

## SUPPLEMENTARY SECTION

### I. DRYING EVOLUTION

The measured and the fitting parameters of drying protein droplets (BSA+DI and Lys+DI) are tabulated.

TABLE T1. Data from  $\theta(t)$  vs.  $t$  graph of drying droplet at each  $\phi$  (initial protein concentration in wt%) for BSA+DI with fitting parameters:  $\theta_0$  (contact angle at  $t = 0$  in degrees),  $1/\tau$  (characteristic rate in  $s^{-1}$ ), and  $R^2$  (adjusted R-square of the fit).

$\phi$ (wt%)	$\theta_0$ (°)	$1/\tau \times 10^{-3}$ ( $s^{-1}$ )	$R^2$
1	$31.43 \pm 0.04$	$1.560 \pm 0.004$	0.998
5	$39.96 \pm 0.08$	$1.500 \pm 0.005$	0.995
9	$39.10 \pm 0.07$	$1.510 \pm 0.005$	0.996
13	$42.90 \pm 0.07$	$1.540 \pm 0.004$	0.997

TABLE T2. Data from  $\bar{r}(t)$  vs.  $t$  graph of drying droplet at each  $\phi$  (initial protein concentration in wt%) for BSA+DI with measured parameters:  $R$  (radius of droplet in mm),  $w$  (rim width in mm),  $t_d$  (time in seconds after which the contact line radius begins to shrink from droplet radius),  $t_s$  (time in seconds at which two linear fits merge); and fitting parameters:  $m_1$  (slope of first linear fit in  $\mu\text{m/s}$ ),  $R_1^2$  (adjusted R-square of first linear fit),  $m_2$  (slope of second linear fit in  $\mu\text{m/s}$ ),  $R_2^2$  (adjusted R-square of the second linear fit). The negative sign in the slope values corresponds to the decrease in the radius of the contact line.

$\phi$ (wt%)	$R$ (mm)	$w$ (mm)	$t_d \pm 30$ (s)	$t_s \pm 20$ (s)	$m_1$ ( $\mu\text{m/s}$ )	$R_1^2$	$m_2$ ( $\mu\text{m/s}$ )	$R_2^2$
1	$1.265 \pm 0.025$	$0.3194 \pm 0.043$	254	<i>n/a</i>	-1.2	0.9604	<i>n/a</i>	<i>n/a</i>
3	$1.129 \pm 0.025$	$0.292 \pm 0.072$	307	<i>n/a</i>	-0.9	0.980	<i>n/a</i>	<i>n/a</i>
5	$1.215 \pm 0.028$	$0.507 \pm 0.051$	186	470	-0.8	0.979	-2.9	0.978
7	$1.135 \pm 0.036$	$0.465 \pm 0.071$	312	524	-0.7	0.983	-2.8	0.967
9	$0.814 \pm 0.017$	$0.378 \pm 0.029$	224	429	-1.2	0.977	-3.4	0.993
11	$1.08 \pm 0.016$	$0.543 \pm 0.022$	225	503	-1.0	0.996	-2.6	0.967
13	$0.854 \pm 0.014$	$0.462 \pm 0.021$	235	477	-1.1	0.983	-2.4	0.970

TABLE T3. Data from  $\theta(t)$  vs.  $t$  graph of drying droplet at each  $\phi$  (initial protein concentration in wt%) for Lys+DI with fitting parameters:  $\theta_0$  (contact angle at  $t = 0$  in degrees),  $1/\tau$  (characteristic rate in  $s^{-1}$ ), and  $R^2$  (adjusted R-square of the fit).

$\phi$ (wt%)	$\theta_0$ ( $^\circ$ )	$1/\tau \times 10^{-3}$ ( $s^{-1}$ )	$R^2$
1	$43.83 \pm 0.15$	$1.500 \pm 0.008$	0.987
5	$36.64 \pm 0.01$	$0.900 \pm 0.001$	0.999
9	$41.89 \pm 0.10$	$1.400 \pm 0.005$	0.994
13	$40.38 \pm 0.08$	$1.140 \pm 0.004$	0.994

TABLE T4. Data from  $\bar{r}(t)$  vs.  $t$  graph of drying droplet at each  $\phi$  (initial protein concentration in wt%) for Lys+DI with measured parameters:  $R$  (radius of droplet in mm),  $w$  (rim width in mm),  $t_d$  (time in seconds after which the contact line radius begins to shrink from droplet radius),  $t_s$  (time in seconds at which two linear fits merge); and fitting parameters:  $m_1$  (slope of first linear fit in  $\mu\text{m/s}$ ),  $R_1^2$  (adjusted R-square of first linear fit),  $m_2$  (slope of second linear fit in  $\mu\text{m/s}$ ),  $R_2^2$  (adjusted R-square of second linear fit). The negative sign in the slope values corresponds to the decrease in the radius of the contact line.

$\phi$ (wt%)	$R$ (mm)	$w$ (mm)	$t_d \pm 30$ (s)	$t_s \pm 20$ (s)	$m_1$ ( $\mu\text{m/s}$ )	$R_1^2$	$m_2$ ( $\mu\text{m/s}$ )	$R_2^2$
1	$0.803 \pm 0.017$	$0.195 \pm 0.037$	395	459	-1.0	0.987	-17.2	0.974
3	$0.895 \pm 0.014$	$0.320 \pm 0.043$	492	655	-1.0	0.973	-14.2	0.971
5	$0.919 \pm 0.020$	$0.383 \pm 0.033$	320	569	-1.0	0.974	-11.7	0.987
7	$1.213 \pm 0.028$	$0.411 \pm 0.073$	427	642	-0.9	0.978	-12.0	0.999
9	$1.149 \pm 0.034$	$0.491 \pm 0.048$	199	490	-0.9	0.941	-11.8	0.989
11	$1.095 \pm 0.016$	$0.481 \pm 0.075$	213	526	-1.1	0.955	-11.3	0.964
13	$1.153 \pm 0.016$	$0.438 \pm 0.061$	303	639	-1.1	0.948	-8.4	0.985

## II. STATISTICAL ANALYSIS

In this section, the outcome of Mann-Whitney U test with the parameters U, z, p are tabulated. An asterisk [\*] indicates that the interaction between the respective pairs (between BSA and Lys droplets) are significantly different at different initial protein concentration  $\phi$ . The difference will be said statistically significant in terms of  $x_c$ , if p (calculated prob-

ability) is less than  $\alpha$  (level of significance) being 0.05. This tells that the crack spacing is significantly different for BSA and Lys at every concentration  $\phi$ .

TABLE T5. Outcome of Mann-Whitney U test with an asterisk [\*] indicating significant interaction between Lys, and BSA in terms of  $x_c$  at different  $\phi$  (wt %).

$\phi$ (wt%)	U	z	p
7	2893.500	-2.119	0.034*
9	2666.500	-5.331	$\leq 0.001^*$
11	1732.500	-5.752	$\leq 0.001^*$
13	1643.500	-5.439	$\leq 0.001^*$

### III. SPIRAL CRACKS

In this section, the spirals which are found to be prominently present in the layer of lysozyme droplets at  $\phi$  of 13 wt% are shown. The fitting parameters are tabulated for each spiral. The different spirals are focused by using  $50\times$  objective lens. There is no preference of clockwise or counter-clockwise found in each droplet, hence, we have generalized the direction of the spiral by flipping the spiral images so that the spirals become consistent every time with the starting spiral revolution line.

TABLE T6. Characteristic fitting parameters of spiral cracks at  $\phi$  of 11, and 13 wt% for  $\ln s(\theta) = \ln a + b\theta$  are tabulated: “b” denotes the spiral tightness in  $\mu\text{m}/\text{rad}$ , and “a” denotes apparent length in  $\mu\text{m}$ .  $s(\theta)$  is the distance from the spiral center, and  $\theta$  is the angle which is not restricted to  $2\pi$ .

$\phi$ (wt%)	Spiral no	Spiral parameters		$R^2$
		b ( $\mu\text{m}/\text{rad}$ )	a ( $\mu\text{m}$ )	
11	1	0.0548	13.1800	0.9460
13	1	0.0398	12.3407	0.9002
13	2	0.0437	13.3631	0.9373
13	3	0.0359	14.8322	0.9521
13	4	0.0376	16.7049	0.9569

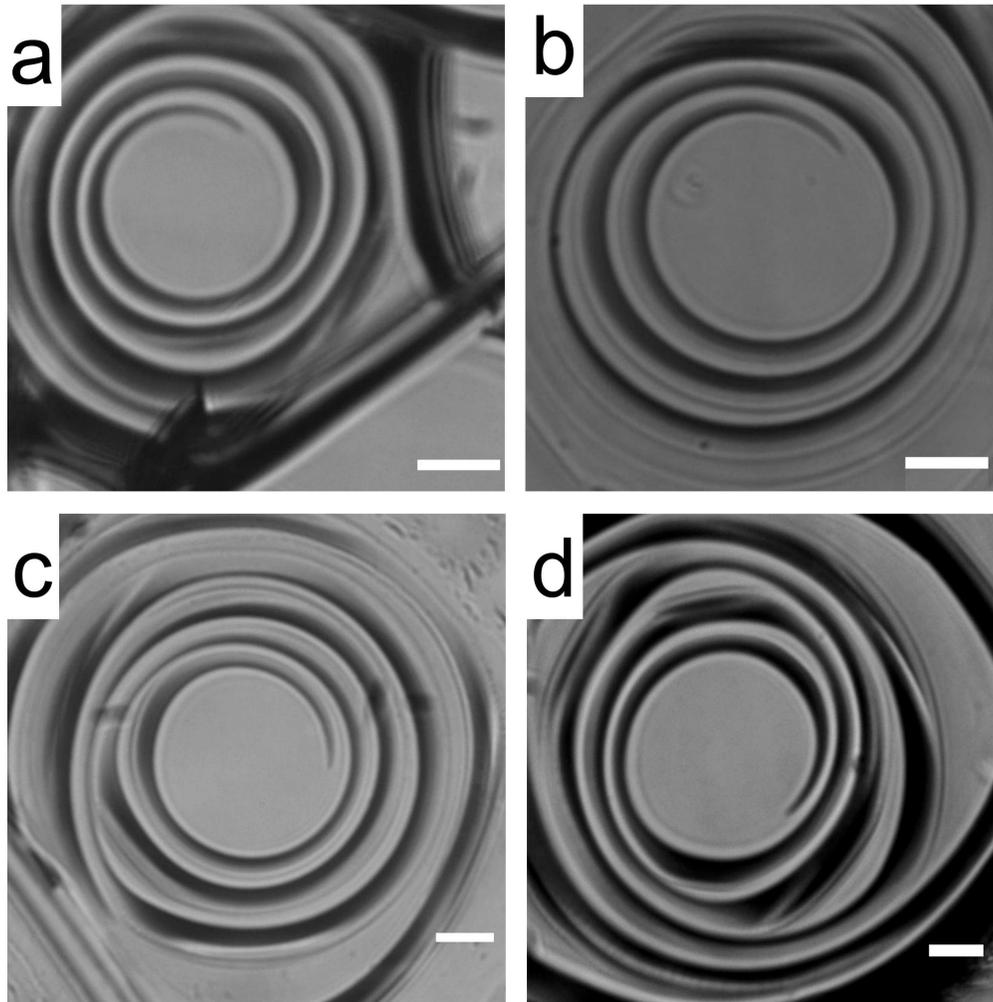


FIG. S1. (a)-(d) show the spirals from 1 to 4 are shown at  $\phi$  of 13 wt% respectively, with the scale bar of length  $10 \mu\text{m}$ .