Physics-informed machine learning for hyperpolarized solid-state nuclear magnetic resonance

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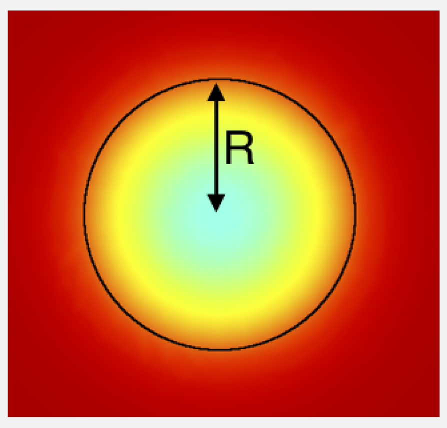
Motivations

- Nuclear magnetic resonance for imaging in the μm range [1].
- Simultaneously solve spin diffusion PDE and learn shape parameters.
- Preliminary results for a spherical shape.

Context

- Setting: spherical solid with unknown radius R in a liquid upon microwave irradiation



- Quantity of interest: polarization $P(r,t)$ at position r and time t.
- Fick diffusion PDE

$$\mathcal{F}\left(P,r,t,R\right)=0 \quad (1)$$

where
$$F\left(P,r,t,R\right)=C_R\left(r\right)\partial_t P\left(r,t\right)-\nabla\left(D_R\left(r\right)C_R\left(r\right)\nabla P\left(r,t\right)\right)+C_R\left(r\right)\frac{P\left(r,t\right)-P_0(t)}{T_R\left(r\right)}$$

with concentration C_R, diffusion rate D_R, relaxation time T_R, local equilibrium polarization P_0.

- Data: for $i\in\{1,\dots,n\}$,
  - $y_i=\frac{1}{R^3}\int_0^R P(r,t)r^2dr$: integral of polarization over the solid.
  - z_i: constant polarization over the liquid (assuming $P(r,t)=P(r',t),\forall r,r'\geq R,\forall t$)
- Goal: estimate radius R and polarization $P(r,t)$ at any position r and time t.

Problem formulation

Change of variable to avoid integration in the optimization process:

$$G\left(r,t\right)=\frac{3}{r^3}\int_0^r P\left(\rho,t\right)\rho^2 d\rho,$$

Problem: solve eq. (1) in G and R with

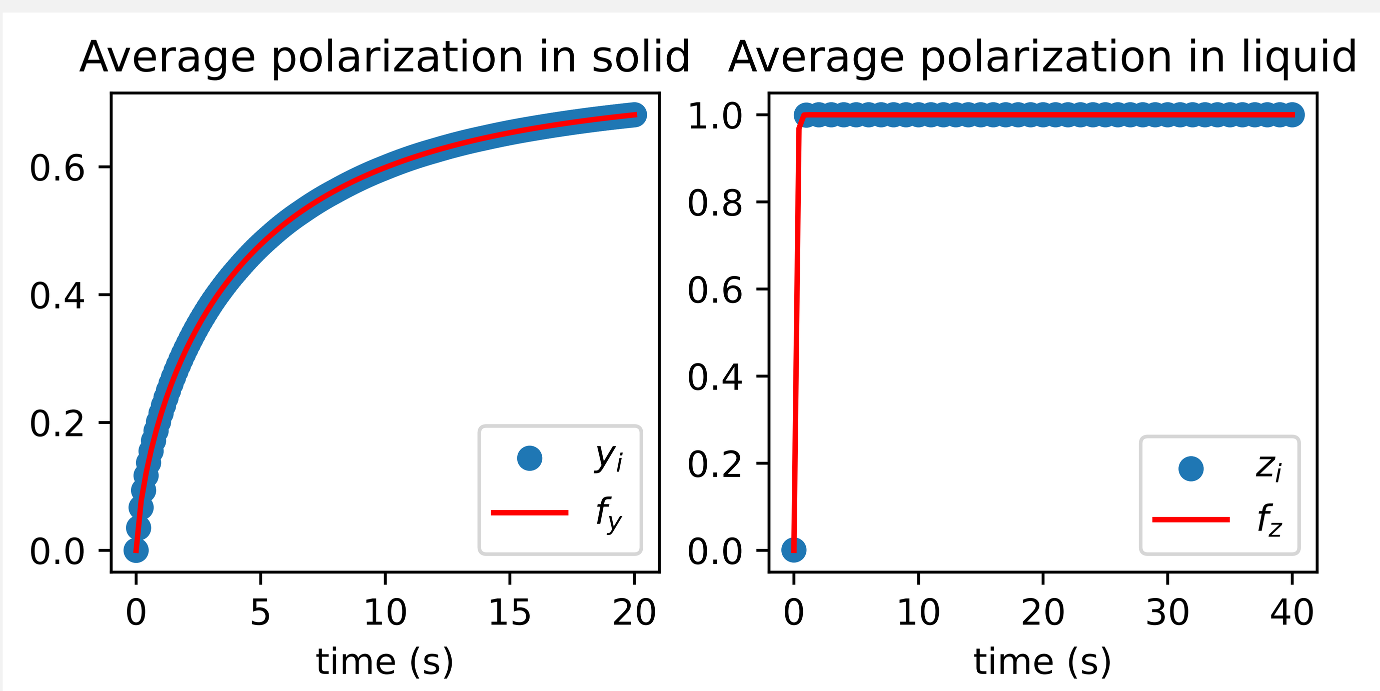
$$\begin{aligned}P\left(r,t\right)&=\frac{r}{3}\partial_r G\left(r,t\right)+G\left(r,t\right)\\ G\left(R,t_i\right)&=y_i,\forall i\in\{1,\dots,n\}\\ P\left(R,t_i\right)&=z_i,\forall i\in\{1,\dots,n\}\\ G\left(r,0\right)&=0,\forall r\end{aligned}$$

Data augmentation

Data $y=\{y_i\}$ (resp. $z=\{z_i\}$) can be fitted by a streched-exponential law

$$f_x(t)=C_x\left(1-e^{-\left(\frac{t}{T_x}\right)^{\beta_x}}\right)\quad\text{for } x=y \text{ (resp. } x=z).$$

- fit parameters C_y,β_y,C_z,β_z from $\{y_i\}$ and $\{z_i\}$
- generate data $f_x(t)$ for any time t



Physics-informed neural network

Based on [2], learn a multilayer perceptron G_θ with parameter θ by minimizing

$$\mathcal{L}\left(\theta,R\right)=\mathcal{L}_{\text{pde}}\left(\theta,R\right)+\mathcal{L}_y\left(\theta,R\right)+\mathcal{L}_z\left(\theta,R\right)+\mathcal{L}_{\text{init}}\left(\theta\right)+\mathcal{L}_{\text{bnd}}\left(\theta\right)$$

where

$$\mathcal{L}_{\text{pde}}\left(\theta,R\right)=\frac{1}{|\mathcal{S}_{\text{pde}}|}\sum_{(t,r)\in\mathcal{S}_{\text{pde}}}\mathcal{F}\left(P_\theta,r,t,R\right)^2$$

with $P_\theta\left(r,t\right)=\frac{r}{3}\partial_r G_\theta\left(r,t\right)+G_\theta\left(r,t\right)$

$$\mathcal{L}_y\left(\theta,R\right)=\frac{1}{|\mathcal{S}_y|}\sum_{t\in\mathcal{S}_y}\left(f_y\left(t\right)-G_\theta\left(R,t\right)\right)^2$$

$$\mathcal{L}_z\left(\theta,R\right)=\frac{1}{|\mathcal{S}_z|}\sum_{t\in\mathcal{S}_z}\left(f_z\left(t\right)-P_\theta\left(R,t\right)\right)^2$$

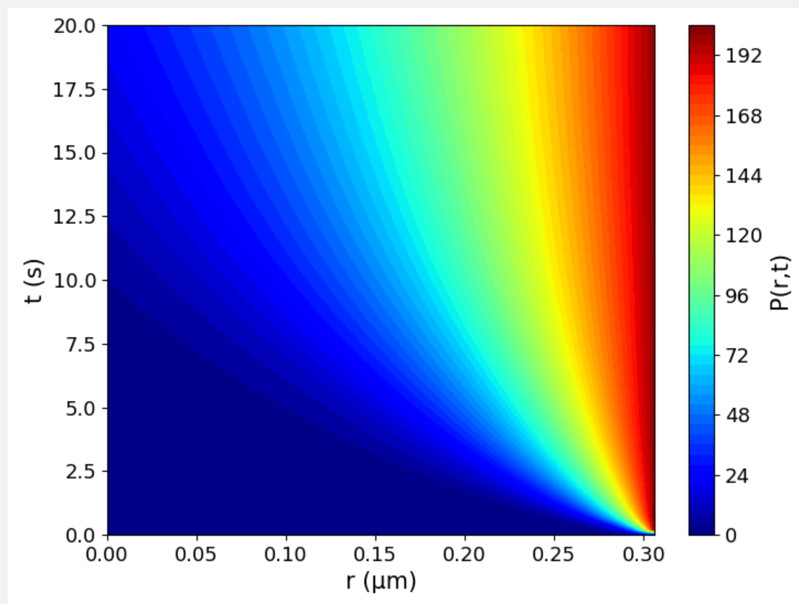
$$\mathcal{L}_{\text{init}}\left(\theta\right)=\frac{1}{|\mathcal{S}_{\text{init}}|}\sum_{r\in\mathcal{S}_{\text{init}}}\left(G_\theta\left(r,0\right)\right)^2$$

for some samplings $\mathcal{S}_{\text{pde}},\mathcal{S}_y,\mathcal{S}_z,\mathcal{S}_{\text{init}}$ of the time and/or space domains.

Experiment

Using an MLP with 2 hidden layers and 32 neurons per layer, and a regular grid for $\mathcal{S}_{\text{pde}},\mathcal{S}_y,\mathcal{S}_z,\mathcal{S}_{\text{init}}$ with 30 spatial samples and 600 time samples,

Synthetic data		
	$R_{\text{true}}=25nm$	$R_{\text{true}}=2.5\mu m$
Homogeneous concentration	$\mathcal{L}=2\cdot10^{-6}$ $R=36.6nm$ (+46%)	$\mathcal{L}=9\cdot10^{-5}$ $R=2.5\mu m$ (0%)
Heterogeneous concentration	$\mathcal{L}=4\cdot10^{-5}$ $R=64.1nm$ (+156%)	$\mathcal{L}=1\cdot10^{-4}$ $R=2.56\mu m$ (+2.4%)



→ Radius is overestimated to compensate for the simplistic assumption that the polarization is constant over the liquid.
→ Need to model the spin diffusion in frozen liquid.
→ Hard to optimize over θ and R.

Perspectives

- Conduct experiments on true polystyrene balls
- Need to model diffusion in both solid & liquid, by considering the space for $r<R'$ where R' is the known radius of the liquid,

$$G\left(R',t_i\right)=\frac{R'^3-R^3}{R'^3}z_i+\frac{R^3}{R'^3}y_i,\forall i\in\{1,\dots,n\}$$

and by replacing loss component $\mathcal{L}_z$ by

$$\mathcal{L}_{yz}\left(\theta,R\right)=\frac{1}{|\mathcal{S}_{yz}|}\sum_{t\in\mathcal{S}_{yz}}\left(\frac{R'^3-R^3}{R'^3}f_z\left(t\right)+\frac{R^3}{R'^3}f_y\left(t\right)-G_\theta\left(R',t\right)\right)^2$$

- Improve joint optimization over neural network and shape parameters.
- Address various parametric shapes.

References

[1] S. F. Cousin, C. E. Hughes, F. Ziarelli, S. Viel, G. Mollica, K. D. M. Harris, A. C. Pinon, and P. Thureau. Exploiting solid-state dynamic nuclear polarization nmr spectroscopy to establish the spatial distribution of polymorphic phases in a solid material. *Chem. Sci.*, 14:10121–10128, 2023.

[2] M. Raissi, P. Perdikaris, and G. Karniadakis. Physics-informed neural networks: A deep learning framework for solving forward and inverse problems involving nonlinear partial differential equations. *Journal of Computational Physics*, 378:686–707, 2019.