





Scintillating Methods to Detect Dark Matter from Nano to Astro Scales



CARLOS BLANCO

MW Dark Matter Halo



Local dark matter density $ho_{\rm DM} = 0.4 \, {\rm GeV/cm}^3$

Local dark matter velocity $\langle v_{\rm DM} \rangle \approx 300 \, {\rm km/s}$

Lighter mass \rightarrow more particles $n_{\rm DM} = \rho_{\rm DM}/m_{\rm DM}$

More particles \rightarrow larger fluxes $\phi_{\rm DM} \sim v_{\rm DM} n_{\rm DM}$

Dark Matter in The Lab







Energy Budget $E \sim v^2 m_{\chi} \approx \mathcal{O}(\text{keV}) \left(\frac{m_{\chi}}{1 \text{ GeV}}\right)$ Momentum Budget $q \sim v m_{\chi} \approx \mathcal{O}(\text{MeV}) \left(\frac{m_{\chi}}{1 \text{ GeV}}\right)$

Direct Detection



Need to delve deep and search wide Minimize threshold energies ΔE_r Maximize signal efficiencies ξ Minimize backgrounds $R_{\rm bkg}$

 $m_{\rm DM} \, [{\rm GeV}]$

Mass Range & Detection Methods



WIMPs: The Miracle



Griest et. al: Phys Rep. 1996

Present-day abundance set by *freeze-out*

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

The appeal Weak-scale masses and cross sections
$$\langle \sigma v
angle_{
m FO} = 3 imes 10^{-26} {
m cm}^3 {
m /s}$$

$$\approx \frac{\alpha_{\rm W}^2}{(100\,{\rm GeV})^2}$$

WIMPs: The Resounding Success



Liquid Noble Gases Effective when $m_{\chi} > (O)(\text{GeV})$



Akerib, D. S., et al. "*Snowmass2021 Cosmic Frontier Dark Matter Direct Detection to the Neutrino Fog.*" arXiv:2203.08084 (2022).

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On to Non-WIMPy Dark Matter



Interaction Rate

Rate *spectrum* (events in detector)

$$\Gamma \sim \int \frac{d^3 \vec{q}}{q} \eta(v) |F_{\rm DM}(q)|^2 |f_{i \to f}(q)|^2$$

Mean inverse velocity (Astrophysics) Transition form factor (Condensed matter / Chemistry) $\eta(v)$ $f_{i \to f}(q) = \langle \tilde{\Psi}_f(k+q) | \tilde{\Psi}_i(k) \rangle$

> Dark matter form factor (Particle physics) $F_{\rm DM}(q) \propto \begin{cases} 1 & , \text{ Contact interaction} \\ \left(\frac{1}{q}\right)^2, \text{ Long-range interaction} \end{cases}$

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Sub-GeV Direct Detection



- R: Nuclei
- r: Electrons
- χ : Dark matter

Imparted momentum in scattering $q \approx \mathcal{O}(\text{keV}) \left(\frac{m_{\chi}}{1 \text{ MeV}}\right)$

Energy Budget $\Delta E \approx \mathcal{O}(\text{eV}) \left(\frac{\text{m}_{\chi}}{1 \text{ MeV}}\right)$

Energy equals momentum in *absorption*

$$\Delta E = q = \left(\frac{m_{\chi}}{1 \text{eV}}\right)$$

Electron Recoil --> Photon Signal



Molecules and Atoms (Quantized) $|\psi_i\rangle \sim \psi_{lcao}(r) \quad |\psi\rangle^* \sim \psi^*_{lcao}(r)$ HOMO \rightarrow LUMO

Crystals (Continuous) $|\psi_i\rangle \sim u_v(r)e^{ik\cdot r} |\psi\rangle^* \sim u_c(r)e^{ik\cdot r}$

Valence \rightarrow Conduction

Chromophores Electronic excitation \rightarrow Unstable state \rightarrow Photon emission.

Carbon: a Natural Candidate



spread out

Organic Chromophores

 σ electrons form *rigid* network



 π electrons *delocalized* on ring

excite to produce photoemission

Delocalized $\overline{\pi}$ networks in molecules are natural DM detector candidates due to kinematic matching $q_e \approx 1/a_0 \sim \mathcal{O}(\text{keV}) \qquad q \sim m_\chi v_\chi \approx \mathcal{O}(\text{keV}) \left(\frac{m_\chi}{1 \text{ MeV}}\right)$

Fluorescence with DM

fluorescence spectra 1.0 Absorption/emission Probability 0.8 ntensity (Normalized) Absorption 0.6 0.4 0.2 Ja. 0.0 250 300 350 400 450 500 λ (nm)

Chromophore:



Decreasing energy $(E) \rightarrow$

Probability for the photon to free stream $\Phi_{\rm FB} \sim (1 - a_{xx})$ e.g. molecular crystals: $\Phi_{\rm FB} \approx 65\%$

Pilot Experiment

Problem

Describe the interaction between DM (BSM) with molecules (chemistry)

Many-body ground state is easy. $\Psi_G = |\psi_2 \overline{\psi_2} \psi_1 \overline{\psi_1} \psi_{1\prime} \overline{\psi_{1\prime}}|$

How to compute the excited states?

The standards in the field:

FCI Hartree-Fock or Density Functional Theory

Molecular scintillators



First Experimental Setup



1L of EJ-301

Ancient lead & Vetos

Surface run at UChicago

[CB, Collar, Kahn, Lillard: 1912.02822]

Results: EJ-301

(Contact interaction)

(Long-range interaction)



Theory \rightarrow Experiment \rightarrow Results in about 6 months

The Field in Context



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





Experimental results

Zero-background projection

Outlook and Potential Reach





Experimental results

Zero-background projection

Outlook and Potential Reach

[CB, Collar, Kahn, Lillard: 1912.02822]



The obstacle **Backgrounds**

6 orders of magnitude of potential reach awaiting

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

Fluorescence with DM Works



Next Step Minimize background Option 1 Reduce background in the excitation.

Molecular crystals Anisotropic excitation → Time-varying DM signal

Option 2 Reduce background in the emission.

Quantum dots Multiple excitons \rightarrow Time-coincident DM signal

Directional Detection



Effective dark matter "wind" from $ec{\hat{v}_\oplus}(t)$



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

Trans-Stilbene







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

Delocalized and planar network of double bonds

Molecular planes oriented in crystal lattice

Large optical-quality crystals

Daily Modulation

(Contact interaction)



(Long-range interaction)

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

Improved Sensitivity & Reach



[CB, Kahn, Lillard, McDermott: 2103.08601]

Improvement of *two orders of magnitude* & potential for discovery

of this room

Nuclear Recoil --> Photon Signal



Next step in *complexity*: Nuclear recoil \rightarrow ??? \rightarrow Radiative emission

Ionizing Atoms Through Nuclear Recoil:



 $||\psi_i\rangle \sim e^{i\frac{m_e}{M_N}\vec{q}\cdot\vec{r}}\psi_{\rm AO}(r_\beta)$

Initial nuclear recoil

Nucleus moves faster than electrons

Electronic transition to ionized state

The Migdal effect Extend the sensitivity of detectors to lower masses. e.g. Xenon

$$f_{i \to f} \approx \frac{m_e}{M_N} \vec{q} \cdot \langle \vec{r} \rangle_{i \to f}$$

Xe

Kinematic *penalty*

The Molecular Migdal Effect(s)

Center of mass recoil (CMR)

Cause by center of mass motion



Analogous to atomic Migdal effect

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

Moving whole molecule \rightarrow BIG penalty

Blanco '22: 2208.09002



Non-adiabatic coupling (NAC)

Caused by relative motion



Effect beyond Born-Oppenheimer

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

Crumpling molecule \rightarrow small Penalty

The Molecular Migdal Effect(s)



Si rate is calculated using the *CMR*-equivalent Migdal effect. Is there an *NAC*-equivalent in Si?

Center of mass recoil
 Subdominant at all masses.

Non-adiabatic coupling
 Favorable kinematic factor.

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

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

Directional Molecular Migdal Effect



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

Molecular alignment \rightarrow Directional electronic excitation \rightarrow Directional molecular Migdal effect(s)

Experimental Evidence for NAC



Claim NAC is visible in existing data

Photon absorption probability $\langle \Psi_f | \vec{r} | \Psi_i \rangle = 0$ e.g. toluene dipole transition

Classically forbidden by symmetry No dipole moment \rightarrow Classically forbidden UV absorption

Experimental Evidence for NAC



NAC form factor How electrons respond to nuclear deformation. $f_{e,NAC} \sim \langle \psi_f | \nabla_R | \psi_i(r) \rangle$

Photon absorption probability $\epsilon \sim \left< \Psi_f \right| \vec{r} \left| \Psi_i \right> = 0 \quad \text{e.g. toluene}$ dipole transition

NAC-induced UV absorption $\epsilon \sim \langle \psi_f(r; R) | \vec{r} | \psi_i(r; R) \rangle |_{R \neq R_0} \neq 0$

However

Nuclear deformation \rightarrow Non-adiabatic dipole

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

Finding Optimal Targets



Find molecules with max NAC Large NAC \rightarrow Large DM interactions

Molecular symmetries \rightarrow Forbidden transitions

Measured with vibrational substructure \rightarrow Evidence of NAC

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 predictionMolecular GenerationEnergies & Matrix elementsSample latent space \rightarrow new molecules



[[]CB, Cook*, Smirnov: 2404.xxxx]

Using exhaustive database (< 9 atoms) Characterize neural nets → Possible to learn from small subsample

Next: Large but sparse dataset up to 10s of atoms Scale architecture → Generate candidate molecule *shortlist*

Future Experiment

Experimental Deployment

The DIANA experiment

Daily modulation from an Intrinsically ANisotropic Array



Fig: Dan Baxter Collab: Uchicago, FermiLab, Northwestern, MIT, and UIUC

Fluorescence with DM Works



Focus Minimize background

Option 1 Reduce background in the excitation.

Molecular crystals Anisotropic excitation → Time-varying DM signal

Option 2 Reduce background in the emission.

Quantum dots Multiple excitons → Time-coincident DM signal

Nanocrystals: Quantum Dots

Quantum confinement affects long-wavelength physics

Quantum confinement



Quantum Dots: Coincident Signal



Absorption \rightarrow Very *energetic* exciton

Multi-exciton generation \rightarrow several excitons If energy is greater than twice the bandgap

Radiative recombination \rightarrow coincident photons Band-edge excitons produce light



 $\sim 1L$ (10 g QDs)

PbS Quantum Dots

(Contact interaction)

(Long-range interaction)



Background-free signal @ this scale

[CB, Essig, Fernandez-Serra, Ramani, Slone: 2208.05967]

With realistic dark rate for photodetectors.

Future Experiment

Experimental Deployment

DarkDot & The QUADRA experiment



Figure 2: (Left:) Inner structure of the current SUXESs facility. (Right:) A diagram of a single module for the proposed detector.

Fig: Joern Mahlstedt Collab: Stockholm U, MIT, & Stony Brook

Deployment



Hybrid Detectors





Molecular excitation (Singlet transitions)



Conduction

Valence





46

 10^{2}

ODs 17

 αm_e

 10^{2}



(Singlet transitions)



Beyond direct detection



Astrophysical volume of molecules

Same theoretical techniques \rightarrow Predict rates in *astrophysical* objects

Beyond direct detection





Cold molecular cloud

Same theoretical techniques \rightarrow Predict rates in *astrophysical* objects

Dark matter in Molecular Clouds



Dense cold molecular clouds are almost entirely opaque. $n_{\rm H_2} \sim O(10^2) {\rm cm}^{-3}$

Ionization from CR produces ionization fraction: ζ^{H_2} CR + H₂ \rightarrow CR + H₂⁺ + e⁻

Well measured through astro-spectroscopy of tracer molecules (line intensity measurements)

DM scattering \rightarrow Add ionized SM particles

$$\zeta_i^{\mathrm{H}_2} = 2\pi \int \frac{dN_i}{dE} (E) \sigma_i(E) dE$$

Dark matter in Molecular Clouds

Constraints on DM w/ ultra-light mediator



Otherwise open parameter space

Strongly-coupled dark matter is stopped before reaching experiments.

*Uncertainty from inferred CR ionization rate due to gas depletion onto grain surfaces.

Dark matter in Molecular Clouds





The Molecular Migdal effect(s) *in space*

Conclusions

- Successful campaign for WIMPs \rightarrow Now we must look beyond.
- By developing the formalism that describes the interaction between dark matter and molecules or nano-materials, we can propose detection strategies capable of *delving deep* and *searching wide* across the dark matter parameter space.
- This remains one of the few ways to probe high-energy physics at the *bench-top* scale.
- Stay tuned for hybrid methods giving multiplicative improvements to sensitivity.

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The DFT Problem



[Peterson, Watkins, Lane, Zhu '23: 2310.00147]

DFT is a ground-state theory

The systematic uncertainty from *out-of-the-box* DFT can be very large.

The Alternative Self-consistent molecular orbital theory

Excited states

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

Many Methods in One Program



PbS QDs



[CB, Essig, Fernandez-Serra, Ramani, Slone: 2208.05967]

In the case of eV-scale dark photon absorption, we can use existing *data* to predict the sensitivity of QD-based detectors.

Key conclusions of QD analysis

1) The interaction rate in a semiconductor generated by DM is the same if the semiconductor is *monolithic* or *nanoscopically* disperse.

2) In a QD-based experiment, the readout is independent of the target.

3) The signal can be tuned through control of quantum confinement.

Dark photon kinetic coupling

QDs – Cheap, tunable and scalable

QDot™ PbS Quantum Dots



\$399.00 - \$2,500.00



QDot[™] PbS (Lead Sulfide) Quantum Dots, oleic acid capped, absorb the light from high energy photons up to near-infrared (NIR) range and re-emit in NIR range. The absorption/emission profiles can be tuned from 800 to 2200 nm, simply by changing nanoparticle sizes from 2 to 12 nm. This material has outstanding light absorption and photoelectrical properties, and is utilised for for near-infrared (NIR) or short-wave infrared (SWIR) image sensors. For specific application convenience, two lines of QDs are available:

- With specific absorption peak in 800 2200 nm range
- With specific emission peak in 900 1600 nm range Read more





Φ dependence on the solution concentration for 3nm and 3.3nm PbS QDs in toluene.

 $n_e \sim 10^{20} \text{ cm}^{-3} = 10^{23} \text{ L}^{-1}$

Strongly Confining Quantum Dots

Semiconducting nano-spheres



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

 $E_{kin} \sim \frac{1}{r^2}$ $E_{coulomb} \sim \frac{1}{r}$

60

Fluorescence: Binary Scintillators

- Solvent: Primary target starts the signal
- Solute: Dilute fluor gets the signal out of the bulk



Energy



PbS QDs: Improvements



"Blind" mode

"Active" mode

PbS QDs: Improvements



PbS QDs: Optimism for comparison



Blanco '22: 2208.05967

DM-Electron Scattering (no background 1-photon signal)

Daily Modulation: Small Mass



Molecular form factors and modulating rates for DM masses near threshold, $m_{\chi} = 2$ MeV. In the contour plots, the gridded shaded regions indicate the kinematically accessible momentum transfers \vec{q} for the four molecules that comprise the unit cell of the crystal, shown at t = 0 and t = 10 h. Here, \vec{q} is given in the molecular basis, $q_x = \vec{q} \cdot \hat{\vec{L}}, q_y = \vec{q} \cdot \hat{\vec{M}}$, and the kinematically accessible region is defined by $v_-(\vec{q}) < v_{\rm esc}$.

[CB, Kahn, Lillard, McDermott: 2103.08601]

Daily Modulation: Large Mass



Same as previous figure but for large DM masses, $m_{\chi} = 100$ MeV. Only the nearly-spherical region near $q \sim 0$ with inner boundary $q_{\min} \simeq$ 1.6 keV is kinematically forbidden. As a result, the daily modulation amplitude is smaller, driven by the anisotropy of the inner secondary peaks and the tails of the primary peaks.

[CB, Kahn, Lillard, McDermott: 2103.08601]

Electron Recoil: Charge Signal



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

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

What has such transition energies?

- Semiconductor band gaps
- Maybe atomic ionization

Electrons in crystals (exciton generation)

2

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

Electrons in atoms (ionization)

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

Semiconductor CCDs



Essig R., et al. "Snowmass2021 Cosmic Frontier The landscape of low-threshold dark matter direct detection in the next decade" arXiv:2203.08297 (2022).

Nuclear Recoil: Phonon Signal



Calorimeters





Essig R., et al. "Snowmass2021 Cosmic Frontier The landscape of low-threshold dark matter direct detection in the next decade" arXiv:2203.08297 (2022).

Trans-Stilbene

s	Platt Symbol	Symmetry	$\Delta E \left[\mathrm{eV} \right]$	Configuration amplitudes			
s_1	^{1}B	B_u	4.240	$d_{7,8} = 0.94,$	$d_{4,11} = -0.24$		
s_2	${}^{1}G^{-}$	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	${}^{1}G^{-}$	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	$^{1}H^{+}$	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	${}^{1}G^{+}$	A_g	6.264	$d_{7,9} = 0.68,$	$d_{6,8} = -0.68$		
s_7	${}^{1}C^{-}$	A_g	6.013	$d_{7,11} = 0.66,$	$d_{4,8} = 0.54,$		
s_8	$^{1}G^{+}$	$\overline{B_{u}}$	6.439	$d_{7,10} = 0.65,$	$d_{5,8} = -0.65$		

Table 1: The first eight excited states $s_{n=1...8}$, 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.

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

$$f_{g \to s_n}(\vec{q}) = \left\langle \psi_{s_n}(\vec{r}_1 \dots \vec{r}_{14}) \left| \sum_{m=1} e^{i\vec{q} \cdot \vec{r}_m} \right| \psi_G(\vec{r}_1 \dots \vec{r}_{14}) \right\rangle$$

$$= \sum_{ij} d_{ij}^{(n)} \left\langle \psi_i^j \left| e^{i\vec{q} \cdot \vec{r}} \right| \psi_G \right\rangle$$

$$= \sqrt{2} \sum_{ij} d_{ij}^{(n)} \left\langle \Psi_j(\vec{r}) \right| e^{i\vec{q} \cdot \vec{r}} |\Psi_i(\vec{r})\rangle.$$

$$T_1$$

Daily Modulation: Light Mediator



Same as previous figures (top) for a light mediator DM form factor $F_{\rm DM} =$ $(\alpha m_e/q)^2$. Here, the contour plots show $F_{\rm DM}^2 |f(s_1)|^2$ which appears in the rate integrand; the scattering is dominated by the smallest kinematicallyallowed 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.
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.