Supplementary Material

CONTENTS

S1. CALIBRATION OF THE EPR REOSNANCE

In this section, we detail the way in which we track and account for time-variation in the response function of the sensor, primarily due to decay of the helium polarization during a particular run. For each run at a given constant axial magnetic field, we characterize the response function to oscillatory magnetic field measured before and after the recording, denoted with subscript "pre" and "post" respectively. These two measured response functions, are denoted by $Y_{\rm pre}(f)$ ($Y_{\rm post}(f)$) and are characterized in the frequency domain in units of V/G. For clarity, the black curves exemplified in Fig. 3 are the $Y_{\rm pre}(f)$ functions at two different magnetic fields. We construct them by measuring the response to oscillatory transverse magnetic fields at several $(10-30)$ sampled points and interpolate between the sampled points. We characterize their resonance frequency by the location of their peak response $f_{\rm pre}$ and $f_{\rm post}$ as well as the linewidth $\Gamma_{\rm pre}$ and $\Gamma_{\rm post}$. Using these parameters, we can describe the corresponding normalized response functions $y_{\rm pre}$ and $y_{\rm post}$ using a single unitless parameter η

$$y_{\rm pre}(\eta) = Y_{\rm pre}(\eta\cdot\Gamma_{\rm pre}+f_{\rm pre})/\max(Y_{\rm pre}), \tag{S1}$$
$$y_{\rm post}(\eta) = Y_{\rm post}(\eta\cdot\Gamma_{\rm post}+f_{\rm post})/\max(Y_{\rm post}). \tag{S2}$$

The normalized functions are a shifted and rescaled version of their parent response functions. They are defined such that each normalized response has a maximal unity response at $\eta=0$ and a width of 1.

We construct the estimated amalgam during the experiment $Y_x(f)$ as a linear interpolation between the initial and final measured responses. We use a single parameter $x(t)\in[0,1]$ and take the following interpolation

$$Y_x(f) = Y_{\max}(x)\left(x y_{\rm pre}\left(\frac{f-f_{\rm res}(x)}{\Gamma(x)}\right)+(1-x)y_{\rm post}\left(\frac{f-f_{\rm res}(x)}{\Gamma(x)}\right)\right), \tag{S3}$$

using the definitions

$$Y_{\max}(x) = x\max(Y_{\rm pre})+(1-x)\max(Y_{\rm post}) \tag{S4}$$
$$\Gamma(x) = x\Gamma_{\rm pre}+(1-x)\Gamma_{\rm post} \tag{S5}$$
$$f_{\rm res}(x) = x f_{\rm pre}+(1-x)f_{\rm post}. \tag{S6}$$

It is constructed to be a weighted average between two functions of the same central value, width and height. The weight, height of the central value of the function is the weighted sum of the weights, heights and central values of the two measurements respectively, where the only free parameter is the how to weight the two measurements. To observe the slow change of x in a continuous manner, we estimate the value of x every $1-5$ minutes by taking the absolute value of the Fourier transform of the measured data in a window of about 1 second and fitting to the amalgam Y_x. Importantly, this low-resolution procedure does not resolve the ALP spectrum and does not introduce bias to the measurement. We also note that $Y=0$ was taken outside the measured frequency window, thus only underestimating the sensitivity of the detector in this exclusion search.

S2. LIKELIHOOD FUNCTION

Our analysis constructs and uses the same likelihood functions presented in detail in the Supplementary Material in Ref. [14], except for a different construction of the sensitivity matrices α, which we detail in this section.

The matrices α link the ALP vector $\mathcal{A}/\sqrt{2\rho_a}$ at a given frequency with the data vector $\mathbf{d}$ in the frequency domain where $\rho_a = 0.4\ {\rm GeV/cm^3}$. In this work, the rotating-wave approximation is not assumed, and the contribution of negative frequencies is explicitly considered. Denoting the full sensitivity matrices by α^Σ, their matrix element is given by

$$\alpha^\Sigma_{mn,i} = (\alpha_i(f_m,f_n)+\alpha_i^*(-f_m,-f_n)), \tag{S7}$$

where each $\alpha_i(f_m,f_n)$ is computed by integrating over the response

$$\alpha_i(f_m,f_n) = \frac{\sqrt{2\rho_{\rm a}}}{T}\int_0^T dt\ g_{\rm eff}(f_n,t)e^{2i\pi(f_n-f_m)t}\Xi(t)(\hat{x}(t)+i\hat{y}(t))_i\mathcal{Y}_{x(t)}(f_n). \tag{S8}$$

Here t is the time from the beginning of the measurement, and $\hat{x}(\hat{y})$ is the direction of the probe beam (the direction perpendicular to the probe and pump beams), projected to the $i^{\rm th}$ direction in galactic coordinates (relying on Ref. [45]). T is the total measurement time, and f_m, f_n are two frequencies in the fourier series of the data in the $[0,T]$ interval. $g_{\rm eff}(f_n,t)$ is defined in Eq. 8 of the main text, and $\Xi(t)$ is the binary quality cut function ($\zeta(t)$ in Ref. [14]). We define $\mathcal{Y}_{x(t)}(f)$ below in Eq. S9, and it represents the response to magnetic fields to a single (positive or negative) frequency f.

The measured response function $Y_x(|f|)$ originates from an input sinusoidal function, which includes both positive and negative frequencies. To construct the response function that explicitly differentiates between positive and negative frequencies, we assume that the response is a sum of two Lorentzian functions centered around $\pm f_{\rm res}(x)$ with width $\Gamma(x)$. We further allow the two Lorentizan functions to have any positive common scaling factor that depends on $|f|$, to allow for any frequency dependent response of the electronics of the measurement circuits. We then find that the modified response is

$$\mathcal{Y}_x(f) = Y_x(|f|) \left| \frac{\Gamma + if + if_{\rm res}}{\Gamma + if_{\rm res}} \right| \frac{\Gamma + i(f_{\rm res} - f)}{|\Gamma + i(f_{\rm res} - f)|}. \tag{S9}$$

The second term produces the correct phase for the response, and is only important when $f_{\rm res}$ crosses f. We use a slightly simpler function for the phase in our numerical calculations that reproduces its time dependence.