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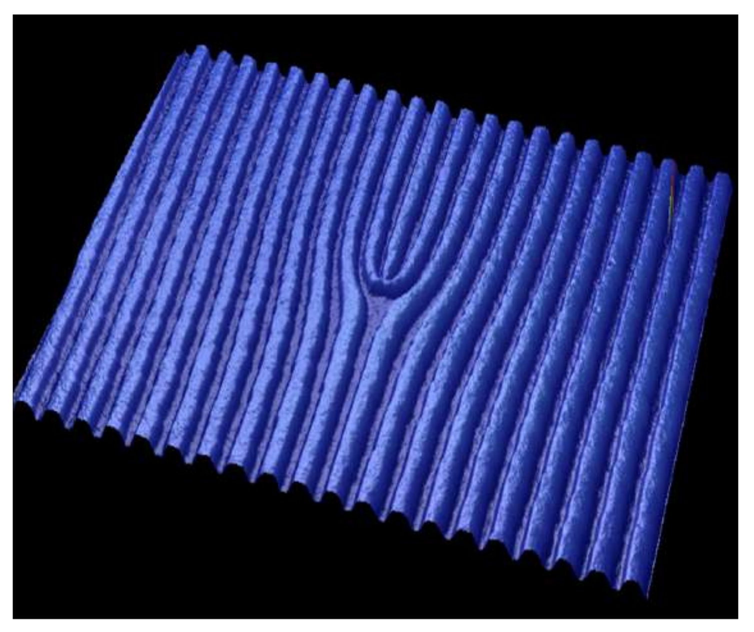

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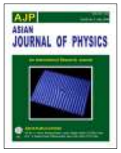

Tunable optical submicron structures based on soft matter

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Tunable, active, adaptive and variable optics has experienced a steep development during the last two decades. While initially in adaptive optics, the individual segments of, for example large mirror were actuated individually by mechanical means, a current trend goes towards liquid and soft optical elements. Soft matter brings along with it a wealth of effects that are new in the field of optics. This article, which is intended at the same time an introductory tutorial and a short review, describes the current status in a sub-field of tunable optics and focuses on optical elements based on submicron structures. This scope encompasses all types of diffracting elements, Bragg mirrors and filters, and photonic bandgap elements. After the introduction, we give a concise overview on the material basics and the fundamentals of the tuning effects. The following section presents an overview on the state of the field as found in scientific literature. The article closes with some concluding remarks and the references..© Anita Publications. All rights reserved.

Keywords: Photonics, Nanooptics, Nano-photonics, Photonic crystal,Grating, Bragg grating,

1 Introduction

Tunable, active, adaptive and variable optics has experienced a steep development during the last two decades. While initially in adaptive optics, the individual segments of for example large mirror were actuated individually by mechanical means a current trend goes towards liquid and soft optical elements. Soft matter brings along with it a wealth of effects that are new in the field of optics. This approach has turned out as highly fruitful in particular in the field of refractive optics. There, liquid and elastomeric tunable lenses have been developed and currently find their first real-world applications in industrial sensor technology and in adaptive automotive headlights. The latest status in this area has been published recently in an edited book [1].

Beyond the developments presentend in [1], optical elements based on submicron structures, too, have profited from these developments. These include all types of diffractive elements, hybrid refractive-diffractive elements, Bragg mirrors and filters, and photonic bandgap structures. A discussion of current application trends in diffractive optics may be found in [2] and [3]. As discussed in these articles, applications of diffractive elements are currently classical spectroscopic gratings, security features in banknotes and other documents, various high-performance optical instruments, semiconductor manufacturing, and intra-ocular lenses. Patent literature shows increasing interest of companies in the application of diffractive optical elements for imaging lenses, e.g. in photography.

The goal of this article is to present an addition to the edited work [1] and to give an overview on recent research literature on tunable submicron-structure optical elements based on soft matter and its characteristic effects. In contrast to [2] and [3], the studies cited in this article report of elements in a laboratory status, but not in applications relevant for industry.

This article is intended both as an introductory tutorial and a short review. In section 2, I give a

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concise overview on the material basics and the fundamentals of the tuning effects. Section 3 presents an overview on the state of the field as found in scientific literature. The article closes with some concluding remarks and the references.

2 Basics

This section briefly reviews materials and mechanisms suited for manufacturing and tuning of optical elements relying on submicron structures.

2.1 Soft matter

The term soft condensed matter describes matter in a condensed state that is neither a simple liquid nor a crystalline solid [4]. This definition has a twofold consequence. Firstly, soft matter is usually not treated in a typical university course on solid-state physics. Nevertheless, the last decades have witnessed an enormous research activity in this field, and several excellent textbooks on this field exist. As my personal favourites, I would like to recommend tot he interested reader those by Israelachvili [5], Evans and Wennerström [6], Chaikin and Lubensky [7], Davis [8], and Jones [4]. Secondly, the term “soft matter” covers an enormously wide variety of natural, artificial, as well as technical matter and materials, among them blood, milk, juices, cosmetics, tyres, paints, lubricants, adhesives, detergents, and liquid crystals in displays

To dare a classification of soft condensed matter from a material point of view, we may group it into several sub-species:

- *Colloidal suspensions* are complex liquids and consist of dispersed liquid or solid particles with a typical size in the submicron range in a second liquid. In contrast to atomic systems, the interaction potential between the dispersed particles can be tailored in colloids.
- *Polymers* are macromolecules consisting of small molecular sub-units called monomers. The connectivity and the molecular weight of a polymer lead to phenomena that markedly differ from that of both simple liquids and crystalline solids.
- *Liquid crystals* are molecules exhibiting an anisotropic molecular shape. By this anisotropy, a number of states with an order in between an isotropic liquid and crystalline solid occur.
- *Surfactants* are amphiphilic molecules, which are able to aggregate into more complex systems, such as micelles, vesicles, and membranes.
- *Composites* usually consist of a soft or polymeric matrix with embedded solid particles. These particles dramatically influence various properties of the composite material, such as mechanical, electrical, magnetic, or optical properties.

Among these, the research studies reported in this article concentrate on polymers and composites. Liquids, in contrast, are of lesser utility for optical elements based on submicron structures, but are regularly applied for tunable refractive optical elements.

2.2 Materials

Since many optical components in real-world applications as well as research studies reported here are manufactured from entirely or in essential parts from polymeric materials we recall some essential material facts of polymers to take into account in any optical application. Generally, when taking about polymeric optical materials, one should distinguish between polymer melts and cross-linked polymers. A *polymer melt* or *thermoplastic polymer* consists of individual polymer chains (in most cases of different length) that are not connected by covalent bonds. The resulting solid material is thus an undercooled melt rather than a truesolid and exhibits a glass transition temperature instead of a melting point. Upon an increase in temperature, the polymer melt undergoes a glass transition, i.e., its shear viscosity decreases by several orders of magnitude.

This behaviour makes polymer melts to ideal candidates for shaping processes such as injection molding and hot embossing. The microscopic structure of a polymer melt can exhibit both amorphous and partly crystalline regions, in which the polymer chains are aligned to each other. It should be noted here that the microscopic structure of a solid polymer material strongly depends on its fabrication process (extrusion, molding). Thus, the optical properties of a polymer component and the refractive index in particular exhibit a dependence of the material used (e.g., extruded or cast PMMA). Furthermore, in some fabrication processes, such as laser machining, extruded and cast polymer materials show different behaviours. Material examples of polymer melts relevant to optical applications are those used for fabrication of most plastic optical components, i.e. polymethylmethacrylate (PMMA), polycarbonate (PC), and polystyrene (PS). Upon exposure to a solvent, a polymer melt is dissolved. In crosslinked polymers or duroplastic polymers, in contrast, the polymer chains are connected by covalent bonds. This crosslinking generally involves a chemical reaction. Thus, when exposed to a solvent, a crosslinked polymer sample can swell, but is not dissolved. Upon a temperature increase, it does not undergo a glass transition, i.e., it does not get softer. A notable example of this class of polymers is silicone rubber. Typically, these silicone rubbers are shipped as two-component liquids, which are mixed and subsequently crosslinked at elevated temperature to result in an elastic solid material.

The refractive index *n* represents one the most important optical material properties and it should be stressed that it depends on wavelength and on temperature. Typical figures for the refractive index of polymers are $n \approx 1.4\text{-}1.6$ and vd ≈ 30-60 for Abbe numbers. Examples formeasured data of refractive indices of several optical standard polymers (PC, PMMA, PC) are plotted in Fig 1, together with data of several liquids for later reference.

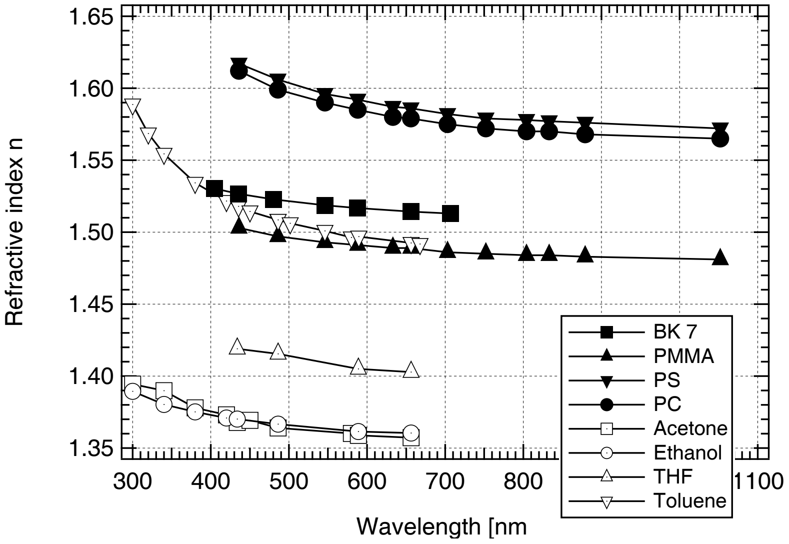

Fig 1. Refractive index as a function of wavelength for BK 7 optical glass, some frequently used optical polymers (PMMA, PS, PC), and some organic solvents.

The influence of temperature *T* on the refractive index *n* is, in general, more pronounced in the case of soft matter, including polymers, than for inorganic glasses. A quantitative expression for dn/dT may be obtained from the Lorentz-Lorenz-, or Clausius-Mossotti formula [9] by implicit differentiation. The result is:

$$\frac{dn}{dT} = \frac{(n^2 - 1)(n^2 + 2)}{6n} \times \left[\frac{1}{\alpha}\frac{\partial\alpha}{\partial T} - CTE\right] \tag{1}$$

Here, α is the molecular polarizability, and CTE the coefficient of thermal expansion, which is defined by the molar volume V_m or by the mass density ρ in the following way:

$$CTE = \frac{1}{V_m}\frac{\partial V_m}{\partial T} = -\frac{1}{\rho}\frac{\partial \rho}{\partial T}$$

By that, the mass density ρ as a function of temperature is $\rho(T) = \rho_0\ (1 - CTE \times \mathrm{T})$. Equation (1) shows that the temperature dependence of the refractive index is composed of the difference of two temperature-dependent quantities and thus is given by an interplay between them. In result, the refractive index of materials with a high CTE typically decreases with temperature. Many polymers exhibit a negative thermo-optical coefficient, i.e. $dn / dT < 0$. Numerical data for various polymers as studied by waveguide devices may be found in a study by Moshrefzadeh *et al* [10].

For later reference let us finally consider a mixture of two components, e.g. a solvent and a polymer, with number densities N_1 and N_2, and a molecular polarizability of α_1 and α_2,

respectively. The total molecule number density N_{tot} of the mixture is then $N_{tot} = N_1 + N_2$, and the total molecular polarizability α_{tot} is given by

$$\alpha_{\mathrm{tot}} = \frac{N_1}{N_1 + N_2}\alpha_1 + \frac{N_2}{N_1 + N_2}\alpha_2 = \phi_1\alpha_1 + \phi_2\alpha_2$$

in which ϕ_1 and ϕ_2 denote the volume fraction of component 1 and 2 in the mixture, respectively. From that, the refractive index of the mixture n_{tot} may be calculated from the refractive indices of the components, n_1 and n_2, by

$$\frac{n_{\mathrm{tot}}^2 - 1}{n_{\mathrm{tot}}^2 + 2} = \phi_1\frac{n_1^2 - 1}{n_1^2 + 2} + \phi_2\frac{n_2^2 - 1}{n_2^2 + 2}$$

2.3 Mechanisms

Polymers represent the material class applied for many tunable submicron-structured optical elements. Thus, we discuss in this section the most important tuning mechanisms and actuation principles for polymer-based optical elements.

Elasticity

The elastic properties of solids (glass, metals, semiconductors) on the one hand and polymeric materials on the other hand significantly differ. Young's elastic moduli for polymeric materials are in the range of approximately 0.001-1 GPa, and those of glass and metals in the range of approximately 50-500 GPa, typically. Thus, plastic optical elements can be detuned in their shape by comparatively small forces, and the elasticity of the optical component itself may be used for tuning their optical properties. For these purposes, standard force actuators, such as voice coils, may be employed.

Elastic stretching as a tuning mechanism seems quite straightforward from a conceptual point of view. Moreover, Hookeian elasticity theory has been known for decades. However, accurate modelling of elastic optical elements is all but easy. Severe difficulties arise from – firstly – the lack or availability of accurate material models taking into account even hyper-elastic effects and – secondly – from the fact that the properties of an individual plastic optical component strongly depend on the batch of base material used and the subsequent manufacturing process. Moreover, effects like stress birefringence are highly difficult to implement in detail in standard construction and optical design software tools.

Electric interactions

For many applications actuation principles based on electric signals are highly advantageous. Regarding polymers, there is a broad variety of electro-mechanical effects that may serve as actuation principles in tunable optical components.

In the simplest configuration, an elastic polymer In the simplest configuration, an elastic polymer with a relative dielectric permittivity ε and a thickness *t* may be used as the dielectric in a parallel-plate capacitor. Upon applying a voltage *U*, the electrodes exert a pressure *p* onto each other, which is given by

$$p = \quad \varepsilon\varepsilon_0 \frac{U^2}{t^2} \tag{2}$$

Under this load a compliant dielectric is compressed in longitudinal direction (electrostriction) and expanded in transverse direction, and the transversal expansion is described by Poisson's number. Both the transverse expansion and the longitudinal compression may be applied as electro-mechanic actuation principles for tunable optical components.

Clarifying the details of the electro-mechanic behaviour of soft matter requires in-depth knowledge of the molecular composition, and thus a number of different effects and material classes can be distinguished. Regarding the effects, the inverse piezo-electric effect involving a molecular re-ordering upon applying an electric field should be distinguished from mere electrostriction for research success in this field. A clear application perspective, in addition, will to the author's viewpoint, be the sufficient condition for success not only in research, but also in industrial applications. Clarifying the details of the electro-mechanic behaviour of soft matter requires in-depth knowledge of the molecular composition, and thus a number of different effects and material classes can be distinguished. Regarding the effects, the inverse piezo-electric effect involving a molecular re-ordering upon applying an electric field should be distinguished from mere electrostriction as described in the preceding paragraph. In the field of polymers, polyvinylidene fluoride (PVDF) is known for its strong piezo-electric material properties [11]. Electro-active polymers [12] generally find enormous interest as electro-mechanical actuators and artificial muscles. Liquid crystal elastomers, [13,14] which combine the ordered phases of liquid crystals with the excellent elasticity properties of elastomers, currently find even applications for tunable optical elements [15].

Swelling

Swelling may be understood as a mutual diffusion process: Solvent molecules diffuse into the polymer phase, and polymer chains reach out into the solvent phase. Upon swelling, the volume of the polymer sample increases and its refractive index changes. The solvent might consists of small molecules or un-crosslinked polymer chains. Swelling has an enormous technical relevance, mostly in the context of material failure and reliability issues, such as failing glue bonds and delamination of compound materials.

Throughout this article, index 1 refers to the solvent, and index 2 to the polymer. By an index 0, a quantity referring to the un-swollen state is understood. A scientific discussion of swelling may be found in textbooks on polymers, such as the classic text by Paul Flory [16], or more recent textbooks [17, 18], and in comprehensive articles, for example [19]. For a modern discussion of polymer swelling, including theories accounting for different network models, see [20].

The change in refractive index has already been discussed in the preceding section, so let us consider here the volume increase of a polymer sample upon swelling. For modelling the swelling effect the number of dimensions d, into which a polymer sample is free to expand, is an important figure: For a free volume sample, $d = 3$, and for surface attached thin films, $d = 1$. Depending on the number of dimensions in which the polymer is free to swell, the volume increase is described by a linear swelling degree α or a volumetric swelling degree S defined by

$$\alpha = \frac{h}{h_0},\ S = \frac{V}{V_0},$$

in which h_0 and V_0 are the film thickness and the volume, respectively, in the initial state, and h and V those in the swollen state. Instead of the thickness of a polymer film h, any other characteristic length scale describing a polymer microstructure may be used, for example the lattice constant in a periodic structure. Using the volume fraction of the polymer in the swollen state, ϕ_2, the linear and the volumetric swelling degree may be expressed by

$$\alpha^{\mathrm{d}} = S = \frac{V}{\phi_2}.$$

For a theoretical description, a lattice model of the polymer may be used, in which the sites of the lattice can either be occupied by a monomer unit or a solvent molecule. Additionally, certain assumptions on the crosslinks of the polymer chains have to be made. In the simplest model, the Flory-Huggins sorption model, elastic contributions to the total free energy of the polymer are neglected, and only the contribution of mixing of the polymer chains with the solvent molecules are calculated and equated to the free energy of an ideal gas as a model for the solvent. The result is:

$$ln \frac{P}{P_0} = ln\phi_1 + (1 - \phi_1) + \chi(1 - \phi_2)^2\ , \tag{3}$$

in which χ is the dimensionless Flory-Huggins interaction parameter describing the specific interaction of between polymer segments and solvent molecules in a particular polymersolvent system. It should be noted that χ is dependent on the solvent concentration; concentration-dependent values of χ for specific polymer-solvent systems are given in [21]. A more detailed discussion of χ may be found in [22]. As long as the elastic contribution to the free energy is small compared to the mixing contribution, Equation (3) does describe swelling well. For purposes of analysing swelling submicron-structured optical elements, Eq (3) may be transformed into

$$\frac{P}{P_0} = \left(1 - \frac{1}{\alpha^d}\right) \times \exp\left\{\frac{1}{\alpha^d} + \frac{\chi}{\alpha^{2d}}\right\} \tag{4}$$

Regarding the free interface of a crosslinked polymer film upon swelling, it is found that experimental data may be explained well by assuming a polymer volume fraction ϕ_2 described by an error function [23, 24]. An additional contribution to the free energy due to elastic stretching of the crosslinked polymer upon swelling is considered in the Flory-Rehner theory of swelling [25, 26], which has to be applied if large swelling degrees ($S \approx 10\ldots100$) are considered [27].

3 State of research

This section gives an overview on tunable optical components based on soft materials and characterized by a periodic submicron structure in at least one dimension. This includes gratings, multilayer Bragg structures, photonic crystals in two and three dimensions, and photonic bandgap fibers. Micro-electro-mechanical diffractive optical systems fabricated in semiconductors are, however, not in the scope of this article.

3.1 Tunable gratings

Optical gratings are among the oldest micro-optical components and have traditionally been fabricated by precise mechanical ruling of a metal layer on a glass substrate, and by laser interference lithography (LIL). In LIL, two expanded laser beams intersect at a certain angle, and a photosensitive material, in particular a photoresist, is exposed to the resulting interference pattern. Subsequent process steps are developing of the photoresist and pattern transfer by etching. LIL is a highly versatile process, and the superposition of three or four beams even allows to generate three-dimensional patterns for photonics crystals [28].

Fabrication of optical gratings in soft materials opens up the possibility of tuning the grating period by a number of effects as discussed in the preceding section. A straightforward tuning method for gratings made from elastic materials is mechanical distension [29]. In [29], the authors fabricated distensible viscoelastic phase diffraction gratings with triangular and rectangular profiles in a commercial silicone elastomer (Dow Corning Sylgard 184). The gratings in this study had a thickness of 0.4 mm and periods 4.46 µm and 32 µm, respectively. The angular shift for the ±1st diffraction order upon mechanical elongation was ≈ 1° in this study, and the grating could be driven periodically with a maximum frequency of 1 kHz. Up to a relative distension of 18% the authors found no effect on the diffraction efficiency. Generally, however, since in this approach not only the period but also the grating profile is changed upon elongation, the angular shift of the diffraction orders is accompanied with a change in the diffraction efficiency, too.

Similar studies report of a diffraction grating written in a stretched azobenzene elastomer film by UV irradiation [30]. By stretching the material, the liquid-crystalline azobenzene mesogens align. Upon irradiation with UV light, this stretching-induced orientation is erased again due to a cis-trans-photoisomerization and may even be recovered by a subsequent irradiation. The grating in this study consisted of oriented and isotropic domains in the elongated film. The grating was reported to be still present after relaxing the mechanical strain, and the period of the grating was narrower in agreement to the stretching ratio.

As discussed above, electrostriction is an interesting effect since compression in one direction leads to an expansion of the sample perpendicular to this direction. Many elastomers exhibit a Poisson's number close to 0.5, which means that the sample's volume is conserved when compressed. Thus, by applying a voltage to an elastomeric material between two compliant electrodes, a lateral distension of the elastomeric material can be accomplished.

Electrostrictive polymers have been shown to be capable of efficient and fast response with high strains (up to 30-40 %) and good actuation pressures (up to 1.9 MPa) [31]. The effect may even be improved by pre-straining the polymers, and actuated strains of up to 117 % with silicone elastomers and up to 215 % with acrylic elastomers, using biaxially and uniaxially pre-strained films, have been measured [32]. For an overview of this technology and its possibilities, including considerations for the different suitable polymers and their material properties, see [33].

Electrostriction has been demonstrated as a suitable effect for tuning of diffraction gratings. In these devices, a pre-strained elastomer film featured with a diffraction grating is compressed by electrostriction, leading to lateral expansion or compression of the grating period, depending on the design of the device. Actuation voltages are in the range of 0-5 kV. Typical strains of up to 20 % are achieved, thereby exceeding the value achieved with piezoelectric actuation by a factor of approximately 100 [34-36]. This technology is currently commercialized by the company Optotune, Switzerland. In the elastomeric grating device presented in [34], for example, the grating period is expanded as voltage is applied. Here, the measured grating period, Λ, as a function of voltage, U, is perfectly described by the equation

$$\Lambda\,(U) = \frac{\Lambda_0}{\sqrt{1 - \varepsilon\varepsilon_0 \dfrac{U^2}{Et^2}}},$$

which readily is derived from Eq (2). This equation requires that the elastic strain in the film is small and isotropic. In this equation, Λ_0 is the original period of the grating, E is Young's modulus of the elastomer film, t its thickness and ε its relative dielectric permittivity. Similar studies on elastic gratings report of compression of optical transmission gratings [37] on electro-active elastomeric substrates and on reflective elastic gratings, which were rendered reflective by silver nanoparticles on the originally transparent polydimethylsiloxane (PDMS) elastomer [38].

Besides electrostriction, swelling in solvents may also be used to change the period of transmission phase gratings manufactured in elastomers. As an example, Fig 2 shows experimental data of a freely suspended silicone elastomer membrane with a surface phase grating swelling in organic solvents. The grating had a period of Λ_0 = 1.14 µm, a thickness of t = 50 µm and was fabricated in Dow Corning Sylgard 184 silicone elastomer. The actual period of the grating was measured by the angle of the 1st diffraction order, and the corresponding solvent partial pressure (normalized to the saturation pressure at laboratory temperature, p_0) was determined from the dew point of the atmosphere (nitrogen plus solvent vapour) surrounding the grating. As can be seen from the figure, for both solvents used (toluene and *n*-heptane) the experimental data are perfectly matched by the Flory-Huggins sorption model as was used in Eq (4). Advantageously, the diffraction angle does depend on the period of the grating only, and not on the refractive index of the grating. Thus, a change in refractive index had not to be considered in the analysis of the data. Linear swelling degrees corresponding to a relative expansion of the grating period in the range of 20-30% could be achieved. The swelling process in this experiment is perfectly reversible and repeatable. It should, however, be noted that fast tuning could not be achieved in this experiment since it requires thermodynamic equilibrium to be described by the Flory-Huggins model, and since dynamics is controlled by slow diffusion processes here.

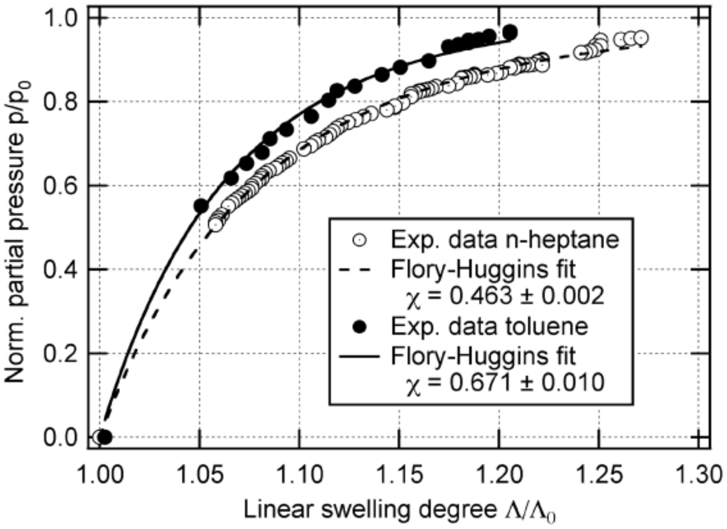

Fig 2. Experimental data of swelling of a freely suspended 2d surface phase grating in nheptane and toluene, respectively. The solvent partial pressure of the nitrogen-solvent gas mixture is normalized to the saturation pressure, and the linear swelling degree is expressed as the fraction of the actual (swollen) and the original (un-swollen) grating period. Experimental data are perfectly explained by the Flory-Huggins sorption model.

3.2 Tunable optical Bragg devices

Optical Bragg structures consist of a periodic stack of dielectric layers with an optical thickness (nt) equal to a desired fraction of the operation wavelength λ_0. For example, a Bragg mirror is made up of an alternating stack of layers of high (n_2) and low (n_1) refractive index, each layer exhibiting an optical thickness $n_1t_1 = n_2t_2 = \lambda/4$. Their high mirror reflectance, which routinely exceeds that of metal mirrors, and the freedom of designing the transmission characteristics to desired specifications have predestined them for a number of extremely demanding optical applications, such as wide-field imaging spectroscopy in astronomy, sensors, or as line filters for optical telecommunications, Raman spectroscopy, and biophotonics. As a further essential advantage, Bragg filters may be tuned by introducing mechanisms for changing the thickness or refractive index of the multi-layer structures.

The dominant material class for the fabrication of all types of high-performance optical Bragg structures, including tunable structures, are semiconductors and inorganic dielectrics because highly accurate thin film deposition processes are well established for these materials. Tunable filters have been demonstrated in all-silicon technology [39] as well as in III-V material systems [40]. III-V-based vertical cavity surface emitting lasers (VSECLs) and reflective cavity LEDs (RCLEDs) are also based on Bragg filters [41, 42]. By the multilayer structure of Bragg mirrors in VCSELs, the mirror's reflectance spectrum can be adapted to the gain profile of the semiconductor material in the cavity. A thin optical cavity layer, moreover, facilitates single longitudinal mode operation.

Since the accuracy of layer thickness definition in typical liquid processes such as spin coating or dip coating is not inferior to many semiconductor deposition processes one may wonder why the number of studies of polymer multilayer fabrication is so small. This fact may perhaps be attributed more to the materials than to the lacking accuracy of the processes. Studies in this field include the fabrication of polystyrene/collodion and polystyrene/polyvinylalcohol multilayers by spin coating [43], or polymer co-extrusion [44-46]. Polymer co-extrusion has been demonstrated as an efficient tool for the fabrication of interference filters consisting of up to 1,000 layers, and this method, though requiring specialized equipment, was exploited for incorporation of layer thickness variations in order to fabricate giant-birefringence structures. Polyelectrolytes, deposited in a layer-by-layer process together with in-situ growth of silver nanoparticles, have successfully been used for the fabrication of rugate filters in which the continuous and periodic refractive index profile was approximated digitally [47]. In this study, poly(allylamine hydrochloride) (PAH) and poly(acrylic acid) (PAA) bilayer regions, in which the subsequent nanoparticle growth reaction was performed, made up the high refractive index regions, whereas unmodified PAH and poly(styrenesulfonate) (PSS) bilayer regions served as the low index regions of the rugate structure.

More interesting than the mere fabrication are the possibilities of transmission wavelength tuning achieved with soft materials. In the cited early study by Kimura *et al* [43], mechanical compression was already shown to be a first example of a tuning mechanism. If the thickness of the layers is much smaller than the wavelength of the light, the transmitted light experiences an effective index of refraction. This effective index has been shown to be tunable by mechanical compression, too [48], in a multilayer film consisting of 1024 alternating layers of an elastomer and a glassy polymer.

Other tuning mechanisms comprise the application of external electric fields to Bragg filters fabricated using poled polymers [49] or liquid crystals for the cavity layer [50] or liquid crystal micro-domains (droplets) separated by cross-linked polymer layers fabricated by holographic photo-polymerization [51]. In these structures, applying of an electric field leads to a change of the transmission spectrum by a field-induced change in the refractive index of one or more layers due to orientation, or the third-order nonlinear optical Kerr effect, as investigated in recent theoretical study [52]. The systems considered in this study are multilayers from composite-composite and composite- dielectric constituents, in which the composites itself consist of metallic nanoparticles dispersed in a dielectric host, leading to a strong third-order nonlinearity in response to externally applied electric fields, allowing extensive and precise tuning of the optical bandgap in one-dimensional photonic bandgap crystals (PBCs) fabricated from these materials.

Finally, optical Bragg filters consisting of alternating multilayers from UV-photo-crosslinked poly(methyl-methacrylate) and poly(styrene) showed a shift of their filter peak wavelength of as much as ≈ 600 nm to higher wavelengths in a nitrogen atmosphere saturated with organic solvents [53]. The materials in this study were chosen such that both polymers swell to approximately the same degree in organic solvents, thereby maintaining the $\lambda/4$-condition for the optical thickness at all values of the linear swelling degree α. It was demonstrated with these structures that polymer swelling with solvents is a robust and sensitive tuning principle for multilayer Bragg structures, which may even be extended to 3d Bragg structures [54].

Conversely, Bragg multilayer system in which only one type of layer swells have been presented, too [55,56]. Kang *et al* [55] used a hydrophobic block-hydrophilic polyelectrolyte block polymer that forms a simple lamellar structure acting as a 1d Bragg device. In this material system, the glassy hydrophobic layer forces expansion of the hydrophilic layer along the layer normal upon swelling in the presence of an aqueous electrolyte. By variation of the salt concentration of the aqueous electrolyte, the authors reported of a shift of the mirror stop band position from (λpeak = 364-1627 nm. Similarly, Karaman *et al* [56] reported of a Bragg system consisting of alternating organic (poly(2-hydroxyethyl-methacrylate, pHEMA, $n = 1.51$ in dry state) and inorganic (titania, $n = 1.81$) layers. In the presence of water, only the polymer swells in this system, thereby forming a hydrogel, while the inorganic titania layers are not affected by water.

Due to a physical or chemical response of the polymer layers to the presence of certain molecules, their concentration may be measured by the optical reflection or transmission spectrum obtained from a polymer Bragg structure, i.e., responsive polymer Bragg structures may be used as optical sensor elements. A number of studies relying on this mechanism exist: For example, a Bragg structure consisting of polyelectrolyte multilayers was reported for gas sensing [57]. In this study, each Bragg layer itself consisted of several tens of polyelectrolyte double layers because of the small layer thickness increment of only a few nanometers achieved in a single polyelectrolyte dip-coating step. Rubner *et al* fabricated Bragg structures from polyelectrolyte multi-layers with alternating porous and fully dense regions. Chemicals adsorbed in the pores change the transmittance spectrum, which allows monitoring of their concentration [58].

3.3 Tunable photonic bandgap structures

Tunable and variable photonic bandgap crystals (PBCs) would be interesting optical devices for tunable semiconductor light sources, or for optical circuits with integrated modulators or switches. In this context, John and Florescu have proven theoretically the all-optical transistor functionality in a PBC [59]. In the following sections, we will present the state of the research in soft matter based tuning mechanisms for PBCs.

Pore infiltration of PBCs

To achieve tunability of the photonic bandgap in a PBC, a frequently employed mechanism is infiltration of the pores with a liquid, often a liquid crystal (LC). The physical background of this mechanism is the possibility of controlling the refractive index of the LC by an external electric field (thereby influencing the orientation of the LC molecules) or by temperature (thereby influencing the ordered phase). The theoretical foundation for bandgap tuning by liquid crystal infiltration was laid in a study of Busch and John [60]. They showed that a inverse opal structure from silicon with LC filled pores exhibits a complete and tunable optical bandgap.

In experiment, this effect was demonstrated by Leonard *et al* [61]. They used a PBC from macroporous silicon with the pores filled with E7 LC, and tuned the refractive index of the LC by changing the temperature. The same mechanism has also been observed in PBCs fabricated from III-V semiconductors [62]. Kubo *et al* [63, 64] exploited the rich phase diagram of LCs for switching the PBC by thermally induced or photo-induced isothermal phase transitions. Their samples were LC infiltrated, inverse opaline PBCs fabricated from silicon dioxide. Alternatively, the refractive index of a nematic LC may also be influenced by application of an external electric field for electric bandgap tuning [65]. Based on this electric tuning mechanism, Escuti et al. have measured the birefringence as a function of the electric field at a LC-polymer composite [66]. The application of tunable 1d and 2d PBCs with LC infiltration for optical switches have been demonstrated by Weiss *et al* [67, 68].

Similarly, the use of a microfluidic network for tuning of PBCs, by deliberate filling of a row of holes with liquids, was demonstrated in the group of D. Psaltis at California Institute of Technology [69]. In this study, the authors present a complete opto-fluidic microsystem comprising fluidic, photonic, and control components. For the liquids they used aqueous solutions of surfactants and inorganic salts.

Pore infiltration is a useful concept for bandgap tuning in photonic bandgap fibers, too. As an example, Haakestad *et al* reported of tunable bandgap guidance in solid-core photonic bandgap fiber, two holes of which had been filled with a nematic liquid crystal. Upon application of an electric field, the bandgap guidance in the fiber could be tuned, and response times in the millisecond range were found [70]. A periodic series of liquid plugs in the cladding holes of a photonic bandgap fiber has been demonstrated to act as a fiber Bragg grating. Upon compression of the distance of the liquids plugs, tunability was achieved in this configuration [71].

Refractive index tuning of PBCs

Except from tuning the refractive index of the filling material in the pores of a PBC structure, the PBC structure itself may be influenced by a number of effects. The effects listed here change the refractive index of the PBC structure and thereby increase the refractive index difference between the structure and the voids in it. Although these effects do not rely specifically on soft matter, they are listed here for completeness' sake.

Tuning of the PBC by magnetic fields relying on the Faraday and Voigt effect for influencing the refractive index of a PBC was reported of in [72]. The piezo effect was exploited by Aoki *et al* [73]; this group fabricated thin PBC films from piezo-electric PLZT ceramics and measured the PBG as a function of the applied electric field. Similarly, ferro-electric tuning of piezo-ceramic PBCs [74] and thermo-optic tuning of 2d silicon-based PBCs [75] have been demonstrated experimentally. In all these effects, the refractive index may be varied typically by up to 10%.

A tunable PBC system based soft-matter has been reported by Escuti *et al* [75]. The photonic crystal from this group consisted of LC-droplets arranged in a fcc lattice and embedded in a polymer matrix. Upon application of an electric field, the authors managed to shift the wavelength of the ± 111 stop band reversibly by 2 % [76]. In general, however, it seems not reasonable to fabricate a PBC structure from soft matter because of the small difference in the refractive indices of the material itself (n = 1.4-1.7, typically) and of the air in the voids. By that, only small bandgaps in certain directions of the reciprocal lattice of the crystal may exist, but a complete bandgap (i.e., in all directions) does not open up.

Lattice constant tuning of PBCs

The lattice constant of a PBC, too, can be influenced by several physical effects. In contrast to the effects in the preceding section, the effects reported here are clearly located in the realm of soft condensed matter, the most popular configuration appearing to be composite systems consisting of an ordered colloidal crystal embedded in a polymeric matrix.

A first example for lattice constant tuning effects is the elastic deformation of a photonic crystal by application of mechanical stress. Feasibility studies of this concept have been presented by several groups. [77, 78]. For example, in the study [78] the authors simulated tuning of the PBG of a silicon PBC embedded in a distensible elastomeric PDMS matrix by mechanical stretching of the lattice period. Experimentally, this effect was demonstrated by Fudouzi and Sawada using a colloid crystal PBC embedded in a elastomeric matrix and on an elastomeric support, respectively [79]. Upon stretching of the sample they observed a shift in the reflectance spectrum. Similarly, Arsenault *et al* have demonstrated experimentally the reversible shift of the reflectance peak of elastomeric PBCs by as much as 100-200 nm through elastic deformation of the lattice constant [80].

An elegant way of influencing the lattice constant of a polymer-based PBC is by external chemical stimuli resulting in swelling of the PBC. Depending on the involved materials and stimuli, a volume increase of the structure by a factor of up to 10 may be accomplished, thereby changing the lattice constant by a factor of approximately 2, corresponding to 100 %. At the same time, the refractive index of the material is changed upon swelling due to the molecules ingested by the polymer network. Compared with the change of the lattice constant, the change of the refractive index upon swelling is of subordinate importance. Polymer swelling has been demonstrated by several groups as a viable effect for tuning the optical bandgap of 3d PBCs. Earlier examples of this mechanism are the studies of Fudouzi and Xia [81], and of Foulger *et al* [82], both demonstrating a reversible change of the reflection spectra from an array of an ordered colloidal crystal embedded in a crosslinked polymer matrix upon swelling in suitable solvents, or by Edrington *et al* reporting of swelling with homopolymer chains [83]. Later on, the Foulger group sandwiched a film (thickness 80 µm) of their photonic bandgap composite system between two electrodes [84]. Upon application of a dc voltage (0.5-3 kV, typically), the compliant system is compressed, leading to a shift in the reflection spectrum. By this configuration, the mechano-chromic behavior of the composite was coupled to an electrostatic actuator, allowing easier control compared to solvent concentration. Photonic crystals fabricated from hydrogels have been demonstrated to react both to temperature and humidity in the surrounding atmosphere by swelling. [85].

More recently, Arsenault *et al* [79] have manufactured PBCs from self-assembled silica bead colloidal crystals embedded in a matrix of lightly crosslinked poly-ferrocenyl-silane (PFS). The swelling degree of this metallo-polymer depends on the solvent and on the oxidation state of the polymer backbone. As solvents, methyl-cyclohexane, benzonitrile, chlorobenzene, and dichloromethane were used [86, 87], and a completely reversible red-shift of the reflectance peak by about 100-200 nm, depending on the solvent and its partial pressure, was measured. For the simulation of the reflectance spectra describing the structural colour of the PBCs, the group employed the scalar wave approximation method [88]. In a further study [89], the same group reported of the dynamic electrical tunability of their system based on the continuously tunable state of oxidation of the iron atoms in the metallo-polymer backbone: When a oxidative potential is applied to the composite PBC, electrons are drawn out of the iron atoms in the PFS backbone, and, to compensate the resulting positive charge, anions together with their solvent shell from the surrounding electrolyte penetrate the polymer network. Both the anions and the solvent molecules cause the polymer network to swell, thereby increasing the distance between the layers of the colloid beads. As a result, a potential-dependent red-shift of the reflected Bragg peak is observed. Based on this mechanism, the authors successfully demonstrated the possibility of using their system in full-colour displays.

Further effects that may be employed for tuning of the optical bandgap and Bragg reflection, respectively, of composite systems consisting of an ordered colloidal crystal in a polymeric matrix comprise optical switching by embedding dyed particles [90], or magnetic tuning by embedding particles containing nanoscopic inclusions of ferromagnetic cobalt ferrites in a poly(acrylamide) network [91]. Conversely, these systems may be regarded as sensor materials providing a measurable optical effect upon certain stimuli [92, 93].

Several groups have succeeded in applying the rich phase behaviour of polymeric systems for influencing the reflection spectrum of optical Bragg structures. Examples comprise lightinduced order-disorder phase transitions based on photochemical processes [94, 95], the reaction of regular arrangements from poly(N-isopropyl-acrylamide) hydrogel nanoparticles on external stimuli such as temperature and pH as presented by the group of L A Lyon (Georgia Institute of Technology)[96-97], or a temperature-induced lamellar ordering transition in a complex solid block- copolymer system exhibiting unusually long lamellar structures responsible for Bragg reflection in its ordered state [98].

4 Concluding remarks

To summarize, researchers from all over the world have shown considerable creativity in conceiving and demonstrating tunability of submicron-structured optical elements. Many of these make use of soft matter as their key constituent, bringing along with it the possibility of exploiting novel effects for tuning. Not all of them appear as reasonable to implement in industrial products. From the numerous interesting studies cited in this article, it appears that close collaboration of preparative organic chemistry and optical engineering is the necessary condition for research success in this field. A clear application perspective, in addition, will to the author's viewpoint, be the sufficient condition for success not only in research, but also in industrial applications.

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