Supporting Information

Anisotropic Scaling Lengths of Colloidal Monolayers near a Water-Air Interface

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1. Calculation of $z,κ^{\left(0\right)}$ and $κ^{\left(1\right)}$

There are two methods to calculate the value of $z$ according to Ref. (Wang & Prabhakar *et al.* 2009) and Ref. (Wang & Prabhakar *et al.* 2011). The value of $κ^{(0)}$ can first be obtained by $κ^{(0)}=6π/α$, which are 14.4, 14.5 and 17.4 for Si1, Si2 and Si3, respectively (listed in **Table** S1, column 4). The first method to calculate the value of $z/a$ is by using equation (1) in the manuscript, the results of which are listed in column 5 of Table S1. A second method is to use the equations (3-4) by Fischer(Fischer & Dhar *et al.* 2006). The results, denoted as $z'/a$, obtained by equation (3) are listed in column 6 of Table S1. The values of $z'/a$ for samples Si1 and Si2 are less than 1.0, which suggests that the particles are partially exposed to the air. This means that the monolayer is maintained at but not near the air-water interface. Such results conflict with the experimental observation: when the upside-down prepared sample cells were placed right-side up, the particles near the interface all dropped under gravity. If the particles remain at the air-water interface, then they will stay at the interface all the time due to the effect of the surface energy of the water interface. The results of $z/a$ in column 5 are more reliable than those of $z'/a$ in column 6. We believe that this inconsistency may be related to the difference between our physical system and Fischer’s: a continuous fluid on one side of the monolayer is replaced by a water layer with thickness $z$.

The deviation of$ κ^{\left(0\right)}$ and $κ^{\left(1\right)}$ using Fischer’s model are estimated as follows: The values of $z/a$ are substituted into equation (3) to recalculate$ κ^{\left(0\right)}$. The results of $κ^{\left(0\right)}$ are listed in column 7 of Table S1. The deviations of$ κ^{\left(0\right)}$ between columns 4 and 7 are approximately 6%. The values of $z/a$ are substituted into equation (4) to calculate$ κ^{\left(1\right)}$. The results of $κ^{\left(1\right)}$ are listed in column 8. Then, the values of$ κ^{\left(1\right)}$ are recalculated by substituting $z^{'}/a$ into equation (4), and the results are listed in column 9. The deviations of $κ^{\left(1\right)}$ between columns 8 and 9 are approximately 16%. The results for $κ^{\left(1\right)}$ in column 8 are used in the manuscript because they were obtained from the values of $z/a$ instead of $z'/a$.

**Table S1.**$ κ^{\left(0\right)}$ and $κ^{\left(1\right)}$ calculated using different methods for all three samples. In column 3,

$α=D\_{s}(0)/D\_{0}$; In column 4, $κ^{\left(0\right)}=6π/α$; In column 5, the value of ${z}/{a}$ is obtained from equation (1); In column 6, the value of ${z'}/{a}$ is obtained from equation (3); In column 7, the value of $κ^{\left(0\right)}$ is obtained through ${z}/{a}$ and equation (3); In column 8, the value of $κ^{\left(1\right)}$ is obtained through ${z}/{a}$ and equation (4); In column 9, the value of $κ^{\left(1\right)}$ is obtained through ${z'}/{a}$ and equation (4).

|  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- |
| 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 |
| Sample | $$a(μm)$$ | $$α$$ | $$κ^{\left(0\right)}$$ | $${z}/{a}$$ | $${z'}/{a}$$ | $$κ^{\left(0\right)}$$ | $$κ^{\left(1\right)}$$ | $$κ^{\left(1\right)}$$ |
| Si1 | $$1.57$$ | 1.31 | 14.39 | 1.21 | 0.85 | 15.3 | 3.55 | 4.17 |
| Si2 | $$1.00$$ | 1.30 | 14.50 | 1.26 | 0.89 | 15.4 | 3.47 | 4.11 |
| Si3 | $$0.60$$ | 1.08 | 17.45 | 4.68 | 2.56 | 18.5 | 1.18 | 2.1 |



Figure S1. The data from Figure 7 in the main text is presented in half-log plot. (a) Universal master curve of $\tilde{D}\_{‖}∙({z}/{a})^{{2}/{3}}$ as a function of $R\_{∥}$ for three samples. (b) Universal master curve of $\tilde{D}\_{⊥}∙({z}/{a})^{{2}/{3}}$ as a function of $R\_{⊥}$ for three samples.

References

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