Optical protection of alkali-metal atoms from spin relaxation

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We describe a new mechanism to continuously protect alkali-metal atoms from spin relaxation using a single off-resonant optical beam. We experimentally demonstrate that state-selective lightshifts can synchronize the Larmor frequencies of the two hyperfine manifolds, and by that form a unique decoherence-free subspace. We report an order of magnitude suppression of the spin decoherence for cesium atoms, simultaneously protecting from random spin-exchange collisions and partially also from spin-relaxation by the interaction with weakly-depolarizing walls. We further report an order of magnitude improvement of the quality factor of the magnetic states. Our results demonstrate the ability to use the multi-level structure of atoms or molecules with accessible optical tools to engineer useful decoherence-free subspaces.

Collisions are a fundamental relaxation mechanism in warm spin gases. At ambient conditions, alkali-metal atoms confined in a glass cell move quickly at hundreds of meters per seconds, collide with the cell walls or randomly pair, and scatter by collisions. Owing to their single valence electron, strong forces acting during collisions correlate the spin state of different colliding atoms [1-3]. In binary collisions the exchange interaction conserves the total spin of the colliding atoms, and does not relax the magnetic moment of the ensemble by itself. In the presence of a magnetic field however, random collisions distribute the atomic state between multiple spin levels whose energy splittings are nonuniform, leading to dephasing of the spin state [4, 5].

Several techniques have been realized to overcome this limitation by operating at a regime known as Spin-Exchange Relaxation Free (SERF). This regime can be accessed by operating under one of a few restricting conditions: setting the absolute magnetic field experienced by the atoms near zero [6–15], applying fast and strong modulation of the magnetic field [17] or maintaining the degree of spin-polarization high [18–20]. Systems operating in this regime are used in a variety of applications including precision sensors [6, 7, 18, 21–25], searches of new physics [26–33], interfacing with the spins noble-gases for imaging and fundamental studies [34–42], and in emerging quantum information applications [2, 10, 15, 43–46]

Continuous suppression of the spin-exchange relaxation using optical methods has been considered via resonant optical-pumping [18, 19], but this approach is limited due to considerable excess relaxation through photons absorption and scattering [47]. Nonetheless, auxiliary off-resonant light fields are an efficient tool to prolong the coherence time of spin ensembles in other configurations, by compensating spatial inhomogenities of external field [48, 49], or by compensating for the inhomogeneous distribution of resonant frequencies of the ensemble's constituents [50–53]. However, for the relaxation by spin-exchange collisions, and partially also by weakly depolarizing walls, the inhomogeneity is related to the internal level-structure, and consequently none of the above techniques directly applies.

Here we describe a new mechanism that provides protection from spin relaxation using an off-resonant auxiliary light. We show that state-selective Zeeman lightshifts applied by an optical beam can protect the alkalimetal spins from spin-exchange relaxation via internal synchronization of the Larmor precession frequencies. We demonstrate the technique for a warm cesium spin ensemble and report up to an order of magnitude suppression of the relaxation rate, providing protection from relaxation of spin-exchange and partially also from the weakly depolarizing walls. Unlike the operation in the standard SERF regime near zero absolute field, which exhibits a small number of measurable spin-precession cycles, here both frequency and number of measurable cycles are kept high. Our technique can be particularly useful in systems using anti-relaxation coated cells where the optical spectrum is well-resolved.

The asynchronous Larmor precession of magnetic moments, which is the underlying cause for the relaxation, is sourced by the level structure of all alkali-metal atoms. Owing to hyperfine interaction between the electron spin S and the nonzero spin I in the nucleus, the spin states in the electronic ground state are collected in two manifolds, characterized with quantum number F = I+S and separated by the gap corresponding to the fast hyperfine frequency ω_{hpf} . In the presence of a magnetic field, the degeneracy of the magnetic levels is lifted by the Zeeman splittings, and the levels are characterized by the additional quantum number $M = S_z + I_z$

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Figure 1. Relaxation of alkali-metal spins and light-shift compensation. a, Spin levels of cesium atoms (I = 7/2)in the electronic ground state. The magnetic levels $|F, M\rangle$ in the F = 4 (F = 3) hyperfine manifolds are Zeeman-splitted by $\hbar\omega_{\rm a}$ ($\hbar\omega_{\rm b}$) for $M \leq |F|$, resulting with asynchronous precession of the magnetic moments; spins precess clockwise at F = 4(blue) but counter-clockwise at F = 3 (cyan). b, Spin-exchange collisions between pairs of atoms, or collisions with weaklydepolarizing walls of the enclosure can lead to random change of the hyperfine manifolds, and give rise to spin-relaxation by the asynchronous precession. c, An off-resonance circularly-polarized optical beam can shift the levels in the two hyperfine manifolds d, Calculated light-shift cross-sections $\sigma_{\rm a}$ (cyan) and $\sigma_{\rm b}$ (blue) of the Zeeman-like shifts in the F = 4 and F = 3manifolds respectively, as a function of the optical frequency ν from the D₁ transitions. Black lines denotes the absorption cross-section of the four transition lines and purple arrow denotes the range for which the protection beam primarily shifts the levels in the F = 3 manifold. e, Applied Zeeman-like shifts can correct for the difference in precession frequencies and protect from dephasing by asynchronous precession.

where |M| < F. The splittings are determined predominantly by the coupling of the magnetic field to the electron spins as shown in Fig. 1. In the upper manifold, the Zeeman splittings between any two adjacent levels M, M+1, and to first order in $g_e B/\omega_{\rm hpf}$ are equal to $\omega_{\rm a} = +\omega_B$. Here g_e is the gyromagnetic ratio of the electron and $\omega_B = g_e B/(2I+1)$ is the Larmor precession rate. In the lower hyperfine manifold however, the Zeeman splitting are inverted and $\omega_{\rm b} = -\omega_B$. The sign change originates from alignment (anti-alignment) of the electron and nuclear spins in the upper (lower) hyperfine manifolds, resulting with clockwise (counterclockwise) precession. For warm gases (at temperature $T \gg \hbar \omega_{\rm hpf}/k_B$, spin-changing collisions redistribute the atomic state between the two hyperfine manifolds at random collisions times as illustrated in Fig. 1b; As $\omega_{\rm a} \neq \omega_{\rm b}$ the spins precession at the two hyperfine manifolds is asynchronous, leading to dephasing of the collective spin-state.

To circumvent this dephasing, we aim to set the

frequencies ω_a and ω_b equal using an auxiliary light field, as shown in Fig. 1c-d. We consider a circularlypolarized beam whose optical axis is aligned with the magnetic field. Setting the optical frequency ν far from the atomic optical transition frequencies ν_i enables to shift the magnetic levels and suppresses photon absorption for $|\Delta_i| \gg \Gamma_e$, where Γ_e is the atomic linewidth and $\Delta_i = \nu - \nu_i$. These Zeeman light-shifts, denoted by $\delta_{\rm a}$ and $\delta_{\rm b}$ for the upper and lower hyperfine manifolds respectively, are proportional to the laser power and depend on the Zeeman light-shift cross-sections $\sigma_{\rm a}(\nu)$ and $\sigma_{\rm b}(\nu)$ [54–56], which are shown in Fig. 1d for the D1 lines of cesium. The relative shifts $\delta_{\rm a}$ and $\delta_{\rm b}$ can therefore be varied by detuning of the optical frequency ν ; This configuration enables synchronization of the Larmor frequencies in the two hyperfine manifolds, $(\omega_{\rm a} = \omega_{\rm b})$ when the resonance condition

$$\omega_B + \delta_a = -\omega_B + \delta_b, \tag{1}$$

is satisfied. It is expected that on resonance, the precession of the magnetic moments would persists contin-



Figure 2. Synchronization of spin precession using light-shifts. a, Fourier spectrum of the measured spin precession of cesium spins at B = 0.43 mG in the presence of the protection beam with power P. The protection beam induces a Zeeman-like field which shifts majorly the precession frequency of the magnetic moment in F = 3 (i.e. ω_b , red dash-dots lines) and to lesser extent also the precession frequency in F = 4 (i.e. ω_a , orange dash line). At the resonance condition $\omega_a = \omega_b$ near P = 9.7 mW the frequencies synchronize and the spectrum is greatly enhanced. **b**, Spin precession without the optical protection beam (top) and in the presence of the protection beam (bottom). At synchronized precession, the coherence time is prolonged five fold.

uously in between sudden collisions, protecting the spins from relaxation.

We study this mechanism using a warm cesium vapor (S = 1/2, I = 7/2) contained in a buffer-gas free and paraffin-coated glass cell. We control the number density of the cesium vapor $n = (2.5 \pm 0.4) \times 10^{11} \,\mathrm{cm}^{-3}$, estimated using absorption spectroscopy [57], by heating of a liquid droplet of cesium. We control the magnetic fields to better than $|B| < 5 \,\mu\text{G}$ using three sets of Helmoltz coils and by magnetically shielding the system from the ambient field using four layers of μ -metal shields. We initially polarize the spins using a weak circularly-polarized optical-pump beam aligned with a background magnetic field along \hat{y} and near resonance with the ν_3 transition. The average degree of spin polarization of the ensemble was kept low to ensure appreciable population of both hyperfine manifolds. Then, we turn off the pump and background field and simultaneously apply a magnetic field B_z and an auxiliary circularly polarized beam that is 12 GHz blue-detuned from the ν_1 transition, selectively exerting different Zeeman light-shifts on the two hyperfine manifold. We monitor the spins' precession using a weak and continuously operating probe beam which propagates along \hat{x} and is linearly polarized. The probe, whose frequency is centered in the D1 lines, experiences polarization rotation that depends on the spin precession of the magnetic moments in both hyperfine manifolds. Using a set of differential photo-diodes in a homodyne configuration we measure the mean spin of the ensemble $\langle S_x(t) \rangle$ [58].

We first measure the spin precession around $B_z =$

0.43 mG as a function of the power of the protection beam P. In Fig. 2a we present the spectral content of the recorded signals, showing the absolute value of the Fast Fourier Transform. For P = 0 the spectral content is peaked at a single frequency, owing to the indistinguishable precession of the two hyperfines which rotate at the same absolute frequency. Increasing P resolves the two precession frequencies $\omega_{\rm a} = \delta_{\rm a}(P) + \omega_B$ and $\omega_{\rm b} = \delta_{\rm b}(P) - \omega_B$, as also indicated by the orange and red lines. At the crossing of these two lines ($\omega_{\rm a} = \omega_{\rm b}$) near P = 9.7mW, the resonance condition is satisfied and strikingly, the spectral amplitude of the signal is greatly enhanced in comparison to lower or zero power of the protection beam, owing to its spectral narrowing. We note that the presented data has no relative normalization between different measurements; all amplitudes were scaled by a single common factor.

In Fig. 2b, we present the time domain signals without the protection beam, i.e. at P = 0 (top), and near the resonance condition at $P = 9.7 \,\mathrm{mW}$ (bottom), corresponding to the black and purple arrows in Fig 2a. We extract the decoherence rate of each measurement by fitting the measured time-domain signal to a single sinusoidal decaying exponent. We find a 5-fold improvement with respect to the decoherence rate without the light-shift, reaching a decoherence rate that is limited by the finite spin-lifetime T_1 . This result demonstrates suppression of spin-relaxation when the precession frequencies of the magnetic moments are synchronized.

We further study the dependence of the relaxation on the magnetic field and repeat the experiment for differ-



Figure 3. Protection from spin relaxation. a, Measured fundamental relaxation rate Γ of cesium vapor as a function of the magnetic field *B* and Power *P* of the optical protection beam. The relaxation is minimized along the resonance line $\omega_{a}(B,P) = \omega_{b}(B,P)$ (white dashed line), providing up to an order of magnitude protection compared to the precession absent light-shifts. b, Calculated fundamental relaxation rate using the hyperfine-Bloch model (see text and [57]). We experimentally observe increased relaxation for $\omega_{b}(B) = 0$ (black dashed line)

ent values of the magnetic field B and light-shift power P. Here we fit the measured signals to a sum of two sinusoidal decaying exponents $\sum_{i=1}^{2} A_i e^{-\gamma_i t} \cos(\omega_i t + \theta_i)$ and present the minimal measured relaxation rate $\Gamma = \min(\gamma_1, \gamma_2)$ in Fig. 3a (see [57]). Evidently, a valley of low relaxation rate Γ is observed along the magnetic fields that satisfy the resonance condition in Eq. (1), as indicated with a white dashed line, protecting from decoherence at magnetic fields that are considerably higher than the values in the SERF regime at P = 0. The relaxation rate weakly increases as a function of the auxiliary light power and independent of the magnetic field, consistent with the measured residual absorption of the auxiliary light. We also measure an increase in the relaxation along the curve for which $\omega_{\rm b}(B, P) = 0$ (black dashed line).

The optically-induced suppression of spin-exchange relaxation is qualitatively different from the suppression without the auxiliary light at low-fields. At low magnetic fields and without optical protection, the number of visible precession cycles, characterized by the unitless parameter $Q = \bar{\omega}/\Gamma$, is about $Q \lesssim 1$ where $\bar{\omega}$ is the precession frequency associated with Γ determined by the fit. In Fig. 4, we present the number of observed cycles in the presence of auxiliary light, where the black line indicates the resonance condition for which $\omega_{\rm a} = \omega_{\rm b}$. Here, the number of visible oscillations along the resonance line is considerably higher, owing to the suppressed relaxation and the higher oscillation frequency $\bar{\omega} = \omega_{\rm a} = \omega_{\rm b}$ which is an increasing function of the magnetic field.

We model the observed phenomena using a simple hyperfine-Bloch picture which is valid in the low-polarization regime [4, 14, 15, 58]. We consider the

dynamics of the ensemble-averaged magnetic spin moments $\langle F_{+}^{(a)} \rangle = \langle F_{x}^{(a)} \rangle + i \langle F_{y}^{(a)} \rangle$ and $\langle F_{+}^{(b)} \rangle = \langle F_{x}^{(b)} \rangle + i \langle F_{y}^{(b)} \rangle$ of the upper and lower hyperfine manifolds respectively. The spin dynamics about a magnetic field and light-shift beam aligned along \hat{z} is given by

$$\frac{d}{dt}\langle F_{+}^{(a)}\rangle = -\left(i\omega_{a} + R_{11}\right)\langle F_{+}^{(a)}\rangle - R_{12}\langle F_{+}^{(b)}\rangle, \quad (2)$$

$$\frac{d}{dt}\langle F_{+}^{(\mathrm{b})}\rangle = -\left(i\omega_{\mathrm{b}} + R_{22}\right)\langle F_{+}^{(\mathrm{b})}\rangle - R_{21}\langle F_{+}^{(\mathrm{a})}\rangle.$$
 (3)



Figure 4. Measured number of visible precession cycles. The number of visible spin-precession cycles, characterized by $Q = \bar{\omega}/\Gamma$, is considerably higher along the resonance line (black dashed line) than the operation at low magnetic-fields where $Q \lesssim 1$. The increase in Q is a unique feature of the optical protection technique which is not limited to low magnetic fields.

The four coefficients R_{11}, R_{12}, R_{21} and R_{22} comprise the relaxation-rate matrix which depends on the nuclear spin I = 7/2; it can be decomposed by the contribution of different relaxation processes, which we estimate in [57] as spin-exchange at a rate $R_{\rm se} = (170 \pm 30) \, {\rm s}^{-1}$, destruction of the electron (total) spin at a rate $R_{\rm sr} =$ $(85 \pm 15) \,\mathrm{s}^{-1} \ (R_{\rm u} = (10 \pm 3) \,\mathrm{s}^{-1})$, both due to collisions with the weakly-depolarizing paraffin-coated walls, and additional relaxation associated with off-resonant absorption of the auxiliary beam's photons $R_{\rm P}$. We take the values of $\omega_{\rm a}(B, P)$ and $\omega_{\rm b}(B, P)$ associated with our experimental settings and diagonalize Eqs. (2-3) for different values of B, P. In Fig. 3b we present the minimal real part of the eigenvalues, which is associated with the relaxation of the least decaying spin mode. Evidently, the model captures the main features of the experimental observation, demonstrating that synchronization of the Larmor precession using vector light-shifts provides protection of the magnetic moments from spin-exchange relaxation.

The hyperfine-Bloch model allows for further comparison between the optical protection and the suppressed relaxation that is realized at low magnetic fields in the SERF regime. Considering the limit of rapid spinexchange collisions $R_{\rm se} \gg R_{\rm sr}, R_{\rm u}$ for simplification, the fundamental relaxation rate is attained when the precession frequencies are synchronous, and is given by $\Gamma_0 = R_{\rm u} + \frac{1}{22}R_{\rm sr}$. This relaxation rate is maintained as long as $|\omega_{\rm a} - \omega_{\rm b}| \lesssim \sqrt{R_{\rm se}\Gamma}$ (see [57]), which characterizes the width of the optically protected valley along the resonance condition line in Fig. 3. In the SERF regime and absent optical protection, $\omega_{\rm a} = -\omega_{\rm b}$ and the precession frequency grows linearly with the magnetic field ($\bar{\omega} \approx \frac{4}{11}\omega_{\rm a}$) but the relaxation rate $\Gamma \approx \Gamma_0 + 0.3 |\omega_{\rm a} - \omega_{\rm b}|^2 / R_{\rm se}$ grows quadratically in the magnetic field, leading to decreased Q for $\sqrt{\Gamma_0 R_{\rm se}} \ll \omega_{\rm a} \ll R_{\rm se}$ [57]. In contrast, in the presence of optical protection $\bar{\omega} \approx \omega_{\rm a}$ and the relaxation is free of spin-exchange and independent of the magnetic field ($\Gamma \approx \Gamma_0$), thus yielding considerably higher Q.

In summary, we have studied the spin-relaxation of a warm spin-gas and showed that it originates from internal asynchronization of the Larmor precession frequencies associated with the universal level structure of alkali-metal atoms. We demonstrated experimentally and theoretically that off-resonant beam can induce relative Zeeman light-shifts which correct for the asynchronized precession and protect from relaxation of the spins, increasing the coherence time by up to an order of magnitude.

Our findings give direct validation to the role of the asynchronous Larmor frequencies on the relaxation, as was first proposed in the statistical model in Ref. [4] for the spin-exchange interaction. Furthermore, this technique can find potential usage in applications and experiments for which the optical spectrum is well resolved (e.g. cells with low buffer-gas pressures) [45, 59–68], allowing for selective action of light-shifts on the different hyperfine manifolds.

We thank Roy Shaham, Ohad Yogev, Gil Ronen, Yaron Artzi and Tal David for fruitful discussions.

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- W. Happer and H. Tang, Spin-exchange shift and narrowing of magnetic resonance lines in optically pumped alkali vapors, Physical Review Letters **31**, 273 (1973).
- [2] K. Mouloudakis and I. Kominis, Spin-exchange collisions in hot vapors creating and sustaining bipartite entanglement, Physical Review A 103, L010401 (2021).
- [3] O. Katz and O. Firstenberg, Synchronization of strongly interacting alkali-metal spins, Physical Review A 98, 012712 (2018).
- [4] W. Happer and A. Tam, Effect of rapid spin exchange on the magnetic-resonance spectrum of alkali vapors, Physical Review A 16, 1877 (1977).
- [5] I. Savukov and M. Romalis, Effects of spin-exchange collisions in a high-density alkali-metal vapor in low magnetic fields, Physical Review A 71, 023405 (2005).
- [6] I. Kominis, T. Kornack, J. Allred, and M. V. Romalis, A subfemtotesla multichannel atomic magnetometer, Nature 422, 596 (2003).
- [7] H. Dang, A. C. Maloof, and M. V. Romalis, Ultrahigh sensitivity magnetic field and magnetization measurements with an atomic magnetometer, Applied Physics Letters 97, 151110 (2010).
- [8] M. Ledbetter, I. Savukov, V. Acosta, D. Budker, and M. Romalis, Spin-exchange-relaxation-free magnetometry with cs vapor, Physical Review A 77, 033408 (2008).
- [9] M. Balabas, T. Karaulanov, M. Ledbetter, and D. Budker, Polarized alkali-metal vapor with minute-long transverse spin-relaxation time, Physical review letters 105, 070801 (2010).
- [10] J. Kong, R. Jiménez-Martínez, C. Troullinou, V. G. Lucivero, G. Tóth, and M. W. Mitchell, Measurementinduced, spatially-extended entanglement in a hot, strongly-interacting atomic system, Nature communications 11, 1 (2020).
- [11] R. Jimenez-Martinez, S. Knappe, and J. Kitching, An optically modulated zero-field atomic magnetometer with suppressed spin-exchange broadening, Review of Scientific Instruments 85, 045124 (2014).
- [12] O. Katz, M. Dikopoltsev, O. Peleg, M. Shuker, J. Steinhauer, and N. Katz, Nonlinear elimination of spinexchange relaxation of high magnetic moments, Physical review letters **110**, 263004 (2013).
- [13] W. Chalupczak, P. Josephs-Franks, B. Patton, and S. Pustelny, Spin-exchange narrowing of the atomic ground-state resonances, Physical Review A 90, 042509 (2014).
- [14] W. Xiao, T. Wu, X. Peng, and H. Guo, Atomic spinexchange collisions in magnetic fields, Physical Review A 103, 043116 (2021).

- [15] K. Mouloudakis, G. Vasilakis, V. Lucivero, J. Kong, I. Kominis, and M. Mitchell, Effects of spin-exchange collisions on the fluctuation spectra of hot alkali-metal vapors, Physical Review A 106, 023112 (2022).
- [16] M. Dikopoltsev, A. Berrebi, U. Levy, and O. Katz, Suppressing the decoherence of alkali-metal spins at low magnetic fields, arXiv preprint arXiv:2209 (2022).
- [17] A. Korver, R. Wyllie, B. Lancor, and T. Walker, Suppression of spin-exchange relaxation using pulsed parametric resonance, Physical review letters **111**, 043002 (2013).
- [18] Y.-Y. Jau, A. Post, N. Kuzma, A. Braun, M. Romalis, and W. Happer, Intense, narrow atomic-clock resonances, Physical review letters **92**, 110801 (2004).
- [19] S. Smullin, I. Savukov, G. Vasilakis, R. Ghosh, and M. Romalis, Low-noise high-density alkali-metal scalar magnetometer, Physical Review A 80, 033420 (2009).
- [20] D. Sheng, S. Li, N. Dural, and M. V. Romalis, Subfemtotesla scalar atomic magnetometry using multipass cells, Physical review letters **110**, 160802 (2013).
- [21] T. Kornack, R. Ghosh, and M. V. Romalis, Nuclear spin gyroscope based on an atomic comagnetometer, Physical review letters 95, 230801 (2005).
- [22] Y. Yang, D. Chen, W. Jin, W. Quan, F. Liu, and J. Fang, Investigation on rotation response of spinexchange relaxation-free atomic spin gyroscope, IEEE Access 7, 148176 (2019).
- [23] J. Fang, S. Wan, J. Qin, C. Zhang, and W. Quan, Spinexchange relaxation-free magnetic gradiometer with dual-beam and closed-loop faraday modulation, JOSA B 31, 512 (2014).
- [24] I. M. Savukov, Spin exchange relaxation free (serf) magnetometers, in *High Sensitivity Magnetometers* (Springer, 2017) pp. 451–491.
- [25] E. Zhivun, M. Bulatowicz, A. Hryciuk, and T. Walker, Dual-axis π -pulse magnetometer with suppressed spinexchange relaxation, Physical review applied **11**, 034040 (2019).
- [26] T. Wang, D. F. J. Kimball, A. O. Sushkov, D. Aybas, J. W. Blanchard, G. Centers, S. R. O'Kelley, A. Wickenbrock, J. Fang, and D. Budker, Application of spinexchange relaxation-free magnetometry to the cosmic axion spin precession experiment, Physics of the dark universe 19, 27 (2018).
- [27] I. M. Bloch, G. Ronen, R. Shaham, O. Katz, T. Volansky, and O. Katz, New constraints on axion-like dark matter using a floquet quantum detector, Science advances 8, eabl8919 (2022).
- [28] G. Vasilakis, J. Brown, T. Kornack, and M. Romalis, Limits on new long range nuclear spin-dependent forces set with a k- he 3 comagnetometer, Physical review letters 103, 261801 (2009).
- [29] D. Budker, P. W. Graham, M. Ledbetter, S. Rajendran, and A. O. Sushkov, Proposal for a cosmic axion spin precession experiment (casper), Physical Review X 4, 021030 (2014).
- [30] M. Safronova, D. Budker, D. DeMille, D. F. J. Kimball, A. Derevianko, and C. W. Clark, Search for new physics with atoms and molecules, Reviews of Modern Physics 90, 025008 (2018).
- [31] D. Budker, T. Cecil, T. E. Chupp, A. A. Geraci, D. F. J. Kimball, S. Kolkowitz, S. Rajendran, J. T.

Singh, and A. O. Sushkov, Quantum sensors for high precision measurements of spin-dependent interactions, arXiv preprint arXiv:2203.09488 (2022).

- [32] Y. Wang, H. Su, M. Jiang, Y. Huang, Y. Qin, C. Guo, Z. Wang, D. Hu, W. Ji, P. Fadeev, *et al.*, Limits on axions and axionlike particles within the axion window using a spin-based amplifier, Physical Review Letters **129**, 051801 (2022).
- [33] M. Padniuk, M. Kopciuch, R. Cipolletti, A. Wickenbrock, D. Budker, and S. Pustelny, Response of atomic spin-based sensors to magnetic and nonmagnetic perturbations, Scientific reports 12, 1 (2022).
- [34] T. R. Gentile, P. Nacher, B. Saam, and T. Walker, Optically polarized he 3, Reviews of modern physics 89, 045004 (2017).
- [35] K. Coulter, A. McDonald, W. Happer, T. Chupp, and M. E. Wagshul, Neutron polarization with polarized 3he, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 270, 90 (1988).
- [36] T. G. Walker and W. Happer, Spin-exchange optical pumping of noble-gas nuclei, Reviews of modern physics 69, 629 (1997).
- [37] T. Chupp and S. Swanson, Medical imaging with laserpolarized noble gases, in Advances in Atomic, Molecular, and Optical Physics, Vol. 45 (Elsevier, 2001) pp. 41–98.
- [38] A. Tsinovoy, O. Katz, A. Landau, and N. Moiseyev, Enhanced coupling of electron and nuclear spins by quantum tunneling resonances, Physical Review Letters 128, 013401 (2022).
- [39] W. T. Lee, G. Zheng, C. L. Talbot, X. Tong, T. D'Adam, S. R. Parnell, M. De Veer, G. Jenkin, G. R. Polglase, S. B. Hooper, *et al.*, Hyperpolarised gas filling station for medical imaging using polarised 129xe and 3he, Magnetic Resonance Imaging **79**, 112 (2021).
- [40] S. Katugampola, C. Jantzi, D. A. Keder, G. W. Miller, V. Nelyubin, H. Nguyen, S. Tafti, W. A. Tobias, and G. D. Cates, Frequency shifts in the epr spectrum of ³⁹k due to spin-exchange collisions with polarized ³he and precise ³he polarimetry, arXiv preprint arXiv:2109.04375 (2021).
- [41] R. Shaham, O. Katz, and O. Firstenberg, Strong coupling of alkali-metal spins to noble-gas spins with an hour-long coherence time, Nature Physics 18, 506 (2022).
- [42] O. Katz, R. Shaham, and O. Firstenberg, Coupling light to a nuclear spin gas with a two-photon linewidth of five millihertz, Science advances 7, eabe9164 (2021).
- [43] O. Katz, R. Shaham, E. S. Polzik, and O. Firstenberg, Long-lived entanglement generation of nuclear spins using coherent light, Physical Review Letters **124**, 043602 (2020).
- [44] O. Katz and O. Firstenberg, Light storage for one second in room-temperature alkali vapor, Nature communications 9, 1 (2018).
- [45] V. Guarrera, R. Gartman, G. Bevilacqua, and W. Chalupczak, Spin-noise spectroscopy of a noisesqueezed atomic state, Physical Review Research 3, L032015 (2021).
- [46] O. Katz, R. Shaham, and O. Firstenberg, Quantum interface for noble-gas spins based on spin-exchange col-

lisions, PRX quantum **3**, 010305 (2022).

- [47] R. Han, M. Balabas, C. Hovde, W. Li, H. M. Roig, T. Wang, A. Wickenbrock, E. Zhivun, Z. You, and D. Budker, Is light narrowing possible with dense-vapor paraffin coated cells for atomic magnetometers?, AIP Advances 7, 125224 (2017).
- [48] A. Radnaev, Y. Dudin, R. Zhao, H. Jen, S. Jenkins, A. Kuzmich, and T. Kennedy, A quantum memory with telecom-wavelength conversion, Nature Physics 6, 894 (2010).
- [49] R. Finkelstein, O. Lahad, I. Cohen, O. Davidson, S. Kiriati, E. Poem, and O. Firstenberg, Continuous protection of a collective state from inhomogeneous dephasing, Physical Review X 11, 011008 (2021).
- [50] C. Cohen-Tannoudji, F. Hoffbeck, and S. Reynaud, Compensating doppler broadening with light-shifts, Optics Communications 27, 71 (1978).
- [51] S. Reynaud, M. Himbert, J. Dupont-Roc, H. Stroke, and C. Cohen-Tannoudji, Experimental evidence for compensation of doppler broadening by light shifts, Physical Review Letters 42, 756 (1979).
- [52] D. Yavuz, N. Brewer, J. Miles, and Z. Simmons, Suppression of inhomogeneous broadening using the ac stark shift, Physical Review A 88, 063836 (2013).
- [53] O. Lahad, R. Finkelstein, O. Davidson, O. Michel, E. Poem, and O. Firstenberg, Recovering the homogeneous absorption of inhomogeneous media, Physical Review Letters **123**, 173203 (2019).
- [54] B. Mathur, H. Tang, and W. Happer, Light shifts in the alkali atoms, Physical Review 171, 11 (1968).
- [55] W. Happer and B. Mathur, Effective operator formalism in optical pumping, Physical Review 163, 12 (1967).
- [56] C. Cohen-Tannoudji and J. Dupont-Roc, Experimental study of zeeman light shifts in weak magnetic fields, Physical Review A 5, 968 (1972).
- [57] See Supplemental Material at [URL inserted by publisher] for further details on the estimation of rates, data analysis of Fig. 3, and analytical calculation of the quality factor.
- [58] O. Katz, O. Peleg, and O. Firstenberg, Coherent coupling of alkali atoms by random collisions, Physical Re-

view Letters **115**, 113003 (2015).

- [59] S. Afach, D. Budker, G. DeCamp, V. Dumont, Z. D. Grujić, H. Guo, D. J. Kimball, T. Kornack, V. Lebedev, W. Li, et al., Characterization of the global network of optical magnetometers to search for exotic physics (gnome), Physics of the Dark Universe 22, 162 (2018).
- [60] S. Afach, B. C. Buchler, D. Budker, C. Dailey, A. Derevianko, V. Dumont, N. L. Figueroa, I. Gerhardt, Z. D. Grujić, H. Guo, *et al.*, Search for topological defect dark matter with a global network of optical magnetometers, Nature Physics **17**, 1396 (2021).
- [61] O. Katz and O. Firstenberg, Transverse optical pumping of spin states, Communications Physics 2, 1 (2019).
- [62] N. Castagna, G. Bison, G. Di Domenico, A. Hofer, P. Knowles, C. Macchione, H. Saudan, and A. Weis, A large sample study of spin relaxation and magnetometric sensitivity of paraffin-coated cs vapor cells, Applied Physics B 96, 763 (2009).
- [63] C. B. Møller, R. A. Thomas, G. Vasilakis, E. Zeuthen, Y. Tsaturyan, M. Balabas, K. Jensen, A. Schliesser, K. Hammerer, and E. S. Polzik, Quantum back-actionevading measurement of motion in a negative mass reference frame, Nature 547, 191 (2017).
- [64] B. Julsgaard, J. Sherson, J. I. Cirac, J. Fiurášek, and E. S. Polzik, Experimental demonstration of quantum memory for light, Nature 432, 482 (2004).
- [65] B. Julsgaard, A. Kozhekin, and E. S. Polzik, Experimental long-lived entanglement of two macroscopic objects, Nature 413, 400 (2001).
- [66] H. Bao, J. Duan, S. Jin, X. Lu, P. Li, W. Qu, M. Wang, I. Novikova, E. E. Mikhailov, K.-F. Zhao, K. Mølmer, H. Shen, and Y. Xiao, Spin squeezing of 1011 atoms by prediction and retrodiction measurements, Nature 581, 159 (2020).
- [67] W. Qu, S. Jin, J. Sun, L. Jiang, J. Wen, and Y. Xiao, Sub-hertz resonance by weak measurement, Nature communications 11, 1 (2020).
- [68] T. Chupp, P. Fierlinger, M. Ramsey-Musolf, and J. Singh, Electric dipole moments of atoms, molecules, nuclei, and particles, Reviews of Modern Physics 91, 015001 (2019).