

Simplified spin dependence in dark matter direct detection

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The interactions of dark matter with Standard Model particles can be systematically studied in the language of effective field theories. We investigate dark matter interactions with Standard Model particles, including spin-dependent interactions, for direct detection experiments and demonstrate that, although the scattering rate generally depends on multiple types of material response functions, certain linear combinations of these material response functions vanish if the initial and final electronic states share the same Hamiltonian. We also find that several other response functions vanish in parity-symmetric materials, making these systems as simple as isotropic detectors in some respects. Finally, we present the scattering rate for an anisotropic, possibly chiral detector, for generic dark matter-electron spin interactions. These relations reduce the number of independent response functions needed, thereby simplifying the computational complexity for a broad class of dark matter models. Our results provide a complete and efficient toolkit for analyzing electron recoil signals in diverse detector materials.

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I. INTRODUCTION

Direct detection experiments provide an opportunity to study the microscopic interactions between dark matter (DM) and Standard Model (SM) particles by directly probing the local DM population in our galaxy. By measuring energy deposits from rare scattering events, these experiments provide powerful constraints on a wide range of DM models that are independent of the DM production mechanism and cosmological distribution. In this paper, we combine the complete nonrelativistic effective theory for DM-SM interactions with a fully three-dimensional model of the detector medium, so as to facilitate *ab initio* calculations of the generic spin-dependent DM scattering rate for the next generations of anisotropic direct detection experiments.

Traditionally, direct detection experiments have primarily focused on DM interactions with nuclei in the target material.

This approach is particularly well suited for probing weakly interacting massive particles (WIMPs), and over the past four decades, experiments have achieved remarkable sensitivities [1–5]. However, despite these advances, no unambiguous DM signals have been observed. In response, DM-electron scattering has emerged as a promising strategy to explore alternative DM candidates, especially those with masses below that of the nucleon (sub-GeV DM) [6,7].

The DM-electron scattering rate depends on the underlying DM model as well as the structure and SM properties of the detector medium. A broadly model-independent framework to describe this rate is provided by nonrelativistic effective field theory (EFT), which classifies interactions through Lagrangian operators that are invariant under rotations and Galilean velocity boosts. Previous studies have cataloged complete sets of invariant operators for DM-electron [8–11] interactions, building on similar efforts for DM-nucleus interactions [12–14], including inelastic DM [15]. In this EFT formalism, the Wilson coefficients of the operators provide a complete accounting of the relevant interactions between nonrelativistic DM and the detector. This formalism is sufficiently precise for DM in the galactic halo, where relativistic corrections on the order of $(v/c)^2 \sim 10^{-6}$ can be safely neglected.

A key ingredient in computing the DM-electron scattering rate is a set of material-dependent response functions (or form factors), which encode the likelihood that momentum,

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energy, and possibly spin transferred from DM to SM particles will excite the target system into a final observable state. Importantly, these response functions depend solely on the SM dynamics, namely the initial and final state wave functions of the electrons, and can therefore be calculated independently of the dark sector.

Initial works on DM–electron scattering focused on spin-independent scattering in an isotropic material, which consists of a single EFT operator and depends on a single, scalar response function [6,16–18]. However, these works were limited to a subset of possible DM models, excluding possibilities such as spin-dependent interactions or DM models with a magnetic or anapole moment. References [19–27] extend the discussion to the direct detection of spin-dependent sub-GeV DM. Building on the work of Refs. [28,29], which demonstrated a connection between the response function and the material dielectric function, Ref. [30] showed that some spin-dependent DM interactions couple to the spin density function, another SM observable. In the context of linear response theory, Refs. [31,32] expanded this connection between response functions to accommodate additional spin-dependent EFT operators. References [8,33] generalized the calculation of DM–electron scattering to a broader set of EFT operators and identified four additional response functions, under the simplifying assumption that the detector materials are isotropic, i.e., spherically symmetric in their response. References [10,11] expanded on this work to find that, with a redefinition of the material response functions, the squared matrix element can be organized into five terms that are a product of the DM response functions, which depend only on the Wilson coefficients of the EFT operators, and a linear combination of the material response functions. For spherically symmetric atomic targets, two of these response functions vanish.

Realistic materials, including those used or proposed for direct detection experiments, can exhibit anisotropy, and in these systems the response functions of Refs. [8,10,11] are insufficient to predict the DM scattering rate. Anisotropy is a desirable detector property: if the DM scattering rate depends on the detector orientation, then an experiment can use the expected sidereal daily modulation to distinguish a possible DM signal from the various SM backgrounds [26,34–48]. In this work, we extend the nonrelativistic formalism to accommodate spin-dependent interactions in *ab initio* calculations for anisotropic detector targets. We find that, in addition to the response functions associated with isotropic detectors, the scattering rate receives a contribution from two additional vectorial response functions, consistent with Ref. [33], raising the number of response functions to seven. Following the organizing principles in Refs. [10,11], we expand on previous work to present for the first time, to our knowledge, the generic nonrelativistic DM–electron scattering rate in a generically anisotropic detector.

Beyond generalizing the formalism, we identify a significant simplification for a broad class of materials.

In systems where the initial and final state wave functions of the SM target particle are eigenstates of the same Hamiltonian, such as in atomic systems or single-particle excitations in crystals, we derive new relationships among the response functions. Specifically, we show that certain spin-dependent contributions can be expressed in terms of the scalar (spin-independent) form factor, up to boundary terms that typically vanish. These results reduce the number of independent response functions required to compute the scattering rate, thereby easing the computational burden for a wide range of DM interaction models and detector materials.

Exceptions to this simplification are found in processes like nonadiabatic Migdal scattering in molecules [41] and crystals [49], which are driven by corrections to the Born-Oppenheimer approximation. Here, the target’s initial and final states are governed by different Hamiltonians due to dynamic coupling between nuclear and electronic degrees of freedom. Collective excitations, such as phonons or plasmons, can also violate the assumptions underpinning our simplification (see, e.g., [31]), whenever electron-electron correlations become important. Nevertheless, for many cases of practical interest, especially atomic DM–electron scattering and single-particle excitations in molecules and crystals, the condition that the initial and final states are governed by the same Hamiltonian, $\hat{H}_i = \hat{H}_f$, provides a good approximation.

We begin in Sec. II by extending previous derivations of the DM–electron scattering rate to anisotropic media, identifying the full set of response functions that contribute and clarifying how they appear in the rate. We conclude this section with a modestly simplified version of the scattering amplitude, for generic DM–SM spin couplings, in a completely anisotropic detector. The expression includes five linear combinations of SM response functions, rather than the seven that might have been expected from [33]. Next, Sec. III demonstrates that two of these combinations vanish in parity-symmetric materials, even if the DM–SM particle interaction violates parity. Finally, in Sec. III C, we derive a novel relationship between the spin-dependent and spin-independent form factors, in the case where the initial and final electronic states are eigenstates of the same single-particle Hamiltonians. We refer to this as “electronic adiabaticity,” and another linear combination of response functions vanishes in the multiparticle systems where it is a good approximation. In the simplest cases, where the detector material is parity-symmetric, e^- -adiabatic, and also spherically symmetric, we show that even the most generic DM–SM spin interactions depend only on the “spin-independent” form factor. Section IV provides a few concrete examples of DM models, with a spin-0 or spin- $\frac{1}{2}$ DM particle coupled to electrons via spin-0 and/or spin-1 mediators. We conclude with an inelastic scalar DM model, as a simple example where all of the DM response functions are nonzero.

II. FORMALISM FOR DM-SM SCATTERING

In this section, we review the formalism for calculating the DM scattering rate as well as the definitions of the material response functions. Section II B concludes with our expression for generically spin-dependent DM-SM interactions in an anisotropic detector medium with unpolarized spins.

A. Scattering rate and matrix element

The total DM-SM scattering rate in a direct detection experiment is given by

$$R_{1 \rightarrow 2} = \frac{N_{\text{cell}} \rho_\chi / m_\chi}{64 \pi^2 m_\chi^2 m_T^2} \int d^3 q d^3 v f_\chi(\vec{v}) \delta \left(E + \frac{q^2}{2m_\chi} - \vec{q} \cdot \vec{v} \right) |\mathcal{M}_{1 \rightarrow 2}(\vec{q}, \vec{v})|^2, \quad (1)$$

where N_{cell} is the number of target cells/molecules/etc. in the detector; m_χ and ρ_χ are the DM particle mass and local mass density; $f_\chi(\vec{v})$ is the DM velocity distribution; $E = E_2 - E_1$ and $\vec{q}$ are the energy and momentum transferred from DM to the SM target particle, which has mass m_T ; and $\mathcal{M}_{1 \rightarrow 2}$ is the transition amplitude. For single-particle scattering,

$$\mathcal{M}_{1 \rightarrow 2}(\vec{q}, \vec{v}) = \int \frac{d^3 k}{(2\pi)^3} \tilde{\psi}_2^*(\vec{k} + \vec{q}) \mathcal{M}_{\text{free}}(\vec{q}, \vec{v}, \vec{k}) \tilde{\psi}_1(\vec{k}). \quad (2)$$

Here ψ_1 and ψ_2 are the initial and final states of the scattered SM particle, with $\tilde{\psi}_i(\vec{k}) = \langle \vec{k} | \psi_i \rangle$ the momentum space wave functions, and $\mathcal{M}_{\text{free}}(\vec{q}, \vec{v}, \vec{k})$ is the amplitude for free particle scattering [16]. To transform from momentum space to position space, we use the normalization convention

$$\psi(\vec{x}) = \int \frac{d^3 k}{(2\pi)^3} e^{i\vec{k} \cdot \vec{x}} \tilde{\psi}(\vec{k}), \quad \tilde{\psi}(\vec{k}) = \int d^3 x e^{-i\vec{k} \cdot \vec{x}} \psi(\vec{x}). \quad (3)$$

Equation (2) assumes that the initial and final states of the SM target reside in the same single-particle Hilbert space. It can also be generalized to a wider class of multiparticle systems, if the (multiparticle) final state Ψ_2 can be written as a linear combination of pairs of one-particle creation and annihilation operators acting on the (multiparticle) initial state Ψ_1 :

$$\Psi_2 \simeq \sum_{ij} T_{ij}^{(1)} c_j^\dagger c_i \Psi_1. \quad (4)$$

Here $T_{ij}^{(1)}$ is the single particle transition density matrix, and it is particularly relevant for correlated electronic systems (see, e.g., [50] for a review). In this notation, the transition amplitude is

$$\mathcal{M}_{1 \rightarrow 2}(\vec{q}, \vec{v}) = T_{ij}^{(1)} \int \frac{d^3 k}{(2\pi)^3} \tilde{\psi}_j^*(\vec{k} + \vec{q}) \mathcal{M}_{\text{free}}(\vec{q}, \vec{v}, \vec{k}) \tilde{\psi}_i(\vec{k}), \quad (5)$$

where $\psi_{i,j}$ are the single particle states $|\psi_i\rangle = c_i^\dagger |0\rangle$, in a basis of states that approaches completeness:

$$\mathbb{1} \simeq \sum_i |\psi_i\rangle \langle \psi_i|. \quad (6)$$

The transition amplitude in Eq. (5) can accurately describe the leading effects from correlations in many relevant multiparticle systems, including electronic transitions in molecules and condensed matter systems [31,39,51]. Single phonon production requires a modified description since the initial state $\Psi_1 = |0\rangle$ is the ground state with no phonons, while the final state $\Psi_2 = \sum_j T_j^{(1)} c_j^\dagger \Psi_1$ contains one phonon. Here, Eq. (4) should be modified so that $c_i \rightarrow \mathbb{1}$ [51,52].

In the special case where the matrix T_{ij} can be written as the outer product of two vectors, Eq. (5) simplifies to Eq. (2). Neither expression is valid for multiply excited final states, where Ψ_2 differs from Ψ_1 by multiple quanta: e.g., $\Psi_2 = \sum_{ij,mn} T_{ij,mn}^{(2)} c_n^\dagger c_m^\dagger c_j c_i \Psi_1$. These multiply excited final states are beyond the scope of this work.

In the nonrelativistic limit, Galilean boost invariance dictates that $\mathcal{M}_{\text{free}}(\vec{q}, \vec{v}, \vec{k})$ should depend only on $\vec{q}$, the momentum transfer, and the elastic transverse relative velocity $\vec{v}_{\text{el}}^\perp$, defined as

$$\vec{v}_{\text{el}}^\perp \equiv \vec{v} - \frac{\vec{q}}{2\mu_{\chi T}} - \frac{\vec{k}}{m_T}, \quad (7)$$

where $\mu_{\chi T}$ is the reduced mass of the DM-SM system. Following the notation of [10], we can expand $\mathcal{M}_{\text{free}}$ to linear order in $\vec{v}_{\text{el}}^\perp$,

$$\mathcal{M}_{\text{free}}(\vec{q}, \vec{v}, \vec{k}) = \mathcal{M}_{\text{free}}(\vec{q}, \vec{v}_{\text{el}}^\perp) \simeq \mathcal{M}_{\text{free}}(\vec{q}, 0) + \vec{v}_{\text{el}}^\perp \cdot (\nabla_{\vec{v}_{\text{el}}^\perp} \mathcal{M}_{\text{free}}(\vec{q}, \vec{v}_{\text{el}}^\perp)|_{\vec{v}_{\text{el}}^\perp=0}) \quad (8)$$

$$\equiv \mathcal{M}_S(\vec{q}) + \vec{v}_{\text{el}}^\perp \cdot \vec{\mathcal{M}}_V(\vec{q}), \quad (9)$$

where $\mathcal{M}_S$ and $\vec{\mathcal{M}}_V \equiv \nabla_{\vec{v}_{\text{el}}^\perp} \mathcal{M}(\vec{q}, \vec{v}_{\text{el}}^\perp)|_{\vec{v}_{\text{el}}^\perp=0}$ each depend only on $\vec{q}$. In this expression, $\vec{v}_{\text{el}}^\perp$ contains all of the $\vec{k}$ dependence in the free particle amplitude $\mathcal{M}_{\text{free}}$. Note that since $\mathcal{M}_S$ and $\mathcal{M}_V$ are independent of the SM particle momentum $\vec{k}$, they can be pulled out of the integral in Eq. (2) and

$$\mathcal{M}_{1 \rightarrow 2}(\vec{q}, \vec{v}) = \mathcal{M}_S(\vec{q}) f_S(\vec{q}) + \vec{\mathcal{M}}_V(\vec{q}) \cdot \vec{f}_V(\vec{q}). \quad (10)$$

All of the $\vec{k}$ -dependent terms are encoded in a scalar form factor, f_S , and a vector form factor, $\vec{f}_V$,

$$f_S(\vec{q}) \equiv \int \frac{d^3k}{(2\pi)^3} \tilde{\psi}_2^*(\vec{k} + \vec{q}) \cdot 1 \cdot \tilde{\psi}_1(\vec{k})$$

$$= \int d^3x e^{i\vec{q}\cdot\vec{x}} \psi_2^*(\vec{x}) \cdot 1 \cdot \psi_1(\vec{x}), \quad (11)$$

$$\vec{f}_V(\vec{q}, \vec{v}) \equiv \int \frac{d^3k}{(2\pi)^3} \tilde{\psi}_2^*(\vec{k} + \vec{q}) \cdot \vec{v}_{\text{el}}^\perp(\vec{k}) \cdot \tilde{\psi}_1(\vec{k})$$

$$= \int d^3x e^{i\vec{q}\cdot\vec{x}} \psi_2^*(\vec{x}) \cdot \left(\vec{v}_0^\perp + \frac{i\nabla}{m_T} \right) \cdot \psi_1(\vec{x}), \quad (12)$$

where for convenience we define

$$\vec{v}_0^\perp \equiv \vec{v} - \frac{\vec{q}}{2\mu_{\chi T}}. \quad (13)$$

The detector response functions, which we define shortly, depend on bilinear products of $(f_S, \vec{f}_V)$ and their complex conjugates.

Specializing to the familiar case of spin-independent SM-DM interactions, the $\mathcal{M}_V$ term vanishes from the rate, leaving

$$R_{1\rightarrow 2} = \frac{N_{\text{cell}} \rho_\chi / m_\chi}{64\pi^2 m_\chi^2 m_T^2} \int d^3q d^3v f_\chi(\vec{v}) \delta\left(E + \frac{q^2}{2m_\chi} - \vec{q} \cdot \vec{v}\right)$$

$$\times |\mathcal{M}_S(\vec{q}, \vec{v})|^2 |f_S(\vec{q})|^2, \quad (14)$$

reproducing the known results [16]. For an anisotropic detector, the scattering rate $R_{1\rightarrow 2}$ is a function of the detector orientation, which can be described by a rotation operator $\mathcal{R} \in SO(3)$ acting on some initial configuration of the detector:

$$R_{1\rightarrow 2}(\mathcal{R}) = \frac{N_{\text{cell}} \rho_\chi}{64\pi^2 m_\chi^3 m_T^2} \int d^3q d^3v f_\chi(\vec{v}) \delta\left(E + \frac{q^2}{2m_\chi} - \vec{q} \cdot \vec{v}\right) \mathcal{R} \cdot |\mathcal{M}_{1\rightarrow 2}(\vec{q}, \vec{v})|^2. \quad (15)$$

Equivalently, the scattering rate can be evaluated in the frame of a fixed detector, with the inverse rotation operator acting on the 3d DM velocity distribution,

$$R_{1\rightarrow 2}(\mathcal{R}) = \frac{N_{\text{cell}} \rho_\chi}{64\pi^2 m_\chi^3 m_T^2} \int d^3q d^3v (\mathcal{R}^{-1} \cdot f_\chi(\vec{v})) \delta\left(E + \frac{q^2}{2m_\chi} - \vec{q} \cdot \vec{v}\right) |\mathcal{M}_{1\rightarrow 2}(\vec{q}, \vec{v})|^2. \quad (16)$$

In both cases, $\mathcal{R} \cdot f$ is an active rotation applied to the function $f(\vec{u})$, i.e.,

$$\mathcal{R} \cdot f(\vec{u}) \equiv f(\mathcal{R}^{-1} \vec{u}). \quad (17)$$

The amplitude $\mathcal{M}$ can be approximated by its non-relativistic limit with negligible loss of accuracy, as long as the SM target particle is also nonrelativistic. In DM-nuclear scattering, the lab-frame speed of the nucleus is typically negligible, but electronic bound states typically have the fastest speeds in the DM-detector system: in terms of the fine-structure constant ($\alpha < 0.01$), $|\vec{k}|/m_e \simeq |\vec{v}_e| \sim \alpha c$. However, even these $\mathcal{O}(v^2) \sim \alpha^2 \sim 10^{-4}$ relativistic corrections for DM-electron scattering are negligible, especially compared to the astrophysical uncertainties on the local DM velocity distribution. For the remainder of the discussion, we will focus on electronic targets but the general framework holds for other targets as well.

B. Expansion in response functions

Now we turn our attention to $\mathcal{M}_S$ and $\vec{\mathcal{M}}_V$, which encode how the DM-SM scattering depends on both the DM model parameters and the SM physics of the detector material. If the detector (or the DM) has polarized spins, then the free particle amplitude may include dot products and cross products between $\vec{f}_V$, $\vec{q}$, and the spin polarization vectors. In a typical direct detection experiment, however, the spins of the electrons and nuclei are unpolarized, as is the DM. Similarly, the usual detector designs might measure the energy transfer $E = E_2 - E_1$, but not the spin state of the scattered SM particle. With this setup, the measured event rate depends only on the spin averaged and summed squared amplitude,

$$\overline{|\mathcal{M}_{1\rightarrow 2}|^2} \equiv \frac{1}{2j_\chi + 1} \frac{1}{2j_{\text{SM}} + 1} \sum_{\text{spins}(i)} \sum_{\text{spins}(f)} |\mathcal{M}_{1\rightarrow 2}|^2, \quad (18)$$

where j_χ is the DM particle spin ($0, \frac{1}{2}, 1$ for scalar, fermion, or vector boson DM), and j_{SM} is the spin of the SM target particle.

The spin-summed $\overline{|\mathcal{M}_{1\rightarrow 2}|^2}$ depends on the scalar products of $\vec{q}$, $\vec{f}_V$, and $\vec{f}_V^*$,

$$\overline{|\mathcal{M}_{1\rightarrow 2}|^2} \supset |f_S|^2, \quad |\vec{f}_V|^2, \quad |\vec{q} \cdot \vec{f}_V|^2,$$

$$\vec{q} \cdot (\vec{f}_V \times \vec{f}_V^*), \quad f_S(\vec{q} \cdot \vec{f}_V^*). \quad (19)$$

For scenarios in which the SM and DM spins are unpolarized and unmeasured, there are no other vectors in the system. Note that any of these terms may receive subleading corrections with additional scalar powers of q^2 , $\vec{q} \cdot \vec{v}_0^\perp$, or $|\vec{v}_0^\perp|^2$; however, because $\vec{f}_V$ is already linear in $\vec{v}_0^\perp$, bilinear combinations such as $\vec{v}_0^\perp \cdot \vec{f}_V$ are already higher order in the $v \ll c$ expansion. Following this logic, Ref. [10] found that $\overline{|\mathcal{M}_{1\rightarrow 2}|^2}$ can be expanded into five terms:

TABLE I. Effective nonrelativistic operators for spin-0 and spin-1/2 DM coupling to an electron following the conventions of Refs. [12,13]. Here we identify the DM and electron spin operators as $\vec{S}_\chi$ and $\vec{S}_e$, respectively along with the identity operators $\mathbb{1}_\chi$ and $\mathbb{1}_e$. Note that we exclude $\mathcal{O}_2$ from the table. As is argued in Ref. [14], $\mathcal{O}_2 = \vec{v}_{\text{el}}^{\perp 2} \mathcal{O}_1$. Since all of the $\vec{v}_{\text{el}}^{\perp}$ dependence can be absorbed into the $\vec{f}_V$ term, including $\mathcal{O}_2$ would thus be redundant.

$\mathcal{O}_1 = \mathbb{1}_\chi \mathbb{1}_e$	$\mathcal{O}_3 = \mathbb{1}_\chi \left(\frac{i\vec{q}}{m_e} \times \vec{v}_{\text{el}}^{\perp} \right) \cdot \vec{S}_e$	$\mathcal{O}_4 = \vec{S}_\chi \cdot \vec{S}_e$
$\mathcal{O}_5 = \vec{S}_\chi \cdot \left(\frac{i\vec{q}}{m_e} \times \vec{v}_{\text{el}}^{\perp} \right) \mathbb{1}_e$	$\mathcal{O}_6 = (\vec{S}_\chi \cdot \frac{\vec{q}}{m_e}) (\frac{\vec{q}}{m_e} \cdot \vec{S}_e)$	$\mathcal{O}_7 = \mathbb{1}_\chi \vec{v}_{\text{el}}^{\perp} \cdot \vec{S}_e$
$\mathcal{O}_8 = \vec{S}_\chi \cdot \vec{v}_{\text{el}}^{\perp} \mathbb{1}_e$	$\mathcal{O}_9 = -\vec{S}_\chi \cdot \left(\frac{i\vec{q}}{m_e} \times \vec{S}_e \right)$	$\mathcal{O}_{10} = \mathbb{1}_\chi \frac{i\vec{q}}{m_e} \cdot \vec{S}_e$
$\mathcal{O}_{11} = \vec{S}_\chi \cdot \frac{i\vec{q}}{m_e} \mathbb{1}_e$	$\mathcal{O}_{12} = -\vec{S}_\chi \cdot (\vec{v}_{\text{el}}^{\perp} \times \vec{S}_e)$	$\mathcal{O}_{13} = (\vec{S}_\chi \cdot \vec{v}_{\text{el}}^{\perp}) (\frac{i\vec{q}}{m_e} \cdot \vec{S}_e)$
$\mathcal{O}_{14} = (\vec{S}_\chi \cdot \frac{i\vec{q}}{m_e}) (\vec{v}_{\text{el}}^{\perp} \cdot \vec{S}_e)$	$\mathcal{O}_{15} = \vec{S}_\chi \cdot \frac{\vec{q}}{m_e} \left[\frac{\vec{q}}{m_e} \cdot (\vec{v}_{\text{el}}^{\perp} \times \vec{S}_e) \right]$	

$$\begin{aligned} |\overline{\mathcal{M}_{1 \rightarrow 2}}|^2 &= a_0 |f_S|^2 + a_1 |\vec{f}_V|^2 + \frac{a_2}{q^2/m_e^2} \left| \frac{\vec{q}}{m_e} \cdot \vec{f}_V \right|^2 \\ &+ ia_3 \frac{\vec{q}}{m_e} \cdot (\vec{f}_V \times \vec{f}_V^*) \\ &+ 2 \text{Im} \left[a_4 f_S \left(\frac{\vec{q}}{m_e} \cdot \vec{f}_V^* \right) \right]. \end{aligned} \quad (20)$$

The DM response functions, $a_{0,\dots,4}(q^2)$, encode all of the relevant microphysical details of the DM particle model through the Wilson coefficients of the nonrelativistic operators (see Table I). We work through a few specific examples in Sec. IV, for fermionic and bosonic DM models. Like f_S , the $a_{0,\dots,4}$ depend on the momentum transfer $\vec{q}$, and $\vec{f}_V$ is the only term that also depends on the DM velocity $\vec{v}$.

The exact forms of the a_i depend on the spin of the DM, but they share some general features. The first four a_i ($a_{0,1,2,3}$) are explicitly real-valued, but a_4 can be complex, in the case of inelastic DM scattering (i.e., the DM initial and final states are different particles, $m_\chi \neq m'_\chi$). Both a_3 and a_4 are generated entirely by interferences between pairs of Wilson coefficients. $a_{0,1,2}$, on the other hand, include terms proportional to $|c_i|^2$ as well as $c_i^* c_j$.

An alternative way of defining the vector material response function is given by [10,33]

$$\begin{aligned} \vec{f}_{1 \rightarrow 2}(\vec{q}) &\equiv \int \frac{d^3k}{(2\pi)^3} \vec{\psi}_2^*(\vec{k} + \vec{q}) \cdot \frac{+i\vec{k}}{m_T} \cdot \vec{\psi}_1(\vec{k}) \\ &= \int d^3x e^{i\vec{q} \cdot \vec{x}} \vec{\psi}_2^*(\vec{x}) \cdot \left(\frac{-i\nabla}{m_T} \right) \cdot \psi_1(\vec{x}), \end{aligned} \quad (21)$$

so that $\vec{v}_0^{\perp}$ contains all of the dependence on the DM velocity $\vec{v}$. Note $\vec{f}_V(\vec{q}, \vec{v}) = \vec{v}_0^{\perp} f_S(\vec{q}) - \vec{f}_{1 \rightarrow 2}(\vec{q})$. From $f_S(\vec{q})$ and $\vec{f}_{1 \rightarrow 2}(\vec{q})$, the scattering rate can be expressed as a linear combination of the DM response functions $a_{0,\dots,4}$ and $\vec{v}$ -independent material response functions $B_i(\vec{q})$ [33]:

$$\begin{aligned} B_1 &= |f_S(\vec{q})|^2, & B_2 &= \frac{\vec{q}}{m_e} \cdot \vec{A}(\vec{q}), \\ B_3 &= |\vec{f}_{1 \rightarrow 2}(\vec{q})|^2, & B_4 &= \left| \frac{\vec{q}}{m_e} \cdot \vec{f}_{1 \rightarrow 2}(\vec{q}) \right|^2, \end{aligned} \quad (22)$$

$$\begin{aligned} B_5 &= i \frac{\vec{q}}{m_e} \cdot (\vec{f}_{1 \rightarrow 2}(\vec{q}) \times \vec{f}_{1 \rightarrow 2}^*(\vec{q})) \\ &= \frac{2\vec{q}}{m_e} \cdot [\text{Re}(\vec{f}_{1 \rightarrow 2}) \times \text{Im}(\vec{f}_{1 \rightarrow 2})], \end{aligned} \quad (23)$$

where $\vec{A}(\vec{q}) \equiv f_S(\vec{q}) \vec{f}_{1 \rightarrow 2}^*(\vec{q})$. Note that B_1 , B_2 , and B_4 are not independent:

$$\begin{aligned} |B_2|^2 &= \left(\frac{\vec{q}}{m_e} \cdot \vec{A}^* \right) \left(\frac{\vec{q}}{m_e} \cdot \vec{A} \right) \\ &= B_1 \left(\frac{\vec{q}}{m_e} \cdot \vec{f}_{1 \rightarrow 2}^* \right) \left(\frac{\vec{q}}{m_e} \cdot \vec{f}_{1 \rightarrow 2} \right) = B_1 B_4, \end{aligned} \quad (24)$$

but the phase of B_2 cannot be extracted from B_1 and B_4 .

For anisotropic detectors, we also need to include the vector response functions:

$$\vec{B}_6 = \vec{A}(\vec{q}), \quad \vec{B}_7 = \frac{\vec{q}}{m_e} \times \vec{A}(\vec{q}). \quad (25)$$

Thus, the most general form of the squared amplitude is given by

$$\begin{aligned} |\overline{\mathcal{M}_{1 \rightarrow 2}}|^2 &= a_0 B_1 + a_1 \left[|\vec{v}_0^{\perp}|^2 B_1 + \frac{m_e}{\mu_{\chi e}} \text{Re} B_2 + B_3 - 2 \text{Re}(\vec{v} \cdot \vec{B}_6) \right] \\ &+ a_2 \frac{m_e^2}{q^2} \left[\left(\frac{\vec{q} \cdot \vec{v}_0^{\perp}}{m_e} \right)^2 B_1 - 2 \left(\frac{\vec{q} \cdot \vec{v}_0^{\perp}}{m_e} \right) \text{Re} B_2 + B_4 \right] \\ &+ a_3 [B_5 - 2 \text{Im}(\vec{v} \cdot \vec{B}_7)] - 2 \text{Re}(a_4) \text{Im} B_2 \\ &+ 2 \text{Im}(a_4) \left[\left(\frac{\vec{q} \cdot \vec{v}_0^{\perp}}{m_e} \right) B_1 - \text{Re} B_2 \right]. \end{aligned} \quad (26)$$

For models of elastic DM, the last line is identically zero as $\text{Im}(a_4) = 0$. Since $\vec{q} \cdot \vec{B}_7 = 0$, the vectorial response functions only appear as the combination $\vec{v} \cdot \vec{B}_{6,7}$ in the squared amplitude.¹ Reference [11] provides the values of $a_{0,\dots,4}(c_i)$ as functions of the Wilson coefficients c_i in their Table 4.

C. Connecting DM microphysics to the detector response

Equation (26) factorizes the SM physics of the detector, captured by the $B_{1\dots 7}(\vec{q})$ response functions, from any dependence on the DM model parameters or the DM velocity, $\vec{v}$. Conveniently, the expansion in $\vec{f}_V$ from Ref. [10] keeps the DM model dependence contained within five coefficients $a_{0,\dots,4}$, rather than the seven we would naively expect from the number of $B_i(\vec{q})$ functions.

Each a_i depends on quadratic combinations of Wilson coefficients, corresponding to Lagrangian operators such as those listed in Table I. Whether or not a given operator can appear in the Lagrangian depends on the spin of the DM particle and the spin of the off-shell mediator that couples the SM and DM sectors. In the simplest examples, when the mediator is a (spin-0) scalar, only a_0 is nonzero, and the only response function contributing to the scattering rate is B_1 . If the mediator is a vector boson, then some of the spin-dependent interactions are permitted even if the DM itself is a scalar. To keep track of which a_i coefficients can be nonzero in each scenario, Figure 1 depicts the mapping between DM particle models and the SM response functions, for spin-0 and spin- $\frac{1}{2}$ DM candidates. We also show which Wilson coefficients are generated by spin-1 DM candidates. Some features are immediately obvious: for example, the a_4 term is generated entirely by interference between pairs of Wilson coefficients, and it would vanish in any analysis that considers only one Wilson coefficient at a time. The spin-independent a_0 term only requires the scalar form factor f_S while $a_{1,\dots,4}$ require the contribution of the vector form factor $\vec{f}_V$. Only renormalizable tree-level couplings between the mediator and the SM/DM particles are shown in the figure. Higher dimensional operators, like the SM anapole moment for Majorana DM, would generate additional connections in the graph.

Before moving on, we briefly comment on how the operators $\mathcal{O}_{12}$ to $\mathcal{O}_{15}$ may be generated at one loop from the diagrams displayed in Fig. 2. First, notice that all of these operators require the DM to be fermionic due to the appearance of the spin $\vec{S}_\chi$. From the form of the operators and the nonrelativistic reduction of the spinor currents, it is straightforward to understand how they are generated. In the case of $\mathcal{O}_{13}$, $\mathcal{O}_{14}$, $\mathcal{O}_{15}$, the DM current and the electron current are not contracted with each other. A scalar mediator

ϕ is needed since it does not carry any Lorentz index, whereas in the case of $\mathcal{O}_{12}$ a vector mediator is needed to couple the two currents. To obtain the nonrelativistic $\vec{S}_{\chi,e} \cdot \vec{v}_{\text{el}}^\perp$ current appearing in $\mathcal{O}_{14}$, $\mathcal{O}_{13}$, the relativistic current $(\bar{u}_{\chi,e} \gamma^\mu \gamma^5 u_{\chi,e})(\bar{u}_e \gamma^\mu u_{e,\chi})$ must contract with an external momentum. The external momentum $(p + p')_\mu$ [or $(k + k')_\mu$] is afforded by the fermion propagators in the loop, while the fermion current is afforded by the vector A'_μ mediator. For $\mathcal{O}_{12}$, $(\bar{u}_\chi \gamma_\mu \gamma^5 u_\chi)(\bar{u}_e \sigma^{\mu\nu} u_e)$ must contract with a nonzero momentum to maintain Lorentz invariance. The first two loop diagrams can generate $\mathcal{O}_{15}$. For the first diagram, $\mathcal{O}_{15}$ is generated from the contraction of $(\bar{u}_\chi \gamma^5 u_\chi)(\bar{u}_e \sigma^{\mu\nu} u_e)$ or $(\bar{u}_\chi \gamma^5 u_\chi)(\bar{u}_e \sigma^{\mu\nu} \gamma^5 u_e)$ with two momenta. For the second diagram, $\mathcal{O}_{15}$ is generated from the contraction of $(\bar{u}_\chi \gamma^5 u_\chi)(\bar{u}_e \gamma^\mu u_e)$ with an external momentum.

Rather than keep the original $\vec{B}_{i=1\dots 7}$ response functions of Ref. [33], we find it more convenient to separate the vectorial form factor $\vec{f}_{1\rightarrow 2}(\vec{q})$ into a radial component, $\hat{q} \cdot \vec{f}_{1\rightarrow 2}$, and a tangential component, $\vec{f}_{1\rightarrow 2}^\perp$:

$$\begin{aligned} \vec{f}_{1\rightarrow 2} &\equiv (f_{1\rightarrow 2}^\parallel) \hat{q} + \vec{f}_{1\rightarrow 2}^\perp, & f_{1\rightarrow 2}^\parallel &\equiv \hat{q} \cdot \vec{f}_{1\rightarrow 2}, \\ \vec{A} &\equiv (A^\parallel) \hat{q} + \vec{A}^\perp, & A^\parallel &\equiv f_S(f_{1\rightarrow 2}^\parallel)^*, \end{aligned} \quad (27)$$

satisfying $\vec{f}_{1\rightarrow 2}^\perp \cdot \vec{q} = 0$ and $\vec{A}^\perp \cdot \vec{q} = 0$ everywhere. Not only does this simplify the mapping between Wilson coefficients and response functions, but also it makes it manifest which of the response functions should vanish in isotropic systems.

With this notation, the material response functions $B_{i \geq 2}$ become

$$\begin{aligned} B_2 &= \frac{q}{m_e} A^\parallel, & B_3 &= |f_{1\rightarrow 2}^\parallel|^2 + |\vec{f}_{1\rightarrow 2}^\perp|^2 = B_3^\parallel + B_3^\perp, \\ B_3^{(\perp, \parallel)} &\equiv |\vec{f}_{1\rightarrow 2}^{(\perp, \parallel)}|^2, & B_4 &= \frac{q^2}{m_e^2} B_3^\parallel, \\ B_5 &= \frac{2\vec{q}}{m_e} \cdot [\text{Re}(\vec{f}_{1\rightarrow 2}^\perp) \times \text{Im}(\vec{f}_{1\rightarrow 2}^\perp)], & \vec{B}_7 &= \frac{\vec{q}}{m_e} \times \vec{A}^\perp. \end{aligned} \quad (28)$$

Finally, the combination $\vec{v} \cdot \vec{B}_6$ that appears in the scattering rate simplifies to

$$\vec{v} \cdot \vec{B}_6 = v_{\min}(q) A^\parallel + \vec{v} \cdot \vec{A}^\perp, \quad (29)$$

where

$$v_{\min}(q) \equiv \frac{E_2 - E_1}{q} + \frac{q}{2m_\chi}. \quad (30)$$

With the exception of the a_4 components, all of the terms in Eq. (20) that depend on $f_{1\rightarrow 2}^\parallel$ but not $\vec{f}_{1\rightarrow 2}^\perp$ appear in the

¹Dot and cross products of $\vec{q}$ with $\vec{B}_6$ are already defined as B_2 and $\vec{B}_7$, respectively, and $\vec{q} \cdot \vec{B}_7 = \vec{A} \cdot (\vec{q} \times \vec{q}) = 0$.

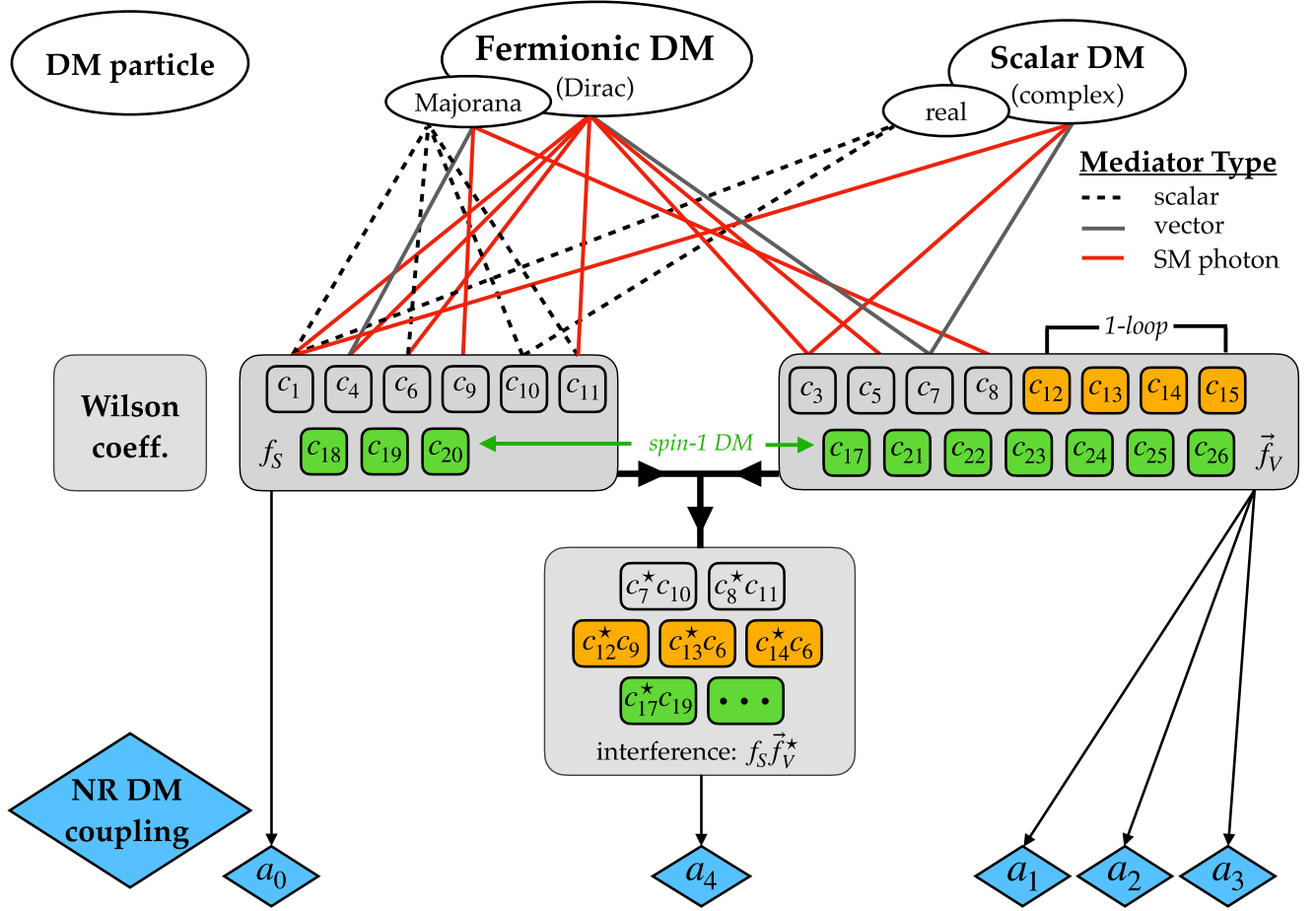


FIG. 1. The mapping of example UV DM models (ovals) to the c_i Wilson coefficients, the material response functions f_S and $\vec{f}_V$ (gray rectangles), and the NR DM couplings $a_{0,\dots,4}$ (blue diamonds). The colored lines join the UV models (scalar real/complex DM or fermionic Dirac/Majorana DM) to the Wilson coefficients that they generate, with the color of the line distinguishing between scalar (dashed black) and vector (solid) mediators. The SM photon (red) is a special case of vector mediator as it only couples to the electron through the vector current. Any c_i that can be generated via the SM photon can also be generated by a generic BSM vector mediator. All lines belonging to “Majorana” (or “real”) also belong to “Dirac” (or “complex”). Note that the coefficients in orange $c_{12}, \dots, c_{15}$ can be generated at one loop as shown in Fig. 2. Shown in green are the coefficients $c_{17}, \dots, c_{26}$ that can only be generated by a spin-1 DM particle (see Table 1 of [11] for the operators they correspond to). The coefficients c_i are classified by their contribution to the scalar form factor f_S (left gray rectangle) or to the vector form factor $\vec{f}_V$ (right gray rectangle). In the central gray box, we show all the five possible interference terms for scalar and fermionic DM. We denote in green the interference terms that are generated by spin-1 DM. In the lower part of the diagram, we show the relation between the c_i coefficients and the $a_{0,\dots,4}$ coefficients.

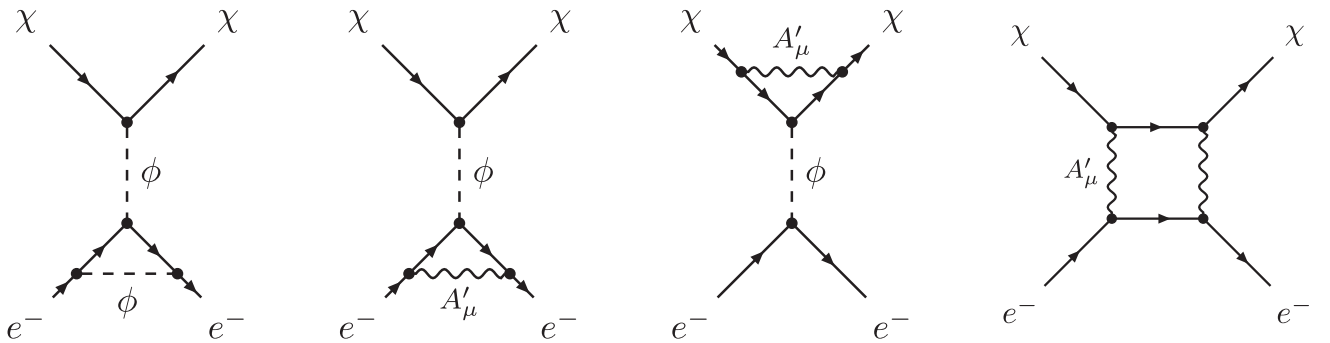


FIG. 2. From left to right, the first two loop diagrams generate $\mathcal{O}_{15}$; the second and third diagrams generate $\mathcal{O}_{14}$ and $\mathcal{O}_{13}$, respectively; and the last diagram generates $\mathcal{O}_{12}$. The $\mathcal{O}_{13,14}$ contributions from these loops vanish as $q \rightarrow 0$.

following combination:

$$B_H \equiv (\hat{q} \cdot \vec{v}_0^\perp)^2 B_1 - 2(\hat{q} \cdot \vec{v}_0^\perp) \text{Re} A^\parallel + B_3^\parallel. \quad (31)$$

This combination of response functions has an important physical significance: we demonstrate in Sec. III C that if the initial and final state wave functions are eigenstates of the same Hamiltonian, then $B_H \rightarrow 0$ vanishes identically.

In conclusion, noting that $|\vec{v}_0^\perp|^2 - (\hat{q} \cdot \vec{v}_0^\perp)^2 = v^2 - v_{\min}^2$, the squared scattering amplitude for a generic detector material is

$$\begin{aligned} |\overline{\mathcal{M}_{1 \rightarrow 2}}|^2 = & a_0 B_1 + a_1 [(v^2 - v_{\min}^2) B_1 - 2 \text{Re}(\vec{v} \cdot \vec{A}^\perp) + B_3^\perp] \\ & + (a_1 + a_2) B_H + a_3 [B_5 - 2 \text{Im}(\vec{v} \cdot \vec{B}_7)] \\ & + \frac{2q}{m_e} [\text{Im}(a_4) [(\hat{q} \cdot \vec{v}_0^\perp) B_1 - \text{Re} A^\parallel] \\ & - \text{Re}(a_4) \text{Im} A^\parallel], \end{aligned} \quad (32)$$

with $\text{Im}(a_4) \rightarrow 0$ for elastic DM scattering, i.e., when the incoming and outgoing DM states are the same. This is the same generic squared amplitude as Eq. (26), reorganized to make it easier to track which terms vanish in each simplifying regime studied in Sec. III.

III. SYMMETRIES AND SIMPLIFYING REGIMES

In all but the simplest detector models, the analytic forms of the wave functions $\psi_{1,2}(\vec{x})$ are not known exactly. In semiconductors and molecules, and (once the electron-electron Coulomb potentials are taken fully into account) in atoms other than hydrogen, the energy eigenstates $\psi_{1,2}$ must generally be obtained numerically. For this reason, the form factors $f_S(\vec{q})$ and $\vec{f}_{1 \rightarrow 2}(\vec{q})$ often represent the most time-consuming part of the calculation: a numerically derived $\psi_{1,2}$ must be integrated over d^3x (or momentum space) once for every value of $\vec{q}$. If $\psi_{1,2}$ are not spherically symmetric, a theory prediction may struggle under the weight of all these integrals. In this light, it is unfortunate that, in addition to B_1 , we must apparently also calculate several new spin-dependent response functions.

Thankfully, many of the relevant, physical systems obey symmetries or properties that allow for simplifications. In this section, we show how parity, isotropy, and adiabaticity simplify the expressions for spin-dependent scattering, to something that is almost as simple as in the spin-independent case. Section III A demonstrates that the a_3 and a_4 terms in Eq. (32) vanish if the detector material is parity symmetric, or invariant under reflections. Section III B investigates two types of isotropic systems: those that are isotropic because their constituent particles are spherically symmetric, as in atomic systems, and those that have become isotropic simply because the unit cells of the material are not aligned, i.e., the target is a fluid or a glass. Finally, in Sec. III C we consider systems where the initial and final state single particle wave

functions ψ_i and ψ_f are eigenstates of the same Hamiltonian. Using their equations of motion, we derive a relationship between $\vec{f}_V$ and f_S , resulting in the a_2 and a_4 terms vanishing.

A. Parity symmetry

The spin-averaged squared amplitude depends on five linear combinations of seven material response functions. In parity-symmetric (nonchiral) materials, however, we demonstrate that several of these response functions vanish: specifically, the a_3 and a_4 terms in Eq. (32), and the related response functions B_5 and $\text{Im}(\vec{B}_7)$. This simplification has previously been observed in atomic systems [8,11], but attributed imprecisely to rotational invariance rather than parity.

Parity, or central inversion symmetry, is common in the symmetry groups of crystals, including those with anisotropic response functions [39]. Thus, these results are relevant to a wide class of detector material candidates. The main counterexamples include chiral crystals [53], or detector systems where a strong external $\vec{B}$ field breaks the parity symmetry.

1. Parity eigenstates

We begin our discussion by focusing on Hamiltonians that commute with the parity operation P ,

$$[P, H] = 0. \quad (33)$$

Here $[P, H] \equiv PH - HP$ is the commutator, and the parity operator P acts on the position space or momentum space coordinate systems to set $\vec{x} \rightarrow -\vec{x}$ and $\vec{k} \rightarrow -\vec{k}$, respectively. Let $\psi_{1,2}$ be eigenstates of Hamiltonians $H_{1,2}$ of the form

$$H_i = -\frac{\nabla^2}{2m_i} + V_i(\vec{x}), \quad (34)$$

i.e., with no coupling to an external magnetic field. At this stage, we do not need to assume $m_1 = m_2$ or $V_1 = V_2$, merely that H_i is real. A parity-symmetric potential $V(\vec{x})$ is invariant when reflected through a particular point: without loss of generality, we choose this point to be the origin in position space, so that $V(\vec{x}) = V(-\vec{x})$.

The eigenstates of H_i , $|\psi_i\rangle$, form an orthogonal basis of the Hilbert space. Any basis function can be redefined by multiplying it by a constant phase, $|\psi_i\rangle \rightarrow e^{i\delta} |\psi_i\rangle$; using this freedom, we can ensure that $\psi_i(\vec{x}_0)$ is real valued at some arbitrary point $\vec{x}_0$ where $\psi_i(\vec{x}_0) \neq 0$. Because each $H(\vec{x})$ is a real-valued operator on position space functions $\psi(\vec{x})$, $H\psi_i = E_i\psi_i$, this phase choice $e^{i\delta}$ ensures that for nondegenerate eigenstates

$$\psi_i(\vec{x}) = \text{Re}\psi_i(\vec{x}) \quad (35)$$

for all $\vec{x}$.

This argument holds even if H has degenerate eigenstates. Take for example the special case where $V(\vec{x}) = 0$ and $[H, k] = 0$. Momentum eigenstates $|\vec{k}\rangle \sim \exp(i\vec{k} \cdot \vec{x})$ are not strictly real valued. However, a generic final state with energy $E = k^2/2m$ is a linear combination of the degenerate energy eigenstates with fixed $|\vec{k}| = k$, including the pair of momentum states $|\pm\vec{k}\rangle$; therefore, the Hilbert space can be spanned by real-valued eigenstates

$$\begin{aligned} \psi_{\pm\vec{k}}(\vec{x}) &\propto |\vec{k}\rangle \pm |-\vec{k}\rangle, & \psi_+(\vec{x}) &\sim \cos(\vec{k} \cdot \vec{x}), \\ \psi_-(\vec{x}) &\sim \sin(\vec{k} \cdot \vec{x}). \end{aligned} \quad (36)$$

Similar examples can be constructed for spherically symmetric potentials, where H commutes with the generators of rotation.

Once we have established a basis of real-valued energy eigenstates ψ_i in position space, we can specialize to parity-symmetric Hamiltonians. If $[P, H] = 0$, a nondegenerate ψ_i must also be a parity eigenstate:

$$P\psi_i = (\pm 1)\psi_i, \quad (37)$$

i.e., $\psi_i(-\vec{x}) = \pm\psi_i(\vec{x})$. If ψ_i is not a P eigenstate ($P\psi_i \neq \pm\psi_i$), but it still satisfies $H\psi_i = E_i\psi_i$ for a parity-symmetric H , then there must be a degenerate energy eigenstate ψ'_i such that

$$P\psi_i = \psi'_i. \quad (38)$$

Otherwise, $[P, H]\psi_i \neq 0$. Given these two degenerate states ψ_i, ψ'_i , it is possible to define a rotated basis $\psi_{\pm} \sim \psi_i \pm \psi'_i$ where $\psi_{\pm}$ are of definite parity. So, if $[P, H] = 0$, then the Hilbert space can be spanned by real-valued $\psi_i(\vec{x})$ that are eigenstates of both parity and energy.

Central potentials $V(x)$, e.g., hydrogenic atoms, provide a familiar example. It is common to label the electronic states of fixed energy according to their angular quantum numbers ℓ, m , i.e., $\psi_i = |n\ell m\rangle$, with $\psi_{n\ell m}(\vec{x}) \propto Y_{\ell}^m(\hat{x})$ or $\psi_{n\ell m}(\vec{k}) \propto Y_{\ell}^m(\hat{k})$. In this case, real-valued parity eigenstates can be constructed via the linear combinations $Y_{\ell}^m \pm Y_{\ell}^{-m}$.

2. Effect of parity on form factors

Having established a basis of eigenstates of P and H , we move on to the scalar and vector form factors, $f_S(\vec{q})$ and $\vec{f}_{1\rightarrow 2}(\vec{q})$. Because $\psi_{1,2}(\vec{x})$ are real valued with definite parity, the same is true for the product $\psi_2^*(\vec{x})\psi_1(\vec{x})$.² From Eq. (3), $f_S(\vec{q})$ is simply the Fourier transform of this function, or rather its complex conjugate:

²Since the ψ_i are real valued, $\psi_i^* = \psi_i$.

$$\begin{aligned} f_S(-\vec{q}) &= \mathcal{F}[\psi_2^*\psi_1] = \mathcal{F}[\psi_2\psi_1] \\ &= \int d^3x e^{-i\vec{q}\cdot\vec{x}} \psi_2(\vec{x})\psi_1(\vec{x}), \end{aligned} \quad (39)$$

$$f_S(\vec{q}) = f_S^*(-\vec{q}). \quad (40)$$

Noting that the Fourier transform commutes with parity,

$$f_S(\vec{q}) = P \cdot f_S(-\vec{q}) = \mathcal{F}[P \cdot \psi_2\psi_1] \equiv \mathcal{F}[(\pm 1)\psi_2\psi_1], \quad (41)$$

$$\equiv \epsilon_{12} f_S(-\vec{q}), \quad (42)$$

where $\epsilon_{12} = +1$ if the function $\psi_2\psi_1$ is even, and $\epsilon_{12} = -1$ if it is odd. Applying Eq. (40) to this result, we see that

$$f_S^*(\vec{q}) = \epsilon_{12} f_S(\vec{q}). \quad (43)$$

Therefore, if $\psi_{1,2}$ have matching parity under central inversion ($\epsilon_{12} = +1$), then f_S is purely real; if $\psi_{1,2}$ have opposite parities ($\epsilon_{12} = -1$), then f_S is purely imaginary.

Next, consider the quantity $i\vec{f}_{1\rightarrow 2}$,

$$\begin{aligned} im_e \vec{f}_{1\rightarrow 2}(\vec{q}) &= \int d^3x e^{i\vec{q}\cdot\vec{x}} \psi_2^*(\vec{x}) \cdot \nabla \psi_1(\vec{x}) \\ &= (\mathcal{F}[\psi_2^* \nabla \psi_1])^* = (\mathcal{F}[\psi_2 \nabla \psi_1])^*. \end{aligned} \quad (44)$$

The ∇ operator flips the parity of the integrand, so

$$\begin{aligned} P \cdot im_e \vec{f}_{1\rightarrow 2}(\vec{q}) &= \mathcal{F}[P \cdot \psi_2 \nabla \psi_1]^* = (-\epsilon_{12}) \mathcal{F}[\psi_2 \nabla \psi_1]^* \\ &= (-\epsilon_{12}) im_e \vec{f}_{1\rightarrow 2}(\vec{q}). \end{aligned} \quad (45)$$

For real-valued $\psi_{1,2}$, the parity operation $P \cdot \vec{f}_{1\rightarrow 2}$ is equivalent to complex conjugation:

$$-im_e \vec{f}_{1\rightarrow 2}^*(\vec{q}) = \mathcal{F}[\psi_2 \nabla \psi_1] = im_e \vec{f}_{1\rightarrow 2}(-\vec{q}), \quad (46)$$

or more simply

$$\vec{f}_{1\rightarrow 2}^*(\vec{q}) = -\vec{f}_{1\rightarrow 2}(-\vec{q}) = +\epsilon_{12} \vec{f}_{1\rightarrow 2}(\vec{q}). \quad (47)$$

Again, we see here that $\epsilon_{12} = \pm 1$ for $\psi_2\psi_1$ even/odd, respectively.³ Therefore, f_S and $\vec{f}_{1\rightarrow 2}$ transform the same under parity, i.e., if f_S is real, then $\vec{f}_{1\rightarrow 2}$ is also real; if f_S is imaginary, then so is $\vec{f}_{1\rightarrow 2}$.

The fact that f_S and $\vec{f}_{1\rightarrow 2}$ in a parity-symmetric system are either purely real or purely imaginary has immediate,

³In this section, we take the parity operator to act on the spatial coordinates. Equations (43) and (47) also follow unchanged if the parity operator acts on the Hilbert space, but the commutation relations between parity and momentum introduce extra minus signs in the intermediate equations.

important consequences for the material response functions. First, parity symmetry enforces that B_5 vanishes. From Eq. (23), the only way to obtain a nonzero B_5 is if $\vec{f}_{1 \rightarrow 2}$ has both real and imaginary components,

$$B_5 = \frac{2\vec{q}}{m_e} \cdot (\text{Re}(\vec{f}_{1 \rightarrow 2}) \times \text{Im}(\vec{f}_{1 \rightarrow 2})), \quad (48)$$

and Eq. (47) implies that either $\text{Re}(\vec{f}_{1 \rightarrow 2}) = 0$ or $\text{Im}(\vec{f}_{1 \rightarrow 2}) = 0$.

Second, parity symmetry implies that $\vec{A}$ is real valued:

$$\vec{A}^* = f_S^* \vec{f}_{1 \rightarrow 2} = (\epsilon_{12} f_S)(\epsilon_{12} \vec{f}_{1 \rightarrow 2}^*) = \epsilon_{12}^2 \vec{A} = \vec{A}, \quad (49)$$

which in turn implies that B_2 , B_6 , and B_7 are real valued. This allows us to simplify several of the terms in Eq. (26). The a_3 component of $|\overline{\mathcal{M}_{1 \rightarrow 2}}|^2$ vanishes completely:

$$ia_3 \frac{q}{m_e} \cdot (\vec{f}_V \times \vec{f}_V^*) = a_3 [B_5 - 2 \text{Im}(\vec{v} \cdot \vec{B}_7)] = 0. \quad (50)$$

If a_4 is real valued, then the a_4 component of $|\overline{\mathcal{M}_{1 \rightarrow 2}}|^2$ also vanishes. Finally, for real B_2 , Eq. (24) becomes

$$(B_2)^2 = B_4 B_1. \quad (51)$$

This modestly simplifies the a_2 term.

3. Matrix element

In conclusion, the scattering rate in a parity-symmetric material can be described by just three linear combinations of the $B_{1 \dots 3}$ and $\vec{B}_6$ material response functions,

$$\begin{aligned} |\overline{\mathcal{M}_{1 \rightarrow 2}}|^2 &= a_0 |f_S|^2 + a_1 |\vec{f}_V|^2 + \frac{a_2}{q^2/m_e^2} \left| \frac{\vec{q}}{m_e} \cdot \vec{f}_V \right|^2 \\ &= a_0 B_1 + a_1 [(v^2 - v_{\min}^2) B_1 - 2\vec{v} \cdot \vec{A}^\perp + B_3^\perp] \\ &\quad + (a_1 + a_2) B_H. \end{aligned} \quad (52)$$

This result applies to crystals composed of P -symmetric molecules, e.g., [39,54], as well as semiconductors with P -symmetric lattices. Parity symmetry in crystalline silicon explains the vanishingly small contributions from $\text{Im} B_2$ and B_5 in Ref. [33], for example. If the Hamiltonian H permits degenerate excited states, the cancellation between the a_3 and a_4 contributions only becomes apparent when summing over the degenerate final states: i.e.,

$$\begin{aligned} \sum_f |\overline{\mathcal{M}_{1 \rightarrow f}}|^2 &= \sum_f \left(a_0 |f_S|^2 + a_1 |\vec{f}_V|^2 + \frac{a_2}{q^2/m_e^2} \left| \frac{\vec{q}}{m_e} \cdot \vec{f}_V \right|^2 \right), \end{aligned} \quad (54)$$

summing over all ψ_f satisfying $H\psi_f = E_2\psi_f$.

B. Isotropic detectors

Another common symmetry found in materials is isotropy. A material may be isotropic because its $\psi_{1,2}$ wave functions are eigenstates of a spherically symmetric Hamiltonian, with $V(\vec{x}) = V(x)$. Alternatively, as in the cases of fluids and glasses, the material is isotropic only because its constituents are randomly aligned (e.g., liquid molecular scintillators [54]). The total scattering rate is then proportional to isotropic averages of the anisotropic material response functions. In this section, we explore how isotropy simplifies the scattering rate, even in chiral materials.⁴

1. Rotation operator

Following Eq. (15), an isotropic detector can be described as a collection of unit cells, each with a different orientation $\mathcal{R}$, sampled uniformly on the $SO(3) \equiv S^3/Z_2$ manifold. To begin, we review how the rotation operator acts on the wave functions $\psi_{1,2}$. We take $\mathcal{R}$ to act on the detector, rather than the DM velocity distribution, so that $\mathcal{R}$ acts only on the wave functions $\psi_{1,2}$:

$$\mathcal{R} \cdot \psi(\vec{u}) = \psi(\mathcal{R}^{-1} \cdot \vec{u}), \quad (55)$$

for $\vec{u} = \vec{x}$ or $\vec{u} = \vec{k}$ in position or momentum space. However, by a change of variable, this is equivalent to acting on the $\vec{q}$ -dependent form factors $f_S(\vec{q})$ and $\vec{f}_{1 \rightarrow 2}(\vec{q})$:

$$\begin{aligned} \mathcal{R} \cdot f_S(\vec{q}) &= f_S(\mathcal{R}^{-1} \vec{q}), \\ \mathcal{R} \cdot \vec{f}_{1 \rightarrow 2}(\vec{q}) &= [\mathcal{R} \vec{f}_{1 \rightarrow 2}](\mathcal{R}^{-1} \vec{q}). \end{aligned} \quad (56)$$

Rotations on a vector field $\vec{f}_{1 \rightarrow 2}$ change the direction of the vector, $\mathcal{R}\vec{f}$, as well as its dependence on its argument. After simplifying the dot products $\vec{q} \cdot \vec{f}(\vec{q}) \rightarrow \vec{q} \cdot \mathcal{R}\vec{f}(\mathcal{R}^{-1}\vec{q}) = (\mathcal{R}^{-1}\vec{q}) \cdot \vec{f}(\mathcal{R}^{-1}\vec{q})$, all the scalar response functions $B_{1 \dots 5}$ transform as

$$\mathcal{R} \cdot B_i(\vec{q}) = B_i(\mathcal{R}^{-1} \vec{q}). \quad (57)$$

2. Isotropic averaging of form factors

In a detector with $N_{\text{cell}} \sim N_A$ many randomly aligned constituent particles or unit cells, it is an excellent approximation to ignore the $1/\sqrt{N_{\text{cell}}}$ statistical fluctuations away from spherical symmetry. Each material response function can then be replaced by its isotropic average:

$$\langle f(\vec{q}) \rangle \equiv \frac{1}{V_{SO(3)}} \int_{SO(3)} d\mathcal{R} (\mathcal{R} \cdot f(\vec{q})), \quad (58)$$

⁴Chirality is rotation invariant, so a liquid of chiral molecules retains its chirality.

an equally weighted sum over all $\mathcal{R} \in SO(3)$, normalized by the volume $V_{SO(3)} = \frac{1}{2}2\pi^2$. If $f(\vec{q}) = f(q)$ is already spherically symmetric, then each $\mathcal{R}f = f$ acts trivially on the response functions. We note that the isotropic average is a function only of $q = |\vec{q}|$:

$$B_i^{\text{iso}}(q) \equiv \langle B_i(\vec{q}) \rangle, \quad (59)$$

for $i = 1, \dots, 5$.

For the vectorial response functions $\vec{B}_{6,7}$, on the other hand, we must track how $\mathcal{R}$ acts on the vector field $\vec{A}(\vec{q})$. Using the index notation with unit vectors $\hat{e}_i$, where repeated indices imply summation, $\vec{A}(\vec{q}) = A_i(q_j \hat{e}_j) \hat{e}_i$ transforms under rotations as

$$\mathcal{R} \cdot \vec{A}(\vec{q}) = A_i(q_j \mathcal{R}_{jb} \hat{e}_b) \mathcal{R}_{ia}^{-1} \hat{e}_a, \quad (60)$$

where $\mathcal{R}^{-1}$ is the inverse of the 3×3 matrix representation of the active rotation $\mathcal{R} \in SO(3)$. A generic $\vec{A}$ has a radial component parallel to $\vec{q}$, which we denote as $A^{\parallel} \hat{q}$, and a tangential component $\vec{A}^{\perp}$ satisfying $\vec{q} \cdot \vec{A}^{\perp} = 0$:

$$\vec{A}(\vec{q}) = A^{\parallel}(\vec{q}) \hat{q} + \vec{A}^{\perp}(\vec{q}). \quad (61)$$

In the isotropic average of $\vec{A}$, $\vec{A}_{\text{iso}}(q) = \langle \vec{A}(\vec{q}) \rangle$, the contributions from $\vec{A}^{\perp}$ vanish. This can be seen most simply by the requirement that $\langle \vec{A}^{\perp} \rangle$ should be rotationally invariant. If $\langle \vec{A}^{\perp}(\vec{q}) \rangle \neq 0$ at some point $\vec{q}$, then we can apply a 180° degree rotation about the $\hat{q}$ axis:

$$\mathcal{R}_{\pi}(\vec{q}) \cdot \langle \vec{A}^{\perp}(\vec{q}) \rangle = -\langle \vec{A}^{\perp}(\vec{q}) \rangle = 0. \quad (62)$$

This leaves $\vec{q}$ fixed, but rotates $\vec{A}^{\perp} \rightarrow -\vec{A}^{\perp}$. So, a rotationally invariant $\vec{A}^{\perp}$ must be zero.⁵ In the isotropic average, then,

$$\vec{B}_6^{\text{iso}}(q) \equiv \vec{A}_{\text{iso}}(q) = A_{\text{iso}}^{\parallel}(q) \hat{q} \quad (63)$$

can include only a radial component, and

$$\vec{B}_7^{\text{iso}}(q) = \frac{1}{m_e} \langle \vec{q} \times \vec{A}(\vec{q}) \rangle = \left\langle \frac{\vec{q}}{m_e} \times (A^{\parallel}(\vec{q}) \hat{q}) \right\rangle = 0 \quad (64)$$

vanishes identically. While $\vec{B}_6^{\text{iso}}$ does not generally vanish, it is a simple function of $\vec{q} B_2(\vec{q})$:

$$\vec{B}_6^{\text{iso}} = \vec{A}_{\text{iso}}(q) = \frac{\vec{q}}{q^2} (\vec{q} \cdot \vec{A}_{\text{iso}}(q)) = \frac{\vec{q}}{m_e} \frac{m_e^2}{q^2} B_2^{\text{iso}}(q), \quad (65)$$

so that

$$\vec{v} \cdot \vec{B}_6^{\text{iso}}(\vec{q}) = \frac{\vec{v} \cdot \vec{q}}{m_e} \frac{m_e^2}{q^2} B_2^{\text{iso}}(q). \quad (66)$$

So, the combination of B_2 and $\vec{B}_6$ that appears in Eq. (26) can be simplified to

$$\begin{aligned} & \left[\frac{m_e}{\mu_{\chi e}} B_2 - 2\vec{v} \cdot \text{Re} \vec{B}_6 \right]_{\text{iso}} \\ &= -\frac{2m_e}{q^2} (\vec{q} \cdot \vec{v}_0^{\perp}) \text{Re} B_2^{\text{iso}}(q) + \frac{m_e}{\mu_{\chi e}} \text{Im}(B_2^{\text{iso}}). \end{aligned} \quad (67)$$

Although $\langle \vec{A}^{\perp} \rangle = 0$ in the isotropic average, this does not mean that $\vec{A}^{\perp}$ can be completely ignored: B_3 , for example, is quadratic in $\vec{f}_{1 \rightarrow 2} \propto \vec{A}$, and

$$|\vec{A}|^2 = |\hat{q} \cdot \vec{A}|^2 + (\vec{A}^{\perp})^* \cdot \vec{A}^{\perp}, \quad (68)$$

$$\langle B_1 B_3 \rangle = \langle |A^{\parallel}|^2 + |\vec{A}^{\perp}|^2 \rangle. \quad (69)$$

Finally, let us consider $\langle B_5(\vec{q}) \rangle$. Note that B_5 can be decomposed as

$$B_5^{\text{iso}}(q) = \left\langle \frac{2\vec{q}}{m_e} \cdot \text{Re}(\vec{f}_{1 \rightarrow 2}) \times \text{Im}(\vec{f}_{1 \rightarrow 2}) \right\rangle. \quad (70)$$

In isotropic configurations of anisotropic unit cells, there can be tangential components of $\vec{f}_{1 \rightarrow 2}(\vec{q})$, and in a chiral system there is no requirement for $\text{Re}(\vec{f}_{1 \rightarrow 2}) \times \text{Im}(\vec{f}_{1 \rightarrow 2})$ to vanish. Therefore, B_5 can contribute to isotropic-but-chiral detector materials.

3. Matrix element

After the isotropic average, then, the spin averaged squared amplitude is

$$\begin{aligned} \overline{|\mathcal{M}_{1 \rightarrow 2}|^2} &= a_0 B_1^{\text{iso}}(q) + a_1 \left[|\vec{v}_0^{\perp}|^2 B_1^{\text{iso}}(q) - \frac{2m_e^2}{q^2} \frac{\vec{q} \cdot \vec{v}_0^{\perp}}{m_e} \text{Re} B_2^{\text{iso}}(q) + B_3^{\text{iso}}(q) \right] \\ &+ a_2 \frac{m_e^2}{q^2} \left[\left(\frac{\vec{q} \cdot \vec{v}_0^{\perp}}{m_e} \right)^2 B_1^{\text{iso}}(q) - 2 \left(\frac{\vec{q} \cdot \vec{v}_0^{\perp}}{m_e} \right) \text{Re} B_2^{\text{iso}}(q) + B_4^{\text{iso}}(q) \right] \\ &+ a_3 B_5^{\text{iso}}(q) - 2 \text{Re}(a_4) \text{Im} B_2^{\text{iso}}(q). \end{aligned} \quad (71)$$

⁵This is a corollary of the fuzzy ball theorem for vector fields defined on the S^2 sphere: if $\vec{A}^{\perp}$ is continuous, then there must be at least two points at which $\vec{A}^{\perp}(\vec{q}) = 0$. Requiring that $\vec{A}_{\text{iso}}^{\perp}(\vec{q})$ is also rotationally invariant implies that $\vec{A}_{\text{iso}}^{\perp} = 0$ everywhere.

Recall that $a_i(q)$ are functions of $q = |\vec{q}|$, but not $\vec{v}$. In a parity-symmetric, rotationally invariant detector,

$$\begin{aligned} \langle |\overline{\mathcal{M}_{1 \rightarrow 2}}|^2 \rangle &= a_0 B_1^{\text{iso}}(q) + a_1 \left[|\vec{v}_0^\perp|^2 B_1^{\text{iso}}(q) - \frac{2m_e^2 \vec{q} \cdot \vec{v}_0^\perp}{q^2 m_e} B_2^{\text{iso}}(q) + B_3^{\text{iso}}(q) \right] \\ &+ a_2 \frac{m_e^2}{q^2} \left[\left(\frac{\vec{q} \cdot \vec{v}_0^\perp}{m_e} \right)^2 B_1^{\text{iso}}(q) - 2 \left(\frac{\vec{q} \cdot \vec{v}_0^\perp}{m_e} \right) B_2^{\text{iso}}(q) + B_4^{\text{iso}}(q) \right]. \end{aligned} \quad (72)$$

Although $B_4 = B_2^2/B_1$, it is not necessarily true that $B_4^{\text{iso}} = (B_2^{\text{iso}})^2/B_1^{\text{iso}}$. That would only be guaranteed if $B_2(\vec{q})$ and $B_1(\vec{q})$ are both spherically symmetric, which is a much more restrictive assumption.

4. Discrete rotational symmetries

Many crystal groups include one or more discrete rotational symmetries, e.g., under rotations of 180° about some axis $\hat{b}$. While the linear terms in $\vec{A}^\perp$ (i.e., $\vec{B}_{6,7}$) cannot *generally* be ignored, we can see immediately that $\vec{A}^\perp(q\hat{b}) = 0$ for $\vec{q} = q\hat{b}$ along the axis of symmetry. In the case of degenerate E_2 final states, this argument assumes that the energy eigenstates are labeled using eigenstates of the discrete rotation operator; otherwise, the cancellation of $A^\perp(q\hat{b})$ arises later in the calculation, in the sum over degenerate states. $\vec{A}^\perp$ still needs to be evaluated for generic off-axis values of $\vec{q}$, however.

C. Electronic adiabaticity

In this section, we derive the relationships between B_1 and some of the $B_{i \geq 2}$ response functions for the class of physical systems where ψ_1 and ψ_2 are eigenstates of a Hamiltonian that acts on position space $\psi_{1,2}(\vec{x})$ as

$$H_0 = -\frac{\nabla^2}{2m} + V(\vec{x}), \quad (73)$$

with a potential $V(\vec{x})$ that depends only on $\vec{x}$, and not its conjugate momentum. Here, depending on the system, m can represent, i.e., the reduced or effective mass of the system. We will show that when $V(\vec{x})$ is the same for the initial and final states, we can use our derivation to replace all the appearances of $\vec{q} \cdot \vec{f}_{1 \rightarrow 2}$ with simply f_S . For $\vec{q}$ and $\vec{v}$ satisfying the energy conservation condition

$$\vec{q} \cdot \vec{v} = E_2 - E_1 + q^2/(2m_\chi), \quad (74)$$

we show that the terms multiplying a_2 and a_4 in Eq. (26) vanish. We refer to this special case with unchanging $V_1(\vec{x}) = V_2(\vec{x})$ as “electronic adiabaticity,” because it implies that the Coulomb potential from the atomic nuclei *and* the other electrons in the system is unchanged by the DM-induced excitation of one electron. Aside from single-electron systems like the hydrogen atom or the He^+ ion, examples of approximate e^- adiabaticity include the many atomic or molecular systems where the excitation above the

ground state is well described by a single pair of Hartree-Fock eigenstates [55,56]. A sufficient condition for adiabaticity can be obtained from the transition density matrix T_{ij} of Eq. (4). If the $\Psi_1 \rightarrow \Psi_2$ transition is dominated by a single element of T_{ij} , in a basis where $|\psi_i\rangle$ and $|\psi_j\rangle$ diagonalize the same single-particle Hamiltonian, then Eq. (73) is a good approximation. Common examples include the first excited state in many aromatic organic molecules [39,54], or quantum dots in the strongly confining regime [57] where the effective Bohr radius of the electron-hole pair is larger than the quantum dot.

Section III C 1 begins without assuming that $V(\vec{x})$ has any spatial symmetries, showing that the spin-averaged amplitude squared in Eq. (26) reduces to just the a_0 , a_1 , and a_3 terms. In Sec. III C 7, we specialize to parity-symmetric and isotropic systems, using our results to further simplify the dependence on the material response functions.

1. Integration by parts

To simplify the presentation of our derivation, we define a symmetric vector form factor $\vec{f}_{\leftrightarrow}$:

$$\vec{f}_{\leftrightarrow}(\vec{q}) \equiv \frac{-i}{m} \int d^3x e^{i\vec{q} \cdot \vec{x}} (\psi_2^* \vec{\nabla} \psi_1), \quad (75)$$

where

$$\psi_2^* \vec{\nabla} \psi_1 \equiv \psi_2^* \nabla \psi_1 - \psi_1 \nabla \psi_2^*. \quad (76)$$

Up to a surface integral, this $\vec{f}_{\leftrightarrow}$ is related to the original $\vec{f}_{1 \rightarrow 2}$ and f_S via integration by parts:

$$\vec{f}_{\leftrightarrow}(\vec{q}) = \vec{f}_{1 \rightarrow 2}(\vec{q}) + \frac{i}{m} \int d^3x e^{i\vec{q} \cdot \vec{x}} \psi_1 \nabla \psi_2^* \quad (77)$$

$$\begin{aligned} &= 2\vec{f}_{1 \rightarrow 2}(\vec{q}) + \frac{\vec{q}}{m} f_S(\vec{q}) \\ &+ \frac{i}{m} \int_{\partial V} d\Gamma \vec{n} e^{i\vec{q} \cdot \vec{x}} \psi_2^* \psi_1, \end{aligned} \quad (78)$$

where $\vec{n}$ is the unit normal vector to the surface ∂V on the boundary of the integration volume, and $d\Gamma$ is an infinitesimal surface element on ∂V .

In isolated systems (e.g., nuclei, atoms, molecules) the $\int d^3x$ integral usually extends to $|\vec{x}| \rightarrow \infty$, and so the boundary integral vanishes because $\psi_1(\vec{x} \rightarrow \infty) = 0$ for

the ground state. For crystals, $\int d^3x$ may be applied instead to a single unit cell, with $\psi_{1,2}(\vec{x})$ given periodic boundary conditions at the finite surface ∂V . In this example, assuming $\psi_{1,2}$ are continuous and topologically trivial, the surface integrals cancel pairwise: neighboring unit cells have matching $e^{i\vec{q}\cdot\vec{x}}\psi_2^*(\vec{x})\psi_1(\vec{x})$ at the boundary, with opposite-sign $\pm\hat{n}$ normal vectors. For completeness, we include the boundary terms in the following results. However, as long as there are no topologically nontrivial edge states, which could appear in Weyl semimetals (such as TaAs [58,59]) or topological insulators (such as Bi₂Se₃ [60,61]), we expect these surface integrals to vanish.

2. Simplifying B_2 and B_4

We note that $\vec{q} \cdot \vec{f}_{\leftrightarrow}$ can be written as an integral involving a gradient of $e^{i\vec{q}\cdot\vec{x}}$:

$$\begin{aligned} -m\vec{q} \cdot \vec{f}_{\leftrightarrow} &= \int d^3x e^{i\vec{q}\cdot\vec{x}} i\vec{q} \cdot (\psi_2^* \vec{\nabla} \psi_1) \\ &= \int d^3x \nabla(e^{i\vec{q}\cdot\vec{x}}) \cdot (\psi_2^* \vec{\nabla} \psi_1). \end{aligned} \quad (79)$$

Integrating this expression by parts, we find

$$\begin{aligned} \vec{q} \cdot \vec{f}_{\leftrightarrow}(\vec{q}) &= - \int_{\partial V} d\Gamma e^{i\vec{q}\cdot\vec{x}} \frac{\vec{n}}{m} \cdot (\psi_2^* \vec{\nabla} \psi_1) \\ &\quad + 2 \int d^3x e^{i\vec{q}\cdot\vec{x}} \frac{\psi_2^* \nabla^2 \psi_1 - \psi_1 \nabla^2 \psi_2^*}{2m}. \end{aligned} \quad (80)$$

If $\psi_{1,2}$ are both solutions to second-order differential equations of the form Eq. (73), i.e., $H_i\psi_i = E_i\psi_i$ with $\nabla^2/(2m) = -H_i + V_i(\vec{x})$ for potentials $V_{i=1,2}$, then

$$\begin{aligned} \vec{q} \cdot \vec{f}_{\leftrightarrow}(\vec{q}) &= - \int_{\partial V} d\Gamma e^{i\vec{q}\cdot\vec{x}} \frac{\vec{n}}{m} \cdot (\psi_2^* \vec{\nabla} \psi_1) \\ &\quad + 2 \int d^3x e^{i\vec{q}\cdot\vec{x}} [\psi_2^* (V_1 - E_1) \psi_1 \\ &\quad + \psi_1 (E_2 - V_2) \psi_2^*]. \end{aligned} \quad (81)$$

The potentials $V_{1,2}(\vec{x})$ commute with the position-space wave functions, leaving only

$$\begin{aligned} \vec{q} \cdot \vec{f}_{\leftrightarrow}(\vec{q}) &= 2(E_2 - E_1)f_S(\vec{q}) - \int_{\partial V} d\Gamma e^{i\vec{q}\cdot\vec{x}} \frac{\vec{n}}{m} \cdot (\psi_2^* \vec{\nabla} \psi_1) \\ &\quad - 2\langle\psi_2|e^{i\vec{q}\cdot\vec{x}}\delta V|\psi_1\rangle, \end{aligned} \quad (82)$$

where for the generic case $V_2 \neq V_1$ we define

$$\langle\psi_2|e^{i\vec{q}\cdot\vec{x}}\delta V|\psi_1\rangle \equiv \int d^3x e^{i\vec{q}\cdot\vec{x}} (V_2(\vec{x}) - V_1(\vec{x}))\psi_2^*\psi_1. \quad (83)$$

For electronically adiabatic systems, $\langle\psi_2|e^{i\vec{q}\cdot\vec{x}}\delta V|\psi_1\rangle = 0$.

Like the boundary term in Eq. (78), the $(\psi_2^* \vec{\nabla} \psi_1)$ part of the integrand vanishes for isolated systems, as ψ_1 and $\nabla\psi_1$ both vanish in the $|\vec{x}| \rightarrow \infty$ limit. For periodic crystals, the $\pm\vec{n}$ cancellation from adjacent unit cells also applies to the new integrand, as long as $(\psi_2^* \vec{\nabla} \psi_1)$ is continuous at the boundary of the unit cell (i.e., assuming there is no infinitely dense surface charge distribution at ∂V). Topologically nontrivial wave functions at the physical surface of the detector volume could in principle generate a nonzero boundary term.

With Eq. (78), it is easy to convert $\vec{q} \cdot \vec{f}_{\leftrightarrow} = 2Ef_S$ into a similar expression for $\vec{q} \cdot \vec{f}_{1\rightarrow 2}$,

$$\begin{aligned} \vec{q} \cdot \vec{f}_{1\rightarrow 2} &= \left(E_2 - E_1 - \frac{q^2}{2m}\right)f_S \\ &\quad - \frac{1}{2m} \int_{\partial V} d\Gamma e^{i\vec{q}\cdot\vec{x}} \psi_2^* (\vec{n} \cdot \vec{\nabla} + i\vec{q} \cdot \vec{n}) \psi_1, \end{aligned} \quad (84)$$

which can in turn be used to write B_2 and B_4 in terms of the simpler B_1 response function. Using the usual arguments to drop the boundary terms, and Eq. (74) we find

$$\vec{q} \cdot \vec{f}_{1\rightarrow 2} = (\vec{q} \cdot \vec{v}_0^\perp)f_S, \quad \vec{q} \cdot \vec{A} = (\vec{q} \cdot \vec{v}_0^\perp)B_1, \quad (85)$$

$$B_2 = \frac{\vec{q} \cdot \vec{v}_0^\perp}{m} B_1, \quad B_4 = \left(\frac{\vec{q} \cdot \vec{v}_0^\perp}{m}\right)^2 B_1. \quad (86)$$

Notice that B_2 is now explicitly real valued.

For generic anisotropic detectors, the part of $\vec{f}_{1\rightarrow 2}$ that is perpendicular to $\vec{q}$ (i.e., $\vec{q} \times \vec{f}$) is not simplified by this trick, so we still expect to need B_3 , B_5 , $\vec{B}_6$, and $\vec{B}_7$. This is where the organization scheme of Ref. [11] becomes particularly convenient. Rather than $\vec{q} \cdot \vec{f}_{1\rightarrow 2}$, consider instead the velocity-dependent vector form factor, $\vec{q} \cdot \vec{f}_V$:

$$\vec{q} \cdot \vec{f}_V(\vec{q}, \vec{v}) = -\left(E_2 - E_1 + \frac{q^2}{2m_\chi} - \vec{q} \cdot \vec{v}\right)f_S(\vec{q}). \quad (87)$$

This combination of E , q^2 , and $\vec{q} \cdot \vec{v}$ is the same one that appears in the argument of the energy-conserving δ function in Eq. (1). Consequently, for on-shell momenta $\vec{q}$, for any nonrelativistic $\vec{v}$, we find

$$\vec{q} \cdot \vec{f}_V \rightarrow 0, \quad (88)$$

up to some surface integrals that vanish under the typical assumptions.

As a verification of our results, we test Eq. (85) in two single particle quantum systems where we know *a priori* that $\delta V = 0$. These examples are provided in the Appendix. First, as a crude approximation of a crystal, we take $\psi_{1,2}$ to be energy eigenstates of a particle in an infinite rectangular well, with arbitrary box dimensions (L_x, L_y, L_z) . Next, we consider an isolated hydrogen atom, with Ψ_2 any of the excited states. Both of these examples permit analytic solutions for f_S and $\vec{f}_{1 \rightarrow 2}$, as well as E_1 and E_2 . As expected, we find

$$\vec{q} \cdot \vec{f}_{1 \rightarrow 2}(\vec{q}) = \left(E_2 - E_1 - \frac{q^2}{2m} \right) f_S(\vec{q}). \quad (89)$$

3. Simplifying B_3 and $\vec{B}_6$

For the remainder of our discussion, it is helpful to separate the $\vec{f}_{1 \rightarrow 2}(\vec{q})$ form factor into a radial part, $\propto \hat{q}$, and a tangential part $\vec{f}_{1 \rightarrow 2}^\perp$, as in Eq. (28):

$$\vec{f}_{1 \rightarrow 2} = (\hat{q} \cdot \vec{f}_{1 \rightarrow 2}) \hat{q} + \vec{f}_{1 \rightarrow 2}^\perp. \quad (90)$$

The radial part, though not $\vec{f}_{1 \rightarrow 2}^\perp$, is related to f_S via integration by parts:

$$\vec{f}_{1 \rightarrow 2}(\vec{q}) = \frac{\vec{q} \cdot \vec{v}_0^\perp}{q} f_S(\vec{q}) \hat{q} + \vec{f}_{1 \rightarrow 2}^\perp(\vec{q}), \quad (91)$$

$$\vec{A}(\vec{q}) = f_S(\vec{q}) \vec{f}_{1 \rightarrow 2}^\star(\vec{q}) = \frac{\vec{q} \cdot \vec{v}_0^\perp}{q} B_1(\vec{q}) \hat{q} + \vec{A}^\perp(\vec{q}), \quad (92)$$

where $\vec{A}^\perp = f_S(\vec{f}_{1 \rightarrow 2}^\perp)^\star$. So, B_3 and $\vec{B}_6$ separate into a term proportional to B_1 plus a $B_{3,6}^\perp$ term generated only by $\vec{f}_{1 \rightarrow 2}^\perp$ or $\vec{A}^\perp$:

$$B_3 = \left(\frac{\vec{q} \cdot \vec{v}_0^\perp}{q} \right)^2 B_1 + B_3^\perp, \quad B_3^\perp \equiv |\vec{f}_{1 \rightarrow 2}^\perp|^2, \quad (93)$$

$$\vec{v} \cdot \vec{B}_6 = \frac{\vec{v} \cdot \vec{q}}{q} \frac{\vec{q} \cdot \vec{v}_0^\perp}{q} B_1 + \vec{v} \cdot \vec{B}_6^\perp, \quad \vec{B}_6^\perp \equiv \vec{A}^\perp. \quad (94)$$

Note that the magnitudes of $B_6^\perp$ and $B_3^\perp$ are related:

$$|\vec{B}_6^\perp|^2 = B_1 B_3^\perp. \quad (95)$$

Because B_5 and $\vec{B}_7$ are independent of the radial part of $\vec{f}_{1 \rightarrow 2}$,

$$B_5 = \frac{2\vec{q}}{m_e} \cdot (\text{Re}(\vec{f}_{1 \rightarrow 2}^\perp) \times \text{Im}(\vec{f}_{1 \rightarrow 2}^\perp)), \quad \vec{B}_7 = \frac{\vec{q}}{m_e} \times \vec{A}^\perp, \quad (96)$$

they are not simplified by the integration by parts trick.

4. Matrix element

With the identification that $|\vec{v}_0^\perp|^2 - (\hat{q} \cdot \vec{v}_0^\perp)^2 = v^2 - v_{\min}^2(q)$, we can write the simplified spin-averaged squared amplitude for a system where $\psi_{1,2}$ are eigenstates of the same Hamiltonian:

$$\overline{|\mathcal{M}_{1 \rightarrow 2}|^2} = a_0 B_1 + a_1 [(v^2 - v_{\min}^2) B_1 + B_3^\perp - 2\text{Re}(\vec{v} \cdot \vec{B}_6^\perp)] + a_3 [B_5 - 2\text{Im}(\vec{v} \cdot \vec{B}_7)]. \quad (97)$$

From Eqs. (93) and (94), we see that $B_3^\parallel = (\hat{q} \cdot \vec{v}_0^\perp)^2 B_1$ and $B_6^\parallel = (\hat{q} \cdot \vec{v}_0^\perp) B_1$,

$$B_H = (\hat{q} \cdot \vec{v}_0^\perp)^2 B_1 - 2(\hat{q} \cdot \vec{v}_0^\perp) B_6^\parallel + B_3^\parallel \longrightarrow 0. \quad (98)$$

The $a_1 B_1$ term is now explicitly non-negative, vanishing when $v \rightarrow v_{\min}(q)$ approaches the kinematic threshold for exciting the E_2 state.

5. Multiparticle wave functions and nonadiabaticity

In a system with multiple electrons, one generally expects a change to one electronic wave function to have some impact on the other electrons in the system. When the wave functions of the other electrons change, the Coulomb potential $V_2(\vec{x})$ acting on ψ_2 changes, and $V_2 \neq V_1$ for the e^- scattered by the DM. Even so, there are many cases where DM- e^- scattering is well approximated by $V_1 \approx V_2$. Precise *ab initio* calculations, e.g., post-Hartree-Fock methods or time-dependent density functional theory, can reveal whether or not $V_1 = V_2$ is a good approximation for a given system.

If ψ_2 and ψ_1 are eigenstates of *different* Hamiltonians, but with the same effective particle mass m , then

$$-\frac{\nabla^2}{2m} = H_1 - V_1 = H_2 - V_2, \quad (99)$$

with $V_1 \neq V_2$. We no longer expect perfect cancellation in $\vec{q} \cdot \vec{f}_V$. Instead, continuing to drop the $\int_{\partial V}$ surface integrals and assuming the kinematics of elastic DM, integration by parts gives us

$$\vec{q} \cdot \vec{f}_{1 \rightarrow 2} = (\vec{q} \cdot \vec{v}_0^\perp) f_S - \langle \psi_2 | e^{i\vec{q} \cdot \vec{x}} \delta V | \psi_1 \rangle, \quad (100)$$

for the $\langle \psi_2 | e^{i\vec{q} \cdot \vec{x}} \delta V | \psi_1 \rangle$ defined in Eq. (83). For the velocity-dependent vector form factor, we find

$$\vec{q} \cdot \vec{f}_V = \langle \psi_2 | e^{i\vec{q} \cdot \vec{x}} \delta V | \psi_1 \rangle. \quad (101)$$

A nonzero effective δV generally arises in electronic systems whenever multiparticle effects are important. For example, in a system like trans-stilbene, the lowest energy transition is often dominated by one single particle process [39,62]. However, the higher-energy transitions can receive substantial contributions from multiple pairs of Hartree-

Fock eigenstates and therefore it is not guaranteed that $\delta V \approx 0$ is a good approximation for these transitions. Other examples where δV cannot be neglected include non-adiabatic Migdal scattering in molecules [41] and crystals [49], as well as systems where exciton-hole interactions are important such as nanorods and quantum dots. In these systems, the initial and final states have somewhat different arrangements of the atomic nuclei, and the two electronic states have appreciably different Coulomb potentials.

6. Inelastic DM

In the case of inelastic DM with a mass splitting δ , the kinematic constraint relating $E_2 - E_1$ and $\vec{q}$ is modified to

$$E_2 - E_1 = -m_\chi \epsilon + \frac{\epsilon}{1 + \epsilon} \frac{1}{2} m_\chi v^2 + \frac{\vec{q} \cdot \vec{v}}{1 + \epsilon} - \frac{q^2}{2m_\chi(1 + \epsilon)}, \quad (102)$$

where $\epsilon = \delta/m_\chi$. Applying these modified kinematics to Eq. (87), we find instead

$$\vec{q} \cdot \vec{f}_V = \left(\frac{\epsilon}{1 + \epsilon} \vec{q} \cdot \vec{v} - \frac{q^2}{2m_\chi} \frac{\epsilon}{1 + \epsilon} + m_\chi \epsilon - \frac{\epsilon}{1 + \epsilon} \frac{1}{2} m_\chi v^2 \right) f_S + \langle \psi_2 | e^{i\vec{q} \cdot \vec{x}} \delta V | \psi_1 \rangle. \quad (103)$$

If ψ_1 is the ground state of the SM system, the necessary energy for the $m_\chi \rightarrow m_\chi + \delta$ transition must come from the DM kinetic energy, $\propto v^2 m_\chi$, with $v^2 \sim 10^{-6}$ for the galactic DM halo. Expanding to linear order in v^2 and $\epsilon \lesssim 10^{-5}$, then,

$$\vec{q} \cdot \vec{f}_V \simeq \epsilon \left[m_\chi - \frac{q^2}{2m_\chi} + \vec{q} \cdot \vec{v} \right] f_S + \langle \psi_2 | e^{i\vec{q} \cdot \vec{x}} \delta V | \psi_1 \rangle + \mathcal{O}(\epsilon^2, \epsilon v^2). \quad (104)$$

7. Adiabaticity, parity, and isotropy

Section III C 1 has shown that when $H_0 \psi_{1,2} = E_{1,2} \psi_{1,2}$ share the same Hamiltonian, the squared matrix element [Eq. (97)] is substantially simplified, compared to the generic spin-dependent case. When this approximation can be applied to spherical or parity-symmetric systems, the complexity is reduced even further.

If parity (under central inversion, $\vec{x} \rightarrow -\vec{x}$) is a good symmetry of the detector material, however, then Sec. III A has shown that $\vec{A} = \vec{B}_6$ is real valued. In this case, the two contributions to a_3 vanish separately, leaving only a_0 and a_1 :

$$|\overline{\mathcal{M}_{1 \rightarrow 2}}|^2 = a_0 B_1 + a_1 [(v^2 - v_{\min}^2) B_1 + B_3^\perp - 2(\vec{v} \cdot \vec{B}_6^\perp)], \quad (105)$$

for a parity-symmetric, electronically adiabatic detector. If H_0 admits degenerate excited states, $H_0 \psi_2 = E_2 \psi_2$ and

$H_0 \psi'_2 = E_2 \psi'_2$, then this result holds as long as we label the final states using a basis where ψ_2 and ψ'_2 have definite parity. Otherwise, the cancellation of the a_3 term is only apparent in the sum over degenerate final states, $\sum_f |\overline{\mathcal{M}_{1 \rightarrow f}}|^2$.

Alternatively, consider the special case of an isotropic detector. As usual, we do not require that $V(\vec{x}) = V(x)$ itself is spherically symmetric, only that the material itself has no preferred directions, as in a fluid or glass. In such a system the scattering rate depends only on the isotropic average of $|\overline{\mathcal{M}_{1 \rightarrow 2}}|^2$, as in Eq. (71). Applying the rotational average to Eq. (97), we find

$$\langle |\overline{\mathcal{M}_{1 \rightarrow 2}}|^2 \rangle = (a_0 + a_1(v^2 - v_{\min}^2)) B_1^{\text{iso}} + a_1 \langle B_3^\perp \rangle + a_3 \langle B_5 \rangle, \quad (106)$$

for an isotropic, possibly chiral, electronically adiabatic detector. Finally, if we assume that our adiabatic, isotropic system is also nonchiral:

$$\langle |\overline{\mathcal{M}_{1 \rightarrow 2}}|^2 \rangle = (a_0 + a_1(v^2 - v_{\min}^2)) B_1^{\text{iso}} + a_1 \langle B_3^\perp \rangle. \quad (107)$$

Our assumption that $\psi_{1,2}$ are eigenstates of the same Hamiltonian has substantially simplified the spin-dependent form factor structure for isotropic systems, beyond any results in the literature.

If the detector is isotropic not because it is a fluid or glass, but because its constituent particles or unit cells are themselves spherically symmetric, then $B_3^\perp \rightarrow 0$. In this simplest of e^- -adiabatic cases, the generic spin-dependent scattering rate depends only on the “spin-independent” material form factor $B_1(|\vec{q}|)$,

$$\langle |\overline{\mathcal{M}_{1 \rightarrow 2}}|^2 \rangle \longrightarrow (a_0 + a_1(v^2 - v_{\min}^2)) B_1^{\text{iso}}, \quad (108)$$

up to corrections proportional to $\epsilon = \delta/m_\chi$ in the case of inelastic DM. Notably, this simplification applies to all of the spin-dependent operators shown in Fig. 1, including the couplings to the electron spin, regardless of whether the DM is spin-0, spin- $\frac{1}{2}$, or spin-1. In Table II, we provide a summary of the material response functions in the three simplifying regimes discussed in this section.

IV. CASE STUDIES

In this section, we give explicit examples of the coefficients $a_{0,\dots,4}$ as in Eq. (20) as functions of the Wilson coefficients c_i generated by three different UV models: anapole DM, Majorana DM, and scalar (inelastic) DM. The Wilson coefficients c_i are those of the effective nonrelativistic operators $\mathcal{O}_1, \dots, \mathcal{O}_{15}$ defined in Table I. Our results are summarized in Table III. In Fig. 1, we show the effective operators that are generated from scalar and fermionic DM,

TABLE II. Summary of which material response functions contribute to the rate under the symmetries or simplifying regimes discussed in Sec. III. Note that parity symmetry ensures that $\vec{B}_7$ is real; however, because $\text{Re}(\vec{B}_7)$ does not appear in Eq. (26), $\vec{B}_7$ has no contribution to the scattering rate in this case.

Material property	B_1	B_2	B_3	B_4	B_5	$\vec{B}_6$	$\vec{B}_7$
Parity symmetry $[H, P] = 0$	✓	Real	✓	B_1, B_2	✗	Real	✗
Isotropy $V(\vec{q}) = V(q)$	✓	✓	✓	✓	✓	B_2	✗
e^- adiabaticity $\hat{H}_1 = \hat{H}_2$	✓	B_1	✓	B_1	✓	Real	Imaginary

as well as which $a_{0,\dots,4}$ coefficients contribute to the response functions in Eq. (32).

A. Kinematics

Throughout the following case studies, we define p^μ and p'^μ as the incoming and outgoing four-momentum of the DM and k^μ and k'^μ as the incoming and outgoing four-momentum of the electron. In the center of momentum frame, we calculate the following kinematic quantities at linear order in $\vec{v}_{\text{el}}^\perp$:

$$\vec{p} = -\vec{k} = \mu_{\chi e} \vec{v}_{\text{el}}^\perp + \frac{\vec{q}}{2}, \quad (109)$$

$$\vec{p}' = \vec{k}' = \mu_{\chi e} \vec{v}_{\text{el}}^\perp - \frac{\vec{q}}{2}, \quad (110)$$

$$p^0 = E_p \simeq m_\chi \sqrt{\beta} + \frac{1}{\sqrt{\beta}} \frac{\mu_{\chi e}}{2m_\chi} (\vec{q} \cdot \vec{v}_{\text{el}}^\perp), \quad (111)$$

$$p'^0 = E_{p'} \simeq m_\chi \sqrt{\beta} - \frac{1}{\sqrt{\beta}} \frac{\mu_{\chi e}}{2m_\chi} (\vec{q} \cdot \vec{v}_{\text{el}}^\perp), \quad (112)$$

where

$$\beta \equiv 1 + \frac{q^2}{4m_\chi^2}. \quad (113)$$

B. Anapole dark matter

Consider as an example the Lagrangian for spin-1/2 DM coupled to a massive vector through the anapole interaction

$$\mathcal{L} \supset \frac{g_\chi}{4m_\chi^2} \bar{\chi} \gamma^\mu \gamma^5 \chi \partial^\nu F'_{\mu\nu} + g_e \bar{e} \gamma^\mu e A'_\mu. \quad (114)$$

In addition to the dark sector fermion χ (with mass m_χ), we include a vectorlike mediator A'_μ , with mass $m_{A'}$ and field strength tensor $F'_{\mu\nu} = \partial_\mu A'_\nu - \partial_\nu A'_\mu$. This DM candidate interacts through its anapole moment under a spontaneously broken dark $U(1)$ gauge group. For a DM candidate with an anapole moment under the SM electromagnetism, simply set $m_{A'} \rightarrow 0$ and replace g_e with the electric charge of an electron, $g_e \rightarrow -e$.

We start by finding the amplitude in the Feynman gauge for the relevant DM-electron scattering process $e^- \chi \rightarrow e^- \chi$:

$$i\mathcal{M} = \bar{u}_\chi(p') \left[\frac{ig_\chi}{4m_\chi^2} \gamma^\mu \gamma^5 (q_\alpha q^\alpha g_{\mu\nu} - q_\nu q_\mu) \right] u_\chi(p) \\ \times \frac{-ig^{\rho\mu}}{(p' - p)^2 - m_{A'}^2} \bar{u}_e(k') ig_e \gamma_\rho u_e(k). \quad (115)$$

Taking the nonrelativistic limit we obtain the amplitude as a function of the nonrelativistic operators listed in Table I:

$$i\mathcal{M} \approx 2ig_\chi g_e \frac{m_e^2}{q^2 + m_{A'}^2} \frac{m_e}{m_\chi} \\ \times \left[\left(1 + \frac{1}{\sqrt{\beta}} \frac{m_e}{m_\chi} \right) \frac{\mu_{\chi e}}{m_e} \left(\frac{i\vec{q}}{m_e} \cdot \vec{v}_{\text{el}}^\perp \right) \langle \mathcal{O}_{11} \rangle \right. \\ \left. + \frac{q^2}{m_e^2} \langle \mathcal{O}_8 \rangle - \frac{q^2}{m_e^2} \langle \mathcal{O}_9 \rangle \right]. \quad (116)$$

Here $\langle \mathcal{O}_i \rangle = \langle \xi_2 | \mathcal{O}_i | \xi_1 \rangle$ is the amplitude of $\mathcal{O}_i$ between initial and final spin states $\xi_{1,2}$. We define the following

TABLE III. The values of a_i , for each of the three case studies in Sec. IV. For conciseness, we define $x_e \equiv (q/m_e)^2$. The coefficients $c_1, \dots, c_{15}$ are those corresponding to the effective nonrelativistic operators $\mathcal{O}_1, \dots, \mathcal{O}_{15}$ defined in Table I, as they appear in the scattering amplitudes Eqs. (116), (129), and (142). We note that in the case of scalar *elastic* DM, all coefficients c_i are real, and the terms proportional to $c_1^\perp$ will vanish.

Model	a_0	a_1	a_2	a_3	a_4
Anapole	$\frac{1}{8} c_9 ^2 x_e$	$\frac{1}{4} c_8 ^2$	$\frac{1}{4} c_{11} ^2 x_e^2 + \frac{1}{2} \text{Re}(c_8 c_{11}^*)$	0	0
Majorana	$\frac{3}{16} c_4 ^2 + \frac{1}{8} c_9 ^2 x_e$	$\frac{1}{4} c_8 ^2$	0	0	0
Scalar Inelastic	$ c_1^0 ^2 + \frac{1}{4} c_{10} ^2 x_e$	$\frac{1}{4} c_3 ^2 x_e + \frac{1}{4} c_7 ^2$	$ c_1^\perp ^2 x_e - \frac{1}{4} c_3 ^2 x_e$	$\frac{1}{2} \text{Re}(c_3 c_7^*)$	$c_1^0 c_1^{\perp*} - \frac{1}{4} c_{10} c_7^*$

Wilson coefficients:

$$\begin{aligned} c_8 &= 2g_\chi g_e \frac{q^2}{q^2 + m_{A'}^2} \frac{m_e}{m_\chi}, \\ c_9 &= -2g_\chi g_e \frac{q^2}{q^2 + m_{A'}^2} \frac{m_e}{m_\chi}, \\ c_{11} &= 2g_\chi g_e \frac{m_e^2}{q^2 + m_{A'}^2} \frac{\mu_{\chi e}}{m_\chi} \left(1 + \frac{1}{\sqrt{\beta}} \frac{m_e}{m_\chi}\right), \end{aligned} \quad (117)$$

to obtain

$$\mathcal{M} = c_8 \langle \mathcal{O}_8 \rangle + c_9 \langle \mathcal{O}_9 \rangle + c_{11} \left(\frac{i\vec{q}}{m_e} \cdot \vec{v}_{\text{el}}^\perp \right) \langle \mathcal{O}_{11} \rangle. \quad (118)$$

We now expand the matrix element as a first-order power series in $\vec{v}_{\text{el}}^\perp$ and find

$$\mathcal{M}_S f_S = -c_9 \left\langle \vec{S}_\chi \cdot \left(\frac{i\vec{q}}{m_e} \times \vec{S}_e \right) \right\rangle f_S, \quad (119)$$

$$\vec{\mathcal{M}}_V \cdot \vec{f}_V = c_8 \langle \vec{S}_\chi \cdot \vec{f}_V \mathbb{1}_e \rangle + c_{11} \left(\frac{i\vec{q}}{m_e} \cdot \vec{f}_V \right) \left\langle \vec{S}_\chi \cdot \frac{i\vec{q}}{m_e} \mathbb{1}_e \right\rangle. \quad (120)$$

The total amplitude to first order in $\vec{v}_{\text{el}}^\perp$ is given by summing the terms:

$$\begin{aligned} \overline{|\mathcal{M}_{1 \rightarrow 2}|^2} &= \overline{|\mathcal{M}_S f_S|^2} + \overline{|\vec{\mathcal{M}}_V \cdot \vec{f}_V|^2} \\ &\quad + 2\text{Re}(\overline{\vec{\mathcal{M}}_V \cdot \vec{f}_V \mathcal{M}_S^* f_S^*}). \end{aligned} \quad (121)$$

Keeping only the terms that are linear in $\vec{v}_{\text{el}}^\perp$ yields

$$\overline{|\mathcal{M}_S f_S|^2} = \frac{1}{8} |c_9|^2 \frac{q^2}{m_e^2} |f_S|^2, \quad (122)$$

$$\begin{aligned} \overline{|\vec{\mathcal{M}}_V \cdot \vec{f}_V|^2} &= \frac{1}{4} |c_8|^2 |\vec{f}_V|^2 + \frac{1}{4} \frac{|c_{11}|^2 q^4}{q^2 + m_e^2} \left| \frac{\vec{q}}{m_e} \cdot \vec{f}_V \right|^2 \\ &\quad + \frac{1}{2} \text{Re}(c_8 c_{11}^*) \left| \frac{\vec{q}}{m_e} \cdot \vec{f}_V \right|^2, \end{aligned} \quad (123)$$

and

$$2\text{Re}(\overline{\vec{\mathcal{M}}_V \cdot \vec{f}_V \mathcal{M}_S^* f_S^*}) = 0. \quad (124)$$

The resulting a_i coefficients are listed in Table III.

C. Majorana dark matter

For our next example, we consider the case of a Majorana fermion, χ , with a vector mediator, A'^μ . The renormalizable interaction terms are

$$\mathcal{L} \supset -\frac{1}{2} \lambda_1 \bar{\chi} \gamma^\mu \gamma^5 \chi A'_\mu - h_1 \bar{e} \gamma^\mu e A'_\mu - h_2 \bar{e} \gamma^\mu \gamma^5 e A'_\mu. \quad (125)$$

We find the amplitude for DM-electron scattering in Feynman gauge to be

$$\begin{aligned} i\mathcal{M} &= \bar{u}_\chi(p') i(\lambda_1 \gamma_\mu \gamma^5) u_\chi(p) \frac{-ig^{\mu\nu}}{(p' - p)^2 - m_{A'}^2} \\ &\quad \times \bar{u}_e(k') i(h_1 \gamma_\nu + h_2 \gamma_\nu \gamma^5) u_e(k). \end{aligned} \quad (126)$$

In the nonrelativistic limit, this reduces to

$$i\mathcal{M} \approx -i \frac{8m_\chi m_e}{q^2 + m_{A'}^2} [\lambda_1 h_1 \langle \mathcal{O}_8 \rangle - \lambda_1 h_1 \langle \mathcal{O}_9 \rangle - 2\lambda_1 h_2 \langle \mathcal{O}_4 \rangle]. \quad (127)$$

Next, we define the following Wilson coefficients:

$$\begin{aligned} c_4 &= \frac{16m_\chi m_e}{q^2 + m_{A'}^2} \lambda_1 h_2, \\ c_8 &= -\frac{8m_\chi m_e}{q^2 + m_{A'}^2} \lambda_1 h_1, \\ c_9 &= \frac{8m_\chi m_e}{q^2 + m_{A'}^2} \lambda_1 h_1 \end{aligned} \quad (128)$$

to obtain

$$\mathcal{M} = c_4 \langle \mathcal{O}_4 \rangle + c_8 \langle \mathcal{O}_8 \rangle + c_9 \langle \mathcal{O}_9 \rangle. \quad (129)$$

This yields

$$\mathcal{M}_S f_S = c_4 \langle \vec{S}_\chi \cdot \vec{S}_e \rangle f_S - c_9 \left\langle \vec{S}_\chi \cdot \left(\frac{i\vec{q}}{m_e} \times \vec{S}_e \right) \right\rangle f_S \quad (130)$$

and

$$\vec{\mathcal{M}}_V \cdot \vec{f}_V = c_8 \langle \vec{S}_\chi \cdot \vec{f}_V \mathbb{1}_e \rangle. \quad (131)$$

Thus, we have

$$\overline{|\mathcal{M}_S f_S|^2} = \frac{3}{16} |c_4|^2 |f_S|^2 + \frac{1}{8} |c_9|^2 \frac{q^2}{m_e^2} |f_S|^2, \quad (132)$$

$$\overline{|\vec{\mathcal{M}}_V \cdot \vec{f}_V|^2} = \frac{1}{4} |c_8|^2 |\vec{f}_V|^2, \quad (133)$$

and

$$2\text{Re}(\overline{\vec{\mathcal{M}}_V \cdot \vec{f}_V \mathcal{M}_S^* f_S^*}) = 0. \quad (134)$$

The resulting a_i coefficients are listed in Table III.

D. Scalar (inelastic) dark matter

Next, we study a case that populates all a_i coefficients, including the imaginary part of a_4 . In this model the DM is a complex scalar S_1 with mass m_S , but there is another dark scalar S_2 with mass $m_S + \delta$, with a small mass splitting $\delta \ll m_S$. Coupling the dark scalars to the SM electron via a scalar mediator ϕ and a vector mediator A'_μ , the allowed interactions are

$$\begin{aligned} \mathcal{L} \supset & -g_1^* m_S S_1^\dagger S_2 \phi - g_1 m_S S_2^\dagger S_1 \phi \\ & -ig_2 S_1^\dagger (\partial_\mu S_2) A'^\mu + ig_2^* (\partial_\mu S_2^\dagger) S_1 A'^\mu \\ & -ig_3 S_2^\dagger (\partial_\mu S_1) A'^\mu + ig_3^* (\partial_\mu S_1^\dagger) S_2 A'^\mu \\ & -ih_2 \bar{e} \gamma^5 e \phi - h_1 \bar{e} e \phi - h_3 (\bar{e} \gamma_\mu e) A'^\mu \\ & -h_4 (\bar{e} \gamma_\mu \gamma^5 e) A'^\mu. \end{aligned} \quad (135)$$

We allow the $U(1)_D$ gauge symmetry to be broken both by the mass term of the A'_μ and by the couplings g_2, g_3 of the DM to the A'_μ . In Feynman gauge, we find the scattering amplitude for $S_1 e^- \rightarrow S_2 e^-$ to be

$$\begin{aligned} i\mathcal{M} = & (ig_1 m_S) \left(\frac{i}{(p' - p)^2 - m_\phi^2} \right) \\ & \times \bar{u}'(k') i(h_1 + ih_2 \gamma^5) u^r(k) \\ & + (ig_3 p_\mu + ig_2^* p'_\mu) \left(\frac{-ig^{\mu\nu}}{q^2 - m_{A'}^2} \right) \\ & \times \bar{u}'(k') (ih_3 \gamma_\nu + ih_4 \gamma_\nu \gamma^5) u^r(k). \end{aligned} \quad (136)$$

For inelastic DM, $\vec{v}_{\text{el}}^\perp$ is no longer Galilean invariant as defined in Eq. (7). Here we define a new Galilean invariant velocity,

$$\vec{v}_{\text{in}}^\perp = \frac{\vec{p}}{2m_S} + \frac{\vec{p}'}{2(m_S + \delta)} - \frac{\vec{k} + \vec{k}'}{2m_e}. \quad (137)$$

We remark that $\vec{q}$ itself is not Galilean invariant. Since positive δ is kinematically constrained to be $\delta \sim O(v^2)$, focusing on this case allows us to neglect terms appearing at order $O(v^3)$ that break the Galilean symmetry.

In the center of momentum frame, the previously defined kinematic quantities receive the following corrections at leading order in $\epsilon = \delta/m_S$:

$$\vec{p} = -\vec{k} = \mu_{Se} \vec{v}_{\text{in}}^\perp + \frac{\vec{q}}{2} \left(1 + \epsilon \frac{\mu_{Se}}{m_S} \right), \quad (138)$$

$$\vec{p}' = \vec{k}' = \mu_{Se} \vec{v}_{\text{in}}^\perp - \frac{\vec{q}}{2} \left(1 - \epsilon \frac{\mu_{Se}}{m_S} \right), \quad (139)$$

$$p^0 = E_p \simeq m_S \sqrt{\beta} + \frac{q^2 \mu_{Se}}{4\sqrt{\beta} m_S^2} \epsilon + \frac{1}{\sqrt{\beta}} \frac{\mu_{Se}}{2m_S} (\vec{q} \cdot \vec{v}_{\text{in}}^\perp), \quad (140)$$

$$p'^0 = E_{p'} \simeq m_S \sqrt{\beta} - \frac{q^2 \mu_{Se}}{4\sqrt{\beta} m_S^2} \epsilon - \frac{1}{\sqrt{\beta}} \frac{\mu_{Se}}{2m_S} (\vec{q} \cdot \vec{v}_{\text{in}}^\perp). \quad (141)$$

Thus, in the nonrelativistic limit, we find

$$\begin{aligned} \mathcal{M} \simeq & \left[c_1^0 + c_1^\perp \left(\frac{i\vec{q}}{m_e} \cdot \vec{v}_{\text{in}}^\perp \right) \right] \langle \mathcal{O}_1 \rangle + c_3 \langle \mathcal{O}_3 \rangle \\ & + c_7 \langle \mathcal{O}_7 \rangle + c_{10} \langle \mathcal{O}_{10} \rangle, \end{aligned} \quad (142)$$

where the Wilson coefficients are given by

$$\begin{aligned} c_1^0 = & \frac{2m_S m_e}{q^2 + m_\phi^2} g_1 h_1 - \frac{m_S m_e}{q^2 + m_{A'}^2} h_3 \left[2(g_3 + g_2^*) \sqrt{\beta} \right. \\ & \left. + (g_3 - g_2^*) \epsilon \frac{q^2 \mu_{Se}}{2m_S^2 m_e} \left(1 + \frac{m_e}{\sqrt{\beta} m_S} \right) \right], \end{aligned} \quad (143)$$

$$c_1^\perp = \frac{im_S m_e}{q^2 + m_{A'}^2} h_3 (g_3 - g_2^*) \frac{\mu_{Se}}{m_S} \left(\frac{m_e}{m_S \sqrt{\beta}} + 1 \right), \quad (144)$$

$$c_3 = \frac{2m_S m_e}{q^2 + m_{A'}^2} h_3 (g_3 + g_2^*) \frac{\mu_{Se}}{m_S}, \quad (145)$$

$$c_7 = \frac{4m_S m_e}{q^2 + m_{A'}^2} h_4 (g_3 + g_2^*) \frac{\mu_{Se}}{m_S} \left(\frac{m_S}{m_e} \sqrt{\beta} + 1 \right), \quad (146)$$

$$\begin{aligned} c_{10} = & \frac{-2m_S m_e}{q^2 + m_\phi^2} g_1 h_2 - 2ih_4 \frac{m_S m_e}{q^2 + m_{A'}^2} \left[(g_3 + g_2^*) \epsilon \frac{\mu_{Se} m_e}{m_S m_S} \right. \\ & \left. \times \left(1 + \frac{m_S}{m_e} \sqrt{\beta} \right) + (g_3 - g_2^*) \frac{m_e}{m_S} \right]. \end{aligned} \quad (147)$$

This yields

$$\mathcal{M}_S f_S \simeq \left(c_1^0 \langle \mathbb{1}_S \mathbb{1}_e \rangle + c_{10} \left\langle \mathbb{1}_S \frac{i\vec{q}}{m_e} \cdot \vec{S}_e \right\rangle \right), \quad (148)$$

$$\begin{aligned} \vec{\mathcal{M}}_V \cdot \vec{f}_V \simeq & c_1^\perp (\vec{q} \cdot \vec{f}_V) \langle \mathbb{1}_S \mathbb{1}_e \rangle + c_3 \left\langle \mathbb{1}_S \left(\frac{i\vec{q}}{m_e} \times \vec{f}_V \right) \cdot \vec{S}_e \right\rangle \\ & + c_7 \langle \mathbb{1}_S \vec{f}_V \cdot \vec{S}_e \rangle, \end{aligned} \quad (149)$$

such that

$$|\overline{\mathcal{M}_S f_S}|^2 = \left(|c_1^0|^2 + \frac{|c_{10}|^2}{4} \frac{\vec{q}^2}{m_e^2} \right) |f_S|^2, \quad (150)$$

$$\begin{aligned} |\overline{\vec{\mathcal{M}}_V \cdot \vec{f}_V}|^2 = & \left(|c_1^\perp|^2 - \frac{|c_3|^2}{4} \right) \left| \frac{\vec{q}}{m_e} \cdot \vec{f}_V \right|^2 \\ & + \frac{1}{4} \left(|c_3|^2 \frac{\vec{q}^2}{m_e^2} + |c_7|^2 \right) |\vec{f}_V|^2 \\ & + i \frac{1}{2} \text{Re}(c_3 c_7^*) \frac{\vec{q}}{m_e} \cdot (\vec{f}_V \times \vec{f}_V^*), \end{aligned} \quad (151)$$

TABLE IV. Starting from the generic scalar inelastic DM model, we show how the real and/or imaginary parts of a_4 can vanish in certain limits. If g_2, g_3 are complex and $\epsilon \neq 0$, all four coefficients $c_1^0, c_1^\perp, c_7, c_{10}$ are complex, leading to complex a_4 in both cases (i) and (ii). Note that if the vector mediator is removed, then a_4 is identically zero in this model.

Scalar inelastic DM		c_1^0	$c_1^\perp$	c_7	c_{10}	a_4
(i) Scalar and vector mediators present	g_2, g_3 real	real	imaginary	real	complex	complex
	elastic ($\epsilon \rightarrow 0, g_3 \rightarrow g_2$)	real	real	real	real	real
	g_2, g_3 real and elastic	real	zero	real	real	real
(ii) Only vector mediator present ($h_1 = h_2 = 0$)	g_2, g_3 real	real	imaginary	real	imaginary	imaginary
	elastic ($\epsilon \rightarrow 0, g_3 \rightarrow g_2$)	real	real	real	real	real
	g_2, g_3 real and elastic	real	zero	real	zero	zero

and

$$2\text{Re}(\mathcal{M}_S f_S (\mathcal{M}_V \cdot \vec{f}_V)^*) \\ = 2\text{Im} \left[\left(c_1^0 c_1^{\perp*} - \frac{1}{4} c_{10} c_7^* \right) f_S \left(\frac{\vec{q}}{m_e} \cdot \vec{f}_V^* \right) \right]. \quad (152)$$

In general, a_4 is complex. In particular, the phase in a_4 is not simply generated from g_2, g_3 being complex since even if g_2 and g_3 were both real, a relative phase would still be present in c_{10} . In the case of elastic scattering, $\epsilon \rightarrow 0$, and since we have only one species of DM, $g_3 \rightarrow g_2$, so a_4 becomes real. For $a_4 \neq 0$, (i) both scalar and vector mediators should be present, or (ii) if only the vector mediator is present, at least one of g_2, g_3 should be complex or DM should be inelastic, as can be seen in Eqs. (143)–(147). The different possibilities are summarized in Table IV.

Finally, we emphasize that both the electron vector and axial vector currents must couple to the spin-1 mediators in order to generate a_3 .

V. DISCUSSION AND CONCLUSIONS

In this work, we have extended the general framework for computing spin-dependent DM scattering rates beyond the conventional assumption of isotropic detector materials, establishing a consistent set of conventions applicable to anisotropic targets. We find that the most general squared amplitude governing nonrelativistic DM-SM interactions can be expressed in terms of the seven distinct material response functions and five DM response functions that appear in Eq. (26). Despite the fact that these material response functions depend entirely on SM physics, it is often computationally challenging to calculate them from first principles. Under common simplifying conditions, namely parity conservation, isotropy, or electronic adiabaticity, we have uncovered new relations between these functions, reducing the computational burden of the spin-dependent scattering rate prediction.

While a reduction in computational effort is always welcome, our simplified form of the spin-dependent scattering cross section from Eq. (32) also provides valuable

insights for the design of future detectors. For example, a parity-symmetric detector medium is completely insensitive to the a_3 DM response function. A pair of direct detection experiments with chiral and achiral detector targets would be able to isolate the a_3 component of a DM signal. Similarly, as we demonstrated in Eq. (97), the a_2 and a_4 contributions to the rate both vanish in electronically adiabatic systems: to detect DM scattering through these DM response functions, a detector medium must have substantial difference between the Coulomb potentials in its initial and final states.

Altogether, our results provide a systematic foundation for incorporating anisotropic material effects into DM direct detection calculations, and a road map for connecting microscopic DM models to experimentally accessible observables.

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DATA AVAILABILITY

No data were created or analyzed in this study.

APPENDIX: EXPLICIT COMPUTATION OF FORM FACTORS

In this appendix, we verify explicitly that the electronic adiabaticity relation in Eq. (85), $\vec{q} \cdot \vec{f}_{1 \rightarrow 2} = [\Delta E - q^2/(2m_e)]f_S$ with $\Delta E = E_2 - E_1$, holds for a particle-in-a-box model and the hydrogen atom.

1. Particle in a box

For a rectangular box of dimensions (L_x, L_y, L_z) and volume $V = L_x L_y L_z$, the eigenstates are given by

$$\psi_{\vec{n}} = \frac{2^{3/2}}{\sqrt{V}} \sin \frac{\pi n_x x}{L_x} \sin \frac{\pi n_y y}{L_y} \sin \frac{\pi n_z z}{L_z} \quad (\text{A1})$$

where $\vec{n} = (n_x, n_y, n_z)$, with $n_i \in \mathbb{N}$, labels the possible excited states starting $(1, 1, 1)$ as the ground state. If ψ_1 is the ground state and ψ_2 is a state with arbitrary $\vec{n}$, then the excited state energy is given by

$$\Delta E = \frac{\pi^2}{2m_e} \left(\frac{n_x^2 - 1}{L_x^2} + \frac{n_y^2 - 1}{L_y^2} + \frac{n_z^2 - 1}{L_z^2} \right). \quad (\text{A2})$$

Since the integral for the scalar form factor factorizes, we can write

$$f_S = f_x(q_x) f_y(q_y) f_z(q_z) \quad (\text{A3})$$

where

$$f_j(q_j) = \int_0^L dx_j e^{iq_j x_j} \frac{2}{L_j} \sin \left(\frac{\pi n_j x_j}{L_j} \right) \sin \left(\frac{\pi x_j}{L_j} \right) \quad (\text{A4})$$

for $j = x, y, z$. We compute each $f_j(q_j)$ and find

$$f_j(q_j) = -i4\pi^2 n_j q_j L_j \times \frac{[1 + (-1)^{n_j} e^{iq_j L_j}]}{[(q_j L_j)^2 - \pi^2(n_j - 1)^2][(q_j L_j)^2 - \pi^2(n_j + 1)^2]}. \quad (\text{A5})$$

Similarly, the vector form factor factorizes

$$\vec{f}_{1 \rightarrow 2} = f_{\vec{\nabla}}^{(x)}(q_x) f_y(q_y) f_z(q_z) \hat{x} + f_x(q_x) f_{\vec{\nabla}}^{(y)}(q_y) f_z(q_z) \hat{y} + f_x(q_x) f_y(q_y) f_{\vec{\nabla}}^{(z)}(q_z) \hat{z}, \quad (\text{A6})$$

where

$$f_{\vec{\nabla}}^{(j)}(q_j) = -\frac{i}{m_e} \int_0^L dx_j e^{iq_j x_j} \frac{2}{L_j} \frac{\pi}{L_j} \sin \left(\frac{\pi n_j x_j}{L_j} \right) \cos \left(\frac{\pi x_j}{L_j} \right), \quad (\text{A7})$$

resulting in

$$f_{\vec{\nabla}}^{(j)}(q_j) = i \frac{2\pi^2 n_j}{m_e L_j} \frac{[1 + (-1)^{n_j} e^{iq_j L_j}][(q_j L_j)^2 - \pi^2(n_j^2 - 1)]}{[(q_j L_j)^2 - \pi^2(n_j - 1)^2][(q_j L_j)^2 - \pi^2(n_j + 1)^2]}. \quad (\text{A8})$$

Noting that

$$\frac{f_{\vec{\nabla}}^{(j)}(q_j)}{f_j(q_j)} = \frac{1}{2m_e q_j L_j^2} [\pi^2(n_j^2 - 1) - (q_j L_j)^2] \quad (\text{A9})$$

it is then straightforward to verify the relation $\vec{q} \cdot \vec{f}_{1 \rightarrow 2} = [\Delta E - q^2/(2m_e)] f_S$ holds.

2. Hydrogen atom

Here we consider the $1/r$ potential of the hydrogen atom. Using the formulas in the appendixes of [8,63], the form factors for the hydrogen atom can be found for a transition from an initial state $|n, l, m\rangle$ to a final state $|n', l', m'\rangle$. In order for this section to be self-contained, we recall here the expressions for the form factors:

$$f_S = \sqrt{4\pi} \sum_{L=|l-l'|}^{l+l'} i^L I_1^L \sum_{M=-L}^L Y_L^{M*} (-1)^{m'} \sqrt{(2l+1)(2l'+1)(2L+1)} \begin{pmatrix} l & l' & L \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} l & l' & L \\ m & -m' & M \end{pmatrix}, \quad (\text{A10})$$

$$\vec{f}_{1 \rightarrow 2} = -i \frac{\sqrt{4\pi}}{m_e} \sum_{i=1}^3 \hat{e}_i \sum_{\hat{l}=l-1}^{l+1} \sum_{\hat{m}=m-1}^{m+1} \sum_{L=0}^{\infty} \sum_{M=-L}^L Y_L^{M*} i^L (-1)^{m'} \sqrt{(2\hat{l}+1)(2l'+1)(2L+1)} \times \begin{pmatrix} \hat{l} & l' & L \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} \hat{l} & l' & L \\ \hat{m} & -m' & M \end{pmatrix} (c_{\hat{l}\hat{m}}^{(i)} I_2^L + d_{\hat{l}\hat{m}}^{(i)} I_3^L), \quad (\text{A11})$$

where Y_L^M are the spherical harmonics and $c_{\hat{l}\hat{m}}^{(i)}$ and $d_{\hat{l}\hat{m}}^{(i)}$ are both constants that only depend on the summation indices.⁶ The energy difference between the states is given by

$$\Delta E = -\frac{1}{2m_e a^2} \left(\frac{1}{n'^2} - \frac{1}{n^2} \right) \quad (\text{A12})$$

where m_e is the electron mass and a is the Bohr radius.

⁶Note that there is a relative minus sign difference between the formula in Ref. [8] and the definition in Eq. (21).

For a central potential, the radial integrals are factorized. Reference [8] gives the following expressions:

$$I_1^L = \int_0^\infty dr r^2 R_{n'l'}^* R_{nl} j_L(qr), \quad (\text{A13})$$

$$I_2^L = \int_0^\infty dr r^2 R_{n'l'}^* \frac{dR_{nl}}{dr} j_L(qr), \quad (\text{A14})$$

$$I_3^L = \int_0^\infty dr r R_{n'l'}^* R_{nl} j_L(qr), \quad (\text{A15})$$

where L is the angular momentum (e.g., for the scalar form factor $L = |l - l'|, \dots, l + l'$). Due to the form of hydrogen radial wave functions, we can write

$$I_1^L = \sum_{k=l+l'}^{(n-1)+(n'-1)} \alpha_k^{nl,n'l'} \int_0^\infty dr r^2 \left(\frac{r}{a}\right)^k e^{-r/(pa)} j_L(qr), \quad (\text{A16})$$

$$I_2^L = \sum_{\substack{k=l+l'-1 \\ k \geq 0}}^{(n-1)+(n'-1)} \beta_k^{nl,n'l'} \int_0^\infty dr r^2 \left(\frac{r}{a}\right)^k e^{-r/(pa)} j_L(qr), \quad (\text{A17})$$

$$I_3^L = \sum_{k=l+l'-1}^{(n-1)+(n'-1)-1} \gamma_k^{nl,n'l'} \int_0^\infty dr r^2 \left(\frac{r}{a}\right)^k e^{-r/(pa)} j_L(qr), \quad (\text{A18})$$

with $p = (1/n + 1/n')^{-1}$. Here, $\alpha_k, \beta_k, \gamma_k$ are coefficients that depend only on n, l, n', l' , and a . We will suppress the superscripts $nl, n'l'$ henceforth for ease of notation. I_1^L is the radial integral for the scalar form factor f_S whereas the vector form factor has contributions from both I_2^L and I_3^L . The building block for these integrals is analytically computed to be

$$\int_0^\infty dr r^2 \left(\frac{r}{a}\right)^k e^{-r/(pa)} j_L(qr) = h_k^L {}_2F_1\left(\frac{L-k}{2}, \frac{L-k-1}{2}, \frac{3}{2} + L, -X\right) \quad (\text{A19})$$

with ${}_2F_1$ the hypergeometric function, $X = a^2 p^2 q^2$, and where we define the coefficient h_k^L to be

$$h_k^L \equiv 2^{-1-L} p^{3+L+k} a^{3+L} \sqrt{\pi} \frac{q^L}{(1+X)^{2+k}} \frac{\Gamma(3+L+k)}{\Gamma(3/2+L)}. \quad (\text{A20})$$

In the following, ϕ is the azimuthal angle of $\vec{q}$ and we employ the notation $c_\theta \equiv \cos \theta$ and $s_\theta \equiv \sin \theta$ where θ is the polar angle of the vector $\vec{q}$. As an example, taking $n' = 3, l' = 2, m' = 2$, and $n = 2, l = 1, m = 1$, the radial wave functions are

$$R_{21} = \mathcal{N}_{21} a^{-3/2} \frac{r}{a} e^{-r/(2a)}, \quad (\text{A21})$$

$$R_{32} = \mathcal{N}_{32} a^{-3/2} \left(\frac{r}{a}\right)^2 e^{-r/(3a)}, \quad (\text{A22})$$

with $\mathcal{N}_{21} = 1/(2\sqrt{6})$ and $\mathcal{N}_{32} = 4/(81\sqrt{30})$. The non-zero coefficients are given by

$$\alpha_3 = \mathcal{N}_{21} \mathcal{N}_{32} a^{-3}, \quad (\text{A23})$$

$$\beta_2 = \alpha_3/a, \quad (\text{A24})$$

$$\beta_3 = -\frac{1}{2} \alpha_3/a, \quad (\text{A25})$$

$$\gamma_2 = \alpha_3/a. \quad (\text{A26})$$

We find for the scalar form factor

$$f_S = \sqrt{4\pi} \alpha_3 \sum_{L=1}^3 h_3^L i^L \sqrt{15(2L+1)} \begin{pmatrix} 1 & 2 & L \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} 1 & 2 & L \\ 1 & -2 & 1 \end{pmatrix} Y_L^{1*} {}_2F_1\left(\frac{L-3}{2}, \frac{L-4}{2}, \frac{3}{2} + L, -X\right) \quad (\text{A27})$$

where $p = 6/5$. Here the $L = 2$ term will not contribute to the summation as the Wigner $3-j$ symbol evaluates to 0. We simplify each of the components of the vector form factor in turn. We obtain for the x, y components

$$f_{1 \rightarrow 2, (x,y)} = i \frac{\sqrt{4\pi}}{m_e} (Y_2^2)^* h_2^2 \left[(c_{00}^{(x,y)} \beta_2 + d_{00}^{(x,y)} \gamma_2) + c_{00}^{(x,y)} \beta_3 \frac{p}{(1+X)} (7-X) \right] \quad (\text{A28})$$

$$- \left[\frac{i\sqrt{4\pi}}{m_e} \sum_{\hat{m}=0,2} \sum_{L=0,2,4} g^L(\hat{m}) h_2^L (Y_L^{2-\hat{m}})^* c_{2\hat{m}}^{(x,y)} \beta_3 \frac{p}{(1+X)} (5+L) {}_2F_1\left(\frac{L-3}{2}, \frac{L-4}{2}, \frac{3}{2} + L, -X\right) \right] \quad (\text{A29})$$

where the first term comes from the $\hat{l} = 0, L = 2, M = 2$ term and the rest is from the $\hat{l} = 2$ portion of Eq. (A11). Similarly, the z component is given by

$$f_{1 \rightarrow 2, z} = -i \frac{\sqrt{4\pi}}{m_e} \sum_{L=2,4} (Y_L^1)^* g^L(1) h_2^L c_{21}^{(z)} \beta_3 \frac{p}{(1+X)} (5+L) {}_2F_1 \left(\frac{L-3}{2}, \frac{L-4}{2}, \frac{3}{2} + L, -X \right) \quad (\text{A30})$$

where we have also used that $c_{2\hat{m}}^{(x,y)} \beta_2 + d_{2\hat{m}}^{(x,y)} \gamma_2 = c_{21}^{(z)} \beta_2 + d_{21}^{(z)} \gamma_2 = 0$ for $\hat{m} = 0, 2$. Evaluating the Wigner $3j$ symbols for these values of L , $\hat{m}$ gives

$$g^L(\hat{m}) = (-1)^{\hat{m}} 2\sqrt{6}(L^3 + 3L^2 - 10L - 24) \frac{(L-1)}{(4-L)!(5+L)!} \sqrt{\frac{(2+L-\hat{m})!(2+\hat{m})!}{(2-\hat{m})!(L-2+\hat{m})!}} i^L 5\sqrt{(2L+1)} \quad (\text{A31})$$

while the $c_{i\hat{m}}^{(i)}$, $d_{i\hat{m}}^{(i)}$ coefficients are defined as in Ref. [8]:

$$c_{00}^{(x)} = -i c_{00}^{(y)} = -\frac{1}{\sqrt{6}}, \quad (\text{A32})$$

$$d_{00}^{(x)} = -i d_{00}^{(y)} = -\sqrt{\frac{2}{3}}, \quad (\text{A33})$$

$$c_{20}^{(x)} = -i c_{20}^{(y)} = \frac{1}{\sqrt{30}}, \quad (\text{A34})$$

$$c_{22}^{(x)} = i c_{22}^{(y)} = -\frac{1}{\sqrt{5}}, \quad (\text{A35})$$

$$c_{21}^{(z)} = \frac{1}{\sqrt{5}}. \quad (\text{A36})$$

For $L \leq 4$, the hypergeometric function evaluates to a polynomial. This allows us to make the replacement

$${}_2F_1 \left(\frac{L-3}{2}, \frac{L-4}{2}, \frac{3}{2} + L, -X \right) \rightarrow 1 - X \frac{(L-4)(L-3)[20 + 8L - X(L-1)(L-2)]}{8(3+2L)(5+2L)} \quad (\text{A37})$$

in Eqs. (A27), (A28), and (A30).

Numerically, the scalar form factor evaluates to

$$f_S = -\frac{829440 i a q e^{-i\phi} s_\theta (36a^2 q^2 c_{2\theta} + 25)}{(36a^2 q^2 + 25)^5} \quad (\text{A38})$$

whereas the results in Cartesian coordinates for the vector form factor are given by

$$f_{1 \rightarrow 2, x} = -\frac{2304i[(25 - 18a^2 q^2)^2 + 36a^2 q^2 e^{-2i\phi} (3e^{2i\phi} (9a^2 q^2 c_{4\theta} + 25c_{2\theta}) + 36a^2 q^2 s_\theta^2 (1 - 3c_{2\theta}) - 50s_\theta^2)]}{am_e (36a^2 q^2 + 25)^5} \quad (\text{A39})$$

$$f_{1 \rightarrow 2, y} = -i f_{1 \rightarrow 2, x} - 331776 a q^2 e^{-2i\phi} \frac{[25s_\theta^2 - 18a^2 q^2 s_\theta^2 (1 - 3c_{2\theta})]}{m_e (36a^2 q^2 + 25)^5} \quad (\text{A40})$$

$$f_{1 \rightarrow 2, z} = \frac{248832 i a q^2 e^{-i\phi} s_{2\theta} (36a^2 q^2 c_{2\theta} + 25)}{m_e (36a^2 q^2 + 25)^5}. \quad (\text{A41})$$

This gives

$$\vec{q} \cdot \vec{f}_{1 \rightarrow 2} = 11520 i q s_\theta e^{-i\phi} \frac{(36a^2 q^2 - 5)(36a^2 q^2 c_{2\theta} + 25)}{am_e (36a^2 q^2 + 25)^5} \quad (\text{A42})$$

$$= \left(\frac{5}{72a^2 m_e} - \frac{q^2}{2m_e} \right) \left(-\frac{829440 i a q e^{-i\phi} s_\theta (36a^2 q^2 c_{2\theta} + 25)}{(36a^2 q^2 + 25)^5} \right) \quad (\text{A43})$$

$$= \left[\Delta E - \frac{q^2}{2m_e} \right] f_S, \quad (\text{A44})$$

satisfying Eq. (85). Similarly, we can verify that the relation holds for other initial and final states.

a. Parity

We also verify the statement made in Sec. III A namely that if $\psi_{1,2}$ have matching parity, then f_S is purely real whereas if $\psi_{1,2}$ have opposite parities, then f_S is purely imaginary.

As an example, we will take $|\psi_2\rangle = |3, 2, 1\rangle + |3, 2, -1\rangle$ (parity even) and $|\psi_1\rangle = |2, 1, 1\rangle + |2, 1, -1\rangle$ (parity odd). This gives

$$f_S = i \frac{1658880 a q c_\theta [72 a^2 q^2 (2 s_\theta^2 c_{2\theta} + c_{2\theta}) - 36 a^2 q^2 + 25]}{(36 a^2 q^2 + 25)^5} \quad (\text{A45})$$

which is purely imaginary.

On the contrary, if we take $|\psi_2\rangle = |3, 1, 1\rangle + |3, 1, -1\rangle$ (parity odd) keeping ψ_1 the same, we obtain

$$f_S = \frac{9216 [-54 a^2 q^2 (36 a^2 q^2 - 175) (2 s_\theta^2 c_{2\theta} + c_{2\theta}) + 648 a^4 q^4 - 5850 a^2 q^2 + 3125]}{\sqrt{5} (36 a^2 q^2 + 25)^5} \quad (\text{A46})$$

which is purely real. We also note that both of the above examples satisfy the relation of Eq. (42).

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