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Supplementary Information Accompanying: "Water Diffusion Measurements of Single Charged Aerosol Using H_2O/D_2O Isotope Exchange and Raman Spectroscopy in an Electrodynamic Balance"

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The Raman spectra are fit using a 7-term Gaussian expansion of the form:

$$Fit = \sum_{i=1}^{7} a exp\left(-\frac{(b-x)^2}{c^2}\right) \tag{S.1}$$

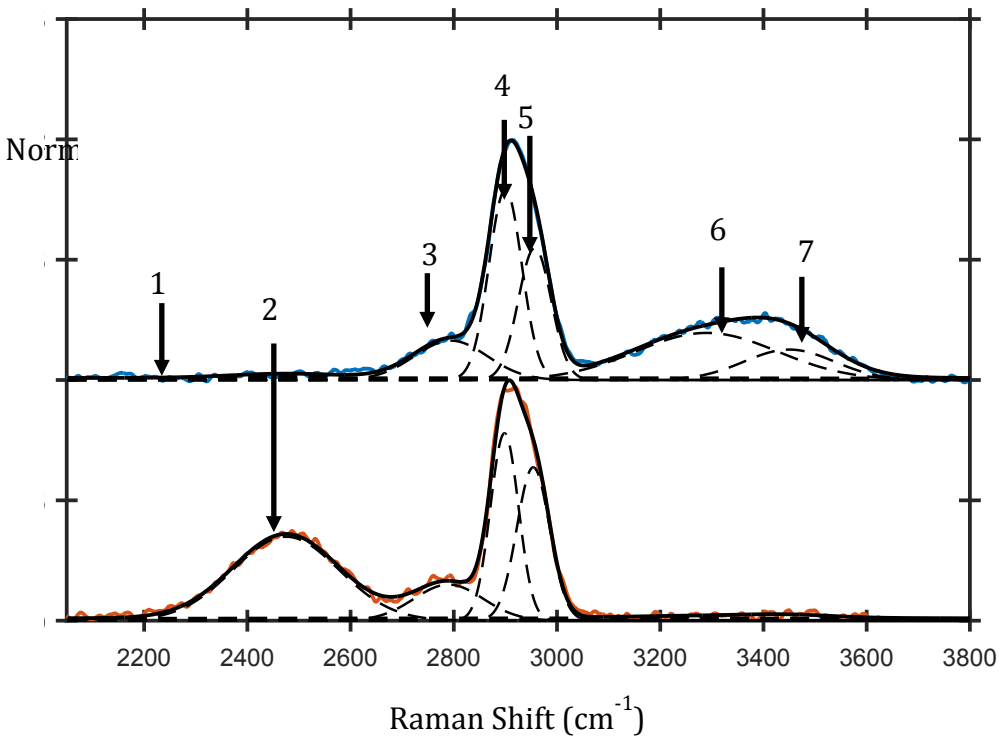

Figure S1. Raman spectra collected during an isotope exchange experiment carried out at 50% RH at early (top) and late (bottom) time points. The spectra are deconstructed into 7 Gaussians as described in the manuscript, which are shown here individually. The C-H stretches, peaks 4 and 5 peaking at 2930 cm^{-1}, are used for normalization.

Table S1. Gaussian fit parameters for each of the 7 Gaussians in a single sucrose droplet spectrum accompanying Figure S1 where *a*, *b,* and *c* follow Equation S1. The full dataset of fit parameters will be available in the Digital Collection.

Upper	**Peak**	**a**	**b**	**c**
	1	0.010	2915	2000
	2	0.017	2475	150
	3	0.164	2794	100
	4	0.791	2900	45
	5	0.549	2957	45
	6	0.196	3290	194
	7	0.127	3450	123
Lower	**Peak**	**a**	**b**	**c**
	1	0.012	2915	2000
	2	0.349	2475	145
	3	0.150	2790	95
	4	0.779	2898	39
	5	0.637	2954	45
	6	0.009	3290	200
	7	0.010	3450	120

The area of the ν(O-D), α, ν (C-H), and ν (O-H) peaks are calculated by the following equations:

$$Area_{OD} = Area_{Peak2} \quad \text{(S.2)}$$

$$Area_{\alpha} = Area_{Peak3} \quad \text{(S.3)}$$

$$Area_{CH} = Area_{Peak4} + Area_{Peak5} \quad \text{(S.4)}$$

$$Area_{OH} = Area_{Peak6} + Area_{Peak7} \quad \text{(S.5)}$$

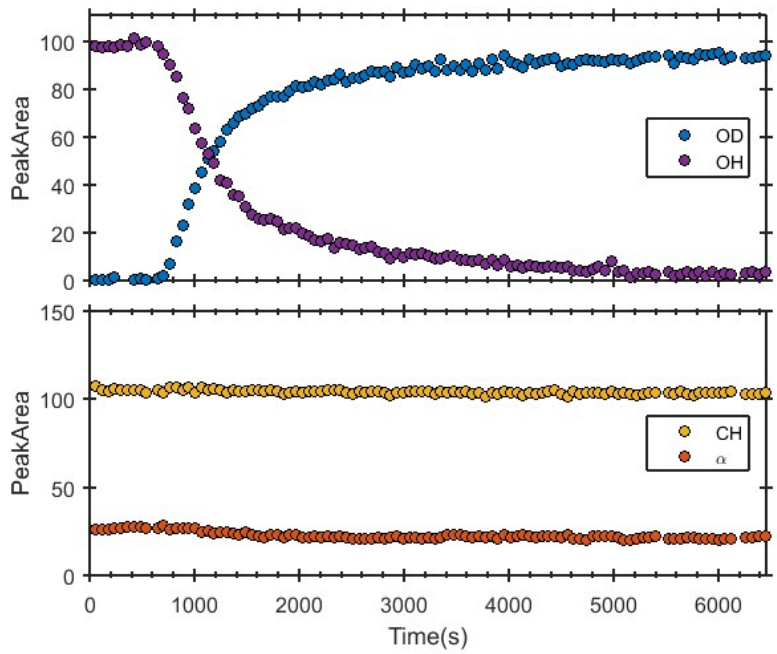

Figure S2. Peak area calculations for an experiment carried out at 54% RH which shows the growth of the ν(O-D), decay of the ν(O-H), and the constant contributions for the ν(C-H) and α over time.

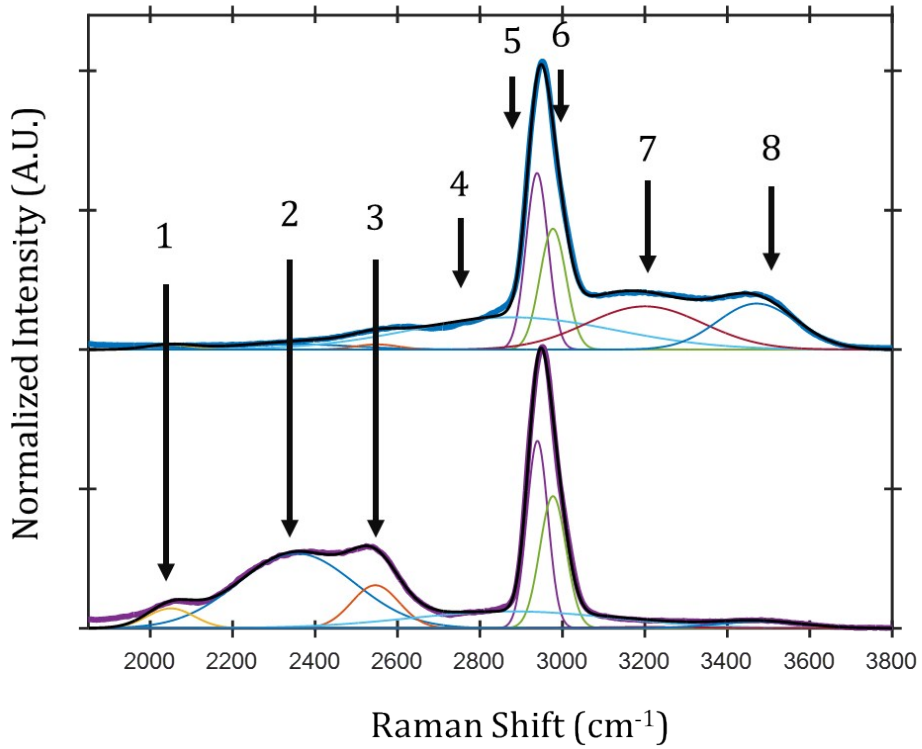

Figure S3. For CA droplets, the spectra were fit with 8 Gaussians. Spectra for the early stage of exchange (top, blue line) and near the completion of exchange (bottom, purple line) conducted at 21% RH are shown. Black curves represent the overall Gaussian fitting as the sum of the 8 Gaussian functions with different colors. Normalization of the data was done using the C-H stretch feature peaking at 2950 cm^{-1}. Representative Gaussian fit parameters for these data are shown in Table S2.

Table S2. Gaussian fit parameters for Figure S3, two spectra of CA droplets showing pre/post H_2O/D_2O exchange. 8 sets of parameters are used, and each parameter is analogous to Table S1.

Upper	Peak	a	b	c
	1	0.0194	2050	80
	2	0.0197	2350	210
	3	0.0163	2550	80
	4	0.1215	2880	350
	5	0.6362	2938	38
	6	0.4267	2977	45
	7	0.1537	3202	200
	8	0.1811	3473	198

Lower	Peak	a	b	C
	1	0.0753	2050	80
	2	0.2810	2354	206
	3	0.1742	2547	81
	4	0.0626	2880	350
	5	0.6702	2935	36
	6	0.4779	2977	45
	7	0.0000	3200	200
	8	0.0225	3480	140

The area of the ν(O-D), ν (C-H), and ν (O-H) peaks are calculated by the following equations (no α peak observed in CA droplets):

$$Area_{OD} = Area_{Peak1} + Area_{Peak2} + Area_{Peak3} \quad \text{(S.6)}$$

$$Area_{CH} = Area_{Peak5} + Area_{Peak6} \quad \text{(S.7)}$$

$$Area_{OH} = Area_{Peak4} + Area_{Peak7} + Area_{Peak8} \quad \text{(S.8)}$$

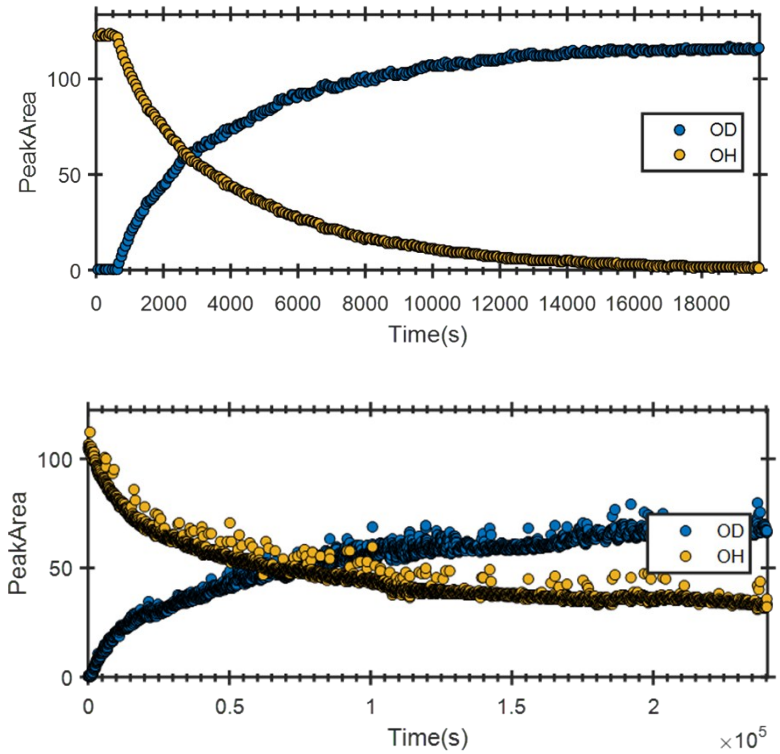

Figure S4. Peak area calculations for CA droplet at 21% (top) and 11% RH (bottom). Change in timescale shows the dramatic decrease in water diffusion rate.

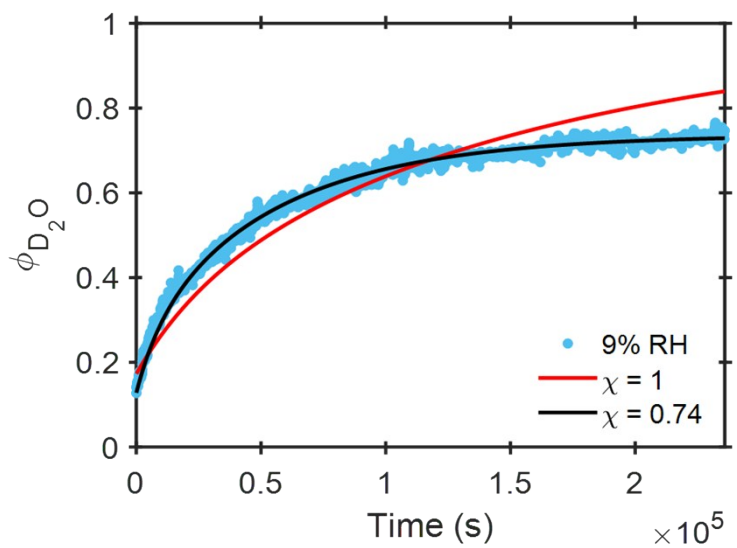

Figure S5. The result of using 1-parameter (eq. 7, red) and 2-parameter (eq. 9, black) fitting to ϕ_{D_2O} for CA droplet at 11% RH. Setting $\chi < 1$ represents the physical behavior better than assuming complete H_2O/D_2O exchange ($\chi = 1$).