Dynamics of electronic dephasing in the Fenna-Matthews-Olson complex:
Supplementary data
Figure 1. In the left panel, the amplitude of the signal from the frequency-time spectrum taken at $T = 300$ fs at the rephasing frequency corresponding to exciton 3 ($12,344 \text{ cm}^{-1}$) is plotted against positive coherence time (blue line). Coherence time is plotted using the same scale as in figure 3(c) from the text to emphasize the disparity between the dephasing times for one-quantum and zero-quantum coherences. Because individual excitons cannot be resolved in FMO, the beating signal should not contain only the exciton 3 optical frequency. Accordingly, the signal was fit to the sum of two independently decaying sine waves (red line) with one frequency fixed at $12,344 \text{ cm}^{-1}$ and the other varied as a fitting parameter. A magnified view of the signal and fit is given in the right panel. The fit is plotted every 4 fs in both panels in order to match the experimental sampling frequency, which is below the optical Nyquist frequency. This analysis gives a dephasing rate for the exciton 3 one-quantum coherence of $\gamma_3 = 320 \pm 8 \text{ cm}^{-1}$. The lifetime of this coherence, about 100 fs, is an order of magnitude less than the lifetimes of the zero-quantum coherences analyzed by the same method.
Figure 2. A comparison of least-squares fits of the waiting time beating signal to the sum of two independently decaying sine waves using variable difference frequencies (top panel, taken from figure 3(c) from the text) and difference frequencies fixed to the values taken from the Hamiltonian calculated by Adolphs and Renger for exciton 1-2 and 1-3 coherences (bottom panel). In both panels, the integrated amplitude of the cross-peak is plotted against waiting time after removal of exponential population decay (blue line) along with the fit (red line).