

# Supplemental Material for “Theory of rotationally-resolved two-dimensional infrared spectroscopy”

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## I. R-branch diagrams

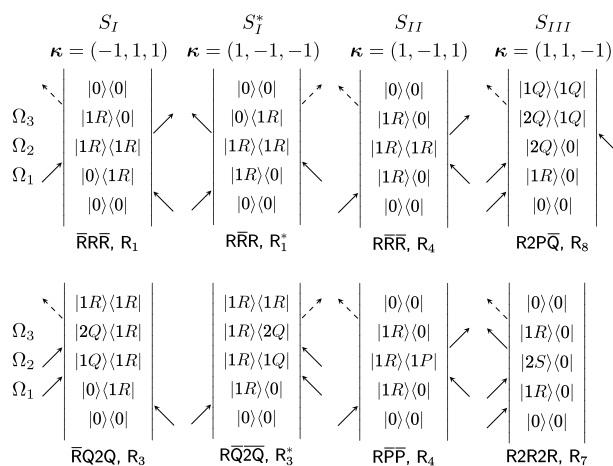


Figure S1: R-branch analogue of Fig. 1 in main article. Diagrams of third-order rovibrational pathways phase-matched in direction:  $S_I$ , first column;  $S_I^*$ , second column;  $S_{II}$ , third column;  $S_{III}$ , fourth column. First row contains pathways not in a rotationally-coherent (RC) state during waiting time. Second row contains RC pathways.

## II. Diagonal branch and subbranches

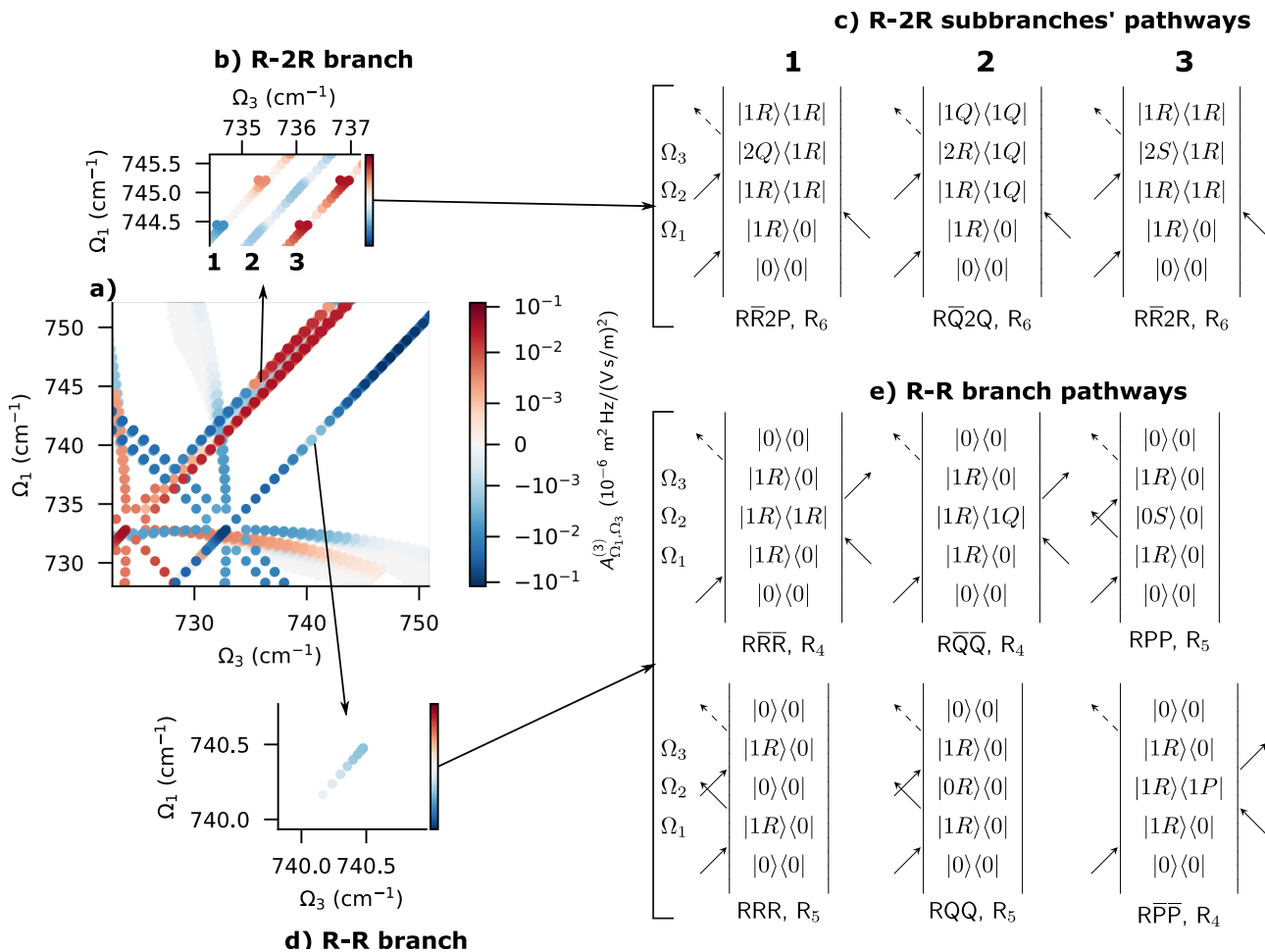


Figure S2: Subbranches in  $S_{11}$  direction. a) Zoom in on the top right part of Fig. 4 of the main article. b) R-2R branch is split into three subbranches (1, 2, 3). Subbranches 1 and 2 are rotationally-coherent. c) Each subbranch comprises one third-order pathway. The subbranches can be identified by the  $\Omega_3$  coherence, which are:  $|2Q\rangle\langle 1R|$  for 1,  $|2R\rangle\langle 1Q|$  for 2 and  $|2S\rangle\langle 1R|$  for 3. d) For each  $J_i$  there are multiple resonances corresponding to different projection of the rotational angular momentum onto molecular axis,  $K_m$ . e) Each R-R resonance is composed of 6 pathways which differ only by  $\Omega_2$  coherence.

### III. Rotational coherence frequencies - $S_{II}$

Table S1: Frequencies of rotational coherences for pathways phase-matched in the  $S_{II}$  direction.

Branch	Subbranch ( $\Omega_1, \Omega_3$ )	Rot. coherence	Pathway label	Exact frequency	Approx. frequency
P-2P	$ 0\rangle\langle 1P ,  1Q\rangle\langle 2P $	$ 1Q\rangle\langle 1P $	$\overline{PQ}2Q$	$-F(v_1, J_i, K) + F(v_1, J_i - 1, K)$	$-2B_1 J_i$
P-2P	$ 0\rangle\langle 1P ,  1R\rangle\langle 2Q $	$ 1R\rangle\langle 1P $	$\overline{PR}2R$	$F(v_1, J_i - 1, K) - F(v_1, J_i + 1, K)$	$-2B_1 (2J_i + 1)$
P-2Q	$ 0\rangle\langle 1P ,  1Q\rangle\langle 2Q $	$ 1Q\rangle\langle 1P $	$\overline{PQ}2R$	$-F(v_1, J_i, K) + F(v_1, J_i - 1, K)$	$-2B_1 J_i$
P-P	$ 0\rangle\langle 1P ,  0\rangle\langle 1P $	$ 0\rangle\langle 0O $	PRR	$-F(v_0, J_i, K) + F(v_0, J_i - 2, K)$	$-2B_0 (2J_i - 1)$
P-P	$ 0\rangle\langle 1P ,  0\rangle\langle 1P $	$ 0\rangle\langle 0P $	PQQ	$-F(v_0, J_i, K) + F(v_0, J_i - 1, K)$	$-2B_0 J_i$
P-P	$ 0\rangle\langle 1P ,  0\rangle\langle 1P $	$ 1Q\rangle\langle 1P $	$\overline{PQ}Q$	$-F(v_1, J_i, K) + F(v_1, J_i - 1, K)$	$-2B_1 J_i$
P-P	$ 0\rangle\langle 1P ,  0\rangle\langle 1P $	$ 1R\rangle\langle 1P $	$\overline{PR}R$	$F(v_1, J_i - 1, K) - F(v_1, J_i + 1, K)$	$-2B_1 (2J_i + 1)$
P-Q	$ 0\rangle\langle 1P ,  0P\rangle\langle 1P $	$ 1Q\rangle\langle 1P $	$\overline{PQ}R$	$-F(v_1, J_i, K) + F(v_1, J_i - 1, K)$	$-2B_1 J_i$
P-Q	$ 0\rangle\langle 1P ,  0\rangle\langle 1Q $	$ 0\rangle\langle 0P $	PQR	$-F(v_0, J_i, K) + F(v_0, J_i - 1, K)$	$-2B_0 J_i$
Q-2P	$ 0\rangle\langle 1Q ,  1R\rangle\langle 2Q $	$ 1R\rangle\langle 1Q $	$\overline{QR}2Q$	$F(v_1, J_i, K) - F(v_1, J_i + 1, K)$	$-2B_1 (J_i + 1)$
Q-2Q	$ 0\rangle\langle 1Q ,  1P\rangle\langle 2P $	$ 1P\rangle\langle 1Q $	$\overline{QP}2P$	$F(v_1, J_i, K) - F(v_1, J_i - 1, K)$	$2B_1 J_i$
Q-2Q	$ 0\rangle\langle 1Q ,  1R\rangle\langle 2R $	$ 1R\rangle\langle 1Q $	$\overline{QR}2R$	$F(v_1, J_i, K) - F(v_1, J_i + 1, K)$	$-2B_1 (J_i + 1)$
Q-2R	$ 0\rangle\langle 1Q ,  1P\rangle\langle 2Q $	$ 1P\rangle\langle 1Q $	$\overline{QP}2Q$	$F(v_1, J_i, K) - F(v_1, J_i - 1, K)$	$2B_1 J_i$
Q-P	$ 0\rangle\langle 1Q ,  0R\rangle\langle 1Q $	$ 1R\rangle\langle 1Q $	$\overline{QR}Q$	$F(v_1, J_i, K) - F(v_1, J_i + 1, K)$	$-2B_1 (J_i + 1)$
Q-P	$ 0\rangle\langle 1Q ,  0\rangle\langle 1P $	$ 0\rangle\langle 0P $	QRQ	$-F(v_0, J_i, K) + F(v_0, J_i - 1, K)$	$-2B_0 J_i$
Q-Q	$ 0\rangle\langle 1Q ,  0\rangle\langle 1Q $	$ 0\rangle\langle 0P $	QRR	$-F(v_0, J_i, K) + F(v_0, J_i - 1, K)$	$-2B_0 J_i$
Q-Q	$ 0\rangle\langle 1Q ,  0\rangle\langle 1Q $	$ 0\rangle\langle 0R $	QPP	$-F(v_0, J_i, K) + F(v_0, J_i + 1, K)$	$2B_0 (J_i + 1)$
Q-Q	$ 0\rangle\langle 1Q ,  0\rangle\langle 1Q $	$ 1P\rangle\langle 1Q $	$\overline{QP}P$	$F(v_1, J_i, K) - F(v_1, J_i - 1, K)$	$2B_1 J_i$
Q-Q	$ 0\rangle\langle 1Q ,  0\rangle\langle 1Q $	$ 1R\rangle\langle 1Q $	$\overline{QR}R$	$F(v_1, J_i, K) - F(v_1, J_i + 1, K)$	$-2B_1 (J_i + 1)$
Q-R	$ 0\rangle\langle 1Q ,  0P\rangle\langle 1Q $	$ 1P\rangle\langle 1Q $	$\overline{QP}Q$	$F(v_1, J_i, K) - F(v_1, J_i - 1, K)$	$2B_1 J_i$
Q-R	$ 0\rangle\langle 1Q ,  0\rangle\langle 1R $	$ 0\rangle\langle 0R $	QPQ	$-F(v_0, J_i, K) + F(v_0, J_i + 1, K)$	$2B_0 (J_i + 1)$
R-2Q	$ 0\rangle\langle 1R ,  1Q\rangle\langle 2Q $	$ 1Q\rangle\langle 1R $	$\overline{RQ}2P$	$-F(v_1, J_i, K) + F(v_1, J_i + 1, K)$	$2B_1 (J_i + 1)$
R-2R	$ 0\rangle\langle 1R ,  1P\rangle\langle 2Q $	$ 1P\rangle\langle 1R $	$\overline{RP}2P$	$-F(v_1, J_i - 1, K) + F(v_1, J_i + 1, K)$	$2B_1 (2J_i + 1)$
R-2R	$ 0\rangle\langle 1R ,  1Q\rangle\langle 2R $	$ 1Q\rangle\langle 1R $	$\overline{RQ}2Q$	$-F(v_1, J_i, K) + F(v_1, J_i + 1, K)$	$2B_1 (J_i + 1)$
R-Q	$ 0\rangle\langle 1R ,  0R\rangle\langle 1R $	$ 1Q\rangle\langle 1R $	$\overline{RQ}P$	$-F(v_1, J_i, K) + F(v_1, J_i + 1, K)$	$2B_1 (J_i + 1)$
R-Q	$ 0\rangle\langle 1R ,  0\rangle\langle 1Q $	$ 0\rangle\langle 0R $	RQP	$-F(v_0, J_i, K) + F(v_0, J_i + 1, K)$	$2B_0 (J_i + 1)$
R-R	$ 0\rangle\langle 1R ,  0\rangle\langle 1R $	$ 0\rangle\langle 0R $	RQQ	$-F(v_0, J_i, K) + F(v_0, J_i + 1, K)$	$2B_0 (J_i + 1)$
R-R	$ 0\rangle\langle 1R ,  0\rangle\langle 1R $	$ 0\rangle\langle 0S $	RPP	$-F(v_0, J_i, K) + F(v_0, J_i + 2, K)$	$2B_0 (2J_i + 3)$
R-R	$ 0\rangle\langle 1R ,  0\rangle\langle 1R $	$ 1P\rangle\langle 1R $	$\overline{RP}P$	$-F(v_1, J_i - 1, K) + F(v_1, J_i + 1, K)$	$2B_1 (2J_i + 1)$
R-R	$ 0\rangle\langle 1R ,  0\rangle\langle 1R $	$ 1Q\rangle\langle 1R $	$\overline{RQ}Q$	$-F(v_1, J_i, K) + F(v_1, J_i + 1, K)$	$2B_1 (J_i + 1)$

#### IV. Rotational coherence frequencies - $S_I$

Table S2: Frequencies of rotational coherences for pathways phase-matched in the  $S_I$  direction.

Branch	Subbranch ( $\Omega_1, \Omega_3$ )	Rot. coherence	Pathway label	Exact frequency	Approx. frequency
P-2Q	$ 0\rangle\langle 1P ,  2P\rangle\langle 1P $	$ 1Q\rangle\langle 1P $	$\overline{PQ2P}$	$-F(v_1, J_i, K) + F(v_1, J_i - 1, K)$	$-2B_1 J_i$
P-2R	$ 0\rangle\langle 1P ,  2Q\rangle\langle 1P $	$ 1Q\rangle\langle 1P $	$\overline{PQ2Q}$	$-F(v_1, J_i, K) + F(v_1, J_i - 1, K)$	$-2B_1 J_i$
P-2R	$ 0\rangle\langle 1P ,  2Q\rangle\langle 1P $	$ 1R\rangle\langle 1P $	$\overline{PR2P}$	$F(v_1, J_i - 1, K) - F(v_1, J_i + 1, K)$	$-2B_1 (2J_i + 1)$
P-Q	$ 0\rangle\langle 1P ,  1P\rangle\langle 0P $	$ 0\rangle\langle 0P $	$\overline{PQP}$	$-F(v_0, J_i, K) + F(v_0, J_i - 1, K)$	$-2B_0 J_i$
P-Q	$ 0\rangle\langle 1P ,  1Q\rangle\langle 0 $	$ 1Q\rangle\langle 1P $	$\overline{PQP}$	$-F(v_1, J_i, K) + F(v_1, J_i - 1, K)$	$-2B_1 J_i$
P-R	$ 0\rangle\langle 1P ,  1P\rangle\langle 0O $	$ 0\rangle\langle 0O $	$\overline{PRP}$	$-F(v_0, J_i, K) + F(v_0, J_i - 2, K)$	$-2B_0 (2J_i - 1)$
P-R	$ 0\rangle\langle 1P ,  1Q\rangle\langle 0P $	$ 0\rangle\langle 0P $	$\overline{PQQ}$	$-F(v_0, J_i, K) + F(v_0, J_i - 1, K)$	$-2B_0 J_i$
P-R	$ 0\rangle\langle 1P ,  1Q\rangle\langle 0P $	$ 1Q\rangle\langle 1P $	$\overline{PQQ}$	$-F(v_1, J_i, K) + F(v_1, J_i - 1, K)$	$-2B_1 J_i$
P-R	$ 0\rangle\langle 1P ,  1R\rangle\langle 0 $	$ 1R\rangle\langle 1P $	$\overline{PRP}$	$F(v_1, J_i - 1, K) - F(v_1, J_i + 1, K)$	$-2B_1 (2J_i + 1)$
Q-2P	$ 0\rangle\langle 1Q ,  2P\rangle\langle 1Q $	$ 1P\rangle\langle 1Q $	$\overline{QP2Q}$	$F(v_1, J_i, K) - F(v_1, J_i - 1, K)$	$2B_1 J_i$
Q-2Q	$ 0\rangle\langle 1Q ,  2Q\rangle\langle 1Q $	$ 1P\rangle\langle 1Q $	$\overline{QP2R}$	$F(v_1, J_i, K) - F(v_1, J_i - 1, K)$	$2B_1 J_i$
Q-2Q	$ 0\rangle\langle 1Q ,  2Q\rangle\langle 1Q $	$ 1R\rangle\langle 1Q $	$\overline{QR2P}$	$F(v_1, J_i, K) - F(v_1, J_i + 1, K)$	$-2B_1 (J_i + 1)$
Q-2R	$ 0\rangle\langle 1Q ,  2R\rangle\langle 1Q $	$ 1R\rangle\langle 1Q $	$\overline{QR2Q}$	$F(v_1, J_i, K) - F(v_1, J_i + 1, K)$	$-2B_1 (J_i + 1)$
Q-P	$ 0\rangle\langle 1Q ,  1P\rangle\langle 0 $	$ 1P\rangle\langle 1Q $	$\overline{QPQ}$	$F(v_1, J_i, K) - F(v_1, J_i - 1, K)$	$2B_1 J_i$
Q-P	$ 0\rangle\langle 1Q ,  1Q\rangle\langle 0R $	$ 0\rangle\langle 0R $	$\overline{QPQ}$	$-F(v_0, J_i, K) + F(v_0, J_i + 1, K)$	$2B_0 (J_i + 1)$
Q-Q	$ 0\rangle\langle 1Q ,  1P\rangle\langle 0P $	$ 0\rangle\langle 0P $	$\overline{QRP}$	$-F(v_0, J_i, K) + F(v_0, J_i - 1, K)$	$-2B_0 J_i$
Q-Q	$ 0\rangle\langle 1Q ,  1P\rangle\langle 0P $	$ 1P\rangle\langle 1Q $	$\overline{QPR}$	$F(v_1, J_i, K) - F(v_1, J_i - 1, K)$	$2B_1 J_i$
Q-Q	$ 0\rangle\langle 1Q ,  1R\rangle\langle 0R $	$ 0\rangle\langle 0R $	$\overline{QPR}$	$-F(v_0, J_i, K) + F(v_0, J_i + 1, K)$	$2B_0 (J_i + 1)$
Q-Q	$ 0\rangle\langle 1Q ,  1R\rangle\langle 0R $	$ 1R\rangle\langle 1Q $	$\overline{QRP}$	$F(v_1, J_i, K) - F(v_1, J_i + 1, K)$	$-2B_1 (J_i + 1)$
Q-R	$ 0\rangle\langle 1Q ,  1Q\rangle\langle 0P $	$ 0\rangle\langle 0P $	$\overline{QRQ}$	$-F(v_0, J_i, K) + F(v_0, J_i - 1, K)$	$-2B_0 J_i$
Q-R	$ 0\rangle\langle 1Q ,  1R\rangle\langle 0 $	$ 1R\rangle\langle 1Q $	$\overline{QRQ}$	$F(v_1, J_i, K) - F(v_1, J_i + 1, K)$	$-2B_1 (J_i + 1)$
R-2P	$ 0\rangle\langle 1R ,  2Q\rangle\langle 1R $	$ 1P\rangle\langle 1R $	$\overline{RP2R}$	$-F(v_1, J_i - 1, K) + F(v_1, J_i + 1, K)$	$2B_1 (2J_i + 1)$
R-2P	$ 0\rangle\langle 1R ,  2Q\rangle\langle 1R $	$ 1Q\rangle\langle 1R $	$\overline{RQ2Q}$	$-F(v_1, J_i, K) + F(v_1, J_i + 1, K)$	$2B_1 (J_i + 1)$
R-2Q	$ 0\rangle\langle 1R ,  2R\rangle\langle 1R $	$ 1Q\rangle\langle 1R $	$\overline{RQ2R}$	$-F(v_1, J_i, K) + F(v_1, J_i + 1, K)$	$2B_1 (J_i + 1)$
R-P	$ 0\rangle\langle 1R ,  1P\rangle\langle 0 $	$ 1P\rangle\langle 1R $	$\overline{RPR}$	$-F(v_1, J_i - 1, K) + F(v_1, J_i + 1, K)$	$2B_1 (2J_i + 1)$
R-P	$ 0\rangle\langle 1R ,  1Q\rangle\langle 0R $	$ 0\rangle\langle 0R $	$\overline{RQQ}$	$-F(v_0, J_i, K) + F(v_0, J_i + 1, K)$	$2B_0 (J_i + 1)$
R-P	$ 0\rangle\langle 1R ,  1Q\rangle\langle 0R $	$ 1Q\rangle\langle 1R $	$\overline{RQQ}$	$-F(v_1, J_i, K) + F(v_1, J_i + 1, K)$	$2B_1 (J_i + 1)$
R-P	$ 0\rangle\langle 1R ,  1R\rangle\langle 0S $	$ 0\rangle\langle 0S $	$\overline{RPR}$	$-F(v_0, J_i, K) + F(v_0, J_i + 2, K)$	$2B_0 (2J_i + 3)$
R-Q	$ 0\rangle\langle 1R ,  1Q\rangle\langle 0 $	$ 1Q\rangle\langle 1R $	$\overline{RQR}$	$-F(v_1, J_i, K) + F(v_1, J_i + 1, K)$	$2B_1 (J_i + 1)$
R-Q	$ 0\rangle\langle 1R ,  1R\rangle\langle 0R $	$ 0\rangle\langle 0R $	$\overline{RQR}$	$-F(v_0, J_i, K) + F(v_0, J_i + 1, K)$	$2B_0 (J_i + 1)$



## VI. Decomposition of molecular response into polarization classes

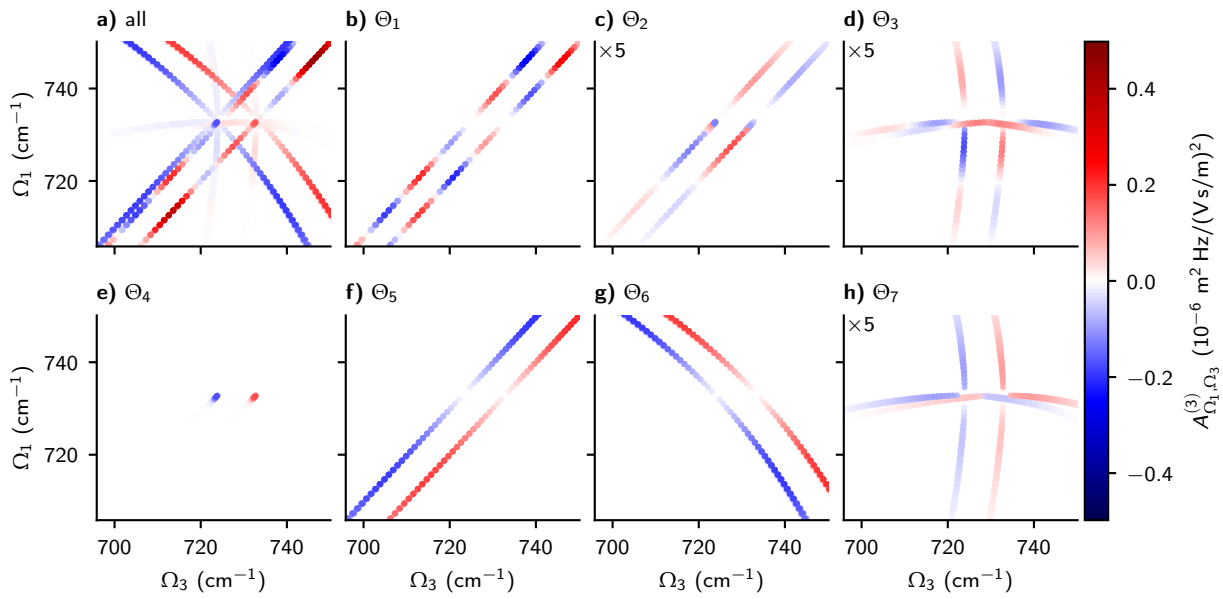


Figure S4: Polarization decomposition of third-order response of methyl chloride  $\nu_3$  vibrational mode phase-matched in the  $S_{III}$  direction. The plotted quantity is the 2D resonance amplitude defined in Eq. 22. a) total response; b–h) subsets of response associated with polarization classes  $\Theta_1$ – $\Theta_7$ , see Tab. I.

## VII. Table of G-factors

Table S3: Reduced G-factors  $(2J_i + 1)^{3/2}G(J_i, J_i + \Delta J_j, J_i + \Delta J_k, J_i + \Delta J_l; k)$  for different sequences of arguments, see Eq. (29).

$(\Delta J_j, \Delta J_k, \Delta J_l)$	$k = 0$	$k = 1$	$k = 2$
(-1, -2, -1)	0	0	$\sqrt{5}(2J_i + 1) / (5(2J_i - 1))$
(1, 2, 1)	0	0	$\sqrt{5}(2J_i + 1) / (5(2J_i + 3))$
(-1, 0, -1)	1/3	$-\sqrt{3}(J_i + 1) / 6J_i$	$\sqrt{5}(J_i + 1)(2J_i + 3) / (30J_i(2J_i - 1))$
(1, 0, 1)	1/3	$-\sqrt{3}J_i / (6(J_i + 1))$	$\sqrt{5}J_i(2J_i - 1) / (30(J_i + 1)(2J_i + 3))$
(1, 0, -1)	1/3	$\sqrt{3}/6$	$\sqrt{5}/30$
(-1, 0, 1)	1/3	$\sqrt{3}/6$	$\sqrt{5}/30$
(-1, -1, -1)	0	$\sqrt{3}(J_i - 1)(2J_i + 1) / (6J_i(2J_i - 1))$	$-\sqrt{5}(J_i + 1)(2J_i + 1) / (10J_i(2J_i - 1))$
(0, -1, -1)	0	$-\sqrt{3}\sqrt{J_i - 1}\sqrt{J_i + 1}\sqrt{2J_i + 1} / (6J_i\sqrt{2J_i - 1})$	$-\sqrt{5}\sqrt{J_i - 1}\sqrt{J_i + 1}\sqrt{2J_i + 1} / (10J_i\sqrt{2J_i - 1})$
(0, -1, 0)	0	$\sqrt{3}(J_i + 1) / 6J_i$	$-\sqrt{5}(J_i - 1) / 10J_i$
(0, 0, -1)	-1/3	$\sqrt{3}/6J_i$	$\sqrt{5}(2J_i + 3) / 30J_i$
(0, 0, 0)	1/3	$-\sqrt{3}/(6J_i(J_i + 1))$	$\sqrt{5}(2J_i - 1)(2J_i + 3) / (30J_i(J_i + 1))$
(0, 1, 0)	0	$\sqrt{3}J_i / (6(J_i + 1))$	$-\sqrt{5}(J_i + 2) / (10(J_i + 1))$
(1, 0, 0)	-1/3	$-\sqrt{3}/(6(J_i + 1))$	$\sqrt{5}(2J_i - 1) / (30(J_i + 1))$
(1, 1, 0)	0	$-\sqrt{3}\sqrt{J_i}\sqrt{J_i + 2}\sqrt{2J_i + 1} / (6(J_i + 1)\sqrt{2J_i + 3})$	$-\sqrt{5}\sqrt{J_i}\sqrt{J_i + 2}\sqrt{2J_i + 1} / (10(J_i + 1)\sqrt{2J_i + 3})$
(1, 1, 1)	0	$\sqrt{3}(J_i + 2)(2J_i + 1) / (6(J_i + 1)(2J_i + 3))$	$-\sqrt{5}J_i(2J_i + 1) / (10(J_i + 1)(2J_i + 3))$
(-1, -1, 0)	0	$-\sqrt{3}\sqrt{J_i - 1}\sqrt{J_i + 1}\sqrt{2J_i + 1} / (6J_i\sqrt{2J_i - 1})$	$-\sqrt{5}\sqrt{J_i - 1}\sqrt{J_i + 1}\sqrt{2J_i + 1} / (10J_i\sqrt{2J_i - 1})$
(-1, 0, 0)	-1/3	$\sqrt{3}/6J_i$	$\sqrt{5}(2J_i + 3) / 30J_i$
(0, 0, 1)	-1/3	$-\sqrt{3}/(6(J_i + 1))$	$\sqrt{5}(2J_i - 1) / (30(J_i + 1))$
(0, 1, 1)	0	$-\sqrt{3}\sqrt{J_i}\sqrt{J_i + 2}\sqrt{2J_i + 1} / (6(J_i + 1)\sqrt{2J_i + 3})$	$-\sqrt{5}\sqrt{J_i}\sqrt{J_i + 2}\sqrt{2J_i + 1} / (10(J_i + 1)\sqrt{2J_i + 3})$

Table S4: Reduced G-factors  $(2J_i + 1)^{3/2}G(J_i, J_i + \Delta J_j, J_i + \Delta J_k, J_i + \Delta J_l; k)$  in the high- $J_i$  limit for different sequences of arguments, see Eq. (29).

$(\Delta J_j, \Delta J_k, \Delta J_l)$	$k = 0$	$k = 1$	$k = 2$
(-1, -2, -1)	0	0	$\sqrt{5}/5$
(1, 2, 1)	0	0	$\sqrt{5}/5$
(-1, 0, -1)	1/3	$-\sqrt{3}/6$	$\sqrt{5}/30$
(1, 0, 1)	1/3	$-\sqrt{3}/6$	$\sqrt{5}/30$
(1, 0, -1)	1/3	$\sqrt{3}/6$	$\sqrt{5}/30$
(-1, 0, 1)	1/3	$\sqrt{3}/6$	$\sqrt{5}/30$
(-1, -1, -1)	0	$\sqrt{3}/6$	$-\sqrt{5}/10$
(0, -1, -1)	0	$-\sqrt{3}/6$	$-\sqrt{5}/10$
(0, -1, 0)	0	$\sqrt{3}/6$	$-\sqrt{5}/10$
(0, 0, -1)	-1/3	0	$\sqrt{5}/15$
(0, 0, 0)	1/3	0	$2\sqrt{5}/15$
(0, 1, 0)	0	$\sqrt{3}/6$	$-\sqrt{5}/10$
(1, 0, 0)	-1/3	0	$\sqrt{5}/15$
(1, 1, 0)	0	$-\sqrt{3}/6$	$-\sqrt{5}/10$
(1, 1, 1)	0	$\sqrt{3}/6$	$-\sqrt{5}/10$
(-1, -1, 0)	0	$-\sqrt{3}/6$	$-\sqrt{5}/10$
(-1, 0, 0)	-1/3	0	$\sqrt{5}/15$
(0, 0, 1)	-1/3	0	$\sqrt{5}/15$
(0, 1, 1)	0	$-\sqrt{3}/6$	$-\sqrt{5}/10$

### VIII. Table of R-factor coefficients

Table S5: Coefficients defining polarization-angular momentum dependence factors,  $R_0^{(0)}(\vec{\epsilon}; \vec{J})$ . See Eq. (40).

Label	$c_{00}$	$c_{12}$	$c_{13}$	$c_{14}$
P2P2P, P2P $\bar{P}$ , PR $\bar{P}$ , PRR	$(2J_i + 1)/(2J_i - 1)$	6	1	1
P2Q2P, P $\bar{Q}$ 2 $\bar{P}$ , PQR, P $\bar{Q}$ $\bar{R}$ , Q2P2Q, QP $\bar{Q}$ , QP2 $\bar{Q}$ , Q $\bar{R}$ Q	$-\sqrt{J_i - 1}\sqrt{J_i + 1}\sqrt{2J_i + 1}/(J_i\sqrt{2J_i - 1})$	3	-2	3
PPP, P2R2R	$1/(J_i(2J_i - 1))$	$2J_i^2 + 5J_i + 3$	$12J_i^2 - 2$	$2J_i^2 - 5J_i + 3$
PPQ, P2R2Q, QQP, Q $\bar{Q}$ 2 $\bar{P}$ , Q2Q2R, Q $\bar{Q}$ R	$1/J_i$	$2J_i + 3$	$-3J_i - 2$	$3 - 3J_i$
PPR, P2R2P, R2P2R, RRP	1	1	1	6
PP $\bar{P}$ , P2R $\bar{P}$	$1/(J_i(2J_i - 1))$	$2J_i^2 + 5J_i + 3$	$2J_i^2 - 5J_i + 3$	$12J_i^2 - 2$
PP $\bar{Q}$ , P2R $\bar{Q}$ , QQP $\bar{P}$ , Q2Q $\bar{P}$ , Q $\bar{Q}$ 2P, Q $\bar{Q}$ R	$1/J_i$	$2J_i + 3$	$3 - 3J_i$	$-3J_i - 2$
PP $\bar{R}$ , P2R $\bar{R}$ , R2P $\bar{P}$ , RR $\bar{P}$	1	1	6	1
PQQ, P2Q2Q	$-(2J_i + 1)/(J_i(2J_i - 1))$	$3J_i + 3$	$3J_i - 2$	$3 - 2J_i$
PQ $\bar{P}$ , P2Q $\bar{P}$	$-(2J_i + 1)/(J_i(2J_i - 1))$	$3J_i + 3$	$3 - 2J_i$	$3J_i - 2$
PQ $\bar{Q}$ , P2Q $\bar{Q}$ , P $\bar{Q}$ Q, P $\bar{Q}$ 2Q, Q2P $\bar{P}$ , QP2P, Q $\bar{P}$ R, Q $\bar{P}$ R	$-\sqrt{J_i - 1}\sqrt{J_i + 1}\sqrt{2J_i + 1}/(J_i\sqrt{2J_i - 1})$	3	3	-2
PP2P, P $\bar{P}$ R	$(2J_i + 1)/(2J_i - 1)$	1	6	1
PP $\bar{P}$ , PP2R	$1/(J_i(2J_i - 1))$	$2J_i^2 - 5J_i + 3$	$2J_i^2 + 5J_i + 3$	$12J_i^2 - 2$
PP $\bar{Q}$ , PP2Q	$(2J_i + 1)/(J_i(2J_i - 1))$	$2J_i - 3$	$-3J_i - 3$	$2 - 3J_i$
PP2 $\bar{P}$ , P $\bar{P}$ R	$(2J_i + 1)/(2J_i - 1)$	1	1	6
PP $\bar{P}$ , PP2 $\bar{R}$	$1/(J_i(2J_i - 1))$	$2J_i^2 - 5J_i + 3$	$12J_i^2 - 2$	$2J_i^2 + 5J_i + 3$
PP $\bar{Q}$ , PP2 $\bar{Q}$	$(2J_i + 1)/(J_i(2J_i - 1))$	$2J_i - 3$	$2 - 3J_i$	$-3J_i - 3$
PQ $\bar{P}$ , P $\bar{Q}$ 2R, Q2P $\bar{Q}$ , Q $\bar{P}$ Q, Q $\bar{P}$ 2Q, QR $\bar{Q}$	$-1/J_i$	$3J_i - 3$	$-2J_i - 3$	$3J_i + 2$
PQ $\bar{Q}$ , P $\bar{Q}$ 2 $\bar{Q}$ , Q2P2P, Q $\bar{P}$ $\bar{P}$ , Q $\bar{P}$ 2 $\bar{R}$ , QRR	$-1/J_i$	$3J_i - 3$	$3J_i + 2$	$-2J_i - 3$
PR $\bar{P}$ , P $\bar{R}$ 2 $\bar{P}$ , P $\bar{R}$ 2R, P $\bar{R}$ $\bar{R}$ , R $\bar{P}$ 2P, R $\bar{P}$ $\bar{P}$ , R $\bar{P}$ R, R $\bar{P}$ 2 $\bar{R}$	1	6	1	1
Q2Q2P, Q $\bar{Q}$ $\bar{P}$ , QQR, Q $\bar{Q}$ 2 $\bar{R}$ , R2P2Q, RRQ	$1/(J_i + 1)$	$2J_i - 1$	$-3J_i - 1$	$-3J_i - 6$
QPP, Q $\bar{R}$ 2 $\bar{P}$ , Q2R2R, Q $\bar{R}$ $\bar{R}$ , R $\bar{Q}$ Q, R $\bar{Q}$ 2Q	$-1/(J_i + 1)$	$3J_i + 6$	$3J_i + 1$	$1 - 2J_i$
QPQ, Q2R2Q, Q $\bar{R}$ Q, Q $\bar{R}$ 2Q, RQP, RQP, R2Q2R, R $\bar{Q}$ 2 $\bar{R}$	$-\sqrt{J_i}\sqrt{J_i + 2}\sqrt{2J_i + 1}/((J_i + 1)\sqrt{2J_i + 3})$	3	-2	3
QP $\bar{Q}$ , Q2R $\bar{Q}$ , Q $\bar{R}$ Q, Q $\bar{R}$ 2Q, R $\bar{Q}$ 2P, R $\bar{Q}$ R	$-1/(J_i + 1)$	$3J_i + 6$	$1 - 2J_i$	$3J_i + 1$
QP $\bar{R}$ , Q $\bar{R}$ P, Q2R $\bar{R}$ , Q $\bar{R}$ 2R, RQ $\bar{Q}$ , R2Q $\bar{Q}$ , R $\bar{Q}$ Q, R $\bar{Q}$ 2Q	$-\sqrt{J_i}\sqrt{J_i + 2}\sqrt{2J_i + 1}/((J_i + 1)\sqrt{2J_i + 3})$	3	3	-2
QQQ, Q2Q2Q, Q $\bar{Q}$ Q, Q $\bar{Q}$ 2 $\bar{Q}$	$1/(J_i(J_i + 1))$	$4J_i^2 + 4J_i - 3$	$4J_i^2 + 4J_i + 2$	$4J_i^2 + 4J_i - 3$
QQ $\bar{Q}$ , Q2Q $\bar{Q}$ , Q $\bar{Q}$ Q, Q $\bar{Q}$ 2Q	$1/(J_i(J_i + 1))$	$4J_i^2 + 4J_i - 3$	$4J_i^2 + 4J_i - 3$	$4J_i^2 + 4J_i + 2$
Q $\bar{Q}$ P, Q $\bar{Q}$ R, Q2Q $\bar{R}$ , Q $\bar{Q}$ 2R, R2P $\bar{Q}$ , RRQ	$1/(J_i + 1)$	$2J_i - 1$	$-3J_i - 6$	$-3J_i - 1$
R2P2P, RRR	$1/((J_i + 1)(2J_i + 3))$	$2J_i^2 - J_i$	$12J_i^2 + 24J_i + 10$	$2J_i^2 + 9J_i + 10$
R2P $\bar{R}$ , RRR $\bar{R}$	$1/((J_i + 1)(2J_i + 3))$	$2J_i^2 - J_i$	$2J_i^2 + 9J_i + 10$	$12J_i^2 + 24J_i + 10$
RPP, RP $\bar{R}$ , R2R2R, R2R $\bar{R}$	$(2J_i + 1)/(2J_i + 3)$	6	1	1
RQQ, R2Q2Q	$-(2J_i + 1)/((J_i + 1)(2J_i + 3))$	$3J_i$	$3J_i + 5$	$-2J_i - 5$
RQ $\bar{R}$ , R2Q $\bar{R}$	$-(2J_i + 1)/((J_i + 1)(2J_i + 3))$	$3J_i$	$-2J_i - 5$	$3J_i + 5$
R $\bar{R}$ 2P, RRR	$1/((J_i + 1)(2J_i + 3))$	$2J_i^2 + 9J_i + 10$	$2J_i^2 - J_i$	$12J_i^2 + 24J_i + 10$
R $\bar{R}$ P, R $\bar{R}$ 2R	$(2J_i + 1)/(2J_i + 3)$	1	6	1
R $\bar{R}$ Q, R $\bar{R}$ 2Q	$(2J_i + 1)/((J_i + 1)(2J_i + 3))$	$2J_i + 5$	$-3J_i$	$-3J_i - 5$
R $\bar{R}$ 2 $\bar{P}$ , RRR $\bar{R}$	$1/((J_i + 1)(2J_i + 3))$	$2J_i^2 + 9J_i + 10$	$12J_i^2 + 24J_i + 10$	$2J_i^2 - J_i$
R $\bar{R}$ P, R $\bar{R}$ 2 $\bar{R}$	$(2J_i + 1)/(2J_i + 3)$	1	1	6
R $\bar{R}$ Q, R $\bar{R}$ 2Q	$(2J_i + 1)/((J_i + 1)(2J_i + 3))$	$2J_i + 5$	$-3J_i - 5$	$-3J_i$