## Doped Cryocrystals for Ultrasensitive EDM Measurements: Snowmass LOI

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Atoms and molecules gently encased in the "artifical vacuum" of an inert cryogenic crystal matrix are extremely promising as quantum sensors for fundamental physics tests, such as searches for EDMs and time-varying, dark-matter-related EDMs. They are also a promising technology for single molecule NMR and MRI. The key remaining challenge is producing single-crystal samples with high dopant densities.

The Standard Model (SM) of particle physics is the most precisely tested theory of physical reality [1]. For example, its prediction for the electron magnetic moment (including the Dirac prediction, quantum electrodynamics, hadronic and weak contributions) agrees to 1 part per billion with exceptionally precise measurements [2], though an intriguing 2.4 standard deviation discrepancy is now prompting new measurements and theoretical investigations. Despite its successes, there is a preponderance of evidence that the SM is incomplete. It fails to account for several basic phenomena of the universe: Why is there more matter than anti-matter in the universe? What is Dark Matter, which is five times more abundant than the ordinary matter described by the SM? What is Dark Energy? How can gravity be incorporated into our understanding of the other fundamental forces in the SM? The traditional approach towards answering these questions has been to increase the collision energy achievable at particle colliders, allowing new particles and phenomena to be produced and studied. While this approach has been enormously successful during the development and confirmation of the SM, colliders have thus far failed to find any of the new physics Beyond the Standard Model (BSM) described above. As the cost of continuing to increase the energy frontier at colliders grows, searches at the precision frontier of particle physics provide an alternative approach [3–5]. At the precision frontier, sensitive laboratory-scale experiments may be able to provide insight into these questions by detecting tiny deviations arising from higher energy scales (or weaker couplings) than can currently be reached by collider experiments.

Atom-like systems in the solid state, such as NV centers in diamond or phosphor donors in silicon, have shown great capabilities as quantum sensors [6]. These "artifical atoms" have demonstrated high sensitivity as ensemble sensors, leveraging the advantage of the large number of atoms trapped within the solid [6]. They also have demonstrated high sensitivity as single-atom sensors for nanoscale NMR and MRI, due to advantages from the angstrom-scale localization provided by the solid-phase host [6]. Unfortunately, the atoms used in these systems to date have poor sensitivity to physics beyond the standard model.

Fortunately, certain carefully-chosen gas-phase atoms and molecules are known to offer great sensitivity to physics beyond the standard model [3, 7]. Trapping such atoms and molecules within the "artifical vacuum" of an inert cryogenic crystal matrix is extremely promising for developing quantum sensors for fundamental physics tests. Since Pryor and Wilczek [8], many have proposed confining dopant atoms and molecules within the benign environment of inert, cm-sized [9], cryogenic crystals [10–16]. The benefits of trapping in a cryogenic matrix are clear: the interaction times, densities, and number of dopants are all orders of magnitude larger than what can be achieved with gas-phase techniques. However, these advantages can only be realized if the trapped atoms retain their essential properties for quantum sensing: efficient optical control and readout of spin states, and spin superposition states with long coherence times [6]. Work with alkali atoms in solid hydrogen and solid helium have demonstrated all these properties [17–22]. While solid helium has not demonstrated high dopant densities, solid parahydrogen has. Moreover, electron spin coherence times approaching  $T_2 = 0.1$  s have been demonstrated for rubidium atoms in parahydrogen

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[19]; this is a longer spin coherence time than that of ACME-II, the experiment that produced the best limit on the electron EDM [7].

While these preliminary results are extremely promising, the ensemble dephasing time  $T_2^*$  — crucial for ensemble-based fundamental physics measurements — has been limited by the polycrystalline nature of the samples grown to date [17, 18]. The inhomogeneous nature of the host limits the spin  $T_2^*$  to a value many orders of magnitude lower than the single-spin coherence time  $T_2$ , and prevents efficient optical addressing of all the trapped atoms, reducing the effective number of the implanted species. Thus, the key remaining experimental challenge is to create single-crystal doped cryocrystals with a well-defined crystal axis, as has been done for NV centers in diamond. This would enable the full potential of the system to be realized.

This goal should be achievable: single-crystal cryocrystals with a well defined crystal axis have previously been created [23–25]. But the methods used to grow single-crystal samples are incompatible with the methods currently employed to produce samples with high dopant densities [26, 27]. We propose the creation of a facility optimized for the development of new techniques to produce single-crystal cryocrystal samples with high dopant densities. We will perform an exhaustive and systematic study of the growth and diagnosis of doped cyrocrystals of helium, hydrogen, neon, and argon. We will use optical techniques as well as the electron spin  $T_2^*$  of a simple atomic dopant [18] as diagnostics.

The eventual creation of doped single crystals will unlock their full potential for the creation of quantum sensors for exploring physics beyond the standard model. The number of dopants that can be distributed throughout a cryocrystal with a magnetic inhomogeneity low enough to allow a 1 ms spin coherence time is large enough that the 350 hour data set that produced the best electron EDM value (ACME-II [7]) could in principle be accumulated in seconds. Greatly improved sensitivity for measuring EDMs and time-varying EDMs (e.g. due to dark matter), should result, along with improvements in "conventional" quantum sensing, such as single-molecule NMR and MRI [19].

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