



## Photo-induced deformation of active polymer films: Single spot irradiation

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### ABSTRACT

Light-activated polymers can undergo complex deformation in response to the combination of mechanical and optical stimuli. These materials are attractive for remote actuation and sensing applications. However, the behavior of such materials subjected to photomechanical patterning is not well understood. In this paper we consider a polymer that operates by photoactivated stress relaxation; at the molecular level, photoinitiation of residual initiator molecules generate free radicals that break and then reform in-chain functionalities of stretched chains in an elastomeric network, which results in macroscopic stress relaxation. We carry out experiments and finite element calculations that demonstrate the sequence of deformation events culminating in the formation of a buckled spot as a result of biaxially stretching the elastomeric film then irradiating a circular region followed by releasing the mechanical constraint. In order to better understand the photomechanics, we analyze a simpler model problem wherein a linear elastic, stress relaxing disk is subjected to (i) radial extension, (ii) irradiation of a concentric circular region, and (iii) release of the applied displacements in (i), which results in deformation and stress redistribution. In the final step, the deformation may transition from planar to buckling out of the plane depending on system parameters. Companion finite element calculations are performed against which our analytical results are in good agreement. Although not directly comparable, the analytic model qualitatively agrees with the experiments. The results of this work provide a useful foundation from which to explore more interesting behavior of periodically photo-mechanically patterned films and other more challenging actuation problems.

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### 1. Introduction

Mechanically-responsive, light-activated polymers (LAP) are an exciting class of environmentally activated materials that undergo large deformations in response to irradiation at particular wavelengths (Finkelmann et al., 2001; Ikeda et al., 2003; Li et al., 2003; Warner and Mahadevan, 2004; Lendlein et al., 2005; Jiang et al., 2006). One critical feature that distinguishes LAPs from other environmentally activated materials, such as thermally driven shape memory in polymers (Tobushi et al., 1996; Liu et al., 2006; Behl and Lendlein, 2007; Qi et al., 2008) or swelling in temperature-sensitive hydrogels (Qiu and Park, 2001; Yu et al., 2001; Westbrook and Qi, 2008; Guvendiren et al., 2009), is that light as stimulus offers remote application with precise spatial control that can be readily realized with existing optical technologies. Such capabilities make possible complex photo-mechanical applications for sensing and actuation that are not feasible with other systems.

In this article, we focus on the behavior of a photo-induced network rearranging (PNR) elastomer (Scott et al., 2005, 2006), which

exhibits macroscopic photo-induced stress relaxation and operates as follows (Long et al., 2009). The material is deformed in a prescribed manner, held in the deformed shape, and irradiated with light at a prescribed material-dependent wavelength. A sequence of photo-chemical reactions ensues that continuously fragments and reforms polymer chains in the network, ultimately rearranging the connectivity of the network, evolving it to a stress-free configuration. Subsequently, when the mechanical constraints are released, the material deforms into a shape that is different from the original shape in order to satisfy mechanical equilibrium. We idealize the material's complex photomechanics via a rule-of-mixtures approach wherein the continuum mechanical behavior of a material point is represented by the volume-fraction weighted sum of the behaviors of two networks that are stress free in different configurations. The original network is stress free in the reference state of the body while the reformed network is stress free in an intermediate configuration determined by the irradiation history. Irradiation and subsequent network fragmentation and reformation drive the evolution of the respective network volume fractions as well as the stress free configuration of the reformed network.

Realizing applications using these novel materials requires experimentally validated theory and attendant computational

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