I. Substrate response

The substrate used in our work for the epitaxial growth of CoFe$_2$O$_4$ is MgAl$_2$O$_4$. Based on the chemical composition of the substrate and prior spectroscopic work at low magnetic fields, it is generally assumed that MgAl$_2$O$_4$ (which is also a spinel) is magnetically and magnetooptically silent.\(^1\) This assumption has, of course, not been tested in high magnetic fields. It is always a good idea to carry out a full set of substrate measurements because substrates can have small impurity concentrations that may introduce spurious features into the spectrum. For instance, even SrTiO$_3$ has magneto-optical signatures at high magnetic fields.\(^2\)

Figure 1(a) shows MCD response of MgAl$_2$O$_4$ substrate varies across the energy spectrum at \(\pm 10\) T. The substrate was a 100 \(\mu\)m thick. These data are obtained by measuring the spectra at 10 T, 0 T going down, -10 T, and finally at 0 T going up. Once each spectrum is measured the average of the 0 T spectra is calculated and subtracted from the \(\pm 10\) T spectra. Importantly, the result is quite small - on the order of \(10^{-4}\) - so the substrate response does not impact results on CoFe$_2$O$_4$. Figure 1(b) displays a similar style of spectrum but with an average of the 0 T spectra subtracted from the individual 0 T spectra, red and blue lines.

II. A closer look at the derivative relationship between magnetic circular dichroism and optical absorption

As described in the main text, \(I_{\text{MCD}}\) is closely connected with the energy derivative of the optical absorption according to the following relationship:

\[
I_{\text{MCD}} \approx \left( \frac{\alpha_+ (E) - \alpha_- (E)}{2} \right) \approx \frac{\Delta E}{2} \frac{1}{\alpha (E)} \frac{d\alpha (E)}{dE}.
\]

In order to illustrate this connection more clearly, we plot \(I_{\text{MCD}}\), \(\alpha (E)\), and the derivative of the absorption spectrum together in Fig. 2. By comparing the information in the panels, one can easily see the indicated correspondence.

III. Lineshape analysis

Figure 3 shows an example of our lineshape analysis. Here, the Lorentzian oscillators (red, blue, magnenta, and green lines) are superimposed to create a total fit (orange lines) which compares well with the measured \(I_{\text{MCD}}\) (black line). The total line shape is thus the sum of Lorentzians.

References


Figure 1. The MCD response of MgAl$_2$O$_4$ is shown to be magneto-optically silent. (a) ±10 T MCD response of the substrate is effectively flat across spectrum. (b) The MCD response of the substrate at 0 T, the distinction between the two is the direction that the field was before reaching 0 T.

Figure 2. This panel illustrates the connection between the optical absorption, the energy derivative of the absorption, and $I_{MCD}$. (a) This data is reproduced from the main text. (b) The relationship between the optical absorption and the derivative of the absorption is shown here.

Figure 3. (a) Shown here is a fit of the 10 T $I_{MCD}$ response of CoFe$_2$O$_4$. Four main features are fit with Lorentzian functions. (b) The same process is used to fit the 2 T $I_{MCD}$ response.