Simulation study of the electrical tunneling network conductivity of suspensions of hard spherocylinders

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Using Monte Carlo simulations, we investigate the electrical conductivity of networks of hard rods as a function of the volume fraction for two tunneling conductance models. For a simple, orientationally independent tunneling model, we observe non-monotonic behaviour of the bulk conductivity as a function of volume fraction at the isotropic-nematic transition. However, this effect is lost if one allows for anisotropic tunneling. We also compute the mesh number of the Kirchhoff network, which turns out to be a simple alternative to the computationally expensive conductivity of large systems in order to get a qualitative estimate.

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I. INTRODUCTION

At the percolation transition, clusters of connected objects merge to form one system-spanning network. This transition has first been studied in the context of the flow of water through porous rock. By now, due to the technological relevance of transport processes in disordered media, in general, as well as the mathematical properties of percolation transition, percolation has become a topic of interest in a wide range of different research fields.\(^{2,60}\) In this article, we focus on the formation of networks of electrically conducting fibres, as they would be used e.g. in a composite material that is designed to undergo a transition from an insulating state to a conductive state. These types of composites have been studied extensively for almost 50 years in experiment, simulation and theory. A complete literature overview is hence beyond the scope of this paper. Instead, we refer the reader to a selection of relevant articles in the context of the present work: For reviews on conductive composites see e.g. refs.\(^{23,40,49,53,55,59}\), for theoretical approaches to connectivity percolation in rods see e.g. refs.\(^{5–7,11,12,17,18,24,30,34,35,38,47,56,64,65}\), for simulation e.g. refs.\(^{3,4,9,21,29,37,39,41,50,51,54,58}\) and for experiments refs.\(^{8,10,15,22,26,27,31,32,43,52,61}\).

In suspension, anisotropic particles form liquid crystalline phases. In particular, on increasing the concentration of rod-like particles, they undergo a transition from an isotropic phase to an orientationally ordered, but positionally disordered phase (called the "nematic" phase)\(^{16,46}\). In general, the electrical conductivity of a suspension of aligned particles is different from that of an isotropic system. The effect of alignment on networks of conductive fillers has been studied in experiment\(^{1,14,19,62,63}\) and in theory and simulation\(^{20,25,28,48,66}\). However, these studies considered alignment by means of an external field such as e.g. shear and not alignment due to a phase transition that is intrinsic to the system. To our knowledge there is no systematic study of the behaviour of the conductivity across the isotropic-nematic (I-N) phase transition yet. It is the aim of our study to fill this gap.

II. MODELS AND SIMULATIONS

We have modeled only the filler particles and neglected the host environment of the nanocomposite. We used hard spherocylinders ("rods") with a cylindrical part of length \(L\) and diameter \(D\), capped on both ends with hemispheres of diameter \(D\). The rods are
'coated' by a spherocylindrical penetrable contact shell of diameter $\lambda$ which serves as a means of establishing a geometric connectivity criterion between pairs of rods, where then simply, if two rods have overlapping contact shells they are considered to be a connected pair. By extension then, a cluster can be formed by a contiguous sequence of such pairwise connected rods.

We have generated configurations of hard rods using canonical Monte Carlo simulations (NVT-MC). The simulations have been carried out for systems of aspect ratio $L/D = 20$ primarily, using a non-cubic simulation box of dimensions $L_{x,y} = 3L, L_z = 4L$, (in which the director of the nematic phase was roughly parallel to the $z$-direction) and with particle numbers ranging from $N = 1000$ to $N = 5000$ in order to cover a range of volume fractions that span configurations in both the isotropic and the nematic phase. The volume fraction is defined as $\eta = N v_{\text{core}} / V$, with $N$ the number of rods in suspension, $V$ volume of the simulation box, and $v_{\text{core}} = \pi D^2(L/4 + D/6)$ denotes the volume of one rod. Furthermore, we have chosen to work with two different contact shell diameters, $\lambda = 1.1D$ and $\lambda = 1.2D$. This choice is motivated by the fact that the former is small enough such that irrespective of the volume fraction we simulated the suspension never exhibit a percolating cluster, while conversely for the latter choice of $\lambda$ we were always in the percolated state. As a scalar order parameter to distinguish between the isotropic and nematic phase, we use $S_2$, the maximum eigenvalue of the orientation tensor $Q$ as given by:

$$Q_{ij} = \frac{1}{2N} \sum_{\alpha=1}^{N} (3v_{i}^{\alpha}v_{j}^{\alpha} - \delta_{ij})$$

(1)

where $v_i^{\alpha}$ and $v_j^{\alpha}$ are the $i$th and $j$th components of the normalized orientation vector of rod $\alpha$ respectively and $\delta_{ij}$ is the Kronecker delta.

For the conductance $g_{ij}$ between a given pair of rods, we use the commonly adopted model of single-electron tunneling as the dominant process for electron transfer in composite systems dispersed with nano-sized conductive filler particles. The general form of the model is given by a simple exponential decay function $g$:

$$g(d_{ij}) = g_0 \exp\left(-\frac{2d_{ij}}{\xi}\right)$$

(2)

where $d_{ij}$ denotes the distance between the surfaces of rods $i$ and $j$, and $\xi$ is the tunneling decay length (which is usually taken to be in the order of magnitude of a few nanometers
and in our simulations it is simply taken as difference between the penetrable shell diameter \( \lambda \) and the impenetrable rod diameter \( D \). Conventionally, we set the conductance between two rods whose surfaces are touching to be equal to unity which resolves the pre-factor in (2) into being unity as well \( g(0) = 1 \). Before proceeding, it is important to discuss the range of validity of Eq. (2), which is sometimes also referred to as *normal tunneling*. Briefly, the limit of validity of normal tunneling as the dominant process of electron transport between nano-sized conductive filler particles, is directly related to the cross-section area of the tunneling junction between them, namely, the narrower the junction the higher the probability for events of single uncorrelated electrons tunneling through the junction barrier. The cross-section area depends primarily on the shape of the electrodes, which in our case are spherocylindrical. Furthermore, for the general working temperature of nanocomposites, namely \( T < 500K \), normal tunneling is expected to remain valid. But given that hard systems are athermal together with the fact that the area of the junction is then only dependent on particle geometry and relative orientation with a neighbouring particle, the former validity criterion can be translated into one on the aspect ratio, \( L/D << 200 \). For our purposes, our choice of \( L/D = 20 \) remains then reasonable if we are to adopt Eq. (2) in order to describe pairwise conductances. For an in-depth discussion on the validity of normal tunneling in systems of elongated particles, we refer the reader to the work of Sherman et al [57].

Next to Eq. (2), we also applied a second conductance model which deviates from Eq. (2) by its explicit incorporation of the relative orientation between a pair of rods in order to account for the cross section of the tunneling junction between the pair. More precisely, G. Negro and C. Grimaldi [42] have recalculated the matrix element for electron tunneling between rod-like particles and have shown that in general the tunneling between two parallel rods (e.g., in the nematic phase) is more probable by a factor of \( L/\sqrt{\xi D} \) compared to two rods perpendicular to one another. Moreover, they show that for disordered phases (e.g., in the isotropic phase) the orientationally dependent contributions to the conductance become negligible once an averaging over all solid angles is performed. However, for strongly oriented phases of suspension of rod-like particles, the enhanced tunneling between parallel rods is expected to have a significant influence on the conductivity of the network of rods. For comparison with our main results which are obtained according to Eq. (2), we will adopt a simplified form of the anisotropic tunneling conductance which is obtained with the un-
derlying assumption that the centres of mass of two connected rods are close to their line of shortest distance: 

\[ g(d_{ij}, \gamma_{ij})/g_0 = \frac{\exp \left( -2d_{ij}/\xi \right)}{2\pi D \xi + \sin^2(\gamma_{ij})} \]  

(3)

where \( \gamma_{ij} \) denotes the angle between the long axes of the rods. We note that, our comparison aims merely at probing the effects on the bulk conductivity resulting from a simple deviation from the common model Eq. (2), one that would account in an explicit manner for the relative orientation when estimating the tunneling conductance between two rods. Therefore, we take one of the simplest forms of the corrections derived by G. Nigro and C. Grimaldi, which offers the added advantage of maintaining the numerical computations rather simple. Moreover considering that on the one hand, we have only chosen to work with short aspect ratio rods, namely of 20 and 10, contained in simulation boxes of dimensions 4 and 6 times larger than \( L \) respectively, and that on the other hand, only small tunneling decay lengths (10 – 20% of rod diameter) are considered, the assumptions behind Eq. (3) remain for our purposes safe to work with.

Lastly, we briefly discuss our method for estimating the network conductivity \( \sigma \) of percolating clusters of hard rods. The resistor (conductance) network is built by assigning a node to each rod and a resistor between all pairs of nodes in the system whose value is given by the inverse of their corresponding conductance either given by Eq. (2) or (3). In order to reduce the computational efforts, we set the tunneling conductance of pairs of rods whose shortest distance is greater than \( \lambda^2/D \) to zero. Finally, two electrodes are placed on the most distant nodes, and in order to solve the Kirchhoff equations (i.e., to obtain the equivalent resistance or equivalently the overall conductivity) of the corresponding tunneling-based resistor network, we apply once an exact numerical decimation using the Star-Mesh transform which entails applying Rosen’s formula until all nodes except the electrode-nodes are eliminated, and as a second method for comparison, we measure the equivalent resistance of the network between the two electrode-nodes by performing a DC simulation on the network in Qucs. The final conductivity value for each volume fraction is averaged over an ensemble of 200 independent largest clusters.

In an independent calculation, we counted the number of meshes of the Kirchhoff network.

We recorded the edge-vertex-incidence matrix of the network corresponding to the largest
cluster considered as an undirected graph, and then computed the mesh number from the dimension of its kernel, i.e., by subtracting its rank from the number of resistors (size of the graph).

III. RESULTS

In Fig. 1 examples of percolating clusters in the isotropic and nematic phase are visualized. We start by discussing the bulk conductivity ($\sigma$) as a function of volume fraction ($\eta$) for values in the isotropic and the nematic phase and for two contact shell diameters, $\lambda = 1.1 D, 1.2 D$ and $L/D = 20$. In Figure 2 we show results for normal tunneling (eqn. 2). The conductivity increases with volume fraction for the percolating case (for $\lambda = 1.2 D$), while below the percolation threshold ($\lambda = 1.1 D$) we observe an essentially constant and negligibly small value for $\sigma$. In the latter case, the conductivity of the nano-composite would correspond to that of the polymeric host material alone.

In fig. 2 we also show the nematic order parameter $S_2$ (red dotted line, second y-axis) of the entire system. At the onset of the nematic phase (vertical solid line) and for $\lambda = 1.2D$, despite the increase in volume fraction, we observe a clear drop in $\sigma$ relative to the last value observed in the isotropic phase. This non-monotonic behaviour is due to the rapid increase in orientational order at the phase transition, which has the effect of reducing the excluded volume between neighbouring rods, thus in turn increasing the shortest distance between them and therefore leading to a reduced tunneling conductivity. After the drop at
the onset, \( \sigma \) increases again with \( \eta \), but less strongly than in the isotropic phase.\(^{44}\)

Next we compare the dependence of \( \sigma \) on \( \eta \) for the two different tunneling conductance models given in Eqs. 2 and 3. In Fig. 3, both below and above the percolation threshold, we observe that the anisotropic tunneling model enhances the conductivity, in particular in the nematic phase. The non-monotonic behaviour of the conductivity across the transition is lost for the more realistic, anisotropic model. As shown in Fig. 4, the same behaviour in conductivity as a function of \( \eta \) is also observed for \( L/D = 10 \), based on the model Eq. 3 and using \( \lambda = 1.1D \) which is above the percolation threshold.

The enhancement of the conductivity in nano-composites across the I-N transition of the filler particles has previously been theoretically predicted\(^{66}\) in the context of polymeric fillers of very high aspect ratio (\( L/D = 100 \) and above) under shear flow. Zheng and co-workers showed that the relative conductivity enhancement\(^{45}\), which is the relative conductivity change in the polymeric bulk after and before the dispersion of conductive filler particles, grows linearly with volume fraction both in the isotropic and nematic stable regions.

In order to draw a closer comparison in addition to the general qualitative agreement, we set the conductivity in the host insulating material as \( \sigma_{\text{ins}} = \langle \sigma \rangle_{\lambda=1.1} \) and similarly the conductivity in the conductive state to \( \sigma_{\text{co}} = \sigma_{\lambda=1.2} \), i.e., \( \sigma \) in the presence of percolating clusters. With the latter choices, we define the relative conductivity enhancement as the ratio \( \epsilon = \sigma_{\text{co}}/\sigma_{\text{ins}} \). In Fig. 5, the relative enhancement based on the anisotropic conductance model is plotted as a function of \( \eta \). We clearly observe that the relative enhancement is well described in the nematic phase by an exponential function, \( \epsilon(\eta) \approx \exp(\alpha \eta) \), with \( \alpha > 0 \), i.e. we observe exponential rather than linear scaling. In the inset plot of Fig. 5, a linear fit applied to \( \ln \epsilon \) restricted to the volume fraction range in the nematic phase yields \( \alpha = 19.9 \pm 0.9 \).

We note that due to the fact that in this comparison the aspect ratios are not the same, nor are the considered conductivities in the host insulating environment (more precisely the ratios of \( \sigma \) after and before dispersion), a direct quantitative comparison with ref.\(^{66}\) remains difficult to perform.

Next we briefly discuss our observations on the anisotropy of the bulk conductivity in the aligned phase of the conductive fillers. Using the model Eq. 3 and \( \lambda = 1.2D \), we compare \( \sigma \) as measured in parallel and orthogonal directions to the common director field \( \vec{n} \) of the rods in the nematic phase. The comparison is shown in Fig. 6, where near the nematic coexistence
FIG. 2. Conductivity for the normal tunneling model Eq. 2 as a function of volume fraction for two different contact shell diameters, \( \lambda = 1.1D \) (blue symbols, mean value as a dashed line), and \( \lambda = 1.2D \) (green symbols, solid line). In addition, on the second Y-axis the mean nematic order parameter \( S_2 \) (red, dotted line) values of the entire system (not just the connected clusters) is plotted, and the vertical solid (brown) line indicates the coexistence volume fraction in the nematic phase for \( L/D = 20 \). (colour online)

volume fraction we observe on average the same behaviour for \( \sigma \) along the two directions with respect to \( \vec{n} \). On the other hand, for volume fractions well past this region, i.e., with increasing orientational ordering of the rods, we notice a clear separation in trends where \( \sigma \) values measured along the director field become increasingly larger with \( S_2 \). This reflects the fact that the increased orientational ordering well in the stable region of the nematic phase, does introduce anisotropy (favoured direction) in the bulk conductivity, which has been a consistent observation in various experimental works.\[1,14,19,62,63\]
FIG. 3. Comparison of the dependence of $\sigma$ on $\eta$ for the two tunneling conductance models as given in Eq. 2 (labelled Iso.) and Eq. 3 (labelled Aniso.). Left: $\lambda = 1.1D$, right: $\lambda = 1.2D$. The nematic order parameter $S_2$ is shown on the second Y-axis in each case.

Finally, we compare the trend in $\sigma$ with the mean number of meshes of the tunneling network as a function of $\eta$. This comparison is shown in Fig. 7. We observe that the mean mesh number follows the same trend with $\eta$ as we have observed for $\sigma$ (However, there are clear qualitative differences in the functional form.) This offers the potential for a simpler and computationally less expensive route to extracting the trend of $\sigma$ without explicitly solving the Kirchhoff equations of the tunneling network.

IV. CONCLUSIONS

We have studied the conductivity of networks of hard spherocylinders in the isotropic and the nematic phase. For the widely-used normal tunneling model we observe non-monotonic behaviour of the conductivity at the phase transition. However, for a more realistic anisotropic tunneling model, this effect is lost. Further we observe that the conductivity in the nematic phase is isotropic close to the phase transition, but becomes anisotropic at higher volume fractions. We also analyzed the number of meshes in the Kirchhoff resistor network and found it to be a very rough, but fast to compute, estimator for the conductivity.
FIG. 4. Conductivity for the anisotropic tunnel model Eq. 3 as a function of volume fraction, for $L/D = 10$ and $\lambda = 1.1D$, which is above the percolation threshold for the entire range of chosen volume fractions here. The mean nematic order parameter ($S_2$) is plotted on the second Y-axis (red dashed).

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FIG. 5. The relative conductivity enhancement in percentages for the anisotropic tunneling model as a function of volume fraction. $\sigma_{\text{co}}$ denotes $\sigma$ in the conductive state, i.e., in the presence of percolating clusters ($\lambda = 1.2D$), and $\sigma_{\text{ins}}$ denotes the mean conductivity value in the absence of percolating clusters ($\lambda = 1.1D$). The inset plot shows $\ln(\sigma_{\text{co}}/\sigma_{\text{ins}})$ as a function of $\eta$ restricted to the nematic phase, with the linear fit shown in dashed.

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FIG. 6. Conductivity in the nematic phase for $L/D = 20, \lambda = 1.2D$, using the model Eq. 3, as a function of volume fraction. $\sigma$ measured along the director $\vec{n}$ of the rods (orange, triangular), compared to $\sigma$ measured in the orthogonal direction to $\vec{n}$ (blue, round).

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Note2. More precisely, first an effective expression for the electrical conductivity tensor is derived, then the largest relative enhancement is defined in terms of the maximum eigenvalue of the effective conductivity tensor after the inclusion of conductive filler rods. For a clear account of all details, we refer the reader to the complete work of Zheng et al. [66].

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