Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supporting Information:Polariton Induced Conical Intersection and Berry Phase

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I. Molecular alignments

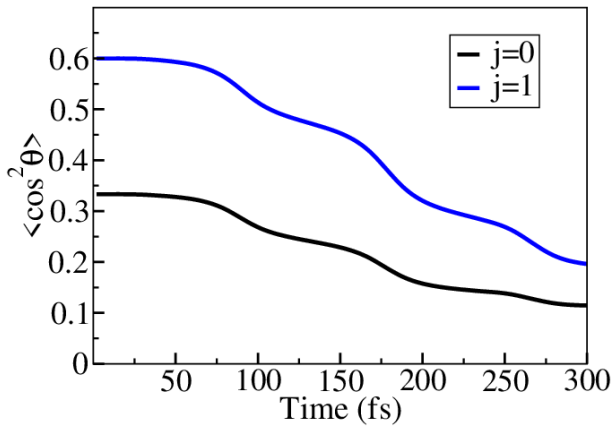

Figure S1: Time evolution of the molecular alignment obtained from the 2D model for the rovibrational wavefunction $J = 0$ and $J = 1$.

The molecular alignment is computed from the expectation value of $\cos^2\theta$ as follows

$$\langle \cos^2\theta \rangle = \langle \Psi(R, \theta, t) | \cos^2\theta | \Psi(R, \theta, t) \rangle \tag{S1}$$

where $\Psi(R, \theta, t)$ is the total polariton wavefunction. Figure S1 presents the molecular orientation as a function of time for the 2D model for the rovibrational wavefunction $J = 0$ and $J = 1$. During the first 50 fs the molecular orientation does not change. This explains why the results of 1D and 2D model are almost identical early time dynamics, for those result presented in Figure 6 of the main text.

II. The nuclear probability density

Figure S2 presents the NPD for the $|-, 0\rangle$ and $|g, 0\rangle$ states obtained from the three models (2D, non-BP, 1D) with the initial rotational quantum number $J = 0$ and $J = 1$. The results in Figure S2 suggest that the transitions of the wavefunction from the $|-, 0\rangle$ surface to $|g, 0\rangle$ surface through the avoided crossing do not affect the wavefunction symmetry.

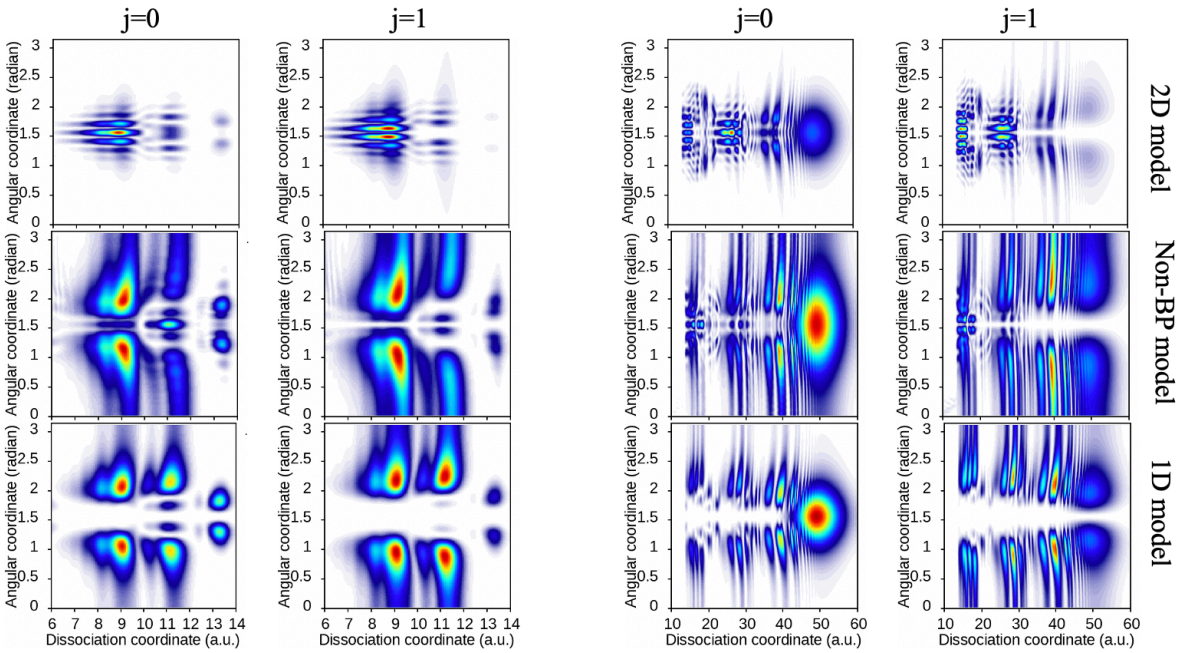

Figure S2: The nuclear probability density of the $|-,0\rangle$ state (left panel) at $t = 240$ fs and $|g,0\rangle$ state (right panel) at $t = 380$ fs with $J = 0$ and $J = 1$.