Supplementary Information:

Chaining of hard disks in nematic needles:
particle-based simulation of colloidal interactions in liquid crystals

David Müller¹, Tobias Alexander Kampmann¹, and Jan Kierfeld¹,*

¹TU Dortmund University, Physics Department, Dortmund, 44227, Germany
*jan.kierfeld@tu-dortmund.de
**Effective interaction and residual elastic interaction**

Fig. 1 supplements Fig. 3 from the main text and gives additional information on the effective interaction $U$ and the residual elastic interaction $\Delta U = U - U_{\text{dep}}$ between two disks by showing cuts through the contour plot in Fig. 3 as a function of $r/\sigma$ or $\vartheta$.

**Figure 1.** (a) Measured effective interaction of two hard disks suspended in hard needles for different disk diameters $\sigma'$. The interaction is mostly attractive with a strength around $\sim 5k_BT$. The interaction minimum is at the disks’ surface at $\vartheta = 0^\circ$. The range of $U(r, \vartheta_0)$ in radial direction (left column) is of order of $l_0$. In angular direction (right column), $U(r_0, \vartheta)$ shows a growing repulsive area at $\vartheta = 90^\circ$ for larger disks. (b) Residual elastic interaction $\Delta U(r, \vartheta_0) = U(\vec{r}) - U_{\text{dep}}(\vec{r})$ of two disks for different diameters $\sigma'$. For larger disks the interaction shows a distorted quadrupolar pattern. For small disks ($\sigma' = 1$) the repulsive area around $\vartheta = 0^\circ/90^\circ$ is missing. (Figure created using matplotlib 3.2.1 (https://matplotlib.org/), python 3.8.2 (https://www.python.org/), inkscape 0.92.5 (https://inkscape.org/))

**Event-chain algorithm**

The event-chain algorithm is a rejection-free MC technique, which is based on global balance by introducing so-called lifting moves. For hard spheres or needles a lifting move is the transfer of a MC displacement from one particle to another particle. This means in a MC move only one particle at a time is active and moved along a line until it contacts another object. Then the remaining MC move distance is lifted to this object, which is then moved further.

Needles are represented by their two endpoints, where only one endpoint is moving at a time. In two dimensions the needle-needle interaction simplifies effectively to a collision of an endpoint with another needle. The remaining MC move distance is lifted to one of the endpoints of this needle. Therefore, we have a fluid of endpoints with an effective 3-particle interaction (two endpoints of a passive needle and the active endpoint). In Ref. 1 the generalization of the event-chain algorithm to N-particle interactions has been worked out. For needles the probability to which endpoint the MC move is lifted is proportional to the the distance to the other endpoint, i.e., it is lifted with higher probability to the closer endpoint. In the presence of additional disks, MC displacement is also lifted to disks if a needle collides with the disk and vice versa.

Effective collision detection is essential for a fast event-chain simulation. The collisions are calculated by intersections of a ray starting at the active particle (either a sphere center or a needle endpoint) with another ray, line segment, or circle. The construction and an overview of all possible cases is shown in Fig. 2 (a) and (b). To speed up the collision detection, we use a special neighbor list design. Each particle is confined to a “container”, which triggers an event when the particle leaves it. Then the neighbor list is updated, which ensures that the neighbor lists are always valid. Particles are added to the neighbor lists of the other particle and vice versa when their containers overlap. This way, for different particles different container shapes can be chosen. For the needles a very narrow rectangle can be used, which limits the computational effort for calculating the
The disk-disk interaction is given by

\[ V_{\text{disk}}(\vec{x}_i - \vec{x}_j) = \sum_{i<j} V_{\text{disk}}(\vec{x}_i - \vec{x}_j, \varphi_i, \varphi_j) \]

Here we present the derivation of the density-dependent depletion interaction including more intermediate steps. We use the results of Biben et al.\(^2\) and generalize them to anisotropic depletants with a rotational degree of freedom \( \varphi \) to get a density-dependent depletion interaction for disks in a suspension of hard needles. We consider a system of hard disks with positions \( \{ \vec{X}_i \} \) and \( N_h \) hard needles with positions \( \{ \vec{x}_i \} \) and orientations \( \{ \varphi_i \} \). Upper case indices refer to disks, s lower case indices to needles. The energy of the system is given by

\[ H = \sum_{i<j} V_{\text{disk}}(\vec{X}_i - \vec{X}_j) + \sum_{i<j} V_{\text{nm}}(\vec{x}_i - \vec{x}_j, \varphi_i, \varphi_j) + \sum_i V_{\text{nn}}(\vec{x}_i - \vec{X}_i, \varphi_i). \]

The disk-disk interaction is given by \( V_{\text{disk}} \), the needle-needle interaction by \( V_{\text{nm}} \) and the disk-needle interaction by \( V_{\text{nn}} \). By integrating over the needle degrees of freedom one can derive the effective interaction \( \gamma'(\{ \vec{X}_i \}) \) between the disks\(^2\),

\[ \beta \gamma'(\{ \vec{X}_i \}) = -\ln \left[ \prod_i d^3x_i d\varphi_i \exp \left( -\beta \left[ \sum_i V_{\text{nn}}(\vec{x}_i - \vec{X}_i, \varphi_i) + \sum_{i<j} V_{\text{nm}}(\vec{x}_i - \vec{x}_j, \varphi_i, \varphi_j) \right] \right) \right] \]

(\( \beta \equiv 1/k_b T \)). The corresponding force \( \mathcal{F}_K(\{ \vec{X}_i \}) \) on disk \( K \) is given by

\[ \mathcal{F}_K(\{ \vec{X}_i \}) = -\nabla_{\vec{X}_K} \gamma'(\{ \vec{X}_i \}) \]

\[ = -\sum_i \left[ \int \prod_{j \neq i} d^3x_j d\varphi_j \exp \left( -\beta \left[ \sum_j V_{\text{nn}}(\vec{x}_i - \vec{X}_j, \varphi_j) + \sum_{i<j} V_{\text{nm}}(\vec{x}_i - \vec{x}_j, \varphi_i, \varphi_j) \right] \right) \right] \times \left[ \int \prod_i d^3x_i d\varphi_i \exp \left( -\beta \left[ \sum_i V_{\text{nn}}(\vec{x}_i - \vec{X}_i, \varphi_i) + \sum_{i<j} V_{\text{nm}}(\vec{x}_i - \vec{x}_j, \varphi_i, \varphi_j) \right] \right) \right]^{-1} \nabla_{\vec{X}_K} V_{\text{nn}}(\vec{x}_i - \vec{X}_K, \varphi_i) d\vec{x}_i d\varphi_i \]

\[ = -\frac{1}{N_h^2} \sum_i \rho^{(1)}(\vec{x}_i, \varphi_i|\{ \vec{X}_i \}) \nabla_{\vec{X}_K} V_{\text{nn}}(\vec{x}_i - \vec{X}_K, \varphi_i) d\vec{x}_i d\varphi_i. \]
We use the superposition approximation where we used where 

The effective potential is the density-dependent depletion interaction, which we further approximate by

By using \( \nabla_{\vec{x}_K} V_{\text{dn}}(\vec{x}_i - \vec{x}_K) \phi_i = -\nabla_{\vec{x}_i} V_{\text{dn}}(\vec{x}_i - \vec{x}_K) \phi_i \), evaluating the sum to a factor \( N_n \) and defining the average over the needle angles as \( \langle A \phi \rangle = \int d\phi A(\phi) \), we get

For the case of two disks at \( \vec{0} \) and \( \vec{r} \) this yields

We use the superposition approximation

where \( \rho(\vec{r}', \phi) \equiv \rho(\vec{r}', \phi) / \rho_n \) is the density distribution around a single disk and \( \rho_n \) is the average needle density. For a single disk the needles are distributed according to the direct interaction potential \( V_{\text{dn}}(\vec{r}, \phi) \),

resulting in

Using this in eq. (1) we arrive at

This effective potential is the density-dependent depletion interaction, which we further approximate by

where we used \( \rho(\vec{r}', \phi) \rho(\vec{r}' - \vec{r}, \phi) \phi \approx \rho(\vec{r}', \phi) \phi \rho(\vec{r}' - \vec{r}, \phi) \phi \), which is valid for the isotropic phase and the ideal nematic phase. Since we investigate the effective interaction in the nematic phase this should be a good approximation.

For the special case of an idealized density that is a step function and either zero or \( \rho_n \) (see Fig. 3), the effective potential essentially becomes the well-known depletion interaction \( \beta U(\vec{r}) = -\rho_n A_{\text{ov}} \), where \( A_{\text{ov}} \) is the overlap area of the excluded areas.

References

1. Harland, J., Michel, M., Kampmann, T. A. & Kierfeld, J. Event-chain Monte Carlo algorithms for three- and many-particle interactions. *EPL* **117**, 30001, DOI: 10.1209/0295-5075/117/30001 (2017).

2. Biben, T., Bladon, P. & Frenkel, D. Depletion effects in binary hard-sphere fluids. *J. Phys. Condens. Matter* **8**, 10799–10821, DOI: 10.1088/0953-8984/8/50/008 (1996).

3. Asakura, S. & Oosawa, F. On interaction between two bodies immersed in a solution of macromolecules. *J. Chem. Phys.* **22**, 1255–1256, DOI: 10.1063/1.1740347 (1954).
Figure 3. Idealized step-like depletion zone around a disk (a) and resulting overlap area $A_{ov}(r, \vartheta)$ (b). (Figure created with Inkscape 0.92.5 (https://inkscape.org/))