Naturally Degradable Photonic Devices with Transient Function by Heterostructured Waxy-Sublimating and Water-Soluble Materials

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Combined dry–wet transient materials and devices are introduced, which are based on water-dissolvable dye-doped polymers layered onto nonpolar cyclic hydrocarbon sublimating substrates. Light-emitting heterostructures showing amplified spontaneous emission are obtained on transient elements and used as illumination sources for speckle-free, full-field imaging, and transient optical labels are realized that incorporate QR-codes with stably encoded information. The transient behavior is also studied at the microscopic scale, highlighting the real-time evolution of material domains in the sublimating compound. Finally, the exhausted components are fully soluble in water thus being naturally degradable. This technology opens new and versatile routes for environmental sensing, storage conditions monitoring, and organic photonics.

Devices that undergo an univocally evolving designed function, possibly including a self-elimination process to harmless end products through mechanical fragmentation and dissolution in water or air, are collectively referred as physically transient electronics and photonics.[1,2] The interest in such technology is rapidly emerging, and various forms of transient electronics have been recently traced,[1] including dissolvable devices that deploy room-temperature liquid metals with high recycling efficiency.[3,4] Indeed, the continuous increase of electronic waste, that will be further enhanced by the daunting amount of sensors and devices to be used for the forthcoming Internet of Things,[5] is motivating significant concern. For these reasons, technologies are highly desired, which might produce naturally degradable devices, that once ended their function do not lead to release of heavy metals or toxic compounds in the environment. Such feature is also of interest for other applications where conventional electronics and photonics are not well-suited such as biodegradable, temporary implants,[6] secure data-storage systems,[7] and unrecoverable remote controls.[8] For instance, physically transient light emitting components provide an interesting option in various domains, such as biophotonics,[9] implantable analytic chips,[10] wireless optogenetics,[11] and full-field imaging.[12] Examples of transient photonic systems include nanoimprinted distributed feedback lasers based on DNA[2] or silk gratings obtained by using fused silica,[13] and fluorescent chemosensors for acid vapor detection.[14] Furthermore, “dry” forms of transient electronics have been recently developed for transistors or solar cells realized by transfer printing onto materials that sublime.[15] In fact, ambient sublimation of device-carrying substrates leads to mechanically disintegrate the remaining materials into microscopic fragments (due to loss of support and/or interlayer adhesion). Dissolution in water could then be deployed to finally eliminate components at the end of their life cycle.

Here, we present the first example of combined dry–wet transient devices in photonics, based on water-soluble compounds
layered onto sublimating substrates. Cyclododecane (CDD), a wax-like nonpolar cyclic hydrocarbon solid, widely used as a temporary consolidant, adhesive, or barrier during archeological recovery,[16] serves as substrate for an optically-active water-dissolvable polymer bilayer encompassing a laser dye, as schematized in Figure 1a. The relevance of this method is twofold, involving both fundamental and practical findings. On one side the compatibility of sublimating substrates with organic photonics is here assessed, that goes beyond previous results on silicon-based electronics,[15] tackling potential issues related to deterioration of light-emitting molecules interacting with CDD or excess light scattering. In addition, heterostructures built on the sublimating substrates are proposed, including both organic lasers and optical labels (QR-codes) with transient behavior and stably encoded information, showing two different application fields for naturally degradable, all organic photonic devices.

As first step of the device fabrication process, grains of CDD paraffin are melted at 140 °C and then rapidly cooled in a refrigerator to form substrates with size 2 × 2 cm² and thickness in the range 1–3 mm (Figure 1b). Silicon molds are used to generate substrates, which can have varied shapes, different dye-doping (Figure 1b–d), and smooth surfaces (root mean square roughness, \( R_q = 70 \) nm). Alternative templates such as glass Petri dishes, and other methods explored such as applying mechanical forces to solid grains, lead to samples with higher roughness (\( R_q = 25 \) µm and 770 nm, respectively, Figure S1, Supporting Information).

The obtained CDD substrates are soft, and can be cut into pieces of the desired size and shape by the use of a scalpel, as shown in Figure 1e,f. Mechanical analysis in compression mode, performed by thrusting downward a flat disk onto the substrate at a rate of 0.5 mm min⁻¹ at room temperature, highlights a roughly linear force-deformation response, i.e., an elastic modulus of (14 ± 1) MPa up to about 30 N (≈1–1.5% compression), followed by irreversible deformation indicative of the material transport and ductile behavior (Figure S2, Supporting Information). At ambient temperature under a fume hood with an air velocity of 1.9 m s⁻¹, and relative humidity of about 50%, a substrate (thickness 1–2 mm) completely disappears in a time of 7–9 days (Figure 1g–i), corresponding to an area reduction rate of about (0.6 ± 0.1) mm² h⁻¹ and to a thickness reduction rate of about (12±1) µm h⁻¹ (Figure 1j,k and Figure S3, Supporting Information). These rates can be controlled to high extent, thus providing routes to realize components with programmable self-destruction, depend-
The intensity of the emitted light on the observation angle, that is imaged within the microscope depth of focus (Figure 2e–g).

Though in absence of significant sample heating (Figure S4, Supporting Information), this average value slowly varies over time (by $\approx 10^2 \text{ µm}^2 \text{ h}^{-1}$), related to free volume redistribution during sublimation.

Light-emitting heterostructures that can operate as optical amplifiers are built by drop-casting bilayers made of poly(vinyl alcohol) (PVA) and polyvinylpyrrolidone (PVP) doped with the red-emitting laser dye [2-[2-[4-(dimethylamino)phenyl]ethenyl]-ethenyl]-6-methyl-4H-pyranyl-4-ylidene]-propanedinitrile (DCM), onto the CDD substrate, according to the scheme in Figure 1a. The device cross-section is shown in the white light and fluorescence micrographs in Figure 3a,b, where a sample with the intermediate PVA intentionally doped with a blue-emitting molecule (stilbene 420) is used to better highlight the neat interfaces between the different layers. The absorption and photoluminescence (PL) spectra of the active film deposited on a reference quartz substrate are displayed in Figure S5 (Supporting Information). During device fabrication, orthogonal solvents allow the transient heterostructures to be built by sequential casting. Specifically, water is used to dissolve PVA, thus enabling the formation of a continuous planarizing coating on top of the CDD. The PVA–CDD interface is found to be stable, and delamination is only occasionally observed from sample corners during heterostructure sample cutting (surface area $\approx 1 \text{ cm}^2$), which could be related to undesired air pockets trapped at the interface formed during polymer casting. Furthermore, since CDD is soluble in polar solvents, the direct deposition of DCM:PVP from a chloroform solution is unviable onto the sublimating material, since this would lead to permeation of both the polymer and the dye into the substrate, as well as to a significant alteration of the dye spectral properties (Figure S6, Supporting Information). Obtaining interfaces of high quality is clearly important for promoting waveguiding of the photoexcited light emitted by the heterostructure, occurring mostly in the dye-doped layer (see Experimental Section). In addition, these heterostructures are compatible with encapsulation methods with optical adhesives, that can be used to strongly enhance the device photostability.

The PL spectra, obtained from the device (Figure 3c) upon pumping with $\approx 10$ ns pulses at 355 nm and with a stripe excitation geometry (Experimental Section), have a linewidth (full width at half maximum, FWHM) down to 25 nm, and a threshold at 0.2 mJ cm$^{-2}$ for amplified spontaneous emission (ASE) as displayed in the light-light (L-L) plot in Figure 3d. The spectra do not exhibit intrinsic polarization (Figure S7, Supporting Information), which is indicative of an isotropic distribution obtained for the dye chromophores upon casting. In Figure 3e, we display the output beam divergence, namely the dependence of the intensity of the emitted light on the observation angle, that

![Figure 2](image_url)

**Figure 2.** Microscopic images of a CDD region during sublimation. a–c) PB confocal micrographs at different times. The white dashed lines highlight changes in the edges of the back-scattering material surface. Scale bar: 100 µm. d) PB intensity versus time, indicative of the sublimation behavior. The red dashed line is a guide for the eyes. e–g) Cross-polarized white light micrographs of CDD substrate at different times, indicative of dimensional changes in the material domains within the depth of focus. Scale bar: 100 µm.
Figure 3. Transient photonic device. a,b) Optical and fluorescence micrographs of the device cross section. The fluorescence image is obtained upon doping the PVA layer with stilbene (blue-emitting). Marker: 100 µm. c) ASE spectra at different excitation fluences in the range: 0.06–0.5 mJ cm$^{-2}$. d) Corresponding L–L plot (red circles, left vertical scale), and dependence of the FWHM on the excitation fluence (blue diamonds, right scale). e) Angular dependence of the emitted intensity (dots). The continuous line is a guide for the eye. f) Dependence of the emission spectrum on number of pulses at 0.3 mJ cm$^{-2}$. g) ASE intensity (red circle, left vertical scale) and FWHM (blue diamonds, right vertical scale) versus the number of excitation pulses (repetition rate: 10 Hz) at 0.3 mJ cm$^{-2}$. The black line is a guide for the eye. Images of a US Air Force resolution test chart taken by h) an Nd:YAG laser and by i) the ASE from the transient device. Scale bar: 115 µm. Top right inset: magnification view of the test patterns in column 7, lines 4–6. j) Comparison of CNRs for test patterns imaged by a narrow band laser (blue triangles) and by a transient photonic device (red dots). Average values estimated on six pattern lines at each group size.

is measured by the experimental geometry shown in Figure S8 (Supporting Information). The spatial profile for ASE (Figure 3e) evidences the coexistence of different light modes which are out-coupled, which is typical of asymmetric emissive layers embedding organic layers with optical gain.\cite{22} Furthermore, the device photostability in air is studied by recording spectra of the emitted light as a function of the number of excitation pulses (i.e., time), at a constant pump fluence of 0.3 mJ cm$^{-2}$ (Figure 3f). The operational lifetime of the device (given by the number of excitation pulses at which the ASE intensity decays by 1/e) is so estimated to correspond to about 1.5 $\times$ 10$^4$ pulses, with a FWHM increasing by a factor 1.5 in a related way (Figure 3g). This performance is fully in line with the photostability measured for organic lasers using well-established architectures, including distributed feedback resonators.\cite{23,24}

The possible applications of these devices before controlled disintegration are numerous. They can be used as high-quality, bright illumination sources for full-field imaging, since unconventional laser architectures with low spatial coherence allow for producing images without speckle artefacts.\cite{25} In previous works, random lasers made of colloidal solutions or perovskite films have been proposed as tool for speckle-free imaging.\cite{12,26,27} In order to analyze this capability for ASE transient devices, we use them to illuminate a 1951 U.S. Air Force (AF) resolution test chart, and compare so-obtained images with those produced by a narrowband laser (Nd:YAG). The experimental set-up is schematized in Figure S9 (Supporting Information). The illuminated images of the test patterns are shown in Figure 3h,i. While textured artifacts due to speckle interference are found in the image collected from by Nd:YAG laser illumination (Figure 3h), the ASE
from the transient device, providing an optical signal with low
spatial coherence, allow clean imaged patterns to be achieved,
free from speckle effects (Figure 3). A quantitative comparison
can be carried out by estimating the contrast to noise ratio, CNR
\( \frac{\langle I_f \rangle - \langle I_b \rangle}{\sigma_f + \sigma_b/2} \), where \( \langle I_f \rangle \) is the average intensity of
a reference bar in the test chart, \( \langle I_b \rangle \) is the average intensity of
the image background, and \( \sigma_f, \sigma_b \) indicate the corresponding stan-
dard deviations of the pixel intensity. By using the transient de-
vice as light source, CNR values are up to four times better than
by using a conventional laser and comparable to those obtained
by colloidal or perovskite lasers (Figure 3).[26,27]

Upon sublimation the CDD material loss can deeply modify
the device morphology. To capture this behavior we perform
accelerated sublimation tests by placing the ASE transient de-
vice on a hot plate at 55 °C and imaging it by both an infrared
thermographic camera (Figure 4a) and by optical photographs
(Figure 4b). The temperature is chosen to avoid local melting
(occurring above 60 °C) of the surface in contact with the hot
plate. A temperature gradient of about 3 °C mm⁻¹ is measured
across the CDD substrate, keeping the active layer at a tempera-
ture significantly lower than the heated parts. In addition, it is
found that sublimation proceeds faster at the edges of the over-
all heterostructure, which is gradually led to take a mushroom-
like shape with the light-emitting region remaining largely unal-
terated at the top of the device. Bending of the PVA/DCM:PVP bi-
layer could occur as a consequence of the substrate sublimation
and the consequent strain energy release in the two polymers.
The amount of bending would depend on the thickness ratio
\( R \) of the two layers (DCM:PVP thickness/PVA thickness), being
less pronounced upon decreasing \( R \) below unity.[28] In Figure 4c,
we show how this aspect can be engineered to keep the light-
emitting structure mechanically stable and unbent, even upon
complete CDD sublimation, by properly choosing the thickness
ratio \( R = 0.4 \). Such \( R \) value engineered for PVA/DCM:PVP can
depend on the environmental relative humidity, due to differential
water vapor absorption and polymer swelling for the two in-
volved layers. Keeping the optically relevant structure mechani-
cally stable is highly desired in order to use the photonic struc-
ture with unchanged overall geometry and output beam direc-
tionality along the component lifetime. For instance, this prop-
erty is highly relevant to realize intelligent, optical labels that are
naturally degraded after read while keeping stably readable the
information encoded in them during use. We provide a demon-
stration of this behavior by realizing QR-code patterns through
inkjet printing of poly(3,4-ethylenedioxythiophene):polystyrene
sulfonate (PEDOT:PSS) onto CDD/PVA/PVP heterostructures
(Figure 5). To this purpose, a Dimatix Materials Printer 2800 (Fu-
jifilm) is employed, developing a direct, drop-on-demand print-
ing process entirely at room temperature in air. A 1 µL cartridge
is used with a drop spacing of 20 µm, while printing six over-

Figure 4. Transient device sublimation and dissolution. a) Infrared images and corresponding photographs b) of the ASE transient device while the
substrate sublimates on a hot plate at 55 °C. Four instants are captured during 48 h. Scale bar: 2 mm. c) Bending behavior of transient devices, realized
with different thickness ratio \( R \) of the two polymer layers, upon sublimation of the CDD substrate. Photographs (top line, scale bar: 0.5 cm) and
fluorescence images of the cross-sections (bottom line, scale bar: 200 µm) of sample with \( R = 0.9 \) (i–iv) and \( R = 0.4 \) (v–viii). Samples are inspected
before (i,iii,v,vii) and after (ii,iv,vi,viii) sublimation, respectively.
During the sublimation of the CDD substrate, the devices do not significantly bend, making the information encoded in them easily readable by the smartphone scanner (Figure 5c,f,i,l, where the words “Crypted Message” correspond to the reading of the QR-code patterns at each time step).

Finally, after device use and complete self-destruction of the CDD layer, the QR-codes are disintegrated and the remaining polymer layers straightforwardly dissolved in water (Figure 5m–p and Figure S10, Supporting Information). While ultimately dissolving the exhausted components, water is not functional to the device operation, thus avoiding uncontrolled flows or undesired variability related to dye quenching or dissolution rates. In this respect, dry transient technologies provide additional self-eliminating capability, that complements those of widely explored wet systems, and is critically useful in environments where little water or moisture are present.

To summarize, our work introduces new physically transient photonic devices with programmable disintegration. Compared to existing transient architectures involving semiconductors for electronics, bioelectronics, and photovoltaics, these systems are based on fully organic components, with extremely low cost associated with the related processing technologies and absence of dissolved metals. The combined use of wet and dry transient materials might lead to systems with further enhanced and versatile transient functions, and to new fabrication routes for organic lasers and encoded labels, among others, with potential application in environmental sensing, storage conditions monitoring, and photonic chips. For instance, the here analyzed time scales (a few days) can be useful for making transient fluorescence excitation sources in disposable lab-on-chip platforms, where the transient devices can be directly coupled with integrated waveguides and interrogation regions. Importantly, the method is also of interest in terms of environmental safety, suppressing costs and hazards associated with discarded devices.

Experimental Section

Transient Devices: CDD was purchased from Kremer Pigmente GmbH & Co. KG (Aichstetten). PVA and PVP were purchased from Sigma Aldrich. DCM was purchased from Exciton Inc. PEDOT:PSS (Clevios PH 1000 purchased from Heraus) was used as antistatic waterborne dispersion based on submicrometer gel particles. Upon CDD melting, substrates were rapidly refrigerated from the liquid phase to obtain a dense amorphous phase. The surface roughness of CDD obtained by using different molds was determined by inspecting sample profile by a stylus profilometer (Bruker Dektak XT). Mechanical measurements in compression mode were performed by a mechanical tester (INSTRON 5500R) with stainless still parallel plates (diameter 5 cm) and equipped with MERLIN software (INSTRON: Norwood, MA, USA). A bilayer of polymer PVA/DCM:PVP (1% wt:wt) of DCM was deposited by casting directly on top of the CDD substrate. The refractive indexes of involved layers (≈1.48 for CDD and PVA and ≈1.53 for PVP) promote waveguiding only in the dye-doped film. The thickness of each deposited polymer layer, in the range 45–90 μm (PVA) and ≈30 μm (DCM:PVP), was measured by observing cross-sections by an optical stereomicroscope in bright and dark field (Leica MZ16 FA). The method was also supported by looking at identical samples upon fluorescence excitation, by doping PVA with the blue-emitting dye stilbene 420 (Exciton Inc.). Transient QR-codes were realized by direct drop-on-demand, inkjet printing of PEDOT:PSS. A Dimatix Materials Printer 2800 (Fujifilm) was used under ambient conditions, with a 16-nozzle cartridge by 1PL single-drop volume, a single nozzle to improve accuracy, and drop spacing of 20 μm. Six overlapping layers of PEDOT:PSS were printed, with a 60 s pause between two consecutive printing steps to allow for effective drying, thus guaranteeing the deposition of a uniform, dark film in the label features.

Microscopy: An inverted microscope, Eclipse Ti (Nikon), equipped with a confocal A1R-MP system (Nikon) was used for the microscopic optical measurements. For the characterization of the light backscattered by the CDD material, a polarized Ar ion laser (wavelength, λ = 488 nm) was used as the light source, while the backscattered signal was collected by a 10 × objective (numerical aperture, N.A. = 0.25) and measured by a photomultiplier. At the beginning of the measurement, the objective is positioned in order to have the focal plane at the surface of the sample.
(air/sample interface), and this objective position is fixed throughout the overall measurement. Upon sublimation from the sample, the light intensity that is backscattered by the air/sample interface and measured by the detector of the confocal microscope is to decrease because of the increasing spatial gap between the air/sample interface and the fixed focal plane. The sublimated thickness was estimated by the focal depth of the confocal systems: \( \Delta z = 2ax/(N.A.)^2 \).\(^{[11]}\) The bright field imaging time-lapse was collected by a DS-R11 color charged-coupled-device (CCD) camera (Nikon) in cross-polarization mode. Analysis was performed by the ImageJ software. QR-codes were imaged by using either a smartphone camera or a CCD camera (Leica) coupled to a long working-distance optical system (MVLM7000, Thorlabs).

**Thermographic Imaging:** Thermal imaging of the devices was carried out by using an infrared camera (FLIR, A655sc). Before each thermographic measurement,\(^{[14]}\) the camera was calibrated following the manufacturer guidelines.

**Spectroscopy:** A JASCO UV–vis spectrophotometer V-550 was used to measure absorption spectra of the active bilayers, while PL spectra were acquired by a Cary Eclipse Fluorescence Spectrophotometer (Varian). The transient photonic devices were excited by the third harmonic (\( \lambda = 355 \text{ nm} \)) of a Nd:YAG laser (Quanta-Ray INDI, Spectra-Physics) with 10 ns pulse duration (FWHM) and 10 Hz repetition rate. The excitation beam was shaped into a stripe with size 5 × 0.5 mm\(^2\). The intensity and spectral properties of the light emitted from the sample edge were characterized by a fiber-coupled monochromator (HR320, JobinYvon) equipped with a Peltier-cooled CCD detector (Symphony, Horiba) and a 600 grooves x mm\(^{-1}\) grating. Rotating a polarizer positioned along the path of the collection of the light emitted by the sample was used for analyzing the polarization state of the emission. The divergence of the light beam emitted by the device was determined by using a mask with a slit aperture (width 1 mm and height 10 mm) positioned perpendicularly to the emission direction (Figure S8, Supporting Information), and measuring the intensity of the light transmitted through the slit as a function of the transversal position of the mask. A negative 1951 US Air Force resolution test chart (Thorlabs) was used for imaging experiments performed with the transient photonic device and with a laser light source (Figure S9, Supporting Information). Images were collected with a 10 × objective lens (M-10X, Newport, N.A. = 0.25) and a CMOS camera (Thorlabs).

**Statistical Analysis:** The area and thickness of CDD substrates during sublimation (Figure 1 and Figure S3, Supporting Information) were normalized at values measured at \( t = 0 \). Sublimation rates at ambient temperature were averaged over 15 different samples. Experiments to measure the elastic modulus were repeated on three different samples realized with the same processing conditions. The lasing behavior was analyzed on five different devices. The average values of CNR (Figure 3j) was estimated on six pattern lines at each group size.

**Supporting Information**

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

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**Conflict of Interest**

The authors declare no conflict of interest.

**Keywords**

microlasers, physically transient devices, smart materials, sublimation, waxes

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