**Negative thermal expansion of pullulan multilayers**

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The change of temperature leads to thermal expansion. Most materials expand, but, some materials contract upon heating, characterized by negative thermal expansion [1]. Such negative thermal expansion material have various applications such as photoelectric devices, fillers in controlled thermal expansion composites, aerospace technology, as dental fillings [2].

We combine holography and phase separation [3] to design complex photonic structures that exhibit negative thermal expansion behavior. The structures were recorded in pullulan, (linear polysaccharide) doped with ammonium dichromate, and consist of a multilayer composed of pullulan and air.

We study optical response during heating and cooling of the synthesized pullulan nanostructures. The reflection wavelength of the hologram was used to characterize the temperature dependence of their shrinkage and swelling behavior. By heating and cooling the sample partially reversible shifts of the photonic band gap could be observed. We have observed that heating leads to a reduction in the thickness of the structure, resulting in the reflectance peak shift towards shorter wavelengths. With temperature decreasing the band gap position shifts back towards longer wavelengths. We reveal temperature sensitive responses of the structure, which are the consequence of the mechanical deformation of air nanolayers upon heating or cooling. The temperature change leads to the increased permeability of air molecules through the pullulan nanolayers, which causes mechanical changes.

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