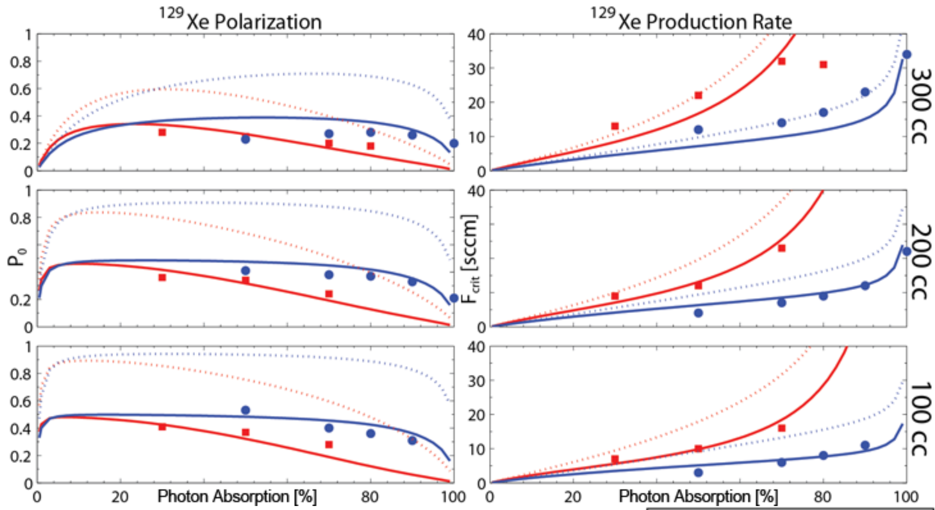
Reconciling the Discrepancy Between Theory and Observed Hyperpolarized ^{129}Xe Polarizer Performance

Matthew S Freeman[1,2], Kiarash Emami[3], and Bastiaan Driehuys[1,2]
[1]*Medical Physics Graduate Program, Duke University, Durham, NC, United States,* [2]*Center for In Vivo Microscopy, Duke University, Durham, NC, United States,*
[3]*Polarean, Inc., Durham, NC, United States*

TARGET AUDIENCE: Hyperpolarized Gas MRI

PURPOSE: Hyperpolarized ^{129}Xe MRI shows great promise as a functional imaging modality, but its range of applications and practical utility still remain limited by polarizer performance. While recent years have seen remarkable demonstrations of high ^{129}Xe polarization[1,2], a fundamental explanation has yet to be provided as to why one polarizer performs better than another. In fact, not a single polarizer described in the literature to date performs in a manner consistent with the "standard model" of optical pumping and spin exchange (SEOP). Absent an explanation of this discrepancy, it is difficult to design polarizers that exhibit optimal ^{129}Xe polarization and production rates in a robust and cost effective manner.

METHODS: A standard ^{129}Xe polarizer (Polarean 9800, Durham, NC) was modified to test its conventional 300 mL cylindrical optical cell, as well as additional custom 200 mL and 100 mL designs. All three cells were pumped using two different lasers – one emitting 111 W, 1.92 nm FWHM (Coherent Dual-FAP, Santa Clara, CA), the other emitting 71 W, 0.39 nm FWHM (QPC, Sylmar, CA). To quantify the performance of each cell/laser combination, we measured ^{129}Xe polarization as a function of the rate at which a lean, 1% Xe mixture flows through the cell from 0.20 to 3.60 standard liters per minute. The resulting flow curves are fit to the function $P(F) = P_0\,(1 - e^{-F_{crit}/F})$ where P_0 is the saturation ^{129}Xe polarization, and F_{crit} is the critical flow rate at which ^{129}Xe atoms spend one spin exchange time constant in the optical cell. These performance metrics were extracted from flow curves acquired at a series of increasing laser absorption levels to evaluate "photon efficiency." These empirical results were compared against those predicted by the most recent standard model, as updated by Norquay, et. al.[4]. Subsequently, the model was extended to include the generation of Rb nanoclusters[3] at a number density proportional to the excited state ($5P_{+½}$) Rb population. Clusters were postulated to have a depolarizing effect on both ^{129}Xe and atomic Rb.



RESULTS: The figure shows the ^{129}Xe polarization and production rate for all 6 systems, overlaid with predictions of the standard model (dashed lines) and the nanocluster model (solid lines). ^{129}Xe polarization is a factor of 2-3 lower than theoretically predicted for most cell/laser combinations at all absorptions. Moreover, ^{129}Xe production rate is reduced by more than 2-fold in the smaller volume optical cells. When the model was revised to include Rb fractal clusters (Rb_n), performance agreed well with observations. The clusters were modeled to reduce ^{129}Xe T_1 in proportion to their number density $[Rb_n]$, with a velocity-averaged cross-section of $\langle\sigma v\rangle = 2 \times 10^{-13}$ cm^{3}s^{-1}, and to cause additional Rb spin-destruction with a velocity-averaged cross-section 4×10^{-7} cm^{3}s^{-1}. This creates a feedback loop whereby optical pumping creates clusters, clusters cause spin destruction, which results in more excited state Rb atoms, and therefore more cluster formation.

DISCUSSION AND CONCLUSIONS: Although a hypothetical construct, the proposed model of Rb_n cluster generation is plausible, as Atutov, et al.[3] recently demonstrated Rb_n as forming in the presence of D1 resonant photons and high [Rb]. Moreover, this single physically-based hypothesis explains a wide range of long-standing discrepancies in high-volume ^{129}Xe polarizer performance. As it stands today, these clusters stand in the way of achieving theoretically predicted arbitrary ability to scale production rates with increasing laser power. A next step is to verify experimentally the presence of such clusters in SEOP systems and develop a clearer understanding of the mechanisms that generate them. This would lead to insight needed to suppress their formation, and thereby recover theoretically predicted performance and scaling of ^{129}Xe production and polarization with laser power. Encouraging recent work of Nikolaou, et al., who report polarization levels close to those predicted by the standard model, suggests that their system does not suffer from cluster generation[2].

ACKNOWLEDGEMENTS: Duke Center for In Vivo Microscopy (NIBIB P41 RR005959), NCI R01 CA142842 **REFERENCES**: **1**. Ruset IC et al.. PRL (2006) 96: 053002. **2**. Nikolaou P, et al. PNAS (2013) 110:35 **3**. Atutov SN, et al. Eur. Phys. J. D (2012) 66: 140 **4**. Norquay G, et al. JAP (2012) 113: 4