SUPPORTING INFORMATION

Two-Dimensional Infrared Spectroscopy of Antiparallel β-Sheet Secondary Structure

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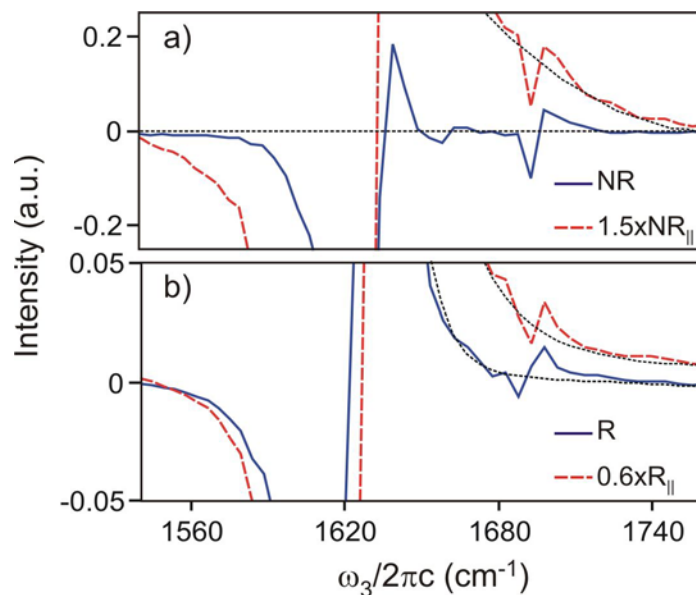**Projection angle between modes α- and α+ for Con A.** Our determination of the projection angle between the α- and α+ transition dipole moments in Con A used the relative amplitude ratios of the cross peaks centered at $(\omega_1, \omega_3) = (1630, 1698)$ cm^{-1}. This cross peak shows relatively minor interference effects with diagonal features, whereas the other $(\omega_1 > \omega_3)$ cross peak interferes strongly with the α− diagonal peak. The cross-peak amplitude ratio between the parallel (ZZZZ) and perpendicular (ZZYY) spectra are compared separately for slices through the constituent rephasing (R) and nonrephasing (NR) spectra.[1] The dependence of the cross peak amplitude ratio $A_\perp/A_\parallel$ on θ is calculated as described in Ref. 1, and is given in Figure 1S.
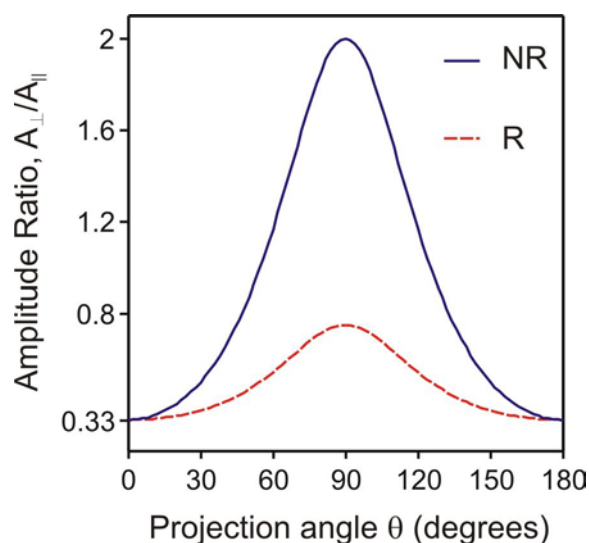
Figure 1S. Cross-peak amplitude ratios $A_\perp/A_\parallel$ as a function of the projection angle between two coupled vibrations for nonrephasing (NR, solid lines) and rephasing (R, dashed line) spectra.

Figures 2S shows two pairs of slices at $\omega_1 = 1630$ cm^{-1} taken from perpendicular and parallel nonrephasing and rephasing spectra of Con A. The ratio of the cross peak amplitudes, $A_\perp/A_\parallel = 1.5 \pm 0.1$ for nonrephasing spectra (Fig. 1S.a) and $A_\perp/A_\parallel = 0.6 \pm 0.05$ for rephasing spectra after correcting for interference with the sloping background signal. These values correspond to a projection angle of 69°±4° and 65°±5°, respectively. We therefore report an average value of 67°±7°. Note that errors in the determination of the

relative amplitudes reflect variation in baseline subtraction, experimental noise and phasing of the 2D spectrum.

Figure 2S. Anisotropy measurements used in the determination of the projection angle between modes α- and α+ for Con A. Vertical slices corresponding to $\omega_1 = 1630$ cm^{-1} taken from perpendicular (solid blue lines) and parallel (dashed red line) polarized spectra. Nonrephasing (NR) spectra in (a) and rephasing spectra (R) in (b). Cross peak interfere with a sloping baseline (short dashed lines), which are subtracted prior to determination of the relative amplitudes. Note that the parallel slices in (a) and (b) are offset by a constant with respect to the perpendicular slices.

REFERENCES

1. M. Khalil, N. Demirdöven, and A. Tokmakoff, J. Phys. Chem. A **107** (27), 5258 (2003).