Electronic Annex for:

Heteroepitaxial growth of cadmium carbonate at dolomite and calcite surfaces:

Mechanisms and rates

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Thermodynamics of the Cd-Ca-CO₃ system

The nearly ideal behavior of the otavite-calcite solid solution is supported by empirical data (Chang and Brice, 1971; Papadopoulos and Rowell, 1988; Konigsberger et al., 1991; Stipp et al., 1992), the similar sizes of Cd²⁺ and Ca²⁺ ions (0.95 Å and 1.00 Å, respectively), and the same crystal structure of the end-members. This implies that the two minerals should be completely miscible over all proportions of Ca and Cd. The degree of non-ideal mixing in the (Cd_x,Ca_{1-x})CO₃ solid solution has been inferred from experimental data (Rock et al., 1994), and can be described by the Guggenheim interaction parameters $a_0 = -0.8$ (Davis et al., 1987; Glynn and Reardon, 1990) and $a_0 = 1.2$ (Tesoriero and Pankow, 1996). These parameters give the activity coefficients of the solid solution endmembers at specific solid solution compositions (Glynn, 2000; Prieto, 2009).

A Lippmann diagram for the (Cd_x,Ca_{1-x})CO₃ solid solution depicts the thermodynamic equilibrium states in the system, i.e., the equilibrium compositions of the solid and the corresponding liquid (Lippmann, 1977, 1980; Gamsjager, 2000; Glynn, 2000). The total solubility products, $\Sigma\Pi$, given by:

$$\log \Sigma \Pi = K_{CdCO_3} X_{CdCO_3} \gamma_{CdCO_3} + K_{CaCO_3} X_{CaCO_3} \gamma_{CaCO_3} \text{ (solidus) and} \quad (\text{EA1})$$

$$\log \Sigma \Pi = 1 / \left[\frac{X_{Cd(aq)}}{K_{CdCO_3} \gamma_{CdCO_3}} + \frac{X_{Ca(aq)}}{K_{CaCO_3} \gamma_{CaCO_3}} \right] \text{ (solutus),} \quad (\text{EA2})$$

where K , X , and γ , are the solubility product constants, mole fractions, and activity coefficients of the end-members, were plotted against the mole fraction, X , of one end-member (in this case, $CdCO_3$) in the solid and in solution, for the ideal and non-ideal cases (Fig. EA1). The solubility products of the end-members vary by several orders of magnitude: $\log K_{sp, CaCO_3} = -8.48$ and $\log K_{sp, CdCO_3} = -12.00$. The shapes of the solidus and solutus curves give rise to the following observations: (1) There is a strong preference of the less soluble phase $CdCO_3$ to go into the solid solution, leading to a Cd-rich solid whose composition is largely invariant with the initial solution compositions, except for extremely Cd-poor solutions ($X_{Cd^{2+}} < 0.002$). (2) For a wide range of aqueous solution compositions (e.g., $1 > X_{Cd^{2+}} > 0.002$), there is a narrower range of solid solution compositions (e.g., $1 > X_{CdCO_3} > 0.9$) that are in equilibrium with this solution. For Cd-poor solutions (e.g., $X_{Cd^{2+}} < 0.002$), small changes in the aqueous composition have large effects on the Cd content of the solid solution (see also Prieto et al., 1997; Perez-Garrido et al., 2007). A solution that is at thermodynamic equilibrium with respect to any $(Cd_x, Ca_{1-x})CO_3$ solid will always be undersaturated with respect to pure calcite and otavite phases (Glynn and Reardon, 1990), as also shown by Gibbs free energy diagrams (Fig. EA2). The various degrees of non-ideality proposed in the literature that may be caused by the relatively large difference in the electronegativities of Cd (1.69) and Ca (1) (e.g., Pavlick, 1999) do not significantly affect the shapes of the solidus and solutus curves (Fig. EA1) because of the large difference between the solubility products of the endmembers (Prieto et al., 1997).

Figure EA1. Lippmann diagram for the ideal (solid black curves) and non-ideal (dotted and dashed black curves) $(\text{Cd}_x\text{Ca}_{1-x})\text{CO}_3$ solid solution. $\log \Sigma \Pi$ are calculated from Eqs. 1 and 2, and are plotted against mole fraction, X , of CdCO_3 . The variation of the activity coefficients of CdCO_3 and CaCO_3 in the non-ideal solid solution is described by empirical coefficient a_0 (Davis et al., 1987; Glynn and Reardon, 1990; Tesoriero and Pankow, 1996). For the ideal case, $a_0 = 0$. A horizontal tie line (solid blue line) gives the equilibrium concentrations (dashed blue lines) of the solid and solution from the solidus and solutus curves, respectively.

A Gibbs free energy diagram was used to determine if the formation of the solid solution is favorable. The Gibbs free energy of mixing for the ideal case,

$$\Delta G_{mixing, ideal} = -T\Delta S_{mixing} = RT(X_{CaCO_3} \ln X_{CaCO_3} + X_{CdCO_3} \ln X_{CdCO_3}), \quad (EA3)$$

was plotted against the mole fraction, X , of $CdCO_3$. The Gibbs free energy, G , of the solid solution is related to ΔG_{mixing} by the expression:

$$\Delta G_{mixing} = G_{solid\ solution} - G_{mechanical\ mixing}, \quad (EA4)$$

and a negative ΔG_{mixing} (Fig. EA2) denotes that the solid solution has a lower free energy than an equivalent mechanical mixture of the two end-members for all compositions, indicating that the system will tend to form the solid solution instead of co-existing separate end-members. There is a favorable thermodynamic force for mixing, and no miscibility gap is predicted.

Figure EA2. Gibbs free energy diagram for the $(Cd_x, Ca_{1-x})CO_3$ solid solution, assuming ideal mixing. A negative ΔG_{mixing} is obtained for all mole fractions, X , of $CdCO_3$ in the solid.

Figure EA3. Supersaturation of the solid solution, β , given in Eq. 3, as a function of the mole fraction, X , of CdCO_3 . In this example, $[\text{Cd}] = 0.01 \text{ mM}$ (dotted curve) or 0.5 mM (solid curve), and $[\text{Ca}] = [\text{CO}_3] = 0.5 \text{ mM}$. The values of maximum supersaturation, β_{max} , which occur at $X_{\text{CdCO}_3} \sim 0.985$ (dotted curve) or >0.99 (solid curve), are indicated by short vertical red lines.

Table EA1. Saturation indices of various phases for a solution containing $[Cd] = [Ca] = [CO_3] = 0.2 \text{ mM}$ ($pH = 7.4$), as calculated with PHREEQC. The solution is supersaturated with respect to otavite, and undersaturated with respect to all other phases. Only saturation indices above -15 are shown.

Phase	SI	log IAP	log K_{sp}
Otavite ($CdCO_3$)	1.57	-10.53	-12.00
Calcite ($CaCO_3$)	-2.02	-10.50	-8.48
Aragonite ($CaCO_3$)	-2.20	-10.50	-8.30
$Cd(OH)_2$	-2.62	11.02	13.64
$Cd(OH)_2$ (am)	-2.71	11.02	13.73
$CdOHCl$	-3.05	0.49	3.54
CO_2 (g)	-3.40	-21.55	-18.15
Monteponite (CdO)	-4.08	11.02	15.10
$CdCl_2 \cdot 2.5H_2O$	-8.13	-10.04	-1.91
$CdCl_2 \cdot H_2O$	-8.35	-10.04	-1.69
O_2 (g)	-8.67	74.42	83.09
$CdCl_2$	-9.38	-10.04	-0.66
Portlandite ($Ca(OH)_2$)	-11.76	11.05	22.8
Thermonatrite	-14.80	-14.16	0.64

81 Table EA2. Saturation indices of various phases for an aqueous solution containing 50 μM
 82 cadmium chloride, as calculated with PHREEQC. The solution is undersaturated with respect to
 83 all phases, including otavite.

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Phase	SI	log IAP	log K_{sp}
CdCO_3	-2.65	-14.65	-12.00
$\text{CO}_2(\text{g})$	-3.42	-21.56	-18.15
CdOHCl	-6.25	-2.72	3.54
$\text{Cd}(\text{OH})_2$	-6.73	6.91	13.64
$\text{Cd}(\text{OH})_2(\text{am})$	-6.82	6.91	13.73
CdO	-8.19	6.91	15.10
$\text{CdCl}_2 \cdot 2.5\text{H}_2\text{O}$	-10.43	-12.35	-1.91
$\text{CdCl}_2 \cdot \text{H}_2\text{O}$	-10.65	-12.35	-1.69
CdCl_2	-11.69	-12.35	-0.66
$\text{Cd}(\text{s})$	-17.68	-4.17	13.51
$\text{CH}_4(\text{g})$	-24.85	-65.89	-41.05
$\text{O}_2(\text{g})$	-60.92	22.16	83.09

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Calibration of absolute reflectivity and determination of scaling parameter

A scaling parameter was used to calibrate the reflectivity signals. The scale factor was determined typically from the samples with the least amounts of Cd, and was fixed to an average value in the final analysis. Alternatively, this parameter was determined individually for samples which have scale factors inconsistent with this average value. For each data set collected at the same beamline and beam energy, the scale factors for calibration of the reflectivity signal were determined as follows. The best-fit scale parameter at Q between $\sim 1.95 - 2.3 \text{ \AA}^{-1}$ and $\sim 4.15 - 4.5 \text{ \AA}^{-1}$ pertain to the “first” and “second” zones, respectively. For data collected at 13-BMC with a beam energy of 14.9 keV, the scale factor was determined from samples having a total Cd occupancy $< 1 \text{ ML}$ (Fig. EA4). For scans at 13-BMC collected at 28.6 keV, and for scans at 13-IDC, the scale factors were determined from the average of best-fit values for Samples XR-13 and XR-7, respectively, which had low Cd occupancies. For data with scale factors that were apparently inconsistent with other data collected at the same XR experimental setup, this parameter was fitted individually, and then fixed in the final analysis.

Figure EA4. Best-fit scale factors for samples with total Cd occupancies < 1 ML. An average value of 0.0205 was applied to data collected at 13-BMC with a beam energy of 14.9 keV.

For 33-IDD scans, scales were obtained from samples with < 2 ML Cd (Fig. EA5). However, unlike for the 13-BMC data, the values for the first and second zones are distinct, presumably due to an unknown source of systematic error. The derived Cd occupancies for individual analyses of the regions around the (104) and (208) Bragg peaks were generally consistent. The average of the second zone scale factor was used in the analysis of the data around the second order film Bragg peaks. The scaling parameter for all the samples are given in Table EA5.

Figure EA5. First and second zone scale factors for samples with total Cd occupancies < 2 ML, collected at 33-IDD.

Figure EA6. *Ex situ* AFM height and phase images at intermediate supersaturation ($1.0 < \log \beta_{max} < 2.0$), and at longer reaction times (1 day) where the dolomite surface is almost fully covered by the overgrowth. (A) and (B) are images of different areas on the sample. Features that are composed of several monolayers nucleating at one location were occasionally formed at the surface, as outlined by yellow box in (B) and shown in more detail in (C). Monolayer islands now have straighter edges. Initial solution: $[Cd^{2+}] = 0.2$, $[Ca^{2+}] = 0.211$ $[CO_3^{2-}] = 0.202$ mM (XR-19); $\log \beta_{max} = 1.48$.

Figure EA7. *In situ* AFM images of a calcite surface during reaction with 50 μM CdCl_2 solution. (A) Initial surface; (B) Same area on the sample after 15 minutes of reaction. Dissolution is evident from the development of pits that are more elongated in shape than the typical rhombohedral pits on calcite dissolved in water; (C-D) Height and phase images of a different area on the same sample, 30 minutes later, showing growth originating at step edges, as well as the formation of fan-shaped islands that are 3 $\text{\AA}$ in height.

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 135 Figure EA8. *Ex situ* AFM height and phase images showing Cd-carbonate overgrowths on
 136 calcite after reaction with solution containing 30 μM Cd. The solution was prepared by adding
 137 80 μL of 1 mM Cd to the 2.5 mL fluid cell containing DIW that had reacted with calcite for 40
 138 minutes. (A) and (B) are images of different areas in the same sample. The morphology of the
 139 overgrowth is similar to that observed in Figs. 7 and EA7.

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141 Figure EA9. *Ex situ* AFM images of calcite after reaction with a solution containing 1 μM
142 CdCl_2 , showing possible overgrowth formation indicated by bright areas at step edges in phase
143 image.

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146 Table EA3. Growth rates from XRF analysis

Sample number ^a	Domain size ^b $\times 10^4$ (Å)	XRF Cd occupancy ^c (ML)	Average growth rate ^d $\times 10^{-2}$ (Cd/Å ² /h)	Step normalized growth rate ^e $\times 10^2$ (Cd/Å/h)
XR-3_1	3.094 (0.543)	0.176 (0.058)	0.015 (5.0E-5)	0.047 (1.74)
XR-3_2	5.395 (0.828)	0.195 (0.057)	0.017 (4.9E-5)	0.090 (2.98)
XR-13_1	3.579 (0.176)	2.821 (0.149)	1.302 (6.9E-4)	4.662 (33.58)
XR-13_2	0.510 (0.055)	16.313 (0.186)	7.532 (8.6E-4)	3.884 (41.40)
XR-13_3	6.053 (0.326)	1.049 (0.121)	0.485 (5.6E-4)	2.933 (37.29)
XR-13_4	1.036 (0.048)	8.051 (0.123)	3.717 (5.7E-4)	3.852 (18.95)
XR-14_1	1.026 (0.064)	1.495 (0.074)	2.429 (1.2E-3)	2.493 (19.85)
XR-14_2	0.804 (0.054)	1.375 (0.085)	2.233 (1.4E-3)	1.795 (16.32)
XR-14_3	1.189 (0.110)	1.516 (0.079)	2.462 (1.3E-3)	2.929 (31.06)
XR-14_4	1.346 (0.078)	1.934 (0.060)	3.142 (9.7E-4)	4.228 (27.68)
XR-18_1	2.732 (0.095)	6.544 (0.146)	2.924 (6.5E-4)	7.987 (32.92)
XR-18_2	3.448 (0.107)	3.757 (0.187)	1.679 (8.4E-4)	5.787 (33.95)
XR-18_3	1.960 (0.063)	8.660 (0.149)	3.869 (6.7E-4)	7.585 (27.47)
XR-28_1	0.723 (0.038)	35.053 (0.233)	61.377 (4.1E-3)	44.38 (233.4)
XR-28_2	0.461 (0.059)	32.454 (0.228)	56.826 (4.0E-3)	26.21 (338.2)
XR-28_3	1.318 (0.070)	31.841 (0.158)	55.752 (2.8E-3)	73.48 (390.0)
XR-28_4	1.061 (0.058)	38.361 (0.239)	67.169 (4.2E-3)	71.26 (390.7)
XR-30_1	4.469 (0.272)	29.600 (0.156)	52.684 (2.8E-3)	235.5 (1438)
XR-30_2	1.424 (0.247)	53.363 (0.189)	94.980 (3.4E-3)	135.2 (2344)
XR-30_3	0.948 (0.068)	40.022 (0.200)	71.234 (3.6E-3)	67.56 (488.4)

147 ^a experiment numbers are as in Table 2.148 ^b substrate domain size, and its uncertainty.149 ^c total Cd occupancies from analysis of XRF data, and its uncertainty.150 ^d growth rate obtained from XRF data, and its uncertainty.151 ^e growth rate obtained from XRF data, normalized with step-spacing (^b), and its uncertainty.

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Analysis of domain size

The broadening of the specular CTR corresponds to the average domain size or step spacing on the surface, which is given by: $L_{\parallel} \sim \frac{2\pi}{Q \Delta\theta}$, where $L_{\parallel}$ is the lateral domain size, Q is the momentum transfer, and $\Delta\theta = \Delta(2\theta)/2$ is the angular width of the scan (Fenter, 2002). The angular width was estimated by fitting a Lorentzian curve to the integrated signal obtained from the Pilatus images, and by using the relation: $\Delta 2\theta = n \times 0.172 \frac{mm}{pixel} \div r$, where n is the number of pixels, and r is the distance of the sample from the detector, which was $r = 1113$ mm (13-BMC), 1108 mm (13-IDC) or 1183 mm (33-IDD). The best-fit Lorentzian widths were obtained by minimizing $\chi^2 = \left(\frac{1}{N}\right) \sum_i [(y_{calc} - y_i)/\sigma_i]^2$, where N , y_{calc} , y_i , and σ_i are the number of data points, best-fit signal, measured signal, and estimated uncertainty of the signal for each data point i , respectively. The results of the domain size analysis are given in Table EA4.

166 Table EA4. Results of substrate surface domain size analysis.

Q ($\text{\AA}^{-1}$)	Sample number	Lorentzian width ($\times 10^{-3}$)	σ , width ($\times 10^{-3}$)	χ^2	Domain size ($\text{\AA}$)	σ , domain size ($\text{\AA}$)
1.187	XR-2-1	0.362	0.041	10.41	29211.9	3281.6
1.187	XR-2-2	5.887	0.256	7.85	1797.9	78.3
1.188	XR-3-1	0.342	0.060	0.91	30944.6	5433.7
1.188	XR-3-2	0.196	0.030	0.91	53947.9	8284.9
1.188	XR-3-3	0.611	0.019	1.15	17317.0	524.7
1.188	XR-3-4	0.273	0.012	1.64	38717.6	1715.4
1.188	XR-3-5	2.981	0.117	1.05	3546.7	139.5
1.188	XR-3-6	2.140	0.090	1.89	4941.9	207.0
1.188	XR-3-7	2.109	0.050	1.10	5014.6	118.0
1.187	XR-4-1	0.208	0.007	1.16	50955.6	1741.9
1.188	XR-5-1	0.367	0.014	1.07	28842.8	1085.7
1.188	XR-5-2	2.076	0.107	2.66	5093.8	261.3
1.188	XR-5-3	0.424	0.026	1.94	24914.7	1532.2
1.188	XR-5-4	0.907	0.026	0.96	11656.7	336.7
1.188	XR-5-5	2.207	0.055	1.39	4790.6	119.2
1.187	XR-6-1	0.228	0.006	8.15	46473.5	1203.7
1.187	XR-6-2	0.799	0.038	41.64	13249.9	631.8
1.187	XR-7-1	2.215	0.039	1.73	4777.7	84.1
1.187	XR-7-2	2.251	0.035	1.39	4700.8	72.2
1.188	XR-8-1	16.202	0.767	1.09	652.6	30.9
1.188	XR-8-2	7.663	0.224	1.46	1379.8	40.4
1.188	XR-8-3	12.657	0.769	1.99	835.4	50.8
1.187	XR-9-1	0.912	0.017	17.96	11612.0	210.2
1.187	XR-9-2	0.574	0.007	4.08	18446.9	225.0
1.187	XR-10-1	0.402	0.019	2.10	26327.1	1224.7
1.187	XR-11-1	1.613	0.025	10.04	6565.4	101.0
1.198	XR-11-2	0.275	0.009	25.16	38101.9	1204.1
1.198	XR-11-3	1.579	0.023	9.14	6645.2	96.4
1.188	XR-12-1	0.736	0.035	2.29	14358.7	686.3
1.188	XR-12-2	2.651	0.106	2.39	3988.8	159.9
1.188	XR-12-3	1.786	0.092	2.74	5922.0	304.1
1.188	XR-13-1	0.295	0.015	0.93	35794.8	1757.0
1.188	XR-13-2	2.072	0.222	1.03	5103.2	546.5
1.188	XR-13-3	0.175	0.009	0.92	60525.4	3256.7
1.188	XR-13-4	1.020	0.048	1.03	10363.4	484.5
1.188	XR-13-5	0.237	0.010	1.33	44577.5	1935.7
1.188	XR-13-6	0.809	0.024	1.38	13071.8	379.8
1.188	XR-13-7	0.266	0.010	1.49	39721.2	1522.0
1.188	XR-13-8	1.129	0.024	1.08	9366.4	199.1
1.188	XR-13-9	0.329	0.011	0.98	32148.9	1045.9
1.188	XR-14-1	1.030	0.064	1.54	10262.8	638.5
1.188	XR-14-2	1.315	0.088	0.94	8040.9	537.5
1.188	XR-14-3	0.889	0.082	1.30	11894.0	1099.8
1.188	XR-14-4	0.786	0.045	1.52	13456.1	777.4
1.188	XR-14-5	1.724	0.059	3.62	6134.0	210.3
1.188	XR-14-6	0.543	0.031	2.08	19472.9	1111.7

1.188	XR-14-7	1.195	0.038	1.42	8850.6	279.3
1.188	XR-15-1	1.732	0.056	1.57	6103.5	195.5
1.188	XR-15-2	0.196	0.015	2.04	54058.2	4256.1
1.188	XR-15-3	0.332	0.011	0.90	31896.8	1010.3
1.188	XR-15-4	0.105	0.025	2.47	100415.8	23840.4
1.198	XR-16-1	0.727	0.023	34.09	14430.4	462.6
1.198	XR-16-2	3.176	0.056	14.11	3302.9	57.7
1.187	XR-17-1	3.963	0.110	2.32	2670.6	74.1
1.187	XR-17-2	0.984	0.039	1.88	10755.6	421.9
1.645	XR-17-3	2.295	0.048	1.93	3329.2	69.6
1.188	XR-18-1	0.387	0.013	2.18	27315.4	945.6
1.188	XR-18-2	0.307	0.010	1.41	34476.0	1067.9
1.188	XR-18-3	0.539	0.017	1.97	19602.9	625.1
1.198	XR-19-1	0.310	0.006	10.72	33891.6	689.9
1.198	XR-19-2	1.479	0.035	17.70	7091.8	165.4
1.187	XR-19-3	1.197	0.032	28.04	8848.0	233.7
1.188	XR-20-1	0.570	0.019	1.64	18563.5	609.4
1.188	XR-20-2	0.895	0.036	1.98	11811.6	480.3
1.187	XR-21-1	0.575	0.005	1.74	18424.4	160.3
1.187	XR-21-2	0.305	0.006	7.54	34687.6	625.1
1.188	XR-22-1	0.279	0.010	1.29	37912.5	1373.0
1.188	XR-22-2	1.341	0.029	1.33	7886.8	167.7
1.188	XR-22-3	0.777	0.021	1.50	13617.2	368.3
1.188	XR-23-2	1.399	0.068	3.25	7558.1	366.8
1.188	XR-24-1	1.100	0.043	1.95	9614.3	373.3
1.188	XR-25-1	1.558	0.065	2.64	6785.5	284.8
1.188	XR-25-2	0.849	0.026	1.70	12452.9	376.9
1.187	XR-26-1	0.511	0.007	5.13	20713.5	287.7
1.187	XR-26-2	0.802	0.017	11.71	13206.9	281.7
1.188	XR-27-1	0.436	0.014	1.53	24268.5	774.2
1.188	XR-27-2	0.985	0.022	1.47	10732.6	242.9
1.188	XR-27-3	0.477	0.012	1.05	22158.0	538.6
1.188	XR-28-1	1.462	0.076	0.91	7230.9	377.3
1.188	XR-28-2	2.292	0.295	0.96	4612.9	594.3
1.188	XR-28-3	0.802	0.042	1.00	13179.3	696.5
1.188	XR-28-4	0.997	0.054	1.06	10608.8	578.0
1.188	XR-28-5	0.615	0.017	1.10	17184.8	474.8
1.188	XR-28-6	1.315	0.032	1.49	8043.3	196.4
1.188	XR-28-7	0.848	0.033	2.09	12475.0	478.3
1.188	XR-29-1	0.414	0.015	1.73	25534.4	931.1
1.188	XR-29-2	1.167	0.038	2.42	9058.3	295.7
1.188	XR-29-3	0.745	0.023	1.81	14189.2	439.8
1.188	XR-30-1	0.237	0.014	1.56	44690.5	2720.0
1.188	XR-30-2	0.743	0.129	1.30	14237.0	2467.1
1.188	XR-30-3	1.115	0.080	0.93	9484.1	683.9
1.188	XR-30-4	0.402	0.024	3.59	26276.8	1593.3
1.188	XR-30-5	0.800	0.026	1.85	13222.2	431.5
1.188	XR-30-6	0.387	0.016	2.29	27322.4	1143.7

169 Figure EA10. Estimation of Lorentzian widths from the signal at $Q = 1.2 \text{ \AA}^{-1}$. The data and
170 Lorentzian curve fit are represented by red circles and black curves, respectively. Sample
171 numbers are indicated.

176 Table EA5. Parameters as in Eq. 4 corresponding to best-fit structure for each XR experiment.

Sample Number ^a	Beamline	Scale Factor	Bragg order	d_{104} (Å)	σ d_{104}	φ	σ φ	z_0	σ z_0	w_0	σ w_0	Δ	σ Δ	χ^2	R	z (Å)
XR-2-1	13-IDC	0.10	1	3	-	0.09	0.005	3	-	0.001	-	2.14	0.06	2.84	0.06	2.14
XR-2-2	13-IDC	0.10	1	3	-	0.12	0.007	3	-	0.001	-	2.51	0.05	10.21	0.12	2.51
XR-3-1*	13-BMC	0.0013	1	3	-	0.17	0.020	3	-	0.001	-	2.68	0.06	4.63	0.17	2.68
XR-3-2*	13-BMC	0.0013	1	3	-	0.31	0.018	3	-	0.001	-	2.83	0.01	1.24	0.09	2.83
XR-3-3	13-BMC	0.0205	1	3	-	0.10	0.001	3	-	0.001	-	2.47	0.02	0.43	0.03	2.47
XR-3-4	13-BMC	0.0205	1	3	-	0.37	0.007	3	-	0.001	-	2.82	0.01	2.21	0.06	2.82
XR-3-5	13-BMC	0.0205	1	3	-	0.14	0.005	3	-	0.001	-	2.67	0.02	1.95	0.05	2.67
XR-3-6	13-BMC	0.0205	1	3	-	0.12	0.002	3	-	0.001	-	2.57	0.02	0.85	0.03	2.57
XR-3-7	13-BMC	0.0205	1	3	-	0.10	0.002	3	-	0.001	-	2.46	0.03	1.06	0.04	2.46
XR-4-1	13-IDC	0.10	1	3	-	0.12	0.010	3	-	0.001	-	1.81	0.03	1.87	0.05	1.81
XR-5-1	13-BMC	0.0205	1	3	-	0.37	0.006	3	-	0.001	-	2.85	0.00	1.49	0.05	2.85
XR-5-2	13-BMC	0.0205	1	3	-	0.40	0.005	3	-	0.001	-	2.83	0.00	1.19	0.04	2.83
XR-5-3	13-BMC	0.0205	1	3	-	0.36	0.007	3	-	0.001	-	2.85	0.00	1.51	0.05	2.85
XR-5-4	13-BMC	0.0205	1	3	-	0.35	0.007	3	-	0.001	-	2.85	0.00	1.26	0.05	2.85
XR-5-5	13-BMC	0.0205	1	3	-	0.10	0.002	3	-	0.001	-	2.41	0.04	1.41	0.04	2.41
XR-6-1	33-IDD	0.345	2	3	-	0.22	0.014	3	-	0.001	-	2.99	0.01	6.17	0.12	2.99
XR-6-2	33-IDD	0.345	2	3	-	1	-	2.80	0.02	3.46	0.09	2.93	0.00	2.34	0.06	4.08
XR-7-1	13-IDC	0.10	1	3	-	1	-	5.58	0.04	3.03	0.05	2.90	0.00	1.33	0.04	4.57
XR-7-2	13-IDC	0.10	1	3	-	1	-	5.87	0.04	2.99	0.05	2.94	0.00	2.02	0.05	4.67
XR-8-1	13-BMC	0.0170	1	3	-	0.46	0.013	3	-	0.001	-	2.90	0.01	5.64	0.09	2.90
XR-8-2	13-BMC	0.0170	1	3	-	0.50	0.005	3	-	0.001	-	2.87	0.01	0.98	0.04	2.87
XR-8-3	13-BMC	0.0170	1	3	-	0.46	0.007	3	-	0.001	-	2.89	0.01	1.53	0.05	2.89
XR-9-1	33-IDD	0.345	2	3	-	1	-	3.16	0.02	3.13	0.06	2.93	0.00	1.49	0.04	3.97
XR-9-2	33-IDD	0.345	2	3	-	1	-	2.40	0.10	8.67	0.31	2.82	0.01	6.18	0.09	7.07
XR-10-1	13-IDC	0.0297	1	2.997	0.003	1	-	59.72	5.20	46.23	5.96	2.52	0.02	42.12	0.24	46.12
XR-11-1	33-IDD	0.345	2	2.995	0.002	1	-	5.96	0.05	4.66	0.07	2.92	0.01	1.69	0.04	5.61
XR-11-2	33-IDD	0.345	2	3	-	0.21	0.020	3	-	0.001	-	3.00	0.00	6.89	0.12	3.00
XR-11-3	33-IDD	0.345	2	2.987	0.002	1	-	6.43	0.05	4.68	0.07	2.93	0.00	1.67	0.04	5.76
XR-12-1	13-BMC	0.0205	1	3	-	0.41	0.005	3	-	0.001	-	2.85	0.00	1.39	0.04	2.85
XR-12-2	13-BMC	0.0205	1	3	-	0.42	0.003	3	-	0.001	-	2.84	0.00	0.62	0.03	2.84
XR-12-3	13-BMC	0.0205	1	3	-	0.42	0.004	3	-	0.001	-	2.87	0.00	1.20	0.04	2.87
XR-13-1*	13-BMC	0.0013	1	3	-	1	-	5.19	0.28	4.14	0.28	2.87	0.03	8.86	0.15	5.05
XR-13-2*	13-BMC	0.0013	1	2.984	0.016	1	-	25.95	1.46	16.08	3.76	2.67	0.05	11.64	0.30	18.39
XR-13-3*	13-BMC	0.0013	1	3	-	0.46	0.010	3	-	0.001	-	2.85	0.01	1.63	0.09	2.85
XR-13-4*	13-BMC	0.0013	1	3.011	0.006	1	-	11.25	0.48	10.41	0.48	3.48	0.02	4.43	0.12	10.91
XR-13-5	13-BMC	0.0205	2	3	-	1	-	0.00	0.48	11.67	0.74	2.78	0.01	5.32	0.14	8.39
XR-13-6	13-BMC	0.0205	2	3.000	0.007	1	-	11.23	0.28	7.26	0.89	2.80	0.03	6.94	0.20	8.66

XR-13-7	13-BMC	0.0205	2	3	-	1	-	0.00	0.32	12.52	0.53	2.76	0.02	5.67	0.17	8.90
XR-13-8	13-BMC	0.0205	2	3.007	0.001	1	-	21.42	0.38	14.12	0.60	2.78	0.01	2.99	0.15	15.83
XR-13-9	13-BMC	0.0205	2	2.998	0.007	1	-	9.72	0.22	6.79	0.72	2.80	0.03	5.40	0.16	7.90
XR-14-1	13-BMC	0.0027	1	3	-	1	-	3.42	0.15	3.18	0.23	2.89	0.01	3.78	0.10	4.03
XR-14-2	13-BMC	0.0013	1	3	-	0.46	0.009	3	-	0.001	-	2.91	0.01	1.69	0.10	2.91
XR-14-3	13-BMC	0.0017	1	3	-	1	-	3.92	0.15	4.01	0.19	2.89	0.01	2.47	0.11	4.64
XR-14-4	13-BMC	0.0025	1	3	-	1	-	3.47	0.09	2.95	0.15	2.91	0.01	1.34	0.07	3.92
XR-14-5	13-BMC	0.0350	2	2.996	0.005	1	-	3.30	0.44	11.49	0.61	2.79	0.01	1.07	0.08	8.99
XR-14-6	13-BMC	0.0350	2	3.016	0.004	1	-	4.58	0.34	12.76	0.55	2.81	0.01	1.52	0.08	10.09
XR-14-7	13-BMC	0.0350	2	3.018	0.005	1	-	1.75	0.44	11.74	0.52	2.83	0.01	1.02	0.07	8.83
XR-15-1	13-BMC	0.0205	1	3	-	0.38	0.003	3	-	0.001	-	2.81	0.00	0.61	0.03	2.81
XR-15-2	13-BMC	0.0205	1	3	-	0.40	0.006	3	-	0.001	-	2.84	0.01	2.15	0.06	2.84
XR-15-3	13-BMC	0.0205	1	3	-	0.36	0.003	3	-	0.001	-	2.81	0.00	0.49	0.03	2.81
XR-15-4	13-BMC	0.0205	1	3	-	0.41	0.003	3	-	0.001	-	2.82	0.00	0.73	0.03	2.82
XR-16-1	33-IDD	0.345	2	3	-	1	-	3.00	0.04	3.49	0.15	2.91	0.01	4.44	0.07	4.13
XR-16-2	33-IDD	0.345	2	2.993	0.002	1	-	8.12	0.07	4.99	0.08	2.91	0.01	2.47	0.05	6.47
XR-17-1	13-IDC	0.10	1	2.992	0.003	1	-	15.88	0.59	8.65	0.27	3.09	0.02	34.18	0.24	11.22
XR-17-2	13-IDC	0.10	1	3.020	0.021	1	-	16.68	1.19	13.66	2.61	2.56	0.04	25.94	0.19	13.83
XR-17-3	13-IDC	0.10	1	3.006	0.003	1	-	13.31	0.40	9.47	0.24	3.33	0.01	11.87	0.13	10.92
XR-18-1 ⁺	13-BMC	0.0205	2	3.008	0.003	1	-	16.19	0.55	14.72	1.37	2.71	0.02	10.56	0.16	14.39
XR-18-2 ⁺	13-BMC	0.0205	2	3.003	0.006	1	-	11.96	0.53	13.00	1.65	2.72	0.02	11.58	0.19	12.11
XR-18-3 ⁺	13-BMC	0.0205	2	3.008	0.003	1	-	20.02	0.75	14.64	1.34	2.73	0.02	13.20	0.20	15.59
XR-19-1	33-IDD	0.345	2	2.999	0.003	1	-	14.31	0.22	11.73	0.59	2.77	0.00	6.48	0.07	12.14
XR-19-2	33-IDD	0.345	2	3.000	0.001	1	-	25.20	0.45	17.94	0.65	2.80	0.00	6.17	0.09	19.19
XR-19-3	33-IDD	0.345	2	3.005	0.002	1	-	18.76	0.49	16.27	0.67	2.82	0.00	7.72	0.09	16.16
XR-20-1	13-BMC	0.0205	2	3.011	0.004	1	-	12.79	0.83	17.50	1.47	2.77	0.02	11.23	0.34	15.09
XR-20-2	13-BMC	0.0205	2	3.014	0.004	1	-	11.84	0.71	16.41	1.26	2.81	0.02	9.20	0.32	14.19
XR-21-1	33-IDD	0.345	2	3	-	1	-	5.15	0.06	4.33	0.10	2.91	0.00	3.97	0.07	5.17
XR-21-2	33-IDD	0.345	2	3	-	1	-	3.99	0.06	3.69	0.13	2.92	0.01	8.12	0.10	4.49
XR-22-1	13-BMC	0.0205	2	3	-	1	-	4.37	0.06	4.58	0.19	2.88	0.01	2.19	0.10	5.08
XR-22-2	13-BMC	0.0205	2	3.000	0.004	1	-	8.94	0.14	6.41	0.39	2.82	0.02	3.02	0.12	7.45
XR-22-3	13-BMC	0.0205	2	2.991	0.004	1	-	7.81	0.13	5.27	0.23	2.89	0.02	2.77	0.10	6.50
XR-23-1	13-IDC	0.0234	1	3.010	0.007	1	-	31.15	3.12	14.39	1.86	2.72	0.02	72.96	0.37	19.58
XR-24-1	13-BMC	0.0036	2	3.009	0.004	1	-	24.58	1.89	25.43	3.14	2.75	0.05	6.84	1.74	23.21
XR-25-1	13-BMC	0.0205	2	3.009	0.002	1	-	18.62	0.54	17.17	1.00	2.77	0.01	5.77	0.20	16.59
XR-25-2	13-BMC	0.0205	2	3.005	0.003	1	-	20.13	0.56	11.68	1.00	2.74	0.01	7.65	0.10	14.08
XR-26-1	33-IDD	0.345	2	3	-	1	-	4.54	0.11	7.05	0.38	2.84	0.01	13.75	0.13	6.59
XR-26-2	33-IDD	0.345	2	3	-	1	-	5.65	0.17	5.79	0.17	2.88	0.01	5.76	0.09	6.15
XR-27-1	13-BMC	0.0205	2	2.988	0.014	1	-	5.46	0.09	3.02	0.22	2.94	0.02	3.81	0.10	4.55
XR-27-2	13-BMC	0.0205	2	3.006	0.004	1	-	9.17	0.21	3.88	0.17	2.93	0.03	5.38	0.10	6.29

XR-27-3	13-BMC	0.0205	1	3	-	1	-	5.32	0.12	3.28	0.13	2.88	0.01	6.15	0.09	4.60
XR-28-1*	13-BMC	0.0013	1	2.995	0.010	1	-	32.11	1.64	16.20	2.90	2.61	0.02	15.97	0.30	20.72
XR-28-2*	13-BMC	0.0013	1	2.988	0.013	1	-	28.38	1.53	15.96	3.21	2.69	0.03	14.73	0.31	19.23
XR-28-3*	13-BMC	0.0013	1	3.008	0.010	1	-	26.33	0.97	13.28	2.07	2.56	0.02	9.78	0.16	17.08
XR-28-4*	13-BMC	0.0013	1	3.008	0.008	1	-	29.01	1.00	13.19	1.75	2.56	0.02	11.13	0.15	18.10
XR-28-5	13-BMC	0.0205	2	3.010	0.002	1	-	24.74	0.66	13.51	0.85	2.77	0.02	6.37	0.20	16.71
XR-28-6	13-BMC	0.0205	2	3.008	0.001	1	-	25.32	0.49	11.77	0.60	2.77	0.01	3.68	0.12	16.12
XR-28-7	13-BMC	0.0205	2	3.008	0.002	1	-	21.93	0.79	19.24	1.26	2.79	0.02	9.16	0.26	18.83
XR-29-1	13-BMC	0.0205	2	3.010	0.003	1	-	18.79	0.56	11.96	0.92	2.77	0.02	8.50	0.16	13.76
XR-29-2	13-BMC	0.0205	2	3.010	0.001	1	-	21.47	0.66	21.53	1.00	2.81	0.01	5.78	0.20	20.03
XR-29-3	13-BMC	0.0205	2	3.010	0.002	1	-	16.52	0.79	23.77	1.53	2.79	0.02	10.53	0.36	19.92
XR-30-1*	13-BMC	0.0013	1	3.003	0.005	1	-	35.02	1.11	17.71	1.81	2.61	0.02	6.49	0.13	22.55
XR-30-2*	13-BMC	0.0013	1	3	-	1	-	20.37	1.46	28.64	3.00	2.75	0.04	19.82	1.53	23.91
XR-30-3*	13-BMC	0.0013	1	2.988	0.008	1	-	33.23	1.96	20.15	2.87	2.71	0.03	14.48	0.30	23.13
XR-30-4	13-BMC	0.0205	2	3.010	0.001	1	-	30.27	0.85	19.87	0.97	2.80	0.01	5.75	0.01	21.98
XR-30-5	13-BMC	0.0205	2	3.012	0.002	1	-	19.33	0.68	21.62	1.23	2.81	0.01	6.75	0.17	19.45
XR-30-6	13-BMC	0.0205	2	3.008	0.003	1	-	14.56	0.92	22.94	2.00	2.76	0.02	13.88	0.35	18.87

177 ^a sample numbers with additional * or ⁺ have corresponding Cd measurements from XRF. “Asterisks” denote measurements at 28.6 keV, and
178 “plus” symbols denote beam energy of 14.9 keV.

Sample number ^a	Domain size ^b × 10 ⁴ (Å)	Cd occupancy (σ) ^c (ML)	Growth rate ^d × 10 ⁻² (Cd/Å ² /h)	Growth rate ^e × 10 ² (Cd/Å/h)
XR-2-1	2.921 (0.328)	0.09 (0.00)	0.004 (2.0E-6)	0.012 (0.15)
XR-2-2	0.180 (0.008)	0.12 (0.01)	0.005 (3.0E-6)	0.001 (0.01)
XR-3-1*	3.094 (0.543)	0.17 (0.02)	0.014 (1.7E-5)	0.044 (0.94)
XR-3-2*	5.395 (0.828)	0.31 (0.02)	0.027 (1.5E-5)	0.144 (2.36)
XR-3-3	1.732 (0.052)	0.10 (0.00)	0.008 (1.1E-6)	0.014 (0.05)
XR-3-4	3.872 (0.172)	0.37 (0.01)	0.032 (6.0E-6)	0.124 (0.60)
XR-3-5	0.355 (0.014)	0.14 (0.01)	0.012 (4.4E-6)	0.004 (0.02)
XR-3-6	0.494 (0.021)	0.12 (0.00)	0.010 (2.0E-6)	0.005 (0.02)
XR-3-7	0.501 (0.012)	0.10 (0.00)	0.009 (1.5E-6)	0.004 (0.01)
XR-4-1	5.096 (0.174)	0.12 (0.01)	0.005 (4.1E-6)	0.026 (0.23)
XR-5-1	2.884 (0.109)	0.37 (0.01)	0.028 (4.8E-6)	0.081 (0.33)
XR-5-2	0.509 (0.026)	0.40 (0.01)	0.030 (3.8E-6)	0.015 (0.08)
XR-5-3	2.491 (0.153)	0.36 (0.01)	0.027 (5.2E-6)	0.067 (0.43)
XR-5-4	1.166 (0.034)	0.35 (0.01)	0.026 (5.2E-6)	0.031 (0.11)
XR-5-5	0.479 (0.012)	0.10 (0.00)	0.007 (1.5E-6)	0.003 (0.01)
XR-6-1	4.647 (0.120)	0.22 (0.01)	0.012 (7.3E-6)	0.054 (0.37)
XR-6-2	1.325 (0.063)	0.71 (0.01)	0.038 (7.1E-6)	0.051 (0.26)
XR-7-1	0.478 (0.008)	1.43 (0.02)	0.061 (6.4E-6)	0.029 (0.06)
XR-7-2	0.470 (0.007)	1.50 (0.02)	0.064 (7.7E-6)	0.030 (0.06)
XR-8-1	0.065 (0.003)	0.46 (0.01)	0.034 (9.6E-6)	0.002 (0.01)
XR-8-2	0.138 (0.004)	0.50 (0.01)	0.038 (4.1E-6)	0.005 (0.02)
XR-8-3	0.084 (0.005)	0.46 (0.01)	0.035 (5.0E-6)	0.003 (0.02)
XR-9-1	1.161 (0.021)	0.75 (0.01)	0.041 (5.0E-6)	0.047 (0.10)
XR-9-2	1.845 (0.023)	1.34 (0.03)	0.072 (1.8E-5)	0.133 (0.36)
XR-10-1	2.633 (0.122)	20.33 (1.77)	0.880 (7.6E-4)	2.316 (22.81)
XR-11-1	0.657 (0.010)	1.64 (0.02)	0.369 (3.9E-5)	0.242 (0.45)
XR-11-2	3.810 (0.120)	0.21 (0.02)	0.048 (4.1E-5)	0.182 (1.68)
XR-11-3	0.665 (0.010)	1.78 (0.02)	0.400 (3.8E-5)	0.266 (0.46)
XR-12-1	1.436 (0.069)	0.41 (0.01)	0.486 (6.0E-5)	0.698 (3.45)
XR-12-2	0.399 (0.016)	0.42 (0.00)	0.502 (3.6E-5)	0.200 (0.82)
XR-12-3	0.592 (0.030)	0.42 (0.00)	0.495 (5.1E-5)	0.293 (1.54)
XR-13-1*	3.579 (0.176)	1.40 (0.11)	0.645 (4.9E-4)	2.310 (20.84)
XR-13-2*	0.510 (0.055)	8.45 (0.40)	3.900 (1.9E-3)	1.990 (23.34)
XR-13-3*	6.053 (0.326)	0.46 (0.01)	0.211 (4.4E-5)	1.278 (7.38)
XR-13-4*	1.036 (0.048)	3.43 (0.13)	1.584 (6.0E-4)	1.641 (9.88)
XR-13-5	4.458 (0.194)	1.34 (0.04)	0.620 (2.0E-4)	2.765 (14.89)
XR-13-6	1.307 (0.038)	3.40 (0.11)	1.570 (5.0E-4)	2.053 (8.84)
XR-13-7	3.972 (0.152)	1.46 (0.04)	0.673 (1.9E-4)	2.675 (12.75)
XR-13-8	0.937 (0.020)	6.86 (0.13)	3.167 (6.0E-4)	2.966 (8.45)
XR-13-9	3.215 (0.105)	2.92 (0.09)	1.349 (4.2E-4)	4.336 (19.61)
XR-14-1*	1.026 (0.064)	0.83 (0.06)	1.349 (1.0E-3)	1.384 (13.75)
XR-14-2*	0.804 (0.054)	0.46 (0.01)	0.748 (1.5E-4)	0.602 (4.21)

XR-14-3*	1.189 (0.110)	1.05 (0.06)	1.698 (9.8E-4)	2.020 (22.02)
XR-14-4*	1.346 (0.078)	0.81 (0.04)	1.322 (6.7E-4)	1.779 (13.67)
XR-14-5	0.613 (0.021)	1.89 (0.03)	3.064 (5.5E-4)	1.879 (7.26)
XR-14-6	1.947 (0.111)	2.28 (0.04)	3.709 (6.2E-4)	7.222 (42.94)
XR-14-7	0.885 (0.028)	1.62 (0.03)	2.630 (5.3E-4)	2.328 (8.74)
XR-15-1	0.610 (0.020)	0.38 (0.00)	0.144 (1.3E-5)	0.088 (0.29)
XR-15-2	5.406 (0.426)	0.40 (0.01)	0.151 (2.4E-5)	0.815 (6.55)
XR-15-3	3.190 (0.101)	0.36 (0.00)	0.138 (1.3E-5)	0.440 (1.45)
XR-15-4	10.042 (2.384)	0.41 (0.00)	0.154 (1.3E-5)	1.549 (36.80)
XR-16-1	1.443 (0.046)	0.76 (0.02)	1.029 (2.8E-4)	1.485 (6.25)
XR-16-2	0.330 (0.006)	2.31 (0.02)	3.110 (3.3E-4)	1.027 (2.11)
XR-17-1	0.267 (0.007)	4.83 (0.19)	2.172 (8.7E-4)	0.580 (2.82)
XR-17-2	1.076 (0.042)	5.46 (0.24)	2.453 (1.1E-3)	2.639 (15.45)
XR-17-3	0.333 (0.007)	3.99 (0.12)	1.794 (5.6E-4)	0.597 (2.25)
XR-18-1 ⁺	2.732 (0.095)	5.37 (0.20)	2.400 (9.1E-4)	6.557 (33.73)
XR-18-2 ⁺	3.448 (0.107)	4.07 (0.17)	1.819 (7.5E-4)	6.272 (32.40)
XR-18-3 ⁺	1.960 (0.063)	6.48 (0.27)	2.894 (1.2E-3)	5.673 (29.57)
XR-19-1	3.389 (0.069)	4.61 (0.08)	1.036 (1.8E-4)	3.510 (9.33)
XR-19-2	0.709 (0.017)	8.22 (0.16)	1.848 (3.5E-4)	1.310 (3.95)
XR-19-3	0.885 (0.023)	6.20 (0.17)	1.393 (3.9E-4)	1.232 (4.71)
XR-20-1	1.856 (0.061)	4.72 (0.24)	0.547 (2.8E-4)	1.015 (6.17)
XR-20-2	1.181 (0.048)	4.35 (0.21)	0.505 (2.4E-4)	0.596 (3.74)
XR-21-1	1.842 (0.016)	1.39 (0.02)	7.516 (1.2E-3)	13.847 (25.92)
XR-21-2	3.469 (0.063)	1.02 (0.03)	5.518 (1.4E-3)	19.139 (59.32)
XR-22-1	3.791 (0.137)	1.22 (0.03)	2.032 (5.0E-4)	7.705 (33.69)
XR-22-2	0.789 (0.017)	2.66 (0.06)	4.409 (9.4E-4)	3.477 (10.46)
XR-22-3	1.362 (0.037)	2.24 (0.05)	3.710 (8.7E-4)	5.053 (18.11)
XR-23-1	0.756 (0.037)	9.98 (1.03)	17.939 (1.8E-2)	13.559 (154.4)
XR-24-1	0.961 (0.037)	8.58 (0.49)	13.807 (7.9E-3)	13.274 (91.74)
XR-25-1	0.679 (0.028)	6.23 (0.19)	2.872 (9.0E-4)	1.949 (10.20)
XR-25-2	1.245 (0.038)	6.37 (0.19)	2.937 (9.0E-4)	3.658 (15.74)
XR-26-1	2.071 (0.029)	1.56 (0.06)	8.404 (3.1E-3)	17.407 (68.22)
XR-26-2	1.321 (0.028)	1.67 (0.04)	9.030 (2.0E-3)	11.926 (36.43)
XR-27-1	2.427 (0.077)	1.38 (0.04)	2.362 (6.8E-4)	5.732 (24.60)
XR-27-2	1.073 (0.024)	2.59 (0.08)	4.429 (1.3E-3)	4.754 (17.67)
XR-27-3	2.216 (0.054)	1.37 (0.05)	2.342 (7.9E-4)	5.189 (21.59)
XR-28-1*	0.723 (0.038)	10.41 (0.54)	18.226 (9.4E-3)	13.179 (96.93)
XR-28-2*	0.461 (0.059)	9.19 (0.48)	16.100 (8.4E-3)	7.427 (103.16)
XR-28-3*	1.318 (0.070)	8.45 (0.31)	14.795 (5.5E-3)	19.499 (125.8)
XR-28-4*	1.061 (0.058)	9.32 (0.33)	16.318 (5.8E-3)	17.311 (112.7)
XR-28-5	1.718 (0.047)	7.88 (0.22)	13.789 (3.9E-3)	23.697 (93.64)
XR-28-6	0.804 (0.020)	8.03 (0.16)	14.053 (2.9E-3)	11.303 (35.96)
XR-28-7	1.247 (0.048)	7.32 (0.27)	12.826 (4.7E-3)	16.000 (85.28)
XR-29-1	2.553 (0.093)	5.95 (0.20)	10.415 (3.4E-3)	26.594 (130.8)
XR-29-2	0.906 (0.030)	7.37 (0.23)	12.905 (4.0E-3)	11.690 (52.61)
XR-29-3	1.419 (0.044)	6.30 (0.28)	11.039 (4.8E-3)	15.664 (84.06)

XR-30-1*	4.469 (0.272)	11.36 (0.37)	20.212 (6.5E-3)	90.328 (622.1)
XR-30-2*	1.424 (0.247)	7.81 (0.62)	13.907 (1.1E-2)	19.799 (377.2)
XR-30-3*	0.948 (0.068)	10.88 (0.65)	19.361 (1.1E-2)	18.362 (171.5)
XR-30-4	2.628 (0.159)	9.84 (0.28)	17.513 (5.0E-3)	46.019 (308.8)
XR-30-5	1.322 (0.043)	6.80 (0.24)	12.098 (4.2E-3)	15.996 (76.14)
XR-30-6	2.732 (0.114)	5.75 (0.30)	10.232 (5.3E-3)	27.957 (186.6)

^a sample numbers with * or + have corresponding Cd measurements from XRF. “Asterisks” denote measurements at 28.6 keV, and “plus” symbols denote beam energy of 14.9 keV.

^b substrate domain size or step spacing, and its uncertainty.

^c total Cd occupancies from analysis of XR data, and its uncertainty.

^d growth rate obtained from XR data, and its uncertainty.

^e growth rate obtained from XR data, normalized with step-spacing (^b), and its uncertainty.

Figure EA11. Change in growth rate of otavite on dolomite in (A) $\text{Cd}/\text{\AA}^2/\text{h}$ and (B) $\text{Cd}/\text{\AA}/\text{h}$ with length of reaction for experiments with $\log \beta_{\max} \sim 1.5$. The data are fit with a line $y = ax + b$. The parameters for the best-fit line (blue lines) are: (A) $a = -4.3 \times 10^{-4}$, $b = 0.03$, $R^2 = 0.52$; and (B) $a = -4.66$, $b = 332$, $R^2 = 0.08$.

193 Table EA7. Comparison of cadmium occupancies from XR and XRF

Sample number ^a	XR Cd occupancy ^b ML	XRF Cd occupancy ^c ML
XR-3_1	0.17 (0.02)	0.176 (0.058)
XR-3_2	0.31 (0.02)	0.195 (0.057)
XR-13_1	1.40 (0.11)	2.821 (0.149)
XR-13_2	8.45 (0.40)	16.313 (0.186)
XR-13_3	0.46 (0.01)	1.049 (0.121)
XR-13_4	3.43 (0.13)	8.051 (0.123)
XR-14_1	0.83 (0.06)	1.495 (0.074)
XR-14_2	0.46 (0.01)	1.375 (0.085)
XR-14_3	1.05 (0.06)	1.516 (0.079)
XR-14_4	0.81 (0.04)	1.934 (0.060)
XR-18_1	5.37 (0.20)	6.544 (0.146)
XR-18_2	4.07 (0.17)	3.757 (0.187)
XR-18_3	6.48 (0.27)	8.660 (0.149)
XR-28_1	10.41 (0.54)	35.053 (0.233)
XR-28_2	9.19 (0.48)	32.454 (0.228)
XR-28_3	8.45 (0.31)	31.841 (0.158)
XR-28_4	9.32 (0.33)	38.361 (0.239)
XR-30_1	11.36 (0.37)	29.600 (0.156)
XR-30_2	7.81 (0.62)	53.363 (0.189)
XR-30_3	10.88 (0.65)	40.022 (0.200)

194 ^a experiment numbers are as in Table.195 ^b total Cd occupancies from analysis of XR data, and its uncertainty.196 ^c total Cd occupancies from analysis of XRF data, and its uncertainty.

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200 Figure EA12. H - K sections at fixed L values of (a) $L = 2.92$; (b) $L = 3.34$; and (c) $L = 1.94$ for
 201 growth of otavite on dolomite (Sample XR-23-1), showing the presence of minor powder-like
 202 otavite growth for the (a) $[0\ 1\ 3]$, (b) $[2\ 1\ 3.44]$, and (c) $[0\ 2\ 2]$ reflections. This phase has the
 203 same d_{104} spacing as the otavite phase that is epitaxially grown on dolomite, but is oriented
 204 differently, e.g., tilted. The signal peak intensity from the incoherent otavite is about 3 order of

205 magnitudes smaller than that for the coherent otavite at H , K values of (a) 0, 1; (b) 2, 1; and (c) 0,

206 2. Reciprocal lattice units H , K , and L are related to momentum transfer, Q , by: $Q =$

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$$[Q_x, Q_y, Q_z] = \left[\left(\frac{2\pi}{a} \right) H, \left(\frac{2\pi}{b} \right) K, \left(\frac{2\pi}{c} \right) L \right]$$

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Figure EA13. (A) Change in $CdCO_3$ saturation index with moles of $CdCO_3$ removed from 30 mL solution, as in dolomite experiments. The initial $[Cd]$, $[Ca]$, and $[CO_3]$ in mM are indicated in the legend. (B) Evolution of $CdCO_3$ (blue curves) and $CaCO_3$ (orange curves) saturation indices with the addition of $CaCO_3$ from the dissolving calcite substrate in water, as in calcite experiments. Initial $[Cd]$ in solutions containing $CdCl_2$ are indicated in the legend.

Figure EA14. *In situ* AFM images of tall islands on the dolomite surface. Elapsed times after exchanging the $[\text{Cd}] = 0.052$, $[\text{Ca}] = [\text{CO}_3] = 0$ solution with DIW are indicated. The islands have a flat top and a height of ~ 2.5 nm, similar to those described by Perez-Garrido et al. (2007) and Xu et al. (2014).

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 228 Figure EA15. Comparison of measured reflectivity data (red circles) from a dolomite sample
 229 (shown in Fig. EA14) with the calculated reflectivity signal assuming that the precipitates are
 230 oriented CdCO_3 islands. (A) Intensities were calculated for a bare dolomite substrate (blue
 231 curve), a free-standing CdCO_3 film (8 layers with d-spacing of $2.95 \text{ \AA}$, and a surface coverage of
 232 20%) (pink curve), and the total intensity that arises from the substrate/film combination (black
 233 curve). (B) Same calculations from (A) are shown in comparison to the XR data (red circles).

234 The data lack of any film Bragg peaks and intensity oscillations predicted by the calculation
235 suggest that the film was not due to a crystalline oriented otavite film.