

Hydrogenated carbon structures as directional sub-GeV dark matter detectors

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We propose hydrogenated carbon structures as targets with a remarkable sensitivity to dark matter–nucleon interactions, in the mass range between 1 MeV and 100 MeV. The ejection of a proton following the interaction with a dark matter particle is a quasielastic process, with an extremely small energy threshold and a clear experimental signature. The proposed detectors are simple, technologically ready, and inexpensive. Yet, they can be considerably more sensitive than current experiments. They also allow strong directionality to be used toward efficient background rejection.

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Introduction. Despite being one of the most compelling problems in fundamental physics, the search for weakly interacting massive dark matter particles (WIMPs) has so far only reported null results [e.g., [1–8]]. This has fueled several efforts toward finding new ways to probe yet unexplored regions both in mass and couplings, as suggested by a number of theoretical models [e.g., [9–17]]. In particular, sub-GeV dark matter particles cannot be detected in elastic scatterings with most of the detector targets, because of the large mass mismatch with the recoiling nuclei. Those experiments that are currently looking into the MeV-to-GeV range, or propose to do so, circumvent this limitation by trying to detect inelastic processes [e.g., [17–19]], such as phonon emission in various materials [e.g., [2,7,20–26]] or the Migdal effect [e.g., [5,27–29]], which, however, have much smaller rates, or by introducing light nuclei such as hydrogen in their targets [e.g., [30–33]].

We propose the use of *hydrogenated* carbon structures, such as graphene, graphite or nanotubes (CNTs), to search for dark matter particles interacting with nucleons. Indeed, when a sub-GeV dark matter particle impinges on the proton belonging to one of the hydrogen atoms, it can

exchange enough energy to release it via an almost *elastic* process. Once emitted in a vacuum enclosure, the charged proton can be accelerated by an electric field, and eventually be collected. This way, the proposed concept does not rely on the detectability of a tiny nuclear recoil energy, but rather on the possibility of accelerating a charged particle up to detectable energies. Consequently, such a detector would have an extremely low energy threshold: as the typical binding energy of a proton onto a carbon structure is of the order of a few eVs [e.g., [34–36]], dark matter particles with masses $m_\chi \sim \mathcal{O}(\text{MeV})$ can eject protons with sizable rates. In the case of vertically aligned CNTs, the expected signal is directional, thus providing a strong handle for background rejection. Graphene and CNTs have been studied both theoretically and experimentally to probe dark matter–electron interactions [37–42], constrain the neutrino mass, and hunt for the elusive relic neutrinos [e.g., [34,36,43]].

The technology behind the hydrogenation of carbon structures is well assessed [e.g., [44–48]], making it possible to engineer the supporting graphene holder in order to optimize it toward the present proposal. Moreover, the proton binding energy can be varied by manipulating the hydrogenation level and/or the local geometry of the substrate [e.g., [34–36]]. Hydrogenated graphene is also stable at room temperature [49], making this detection scheme practically insensitive to thermal effects. This means that it can fit in a relatively small high-vacuum chamber, without the need for expensive infrastructures, such as cryostats. In the proposed detector, the ejected proton emerges from an anode, which is kept at a positive voltage of a few kVs. It can then be easily directed toward a sensor by a suitably shaped electric field. This sensor,

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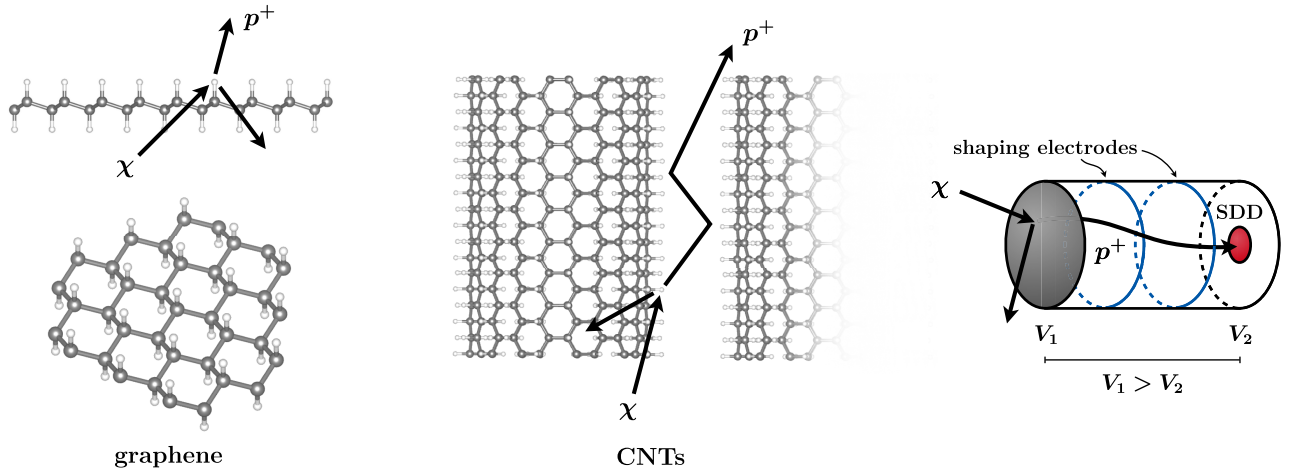


FIG. 1. *Left and central panels:* Schematic representation of the hydrogenated carbon structures at 100% loading, and a pictorial dark matter interaction on a hydrogen nucleus, resulting in the ejection of a proton. Dark and light spheres represent carbon and hydrogen atoms, respectively, with the latter being covalently bound to their carbon site. *Right panel:* Detector concept. After its ejection, the proton is accelerated by an electric field, and its trajectory is focused by shaping electrodes, until it is detected by an SDD.

having a sensitive area much smaller than the hydrogenated substrate, can be a single-channel silicon drift detector (SDD). A SDD can also measure the accelerated proton energy with a resolution sufficient to separate signal from background [50]. (See the Conclusion for a discussion on this.) Such a technique has already been implemented for the collection and detection of electrons, with large efficiencies of the order of 50% [51]. Moreover, the components described above are all readily available technologies, further strengthening the case for the possibility of realizing this idea in the near future, and for a relatively contained cost. The hydrogenated carbon structures considered here, as well as the proposed experimental concept, are shown in Fig. 1.

We consider a simple benchmark model of spin-independent dark matter–nucleon interactions, and we employ density functional theory (DFT) [e.g., [52,53]] to determine the expected rate for the liberation of a proton from the substrate. We do this for both graphene and CNTs. The latter, in addition to offering substantially more target mass per unit area, are expected to exhibit directional sensitivity. In this case, however, one must also estimate the probability that the proton emitted by the dark matter scattering is transmitted to the exterior of the forest, without being lost or captured by the material itself. We do that with a suitably designed Monte Carlo simulation. In all these instances, we find that the proposed materials can outperform by orders of magnitude the best current experimental constraints [e.g., [5,29]].

The dark matter–proton collision. We consider a benchmark model of fermionic dark matter interacting with protons via a spin-independent interaction mediated by a heavy scalar, $\mathcal{L}_{\text{int}} = g_\chi \phi \bar{\chi} \chi + g_p \phi \bar{p} p$, where we assume $m_\phi \gg m_\chi v_\chi$, with v_χ being the typical dark matter velocity.

From the quantum-mechanical viewpoint, this is matched to a contact interaction potential,¹

$$U(\mathbf{x}_\chi, \mathbf{x}_p) = -\frac{g_\chi g_p}{m_\phi^2} \delta(\mathbf{x}_\chi - \mathbf{x}_p). \quad (1)$$

While we choose this model for concreteness and computational simplicity, we stress that a direct laboratory search is completely model independent from the viewpoint of a specific dark matter model, as well as that of its cosmological history.

The initial-state proton is bound to the carbon structure, and it is thus necessary to account for its wave function and binding energy. Realistic CNTs have a radius of order 10 nm, much larger than the size of a unit cell. Consequently, they can also be approximated as locally flat. We will assume a local coverage of 100%, meaning that all carbon sites surrounding a given hydrogen atom are also occupied, and neighboring hydrogen atoms sit on opposite sides of the graphene layer.² This configuration, sometimes dubbed “graphane,” has been discussed in detail in Refs. [34,36] for the case of tritiated graphene. The initial proton wave function is

$$\psi_0(\mathbf{x}_p) = \frac{1}{\pi^{3/4} \lambda_{\parallel} \sqrt{\lambda_{\perp}}} \exp\left(-\frac{x_{p,\parallel}^2}{2\lambda_{\parallel}^2} - \frac{x_{p,\perp}^2}{2\lambda_{\perp}^2}\right), \quad (2)$$

where $x_{p,\parallel}$ and $x_{p,\perp}$ are the directions parallel and perpendicular to the graphene plane, respectively. Upon a

¹In the mass range of interest, the regime of contact interaction is valid for $m_\phi \gg \mathcal{O}(1-100)$ keV. For much larger masses, $m_\phi \gg \mathcal{O}(100)$ MeV, strong bounds are imposed by meson decays [54,55], as well as by direct constraints on the mediators [e.g., [56]].

²Note that a *local* coverage of 100% does not necessarily mean that all carbon sites of the *macroscopic* sample are hydrogenated.

rescaling from the tritium to the proton mass, the parallel and perpendicular spreads are $\lambda_{\parallel} \simeq 0.17 \text{ \AA}$ and $\lambda_{\perp} \simeq 0.11 \text{ \AA}$, while the binding energy is $\varepsilon_0 \simeq -4.5 \text{ eV}$ [36].

In this work, we use graphene as a benchmark two-dimensional material, although large areas might be achieved more easily by leveraging the surface of graphite. The 100% loading configuration described above, however, is not realizable on graphite, as the bottom parts of the carbon sites are bound to another graphene layer, and the hydrogenation is expected to happen all on the same side. A similar situation happens for CNTs. The 100% loading configuration is only realizable on single-wall CNTs, which can be synthesized in lab [e.g., [57,58]]. For the more common multiwall CNTs, hydrogenation happens only on the exterior of the tube. Nonetheless, the parameters characterizing the binding of the proton depend on the loading and spatial distribution only by, at most, a factor of order 1 [34–36]. Our proposal and conclusions are thus unaffected.

For the processes of interest, the final-state proton is free. Its wave function, as well as those of the incoming and outgoing dark matter, are then well approximated by plane waves. We normalize them to unity when working at some finite volume, V . The corresponding matrix element then reads

$$\mathcal{M} \equiv \langle \mathbf{k}', \mathbf{p} | U | \mathbf{k}, \psi_0 \rangle = -\frac{g_{\chi} g_p}{V^{3/2} m_{\phi}^2} \hat{\psi}_0(\mathbf{k} - \mathbf{k}' - \mathbf{p}), \quad (3)$$

where $\mathbf{k}$, $\mathbf{k}'$, and $\mathbf{p}$ are the momenta of the incoming dark matter, the outgoing dark matter, and the outgoing proton, while $\hat{\psi}_0$ is the Fourier transform of the proton's initial wave function. Given this, the rate of ejection of a proton by dark matter is obtained from Fermi's golden rule:

$$d\Gamma = 2\pi |\mathcal{M}|^2 \delta(\varepsilon_0 + E_k - E_{k'} - E_p) \frac{V d\mathbf{k}'}{(2\pi)^3} \frac{V d\mathbf{p}}{(2\pi)^3}, \quad (4)$$

where the E 's are kinetic energies, with obvious definitions. Finally, the expected experimental rate is obtained by accounting for the number of available protons N_H , the mass density of incoming dark matter ρ_{χ} , and its velocity distribution in the Milky Way halo $f(v)$. Specifically, it is given by

$$R(\hat{\mathbf{v}}_e) = V N_H \frac{\rho_{\chi}}{m_{\chi}} \int d\mathbf{v} f(|\mathbf{v} + \mathbf{v}_e|) \Gamma(\mathbf{v}), \quad (5)$$

where Γ is the total rate obtained by integrating Eq. (4), which depends on the dark matter velocity through its initial momentum. We take the velocity distribution to be a truncated Maxwellian as in the standard halo model, with dispersion $v_0 = 230 \text{ km/s}$, escape velocity $v_{\text{esc}} = 600 \text{ km/s}$, and boosted with respect to the Galactic rest frame by the Earth velocity $v_e = 240 \text{ km/s}$ [59,60]. We also take the dark matter mass density to be $\rho_{\chi} = 0.4 \text{ GeV/cm}^3$ [59,61]. We have also anticipated the possible directionality of our signal through the dependence of the rate on the direction of the Earth's

velocity. The direction of the “dark matter wind” is opposite to $\hat{\mathbf{v}}_e$.

2D: Graphene. Equation (5) provides the ideal rate for the liberation of a proton from the surface of the carbon structure. Yet, not all events are experimentally detectable. A possible obstacle lies in the fact that the proton could be ejected together with one of the electrons which were originally localized in its vicinity. This would produce a neutral hydrogen, which is much harder to detect.

We performed a DFT analysis to estimate the probability that the interaction ejects a naked proton, with no electrons around it. Given our flat graphene with a 100% coverage, we computed the probability that, after the scattering, all electrons end up in the ground state of the new graphene sheet (identical to the initial one, but with one fewer proton). As described in detail in Ref. [62], we do this within the sudden approximation for the dark matter–proton interaction. This is valid for all masses of interest, except for those right around the MeV range, where corrections are expected. We conservatively find that the probability of ejecting a naked proton is

$$P_{\text{proton}} \gtrsim 72\%. \quad (6)$$

As far as graphene is concerned, after the proton is liberated without electrons bound to it, it is accelerated with a suitable electric field, and eventually collected by the detector—see again Fig. 1. The final rate is then

$$R_{\text{graphene}}(\hat{\mathbf{v}}_e) = R(\hat{\mathbf{v}}_e) P_{\text{proton}}. \quad (7)$$

We report the corresponding projections in Fig. 2, computed in a configuration with the dark matter wind orthogonal to the graphene layer. The strength of the dark matter–proton interaction is represented with a reference cross section, $\bar{\sigma}_p \equiv g_{\chi}^2 g_p^2 \mu_{\chi p}^2 / (\pi m_{\phi}^4)$, where $\mu_{\chi p}$ is the dark matter–proton reduced mass. We show two instances: a conservative one considering a graphene sheet of area 100 cm^2 , and a more ambitious one with an area of 1 m^2 . Assuming that our graphene sample is globally fully hydrogenated, these correspond respectively to a target mass of $M_H \simeq 0.66$ and $66 \text{ }\mu\text{g}$. As one can see, already with this simple setup, the expected sensitivity is considerably better than the current experimental bounds. The minimal mass that can be probed by this detector is dictated by the initial proton binding energy, which must be overcome by the dark matter kinetic energy. This leads to $m_{\chi}^{\text{min}} = 2|\varepsilon_0|/(v_{\text{esc}} + v_e)^2 \simeq 1.1 \text{ MeV}$.

3D: Nanotubes. Along similar lines, one can consider hydrogenated CNTs. This allows for a three-dimensional target, thus gaining considerably more target mass if compared with a graphene/graphite setup with equal surface area. Arrays of aligned CNTs have already been

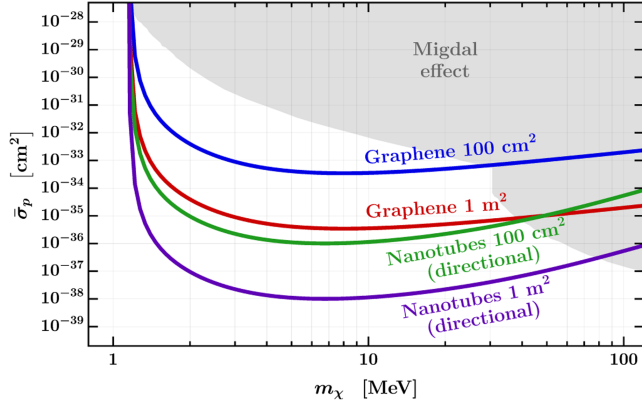


FIG. 2. Projected reach at 90% CL, assuming no background and one year of exposure. The target masses corresponding to graphene of area 100 cm^2 and 1 m^2 are $M_H = 0.66$ and $66 \mu\text{g}$, respectively. For CNTs of area 100 cm^2 and 1 m^2 , they are instead $M_H = 0.84$ and 84 mg . The gray shaded areas correspond to currently excluded region, as obtained using the Migdal effect, whose leading bounds are set by SENSEI [5] and PandaX-4T [29]. The reference cross section is defined as $\bar{\sigma}_p \equiv g_X^2 g_p^2 \mu_{\chi p}^2 / (\pi m_\phi^4)$.

considered as a possible target for direct dark matter detection. In particular, extensive work has been devoted to trying to employ them to hunt for possible dark matter–electron interactions [37–41]. As far as the dark matter–nucleon coupling is concerned, they were originally considered to look for processes where the dark matter ejects a carbon nucleus [63,64]. In that instance, however, the large carbon mass allowed one to probe masses no lighter than a few GeVs.

In our case, instead, the target is a proton. Once emitted, the proton must travel to the outside of the array of CNTs to eventually be detected. Before doing that, however, the vast majority of the protons scatter multiple times on the surfaces of the CNTs. It is therefore necessary to understand the result of these interactions: whether they scatter off elastically, inelastically, or if they get captured and lost.

A number of recent experimental results have investigated the behavior of protons impinging on the surface of graphene, which, we recall, is locally very similar to single-wall CNTs. Among other things, these data show that (a) graphene is somewhat permeable to protons with kinetic energies above about 1 eV [e.g., [65–69]], and (b) proton absorption on the layer is strongly suppressed when their kinetic energy is below about 0.2 eV [70].³ Since the degree

³This behavior differs from that of other ions, which instead are blocked and absorbed by single-layer graphene [e.g., [65–70]]. We also point out that early calculations suggested that protons are neutralized by the interaction with solid surfaces [e.g., [71–74]]. Such results seem to be at odds with the above mentioned experiments. Nonetheless, we notice that they are obtained for proton kinetic energies larger than 100 eV, far from the regime discussed here.

of permeability is hard to quantify at this stage, we adopt a conservative assumption: any proton with energy in the plane orthogonal to the CNT axis, E_{xy} , exceeding $E_{xy} > 0.2 \text{ eV}$ undergoes some inelastic processes, loses energy, and is eventually captured. Only protons with $E_{xy} < 0.2 \text{ eV}$ scatter elastically off the walls of the CNTs. With this at hand, there are three possible fates for one of the protons ejected by the dark matter:

- (1) If its energy is $E_{xy} > 0.2 \text{ eV}$, it gets captured after interacting with the CNT wall.
- (2) If its energy is $E_{xy} < 0.2 \text{ eV}$, it leaves the array from either the bottom or from the sides, preventing its collection and observation.
- (3) If its energy is $E_{xy} < 0.2 \text{ eV}$, it manages to leave the array from the top.

Among these possibilities, only the last one is favorable to our setup. We indicate its probability as $P_{\text{exit}}(\hat{\mathbf{v}}_e)$, anticipating the fact that, as shown below, this depends on the direction of the dark matter wind.

To determine the fraction of protons that manage to leave the CNT forest from the top, we run a suitably designed Monte Carlo simulation, whose details are reported in Ref. [62]. First of all, one of the main outcomes of our simulations is the following: for arrays of area larger than $0.5 \text{ mm} \times 0.5 \text{ mm}$, events lost by later exits are completely negligible. This result is central, as it would remain valid even if one were to consider a more realistic CNT forest, where the spacing between CNTs is somewhat irregular. This conclusion is stronger for larger arrays, as it becomes much more likely for a proton to reach the top before reaching the sides. The results for the probability that a proton escapes from the top are reported in Fig. 3. This probability decreases strongly with increasing dark matter masses. Indeed, higher masses imply a much larger chance for the outgoing proton to have $E_{xy} > 0.2 \text{ eV}$, and thus to fall among those events that we consider as lost. From the large difference between the two curves, we also deduce that we expect order-1 modulations, when the direction of the dark matter wind with respect to the orientation of CNTs changes. This can be used as powerful leverage to discriminate the signal from the (isotropic) background. The modulation decreases for masses $m_\chi \lesssim 10 \text{ MeV}$. This is indeed the regime where the typical dark matter momentum becomes smaller than the inverse spread of the initial proton wave function, $m_\chi v_\chi \lesssim 1/\lambda_\parallel, 1/\lambda_\perp$ —see Eq. (2). Momentum conservation is then maximally broken by the wave function, and the final-state proton retains less information about the direction of the incoming dark matter.

In light of all this, the expected event rate for an array of CNTs is given by

$$R_{\text{CNT}}(\hat{\mathbf{v}}_e) = R(\hat{\mathbf{v}}_e) P_{\text{proton}} P_{\text{exit}}(\hat{\mathbf{v}}_e), \quad (8)$$

where P_{proton} is the probability reported in Eq. (6). The corresponding projected sensitivity is again shown in

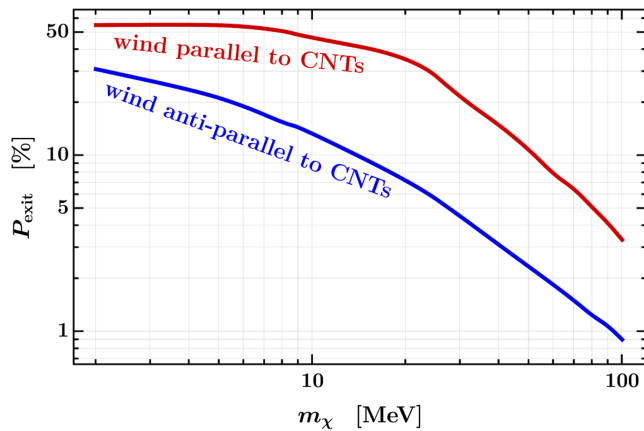


FIG. 3. Probability for a proton to leave from the top side of the array of CNTs as a function of the dark matter mass, computed for a dark matter wind parallel (red curve) and antiparallel (blue curve) to the orientation of the CNTs.

Fig. 2. Specifically, we assume the CNTs to have a diameter of 10 nm, a distance of 50 nm between their centers, and a height of 100 μm [75]. For arrays with area 100 cm^2 and 1 m^2 , the corresponding target masses are $M_H = 0.84$ and 84 mg, respectively.

Conclusion. We proposed the use of hydrogenated graphene, possibly graphite, and carbon nanotubes as targets to probe dark matter–nucleon interactions for masses as light as $m_\chi \sim \mathcal{O}(\text{MeV})$. Thanks to the weak carbon-hydrogen bond and the quasielastic nature of the interactions, this setup leverages the net charge of the emitted proton, which allows us to accelerate it up to detectable energies, thus evading the limits posed by the experimental energy threshold on traditional nuclear recoil. For this reason, the proposed detector would be characterized by a small energy threshold, and a detection scheme based on these materials can be orders of magnitude more sensitive than current experiments.

The setup is expected to be inexpensive and easy to operate. Hydrogenated graphene and graphite are simple systems, with well-understood properties, thus allowing us to envision the realization of this proposal in a short timescale and with a reasonable opportunity to scale up the instrumented target mass. Moreover, CNTs provide substantially more target mass than an equivalent area of graphene/graphite. They are also expected to provide a strongly directional signal, offering a key tool toward background rejection. Nonetheless, they require better understanding, especially regarding the capture probability of protons scattering off their walls, the characterization of irregularities in their distribution and geometry [76], and how these impact the proposed scheme.

Dark matter detectors usually require good shielding against environmental radioactivity and cosmic rays, and

they must be fabricated with weakly radioactive materials. For instance, gamma rays can interact with the carbon atoms of the substrate and generate background Compton electrons reaching the SDD, and mimicking a dark matter signal. Compton electrons, in fact, can have energies as large as 100 keV, and are therefore not stopped by the applied electric field. Nonetheless, we calculated that, for the 100 cm^2 hydrogenated graphene target, it is sufficient to keep the gamma ray flux below a few Hz to have negligible background events in one year of exposure. A further background suppression technique might be based on the use of a localized magnetic field to deflect electrons. Environmental neutrons are also expected to be innocuous. For instance, the typical neutron flux at the underground Gran Sasso laboratory is $\Phi \sim 10^{-6}$ neutrons/ cm^2/s [77]. The neutron-proton cross section is a function of energy. However, even considering only its highest possible value (achieved at very low energy), this is about $\sigma_{np} \sim 20$ barns [78]. A simple calculation shows that the corresponding rate of background events due to environmental neutrons is always completely negligible, except for the largest CNT array considered here, of area 1 m^2 . In that case, the background rate is expected to be $\mathcal{O}(10)$ events/year. Nonetheless, even in this instance, the neutron flux can easily be reduced by several orders of magnitude with proper shielding, as shown, for example, in Ref. [79]. Moreover, signal and background can be discriminated thanks to directionality.

This work suggests a number of further theoretical and experimental investigations. A more accurate theoretical characterization of the materials will come, together with a more refined evaluation of the proton ejection probability, and with the study of different dark matter–proton couplings. These include light mediators and spin-dependent interactions, with the latter allowed thanks to the proton net spin. From the experimental viewpoint, the proposed mechanism of proton ejection can be validated by mimicking the dark matter particles with epithermal neutrons, with energies in the eV range.

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Data availability. The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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