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Communication

Generation of laser-polarized xenon using fiber-coupled laser-diode arrays narrowed with integrated volume holographic gratings

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Generation of hyperpolarized noble gases (both fundamental and hyperpolarized gas applications) within the NMR/MRI imaging and fundamental physics applications.

1. Introduction

Alkali-metal spin exchange optical pumping (AMSE OP) can generate high nuclear spin polarization in noble gases [1] for a variety of NMR/MRI [2,3] and fundamental physics [4] applications. While different types of light sources have been used to prepare spin-polarized gases via AMSE OP (e.g., Refs. [1–12]), laser-diode arrays (LDAs) have become increasingly dominant because of their high photon flux, low costs, and operational simplicity. However, a key drawback of conventional LDAs is the poor quality of the emitted light—particularly the breadth of their spectral output (compared to a conventional LDA) was achieved with the VHG-LDA's center wavelength tuned to a wing of the Rb D 1 line. Additionally, an anomalous dependence of P Xe on the xenon density within the OP cell is reported—including high P Xe values (>10%) at high xenon partial pressures (~1000 torr).

Volume holographic gratings (VHGs) can be exploited to narrow the spectral output of high-power laser-diode arrays (LDAs) by nearly an order of magnitude, permitting more efficient generation of laser-polarized noble gases for various applications. A ~3-fold improvement in 129Xe nuclear spin polarization, P Xe, (compared to a conventional LDA) was achieved with the VHG-LDA's center wavelength tuned to a wing of the Rb D 1 line. Additionally, an anomalous dependence of P Xe on the xenon density within the OP cell is reported—including high P Xe values (>10%) at high xenon partial pressures (~1000 torr).

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2. VHG-LDAs: background and experimental details

External cavity narrowing [14–16] employs planar dispersive gratings to provide feedback to the individual LDA elements; however the optical alignment is critical to maintain feedback to all the elements to ensure acceptable spectral quality and energy effi...
ciency (typically ~40–66% [15,16]), and whether the gratings can withstand sufficiently high intensities is questionable [15]. Alternatively, VHGs (a.k.a. volume Bragg gratings (VBGs)) can achieve LDA spectral narrowing with efficiencies exceeding 90% [20]. VHGs are bulk slabs of photosensitive glass [21] containing Bragg planes of varying index of refraction; in a VHG-based external-cavity diode laser array (VHG-ECDL, Fig. 1(a)) [22], the VHG retro-reflects a narrow band of the laser emission into the LDA elements, forcing them to lase at the injected wavelength. Barlow et al. recently reported a 120 W VHG-ECDL with output near the Rb D1 (794.76 nm) and with a FWHM of 0.24 nm [17].

Details concerning the LDA light sources utilized here are provided immediately below (other experimental aspects are summarized in Section 5). Both VHG-LDAs used in the present work are engineering prototypes from Spectra-Physics/Newport (‘VL1’ is a “Comet” VHG-LDA module, and ‘VL2’ is an “Integra” turn-key laser system). VL1 was mounted to the same TEC / heat sink used by our original 40 W ‘standard’ LDA (Coherent FAP-B), is driven with the same 65 A diode and TEC-drivers (Newport), and fiber-couples to the same circular-polarizer (CP) optics box (Coherent). VL1 employs additional slow-axis collimation (SAC) and achromatic lenses after the VHG (thickness: 1.5 mm[21]) to focus the 19 frequency-locked laser elements into a single-core 200 μm silica-clad fiber; VL1’s spectral profile is narrowed by nearly an order of magnitude compared to that of our previous LDA (Fig 1(b)), while providing nearly the same power.

VL2 combines two VHG-LDA modules (each similar to VL1) but with simplified optical arrangements: Instead of using FAC or SAC lenses, individual optical fibers (400 μm) are brought within ~600 μm of each laser element (giving 19 fibers/module). The close proximity ensures that each diverging fast-axis beam is launched into its fiber, but requires each module’s VHG (placed between the LDA elements and the fibers) to be thin (500 μm)—yielding somewhat reduced spectral narrowing and lineshape quality (Fig. 1(b)). Obtaining the optimal spectral output from VL2 required overdriving the current (~104%), giving nearly twice the power of VL1—albeit with nearly twice the spectral width.

The 1 m optical fibers used with VL1 and the standard LDA maintain much of their original linear polarization; this ‘memory’ of the LDAs’ polarizations affects the power ratio of the two side-by-side circularly-polarized beams emitted from the CP box—causing the ratio of the beam intensities to be sensitive to the strain on the fiber. For the ‘standard’ LDA, VL1 and VL2, the nominal beam intensity ratios (straight vs. angled) were ~72:28, ~80:20, and ~55:45, respectively (because of its design, VL2’s beam ratio is much closer to unity and less amenable to variation by fiber straining).

Whereas tuning of conventional LDAs may be achieved by varying the LDA temperature (~0.3 nm/°C), the nature of the VHGFedback makes tuning VHG-narrowed LDAs a non-trivial function of LDA temperature and VHG temperature (itself primarily determined by the laser power). A change in the VHG temperature induces a slight change in the element spacing within the VHG, altering the laser’s spectral profile. For example, increased driving current for VL1 (Fig. 2(a)) provides greater laser flux in addition to red-shifting of the centroid towards the Rb D1 center (~0.1 nm/amp, with operational range of ~0.3 nm); the linewidth also increases. As with VL1, the output for VL2 red-shifts with increased current, but at a reduced rate (~0.006 nm/amp, as a consequence of its design differences); at 94 A (i.e., 47 A/diode, 100% current, 51.6 W at cell) the centroid is (~0.18 nm from the Rb D1 (‘blue’ side) with a diode temperature of 25 °C, and ~0.09 nm from D1 at 80 °C.

Fig. 2. (a) Spectral output of VL1 as a function of laser diode driving current, showing the lineshape broadening and red-shifting (Δλ) of the centroid as the laser power increases. FWHM examples: at 40 A: ~0.24 nm; 52 A: ~0.27 nm; 62 A: ~0.33 nm. Position of Rb D1 at 794.76 nm (air referenced) is included for comparison. (b) Spectral output of VL2 as a function of laser diode driving current (37.5 W: black; 44.5 W: red; 51.6 W: green). Overall lineshape improves with increasing driving current up to its maximum value, and is slightly red-shifted. Inset: Close-up view of VL2 laser output near the Rb D1 line. The less-efficient implementation of the VHG into the design of VL2 (due in part to the requirement of using thinner VHGs) results in reduced lineshape quality at lower driving currents, particularly regarding the presence of additional features and noise from the main line—indicative of incomplete VHG locking. Such features were not minimized until VL2 was driven at 104% current (96 A, 54.5 W at cell; not shown). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this paper.)
3 Results and discussion

3.1. Dependence of \( P_{\text{Sc}} \) on VHG-LDA excitation profile

With \( \text{VL1} \), \( P_{\text{Sc}} \) is maximized with a driving current of 52 A (Fig. 3(a))—corresponding to an incident power, FWHM, and \( \text{Rb} \) \( \text{D}_1 \) offset of \( \sim 27 \text{ W}, \sim 0.27 \text{ nm}, \) and \( \sim (-3.0) \text{ nm} \), respectively. Despite this blue-shifted offset the laser is absorbed quasi-homogeneously by the cell’s \( \text{Rb} \) vapor (i.e., with little distortion of the spectral profile of the transmitted beam besides a small shift; Fig. 3(b))—indicating that the amount of pressure-broadening (\( \text{D}_1 \) FWHM \( \sim 0.1 \text{ nm} \) [13]) present under these conditions is already sufficient to achieve efficient absorption of the VHG-narrowed light. When utilizing conventional broadband LDAs, the \( \text{Rb} \) \( \text{D}_1 \) absorption line is typically ‘hole-burned’ from the broad and uneven spectral profile—even at the highest described OP cell pressures and temperatures/\( \text{Rb} \) vapor densities. The optimal \( \text{VL1} \) driving current also provides the highest peak intensity (W vs. FWHM; Fig 2(a)); however, it does not give the highest integrated power or the highest intensity at the \( \text{Rb} \) \( \text{D}_1 \). Indeed, higher currents yield increased laser flux, as well as red-shifting of the centroid toward the \( \text{D}_1 \) center—but reduced \( P_{\text{Sc}} \) nearer to true \( \text{Rb} \) \( \text{D}_1 \) resonance (Fig. 3(a)).

The narrowed lineshape of \( \text{VL1} \) allowed small changes in the amount of laser light transmitted through the cell to be recorded when the external magnetic field (\( B_0 \)) was cycled (Fig. 3(b))—thereby potentially providing an \textit{in situ} measure of \( P_{\text{Sc}} \). This effect—resulting from \( \text{Rb} \) \( \text{D}_1 \) ‘bleaching’ due to depletion pumping of the ground-state magnetic sublevels—has been measured previously using high-resolution lasers [8], but would be difficult to observe with conventional broadband sources. More specifically, this first step of the OP process—whereby the \( \text{Rb} \) electrons become spin-polarized—leaves a smaller concentration of \( \text{Rb} \) atoms in the laser-absorbing \( m_I \) state (neglecting \( \text{Rb} \) nuclear spin sublevels). In turn, this population reduction allows slightly more laser light to be transmitted through the cell—allowing \( P_{\text{Sc}} \) to be estimated from the difference.

Here, the spectral profile of the laser light transmitted through the cell was monitored using a near-IR spectrometer via a pinhole behind the cell. Using a simplistic model (see Appendix) the average \( \text{Rb} \) polarization along the cell’s center axis can be estimated using the relation: \(||P_{\text{Sc}}|\rangle = (A/A_0) - 1 \), where \(||P_{\text{Sc}}|\rangle \) is the absolute value of the average \( \text{Rb} \) electron spin polarization along the cell’s center, and \( A \) and \( A_0 \) are the laser absorbances when \( B_0 \) is ‘on’ and ‘off’, respectively (Fig. 3(a)). Despite some run-to-run variability, these results may explain the observed \( \text{D}_1 \) offset dependence of \( P_{\text{Sc}} \): When \( \text{VL1} \) is tuned far from \( \text{D}_1 \) (e.g., with a LDA current of \( \sim 48–50 \) A), \( P_{\text{Sc}} \) tracks \( P_{\text{Sc0}} \) and both are relatively low (while the laser transmittance is high). However when the laser nears resonance (\( \sim 58–60 \) A), \( P_{\text{Sc}} \) along the cell’s center axis is high, but \( P_{\text{Sc0}} \) is relatively low: here the low laser transmittance at the cell’s center directly implies even poorer illumination within a significant portion of the cell’s rear and outer regions. Indeed, \( P_{\text{Sc}} \) is greatest with an intermediate \( \text{D}_1 \) offset where both \( P_{\text{Sc0}} \) and the transmittance are still relatively high—consistent with optimal \( P_{\text{Sc0}} \) requiring a balance between efficient laser absorption and \textit{global} cell illumination sufficient to minimize xenon depolarization from ‘dark’ \( \text{Rb} \) in those regions.

3.2. Comparisons of LDA light sources for Xe AMSE OP

Following switching from the standard LDA to \( \text{VL1} \), we observed a nearly three-fold (W-for-W) improvement in \( P_{\text{Sc}} \) (Fig. 4). The dependences of \( P_{\text{Sc}} \) on cell temperature are similar for the two lasers (with and without VHG-narrowing), with \( \text{VL1} \) peaking at a temperature within ~10 °C of the standard LDA. The relatively mild optimal cell temperature, combined with the \( \text{D}_1 \) offset and the modest fraction of laser light absorbed during optimal conditions, are consistent with the OP being laser-power-limited—despite the high resonant power. Nevertheless, increasing \( \text{VL1} \)’s current to provide greater incident laser flux and improved \( \text{D}_1 \) resonance yielded markedly decreased \( P_{\text{Sc}} \) values across the entire temperature range (Fig. 4; consistent with the results described in Fig. 3 above).

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\[4\] Separately, we note it was also possible to utilize the violet (60–55) emission from \( \text{Rb}_\text{g} \text{Rb} \), energy-pooling processes (e.g., [23]) as a sensitive \textit{in situ} indicator of laser detuning from the \( \text{Rb} \) \( \text{D}_1 \) line (when observing the cell under conditions non-optimal for AMSE OP—i.e., low \( N_2 \) conditions); somewhat surprisingly, the wave-lengths and relative intensities of the \( \text{Rb} \) lines were more sensitive than expected [13] to buffer gas composition—observations that will be pursued and reported in greater detail elsewhere.

\[5\] Note that this work describes several sets of experiments that were performed over time under different conditions; correspondingly, while one may draw quantitative comparisons among data sets within the same experiment (e.g. Fig. 4), it is difficult to meaningfully compare absolute \( P_{\text{Sc}} \) values among different experiments.
In an initial study using VL1, we observed that the reader is referred to the web version of this paper.)

The nearly identical Rb D1 offsets for VL1 and VL2 under nominal conditions (~0.11 and ~0.09 nm, respectively) enabled their performance for LP Xe generation to be compared. To allow a more direct comparison, the optical fiber for VL1 was carefully strained until the CP beam ratio was reduced to that of VL2. The LP Xe NMR spectra obtained at 9.4 T indicated P_xe enhancement factors of ~8800 (P_xe ~ 7.8%) and ~16,000 (P_xe ~ 14.3%) when using VL1 vs. VL2 (respectively). Although VL1 has a narrower spectral output, this advantage translated into only a small W-for-W P_xe benefit; thus at least under the given conditions, VL2's output was sufficiently narrow to allow its higher total power to be exploited.

In later experiments, further OP optimization with VL1 as the light source (simultaneous with improved cell preparation) provided routine P_xe values of ~15–17% and ~21–24% (with and without cryo-collection), consistent with ~75% of P_xe surviving the cryo-collection and transfer processes [23].

3.3. Dependence of Xe cell density and laser power on OP

Lastly, the minimal change in spectral profile of VL2 with increasing current was exploited to examine the dependence of P_xe on laser flux at different Xe partial pressures. P_xe would generally be expected to decrease smoothly and dramatically with increasing Xe density; the xenon polarization becomes 'capped' by corresponding reductions in the values of P_rb that can be achieved (given the strong contribution to the Rb spin-destruction rate from increased Rb/Xe collisions [1,26]). Thus, most AMSE OP setups typically employ relatively low running Xe partial pressures (~1–70 torr) to achieve high (>10%) P_xe values—potentially limiting some applications that simultaneously require both high Xe densities/amounts and high P_xe.

Instead, we have reproducibly observed a different trend (Fig. 5) wherein P_xe initially rises with increasing [Xe]_cell peaks, and then falls off but remains relatively high (>10%)—even at ~1000 torr Xe—to give among the highest P_xe yet reported at such high [Xe]_cell values. We note that a similar qualitative trend was also observed in an initial study using VL1 [19] and the effect does not appear to be an artifact of cell contamination or variances in Xe collection efficiency. Moreover, laser energy-dependent mechanisms (e.g., involving highly elevated internal temperatures of the N2 buffer gas [27]) may affect the OP process, but do not appear to cause the observed dependence of [Xe]_cell on P_xe given that similar trend lines are observed at various incident laser powers (Fig. 5). Indeed, the data shows the potential utility of additional laser flux, as P_xe rises almost linearly with power (except at low [Xe]_cell—Fig. 5 inset). While the origin of the anomalous dependence of P_xe on [Xe]_cell is not yet understood, ongoing studies with in situ low-field NMR polarimetry [28] indicate that the effect involves an interplay between [Xe]_cell and the overall cell temperature—suggesting a route to further P_xe improvements as these effects are explored in greater detail.

4. Conclusions

We have shown that the LDA line-narrowing endowed by VHGs can improve spin-polarized gas production under relatively mild conditions—demonstrating the utility of VHGs for fundamental AMSE OP studies, as well as for a variety of NMR/MRI applications. While altering the spectral profiles of the VHGs utilized here generally requires varying the output power, next-generation VHGs capable of providing true tunability at the Rb D1 wavelength should become available in the near future [28–30]—a desirable capability in light of the observed dependence of P_xe on the D1 offset. Finally, while the present data were derived from 'batch-mode' Rb129 Xe OP, we expect that the demonstrated advantages will translate well to OP setups employing alternate configurations [7,11,12,31–34], alkali metals [1,35], and/or noble gas isotopes [1,36,37].

5. Experimental

The non-LDA components of the Xe OP apparatus have been described previously [23]. Briefly, the OP apparatus—centered around a Rosen [9] cell (75 cc inner cell volume) residing in a Helmholtz coil (HC) pair (22' i.d., ~30 G, Walker Magnetics)—permits operation in either batch or ‘stop-flow’ mode and allows for arbitrary gas mixtures of Xe, N2, and He. Nominal OP conditions: single-batch-mode OP; binary Xe/N2 gas mixture; cell gas pressures: 300 torr Xe, backfilled with N2 gas to give ~2000 torr total pressure; and 15 min. OP time. Following OP, the Xe may be efficiently collected by using a spiral glass liquid-N2-cooled condenser resid-
ing in a strong (~1500 G) field provided by two permanent magnets (Indigo Inst.); alternatively, the contents of the cell can be simply expanded into an evacuated volume including the NMR tube. Optical emission from the lasers and/or excited Rb vapor was monitored via fiber optic probes connected to UV/visible and near-IR portable spectrometers (Ocean Optics USB2000 and HR2000+, respectively). High-field 129Xe NMR spectra were recorded at 9.4 T (110.6 MHz) using a Varian Inova spectrometer. Laser-polarized (LP) Xe spectra were obtained with a single 1 ms rf pulse (\(\alpha = 6.7^{\circ}\)); correspondingly, thermally polarized Xe signals were obtained from the same sample following careful addition of sufficient O2 gas to reduce the 129Xe T1 (to a few s) to permit signal averaging (\(\alpha = 90^{\circ}\); typical recycle delay = 20 s).

**Note Added in Proof**

Very recently, continued optimization of our OP setup with VL1 (aided by low-field nuclear polarimetry and exploitation of the apparent dependence of the optimal cell temperature upon [Xe] cell) has yielded further improvements in the xenon nuclear spin polarization at high xenon densities (including PXe values of ~55%, ~32%, ~23%, and ~11% at 50, 300, 500, and 2000 torr Xe, respectively); these results will be investigated and presented in greater detail elsewhere.

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**Appendix A**

Changes in the spectral profiles of the transmitted laser light caused by cycling \(B_0\) can be integrated to estimate the average Rb polarization along the central axis of the cell \(\langle P_{Rb}(z)\rangle\) using a simplistic model. While the measurement of \(P_{Rb}\) via optical transmission methods has been the subject of considerable previous work (e.g., Refs. [38–40]), the experimental conditions for the present work were sufficiently different to warrant an alternate approach (and corresponding set of assumptions). Here we assume: a near-work were sufficiently different to warrant an alternate approach (e.g., Refs. [38–40]), the experimental conditions for the present situation methods has been the subject of considerable previous work.

\[ A = -\ln T - \ln \left( \frac{l_{hot}}{l_{cold}} \right), \]  

where \(A\) is the absorbance, \(T\) is the transmittance, and \(l_{hot}\) and \(l_{cold}\) are determined by integrating the intensities of the transmitted laser spectra obtained when a gas-loaded cell is, respectively, ‘hot’ and ‘cold’ (i.e., with and without Rb vapor present). Since only those Rb atoms in one of the two ground-state magnetic sublevels can act as absorbers (say \(m_r = +1/2\) with \(\sigma^+\) light), \(A\) is given by:  

\[ A = \sigma(\lambda) I(N^z), \]  

where \(\sigma(\lambda)\) is the (wavelength-dependent) absorption cross-section for the Rb vapor, \(\lambda\) is the path length, and \(N^z\) is the number density of Rb atoms in the \(m_r = +1/2\) spin state (averaged along the long axis of the cell). Next,

\[ \langle P_{Rb} \rangle = \frac{\langle N^z \rangle - \langle N^- \rangle}{N_T}, \]  

where \(N_T = (N^z) + \langle N^- \rangle\). Since \(P_{Rb} = 0\) when \(B_0 = 0\), \(N^-\) under these conditions is given by: \(\langle N^- \rangle = N_T - \langle N^z \rangle\). The terms \(\sigma(\lambda)\) and \(I\) cancel when \(A\) is divided by \(A_0\) (the absorbance when \(B_0 = 0\)) to give \(\langle A/A_0 \rangle = 2\langle N^- \rangle/N_T\); Rearranging Eq. (A3) and combining with the relationship for \(A/A_0\) gives:

\[ \langle |P_{Rb}| \rangle = \frac{A}{A_0} - 1, \]  

where the absolute value is taken because the differential absorbance will be insensitive to the light helicity.

Because the Rb polarization at each position within the cell will rapidly reach steady state with the resonant laser flux (and because the illumination decreases not only along \(z\) (the long axis of the cell) but radially outward from this central axis as well), the Rb polarization will generally be a function of both \(z\) and \(r\) \(P_{Rb}(z, r)\). Thus since in our experiments the transmitted light is sampled via a pinhole in the beam-stop behind the cell, the value of \(P_{Rb}\) that is provided is really a \(z\)-averaged measurement of \(P_{Rb}\) for \(r \approx 0\), and this \(P_{Rb}\) value can be expected to overestimate the cell-averaged value of \(P_{Rb}\)—particularly under conditions of highly non-uniform illumination (e.g., under high-absorbance conditions). Separately, this approach would underestimate \(P_{Rb}\) if a sufficiently-strong residual magnetic field persisted when \(B_0\) is turned off.

**References**


