Colors enable interaction and communication between living species in a myriad of biological and artificial environments. While living organisms feature low-power mechanisms to dynamically control color in soft tissues, man-made color-changing devices remain predominantly rigid and energy intensive. Here, architectured composites that display striking color changes when stretched in selective directions under ambient light with minimum power input are reported. The orientation-dependent color change results from the rotation of reflective coated platelets that are embedded in a soft polymer matrix and pre-aligned in a well-defined architecture. The light reflected by the platelets generates structural color defined by the oxide coating on the platelet surface. By magnetically programming the initial orientation and spatial distribution of selected platelets within the soft matrix, composites with strain-modulated color-changing effects that cannot be achieved using state-of-the-art technologies are created. The proposed concept of strain-induced architectured color can be harnessed to develop low-power smart stretchable displays, tactile synthetic skins, and autonomous soft robotic devices that undergo fast and reversible color changes through the mechano-optic coupling programmed within their soft composite architecture.

1. Introduction

Soft materials that quickly change color under external stimulus have been used to create enticing functional devices, including stretchable displays, tactile sensors, camouflaging soft robots, and damage-reporting structures. Fast color shifts in these materials are driven by electrical fields, fluid flow, temperature changes, or mechanical stress through different coupling mechanisms,[1–7] Transitions in molecular configuration have been used in mechano-responsive color-changing polymers,[8–10] whereas electron transfer mechanisms have been exploited to fabricate electroluminescent robotic skins.[11] Soft materials with stress-tunable structural color have also been developed using aligned nanosheets or organic bilayers in a hydrogel matrix, polymer-infiltrated photonic crystals, and liquid crystalline systems.[4,5,12] Although proof-of-concept materials and devices have been successfully demonstrated, the exploitation of these materials in autonomous and energy-efficient bulk devices is currently hindered by the high energy input needed to induce color change, slow speed, irreversibility, and challenges in upscaling the synthesis and manufacturing processes.

In contrast to man-made devices, animals such as fishes, squids, and chameleons have evolved elegant, energy-efficient intracellular structures to dynamically control color for communication, warning, protection, and disguise.[13–17] In some of these animals, iridescent color arises from the constructive interference of rays reflected by lamellar nanostructures present inside specialized cells called iridophores. Changes in color and brightness result from cell-mediated manipulation of the lamellar spacing and orientation of such reflective structures. For example, the Neon tetra fish changes color from blue-green (≈490 nm) to indigo (≈400 nm) by simply tilting highly reflective guanine platelets using the so-called Venetian blind mechanism (Figure 1A,B and Movie S1, Supporting Information).[13] Driven by electrical stimulation of the iridophores, the color changes are reversible and ultrafast. Since the mechanism relies on the incoming light as power source and the reflected rays are reinforced by constructive interference, these animals can generate strong and dynamically tunable color with minimum energy input.

Lamellar architectures in the form of stacked platelets have also been widely explored to enable structural control of the properties and function of synthetic materials. Inspired by the structure of mollusk shells, clay and inorganic platelets arranged in the brick-and-mortar architecture of nacre can be used to significantly enhance the stiffness and fracture toughness of polymer-based composites.[18–22] Beyond mechanical properties, architectured materials with deliberate platelet orientations have been developed to increase the charging rate of graphite anodes for lithium-ion batteries[23] or enable shape changes in plant-inspired morphing structures[24] and soft robots.[25] Compared to the many assembly processes that can
be applied to create structural color, platelet-laden polymers are generally easier to process and can be readily upscaled and shaped into large area, complex geometries.

Here, we design and manufacture elastomer-based composites featuring a programmable platelet architecture that enables fast and reversible color changes upon mechanical deformation without the need of an external power source. Inspired by the dynamic coloration mechanisms of fish, the color of our composites depends on the local orientation and architecture of reflective platelets embedded in the polymer matrix. As opposed to previous mechanisms that rely on shifting the distance between sheets, color changes in our composites arise from the re-orientation of platelets driven by an externally applied mechanical strain. The concept of architectured color introduced in this study is eventually exploited to create highly stretchable soft materials that take different colors depending on the strain orientation and that can sense mechanical forces based on a simple powerless optical readout.

2. Design and Manufacturing of Composite Architecture

Analogous to architectured materials designed for mechanical applications, our color-changing composites exhibit optical properties that are governed by structural features arising from multiple length scales (Figure 1C,D). At the nanoscale, the individual platelets are covered with a layer of high refractive index material to allow for constructive interference of the reflected light. Moving to coarser length scales, the individual platelets are organized in arrays with a deliberate orientation, which is manipulated by the imposed strain to reversibly change the reflected color. Finally, platelet arrays are organized in patterns with specific orientations to allow for the selective platelet tilting depending on the stretching orientation. Such multiscale platelet architecture is embedded in a highly stretchable elastomer matrix that translates the externally applied strain into a tilting motion of the platelets. By tilting the platelets, the applied strain reconfigures the composite architecture thus changing the intensity and the wavelength of the reflected light, following the Venetian blind mechanism. In contrast to the color changes observed in the Neon tetra fish, structural color in our composites is not created by controlling the separation between platelets. Instead, our platelets are coated with a high refractive index layer, the thickness of which is designed to generate the desired structural color as in a 1D photonic crystal.

To illustrate the mechano-optic behavior of composites with architectured color, we take submicron thick alumina platelets coated with a titania layer as model reflective platelets (Figure 2A). Using platelets with distinct titania layer thicknesses, structural colors varying from turquoise to red were selected for the study. The structural color generated by the
Figure 2. Fabrication and mechano-optic characterization of soft composites with programmed platelet architectures. A) Scanning electron microscopy images displaying the alumina platelets (top) and the titania coating on the surface of the platelets (bottom). Adapted with permission. [37] Copyright 2013, Gerhard Pfaff. Requests for permissions for further reuse should be directed the copyright holder. B) Schematics of the magnetic alignment setup used to control the orientation of platelets in the soft composite. With the help of a rotating permanent magnet, composites with one (left) or more (right) platelets can be biaxially aligned within the liquid monomer mixture before fixation via polymerization. C) Wavelength-dependent diffuse reflectance of a soft composite containing out-of-plane aligned platelets as a function of the applied tensile strain. D) Photographs showing the color change of the soft composite upon stretching. Samples contain turquoise-colored platelets aligned in a plane orthogonal to the stretching direction. The insets depict the orientation of the platelets within the composite in the undeformed state and after stretching by 300%. E) Wavelength-dependent reflectance and macroscopic color change of composites containing red and turquoise platelets in orthogonal orientations. The stretching direction selectively activates the preferential tilting of either turquoise or red platelets, leading to a soft composite with anisotropic coloring effects. The photographs show the unstretched sample (middle) and the sample when stretched along the direction that activates the turquoise (left) or the red (right) colors. The squared insets are optical images of the sample in the three different scenarios. We note that the color of the unstrained sample is dominated by the SPIONs and the red platelets. Scale bars: 30 µm (top), 200 nm (bottom) in (A), 2 cm, 100 µm (insets) in (D), 2 cm, 1 mm (insets) in (E).
coated platelets results from the phenomenon of constructive optical interference, which is often found in biological materials, iridescent pigments, and 1D photonic crystals.[30–32] Composites with controlled platelet orientations are manufactured using a previously reported magnetically assisted assembly approach (Figure 2B).[33,34]

For magnetic-assisted assembly, platelets are first coated with small quantities (0.001–0.005 vol%) of superparamagnetic iron oxide nanoparticles (SPIONs) to become magneto-responsive. A set of platelets designed for one structural color can be used alone or combined with other coated platelets to create distinct colors depending on the strain direction relative to the platelet orientation. When only one specific color is desired, platelets of the selected type are suspended in a cross-linkable liquid phase and exposed to a rotating magnetic field in the order of tens of milliTeslas. This enables biaxial alignment of the suspended platelets in the desired orientation, which is eventually fixed through consolidation of the liquid phase (Figures S1 and S2, Supporting Information). For composites with two colors, we use two sets of platelets with distinct thicknesses of the titania coating. To enable selective orientation, the magnetization level of each set of platelets is tuned by changing the SPION concentration on their surfaces. Multicolor composites are then manufactured by first exposing a suspension containing the two sets of platelets to a stronger magnetic field. This results in the biaxial alignment of all platelets in the direction imposed by the external field. In a subsequent step, the mold is rotated by 90° and the applied magnetic field is reduced to selectively re-align only the set of platelets featuring stronger magnetic response. To create highly stretchable composites with single or dual color changes, we use silicones or hydrogels with high failure strain as the reactive cross-linkable liquid phase. This magnetic alignment technology can potentially also be implemented using 3D printing platforms.[35,36]

3. Strain-Induced Color Change

Silicone-based composites containing 1 wt% of turquoise platelets show a remarkable color change from black to bright shades of blue when uniaxially stretched up to 300% under normal white light (Figure 2D and Figure S3 and Movie S2, Supporting Information). In this particular example, a second layer of silicone filled with black pigment particles is attached to the back of the architectured composite to provide the dark background. The observed mechano-optic effect occurs when the composite is stretched along the axis perpendicular to the plane of the biaxially oriented platelets. Optical microscopy of the composite before and after stretching at 300% reveals that the orientation of platelets changes from vertical to horizontal under high strains, suggesting that the increase in brightness arises from the mechanically imposed tilting of the platelets. Like micromirrors, the platelets reflect an increasing portion of the incoming light due to the increase of the reflecting surface area as the composite is stretched. Because high strains are required for platelet tilting, it is crucial to select a highly stretchable elastomer as the composite polymer matrix. Mechanical tensile tests indicate that the presence of platelets does not compromise the stretchability of the composite, which displays a hyperelastic stress–strain response typical of elastomers (Figure S4, Supporting Information). The high durability and robustness of the commercially available elastomer was found to be fully preserved in the composites, which can keep their mechanical response even after subjected to over 100 loading cycles (Figure S5 and Movie S13, Supporting Information).

We quantify the observed mechano-optic effect by measuring the diffuse reflectance of the composite within the visible spectrum at increasing stretch ratios (Figure 2C). The results show that stretching of the composite above 50% strain leads to a distinguishable peak in diffuse reflectance, which increases in height proportionally to the applied strain. Irrespective of the stretch ratio, the reflectance peaks are centered around the wavelength of 520 nm, which is characteristic of the turquoise color. At a strain of 100%, the diffuse reflectance of the composite is as high as 60% despite the relatively low platelet concentration of 1 wt%. This is in line with a previous theoretical analysis of the reflectance of platelet-laden composites, which showed that the reflection by 20 to 40 platelets through the thickness is sufficient to increase the diffuse reflectance of such composites above 40%.[38]

The deformation-induced reflectance becomes orientation dependent if the composite architecture is designed to display two sets of orthogonally aligned colored platelets. We illustrate this directional mechano-optic response using turquoise- and red-colored platelets aligned within orthogonal planes in the same composite (Figure 2E). Photographs of composites stretched along different orientations indicate that the imposed strain predominantly activates the tilting of platelets that are aligned perpendicular to the loading direction. To quantify this selective color change, we measured the reflectance spectra of the composite when progressively stretched along the two relevant orthogonal orientations. The changes in turquoise and red colors of the sample were assessed by analyzing the normalized reflectance under 100% strain for the arbitrary wavelengths of 540 and 670 nm, respectively. Our results show that stretching the composite along the turquoise-activating direction increases the normalized turquoise reflectance ($R_{540}^{100\%}/R_{540}^{0\%}$) by 136% as compared to a reflectance enhancement of only 37% for the red color ($R_{670}^{100\%}/R_{670}^{0\%}$). Likewise, the relative reflectance increase achieved by stretching the composite in the red-activating direction is 1.54 times higher for the red ($R_{670}^{100\%}/R_{670}^{0\%} = 1.99$) as compared to the turquoise color ($R_{540}^{100\%}/R_{540}^{0\%} = 1.29$).

4. Predictive Model and Simulation

To better understand the relation between the applied strain and the change in reflectance, we experimentally measured the rotation of individual platelets during stretching of the composite (Figure 3). This allows us to further test the hypothesis that macroscopic changes in reflectance originate from the strain-induced microscopic variations in the orientation of platelets within the composite. In this experiment, an individual platelet oriented orthogonal to the composite surface was imaged from different perspectives while the macroscopic sample was globally strained along a specific direction (Figure 3A and Movie S3, Supporting Information). The results confirm a clear rotation of an individual platelet if the composite is progressively stretched.
in the direction perpendicular to the platelet surface. With the help of image analysis, this experiment showed that the angle of the individual platelet increases monotonically with the magnitude of the globally applied strain, providing additional evidence of the mechanisms controlling the color change in the composite (Figure 3C).

The experimentally observed correlation between platelet rotation and the applied global strain was further investigated to establish quantitative tools for the design of composites with tunable architectured color. To this end, we propose a simple geometrical interpretation for the strain-induced platelet rotation and compare it with finite element simulations of a representative volume of the platelet-laden composite. Considering first a purely geometrical approach, the rotation of a platelet upon stretching of the surrounding polymer matrix can be interpreted in terms of the Poisson effect. For an incompressible isotropic material, the Poisson ratio is \( \nu = 0.5 \) and a tensile strain \( \varepsilon_x \) applied along the \( x \) direction results in a compressive strain of magnitude \( \varepsilon_y = -\nu \varepsilon_x = -0.5 \varepsilon_x \) in the orthogonal (\( y \)) direction. The expected shrinkage of the polymer matrix along the sample thickness should eventually lead to the rotation of the embedded platelet. Using this simple argument, one can derive the following relation correlating the angle (\( \theta \)) of the embedded platelet and the strain applied to the polymer along the axis normal to the platelet surface (\( \varepsilon_z \)): \[ \theta = \cos^{-1} \left( \frac{\cos \theta_0}{\sqrt{1 + \varepsilon_z}} \right) \], where \( \theta_0 \) is the initial angle of the platelet (Figure S6, Supporting Information).

By comparing the predictions of this geometrical model with the experimental data, we find that the theoretical analysis slightly overestimates the absolute \( \theta \) values but captures
reasonably well the dependence of the platelet angle on the applied global strain.

A more detailed analysis of the platelet angle as a function of the imposed strain can be obtained by performing finite element simulations of the platelet-laden composite (Figure 3B and Movie S4, Supporting Information). To enable a direct comparison with the experimental observations, a composite containing platelets aligned out-of-plane was first strained along the direction that was mechanically loaded in the experiment (x axis). Top and side views of the platelets at an applied strain of 100% show a good qualitative agreement between the simulations and the experimental microscopy images (Figure 3A,B). With the help of these simulations, we also studied the response of the platelets when the global strain is applied parallel to the alignment plane (y axis). Our simulations indicate that this loading configuration does not lead to any significant rotation of the platelets, which explains the anisotropic nature of the strain-induced color change (Figures 2E and 4A). A quantitative comparison between simulations and experiments for composites loaded along the color-changing direction reveals that the finite element analysis correctly captures the experimentally observed trend between the platelet angle and the applied strains.

By quantifying the effect of the imposed strain on the rotation of an individual platelet, we are able to predict the macroscopic change in reflectance resulting from the collective rotation of platelets upon stretching of the composite. For the idealized scenario of perfectly aligned platelets, one should expect the reflectance to sharply rise when the angle of the platelets approaches 90° relative to the viewing angle of the observer. Because manufacturing limitations do not allow for perfect alignment of all platelets, the reflectance of the composite exhibits in fact an angular dependence that results directly from the angular distribution of the embedded reflecting platelets. To demonstrate this correlation, we experimentally measured the reflectance at a fixed observation angle of 45° for a composite with platelets initially aligned at an average angle of 10° and subjected to stretching (Figure 3D). In this experiment, light is also emitted at an angle of 45° with respect to the sample surface. The results show that the reflectance increases significantly for strains higher than 75%, reaching the highest level at an applied deformation of 100%. On the basis of our experiments on individual platelets, strain levels between 75% and 100% lead to platelet angles in the range 37–41° (Figure 3C), which translates into angles of 82–86° relative to the reflectance probe. Such an analysis suggests that the composite reflectance increases strongly as the majority of platelets approach an angle of 90° relative to the viewer. These findings imply that the shape of reflectance curve (Figure 3D) is directly related to the standard deviation of the platelet angular distribution in the polymer. Complementary experiments on similar composites indicate that the reflectance eventually reaches a plateau at very high strain levels (Figure S7, Supporting Information).

5. Potential Applications

Composites with strain-activated architectured colors can potentially be used as smart stretchable displays, color-changing soft actuators, or as passive mechanical sensors with simple optical readout. To illustrate these possible applications, we manufactured and evaluated the functional performance of demonstrators featuring distinct color architectures made with different types of platelets and polymer matrices (Figure 4). Because color is generated under standard white illumination, the unique attractive feature of such demonstrators is the fact that no external power is needed to induce color changes except for the mechanical energy required to control the orientation of the embedded platelets by stretching. This is interesting in applications that already require mechanical energy for another main function, as is the case for soft robots that rely on pressurization for locomotion. Alternatively, the mechanical input necessary for color change might result from direct interactions with the environment or end users, as expected for displays and tactile sensors.

Soft conformable displays that selectively change color depending on the stretching direction were prepared by magnetically aligning coated platelets in a silicone-based composite with programmed multiscale architecture (Figure 4A,B and Movies S5 and S6, Supporting Information). The architecture is designed at the centimeter scale to exhibit different colored patterns upon mechanical actuation in specific directions. The direction leading to color changes is programmed at the micrometer scale by aligning platelets in particular orientations in the silicone matrix. At the nanoscale, the thickness of the titania coating can be tuned to change the macroscopic structural color arising from the microscopic reflective platelets. The implementation of such a multiscale architecture in more complex macroscopic geometries enables the fabrication of air-driven soft actuators that change color when inflated (Figure 4C and Movie S7, Supporting Information). In contrast to the energy-intensive camouflaging strategies that require external electrical power supply,[11] these soft robotic parts can simultaneously morph and change color using just pressure as the single input.

The concept of architectured color implemented in such demonstrators can also be extended to composites with other polymer matrices. We illustrate this by manufacturing composites with orientation-selective color changes using double-network hydrogels as the continuous polymer phase (Figures 4D–F and Figures S8 and S9, Supporting Information). In this case, the hydrophilic nature of the polymer allows for the removal of the SPIONs by a simple chemical etching procedure at the end of the fabrication process (Figure S10, Supporting Information). In two exemplary architectures, hydrogel-based composites were fabricated with red platelets (Figure 4D) or with orthogonally aligned turquoise and red platelets (Figure 4E). Our experiments show samples with the red platelets becoming colorful when compressed, and specimens with two types of platelets selectively change color depending on the shearing direction. The highly transparent composites generate impressive vivid colors only when mechanically sheared (Figures 4D,E and Movies S8 and S9, Supporting Information) or simply rotated (Movie S10, Supporting Information).

The conversion of mechanical stimulus into color changes can be harnessed to create soft conformal patches that optically sense the topography of rigid surfaces. Using stereolithography 3D printing to create a complex 3D rigid surface, we show that
the topography of the rigid object can be sensed through local color variations in the patch when put in contact with the rigid surface and sheared (Figure 4F and Movie S11, Supporting Information). Such a functionality may find use in synthetic skins that could provide robots with proprioceptive skills to sense, self-orient, and interact with unstructured environments.

To evaluate the sensing capabilities of such a light-based tactile skin, we placed one of our composite patches on top of a set of light-emitting and photosensitive diodes and measured the change in reflectance of the material when subjected to controlled mechanical stimuli (Figure 4G–I). Our experiments reveal that the silicone-based composite can sense normal pressures up to 160 kPa through a relative increase in reflectance between 0% and 10% (Figure 4H). The sensitivity of the sensor increases from 0.02% to 0.24% per kPa as the pressure is reduced from 160 to 5 kPa. With a resolution of 3–70 Pa for the pressure range 5–160 kPa, our sensor can detected pressure levels comparable to that of high-resolution resistive and capacitive tactile systems while keeping a fully conformable elastomeric surface.

Importantly, the orientation-dependent color changes programmed within the composite architecture allows the skin to sense the direction of the imposed mechanical stimulus, a feat that is not easily achieved using state-of-the-art synthetic skins. This is illustrated by measuring the
were magnetically functionalized by the adsorption of SPIONs from elastomer (Ecoflex 00–20, parts A and B, Smooth-On, Inc.). The platelets SPIONs (ferrofluid EMG 605, Ferrotec GmbH), and a silicone-based when strained in one direction were manufactured using titania-coated formable reflective devices.

6. Conclusions

Soft composites with a multiscale architecture of aligned platelets can be designed to undergo dramatic color changes when stretched along the direction normal to the alignment plane. The color change arises from the collective tilting of embedded platelets, which work as micromirrors to enable specular reflection of the incoming light. Because the platelets are coated with a layer of high refractive index oxide, the reflected light produces structural color under standard illumination conditions without requiring additional energy sources. The level of platelet tilting, and the associated color change induced by stretching can be explained in terms of a simple Poisson effect and is also well captured by finite element simulations. The platelet architecture within the soft composite can be programmed to create a broad range of stretchable and conformal devices that generate color when mechanically loaded or uses structural color to sense mechanical stimuli imposed by the environment. The proposed concept of strain-induced architected color may find applications in soft robotic actuators, low-power stretchable displays, smart synthetic skins, or conformable reflective devices.

7. Experimental Section

Silicone-Based Soft Composites: Soft composites that change color when strained in one direction were manufactured using titania-coated alumina platelets (Xirallic T60-25 SW Cosmic Turquoise, Merck KGaA), SPIONs (ferrofluid EMG 605, Ferrotec GmbH), and a silicone-based elastomer (Ecoflex 00–20, parts A and B, Smooth-On, Inc.). The platelets were magnetically functionalized by the adsorption of SPIONs from the ferrofluid following a previously reported protocol.[24] In a typical procedure, 200 g of platelets were first added to 4 L of deionized water to form a platelet suspension. In another flask, 7 mL of the ferrofluid was added to 1 L of deionized water to obtain a diluted suspension of SPIONs. The platelets and SPION suspensions were stirred separately and later mixed with one another. The SPION-coated platelets obtained after mixing were eventually removed by filtration after a stirring time of 24 h. The resulting magnetized platelets were then added to the part A of the elastomer to form a suspension containing either 0.25 or 2 wt% platelets, depending on the concentration desired in the final composite. The suspension was mixed in a planetary mixer (ARE-250, THINKY), added to part B of the elastomer and mixed again before pouring into a mold. By leaving the mold close to the rotating field, the platelets were first aligned parallel to the rotating field. Subsequently, the mold was moved 5 cm further from the magnet and rotated by 90°, allowing only the more responsive red platelets to re-align perpendicular to the previously oriented turquoise platelets (Figure S11, Supporting Information). The mold was left in this position for 2 h for complete curing.

A step-wise curing sequence was used to create stretchable soft composites combining specific patterns and directional color-changing effects (Figure 4A,B). In this case, distinct color changes were programmed within different regions of the composites, so that specific patterns were unveiled upon stretching of the sample. This was accomplished by using two sets of platelets in the same composite and aligning them in different directions depending on the position within the sample. To generate a pattern, the suspension of platelets was first cast into a 3D-printed mold with the negative shape of the desired pattern, namely the logo “ETH” or “SDU.” A rotating magnetic field was used to align the platelets in a specific direction while the suspension was cured for 2 h. The resulting composite with the shape of the pattern was removed from the first mold and placed in a rectangular-shaped mold. In the second step, the rectangular mold containing the first cured sample was filled with a second suspension containing either the same platelets (ETH logo) or platelets with a different color (SDU logo). The final stretchable displays (Figure 4A,B) were generated by imposing a rotating magnetic field to align the platelets orthogonal to the first set of platelets, followed by curing of the second suspension.

Hydrogel-Based Soft Composites: Double-network hydrogels and magnetically functionalized platelets were used to fabricate transparent soft composites with architecured color. Hydrogels were manufactured using an experimental protocol adapted from a previously reported study.[25] In a typical sample, a suspension with the following constituents and mass fractions was prepared: 91.81 wt% distilled water, 1.08 wt% alginate (A2033, Sigma-Aldrich), 0.06 wt% MBAA (N,N′-methylene bisacrylamide, 146072, Sigma-Aldrich), 0.25 wt% TPO (diphenyl(2,4,6-trimethylbenzyl)phosphine oxide, 415952, Sigma-Aldrich), 5.80 wt% acrylicamide (A8887, Sigma-Aldrich), and 1 wt% turquoise or red platelets. Composites were prepared by first mixing the suspension in a planar mixer and pouring it into a mold. To align the platelets while polymerizing the acrylamide, the mold was placed inside a box connected to nitrogen supply and equipped with an ultraviolet light source (wavelength 300–450 nm) adapted to TPO, which cures at wavelengths below 405 nm. The box was moved close to a rotating magnet and the suspension was left 10 min directly exposed to ultraviolet light under nitrogen atmosphere. The polymerized acrylamide gel was removed from the mold and left for 1 h in an aqueous solution of 5 wt% CaSO_4 (calcium sulfate, C3771, Sigma-Aldrich) to cross-link the alginate molecules that form the second gel. While the presence of SPIONs makes the soft composites initially opaque, the hydrophilic nature of the cured hydrogels enables removal of the SPIONs from the gelled composites following a previously reported chemical etching procedure.[24] To achieve transparency, the opaque double-network hydrogel was transferred to a solution of 50 wt% phosphoric acid (345245, Sigma-Aldrich) in distilled water for ≈2 h. The final composite was rinsed with distilled water and stored in the CaSO_4 solution.

Air-Driven Soft Robotic Actuators: Soft actuators that change color upon inflation were fabricated via sequential casting of three distinct silicone resins into a 3D-printed mold. The 3D-printed mold was designed to generate a bending actuator with interconnected cavities
(Pneuvents) that can be inflated with air pressure. To simultaneously bend and change color upon inflation, the actuator featured a stiffener silicone on one side and a softer silicone with architectured color on the other side. This complex 3D structure was manufactured by first depositing an uncured layer of the stiffener elastomer resin (Renew Silicone 20, Smooth-On, Inc.) at the bottom of the mold. Next, a silicone resin (Ecocure 00–20, parts A and B, Smooth-On, Inc.) mixed with black dye pigment (Silc Pig, Smooth-On, Inc.) was also deposited uncured until the mold was completely filled. Curing of these two resins for 2 h led to strong attachment between the resulting silicones. In a second step, a third silicone resin containing turquoise platelets was deposited directly on top of the black outer surface of the actuator to generate the layer with architectured color. The processing route used to fabricate this third layer was similar to the one employed for the preparation of silicone-based soft composites described above. The black pigment present in the silicone beneath the third layer ensured a high color contrast in the inflated state of the actuator.

Characterization: The mechanical properties and the deformation-induced color change of the soft composites (Figure 2) were quantified under tensile mode using a mechanical universal tester (Shimadzu AGS-X) operated at a displacement rate of 2 mm min⁻¹. The tested soft composites were 2 mm thick and 30 mm long. Soft composites were imaged during the mechanical tests using a digital camera (Sony RX100 VI). The reflectance of the composite as a function of applied tensile strain (Figure 2C,E) was measured with a specular reflectance probe and a spectrometer for visible light (Ocean Optics Inc.). For composites manufactured with orthogonally aligned turquoise and red platelets, the reflectance was measured upon stretching along one of the two color-changing perpendicular directions. Cyclic tests (Figure S5 and Movie S13, Supporting Information) were performed using a 100-N load cell (Shimadzu AGS-X). The cycled samples were manufactured with 1 wt% of turquoise platelets in silicone. The imposed strain was varied between 0% and 315% in each cycle. The recorded video (Movie S13, Supporting Information) showed the first 8 out of the 100 cycles performed.

The rotation of individual platelets during stretching of the soft composite (Figure 3A) was investigated in a digital optical microscope (Keyence VHX). To track the rotation of single platelets as a function of the applied strain, a high-magnification lens (Keyence VHZ500R) was coupled to the optical microscope. The platelets were imaged from their top and side views while the composites were stretched by an Arduino-controlled stepper motor (M-23 1.8° 3.0 A, NEMA 23) with 0.25 mm steps.

The reflectance of stretched samples at a fixed observation angle (Figure 3D) was measured with a probe for specular reflectance analysis. The reflectance probe was tilted by 45° with respect to the surface of the composite and was pointed to a spot between the center of the sample and the clamps used to fix it to the stepper motor. This probe tilting angle was chosen so that the platelets were aligned perpendicular to the probe at high strains, thus maximizing the macroscopic reflectance.

The ability of the proposed soft composites to sense mechanical stimuli through optical signals was demonstrated using a photodiode and LEDs (MAX6641, Maxim Integrated) emitting at wavelength 536 nm placed underneath a silicone-based sample (Figure 4H, I). The 2 mm thick soft composite used in these experiments contained 4 wt% turquoise platelets vertically aligned in the elastomer matrix. Experiments were performed by compressing or shearing the sample while a dedicated software (MAX6640EVSYS, Maxim Integrated) recorded the intensity of light generated by the LEDs and reflected by the embedded platelets. The optical signal was acquired with an integration time of 14.8 µs with the LEDs operating in continuous mode. For compressing the soft composites, a patterned surface was mounted on a mechanical universal tester. The pattern consisted of 2 mm tall cylinders spaced from one another by 6 mm. The pressure was applied at a speed of 2 mm min⁻¹ with a maximum force of 70 N (Figure S12C, Supporting Information). To apply shear forces, the upper surface of the sample was fixed to a graduated focusing rail slider (MR 180, Fittest), while a constant compressive pressure of 1 N was applied normal to the sample surface. Shear was imposed parallel to the sample surface by moving the rail of the slider at a constant speed (Figure S12D, Supporting Information). The directional mechano-optical response of the composite was probed by performing the same shear experiment after rotating the sample by 90°.

Simulations: The simulations were carried out using the commercial finite element package ABAQUUS 2017 with the Abaqus/Standard solver. A Python script was developed for pre- and post-processing of the models and a user-material subroutine (UMAT) was employed to assign an incompressible Gent hyperelastic constitutive law to the silicone elastomer with a shear modulus of G = 10 kPa and a stretch limit of 32%⁴⁴. The alumina platelets were much stiffer than the matrix, with a Young’s modulus of 5.3 GPa⁴⁴ and a Poisson’s ratio of 0.25. A cubic representative volume element (RVE) with an edge size of 100 µm was considered for the soft composite. The RVE consists of an elastomer matrix that was populated with 16 hexagonal-shaped platelets with a circumscribed radius of 15 µm and a thickness of 300 nm which resulted in a weight ratio of ~ 1%. The platelets were randomly dispersed in the RVE and were initially aligned at θ₀ = 10°. Periodic boundary conditions were imposed on opposite edges and the unit cells were stretched in different directions under quasistatic conditions under a displacement-control protocol using a static step and considering the geometric nonlinearities. The effective stress-strain response of the composite and the orientation of the platelets were extracted in the post-processing step (Figure S13, Supporting Information).

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors have submitted a patent application based on the results of this research.

Author Contributions

E.P. and A.R. contributed equally to this work. E.P., A.R., and A.R.S. designed the research. E.P., A.R., V.P., and D.F. performed the experiments. A.R. performed the simulations. The manuscript was prepared by E.P. A.R., and A.R.S. All authors discussed and critically assessed the results and their implications and revised the manuscript at all stages.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.
color changing, composites, soft robotics, stretchable devices, structural color

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[1] S. A. Morin, R. F. Shepherd, S. W. Kwok, A. A. Stokes, A. Nemiroski, G. M. Whitesides, Science 2012, 337, 828.
[2] C. Xu, G. T. Stiubianu, A. A. Gorodetsky, Science 2018, 359, 1495.
[3] F. Fu, L. Shang, Z. Chen, Y. Yu, Y. Zhao, Sci. Rob. 2018, 3, eaar8580.
[4] Y. Yue, T. Kurokawa, M. A. Haque, T. Nakajima, T. Nonoyama, X. Li, I. Kajiwara, J. P. Gong, Nat. Commun. 2014, 5, 4659.
[5] G. Isapour, M. Lattuada, Adv. Mater. 2018, 30, 1707069.
[6] J. H. Pikul, S. Li, H. Bai, R. T. Hanlon, I. Cohen, R. F. Shepherd, Science 2017, 358, 210.
[7] C. Laschi, Science 2017, 358, 169.
[8] Y. Sagara, S. Yamane, M. Mitani, C. Weder, T. Kato, Adv. Mater. 2016, 28, 1073.
[9] Q. Wang, G. R. Gossweiler, S. L. Craig, X. Zhao, Nat. Commun. 2014, 5, 4899.
[10] G. R. Gossweiler, C. L. Brown, G. B. Hewage, E. Sapiro-Gheiler, W. J. Trautman, G. W. Welshofer, S. L. Craig, ACS Appl. Mater. Interfaces 2015, 7, 22431.
[11] C. Larson, B. Peele, S. Li, S. Robinson, M. Totaro, L. Beccai, B. Mazzolai, R. Shepherd, Science 2016, 351, 1071.
[12] Y. S. Kim, M. Liu, Y. Ishida, Y. Ebina, M. Osada, T. Sasaki, T. Hikima, M. Takata, T. Aida, Nat. Mater. 2015, 14, 1002.
[13] D. Gur, B. A. Palmer, B. Leshem, D. Oron, P. Fratzl, S. Weiner, L. Addadi, Angew. Chem., Int. Ed. 2015, 54, 12426.
[14] T. L. Williams, S. L. Senft, J. Yeo, F. J. Martin-Martinez, A. M. Kuzirian, C. A. Martin, C. W. Dibona, C.-T. Chen, S. R. Dinneen, H. T. Nguyen, C. M. Gomes, J. C. Rosenthal, M. D. Macmanes, F. Chu, M. J. Buehler, R. T. Hanlon, L. F. Deravi, Nat. Commun. 2019, 10, 1004.
[15] L. M. Mäthger, R. T. Hanlon, Cell Tissue Res. 2007, 329, 179.
[16] J. Teysier, S. V. Saenko, D. Van Der Marel, M. C. Milinkovitch, Nat. Commun. 2015, 6, 6368.
[17] M. Goda, Pigment Cell Melanoma Res. 2017, 30, 368.
[18] H. Le Ferrand, F. Bouville, T. P. Niebel, A. R. Studart, Nat. Mater. 2015, 14, 1172.
[19] L.-B. Mao, H.-L. Gao, H.-B. Yao, L. Liu, H. Colfen, G. Liu, S.-M. Chen, S.-K. Li, Y.-X. Yan, Y.-Y. Liu, S.-H. Yu, Science 2016, 354, 107.
[20] L. J. Bonderer, A. R. Studart, L. J. Gauckler, Science 2008, 319, 1069.
[21] F. Bouville, E. Maire, S. Meille, B. Van De Moortèle, A. J. Stevenson, S. Deville, Nat. Mater. 2014, 13, 508.
[22] A. Walther, I. Bjurhager, J.-M. Malho, J. Pere, J. Ruokolainen, L. A. Berglund, O. Ikala, Nano Lett. 2010, 2742.
[23] J. Billaud, F. Bouville, T. Magrini, C. Villevieille, A. R. Studart, Nat. Energy 2016, 1, 16097.
[24] R. M. Erb, J. S. Sander, R. Grisch, A. R. Studart, Nat. Commun. 2013, 4, 1712.
[25] A. Rafaanjani, K. Bertoldi, A. R. Studart, Sci. Rob. 2019, 4, eaav7874.
[26] F. Barthelat, Int. Mater. Rev. 2015, 60, 413.
[27] S. Shan, S. H. Kang, J. R. Raney, P. Wang, L. Fang, F. Candido, J. A. Lewis, K. Bertoldi, Adv. Mater. 2015, 27, 4296.
[28] T. A. Schaadler, W. B. Carter, Annu. Rev. Mater. Res. 2016, 46, 187.
[29] G. Pfaff, in High Performance Pigments (Eds: E. B. Faulkner, R. J. Schwartz), Wiley-VCH, Weinheim, Germany, 2009, pp. 75–104.
[30] V. Saranathan, C. O. Osuji, S. G. J. Mochrie, H. Noh, S. Narayanan, A. Sandy, E. R. Dufresne, R. O. Prum, Proc. Natl. Acad. Sci. USA 2010, 107, 11676.
[31] A. A. Dolan, B. D. Wilts, S. Vignolini, J. J. Baumburg, U. Steiner, T. D. Wilkinson, Adv. Opt. Mater. 2015, 3, 12.
[32] J. D. Ioannopoulos, S. G. Johnson, J. N. Winn, R. D. Meade, Photonic Crystals: Molding the Flow of Light, 2nd ed, Princeton University Press, Princeton, NJ, USA 2011.
[33] R. M. Erb, J. Segmehl, M. Charilaou, J. F. Löffler, A. R. Studart, Soft Matter 2012, 8, 7604.
[34] R. M. Erb, R. Libanori, N. Rothfuchs, A. R. Studart, Science 2012, 335, 199.
[35] D. Kokkinis, M. Schaffner, A. R. Studart, Nat. Commun. 2015, 6, 8643.
[36] J. J. Martin, B. E. Fiore, R. M. Erb, Nat. Commun. 2015, 6, 8641.
[37] G. Pfaff, qmore articles, “Fascinating displays of colour”, https://q-more.chemeurope.com/q-more-articles/190/fascinating-displays-of-colour.html (accessed: November 2020); chemie&more 2013, 6, 33.
[38] E. Poloni, “Optical reflectance of composites containing aligned engineered microplatelets”, unpublished.
[39] A. Alfadhel, J. Kosek, Adv. Mater. 2015, 27, 7888.
[40] S. Gong, W. Schwabl, Y. Wang, Y. Chen, Y. Tang, J. Si, B. Shirinzadeh, W. Cheng, Nat. Commun. 2014, 5, 3132.
[41] J.-Y. Sun, X. Zhao, W. R. K. Illeperuma, O. Chaudhuri, K. H. Oh, D. J. Mooney, J. J. Vlassak, Z. Suo, Nature 2012, 489, 133.
[42] J. Jorgensen, Parametric Tool to Generate 3D Printable PneuNet Bending Actuator Molds, https://softroboticstoolkit.com/parametric-tool-3d-printed-molds (accessed: November 2020).
[43] H. Wang, J. Kow, N. Raske, G. De Boer, M. Ghajari, R. Hewson, A. Alazmani, P. Culmer, Sens. Actuators, A 2018, 271, 44.
[44] Z. Liao, M. Hossain, X. Yao, Mech. Mater. 2020, 144, 103366.
[45] E. Felden, T. Giovannini, N. Ni, C. Ferraro, E. Saiz, L. Vandeperre, F. Giuliani, Scr. Mater. 2017, 131, 55.