Coherent x-ray studies of spontaneous fluctuations in rare earth nickelates

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Rare-earth nickelates (RNiO₃) exhibit a rich interplay of electronic, magnetic, and structural phase transitions, including a metal-to-insulator transition (MIT), charge ordering, and a symmetry change from orthorhombic to monoclinic structure [1]. While these transitions have been widely studied, the spatio-temporal nature of spontaneous fluctuations across the phase boundary remains largely unexplored. Such fluctuations are increasingly recognized as crucial for stabilizing emergent magnetic textures and for enabling stochastic functionality in neuromorphic computing. Here, we employ X-ray photon correlation spectroscopy (XPCS) [2-3] to directly probe the dynamics of structural and magnetic fluctuations in epitaxial thin films of NdNiO₃ and SmNiO₃. For NdNiO₃, we observe a pronounced slowdown in fluctuation timescales—by an order of magnitude—near the Néel temperature, highlighting strong coupling between structural and magnetic order parameters, independent of epitaxial strain. In contrast, SmNiO₃ shows no such slowdown, emphasizing the distinct dynamics. Unexpectedly, wavevector-dependent measurements reveal that short-range structural fluctuations are significantly slower (by a factor of 3–5) than long-range fluctuations [4]. Our results demonstrate the power of coherent X-ray techniques in capturing nanoscale fluctuation dynamics in quantum materials and provide new insight into the role of fluctuations near phase transitions in complex oxides.

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3. Shpyrko O.G. X-ray photon correlation spectroscopy. Journal of Synchrotron Radiation 21, 1057 (2014)
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Rare earth nickelates RNiO3 display an metal to insulator transition (MIT) which is accompanied by a magnetic transition, charge ordering, and a crystal structure change from orthorhombic low temperature to monoclinic high temperature state. While this system has been widely studied, the nature of fluctuation across the transition, and the associated time- and length-scales is not known. Spontaneous fluctuations are important component for stabilizing topological magnetic structures such as skyrmions in quantum materials. Fluctuations of the order parameter near the phase transition boundary in complex oxides could also be key to designing probabilistic bits and stochastic synapses. Direct probes of fluctuations however are non-trivial due to the requirement of coherent spatio-temporal probes. In our work, we focus on understanding the role of spontaneous fluctuations in rare earth nickelates by employing x-ray photon correlation spectroscopy (XPCS). Our XPCS measurements on NdNiO3 thin films, show complex evolution of fluctuations dependent upon temperature, wait time and wavevector q. Our studies show strong coupling of structural and magnetic order parameters with slowdown of fluctuation timescales by one order of magnitude near Néel temperature irrespective of the epitaxial strain of the film. Our measurements of diffraction wavevector dependence indicate that short range structural fluctuations are surprisingly slow. Our results reveal novel insights into fluctuations enabled by coherent x-rays, expanding the unique capability of spatio-temporal probes to investigate magnetic and structural fluctuations. These finding could have implications, extending beyond the nickelates studied here to other metal-insulator transition systems, and shaping our understanding of fluctuation dynamics near phase transitions in complex materials.

Phase transitions in complex oxides are extensively studied for their potential to enable advancements in neuromorphic computing. Notably, fluctuations of the order parameter near the transition boundary in complex oxides could be key to designing probabilistic bits and stochastic synapses. Direct probes of fluctuations however are non-trivial due to the requirement of coherent spatio-temporal probes. Here, we report the direct measurement of magnetic and lattice fluctuation dynamics in prototypical perovskite nickelate systems of NdNiO3 and SmNiO3 using X-ray photon correlation spectroscopy. For NdNiO3, our studies show strong coupling of structural and magnetic order parameters with slowdown of fluctuation timescales by one order of magnitude near Neel temperature irrespective of the epitaxial strain of the film. For SmNiO3, no such slowdown is observed emphasizing the distinct nature of structural fluctuations. Contrary to the existing theories, our measurements of diffraction wavevector dependence within the Bragg peak, indicate that short range structural fluctuations are surprisingly slow (3-5 X slower) compared to long range fluctuations. Our results reveal novel insights into fluctuations enabled by coherent x-ray techniques, expanding the toolkits to investigate quantum materials resolved in space and time.