

# Measuring the angular dipole spectrum with arbitrary polarized illumination microscopes

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## 1 Forward model in a delta function basis

We develop and analyze a simplified model of polarized excitation fluorescence microscopes with arbitrary angular designs. We simplify the complete problem by considering large objects and ignoring the irradiance pattern on the detector (we effectively integrate over a few pixels). These simplifications are equivalent to the implicit assumptions made by the majority of polarized fluorescence microscopists. Our main goal is to improve our intuition for the angular band limit of arbitrary microscopes and to incorporate light-sheet tilting into our model.

With these simplifications we can model a continuous-to-continuous polarized excitation fluorescence microscope system as

$$g(\hat{\mathbf{s}}_d, \hat{\mathbf{p}}) = \int_{\mathbb{S}^2} d\hat{\mathbf{s}}_o h(\hat{\mathbf{s}}_d, \hat{\mathbf{p}}; \hat{\mathbf{s}}_o) f(\hat{\mathbf{s}}_o), \quad (1)$$

where  $\hat{\mathbf{s}}_o$  is an orientation in the object,  $f(\hat{\mathbf{s}}_o)$  is the angular dipole density (with units of  $\text{sr}^{-1}$ ),  $\hat{\mathbf{p}}$  is the orientation of an excitation polarizer,  $\hat{\mathbf{s}}_d$  is the orientation of the detection optical axis,  $g(\hat{\mathbf{s}}_d, \hat{\mathbf{p}})$  is the measured irradiance with detection optical axis  $\hat{\mathbf{s}}_d$  and excitation polarization  $\hat{\mathbf{p}}$ . Equation (1) assumes that object space is  $\mathbb{U} = \mathbb{L}_2(\mathbb{S}^2)$ —functions on the sphere—and data space is  $\mathbb{V} = \mathbb{L}_2(\mathbb{S}^2 \times \mathbb{S}^1)$ —functions on the product space of a sphere and a circle (a doughnut made of Timbits!).

Our next task is to find the form of the kernel  $h(\hat{\mathbf{s}}_d, \hat{\mathbf{p}}; \hat{\mathbf{s}}_o)$ . We know that the kernel is separable into excitation and detection parts so

$$h(\hat{\mathbf{s}}_d, \hat{\mathbf{p}}; \hat{\mathbf{s}}_o) = h^{\text{exc}}(\hat{\mathbf{p}}; \hat{\mathbf{s}}_o) h^{\text{det}}(\hat{\mathbf{s}}_d; \hat{\mathbf{s}}_o). \quad (2)$$

We restrict ourselves to low-NA excitation (like light-sheet excitation), which gives the following excitation kernel

$$h^{\text{exc}}(\hat{\mathbf{p}} \cdot \hat{\mathbf{s}}_o) \propto (\hat{\mathbf{p}} \cdot \hat{\mathbf{s}}_o)^2. \quad (3)$$

In other words, the probability of exciting a fluorophore oriented along  $\hat{\mathbf{s}}_o$  with light polarized along  $\hat{\mathbf{p}}$  is proportional to  $(\hat{\mathbf{p}} \cdot \hat{\mathbf{s}}_o)^2$ —a familiar  $\cos^2 \theta$  dependence where  $\theta$  is the angle between  $\hat{\mathbf{p}}$  and  $\hat{\mathbf{s}}_o$ . Notice that the excitation kernel is rotationally symmetric, so we can write it as a function of  $\hat{\mathbf{p}} \cdot \hat{\mathbf{s}}_o$ .

The detection kernel is given by

$$h^{\text{det}}(\hat{\mathbf{s}}_d, \hat{\mathbf{s}}_o) = \int_{\mathbb{S}^2} d\hat{\mathbf{s}} \Pi\left(\frac{\hat{\mathbf{s}} \cdot \hat{\mathbf{s}}_d}{\cos \alpha}\right) (\hat{\mathbf{s}} \cdot \hat{\mathbf{s}}_d)^{-1/2} [1 - (\hat{\mathbf{s}} \cdot \hat{\mathbf{s}}_o)^2], \quad (4)$$

where the  $[1 - (\hat{\mathbf{s}} \cdot \hat{\mathbf{s}}_o)^2]$  term models the familiar  $\sin^2 \vartheta$  dependence of dipole radiation where  $\vartheta$  is the angle between  $\hat{\mathbf{s}}_d$  and  $\hat{\mathbf{s}}_o$ , the  $(\hat{\mathbf{s}} \cdot \hat{\mathbf{s}}_d)^{-1/2}$  term models the apodization of an aplanatic objective, and the integral over  $\Pi\left(\frac{\hat{\mathbf{s}} \cdot \hat{\mathbf{s}}_d}{\cos \alpha}\right)$  models the finite aperture of the objective with  $\text{NA} = n_o \sin \alpha$ . In Appendix A we evaluate this integral in closed form to find that

$$h^{\text{det}}(\hat{\mathbf{s}}_d \cdot \hat{\mathbf{s}}_o) \propto (1 - \sqrt{\cos \alpha}) P_0(\hat{\mathbf{s}}_d \cdot \hat{\mathbf{s}}_o) + \frac{1}{5} \left[ \frac{1}{4} \sqrt{\cos \alpha} (7 - 3 \cos(2\alpha)) - 1 \right] P_2(\hat{\mathbf{s}}_d \cdot \hat{\mathbf{s}}_o). \quad (5)$$

The detection kernel is rotationally symmetric, so it can be written as a function of  $\hat{\mathbf{s}}_d \cdot \hat{\mathbf{s}}_o$ . For very low NA the detection kernel reduces to  $[1 - (\hat{\mathbf{s}} \cdot \hat{\mathbf{s}}_o)^2]$  as we would expect.

## 2 Forward model in a spherical harmonic basis

We can rewrite the right-hand side of Eq. (1) in a spherical harmonic basis by applying the generalized Plancherel theorem for spherical functions

$$g(\hat{\mathbf{s}}_d, \hat{\mathbf{p}}) = \sum_{\ell m} H_\ell^m(\hat{\mathbf{s}}_d, \hat{\mathbf{p}}) F_\ell^m, \quad (6)$$

where we have defined the *dipole angular transfer function* as

$$H_\ell^m(\hat{\mathbf{s}}_d, \hat{\mathbf{p}}) = \int_{\mathbb{S}^2} d\hat{\mathbf{s}}_o h(\hat{\mathbf{s}}_d, \hat{\mathbf{p}}; \hat{\mathbf{s}}_o) Y_\ell^{m*}(\hat{\mathbf{s}}_o), \quad (7)$$

and the *dipole angular spectrum* as

$$F_\ell^m = \int_{\mathbb{S}^2} d\hat{\mathbf{s}}_o f(\hat{\mathbf{s}}_o) Y_\ell^{m*}(\hat{\mathbf{s}}_o). \quad (8)$$

Our remaining task is to calculate the dipole angular transfer function. We can start by expanding the kernel into excitation and detection parts

$$H_\ell^m(\hat{\mathbf{s}}_d, \hat{\mathbf{p}}) = \int_{\mathbb{S}^2} d\hat{\mathbf{s}}_o h^{\text{exc}}(\hat{\mathbf{p}} \cdot \hat{\mathbf{s}}_o) h^{\text{det}}(\hat{\mathbf{s}}_d \cdot \hat{\mathbf{s}}_o) Y_\ell^{m*}(\hat{\mathbf{s}}_o). \quad (9)$$

We can expand both the excitation and detection kernels into a sum of spherical harmonics (a Laplace series)

$$H_\ell^m(\hat{\mathbf{s}}_d, \hat{\mathbf{p}}) = \int_{\mathbb{S}^2} d\hat{\mathbf{s}}_o \left[ \sum_{\ell' m'} H_{\ell'}^{m', \text{exc}}(\hat{\mathbf{p}} \cdot \hat{\mathbf{s}}_o) Y_{\ell'}^{m'}(\hat{\mathbf{s}}_o) \right] \left[ \sum_{\ell'' m''} H_{\ell''}^{m'', \text{det}}(\hat{\mathbf{s}}_d \cdot \hat{\mathbf{s}}_o) Y_{\ell''}^{m''}(\hat{\mathbf{s}}_o) \right] Y_\ell^{m*}(\hat{\mathbf{s}}_o), \quad (10)$$

where  $H_{\ell'}^{m', \text{exc}}(\hat{\mathbf{p}} \cdot \hat{\mathbf{s}}_o)$  and  $H_{\ell''}^{m'', \text{det}}(\hat{\mathbf{s}}_d \cdot \hat{\mathbf{s}}_o)$  are the spherical harmonic transforms of the excitation and detection kernels

$$H_{\ell'}^{m', \text{exc}}(\hat{\mathbf{p}}) = \int_{\mathbb{S}^2} d\hat{\mathbf{s}}_o h^{\text{exc}}(\hat{\mathbf{p}} \cdot \hat{\mathbf{s}}_o) Y_{\ell'}^{m'*}(\hat{\mathbf{s}}_o), \quad (11)$$

$$H_{\ell''}^{m'', \text{det}}(\hat{\mathbf{s}}_d) = \int_{\mathbb{S}^2} d\hat{\mathbf{s}}_o h^{\text{det}}(\hat{\mathbf{s}}_d \cdot \hat{\mathbf{s}}_o) Y_{\ell''}^{m''*}(\hat{\mathbf{s}}_o). \quad (12)$$

In Appendix B we evaluate these spherical Fourier transforms as

$$H_{\ell'}^{m', \text{exc}}(\hat{\mathbf{p}}) \propto Y_{\ell'}^{m'*}(\hat{\mathbf{p}}) \left[ \delta_{0\ell'} + \frac{2}{5} \delta_{2,\ell'} \right], \quad (13)$$

$$H_{\ell''}^{m'', \text{det}}(\hat{\mathbf{s}}_d) \propto Y_{\ell''}^{m''*}(\hat{\mathbf{s}}_d) \left[ (1 - \sqrt{\cos \alpha}) \delta_{0,\ell''} + \left( \frac{1}{4} \sqrt{\cos \alpha} (7 - 3 \cos(2\alpha)) - 1 \right) \delta_{2,\ell''} \right]. \quad (14)$$

We continue simplifying Eq. (10) by rewriting it as

$$H_\ell^m(\hat{\mathbf{s}}_d, \hat{\mathbf{p}}) = \sum_{\ell' m'} \sum_{\ell'' m''} H_{\ell'}^{m', \text{exc}}(\hat{\mathbf{p}}) H_{\ell''}^{m'', \text{det}}(\hat{\mathbf{s}}_d) \int_{\mathbb{S}^2} d\hat{\mathbf{s}}_o Y_\ell^{m*}(\hat{\mathbf{s}}_o) Y_{\ell'}^{m'}(\hat{\mathbf{s}}_o) Y_{\ell''}^{m''}(\hat{\mathbf{s}}_o). \quad (15)$$

We can simplify this equation further as

$$H_\ell^m(\hat{\mathbf{s}}_d, \hat{\mathbf{p}}) = \sum_{\ell' m'} \sum_{\ell'' m''} (-1)^m G_{\ell, \ell', \ell''}^{-m, m', m''} H_{\ell'}^{m', \text{exc}}(\hat{\mathbf{p}}) H_{\ell''}^{m'', \text{det}}(\hat{\mathbf{s}}_d), \quad (16)$$

where  $G_{\ell, \ell', \ell''}^{m, m', m''}$  are the Gaunt coefficients (sometimes called the tripling coefficients or triple integrals of the spherical harmonics) defined by

$$G_{\ell, \ell', \ell''}^{m, m', m''} = \int_{\mathbb{S}^2} d\hat{\mathbf{s}}_o Y_\ell^m(\hat{\mathbf{s}}_o) Y_{\ell'}^{m'}(\hat{\mathbf{s}}_o) Y_{\ell''}^{m''}(\hat{\mathbf{s}}_o). \quad (17)$$

Equation (16) is a key result because it allows us to calculate the dipole angular transfer function from the excitation and detection angular transfer functions.

We can draw some general conclusions about the band limits of this type of imaging system by examining the properties of the Gaunt coefficients. First, the Gaunt coefficient can only be non-zero when the coefficients  $\ell$ ,  $\ell'$ , and  $\ell''$  satisfy the *triangle condition*—each of the three coefficients is between the sum and difference of the other two. The most relevant condition for us is

$$|\ell' - \ell''| \leq \ell \leq \ell' + \ell'', \quad (18)$$

which means that we can only have non-zero coefficients for  $0 \leq \ell \leq 4$  since  $0 \leq \ell' \leq 2$  and  $0 \leq \ell'' \leq 2$ . Also, the Gaunt coefficients can only be non-zero when the  $\ell$ ,  $\ell'$ , and  $\ell''$  form an even sum

$$\ell + \ell' + \ell'' = 2n, \quad \text{for } n \in \mathbb{N}, \quad (19)$$

which ensures inversion symmetry. Finally, the Gaunt coefficients can only be non-zero when each  $\ell$  is less than the absolute value of the corresponding  $m$

$$\ell \leq |m|, \quad \ell' \leq |m'|, \quad \ell'' \leq |m''|. \quad (20)$$

These conditions are obvious if you consider that the spherical harmonics are zero (or undefined) when these conditions are not satisfied.

Given that  $0 \leq \ell' \leq 2$  and  $0 \leq \ell'' \leq 2$  for our measurements, the conditions in Eqs. (18)–(20) imply that the dipole angular transfer function has at most 15 terms—1 for the  $\ell = 0$  band, 5 for the  $\ell = 2$  band, and 9 for the  $\ell = 4$  band.

Note that a single choice for a measurement  $\hat{\mathbf{s}}_d$  and  $\hat{\mathbf{p}}$  can have 15 non-zero terms in the dipole angular transfer function if both  $\hat{\mathbf{s}}_d$  and  $\hat{\mathbf{p}}$  do not lie on a node (zero) of any of the  $\ell = 2$  spherical harmonics. I mention this to emphasize that the number of terms in the dipole angular transfer function has little to no bearing on how much we can measure about the object—the choice of coordinate system changes the nodes of the spherical harmonics and our results should never depend on a choice of coordinates. The object space singular vectors are the functions that we care about when we ask: what can this instrument measure about the object?

Notice that all of our results so far are completely coordinate-free—we have used unit vectors everywhere instead of Cartesian or spherical coordinates.

### 3 Continuous-to-discrete model

So far we have modeled the continuous data space  $\mathbb{V} = \mathbb{L}_2(\mathbb{S}^2 \times \mathbb{S}^1)$ . Now we consider discrete samples of this data space with the following forward model

$$g_i = \int_{\mathbb{S}^2} d\hat{\mathbf{s}}_d \int_{\mathbb{S}^1} d\hat{\mathbf{p}} w_i(\hat{\mathbf{s}}_d, \hat{\mathbf{p}}) \int_{\mathbb{S}^2} d\hat{\mathbf{s}}_o h(\hat{\mathbf{s}}_d, \hat{\mathbf{p}}; \hat{\mathbf{s}}_o) f(\hat{\mathbf{s}}_o), \quad (21)$$

where  $g_i$  is a discrete set of  $M$  measurements and  $w_i(\hat{\mathbf{s}}_d, \hat{\mathbf{p}})$  is the “sampling aperture” that indicates which part of data space  $(\hat{\mathbf{s}}_d, \hat{\mathbf{p}})$  we sample during the  $i$ th measurement. For example, if we have a detector along the  $\hat{\mathbf{z}}$  axis and we illuminate with  $\hat{\mathbf{x}}$  polarized light for the first measurement and  $\hat{\mathbf{y}}$  polarized light for the second measurement, then the sampling aperture is

$$w_i^{(0)}(\hat{\mathbf{s}}_d, \hat{\mathbf{p}}) = \delta(\hat{\mathbf{s}}_d - \hat{\mathbf{z}})[\delta(\hat{\mathbf{p}} - \hat{\mathbf{x}})\delta_{i,0} + \delta(\hat{\mathbf{p}} - \hat{\mathbf{y}})\delta_{i,1}]. \quad (22)$$

It is more convenient to rewrite Eq. (21) as

$$g_i = \int_{\mathbb{S}^2} d\hat{\mathbf{s}}_o h_i(\hat{\mathbf{s}}_o) f(\hat{\mathbf{s}}_o), \quad (23)$$

where

$$h_i(\hat{\mathbf{s}}_o) = \int_{\mathbb{S}^2} d\hat{\mathbf{s}}_d \int_{\mathbb{S}^1} d\hat{\mathbf{p}} w_i(\hat{\mathbf{s}}_d, \hat{\mathbf{p}}) h(\hat{\mathbf{s}}_d, \hat{\mathbf{p}}; \hat{\mathbf{s}}_o). \quad (24)$$

In our example, the continuous-to-discrete kernel is given by

$$h_i^{(0)}(\hat{\mathbf{s}}_o) = h(\hat{\mathbf{z}}, \hat{\mathbf{x}}; \hat{\mathbf{s}}_o) \delta_{i,0} + h(\hat{\mathbf{z}}, \hat{\mathbf{y}}; \hat{\mathbf{s}}_o) \delta_{i,1}. \quad (25)$$

## 4 Singular value decomposition

Our continuous-to-discrete forward model can be written in any of the following equivalent forms

$$\mathbf{g} = \mathcal{H}\mathbf{f}, \quad (26)$$

$$g_i = \int_{\mathbb{S}^2} d\hat{\mathbf{s}}_o h_i(\hat{\mathbf{s}}_o) f(\hat{\mathbf{s}}_o), \quad (27)$$

$$g_i = \sum_{\ell, m} H_{i, \ell}^m F_{\ell}^m, \quad (28)$$

$$g_i = \sum_j H_{i, j} F_j. \quad (29)$$

where Eq. (26) is the Hilbert-space form, Eq. (27) is the delta-function basis form, Eq. (28) is the spherical harmonic basis form, and Eq. (29) is the spherical harmonic basis form with a collapsed index  $j(\ell, m) = [\ell(\ell+1)]/2 + m$  (don't confuse the spherical Fourier transform with collapsed coefficients,  $F_j$ , with the Fourier-Legendre transform,  $F_{\ell}$ —I'll consider alternative notations but it should be clear from the context). Notice that the last two forms blur the line between a CD operator and a DD operator—we are still considering a continuous object space and discrete data space, but we have found a basis that allows us to express this mapping in a completely discrete way.

Similarly, we can write the discrete-to-continuous adjoint operator as

$$\mathbf{f} = \mathcal{H}^{\dagger} \mathbf{g}, \quad (30)$$

$$f(\hat{\mathbf{s}}_o) = \sum_i h_i(\hat{\mathbf{s}}_o) g_i, \quad (31)$$

$$F_{\ell}^m = \sum_i H_{i, \ell}^m g_i, \quad (32)$$

$$F_j = \sum_i H_{i, j} g_i, \quad (33)$$

where the forms of Eqs. (30)–(33) match with the forms of Eqs. (26)–(29).

To find the object space singular functions we need to solve the eigenvalue problem in one of its forms

$$\mathcal{H}^{\dagger} \mathcal{H} \mathbf{u}_n = \mu_n \mathbf{u}_n, \quad (34)$$

$$\sum_i h_i(\hat{\mathbf{s}}_o) \int_{\mathbb{S}^2} d\hat{\mathbf{s}}'_o h_i(\hat{\mathbf{s}}'_o) u_n(\hat{\mathbf{s}}'_o) = \mu_n u_n(\hat{\mathbf{s}}_o), \quad (35)$$

$$\sum_i H_{i, \ell}^m \sum_{\ell' m'} H_{i, \ell'}^{m'} U_{n, \ell'}^{m'} = \mu_n U_{n, \ell}^m, \quad (36)$$

$$\sum_i H_{i, j} \sum_{j'} H_{i, j'} U_{n, j'} = \mu_n U_{n, j}, \quad (37)$$

where  $n$  indexes the singular values and singular functions, and the letter “u” is used with varying type and arguments to denote the object space singular functions. We can tighten up the notation by introducing kernels of the operators and denoting them with the letter “k” with varying type and arguments:

$$\mathcal{K}\mathbf{u}_n = \mu_n \mathbf{u}_n, \quad (38)$$

$$\int_{\mathbb{S}^2} d\hat{\mathbf{s}}'_o k(\hat{\mathbf{s}}_o, \hat{\mathbf{s}}'_o) u_n(\hat{\mathbf{s}}'_o) = \mu_n u_n(\hat{\mathbf{s}}_o), \quad (39)$$

$$\sum_{\ell' m'} K_{\ell, \ell'}^{m, m'} U_{n, \ell'}^{m'} = \mu_n U_{n, \ell}^m, \quad (40)$$

$$\sum_{j'} K_{j, j'} U_{n, j'} = \mu_n U_{n, j}, \quad (41)$$

where

$$\mathcal{K} = \mathcal{H}\mathcal{H}^\dagger, \quad (42)$$

$$k(\hat{\mathbf{s}}_o, \hat{\mathbf{s}}'_o) = \sum_i h_i(\hat{\mathbf{s}}_o) h_i(\hat{\mathbf{s}}'_o), \quad (43)$$

$$K_{\ell, \ell'}^{m, m'} = \sum_i H_{i, \ell}^m H_{i, \ell'}^{m'}, \quad (44)$$

$$K_{j, j'} = \sum_i H_{i, j} H_{i, j'}. \quad (45)$$

Now we are in position to solve for the object space singular functions and the singular values. The first step is to calculate the matrix  $K_{j, j'}$  using Eq. (45) and the closed form expressions for the dipole angular transfer function,  $H_{i, j}$ , that we calculated in Sec. 2. As we discussed previously,  $H_{i, j}$  is a  $M \times 15$  matrix so  $K_{j, j'}$  is  $15 \times 15$  and it is computationally trivial (though impossible analytically) to calculate its eigenvalues  $\mu_n$  and eigenvectors  $U_{n, j}$ . With  $U_{n, j}$  in hand we can calculate the object-space singular vectors in a delta function basis using an inverse spherical harmonics transform

$$u_n(\hat{\mathbf{s}}_o) = \sum_j U_{n, j} Y_j(\hat{\mathbf{s}}_o). \quad (46)$$

Before examining specific designs, we briefly examine and interpret the object space singular vectors and singular values. The object space singular vectors span the space of measurable objects—they are orthogonal (and can be chosen to be orthonormal) because the operator  $\mathcal{H}^\dagger \mathcal{H}$  is Hermitian. The singular values tell us “how efficiently” each object space singular vector is transferred through the system—the singular values (the eigenvalues of  $\mathcal{H}^\dagger \mathcal{H}$ ) are real (because  $\mathcal{H}^\dagger \mathcal{H}$  is Hermitian) and positive (because of the form of  $\mathcal{H}^\dagger \mathcal{H}$  see Barrett 1.4.4). In other words,  $\mathcal{H}^\dagger \mathcal{H}$  is positive-semidefinite because the eigenvalues are greater than or equal to zero.

One desirable condition for the operator  $\mathcal{H}^\dagger \mathcal{H}$  is that it has the maximum possible rank— $R = 15$  in our case which is limited by the excitation and detection physics. Achieving  $R = 15$  will always require at least 15 distinct measurements which is far more than we would like, so we search for designs with fewer measurements.

Another desirable condition is that the object space singular vectors span subspaces of  $\mathbb{L}_2(\mathbb{S}^2)$  that are invariant under rotation. The spherical harmonic bands are exactly the subspaces we’re looking for: the  $\ell$ th band spans a  $(2\ell + 1)$ -dimension space that is invariant under rotation—equivalently, the  $\ell$ th band forms a  $(2\ell + 1)$ -dimensional representation for the rotation group. Furthermore, we know that the  $\ell$ th band is an *irreducible* representation of the rotation group, which means that we cannot decompose the  $\ell$ th band of spherical harmonics to find smaller representations of the rotation group. Therefore, if we would like our object space singular vectors to span a rotationally-invariant subspace then we need to take at least 1, 6, or 15 measurements.

## 5 Specific designs (in progress)

one-view epi, one-view lightsheet, two-view lightsheet, two-view lightsheet two-tilt one-side, two-view lightsheet two-tilt two-side, two-view lightsheet three-tilt one-side two-tilt one-side.

## A Spherical integral of the product of two rotationally symmetric functions

Consider two rotationally symmetric functions on the sphere  $f(\hat{\mathbf{s}} \cdot \hat{\mathbf{s}}')$  and  $g(\hat{\mathbf{s}} \cdot \hat{\mathbf{s}}'')$ . In this appendix we will reduce integrals of the form

$$g(\hat{\mathbf{s}}', \hat{\mathbf{s}}'') = \int_{\mathbb{S}^2} d\hat{\mathbf{s}} h(\hat{\mathbf{s}} \cdot \hat{\mathbf{s}}') f(\hat{\mathbf{s}} \cdot \hat{\mathbf{s}}'') \quad (47)$$

to two one-dimensional integrals. The key step is to expand both  $f$  and  $g$  into a Fourier-Legendre series

$$f(x) = \sum_{\ell=0}^{\infty} F_{\ell} P_{\ell}(x), \quad (48)$$

where  $P_{\ell}(x)$  are the Legendre polynomials. We can find the coefficients  $F_{\ell}$  using Fourier's trick—multiply both sides of Eq. (48) by  $P_{\ell'}(x)$  (the Legendre polynomials are real so we can ignore complex conjugates) then integrate over the interval  $[-1, 1]$  and apply the orthogonality relation

$$\int_{-1}^1 dx P_{\ell}(x) P_{\ell'}(x) = \frac{2}{2\ell + 1} \delta_{\ell\ell'}, \quad (49)$$

which implies that the Legendre polynomials are orthogonal but not orthonormal. After applying these steps to Eq. (48) we find that

$$F_{\ell} = \frac{2\ell + 1}{2} \int_{-1}^1 dx f(x) P_{\ell}(x). \quad (50)$$

The coefficients  $F_{\ell}$  are often referred to as the *Fourier-Legendre transform* of  $f(x)$ . Now we can expand  $f$  and  $g$  in Eq. (47) into two Fourier-Legendre series

$$g(\hat{\mathbf{s}}', \hat{\mathbf{s}}'') = \int_{\mathbb{S}^2} d\hat{\mathbf{s}} \left[ \sum_{\ell} H_{\ell} P_{\ell}(\hat{\mathbf{s}} \cdot \hat{\mathbf{s}}') \right] \left[ \sum_{\ell'} F_{\ell'} P_{\ell'}(\hat{\mathbf{s}} \cdot \hat{\mathbf{s}}'') \right], \quad (51)$$

$$= \sum_{\ell} \sum_{\ell'} \int_{\mathbb{S}^2} d\hat{\mathbf{s}} H_{\ell} F_{\ell'} P_{\ell}(\hat{\mathbf{s}} \cdot \hat{\mathbf{s}}') P_{\ell'}(\hat{\mathbf{s}} \cdot \hat{\mathbf{s}}''). \quad (52)$$

The next step is to expand the Legendre polynomials in terms of spherical harmonics using the *spherical harmonic addition theorem*

$$P_{\ell}(\hat{\mathbf{s}} \cdot \hat{\mathbf{s}}') = \frac{4\pi}{2\ell + 1} \sum_{m=-\ell}^{\ell} Y_{\ell}^m(\hat{\mathbf{s}}) Y_{\ell}^{m*}(\hat{\mathbf{s}}'). \quad (53)$$

Applying Eq. (53) to Eq. (52) yields

$$g(\hat{\mathbf{s}}', \hat{\mathbf{s}}'') = \sum_{\ell} \sum_{\ell'} \int_{\mathbb{S}^2} d\hat{\mathbf{s}} H_{\ell} F_{\ell'} \left[ \frac{4\pi}{2\ell+1} \sum_{m=-\ell}^{\ell} Y_{\ell}^m(\hat{\mathbf{s}}) Y_{\ell}^{m*}(\hat{\mathbf{s}}') \right] \left[ \frac{4\pi}{2\ell'+1} \sum_{m'=-\ell'}^{\ell'} Y_{\ell'}^{m'}(\hat{\mathbf{s}}'') Y_{\ell'}^{m'*}(\hat{\mathbf{s}}) \right], \quad (54)$$

$$= \sum_{\ell m} \sum_{\ell' m'} H_{\ell} F_{\ell'} \left[ \frac{4\pi}{2\ell+1} Y_{\ell}^{m*}(\hat{\mathbf{s}}') \right] \left[ \frac{4\pi}{2\ell'+1} Y_{\ell'}^{m'}(\hat{\mathbf{s}}'') \right] \int_{\mathbb{S}^2} d\hat{\mathbf{s}} Y_{\ell}^m(\hat{\mathbf{s}}) Y_{\ell'}^{m'*}(\hat{\mathbf{s}}), \quad (55)$$

$$= \sum_{\ell m} \sum_{\ell' m'} H_{\ell} F_{\ell'} \left[ \frac{4\pi}{2\ell+1} Y_{\ell}^{m*}(\hat{\mathbf{s}}') \right] \left[ \frac{4\pi}{2\ell'+1} Y_{\ell'}^m(\hat{\mathbf{s}}'') \right] \delta_{\ell\ell'} \delta_{mm'}, \quad (56)$$

$$= \sum_{\ell m} H_{\ell} F_{\ell} \left( \frac{4\pi}{2\ell+1} \right)^2 Y_{\ell}^{m*}(\hat{\mathbf{s}}') Y_{\ell}^m(\hat{\mathbf{s}}''), \quad (57)$$

$$g(\hat{\mathbf{s}}' \cdot \hat{\mathbf{s}}'') = \sum_{\ell} \frac{4\pi}{2\ell+1} H_{\ell} F_{\ell} P_{\ell}(\hat{\mathbf{s}}' \cdot \hat{\mathbf{s}}''). \quad (58)$$

where we have (54-55) rearranged the equation, (55-56) used the orthogonality of spherical harmonics, (56-57) used the discrete sifting theorem then, (57-58) applied the spherical harmonic addition theorem.

Eq. (58) is a key result. In words, we can compute the spherical integral of the product of two rotationally symmetric functions by taking their Fourier-Legendre transforms then summing over  $\ell$  with appropriate weights. Perhaps surprisingly, the result only depends on  $\hat{\mathbf{s}}' \cdot \hat{\mathbf{s}}''$  so it is a rotationally symmetric function, too.

We can use this result to evaluate the detection kernel

$$h^{\text{det}}(\hat{\mathbf{s}}_d, \hat{\mathbf{s}}_o) = \int_{\mathbb{S}^2} d\hat{\mathbf{s}} \Pi \left( \frac{\hat{\mathbf{s}} \cdot \hat{\mathbf{s}}_d}{\cos \alpha} \right) (\hat{\mathbf{s}} \cdot \hat{\mathbf{s}}_d)^{-1/2} [1 - (\hat{\mathbf{s}} \cdot \hat{\mathbf{s}}_o)^2]. \quad (59)$$

Comparing Eq. (59) to Eq. (47) means that

$$f(\hat{\mathbf{s}} \cdot \hat{\mathbf{s}}_d) = 1 - (\hat{\mathbf{s}} \cdot \hat{\mathbf{s}}_o)^2, \quad (60)$$

$$h(\hat{\mathbf{s}} \cdot \hat{\mathbf{s}}_d) = \Pi \left( \frac{\hat{\mathbf{s}} \cdot \hat{\mathbf{s}}_d}{\cos \alpha} \right) (\hat{\mathbf{s}} \cdot \hat{\mathbf{s}}_d)^{-1/2}. \quad (61)$$

Evaluating the Fourier-Legendre transform of these functions gives

$$F_{\ell} = \frac{2\ell+1}{2} \int_{-1}^1 dx (1-x^2) P_{\ell}(x) = \frac{2}{3} (\delta_{\ell,0} + \delta_{\ell,2}), \quad (62)$$

$$H_{\ell} = \frac{2\ell+1}{2} \int_{\cos \alpha}^1 dx x^{-1/2} P_{\ell}(x) = (1 - \sqrt{\cos \alpha}) \delta_{\ell,0} + \left[ \frac{1}{4} \sqrt{\cos \alpha} (7 - 3 \cos(2\alpha)) - 1 \right] \delta_{\ell,2} + \dots, \quad (63)$$

Notice that we only need to evaluate the first two terms of  $H_{\ell}$  since only two terms of  $F_{\ell}$  are non-zero. As we've noted before, the physical dipole emission process is the band limiting step here—not the detection process.

Plugging Eqs. (62) and (63) into Eq. (58) gives the closed form result for the detection kernel

$$h^{\text{det}}(\hat{\mathbf{s}}_d \cdot \hat{\mathbf{s}}_o) \propto (1 - \sqrt{\cos \alpha}) P_0(\hat{\mathbf{s}}_d \cdot \hat{\mathbf{s}}_o) + \frac{1}{5} \left[ \frac{1}{4} \sqrt{\cos \alpha} (7 - 3 \cos(2\alpha)) - 1 \right] P_2(\hat{\mathbf{s}}_d \cdot \hat{\mathbf{s}}_o). \quad (64)$$

## B Spherical Fourier transform of a rotationally symmetric function

Consider a rotationally symmetric function on the sphere  $f(\hat{\mathbf{s}} \cdot \hat{\mathbf{s}}')$ . In this appendix we will reduce the spherical Fourier transform

$$F_{\ell}^m = \int_{\mathbb{S}^2} d\hat{\mathbf{s}} f(\hat{\mathbf{s}} \cdot \hat{\mathbf{s}}') Y_{\ell}^{m*}(\hat{\mathbf{s}}) \quad (65)$$

to a single one-dimensional integral. Similar to Appendix A, we expand  $f$  in a Fourier-Legendre series and proceed step by step

$$F_\ell^m = \int_{\mathbb{S}^2} d\hat{\mathbf{s}} \left[ \sum_{\ell'} F_{\ell'} P_{\ell'}(\hat{\mathbf{s}} \cdot \hat{\mathbf{s}}') \right] Y_\ell^{m*}(\hat{\mathbf{s}}), \quad (66)$$

$$F_\ell^m = \sum_{\ell'} F_{\ell'} \int_{\mathbb{S}^2} d\hat{\mathbf{s}} P_{\ell'}(\hat{\mathbf{s}} \cdot \hat{\mathbf{s}}') Y_\ell^{m*}(\hat{\mathbf{s}}), \quad (67)$$

$$F_\ell^m = \sum_{\ell' m'} \frac{4\pi}{2\ell' + 1} F_{\ell'} Y_{\ell'}^{m'*}(\hat{\mathbf{s}}') \int_{\mathbb{S}^2} d\hat{\mathbf{s}} Y_{\ell'}^{m'}(\hat{\mathbf{s}}) Y_\ell^{m*}(\hat{\mathbf{s}}), \quad (68)$$

$$F_\ell^m = \sum_{\ell' m'} \frac{4\pi}{2\ell' + 1} F_{\ell'} Y_{\ell'}^{m'*}(\hat{\mathbf{s}}') \delta_{\ell\ell'} \delta_{mm'}, \quad (69)$$

$$F_\ell^m = \frac{4\pi}{2\ell + 1} Y_\ell^{m*}(\hat{\mathbf{s}}') F_\ell, \quad (70)$$

where we have (66-67) rearranged the equation, (67-68) applied the spherical harmonic addition theorem, (68-69) used the orthogonality of spherical harmonics then, (69-70) used the discrete sifting theorem. Eq. (70) is the result we were seeking—we can find the spherical Fourier transform of a rotationally symmetric function by finding the Fourier-Bessel transform  $F_\ell$  then multiplying by  $\frac{4\pi}{2\ell+1} Y_\ell^{m*}(\hat{\mathbf{s}}')$ .

We can use this result to evaluate the excitation and detection angular transfer functions

$$H_{\ell'}^{m', \text{exc}}(\hat{\mathbf{p}} \cdot \hat{\mathbf{s}}_o) = \int_{\mathbb{S}^2} d\hat{\mathbf{s}}_o h^{\text{exc}}(\hat{\mathbf{p}} \cdot \hat{\mathbf{s}}_o) Y_{\ell'}^{m'*}(\hat{\mathbf{s}}_o), \quad (71)$$

$$H_{\ell''}^{m'', \text{det}}(\hat{\mathbf{s}}_d \cdot \hat{\mathbf{s}}_o) = \int_{\mathbb{S}^2} d\hat{\mathbf{s}}_o h^{\text{det}}(\hat{\mathbf{s}}_d \cdot \hat{\mathbf{s}}_o) Y_{\ell''}^{m''*}(\hat{\mathbf{s}}_o). \quad (72)$$

where

$$h^{\text{exc}}(\hat{\mathbf{p}} \cdot \hat{\mathbf{s}}_o) \propto (\hat{\mathbf{p}} \cdot \hat{\mathbf{s}}_o)^2, \quad (73)$$

$$h^{\text{det}}(\hat{\mathbf{s}}_d \cdot \hat{\mathbf{s}}_o) \propto (1 - \sqrt{\cos \alpha}) P_0(\hat{\mathbf{s}}_d \cdot \hat{\mathbf{s}}_o) + \frac{1}{5} \left[ \frac{1}{4} \sqrt{\cos \alpha} (7 - 3 \cos(2\alpha)) - 1 \right] P_2(\hat{\mathbf{s}}_d \cdot \hat{\mathbf{s}}_o). \quad (74)$$

Starting with the excitation kernel

$$H_{\ell'}^{m', \text{exc}}(\hat{\mathbf{p}}) = \frac{4\pi}{2\ell + 1} Y_{\ell'}^{m'*}(\hat{\mathbf{p}}) \left[ \frac{2\ell' + 1}{2} \int_{-1}^1 dx x^2 P_{\ell'}(x) \right], \quad (75)$$

$$H_{\ell'}^{m', \text{exc}}(\hat{\mathbf{p}}) = \frac{4\pi}{2\ell + 1} Y_{\ell'}^{m'*}(\hat{\mathbf{p}}) \left[ \frac{1}{3} \delta_{0\ell'} + \frac{2}{3} \delta_{2,\ell'} \right], \quad (76)$$

$$H_{\ell'}^{m', \text{exc}}(\hat{\mathbf{p}}) \propto Y_{\ell'}^{m'*}(\hat{\mathbf{p}}) \left[ \delta_{0\ell'} + \frac{2}{5} \delta_{2,\ell'} \right]. \quad (77)$$

Notice that the polarizer orientation  $\hat{\mathbf{p}}$  only affects the  $m$  components of the angular transfer function while the relative contribution to the two  $\ell$  bands is fixed.

Finally, the detection kernel is

$$H_{\ell''}^{m'', \text{det}}(\hat{\mathbf{s}}_d) = \frac{4\pi}{2\ell'' + 1} Y_{\ell''}^{m''*}(\hat{\mathbf{s}}_d) \left[ \frac{2\ell'' + 1}{2} \int_{-1}^1 dx h^{\text{det}}(x) P_{\ell''}(x) \right], \quad (78)$$

$$H_{\ell''}^{m'', \text{det}}(\hat{\mathbf{s}}_d) = \frac{4\pi}{2\ell'' + 1} Y_{\ell''}^{m''*}(\hat{\mathbf{s}}_d) \left[ (1 - \sqrt{\cos \alpha}) \delta_{0,\ell''} + \frac{1}{5} \left( \frac{1}{4} \sqrt{\cos \alpha} (7 - 3 \cos(2\alpha)) - 1 \right) \delta_{2,\ell''} \right], \quad (79)$$

$$H_{\ell''}^{m'', \text{det}}(\hat{\mathbf{s}}_d) \propto Y_{\ell''}^{m''*}(\hat{\mathbf{s}}_d) \left[ (1 - \sqrt{\cos \alpha}) \delta_{0,\ell''} + \left( \frac{1}{4} \sqrt{\cos \alpha} (7 - 3 \cos(2\alpha)) - 1 \right) \delta_{2,\ell''} \right]. \quad (80)$$

Notice that the detection axis  $\hat{\mathbf{s}}_d$  only affects the  $m$  components of the detection axis and the detection NA affects the relative contribution to the two  $\ell$  bands.