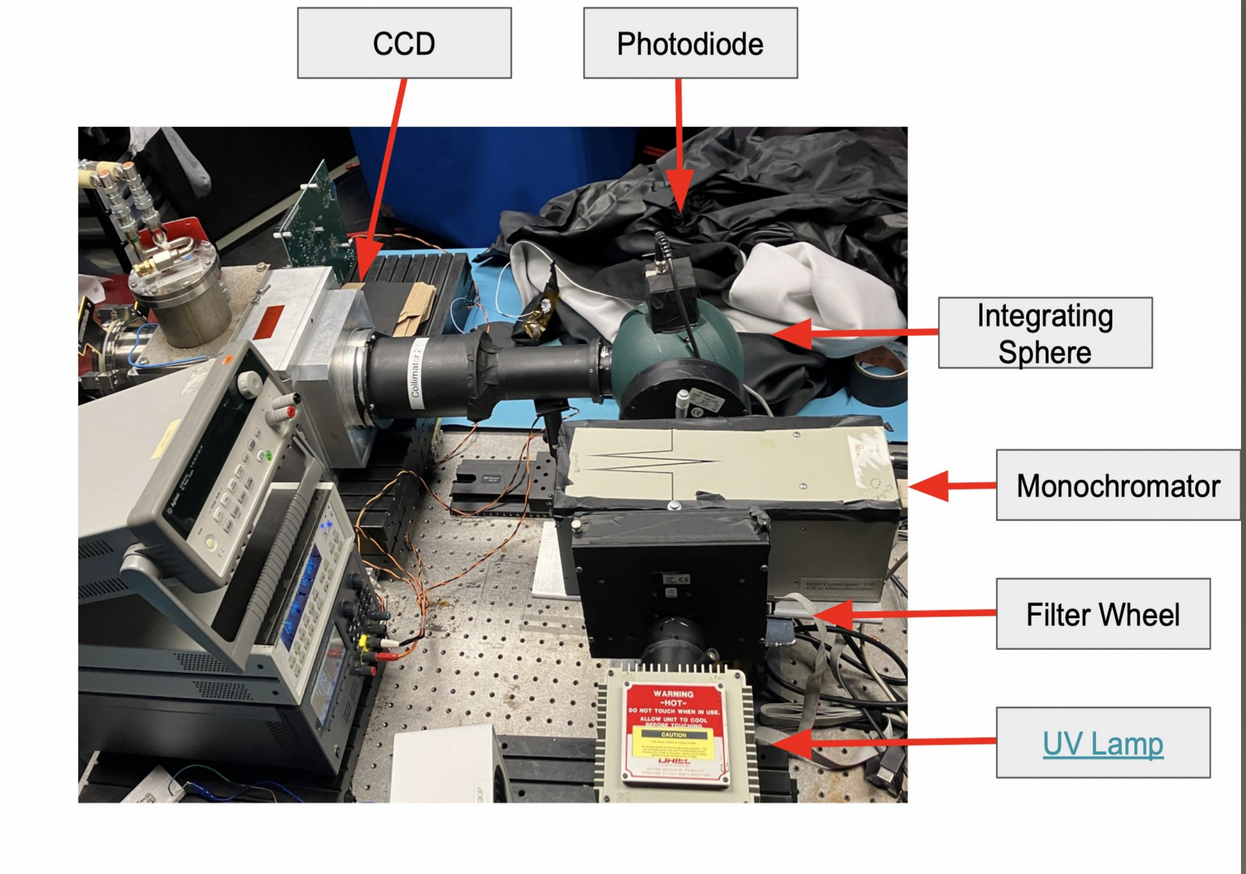
Taking pictures of crystals with a very sensitive CCD

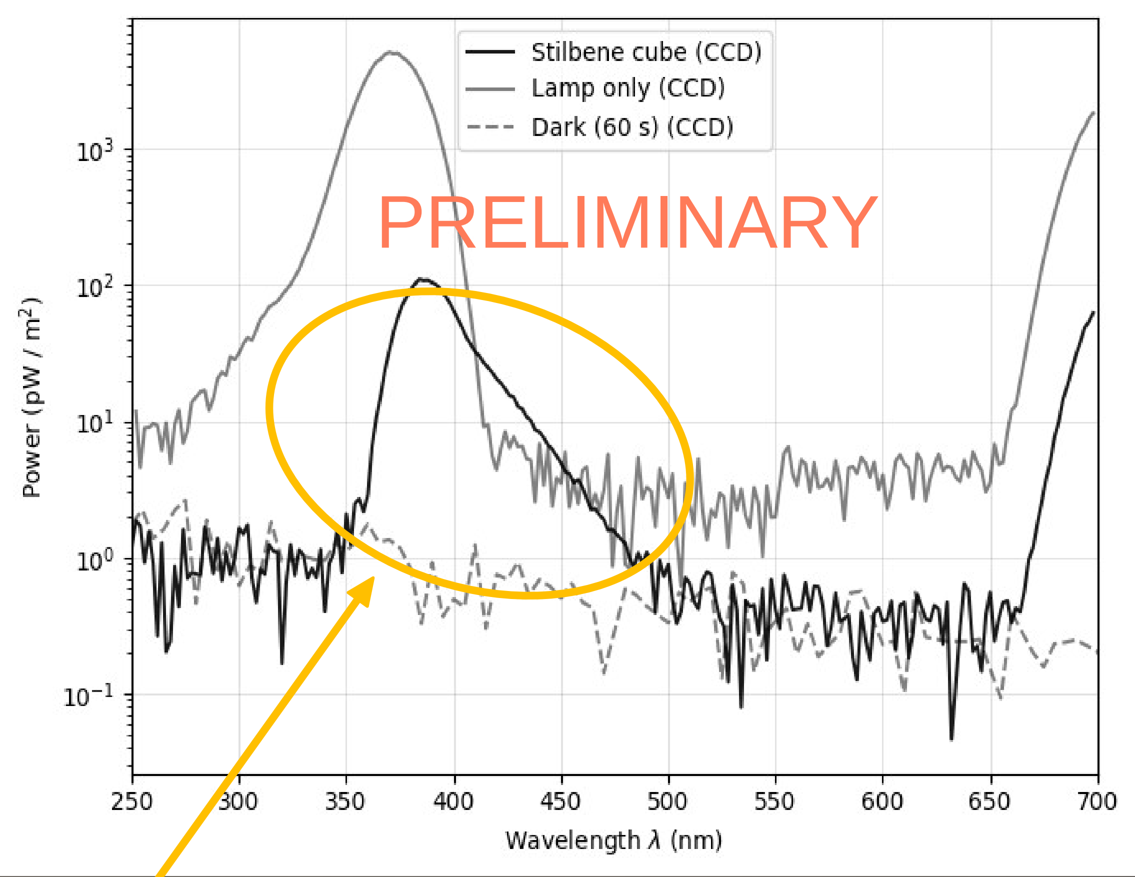
Skipper CCD Measurements of trans-Stilbene

Daniel Baxter, Alex Drlica-Wagner,
Edgar Marrufo, Brandon Roach

CCD Measurement of Signal

Detecting fluorescence with Skipper CCDs



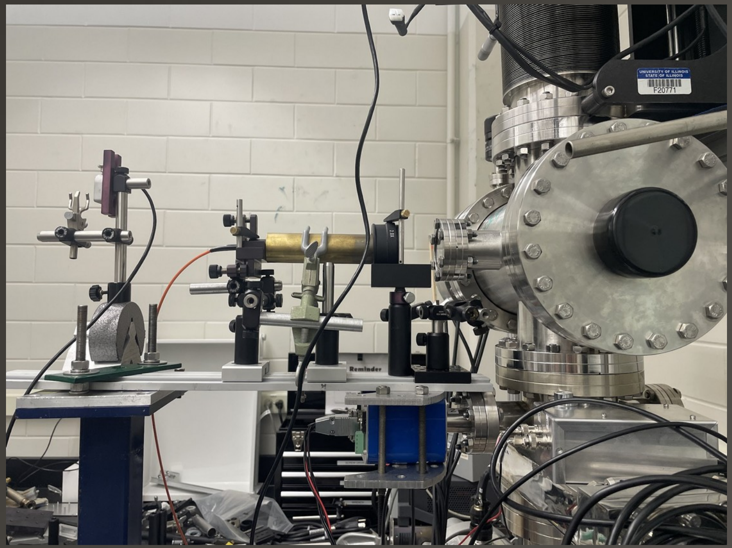


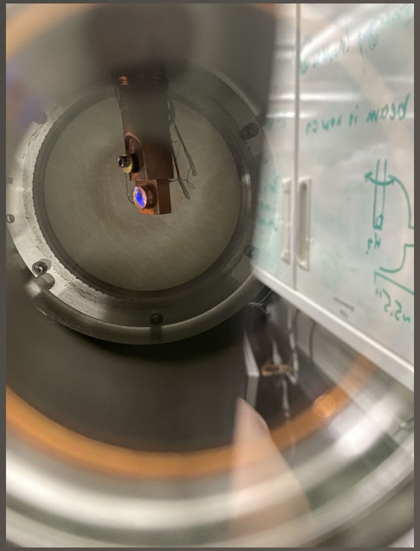
The fluorescence spectrum of t-Stilbene w/ single-photon precision

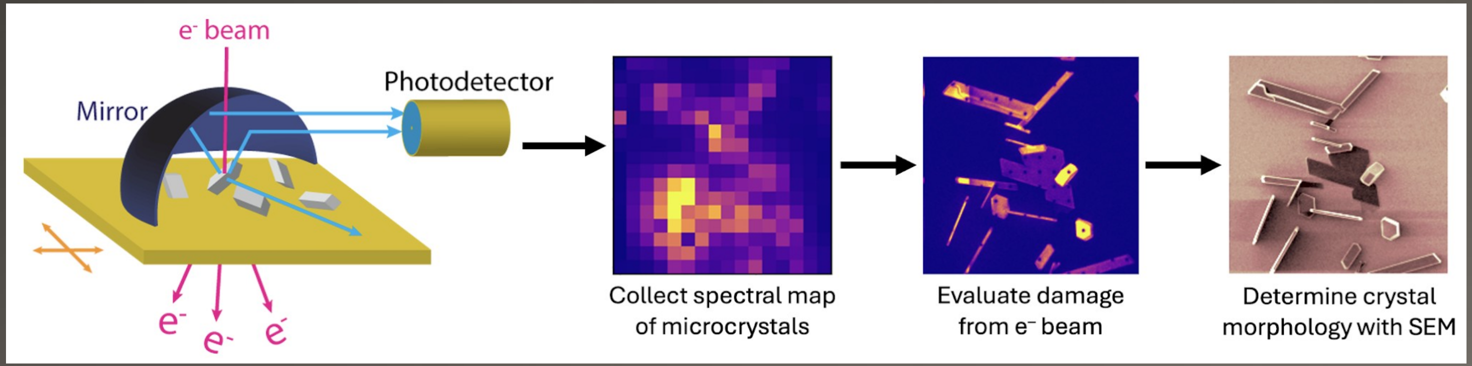
Credit: Dan Baxter, Edgar Marufo, & Brandon Roach (FermiLab)

Measuring Anisotropy

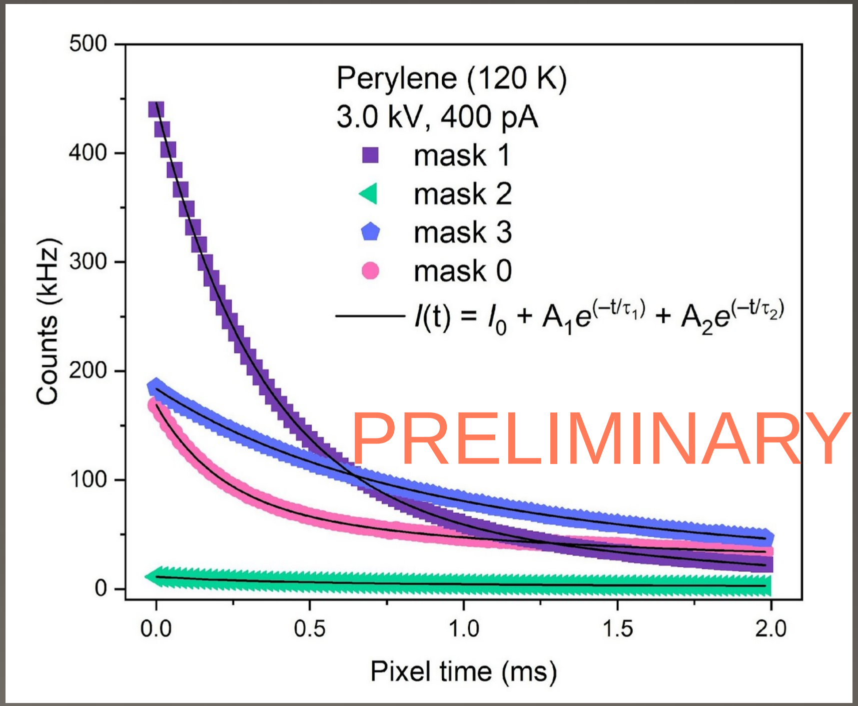
Electron Impact → Cathodoluminescence







Preliminary observation of directionality in experiments using electron impact

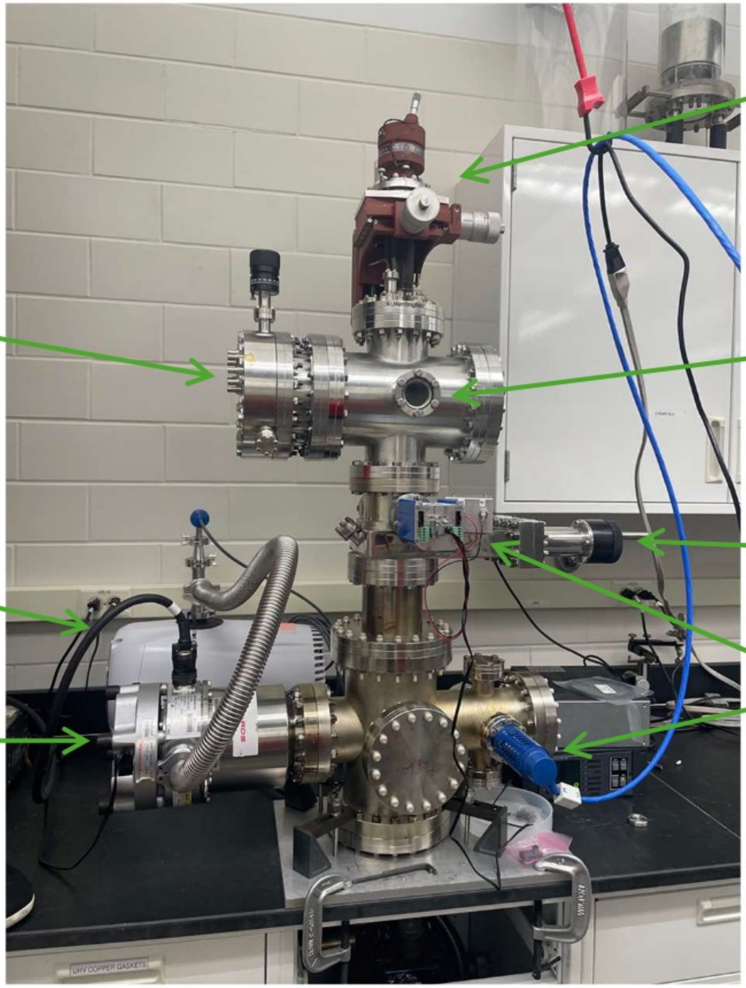


Credit: Yoni Kahn (U Toronto) + Abbamonte Group(UIUC) + Dane Johnson (MIT)

Calibrating Anisotropic Materials

Calibration of Anisotropy via Electron Impact

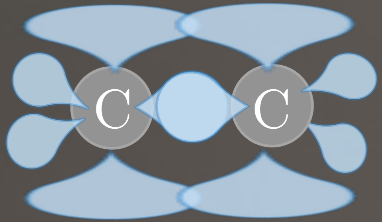
Angle-dependent stilbene fluorescence gizmo (ADSFG)



Abbamonte Group (UIUC)

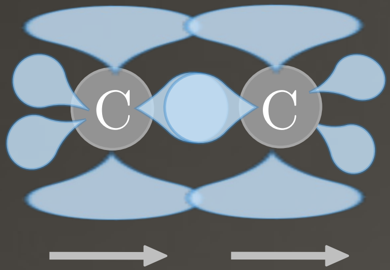
The Molecular Migdal Effect(s)

Sensitivity to Nuclear Recoils



<u>Center of mass recoil (CMR) effect</u>

Caused by center of mass motion



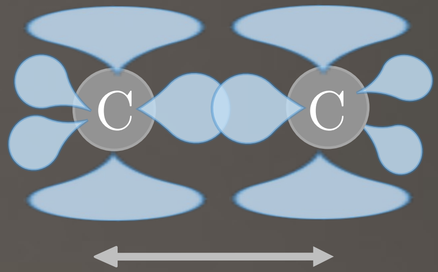
COM recoil effect is the molecular analog of the semiclassical Migdal effect

$$P_{CMR} \sim \frac{m_e}{M_{mol}}$$

Suppressed by kinematic factor due to moving the whole molecule.

<u>Non-adiabatic coupling (NAC) effect</u>

Caused by relative motion



NAC caused by effects beyond Born-Oppenheimer approximation

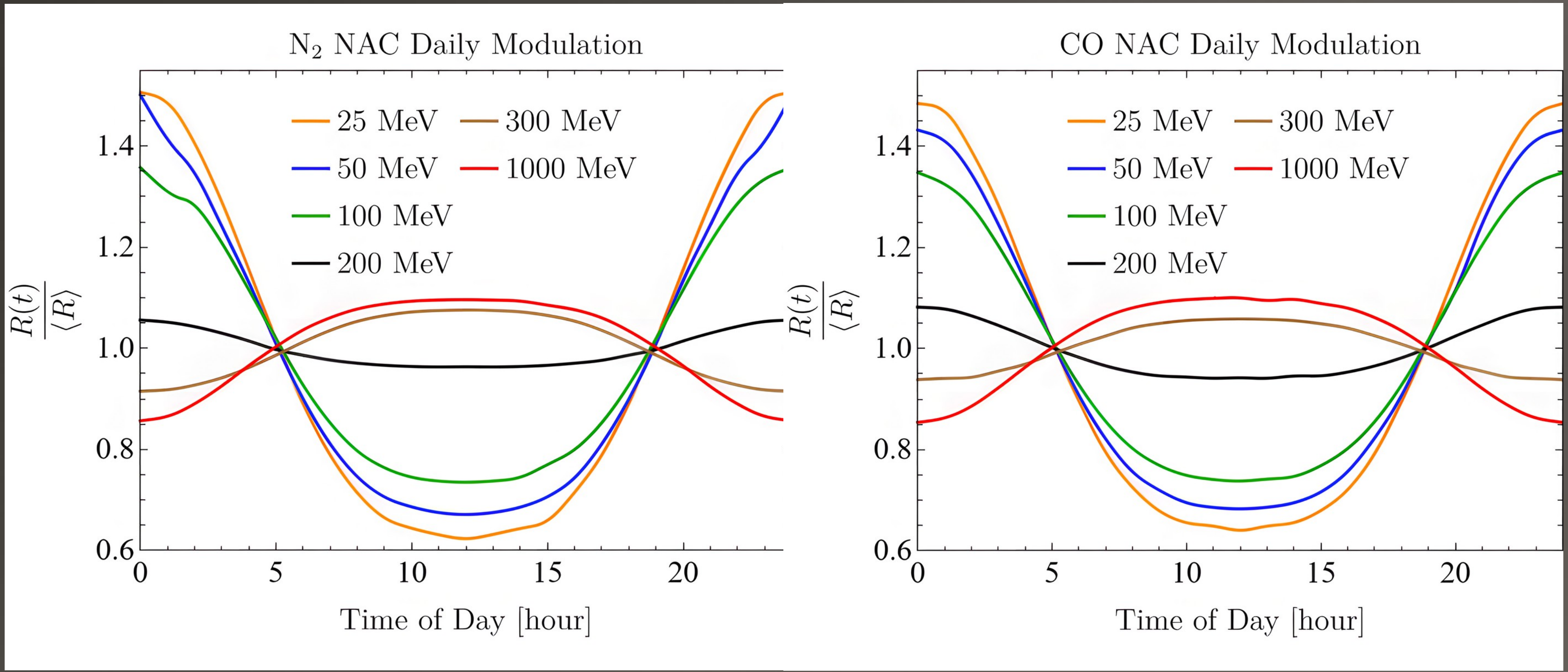
$$P_{NAC} \sim \frac{m_e}{M_N}$$

Suppressed by kinematic factor due to moving a single atom.

Blanco '22: 2208.09002

Directional Molecular Migdal Effect

[CB, Harris*, Kahn, Lillard, Perez-Rios: 2208.09002]



Predicted rate changes by up to 80% throughout the day.

The daily modulation phase is mass dependent.

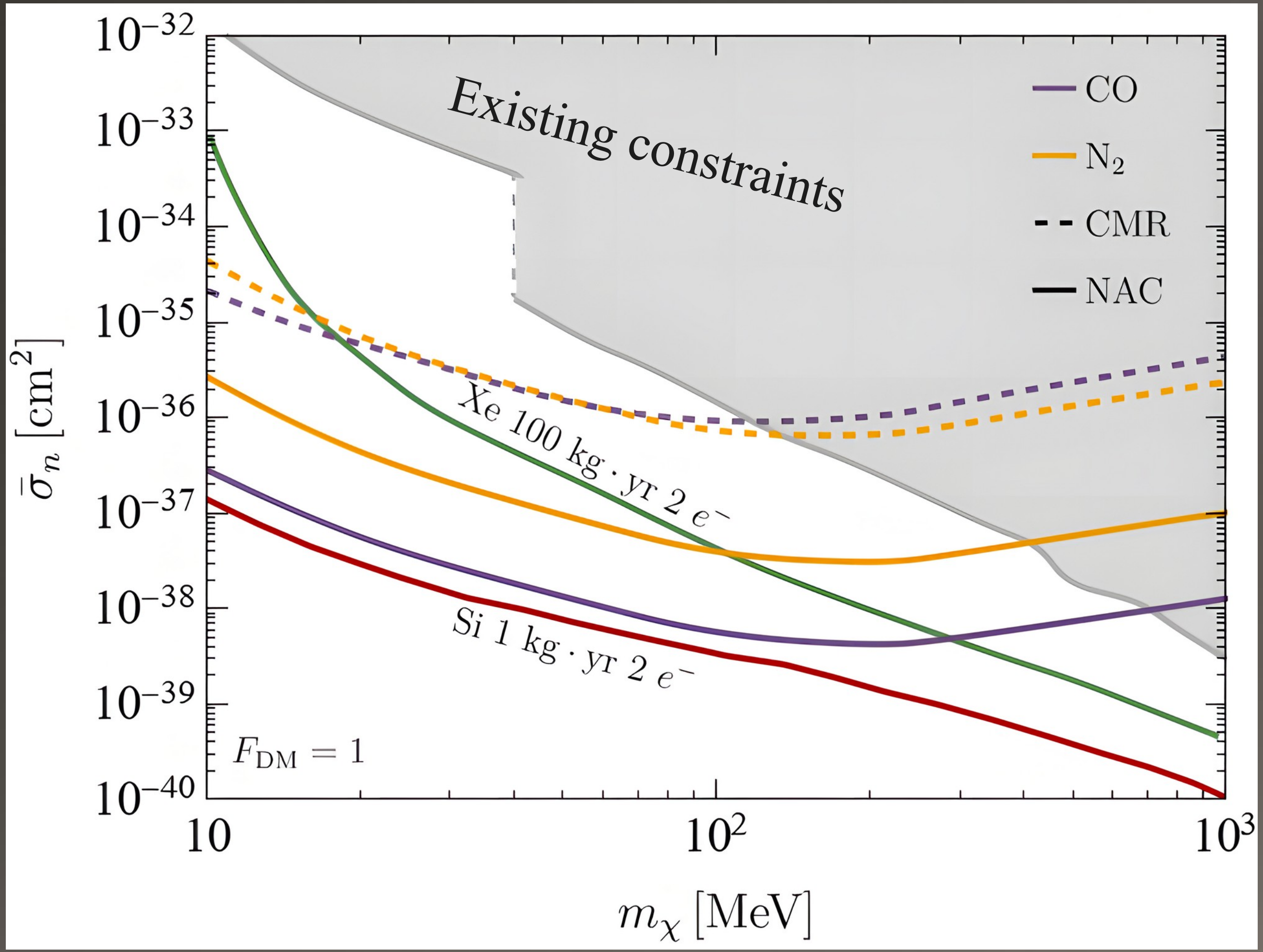
Persistent daily modulation at large dark matter mass is generically predicted for the molecular Migdal effects.

We predict that the same class of molecules that make good directional detectors for electron scattering will also be ideal for nuclear scattering because of the directional molecular Migdal effects.

The Molecular Migdal Effect(s)

Sensitivity to Nuclear Recoils

(Contact interaction)



[CB, Harris*, Kahn, Lillard, Perez-Rios: 2208.09002]

Si rate is entirely due to non-adiabatic processes. [Esposito & Rocchi 2505.08864]

Center of mass recoil effect is predicted to be subdominant at all masses.

Non-adiabatic coupling effect is predicted to dominate due to favorable kinematic factor.

Simplest molecular models already competitive.
Is there an optimal molecular target?

Finding Optimal Targets

Problem: Chemical space is unreasonably large

How many molecules possible with
C, O, N, F, H?

< 9 atoms: 100s of Thousands (DFT Computable)

< 30 atoms: 100s of Billions (Intractable)

...toluene has 15, xylene has 18, t-stilbene has 26

Method
1. Look for known favorable properties - *cheminformatics*
2. Extra(intra)polate onto new molecules – *machine learning*

ML for DM Direct Detection

Property prediction
Energies & Matrix elements

Molecular Generation
Sample latent space → new molecules

Small molecules : Using exhaustive database (< 9 atoms)
Characterize neural nets
→ Prove it's possible to learn from small subsample

Large molecules: Sparse dataset up to 10s of atoms
Scale architecture
→ Generate candidate molecule *shortlist*

Dataset of energies + oscillator strengths:
PubChemQ: a dataset of ~3M organic molecules *computed* using Density Functional Theory

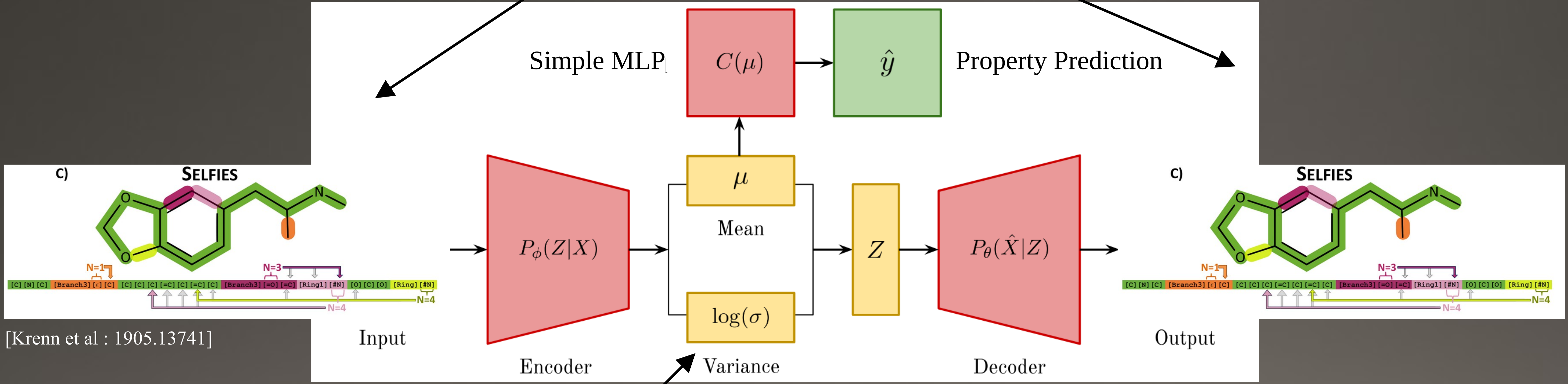
[C. Cook, CB, J. Smirnov 2501.00091]

ML for DM Direct Detection

Molecular Space: Energies & Matrix elements

Variational Autoencoders: Sample latent space → new molecules

Discrete and sparse data space



[Krenn et al : 1905.13741]

Continuous latent space

Regression Model in Action

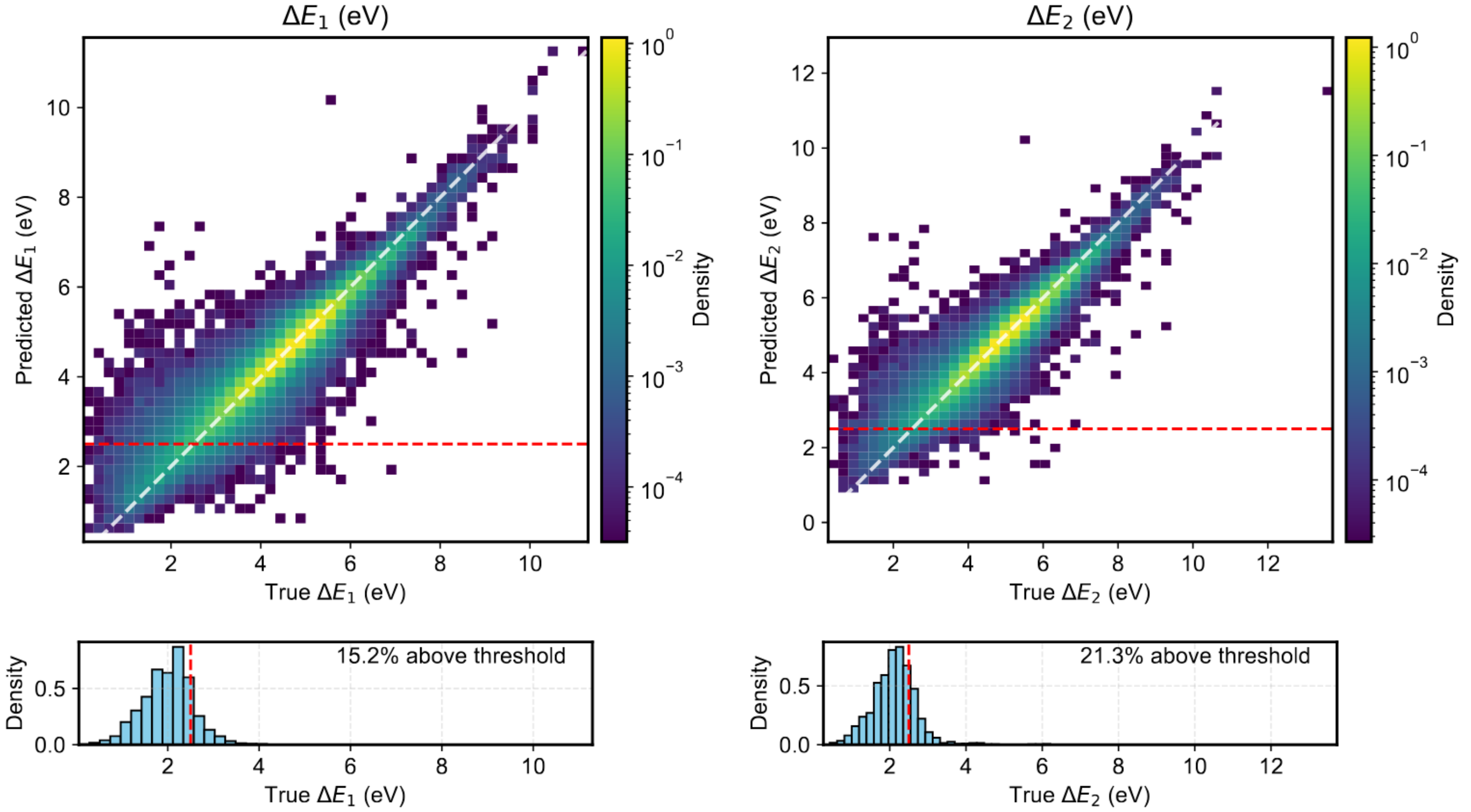


FIG. 19. We show the density of predicted values vs true values for the transition energies of our validation set, with a red line that corresponds to the threshold placed on the predicted value. The Histogram shows the distribution of true values for molecules whose predicted value is above the threshold.

[C. Cook, CB, J. Smirnov 2501.00091]

Regression Model in Action

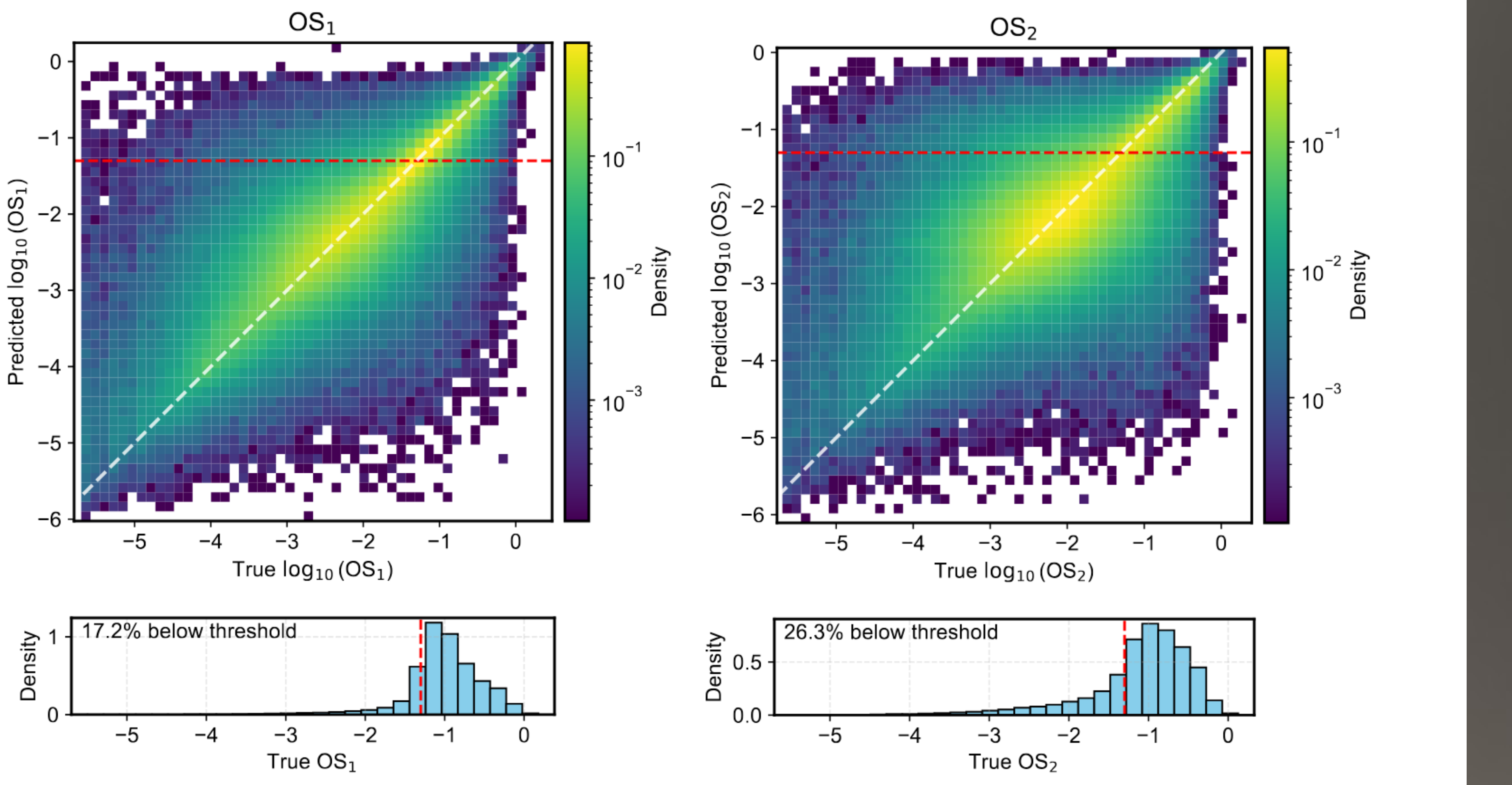


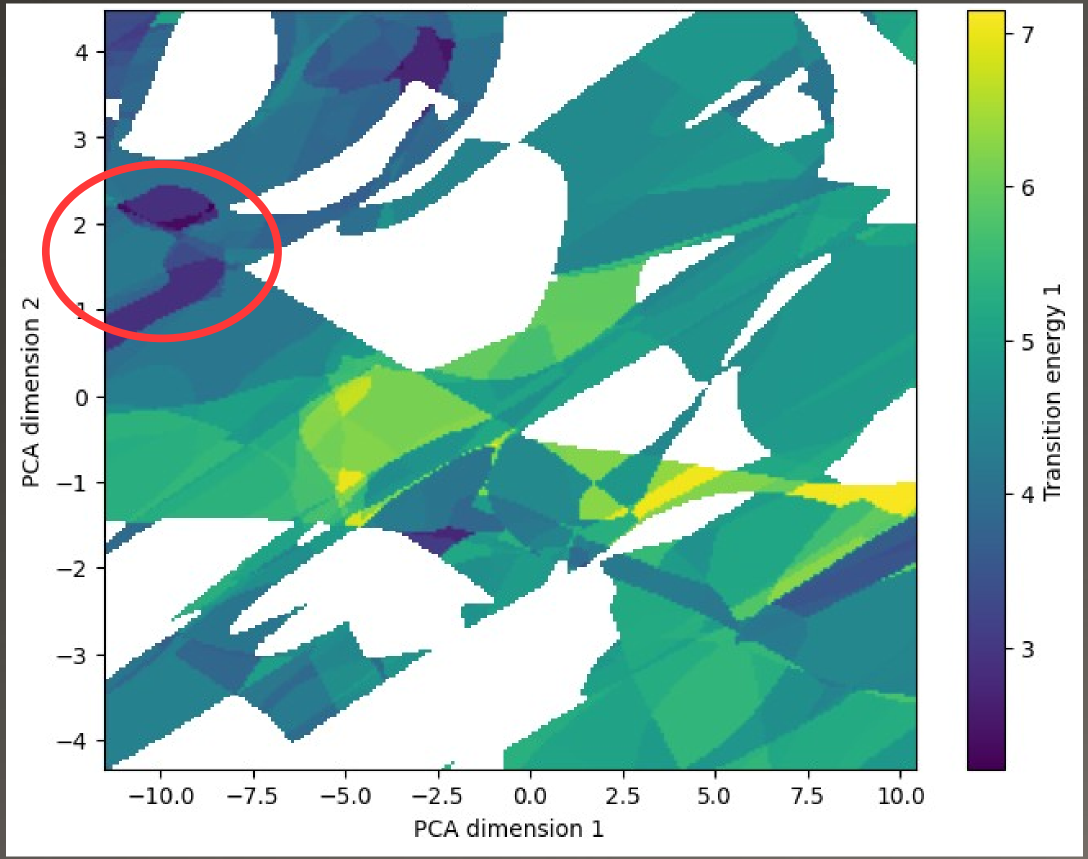
FIG. 20. We show the density of predicted values vs true values for the oscillator strength of our validation set, with a red line that corresponds to the threshold placed on the predicted value. The Histogram shows the distribution of true values for molecules whose predicted value is above the threshold.

[C. Cook, CB, J. Smirnov 2501.00091]

ML for DM Direct Detection

Property prediction: Energies & Matrix elements

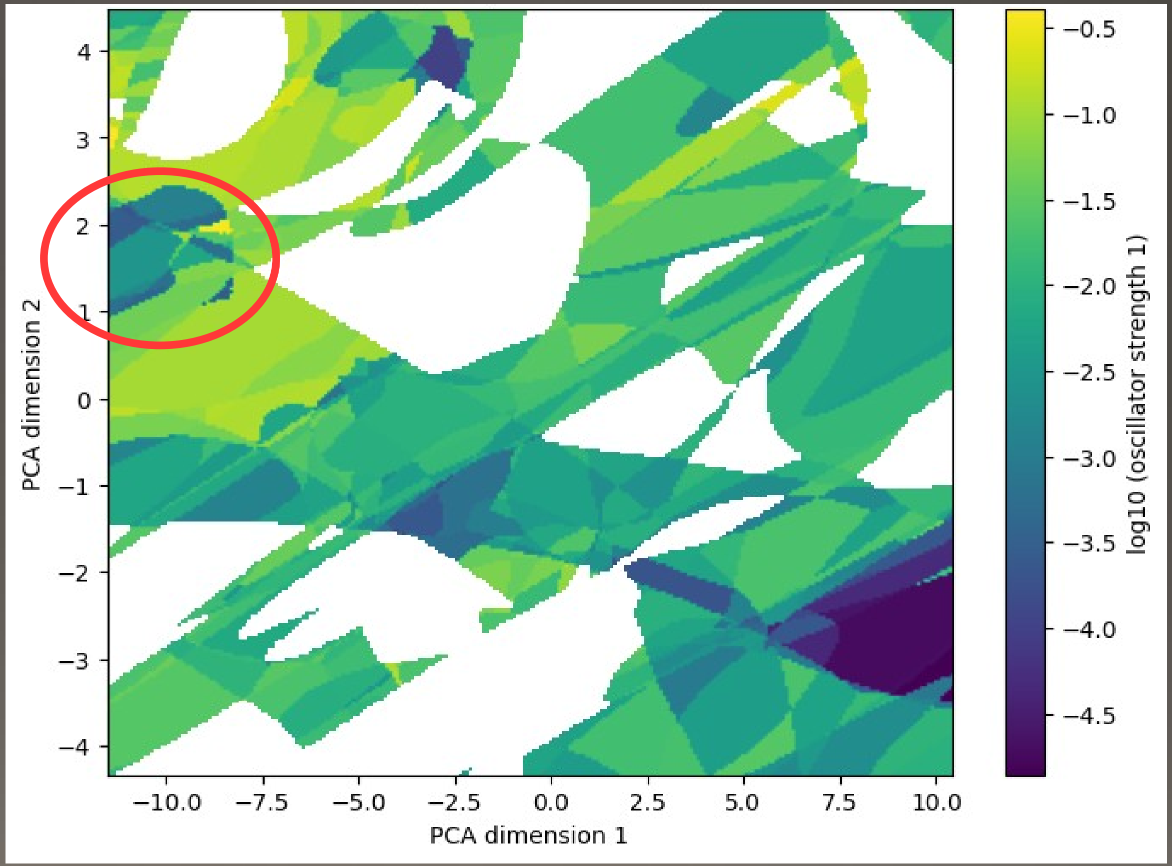
ΔE



Molecular latent space

Sample latent space → new molecules

$\langle r_{01} \rangle \approx |f_{0,1}|/q$



[C. Cook, CB, J. Smirnov 2501.00091]

Clustering: Results

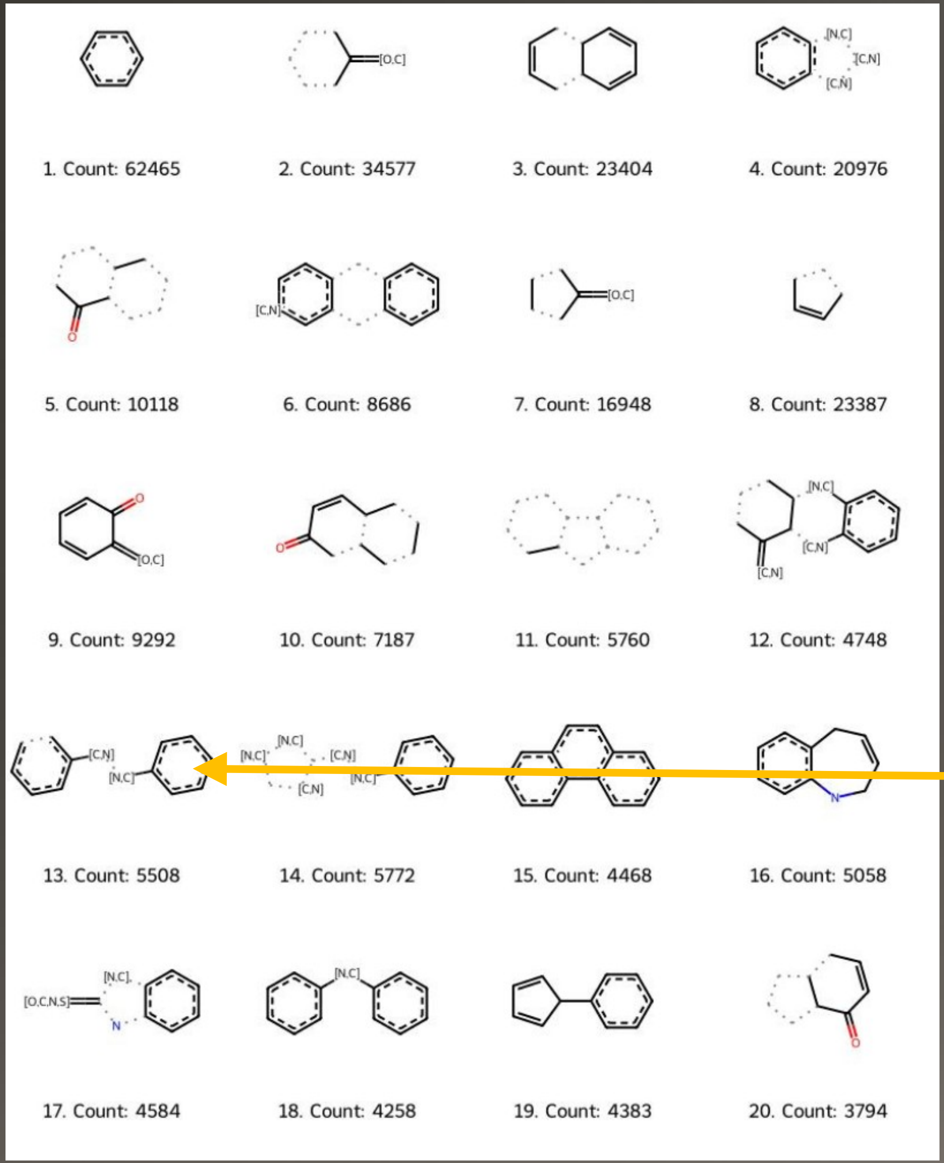
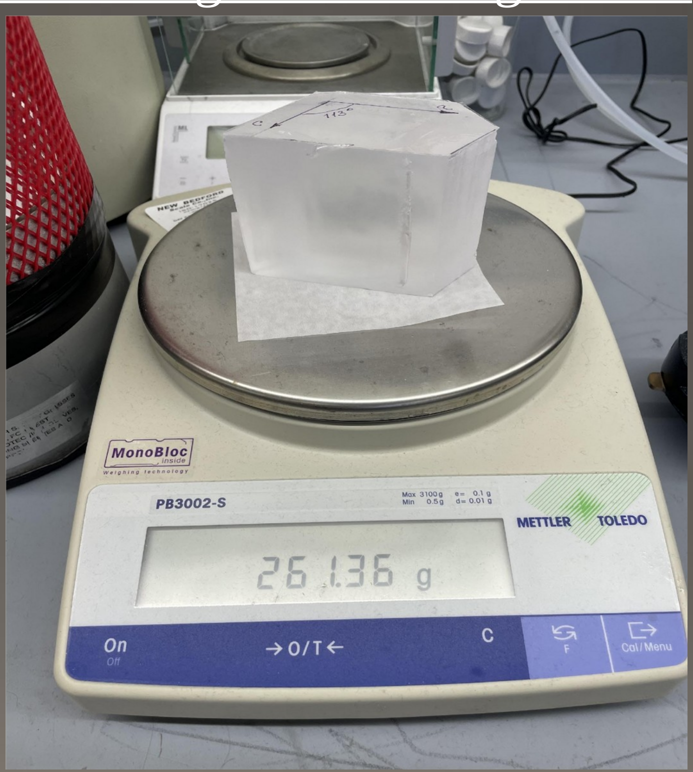


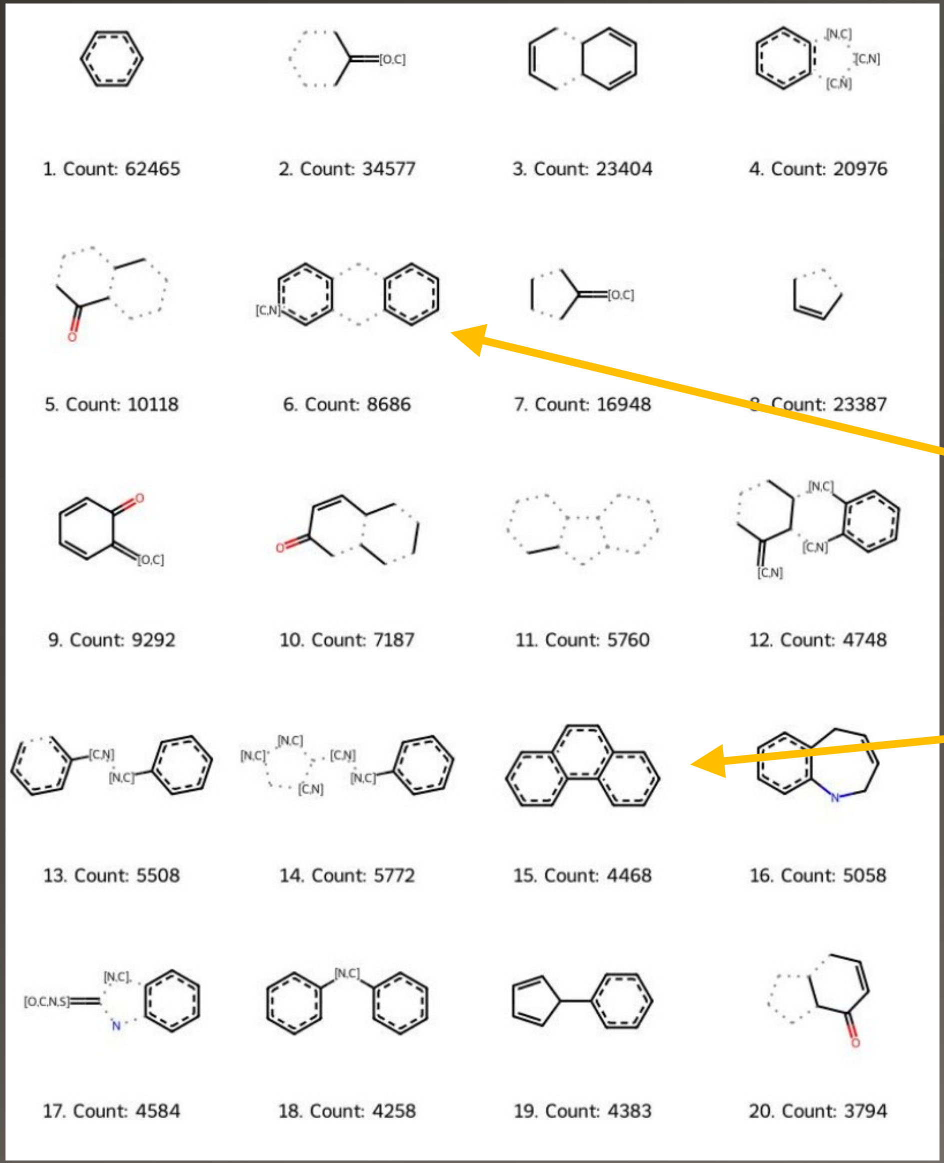
FIG. 7. Top 20 molecular motifs ranked by score. Note that certain atoms can be substituted by [C,N,O,S] as long as the structure remains isoelectronic. Dotted lines indicate that the bond could be a double or single bond.

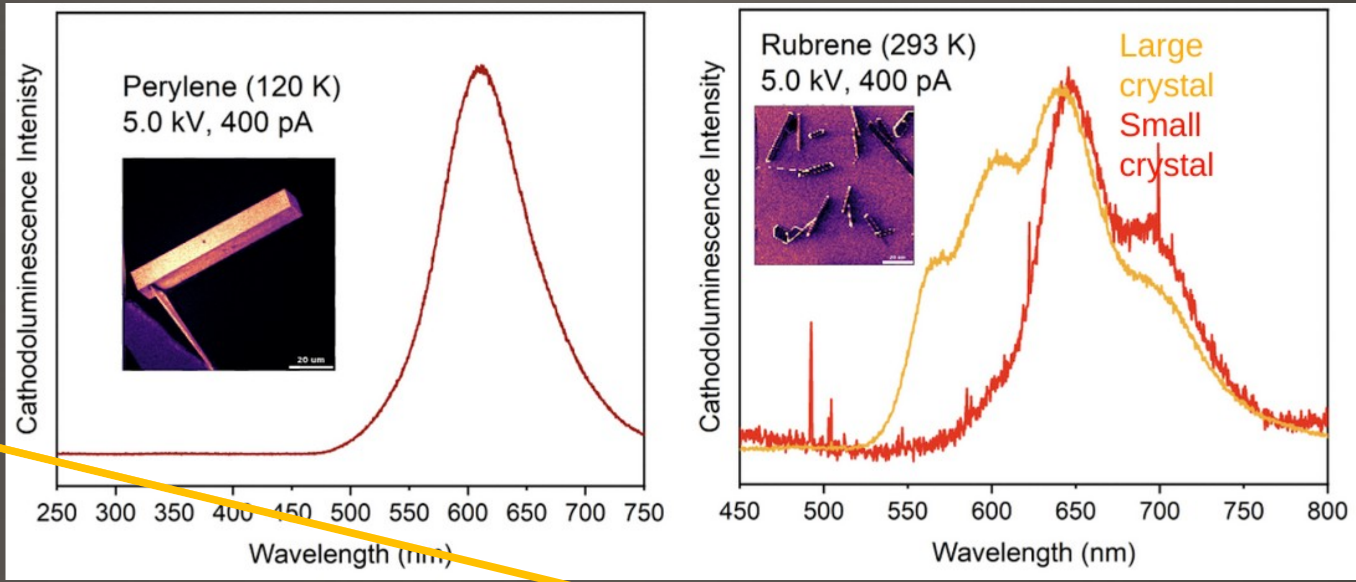
[C. Cook*, CB, J. Smirnov 2501.00091]

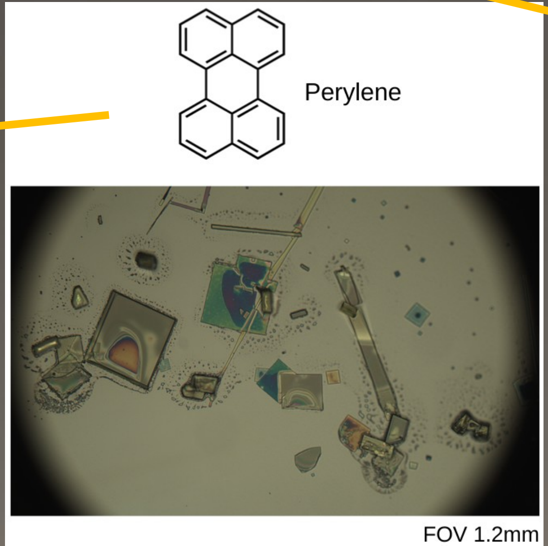
<u>In hindsight: On the right track</u>

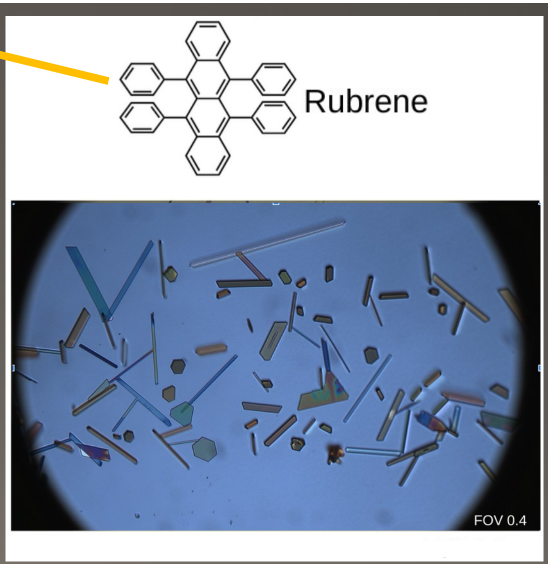


New Target Materials









[C. Cook*, CB, J. Smirnov 2501.00091]

Credit: Dane Johnson (MIT)

Conclusions

1) We have done an extremely effective job looking for WIMPs above a GeV, now we must look beyond.

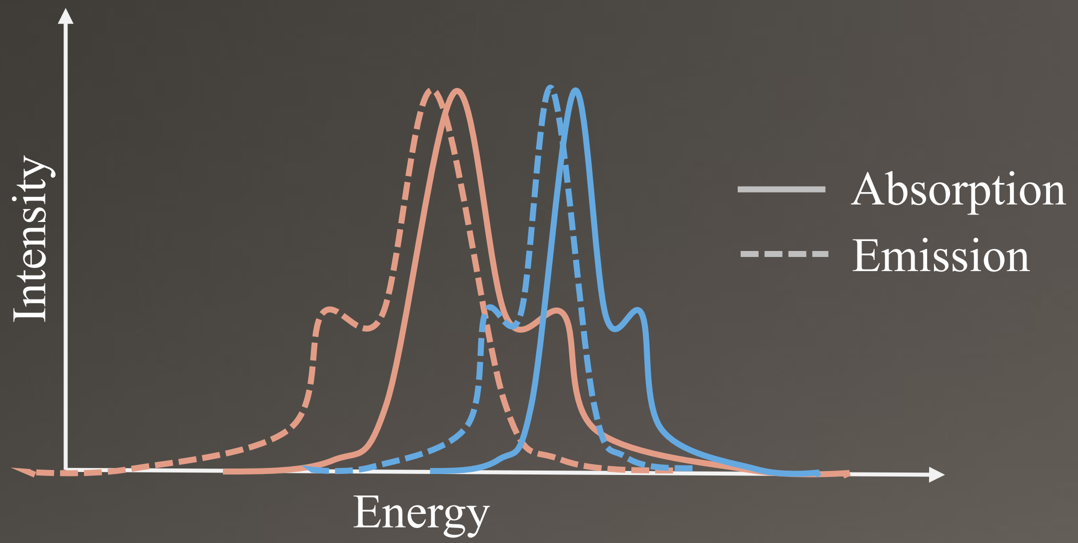
2) By developing the formalism that describes the interaction between dark matter and molecules, we can develop detection strategies capable of *delving deep* and *searching wide* across the dark matter parameter space.

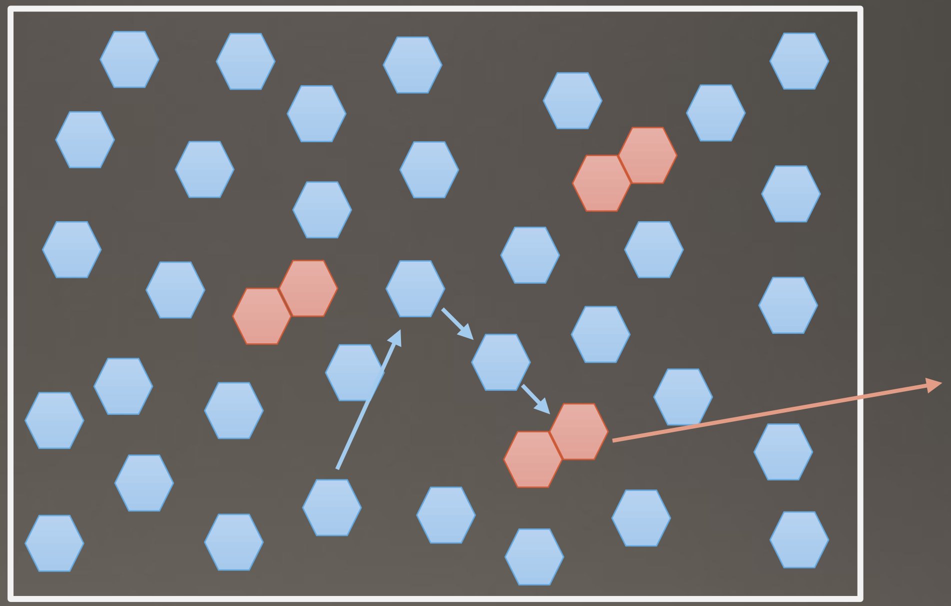
arXiv: 1912.02822

Fluorescence: Binary Scintillators

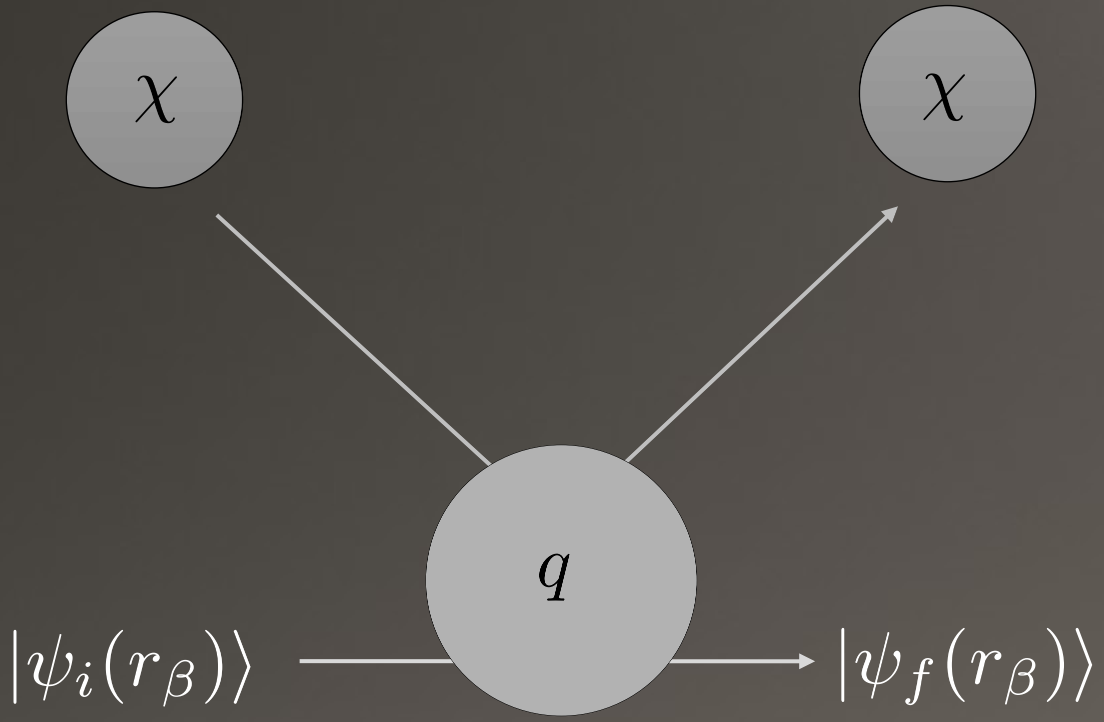
Solvent: Primary target starts the signal

Solute: Dilute fluor gets the signal out of the bulk





Electron Recoil: Charge Signal



Electron scattering

$$\Delta E_r = (m_\chi^2/m_{\mathrm{T}}) \times 10^{-6}$$

$$\Delta E \sim \mathcal{O}(\text{few eV}) \left(\frac{m_\chi}{1\,\text{MeV}}\right)^2$$

What has such transition energies?

- Semiconductor band gaps
- Maybe atomic ionization

Electrons in crystals (exciton generation)

$$|\psi_i\rangle \sim u_v(r)e^{ik'\cdot r} \qquad |\psi_f\rangle \sim u_c(r)e^{ik\cdot r}$$

Electrons in atoms (ionization)

$$|\psi_i\rangle \sim \psi_{\mathrm{STO}}(r_\beta) \qquad |\psi_f\rangle \sim e^{ik\cdot r},\ r \gg a_0$$

Trans-Stilbene

s	Platt Symbol	Symmetry	ΔE [eV]	Configuration amplitudes			
s_1	${}^1B$	B_u	4.240	$d_{7,8} = 0.94,$	$d_{4,11} = -0.24$		
s_2	${}^1G^-$	B_u	4.788	$d_{7,10} = 0.53,$	$d_{5,8} = 0.53,$	$d_{6,11} = 0.37,$	$d_{4,9} = -0.37$
s_3	${}^1G^-$	A_g	4.800	$d_{7,9} = 0.53,$	$d_{6,8} = 0.53,$	$d_{5,11} = 0.37,$	$d_{4,10} = -0.37$
s_4	${}^1(C,H)^+$	A_g	5.137	$d_{7,11} = 0.41,$	$d_{5,9} = -0.41,$	$d_{6,10} = -0.41,$	$d_{4,8} = -0.59$
s_5	${}^1H^+$	B_u	5.791	$d_{5,10} = 0.54,$	$d_{6,9} = 0.54,$	$d_{7,12} = 0.33,$	$d_{3,8} = 0.33$
s_6	${}^1G^+$	A_g	6.264	$d_{7,9} = 0.68,$	$d_{6,8} = -0.68$		
s_7	${}^1C^-$	A_g	6.013	$d_{7,11} = 0.66,$	$d_{4,8} = 0.54,$		
s_8	${}^1G^+$	B_u	6.439	$d_{7,10} = 0.65,$	$d_{5,8} = -0.65$		

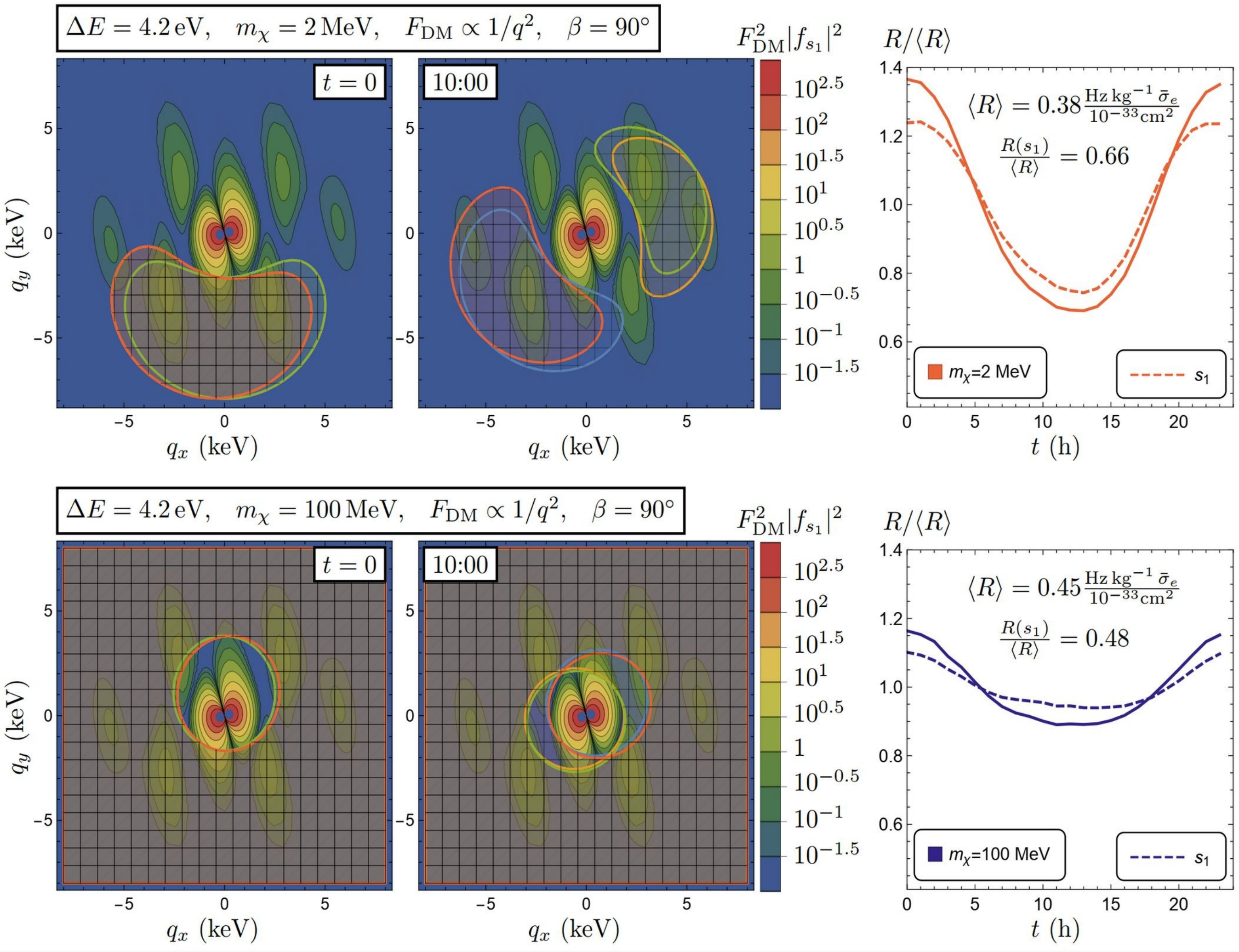
Table 1: The first eight excited states $s_{n=1\ldots8}$, with their energy eigenvalues $\Delta E(s_n)$ with respect to the ground state and coefficients $d_{ij}^{(n)}$ as calculated by Ting and McClure.

$$|s_n\rangle = \sum_{i,j>i} d_{ij}^{(n)} |\psi_i^j\rangle,$$

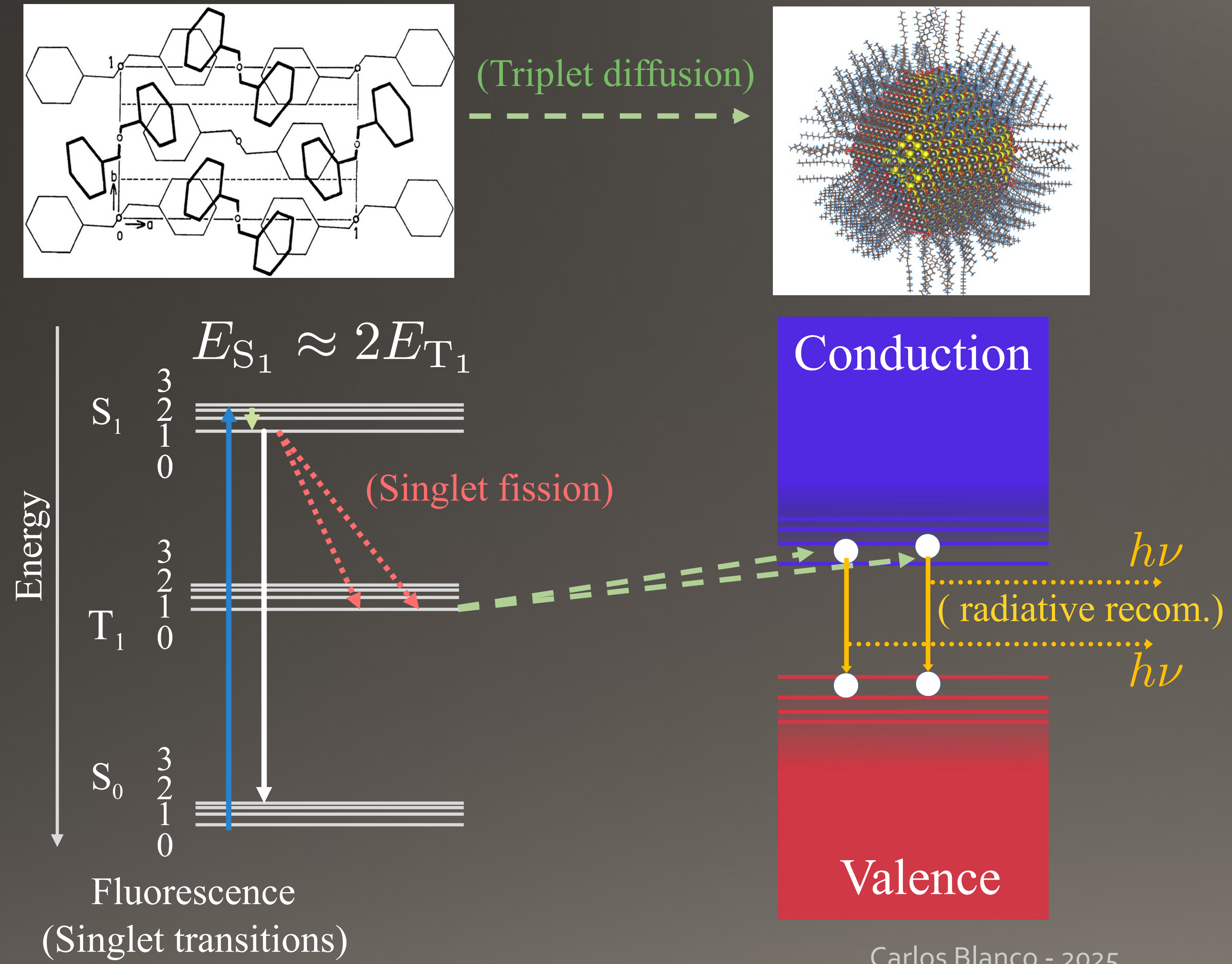
$$\sum_{ij} |d_{ij}^{(n)}|^2 = 1.$$

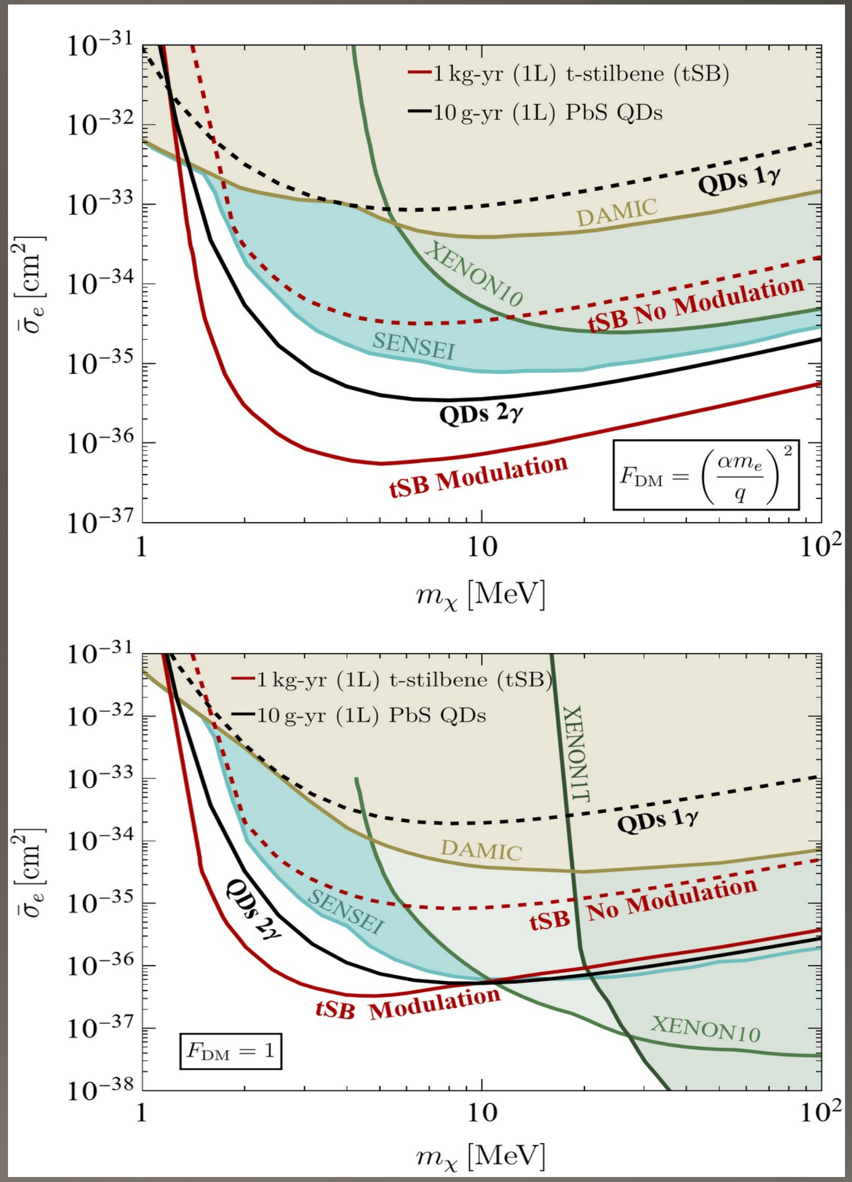
$$\begin{aligned} f_{g\to s_n}(\vec{q}) &= \left\langle \psi_{s_n}(\vec{r}_1 \ldots \vec{r}_{14}) \left| \sum_{m=1}^{14} e^{i\vec{q}\cdot\vec{r}_m} \right| \psi_G(\vec{r}_1 \ldots \vec{r}_{14}) \right\rangle \\ &= \sum_{ij} d_{ij}^{(n)} \left\langle \psi_i^j \left| e^{i\vec{q}\cdot\vec{r}} \right| \psi_G \right\rangle \\ &= \sqrt{2} \sum_{ij} d_{ij}^{(n)} \langle \Psi_j(\vec{r}) | e^{i\vec{q}\cdot\vec{r}} | \Psi_i(\vec{r}) \rangle. \end{aligned}$$

Daily Modulation: Light Mediator

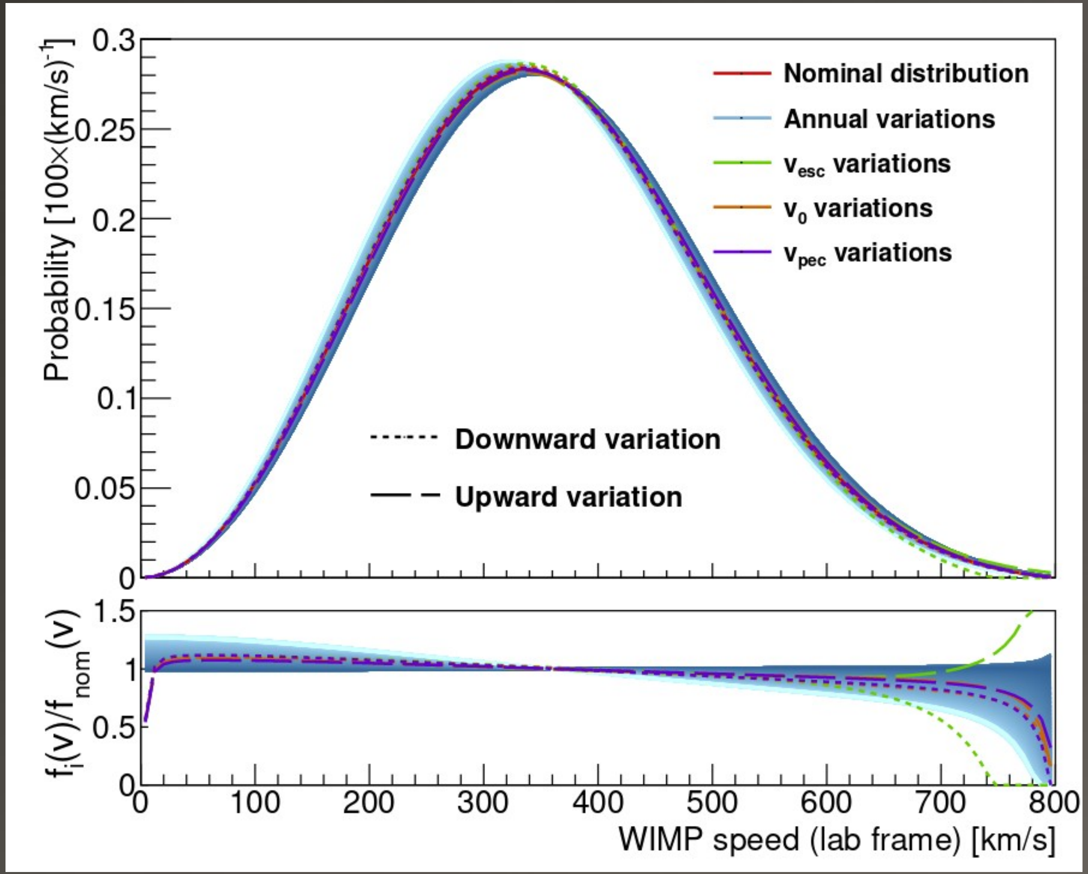


Same as previous figures (top) for a light mediator DM form factor $F_{\mathrm{DM}} = (\alpha m_e/q)^2$. Here, the contour plots show $F_{\mathrm{DM}}^2|f(s_1)|^2$ which appears in the rate integrand; the scattering is dominated by the smallest kinematically-allowed q. **Top:** Molecular form factors with $q_z = 0$ and rate modulations for $m_\chi = 2$ MeV. **Bottom:** Molecular form factors with $q_z = 0$ and rate modulations for $m_\chi = 100$ MeV.

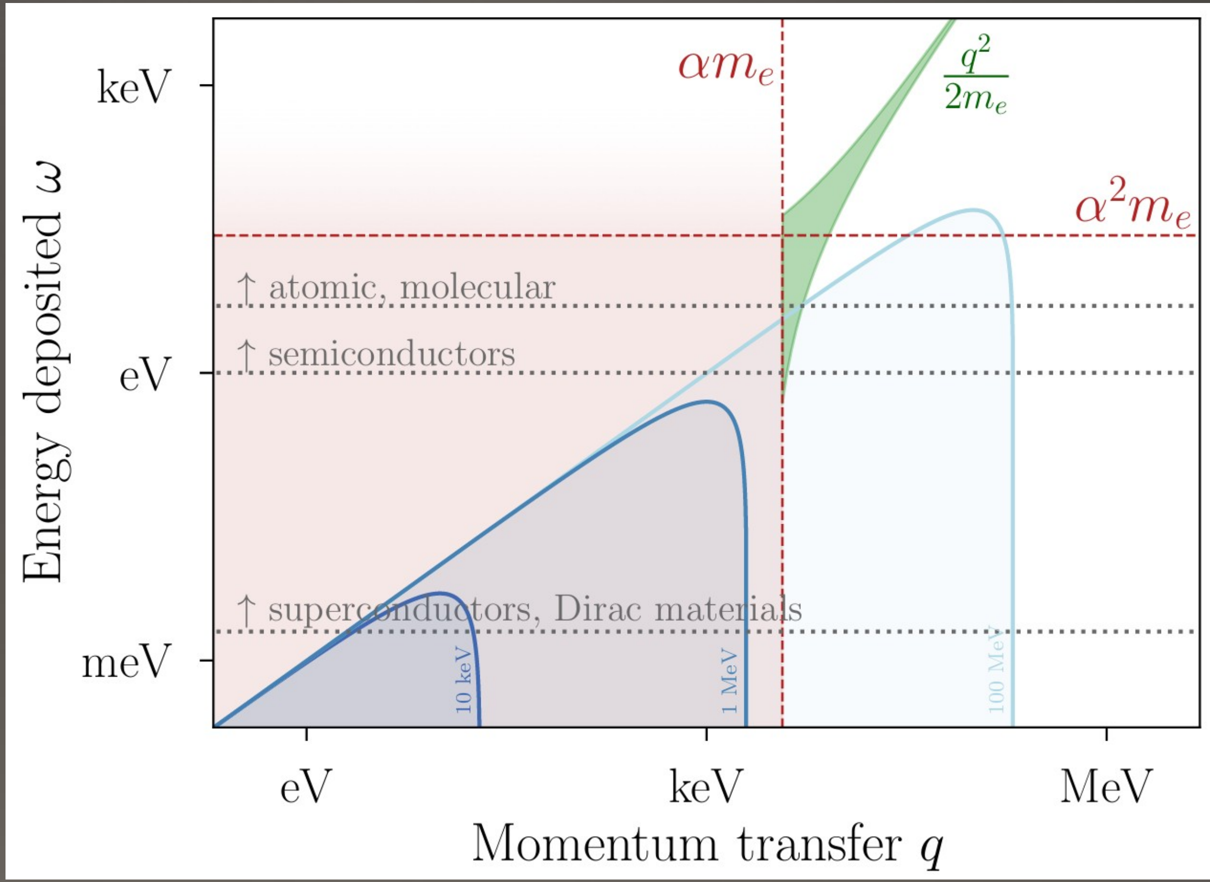

(Triplet diffusion)
$E_{S_1} \approx 2E_{T_1}$
Energy
S_1
T_1
S_0
3
2
1
0
(Singlet fission)
Conduction
Valence
$h\nu$
(radiative recom.)
$h\nu$
Fluorescence
(Singlet transitions)


1 kg-yr (1L) t-stilbene (tSB)
10 g-yr (1L) PbS QDs
QDs 1γ
DAMIC
XENON10
tSB No Modulation
SENSEI
QDs 2γ
tSB Modulation
$F_{DM} = \left(\frac{\alpha m_e}{q}\right)^2$
$\bar{\sigma}_e$ [cm^2]
m_χ [MeV]
XENON1T
$F_{DM} = 1$

Local DM Phase Space



Baxter, D., et al. "*Recommended conventions for reporting results from direct dark matter searches*." The European Physical Journal C 81.10 (2021): 1-19.



Lin, Tongyan. "*Sub-GeV dark matter models and direct detection*." SciPost Physics Lecture Notes (2022): 043.