

**Table S1: Experimental Linear and Circular Polarizance^a for CO₂ + PFPE:
E_{inc} = 10.6(8) kcal/mol & θ_{inc} = 60°**

<i>J</i>	$P_{cir}^{90^\circ} \times 10^{-2}$	<i>J</i>	$P_{lin}^{90^\circ} \times 10^{-2}$	<i>J</i>	$P_{cir}^{0^\circ} \times 10^{-2}$	<i>J</i>	$P_{lin}^{0^\circ} \times 10^{-2}$
0	0.16(50)	0	0.04(60)	0	0.01(37)	0	-0.09(54)
2	-0.71(30)	2	0.25(50)	4	0.03(29)	2	0.23(10)
4	0.27(21)	4	0.14(24)	6	-0.12(24)	4	0.29(12)
6	1.59(90)	6	-0.12(8)	10	0.03(14)	6	0.25(10)
8	0.54(24)	8	-0.17(26)	16	-0.06(30)	8	0.26(16)
10	1.19(32)	10	-0.15(26)	20	0.36(11)	10	0.44(35)
12	1.21(59)	12	0.13(9)	24	0.05(19)	12	0.24(33)
14	1.64(37)	14	0.43(48)	28	-0.10(21)	20	0.10(13)
16	2.35(10)	16	-0.01(10)	32	-0.01(32)	24	0.50(36)
18	3.15(99)	18	0.20(20)	36	0.05(18)	28	0.18(20)
20	3.50(21)	20	0.41(22)	40	0.29(37)	40	0.56(31)
22	3.19(93)	22	0.63(37)	44	-0.29(37)	44	0.21(32)
24	4.41(35)	24	0.75(27)			48	1.25(60)
28	5.81(10)	28	1.15(27)				
30	7.49(95)	32	1.26(38)				
32	7.47(21)	36	1.57(44)				
34	7.62(96)	40	1.79(36)				
36	9.43(73)	44	2.10(24)				
40	11.15(15)	48	2.92(32)				
44	12.97(86)	52	2.05(27)				
48	15.28(95)						
52	16.09(47)						

^a $P_{cir}^{90^\circ}$ and $P_{cir}^{0^\circ}$ are circular polarizances for the $\varphi_{\text{laser}} = 90^\circ$ and 0° configurations, respectively. Likewise, $P_{lin}^{90^\circ}$ and $P_{lin}^{0^\circ}$ are linear polarizances for the two experimental setups shown in Figure 3.

Table S2: Orientation and Alignment Moments for Experimental CO₂ + PFPE

<i>J</i>	<i>A</i> ₀	<i>A</i> ₂₊	<i>O</i> ₁₋
0	0.0003(13)	0.0001(13)	0.0034(74)
2	0.0032(43)	-0.0002(43)	-0.0074(96)
4	0.0039(26)	0.0014(22)	0.0016(13)
6	0.0013(10)	0.0039(6)	0.0098(88)
8	0.0011(34)	0.0048(28)	0.0034(15)
10	0.0034(46)	0.0069(3)	0.0075(19)
12	0.0044(10)	0.0013(8)	0.0078(38)
14	0.0052(41)	-0.0052(41)	0.0106(24)
16	-0.0001(12)	0.0001(12)	0.0152(4)
18	0.0025(24)	-0.0025(24)	0.0204(74)
20	0.0063(25)	-0.0038(17)	0.0228(13)
22	0.0079(32)	-0.0079(32)	0.0209(61)
24	0.0157(50)	-0.0031(3)	0.0288(25)
26	0.0163(42)	-0.0077(16)	0.0346(18)
28	0.0169(34)	-0.0124(21)	0.0382(10)
30	0.0165(32)	-0.0142(29)	0.0493(63)
32	0.0161(34)	-0.0161(34)	0.0492(13)
34	0.0181(28)	-0.0181(28)	0.0503(63)
36	0.0202(22)	-0.0202(22)	0.0623(48)
38	0.0253(42)	-0.0180(32)	0.0642(46)
40	0.0304(68)	-0.0158(38)	0.0736(10)
44	0.0300(51)	-0.0245(21)	0.0860(57)
48	0.0536(82)	-0.0219(13)	0.1010(99)
52	0.0268(25)	-0.0268(25)	0.1064(75)

Table S3: Orientation and Alignment Moments for Simulated CO₂ + F-SAMs

<i>J</i> -Bin	$\langle J \rangle$	A_0	A_{1+}	A_{2+}	O_{1-}
0-5	3	-0.012(27)	-0.028(16)	-0.004(15)	0.008(18)
5-10	8	0.004(17)	-0.006(10)	0.009(9)	0.012(11)
10-15	13	-0.004(14)	0.010(8)	0.007(8)	0.020(8)
15-20	17	0.008(13)	0.020(8)	-0.001(7)	0.028(8)
20-25	23	-0.002(13)	0.034(8)	-0.024(7)	0.056(8)
25-30	27	0.006(13)	0.037(8)	-0.024(8)	0.067(9)
30-35	32	0.014(14)	0.070(8)	-0.040(8)	0.094(9)
35-40	37	0.014(16)	0.089(9)	-0.048(9)	0.118(10)
40-45	42	0.047(17)	0.107(10)	-0.062(9)	0.139(11)
45-50	47	0.057(19)	0.107(11)	-0.072(11)	0.153(13)
50-55	52	0.042(22)	0.141(13)	-0.075(12)	0.218(15)
55-60	57	0.037(26)	0.166(15)	-0.095(15)	0.250(18)
60-70	64	0.009(25)	0.171(14)	-0.148(15)	0.311(18)
70+	76	-0.061(35)	0.176(18)	-0.222(22)	0.416(27)