PROCEEDINGS OF SCIENCE

Spin manipulation in atomic and molecular systems for nuclear spin applications

C. S. Kannis,[a,*] **R. Engels,**[b,c] **T. El-Kordy,**[d,e] **N. Faatz,**[b,c,f] **S. J. Pütz,**[b,c,g] **V. Verhoeven,**[c,h] **T. P. Rakitzis**[i,j] **and M. Büscher**[a,k]

[a] *Institut für Laser- und Plasmaphysik, Heinrich-Heine-Universität Düsseldorf, 40225 Düsseldorf, Germany*
[b] *Insitut für Kernphysik, Forschungszentrum Jülich, 52428 Jülich, Germany*
[c] *GSI Helmholtzzentrum für Schwerionenforschung, 64291 Darmstadt, Germany*
[d] *FH Aachen – University of Applied Sciences, 52066 Aachen, Germany*
[e] *Institute of Technology and Engineering, Forschungszentrum Jülich, 52428 Jülich, Germany*
[f] *III. Physikalisches Institut B, RWTH Aachen University, 52062 Aachen, Germany*
[g] *Institut für Kernphysik, Universität zu Köln, 50937 Köln, Germany*
[h] *Institut für Experimentalphysik I, Ruhr-Universität-Bochum, 44801 Bochum, Germany*
[i] *Department of Physics, University of Crete, 70013 Heraklion–Crete, Greece*
[j] *Institute of Electronic Structure and Lasers, Foundation for Research and Technology–Hellas, 71110 Heraklion–Crete, Greece*
[k] *Peter-Grünberg Institut, Forschungszentrum Jülich, 52428 Jülich, Germany*
E-mail: chrysovalantis.kannis@uni-duesseldorf.de, r.w.engels@fz-juelich.de, tarek.el-kordy@alumni.fh-aachen.de, n.faatz@extern.fz-juelich.de, spuetz12@smail.uni-koeln.de, v.verhoeven@gsi.de, ptr@iesl.forth.gr, m.buescher@fz-juelich.de

Nuclear spin polarization has important applications across various fields, including physics and medicine. It also offers several advantages, such as cross-section enhancement, in the five-nucleon fusion reactions, namely the D–T and D–^{3}He reactions. This work presents a theoretical study of spin dynamics in the hyperfine regime for selected atomic and molecular systems and explores alternative techniques to generate or enhance nuclear spin polarization in these systems. Particular emphasis is placed on coherent spin manipulation strategies and the role of hyperfine interactions in transferring angular momentum from electronic (in atoms) or rotational (in molecules) to nuclear degrees of freedom.

The 26th international symposium on spin physics (SPIN2025)
21–26 September, 2025
Qingdao (Tsingtao), Shandong Province, China

*Speaker

 ISSN 1824-8039 . Published by SISSA Medialab.

PoS(SPIN2025)169

1. Introduction

The development of nuclear spin polarization in atomic and molecular systems is vital in several fields such as physics and medicine. Typical examples include nuclear magnetic resonance (NMR) spectroscopy and magnetic resonance imaging (MRI), whose signals—and therefore their accuracy—can be enhanced by the use of hyperpolarized tracers [1–3]. In physics, beyond applications in fundamental research (e.g., quantum computation [4], electric dipole moment searches [5], etc.), nuclear spin polarization also provides advantages in energy production through nuclear fusion [6]. Specifically, in the five-nucleon reactions, i.e., the D–T and D–^{3}He reactions, polarization of the reactants is known to increase the fusion cross section and allow control over the emission direction of the products, improving reaction dynamics and leading to an overall enhancement of reactor efficiency [7–9].

However, direct production of nuclear polarization in bound systems such as atoms and molecules is not always straightforward and is often limited in the achievable polarization degree. In contrast, it is typically easier to polarize the electron spin of atoms or the rotational angular momentum of molecules. Hyperfine interactions between these degrees of freedom and the nuclear spins enable the transfer of polarization between different spin/angular momentum entities, thereby enhancing nuclear polarization. An appropriately configured external magnetic field can further manipulate and amplify this transfer.

Since the nuclei involved in the five-nucleon reactions have spin quantum numbers $1/2$ (tritons/helions) and 1 (deuterons), this work focuses on hydrogen (H; nuclear spin $1/2$), deuterium (D) atoms, and HD molecules. Assuming specific spin-preparation scenarios for atomic or molecular beams moving with a constant, nonrelativistic velocity v along the z axis, we investigate the dynamics of interacting angular momenta as the beams pass through a static, spatially oscillating, longitudinal magnetic field $B_z = B_0 \sin\left(\frac{2\pi z}{\lambda}\right)$ with B_0 the magnetic field amplitude and λ the wavelength of the oscillation. This magnetic field configuration is inspired by the work of Sona [10].

The calculations are performed using the density operator formalism in the rest frame of the particle, where the external magnetic field takes the form $B_z = B_0 \sin\left(\frac{2\pi v t}{\lambda}\right)$ and the Hamiltonian is therefore time-dependent. As a result, the Liouville–von Neumann equation governing the evolution of the density operator ρ is solved numerically [11]. The density matrix provides direct information about individual state populations and is essential for obtaining average values of observables such as spin projections and spin polarizations.

2. Hydrogen (H) and deuterium (D) atoms

In the following, we investigate the first metastable state $2S_{1/2}$ of H and D atoms. Their electron spin is initially fully (and presumably positively) polarized in a high-field region, and this polarization is subsequently shared between the electron and nuclear spins as the atoms are adiabatically transferred from high magnetic field to the low-field region $B_z \to 0$. Such a spin preparation can be achieved by using solely static fields [12]. The resulting average spin projections for H are $\langle m_{z,e} \rangle = 1/4$ for the electron and $\langle m_{z,p} \rangle = 1/4$ for the proton, while for D they are $\langle m_{z,e} \rangle = 1/6$ and $\langle m_{z,d} \rangle = 1/3$, respectively. The sums of the average electron and nuclear spin projections equal $1/2$, consistent with a fully polarized electron spin and an unpolarized nuclear

spin at the moment when the initial polarization is established in the high-field region, prior to adiabatic transfer. A similar spin preparation can be achieved for ground-state atoms [13], where protons or deuterons traverse optically pumped, polarized Rb vapor, capture polarized electrons in a strong magnetic field, and are then adiabatically transferred to near-zero field.

This specially prepared metastable beam enters (at $t = 0$) the sinusoidal longitudinal magnetic field, described in the previous section. Our analysis focuses on a single period of the trigonometric function, namely the time of flight is $t_f = \lambda/v$. Moreover, the ratio v/λ is adjusted to match the hyperfine splitting (in frequency units) of H ($\nu_{HF} = 177.56$ MHz) and D ($\nu_{HF} = 40.92$ MHz). Thus, the only remaining free parameter of the applied magnetic field is its amplitude B_0, which we vary and evaluate the spin dynamics.

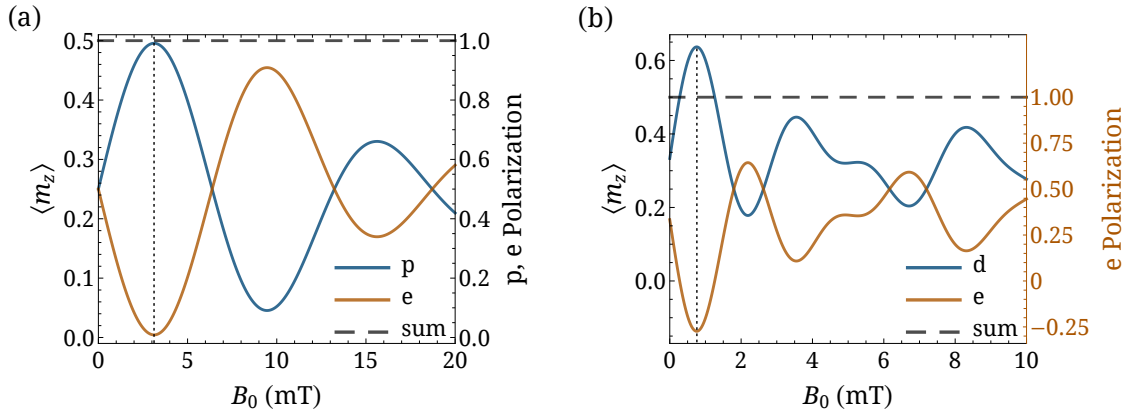

Figure 1: Average spin projections $\langle m_{z,p/d} \rangle$ (blue) and $\langle m_{z,e} \rangle$ (orange) at time t_f as a function of B_0 for metastable H (a) and D (b), with the spins initially prepared as described above. The right vertical axes show the corresponding proton and electron spin polarizations. For deuterium, the deuteron polarization is given directly by $\langle m_{z,d} \rangle$, since its spin is 1.

Figure 1 (a) and (b) show the average spin projections and resulting polarizations for metastable H and D at time t_f as a function of B_0. Polarizations near 100% for protons and approximately 64% for deuterons are expected at 3.1 mT and 0.76 mT, respectively (see vertical dotted lines). The manipulation of these quantities is enabled by magnetic dipole transitions driven by the longitudinal field. These transitions obey the selection rule $\Delta m_F = 0$, i.e., they preserve the sum of spin projections along the z axis (as indicated by the dashed line in Fig. 1). Consequently, the induced redistribution of populations within the hyperfine manifold leads to no polarization loss and does not generate transverse polarization. Comparable results can also be obtained for the ground state of these atoms [11].

3. Hydrogen deuteride (HD) molecule

The sinusoidal-field technique can also be applied to molecular systems. Here, we consider the electronic ground state $^1\Sigma$ of HD, where the relevant degrees of freedom are the nuclear spins (proton and deuteron) and the rotational angular momentum J. A well-defined rotational angular momentum projection, e.g., $m_J = +J$, can be prepared using coherent excitation techniques [14–

16]. However, hyperfine interactions cause the rotational polarization to be transferred to the nuclear spins [17].

In the absence of external perturbations, the maximum achievable nuclear polarization is limited by the hyperfine coupling constants, while applying a constant magnetic field leads to decoupling of the interacting spins/angular momenta [18]. In contrast, the introduced sinusoidal-field method enables the enhancement of a chosen nuclear spin polarization by appropriately tuning the field parameters. For example, by selecting the wavelength (for a fixed molecular-beam velocity) such that the experienced frequency is $1/t_f = 153935$ Hz and setting the field amplitude to $B_0 = 48.6$ mT, a deuteron polarization of $\sim 83\%$ can be achieved starting from the rotationally excited state $J = 1$, $m_J = +1$. A similar enhancement can be achieved for the proton polarization, e.g., in the rotational state $J = 2$. Further details are provided in Ref. [11].

4. Limitations

The introduced spin manipulation technique, together with its computational framework, involves several limitations. First, no entanglement between the internal (spin and angular momentum) and external (position and momentum) degrees of freedom is assumed. The particles in the beam are therefore taken to follow classical trajectories: their external degrees of freedom are treated classically, whereas the internal degrees of freedom are treated quantum mechanically. This approximation is valid for experiments in which any dependence of the spin states on the particle's position or momentum remains unobservable. In addition, the beam particles are assumed to be non-interacting, and the selected beam velocities are nonrelativistic, limiting the kinetic energies to the keV range for the atomic and molecular systems discussed here.

Furthermore, this work focuses exclusively on the longitudinal magnetic field B_z and neglects the radial component B_r, which appears at off-axis positions ($r \neq 0$) and is given by $B_r = -\frac{r}{2}\frac{\partial B_z}{\partial z}$. This approximation is justified for $r \ll \lambda$. If this condition is not met, however, B_r induces magnetic dipole transitions within the same hyperfine manifold according to the selection rule $\Delta m_F = \pm 1$ [19, 20], thereby interfering with the population changes driven by B_z and reducing the total longitudinal spin polarization. The radial field also gives rise to an additional electric field, $\mathbf{E} = \mathbf{v} \times \mathbf{B} = v B_r \hat{\boldsymbol{\phi}}$, whose interaction with the system constitutes the motional Stark effect. This electric dipole interaction does not couple states within the same hyperfine manifold but instead mixes states of opposite parity. A detailed discussion and respective calculations can be found in Ref. [21].

Finally, although only a simple sinusoidal form was assigned to B_z here, the spin-manipulation technique is not restricted to a single field configuration. By appropriately tuning the field parameters, various waveform shapes can be employed to produce comparable or even slightly enhanced nuclear polarization transfer. As an example, results obtained with a sine-cubed waveform are compared with those from a pure sine wave in Ref. [11].

5. Conclusion

We have presented a theoretical study on the influence of a static, spatially oscillating magnetic field on a particle beam composed of interacting spins and/or angular momenta as the beam traverses

the applied field. We have shown that such an external perturbation can enhance the nuclear polarization in an incoherent spin preparation of H and D beams as well as in a coherent rotational-state preparation in HD beams. These manipulations were optimized for nuclear-spin applications and therefore targeted nuclear-polarization enhancement; however, additional manipulations are possible for other purposes.

Since the computations are performed numerically, the spin dynamics can be evaluated for arbitrarily varying magnetic fields that do not necessarily cross zero. This capability is useful for tracking polarization losses as particles move through regions that lack a homogeneous field or they may connect areas of different magnetic-field strengths in experimental setups for the production and storage of spin-polarized particles [22]. Finally, the computational framework employed here is also applicable to systems of interacting angular momenta at rest, where the applied magnetic field varies in time.

Acknowledgments

This work is funded by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) – 533904660.

References

[1] M.P.M. Letertre, P. Giraudeau and P. de Tullio, *Nuclear magnetic resonance spectroscopy in clinical metabolomics and personalized medicine: Current challenges and perspectives*, *Front. Mol. Biosci.* **8** (2021) 698337.

[2] S. Jørgensen, N. Bøgh, E. Hansen, M. Væggemose, H. Wiggers and C. Laustsen, *Hyperpolarized MRI – An update and future perspectives*, *Semin. Nucl. Med.* **52** (2022) 374.

[3] J. Eills, D. Budker, S. Cavagnero, E.Y. Chekmenev, S.J. Elliott, S. Jannin et al., *Spin hyperpolarization in modern magnetic resonance*, *Chem. Rev.* **123** (2023) 1417.

[4] N.A. Gershenfeld and I.L. Chuang, *Bulk spin-resonance quantum computation*, *Science* **275** (1997) 350.

[5] P.G. Harris, C.A. Baker, K. Green, P. Iaydjiev, S. Ivanov, D.J.R. May et al., *New experimental limit on the electric dipole moment of the neutron*, *Phys. Rev. Lett.* **82** (1999) 904.

[6] R.M. Kulsrud, H.P. Furth, E.J. Valeo and M. Goldhaber, *Fusion reactor plasmas with polarized nuclei*, *Phys. Rev. Lett.* **49** (1982) 1248.

[7] G. Ciullo, R. Engels, M. Büscher and A. Vasilyev, eds., *Nuclear Fusion with Polarized Fuel*, Springer Proceedings in Physics 187, Springer International Publishing (2016).

[8] W.W. Heidbrink, L.R. Baylor, M. Büscher, R.W. Engels, A.V. Garcia, A.G. Ghiozzi et al., *A research program to measure the lifetime of spin polarized fuel*, *Front. Phys.* **12** (2024) 1355212.

[9] J. Parisi, A. Diallo and J. Schwartz, *Simultaneous enhancement of tritium burn efficiency and fusion power with low-tritium spin-polarized fuel*, *Nucl. Fusion* **64** (2024) 126019.

[10] P.G. Sona, *New method proposed to increase polarization in polarized ion sources of H^- and D^-*, *Energ. Nucl.* **14** (1967) 295.

[11] C.S. Kannis, R. Engels, T. El-Kordy, N. Faatz, S.J. Pütz, V. Verhoeven et al., *Spin manipulation and nuclear polarization enhancement in particle beams with static magnetic fields*, *Phys. Rev. A* **112** (2025) 012801.

[12] A. Cesati, F. Cristofori, L.M. Colli and P.G. Sona, *Description of a source of polarized negative hydrogen or deuterium ions based on the properties of the $2S_{1/2}$ metastable state: Theory and Experiments*, *Energ. Nucl.* **13** (1966) 649.

[13] A. Kponou, A. Zelenski, S. Kokhanovski and V. Zubets, *Sona transition studies in the RHIC OPPIS*, *AIP Conf. Proc.* **980** (2008) 241.

[14] N.C.M. Bartlett, D.J. Miller, R.N. Zare, D. Sofikitis, T.P. Rakitzis and A.J. Alexander, *Preparation of oriented and aligned H_2 and HD by stimulated Raman pumping*, *J. Chem. Phys.* **129** (2008) 084312.

[15] N. Mukherjee and R.N. Zare, *Preparation of polarized molecules using coherent infrared multicolor ladder excitation*, *J. Chem. Phys.* **132** (2010) 154302.

[16] N.V. Vitanov, A.A. Rangelov, B.W. Shore and K. Bergmann, *Stimulated Raman adiabatic passage in physics, chemistry, and beyond*, *Rev. Mod. Phys.* **89** (2017) 015006.

[17] N.C.-M. Bartlett, D.J. Miller, R.N. Zare, A.J. Alexander, D. Sofikitis and T.P. Rakitzis, *Time-dependent depolarization of aligned HD molecules*, *Phys. Chem. Chem. Phys.* **11** (2009) 142.

[18] C.S. Kannis, G.E. Katsoprinakis, D. Sofikitis and T.P. Rakitzis, *Nuclear-spin-polarization dynamics of H_2, D_2, and HD molecules in magnetic fields*, *Phys. Rev. A* **98** (2018) 043426.

[19] R. Engels, M. Büscher, P. Buske, Y. Gan, K. Grigoryev, C. Hanhart et al., *Direct observation of transitions between quantum states with energy differences below 10 neV employing a Sona unit*, *Eur. Phys. J. D* **75** (2021) 257.

[20] C. Kannis, *Theoretical and experimental investigation of Sona transitions*, Ph.D. Thesis, RWTH Aachen University, Aachen, 2023.

[21] N. Faatz, *Simulation of the occupation numbers of hyperfine substates passing external fields*, Master's Thesis, RWTH Aachen University, Aachen, 2023.

[22] C.S. Kannis, J. Suarez and T.P. Rakitzis, *Macroscopic production of spin-polarised hydrogen atoms from the IR-excitation and photodissociation of molecular beams*, *Mol. Phys.* **120** (2022) e1975053.