Nuclear Instruments and Methods in Physics Research A 1069 (2024) 169951

Contents lists available at ScienceDirect

Nuclear Inst. and Methods in Physics Research, A

journal homepage: www.elsevier.com/locate/nima

Measuring the electric dipole moment of the electron using polar molecules in a parahydrogen matrix

G. Messineo [a,b,*], P. Antonini [c], M. Benettoni [a], A.F. Borghesani [a,d,e], C. Braggio [a,e], R. Calabrese [f,g], G. Carugno [a,b], F. Chiossi [h], A. Dainelli [c], U. Gasparini [a,e], F. Gonella [a], M. Guarise [f,g], A. Khanbekyan [i,j], A. Lombardi [c], E. Mariotti [i,j], J. Pazzini [a,e,k,l], G. Ruoso [c], L. Tomassetti [f,g], M. Zanetti [a,e]

[a] INFN Sezione di Padova, via Marzolo, 8, 35131, Padua, Italy
[b] NQSTI, National Quantum Science and Technology Institute, Piazzale Aldo Moro, 7, 00185, Rome, Italy
[c] INFN Laboratori Nazionali di Legnaro, Viale dell'Università, 2, 35020, Legnaro, Italy
[d] CNISM unit, University of Padua, via Marzolo, 8, 35131, Padua, Italy
[e] Department of Physics and Astronomy, University of Padua, via Marzolo, 8, 35131, Padua, Italy
[f] INFN Sezione di Ferrara, via Saragat, 1, 44122, Ferrara, Italy
[g] Department of Physics and Earth Science, University of Ferrara, via Saragat, 1, 44122, Ferrara, Italy
[h] Institut de Recherche de Chimie Paris, 11, rue Pierre et Marie Curie, 75231, Paris cedex 05, France
[i] Department of Physical Sciences, University of Siena, Via Roma, 56, 53100, Siena, Italy
[j] INFN sezione di Pisa, Via Filippo Buonarroti, 3, 56127, Pisa, Italy
[k] Department of Industrial Engineering, University of Padua, via Venezia, 1, 35131, Padua, Italy
[l] Department of Information Engineering, University of Padua, via Gradenigo 6/b, 35131, Padua, Italy

ARTICLE INFO

Keywords:
Electron electric dipole moment
Parahydrogen
Matrix isolation
Cryogenic crystals

ABSTRACT

This paper outlines the groundwork we have done in preparation for our proposed experiment to measure the electric dipole moment of the electron in barium monofluoride molecules within a cryogenic parahydrogen matrix. We describe the experimental setup we use to grow and characterize the cryogenic crystals and produce a molecular source of barium monofluoride. The technique we present has the potential to improve the sensitivity to the electric dipole moment of the electron by increasing the number of molecules under measurement without affecting their spin coherence times. If successful, this approach would represent a significant advancement in precision measurements using molecules in cryogenic solid matrices.

1. Introduction

Studying the electric dipole moment of the electron (eEDM) is a promising path to explore physics beyond the Standard Model [1–4]. A non-zero eEDM measurement would indicate a violation of time-reversal and CP symmetry, potentially shedding light on unresolved phenomena like matter-antimatter asymmetry, dark matter, and dark energy.

The most common experimental method for measuring the eEDM is to observe changes in the precession rate of electron spins in an electric field. The sensitivity of this measurement depends on the strength of the electric field experienced by the electrons, their spin coherence time, and the number of electrons examined. Currently, the most stringent limit for the eEDM is $d_e \leq 4.1 \times 10^{-30}$ e cm, obtained using electrons confined in HfF^+ molecular ions [5].

To improve upon this limit, the PHYDES Collaboration plans to use heavy polar molecules, such as barium monofluoride (BaF), that have a high internal effective electric field (≈ 10 GV/cm) and embed them in a cryogenic parahydrogen (p-H_2) crystal matrix. By trapping the BaF molecules in a matrix, we can increase the number of molecules in the measurement volume by several orders of magnitude, thus improving the sensitivity to the eEDM effect [6]. Furthermore, the unique properties of the parahydrogen matrix, such as a large intermolecular distance and the absence of permanent electric moments [7], minimize the interaction with the guest molecules, potentially granting them a

* Corresponding author at: INFN Sezione di Padova, via Marzolo, 8, 35131, Padua, Italy.
E-mail address: giuseppe.messineo@pd.infn.it (G. Messineo).

https://doi.org/10.1016/j.nima.2024.169951
Received 30 June 2024; Received in revised form 27 September 2024; Accepted 30 September 2024
Available online 9 October 2024

narrow transition linewidth and long coherence time. Recently, coherence times as long as 0.1 s for the hyperfine transitions of rubidium atoms embedded in p-H_2 have been reported in literature [8].

2. The PHYDES experiment

Current efforts within the PHYDES collaboration have focused on producing and characterizing BaF-doped p-H_2 crystals in preparation for the eEDM measurement. To produce BaF molecules for our experiment, we designed a dedicated setup in which solid BaF_2 powder is evaporated in a glow discharge chamber in a commercial ion source. The resulting BaF_2 molecules are then dissociated and ionized by an electron beam, accelerated to 1 keV, and passed through a velocity filter to isolate a beam of BaF^+. These ions are decelerated and combined with p-H_2 gas to form a solid matrix. In a preliminary test, we achieved a BaF^+ current of about 20 nA at an energy of 5 eV.

To prevent charge accumulation within the matrix and ensure a high density of molecules, the BaF^+ ions must be neutralized. We plan to achieve this by injecting electrons into the matrix. These electrons will be photoextracted by UV laser pulses from a gold layer coated on the substrate where we grow the cryogenic crystal.

To investigate the feasibility of our photoextraction and neutralization approach, we studied the dynamics of quasi-free electrons extracted from a gold photocathode and injected into solid parahydrogen. We measured their thermalization length in the matrix and showed that these low-energy electrons rapidly thermalize within the bulk of the matrix, where they would become available for the neutralization process [9].

The cryogenic system for growing p-H_2 crystal matrices (shown in Fig. 1) follows the methods described in [10,11]. High-purity hydrogen gas cooled to around 20 K by a pulse tube refrigerator flows through a copper line containing hydrous ferric oxide, which enhances the conversion to the p-H_2 state [12]. The p-H_2 gas flows at approximately 10 standard cubic centimeters per minute toward a cold sapphire substrate coated with a 10 nm gold layer, where it condenses to form a solid crystal. The substrate, mounted on a copper plate cooled to around 2.9 K using a two-stage helium pulse tube cryocooler, is housed in a six-way cross stainless steel chamber. This chamber is pumped by a turbolecular system at the bottom and one on the top to expedite hydrogen removal from the upper section of the vacuum chamber. Before starting the growth process, the system is evacuated to a base pressure below 10^{-8} mbar and, during crystal growth, the pressure is maintained constantly below 5×10^{-7} mbar.

The thickness and ortho-hydrogen fraction of the p-H_2 crystal were routinely monitored during growth using infrared absorption spectroscopy, as described in [9]. The crystal growth rate was approximately 100 μm/h with an ortho-hydrogen fraction of about 2%–3%. The crystals were grown to thicknesses of about 500-600 μm.

3. Conclusions

The innovative approach of embedding polar molecules with high internal electric fields in cryogenic crystal matrices shows promise in improving the eEDM detection limits and uncovering physics beyond the Standard Model. In this context, the PHYDES Collaboration is pioneering the combined use of isotopically selected BaF molecules produced in a custom-made molecular source and a pH_2 host matrix. The advantage of using parahydrogen as a host matrix is that it has a larger intermolecular distance and lattice constant compared to other commonly used cryogenic matrices (such as Ne or Ar), so it provides more free space for the guest molecules and weaker interactions with the surrounding host molecules.

We have built a molecular source producing up to 20 nA of BaF+ molecules at an energy of 5 eV and demonstrated the ability to grow optically transparent pH_2 crystals 500-600 μm thick.

Future work will focus on improving the production and characterization of BaF-doped p-H_2 crystals and developing techniques to manipulate and selectively target the electronic states of BaF molecules within the p-H_2 crystal necessary for the eEDM measurement.

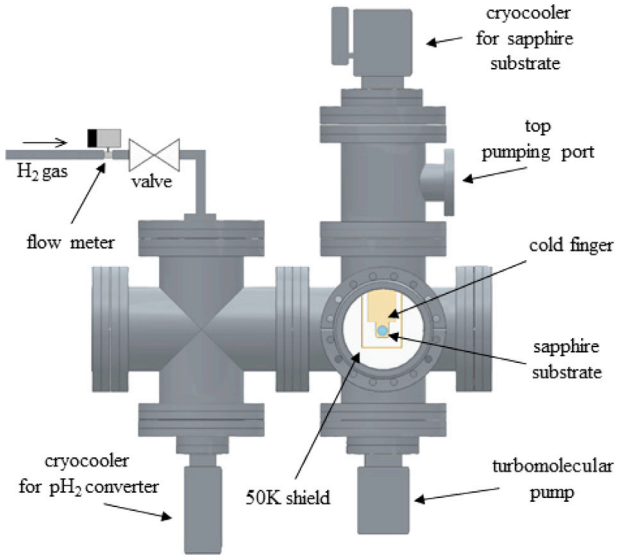

Fig. 1. Schematic illustration of the experimental setup used for crystal growth: hydrogen gas flows through a copper line containing a hydrous ferric oxide catalyst, which is maintained at around 20 K. The substrate on which the p-H_2 is grown is cooled to approximately 2.9 K by another cryocooler. The growth chamber, a six-way cross stainless-steel chamber, is pumped by turbomolecular pumps.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

We acknowledge the support of the PNRR MUR project PE0000023-NQSTI financed by the European Union – Next Generation EU. This work is also supported in part by the "Departments of Excellence grant 2023–2027 - Quantum Frontiers" and by the "Progetti di Ricerca di Rilevante Interesse Nazionale" (PRIN grant 20227F5W4N) of the Italian Ministry of University and Research.

References

[1] J. Baron, W.C. Campbell, D. DeMille, J.M. Doyle, G. Gabrielse, Y.V. Gurevich, P.W. Hess, N.R. Hutzler, E. Kirilov, I. Kozyryev, B.R. O'Leary, C.D. Panda, M.F. Parsons, E.S. Petrik, B. Spaun, A.C. Vutha, A.D. West, Order of magnitude smaller limit on the electric dipole moment of the electron, Science 343 (6168) (2014) 269–272, https://www.science.org/doi/abs/10.1126/science.1248213.

[2] V. Andreev, D.G. Ang, D. DeMille, J.M. Doyle, G. Gabrielse, J. Haefner, N.R. Hutzler, Z. Lasner, C. Meisenhelder, B.R. O'Leary, C.D. Panda, A.D. West, X. West, Improved limit on the electric dipole moment of the electron, Nature 562 (7727) (2018) 355–360, http://dx.doi.org/10.1038/s41586-018-0599-8.

[3] C.J. Ho, J.A. Devlin, I.M. Rabey, P. Yzombard, J. Lim, S.C. Wright, N.J. Fitch, E.A. Hinds, M.R. Tarbutt, B.E. Sauer, New techniques for a measurement of the electron's electric dipole moment, New J. Phys. 22 (5) (2020) 053031, http://dx.doi.org/10.1088/1367-2630/ab83d2.

[4] P. Aggarwal, H.L. Bethlem, A. Borschevsky, M. Denis, K. Esajas, P.A.B. Haase, Y. Hao, S. Hoekstra, K. Jungmann, M.C. Meijknecht, R.G.E. Timmermans, W. Ubachs, L. Willmann, A. Zapara, Measuring the electric dipole moment of the electron in BaF, Eur. Phys. J. D 72 (197) (2018) http://dx.doi.org/10.1140/epjd/e2018-90192-9.

[5] T.S. Roussy, L. Caldwell, T. Wright, W.B. Cairncross, Y. Shagam, K.B. Ng, N. Schlossberger, S.Y. Park, A. Wang, J. Ye, E.A. Cornell, An improved bound on the electron's electric dipole moment, Science 381 (6653) (2023) 46–50, https://www.science.org/doi/abs/10.1126/science.adg4084.

[6] A.C. Vutha, M. Horbatsch, E.A. Hessels, Oriented polar molecules in a solid inert-gas matrix: A proposed method for measuring the electric dipole moment of the electron, Atoms 6 (1) (2018) https://www.mdpi.com/2218-2004/6/1/3.

[7] T. Momose, H. Hoshina, M. Fushitani, H. Katsuki, High-resolution spectroscopy and the analysis of ro-vibrational transitions of molecules in solid parahydrogen, Vib. Spectrosc. 34 (1) (2004) 95–108, http://dx.doi.org/10.1016/j.vibspec.2003.06.001.

[8] S. Upadhyay, U. Dargyte, D. Patterson, J.D. Weinstein, Ultralong spin-coherence times for rubidium atoms in solid parahydrogen via dynamical decoupling, Phys. Rev. Lett. 125 (2020) 043601, https://link.aps.org/doi/10.1103/PhysRevLett.125.043601.

[9] A.F. Borghesani, G. Carugno, G. Messineo, J. Pazzini, Electron thermalization length in solid para-hydrogen at low-temperature, J. Chem. Phys. 159 (10) (2023) 104501, http://dx.doi.org/10.1063/5.0163776.

[10] B.A. Tom, S. Bhasker, Y. Miyamoto, T. Momose, B.J. McCall, Producing and quantifying enriched para-H2, Rev. Sci. Instrum. 80 (1) (2009) 016108, http://dx.doi.org/10.1063/1.3072881.

[11] A. Bhandari, A.P. Rollings, L. Ratto, J.D. Weinstein, High-purity solid parahydrogen, Rev. Sci. Instrum. 92 (7) (2021) 073202, http://dx.doi.org/10.1063/5.0049006.

[12] D.H. Weitzel, W.V. Loebenstein, J.W. Draper, O.E. Park, Ortho-para catalysis in liquid-hydrogen production, J. Res. Natl. Bur. Stand. 60 (3) (1958) 221–227.