GENERAL OUTLOOK

While these four years of work did not yield all the results we hoped for (due to unexpected experimental difficulties and pandemic hiccups), it has not been for naught. This work has shown to be extremely timely and has contributed to the advancement of knowledge in the field, by setting a foundation for further studies on the use of self-assembled networks for functional materials, in general, and neuromorphic materials, in particular. Following is an outlook on the impact of this work on the field and the future of networks for neuromorphic applications.

My collaborators, Jin Xu, Katja Loos, and Beatriz Noheda, were the first to show a combination of polymer imprinting and templating to form $CoFe_2O_4$ nanostructures. [1] Here, I have expanded on this work and shown that this method can be generalized and applied for different metal oxides. And while there is still room for optimization and improvement of the growth conditions, the principles have been established with the present work. I have demonstrated a variety of different metal oxide nanostructures, mostly from complex oxides with more than one cationic site, to be accessible through chemical solution deposition (CSD) with using only one type of block copolymer (BCP). These results show that this same method can be applied to other types of metal oxides; and additional materials and structures can become accessible through the use of different BCPs. Thus, this thesis work shows the great versatility of the technique and forecasts an applicability scope broader than that presented in this thesis. Additionally, further advances could be made by investigating other deposition techniques such as atomic layer deposition (ALD) and specifically sequential infiltration synthesis (SIS) (see Chapter 3.3.1) in order to gain more control over the growth mode and crystal phase of the nanostructures. Based on the work of Sønsteby et al. [2], ALD seems a promising candidate to unlock low-temperature epitaxial growth of oxides, if this method can be generalized beyond the demonstrated nickelates. Growing templated networks through low-temperature ALD unlocks a great potential for future industrial applications, since Back-End-Of-Line (BEOL) compatibility typically requires a maximum processing temperature in the range of 400 °C. [2–5]

As discussed in chapters 4 and 5, networks of different types and morphologies are already investigated for applications in neuromorphic materials but the accessible materials classes are very much limited to noble metals and chalcogenides; with conducting domain walls being a recent addition to this family. The self-assembly of different types of materials in network structures is, therefore, of great interest. If such networks could be properly controlled, they would be very valuable for application purposes, as it would allow the fabrication of complex crosslinked array structures by self-assembly. There are a lot of possibilities of applying such structures into neuromorphic materials (or adaptable electronics, more in general). Once the experimental difficulties of this new avenue have been ironed out, the

main focus should be to obtain the desired material properties: the obtained networks should show hysteretic and non-linear multilevel resistance values; and they should be dynamic, such that connections can be continuously formed and broken. It is important, though, that we learn more about the operation of memristors in a network, especially non-idealized and disorganized networks, to be able to fully harness the power and complexity self-assembled networks have to offer.

Indeed, understanding the behavior and operation of memristors in a network is necessary to extend the use networks beyond the reservoir computing applications that are currently thought of as the main use of such systems.[6–9] In order to fully grasp the operation of memristive networks, it is key to take on an interdisciplinary approach, in which material scientists work closely together with mathematicians, computer scientists and AI experts, in order to combine experimental data with the vast theoretical knowledge on network theory. Once we - as a field - are able to properly model our experimental setups, we will be able to use networks in applications and move beyond crossbar array structures.

BCP templating seems a promising solution that offers the necessary control and the work of this thesis is extremely timely, in view of the publication a couple of months ago by Doerk et al.[10], where they present the use of artificial intelligence (AI) to predict new BCP self-assembly structures, showing a large gamut of interesting new structures that could be applied in network formation. Combining the outputs of the work by Doerk et al.and our work allows for the actual realization of these structures as metal oxide nanostructures and can ultimately give rise to new structures and applications; and, thus, have the potential to open a completely new field in materials science. Since, BCP self-assembly and network formation involve too many parameters to fully grasp and control, the use of AI seems to be the right tool that can take this research to the next level.

Finally, this thesis is part of the required work needed to pave the way to unlock this field as a relevant player in the neuromorphic networks community. Here I have shown that there is, in principle, no limit to the materials that can be processed to form self-organized networks and I have presented the first indications of possible memristive properties in the $La_{0.7}Sr_{0.3}MnO_3$ nanostructures (Chapter 5), providing a foundation for further studies.

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