

# Wearable variable-emittance devices—The future of dynamic personal thermoregulation

Cite as: Appl. Phys. Lett. **125**, 180501 (2024); doi: [10.1063/5.0217725](https://doi.org/10.1063/5.0217725)

Submitted: 6 May 2024 · Accepted: 16 October 2024 ·

Published Online: 30 October 2024



View Online



Export Citation



CrossMark

Ting-Hsuan Chen<sup>1,2</sup>  and Po-Chun Hsu<sup>2,a)</sup> 

## AFFILIATIONS

<sup>1</sup>Thomas Lord Department of Mechanical Engineering and Materials Science, Duke University, Durham, North Carolina 27708, USA

<sup>2</sup>Pritzker School of Molecular Engineering, The University of Chicago, Chicago, Illinois 60637, USA

<sup>a)</sup>Author to whom correspondence should be addressed: [pochunhsu@uchicago.edu](mailto:pochunhsu@uchicago.edu)

## ABSTRACT

Using infrared electrochromism as the strategy to combat the fluctuation of environmental conditions, wearable variable-emittance (WeaVE) devices are able to integrate the functionality of personal thermoregulation and closed-loop control into the future textile, featuring its large tunable range, ultra-low energy consumption, lightweight, and wearability. Recently, this new wearable technology has evolved beyond planar electrochromic cells and is moving closer to woven textiles. To further improve electrochromic performance and wearability, comprehensive progress is necessary from materials science to fabrication techniques. In this Perspective, we elaborate on the mechanisms behind electrochemically active WeaVE devices, analyze how dynamic and fundamental studies may improve the electrochromic performance, and explore the possibility of incorporating nanophotonic designs in the development of this future smart textile through research.

Published under an exclusive license by AIP Publishing. <https://doi.org/10.1063/5.0217725>

## INTRODUCTION

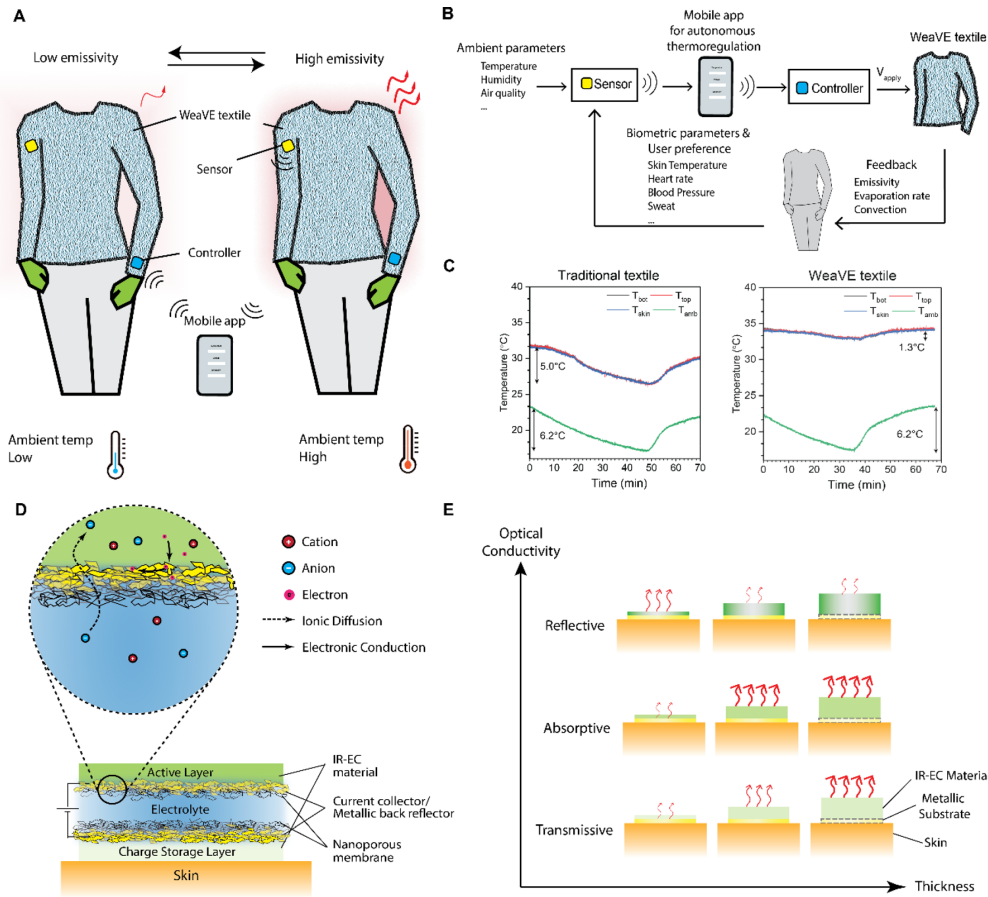
Wearable variable emittance (WeaVE) devices can be defined as actively adaptive personal thermoregulation devices by radiative heat management.<sup>1</sup> In terms of the radiative heat transfer mechanism, WeaVE represents a novel scheme of personal thermoregulation distinct from other thermoregulatory textiles or devices [Fig. 1(a)]. Compared to passive thermoregulation devices, they allow users to personalize and actively control radiative heat transfer in a fluctuating environment by adjusting the overall thermal emittance.<sup>2–8</sup> Infrared (IR) electrochromic materials are used in WeaVE devices due to their variable thermal emissivity when undergoing electrochemical reactions. Organic and carbon-based electrochromic materials have been extensively used to provide variable emittance in WeaVE devices, as their flexible texture significantly increases the integration capability to fabrics or fibers.<sup>1,9–22</sup> For WeaVE devices, no external energy input is required to stabilize skin temperature and maintain at a certain thermal comfort zone within a range of ambient fluctuation.<sup>3</sup> These thermal comfort states can be easily maintained since emissivity is nonvolatile, and the transition between these states is energy-efficient.

Under the context of autonomous personal thermoregulation system, WeaVE device is the crucial element to provide active feedback to maintain thermal comfort based on the environments and biometric parameters [Fig. 1(b)]. The core concept of WeaVE devices is to control the human body heat loss through textiles with electrical signal.

The dynamic thermal emissivity contrast serves as the main variable to stabilize the radiative heat loss from the human body.

Despite the great potential, improving the performance of WeaVE is an interdisciplinary effort due to the complexity of considering all the electron/ion transport,<sup>23–34</sup> molecular structures,<sup>29,30,35–43</sup> optical dispersion,<sup>44–53</sup> and device integration.<sup>1–3,7,17,20,54–61</sup> Dynamic optical and charge transport properties are coupled, and it is difficult to discuss without each other. On the side of fundamental science, developing electrochromic materials is a process to identify and synthesize molecular structures that allows a wide range of tuning of electronic transition and transport in an electrochemically favorable process. Exploring the electrochemical kinetics of tuning is a particularly important research goal for a clear and comprehensive picture of the phenomenon.<sup>23</sup> Advanced structural characterization techniques are the foundation considering the kinetically dependent structural evolution in real time.<sup>62–66</sup> On the other hand, effective textile and fabric design is the key to bringing the performance and wearability closer to the current form of clothing.<sup>9,67–74</sup> Regarding biocompatibility, careful selection of materials and passivation are also crucial in minimizing health hazards.<sup>75</sup>

In addition to electrochromic cells, nanophotonic designs and materials with different tuning mechanisms can also achieve IR tuning. For example, incorporating thermochromic and phase-changing materials into photonic components is a common strategy to enable the tunability in different applications.



**FIG. 1.** Operating principle and schematic diagram for WeaVE devices. (a) Autonomous personal thermoregulation system with WeaVE textile, remote controller, and sensors for biometric and ambient parameters with remote connection. (b) Control loop for autonomous personal thermoregulation with multi-modal feedback from WeaVE textile. (c) Skin temperature stabilization function in real-time thermal performance testing in fluctuating ambient temperature. (d) Example of a planar WeaVE device in cross section. The three-phase interface across the nanoporous membrane is where the electric current and ionic diffusion are generated to enable the electrochemical reactions. (e) Optical response of a WeaVE device with back reflector configuration using IR-EC material with different optical properties. The optical response of WeaVE is dominated by IR-EC material as its thickness increases. \* (c) Reproduced with permission from Chen *et al.*, PNAS Nexus 2(6), pgad165 (2023). Copyright 2023 Authors.

In this Perspective, we will begin with a brief introduction and review of the recent progress in this emerging field, followed by pointing out the opportunities and potential challenges in different domains, hoping to accelerate the realization of actively adaptive personalized thermoregulation in a sustainable way.

#### PERFORMANCE METRICS OF WeaVE DEVICES

Radiation is the main source of heat loss from human body, accounting for up to 40 percent of the total amount. WeaVE devices take this advantage to counteract the ambient temperature fluctuation by acting as textiles with continuously tunable thermal emittance. Their performance is determined by the ability to modulate the radiant exitance emitted from the surface. It is common to define and

maximize the figure of merit,  $\Delta\epsilon$ , as the total hemispherical emissivity contrast between two extreme states ( $\epsilon_{\text{high}} - \epsilon_{\text{low}}$ ). Here,  $\epsilon_{\text{high}}$  corresponds to the high emissivity state of WeaVE to cool down the human body, and  $\epsilon_{\text{low}}$  is the low emissivity state to retain heat with less radiative heat loss. The total hemispherical emissivity at each state is calculated from the weighted integral of spectral emissivity with respect to the blackbody spectral radiance ( $I_{\text{BB}} = \frac{2hc^2}{\lambda^5} \frac{1}{e^{\frac{hc}{\lambda kT}} - 1}$ ) over the entire wavelength,

$$\Delta\epsilon = \frac{\int_0^\infty [\epsilon_{\text{high}}(\lambda) - \epsilon_{\text{low}}(\lambda)] I_{\text{BB}}(\lambda) d\lambda}{\int_0^\infty I_{\text{BB}}(\lambda) d\lambda}. \quad (1)$$

As an approximation to the broadband thermal emission, we typically assume the surface of WeaVE textiles is diffuse with uniform temperature.  $I_{BB}$  at human skin temperature (306 K) is dominated in the range of mid-infrared (mid-IR) from 2.5 to 40  $\mu\text{m}$ , with peak value at around 9.5  $\mu\text{m}$ . Note that in Eq. (1),  $\varepsilon(\lambda)$  at a certain state is used to quantify the thermal emittance from the whole device rather than the emissivity of a single layer of material. Both the material's emissivity and device configuration can be adjusted to control the overall thermal energy emitted from WeaVE on human body.

In practice, the emittance of a WeaVE device is evaluated by experimentally measuring the reflectance spectra with FTIR and integrated sphere at a near-normal angle of incidence using unpolarized light source. For WeaVE devices, the ability to switch to high IR reflectance state is especially important to suppress thermal emittance and retaining heat. Because of the high emissivity of human skin ( $>0.97$ ), transmission tuning between IR transmissive and absorptive will result in the same high  $\varepsilon(\lambda)$  when human skin is underneath. In addition to IR spectra, real-time heat transfer measurement is a demonstration to simulate thermoregulation performance when taking the transient effects in a controlled environmental condition into account [Fig. 1(c)].

#### MODULATING THERMAL EMISSION BY ELECTROCHROMISM

Combining IR-electrochromic (IR-EC) materials, electrolyte, and current collectors, WeaVE device is essentially an electrochemical cell with tunable thermal emittance. Redox reactions and changes in the doping level occur through the charge transfer and ionic diffusion at the interface between electrolyte, current collector, and IR-EC materials [Fig. 1(d)]. Standalone electrochemical cell allows reversible and continuous transition between thermal states by applying voltages through current collectors from portable circuits and power source. The thermal modulation is achieved in a thin design with thickness comparable to that of traditional textiles.

To maximize  $\Delta\varepsilon$ , various configurations can be employed in WeaVE devices. One common strategy is to incorporate transmission tuning IR-EC materials with a back reflector [Fig. 1(e)]. This kind of device can be readily integrated with electropolymerization on metallic substrates. However,  $\Delta\varepsilon$  is therefore limited, because optimizing the thickness is necessary to prevent the device from being too reflective when the IR-EC material is thin and too emissive when it is thick. Another method is to utilize IR-EC materials between reflective and absorptive, although meeting the requirement of high optical conductivity with tunability is more challenging for many materials.

Conducting polymers (CPs) are one of the most popular candidates of IR-electrochromic materials in WeaVE devices [Fig. 2(a) and 2(e)]. Different types of acidic and metal ion dopants, counterions, and variations of CPs have been used to increase the electrochromic performance and stability.<sup>20,54–56,76</sup> In particular, electropolymerized polyaniline (PANI) and polythiophene (PT) based polymers are extensively used due to the simple fabrication procedure and quick response time under a low operating electrochemical potential.<sup>1,9–12,14</sup> Initially, the knowledge of CP-based IR-electrochromic devices was first adopted for spacecraft thermal control by Chandrasekhar *et al.*<sup>54,55</sup> CP-based devices with different designs were later developed for different tunable optical and thermal applications.<sup>10,11,14,16,17,19,57,77,78</sup> In 2023, Chen *et al.* demonstrated the wearability by introducing kirigami design and incorporating autonomous control with temperature sensors and Bluetooth connection function from a smartphone.<sup>1</sup> CPs with a higher

electrical conductivity have also been utilized with chemical or vapor phase polymerization, offering the opportunities to utilize intrinsically reflective CPs to realize multifunctional active metasurfaces.<sup>20,76,79</sup>

Another promising choice is carbon-based nanomaterials. With ultrahigh carrier mobility and electrochemically tunable broadband optical properties, carbon nanotubes (CNT) and multilayer graphene (MLG) have been incorporated into planar WeaVE devices for personal thermoregulation and infrared camouflage applications.<sup>9,15,58,80</sup> Emissivity modulation is enabled by Fermi level engineering through ionic intercalation. Kocabas *et al.* reported the graphene-enabled personal thermoregulation device with WeaVE-like functionality and integration across the broadband [Figs. 2(f) and 2(h)].<sup>15,58,81</sup> Xiao *et al.* developed large-scale CNT film also for radiative thermal modulation afterward [Fig. 2(i)].<sup>9</sup> In 2024, Shi *et al.* further combined CNT grid with CP as dual-band electrochromic device in visible light and mid-IR [Fig. 2(j)].<sup>82</sup>

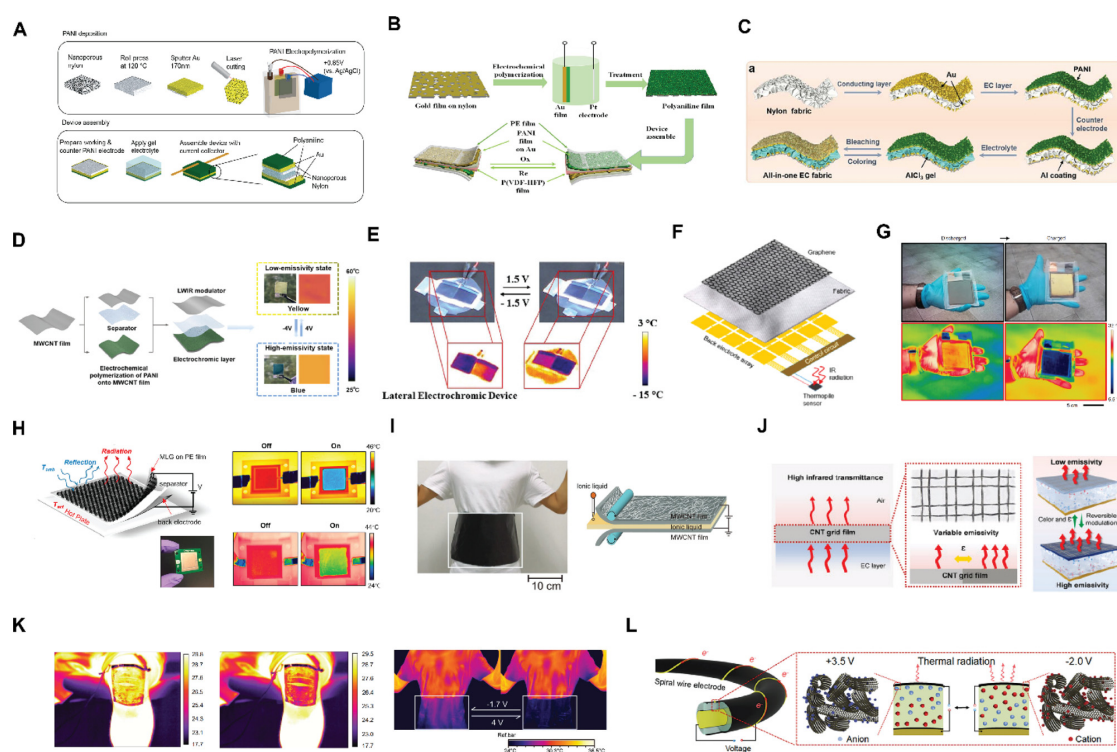
To ensure uniform electrical potential and contact to the electrolyte, these electrochromic materials are usually deposited on metalized nanoporous films. For IR-EC materials switching between transmissive and absorptive (emissive), the metalized layer works as an IR back reflector to render the transmissive state reflective and low-emissivity. Ion permeable membranes and fabrics, such as nylon, polyethylene, and cotton, have been used as the substrate of current collector and mechanical support in planar WeaVE devices.<sup>1,17,20,54–56,58,76</sup> The electrolyte in WeaVE devices is usually a mixture of polymer and cross-linking agent with acidic solution or ionic liquids. Weak protonic acid and polymer blends are often used in PANI-based devices,<sup>54,55,57</sup> and ionic liquid is the more common choice for carbon-based nanomaterials and PT.<sup>9,20,58</sup>

For CP systems, low voltage operation and flexibility in molecular design is useful to incorporate a wide range of applications. Solution processable CPs are also widely available for large scale fabrication on rough surface and different contours. On the other hand, carbon-based nanomaterials offer exceptional EC performance with atomically thin layers. The unique electronic structure also provides electrochemical engineering with high precision. Both families of IR-EC materials have been demonstrated on device with large active area, high performance, straightforward implementation, and human body surface with different curvatures [Fig. 2(k)].<sup>1,9</sup>

#### IMPROVING THE WEARABILITY: FROM PLANAR TO FABRIC WeaVE

While planar WeaVE devices successfully address the issue of radiative thermal management in an efficient and sustainable manner, there are still plenty of opportunities and challenges to increase user comfort and experience through wearability.<sup>83</sup> Many improvements can be achieved by transitioning from planar WeaVE devices to fabric WeaVE devices. For example, woven textile features great flexibility, durability, and continuous fabrication.<sup>67,69–72,84–86</sup> In addition, multi-modal WeaVE devices that allow regulation of water vapor permeability and air convection can provide an additional level of adaptability in responding to different environments.<sup>2,4,5,7,60,61,87,88</sup> Moisture-responsive fabrics and textiles have demonstrated high efficiency in passively adaptive thermoregulation through evaporation and sweat release.<sup>89</sup>

To add dynamic emissivity tunability into textiles, IR-electrochromic fabric can be a promising first step to integrate multiple thermoregulation mechanisms. However, fabrication can be challenging regarding process complexity, uniformity of electrical contact, and



**FIG. 2.** Recent progress of WeaVE-like devices for thermoregulation. (a)–(e) CP-based WeaVE devices with different device configurations. (a) Kirigami-patterned PANI/Nylon, (b) PANI/P(VDF-HFP), (c) PANI/Nylon, (d) PANI/CNT, and (e) PEDOT. (f)–(h) Planar WeaVE devices with carbon nanomaterials. (f) Components and (g) thermoregulation performance of a multi-layer graphene thermoregulation device. (h) MLG/PE, (i) CNT film, and (j) CNT grid/PEDOT devices. (k) Thermal images of CP- and CNT-based WeaVE on human body parts with different curvatures at cooling and heating state. (l) Fiber-based WeaVE device with carbon nanotube as IR-EC coating. \* (a) Reproduced with permission from Chen *et al.*, PNAS Nexus 2(6), pgad165 (2023). Copyright 2023 Authors. (b) Reproduced with permission from Xu *et al.*, Sol. Energy Mater. Sol. Cells 208, 110356 (2020). Copyright 2020 Elsevier. (c) Reproduced with permission from Fu *et al.*, J. Mater. Chem. A 12(11), 6351–6358 (2024). Copyright 2024 Royal Society of Chemistry. (d) Reproduced with permission from Gao *et al.*, ACS Appl. Mater. Interfaces 16(23), 30421–30429 (2024). Copyright 2024 American Chemical Society. (e) Reproduced with permission from Brooke *et al.*, J. Mater. Chem. C 5(23), 5824–5830 (2017). Copyright 2024 the Royal Society of Chemistry. (f) Reproduced with permission from Ergoktas *et al.*, Nano Lett. 20(7), 5346–5352 (2020). Copyright 2020 American Chemical Society. (g) Reproduced with permission from Ergoktas *et al.*, Nat. Photonics 15(7), 493–498 (2021). Copyright 2021 Springer Nature. (h) Reproduced with permission from Yu *et al.*, ACS Nano 17(12), 11583–11592 (2023). Copyright 2023 American Chemical Society. (i) Reproduced from Ref. with permission from Sun *et al.*, Adv. Opt. Mater. 9(3), 2001216 (2020). Copyright 2020 John Wiley and Sons. (j) Reproduced with permission from Shi *et al.*, Mater. Today Chem. 39, 102166 (2024). Elsevier. (k) Reproduced with permission from Chen *et al.*, PNAS Nexus 2(6), pgad165 (2023). Copyright 2023 Authors and Sun *et al.*, Adv. Opt. Mater. 9(3), 2001216 (2020). Copyright 2020 John Wiley and Sons. (l) Reproduced with permission from Fan *et al.*, Adv. Funct. Mater. 34(16), 2310858 (2023). Copyright 2023 John Wiley and Sons.

mechanical strength. One promising material candidate is carbon-based fibers, which exhibit uniform morphology, simple chemical composition, and excellent processability, making them suitable for fabrication in high-temperature treatment and solution process. However, to apply high-performance IR-EC materials onto carbon fibers, one must first overcome solubility and processability issues.<sup>59,90</sup>

In addition to IR-EC materials, development of electrolyte and current collectors with compatible physical and chemical properties is also necessary but needs further exploration. Several fiber structures have been used to accommodate different components of a standalone electrochemical cell while maintaining wearability for different applications, from supercapacitors to displays.<sup>3,90–96</sup> In particular, Fan *et al.* demonstrated tunable woven textile with CNT as IR-EC material for

personal thermoregulation [Fig. 2(l)].<sup>13</sup> In these fiber cells, the configuration, design, choice of materials, and fabrication process are all crucial factors for the electrochemical performance and mechanical property.

For fiber or fabric WeaVE devices, electrical contact and chemical stability overlap with those of other electrochemical devices. However, with the thermal emissivity being a surface property, IR-transparent passivation layer with minimum thickness and electrochromic materials with maximized surface area exposed will be the major design considerations on the device level. For the materials, these requirements also imply the significance of mixed ionic and electronic transport in the three-phase contact interface of current collector, electrolyte, and the active layer.

### CHARGE TRANSPORT IN IR-ELECTROCHROMIC MATERIALS

As an atomic and molecular level fundamental property, the charge transport mechanism in IR-EC materials is the fundamental mechanism that determines the optical properties and is highly dependent on the structure and synthesis [Figs. 3(a) and 3(b)].<sup>28,44,48,97–101</sup> While many performance-driven WeaVE devices have been demonstrated, the huge potential of variable emissivity in IR-EC materials is still not fully utilized. One reason is the difficulty in understanding the charge transport properties in highly disordered IR-electrochromic materials.<sup>32,35,41</sup> In addition, ionic and electronic transport is strongly coupled and associated with processing and synthetic conditions.<sup>24,25,102</sup> Only when these fundamental knowledge gaps are filled can the performance of WeaVE device be improved consequently.

One key phenomenon to understand structure-dependent charge transport is insulator-to-metal transition. Theoretically, considering the wide variety of species and oxidation states from highly insulating to metallic, the mid-IR optical properties of CPs and carbon nanomaterials can vary all the way from transmissive to intrinsically reflective.<sup>48,80,98,100,101,103–105</sup> However, depending on the molecular and electronic structure, this wide tuning range involves transition between several different transport mechanisms, which is complicated to predict when selecting the desired materials.<sup>28,34</sup> The level of challenge can be further increased by the technical barriers in structural characterization and unclear charge distribution profile.<sup>33,106</sup>

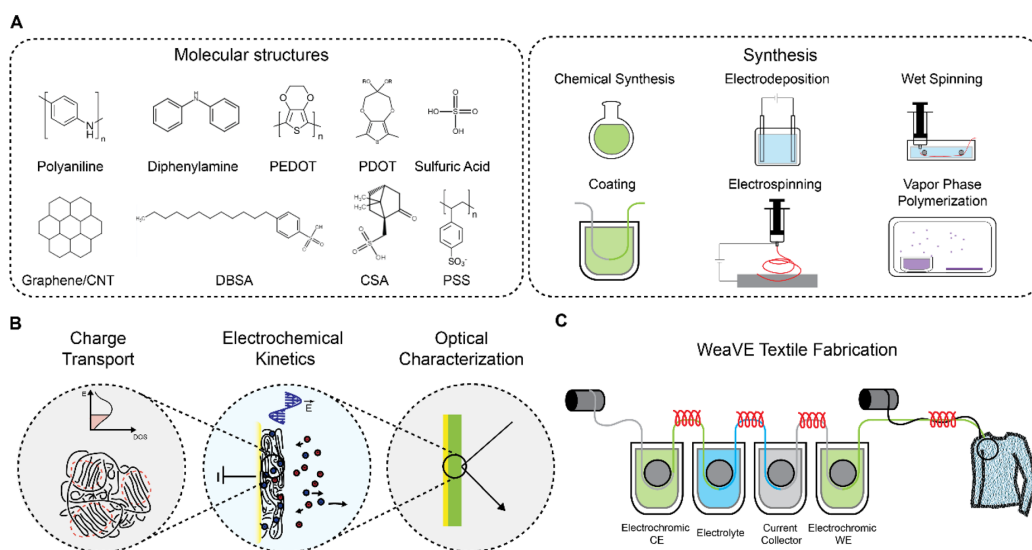
To solve these problems, the knowledge gap can be explored by developing fundamental characterization and theories. Deep understanding and precise molecular design can be more easily achieved when we clearly define the parameters and digitize the fundamental materials' properties.<sup>36,106</sup> Extensive studies have delved into measuring

charge carrier concentration and plasmonic properties for crystalline CPs.<sup>40,44</sup> In-depth studies of weakly localized systems have also been demonstrated both theoretically and experimentally since the discovery of CPs.<sup>45,98</sup> For most active materials in WeaVE devices, a more sophisticated and careful explanation is required since the structural disorder and the correlated interaction between charge carriers are particularly strong. Theoretical studies establishing the dynamic structure-property relation can serve as an important guideline to future materials design for WeaVE devices, since very limited discussion has been focused on the charge transport mechanism under different potential beyond classical band theories.

While it is nontrivial to perform rigorous characterization in atomic-level charge transport for conjugated polymers, we would like to point out that the mid-IR modulation in most WeaVE devices is the electrochemical engineering of delocalization in a disordered macromolecular system.<sup>32,37,38,107</sup> It is also expected that disorder engineering combined with theoretical studies will allow us to cultivate a deeper and more precise understanding of the relationship between different but coupled properties.<sup>29,35,43,108,109</sup> By quantifying the factors contributing to delocalization and combining the knowledge of contemporary solid-state physics and soft matter physics, we can capture the mechanism across strongly localized to metallic transport.

### ELECTROCHEMICAL KINETICS

Electrochemical kinetics on the molecular level is equally important [Fig. 3(b)]. Electrochemical tuning is a transient behavior involving diffusion and chemical reactions in materials, electrolytes, and their interfaces. In IR-electrochromic applications, almost all physical properties, from ionic conductivity to optical response, depend strongly on kinetics. It is also worth noting that although the switching



**FIG. 3.** Design process from molecular engineering to WeaVE device fabrication. (a) Commonly used monomer and dopant building blocks for IR-electrochromic material design with various synthesis methods for structural control. (b) Characterization and mechanism studies at different scales from charge transport, electrochemical kinetics, and dynamic optical properties. (c) Continuous and scalable manufacturing of standalone WeaVE textiles.

speed on the order of second is sufficient for personal thermoregulation, the thermal or optical contrast is also linked to the molecular structures and electrochemical kinetics.<sup>42,103,104</sup> While extensive studies have been carried out to demonstrate the electrochromic performance on the device level, the research on the micro- and mesoscopic behavior is still in its early stage.

Relative studies are emerging and rapidly increasing in the field of organic mixed ionic-electronic conductors (OMIEC).<sup>23,25,35,110</sup> By carefully selecting materials allowing x-ray penetration, *in situ* grazing-incidence wide-angle x-ray scattering (GIWAXS) can reveal the structural evolution.<sup>64–66</sup> For instance, studies suggest crystalline CPs will experience asymmetric dimensional change during the redox reaction, which may result in a long switching time and unreliable performance.<sup>111</sup> This in-depth understanding of the kinetic correlation between charge transport, structural evolution, and optical contrast in CPs is needed to improve their electrochromic performance.

At the atomic and molecular level, electrochemical quartz crystal microbalance with dissipation (EQCM-D) provides accurate mass change on the order of nanogram through mechanical response and vibrational analysis.<sup>112</sup> For electrochemically active soft materials with synergistic effects of ionic and electronic transport, the charge efficiency and electrochemical kinetics will be affected significantly by the structures, volumetric change, and charge transfer at the interface with the electrolyte. On top of that, both the solvent effect and electrochemical reactions will introduce mass change in the active material. By carefully analyzing the results from EQCM-D, ionic diffusion and solvent swelling can be systematically studied and microscopically monitored in real time, even with materials with porous structure and viscoelasticity.<sup>112–114</sup>

By combining the information in electrochemical kinetics and optical properties, we can potentially expand the emissivity contrast and improve the electrochromic figure-of-merits. For example, by comparing IR-electrochromic CPs with different tuning mechanisms, we can observe a trade-off between electrical conductivity and dynamic emissivity contrast.<sup>20</sup> In the visible light regime, recent studies also show the kinetically dependent electrochromic response is a direct result of ion and carrier mobility in different electronic structures.<sup>34,115</sup> The electrochemical kinetics of CNTs and graphene also show significant dependency on structures. Hypotheses and qualitative observations like these can be confirmed by rigorous studies on the electrochemical kinetics at a microscopic scale.

As a result, the understanding of electrochemical kinetics will rely on the structural characterization across multiple scales. Recently, aside from scanning probe microscopy and X-ray scattering, transmission electron microscopy has also joined the toolset for structural studies.<sup>62</sup> From statistical and empirical interpretation of collective pictures to detailed and specific description in significantly higher spatial resolution, complex reasoning is necessary to improve the dynamic tuning range and provide a unifying explanation to connect all the structure-properties relationships in IR-electrochromic materials.

### DYNAMIC OPTICAL PROPERTIES

With the understanding of structural and electrochemical properties, extra focus should be brought to characterize the dynamic optical properties of IR-electrochromic materials [Fig. 3(b)]. The working principle of emissivity tuning can be most accurately quantified with the change of complex optical dispersion in the IR region, and the dynamic optical property is governed by electrochemically driven

insulator-to-metal transition and Fermi level engineering. Rigorous characterization of optical properties can enable numerical design and simulation for different tunable and directional nanophotonic devices. The intermediate states during the continuous transition from one state to another are particularly useful for controlling and predicting the thermal emissivity of WeaVE devices. By combining techniques in different frequency ranges, rigorous optical characterization bridges the gap between polymer science and photonic engineering.

Integrating the dynamic change of complex refractive index along with data on materials structures, electrochemical kinetics, and synthesis processes will cultivate a comprehensive structure-properties relationship. In particular, frequencies of IR waves are lower than visible light but higher than THz and microwaves. The optical conductivities and responses in IR can be perceived as the middle ground, affected by molecular and electronic structures at multiple length scales. The dispersive complex refractive indices of IR-electrochromic materials not only extend the concept of charge transport from DC to AC but also provide vibrational excitation features in IR, such as IR-activated vibrational (IRAV) modes, degree of localization from inelastic scattering, and absorption due to carrier band transitions of polarons and bipolarons.<sup>42,99,116</sup>

With comprehensive results of data on dynamic optical properties, we can accurately identify the origin of thermal emissivity tuning. For example, in IR-electrochromic CPs, the high emissivity state may correspond to either a conducting form or an insulating form, depending on the absorption mechanism of the materials.<sup>117</sup> For highly disordered systems, like electropolymerized CPs, the conducting state has higher absorption due to the strong disorder and the dopant-induced formation of polarons and bipolaron.<sup>99,105,118</sup> On the other hand, the tuning of intrinsically reflecting CPs in IR is likely due to carrier concentration change.<sup>100</sup>

For correlated materials, the change in optical properties dispersion at different states can provide quantitative understanding of doping-level dependent coupling features through light-matter interaction. The modeling of optical properties can also provide information of preferred orientation and anisotropy.<sup>44,119</sup> This is particularly useful for heterogeneous materials like CPs, whose optical properties are sometimes anisotropic and exhibit different electrical and optical conductivity in different chemical and oxidation states.<sup>119</sup>

To date, the dynamic optical dispersion for most electrochromic CPs in the IR region is still almost unexamined. *Ex situ* studies with spectroscopic ellipsometry will be a good starting point in IR. It is a powerful technique to measure the polarization change through light-matter interaction and complex optical dispersion modeling in multilayer samples with uniform thickness. For materials with very complicated optical properties, especially in the intermediate state of insulator-to-metal transition, ellipsometry does not assume the Kramers-Kronig consistency during measurement, and the data can be fitted with customized charge transport models, providing a significant degree of freedom and the ability to consider at different wavelengths. Comparison between samples with different processes, substrates, and thicknesses will also be informative, since the optical properties depend highly on the process and complicated structural effects and disorder scattering.<sup>35</sup>

The goal of optical characterization is *in situ* studies for structural, optical, and chemical evolutions, from static to high-frequency optical limits.<sup>102</sup> The real-time details in kinetics can best reflect the potential

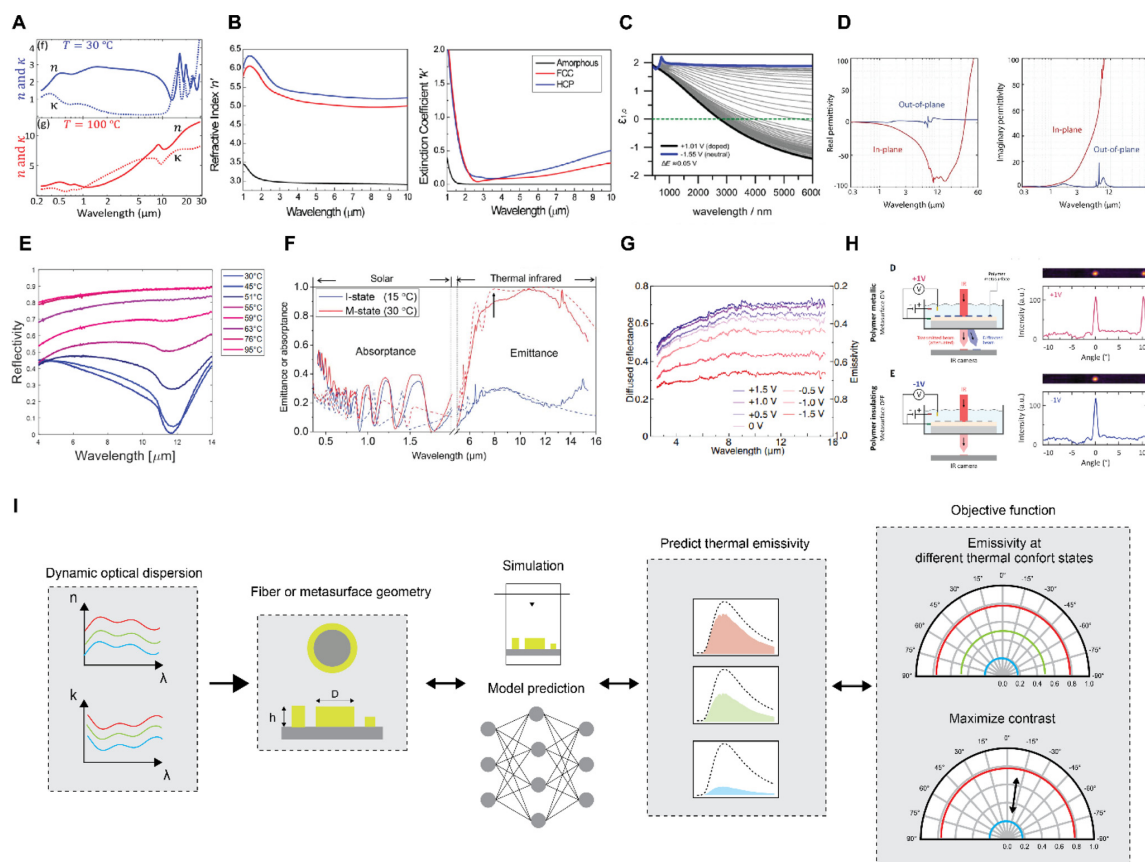
dependency. However, many well-established available in visible light do not apply to IR since the choice of IR-transparent electrolyte is very limited. With extremely high absorbance in mid-IR, a well-designed setup with IR-absorptive electrolyte on the back might be the best solution for future studies.

### DYNAMIC NANOPHOTONIC STRUCTURES AND MATERIALS FOR THERMOREGULATION

Finally, we extend the topic of interest to physics and potential applications by incorporating nanophotonic structures with different materials systems to WeaVE devices based on the established

foundation Dynamic optical properties [Figs. 3(b) and 3(c)]. Usually consisting of periodic blocks of metallic and dielectric materials, metasurfaces manipulate the light wave by creating subwavelength boundary conditions with the contrast of different complex refractive indices. It is achievable exotic optical responses, such as perfect absorption, anomalous reflection and transmission, and generating special wavefronts by utilizing precise design and fabrication based on accurate optical properties data.

While *in situ* characterization of complex refractive indices can be challenging in electrochemically driven materials, the optical properties in IR and microstructure evolution of inorganic phase-changing



**FIG. 4.** Experimental demonstrations and future opportunities for dynamic IR modulation by incorporating nanophotonic applications with IR-tunable materials. (a)–(d) Optical properties of commonly used phase-changing materials and IR-EC CPs (a)  $\text{VO}_2$ , (b)  $\text{GeSbTe}$ , (c)  $\text{PEDOT:PSS}$ , (d)  $\text{PEDOT:Sulf}$ , (e)–(h) thin-film and nanophotonic applications in the IR region, (e) reflectance tuning using  $\text{VO}_2$  thin-film absorber, (f) dynamic building envelope by nanophotonics structure based on  $\text{VO}_2$ , (g) CP and cellulose paper thin-film device for thermal emissivity tuning, (h) electrically tunable beam-steering metasurface with  $\text{PEDOT:PSS}$ , and (i) flow chart for nanophotonic structure design based on experimental data input. Forward and inverse design focused on controlling thermal emissivity and maximizing tuning range. (a) Reproduced with permission from Wan *et al.*, *Ann. Phys.* **531**(10), 1900188 (2019). Copyright 2019 John Wiley and Sons.<sup>121</sup> (b) Reprinted with permission from Julian *et al.*, *Optica* **7**(7), 746 (2020). Copyright 2024 Optica Publishing Group.<sup>125</sup> (c) Reproduced with permission from Dingler *et al.*, *Macromolecules* **55**(5), 1600–1608 (2022). Copyright 2022 American Chemical Society.<sup>51</sup> (d) Reproduced with permission from Chen *et al.*, *Nat. Nanotechnol.* **15**(1), 35–40 (2020). Copyright 2020 Springer Nature.<sup>145</sup> (e) Reproduced with the permission from Larciprete *et al.*, *APL Mater.* **11**(9), 091107 (2023). Copyright 2023 AIP Publishing.<sup>131</sup> (f) Reprinted with permission from Tang *et al.*, *Science* **374**(6574), 1504–1509 (2021). Copyright 2021 AAAS.<sup>136</sup> (g) Reproduced with permission from Kuang *et al.*, *npj Flexible Electron.* **8**(1), 55 (2024). Copyright 2024 Springer Nature.<sup>78</sup> (h) Reproduced with permission from Karst *et al.*, *Science* **374**(6567), 612–616 (2021). Copyright 2021 AAAS.<sup>147</sup>

materials are relatively well studied across different phase-changing mechanisms<sup>120–125</sup> [Figs. 4(a)–4(d)]. These materials are usually thermochromic, exhibiting significant and reversible contrast between metallic and dielectric state in varying temperatures. Their compatibility with existing nanofabrication techniques of also makes them widely adopted choices in nanophotonic devices with small feature sizes.<sup>126–129</sup>

In particular, with the benefit of high IR emissivity and absorption, metasurface and thin-film perfect absorbers have been extensively used as platforms for phase-changing materials in thermal and sustainability applications<sup>123,130–135</sup> [Figs. 4(e) and 4(f)]. For dynamic building envelopes, the thermally induced insulator-to-metal transition of tungsten (W) doped vanadium dioxide (VO<sub>2</sub>) was used in electromagnetic perfect absorber design to manipulate the thermal emission at different ambient temperature.<sup>136,137</sup> For potential applications working at human body temperature, the phase transition temperature in these materials can be effectively decreased by different doping and synthesis strategies.<sup>120,138,139</sup>

Considering the broadband and large tuning range with active control in wearable devices, incorporating soft electrochromic materials with metamaterials allows us to utilize more degrees of freedom and boost the efficiency.<sup>140</sup> Complicated structures can be utilized to maximize the broadband emissivity contrast.<sup>141,142</sup> Using and electrochromic materials in metasurface for thermal and optical applications has also been numerically and experimentally demonstrated for dynamic switching,<sup>143–145</sup> holographic elements,<sup>146</sup> and beam steering applications<sup>129,147</sup> [Figs. 4(g) and 4(h)].

However, the combination of WeaVE devices and nanophotonic structures is almost unexplored. Nanofabrication and patterning while maintaining the electrochemical tunability is the top priority and challenge. Polythiophene-based CPs in organic electrochemical transistors are by far the most popular choices.<sup>143,148–150</sup> Techniques for fabricating other IR-electrochromic materials with large dynamic range at nanoscale are still awaiting for future studies.

Although extensive simulation is often required, the time cost for design and optimization can also be significantly reduced by performing inverse design with smaller amount of data from experiments and simulations<sup>151</sup> [Fig. 4(i)]. Optimizations based on wide-angle emission can be taken into account and facilitated by combining research from co-designing materials structures and nanophotonics. We envision the small feature size with built-in dynamic functionality will pave the way for developing the next generation of WeaVE textiles.

## CONCLUSION

In conclusion, fast, effective, and actively controllable personal thermoregulation with minimal energy input is the ultimate goal for WeaVE devices. Personalized and autonomous thermal feedback with highly efficient IR-EC materials provides the potential for adapting all daily scenarios in one piece of WeaVE textile with less energy consumption. To identify the knowledge gap, we summarize the goals and challenges in developing WeaVE devices, emphasizing the integration of wearability and electrochromism. We then highlight the interdisciplinary efforts required from the angle of charge transport, optical characterization, and electrochemical kinetics, pointing out that the controllable insulator-to-metal transition is the key to developing materials with high mid-IR emissivity contrast. The research will build upon massive amounts of data from in-depth studies and innovative experimental techniques, and researchers will be able to digitize the material's properties for further improvement. Finally, we use the

dynamic metasurface as an example to illustrate the integration of properly defined physical quantities and characterization can enhance the performance of WeaVE devices.

## ACKNOWLEDGMENTS

T.-H.C. and P.-C.H. thank the Pritzker School of Molecular Engineering at the University of Chicago and the National Science Foundation (ECCS Award No. 2145933) for funding support. T.-H.C. thanks Sony Corporation for the WeaVE project.

## AUTHOR DECLARATIONS

### Conflict of Interest

The authors have no conflicts to disclose.

## Author Contributions

**Ting-Hsuan Chen:** Visualization (equal); Writing – original draft (equal); Writing – review & editing (equal). **Po-Chun Hsu:** Writing – original draft (equal); Writing – review & editing (equal).

## DATA AVAILABILITY

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

## REFERENCES

- T. H. Chen, Y. Hong, C. T. Fu, A. Nandi, W. Xie, J. Yin, and P. C. Hsu, *PNAS Nexus* **2**(6), pgad165 (2023).
- R. Wu, T. H. Chen, and P. C. Hsu, *EcoMat* **5**(10), e12396 (2023).
- H. Fan, W. Wei, C. Hou, Q. Zhang, Y. Li, K. Li, and H. Wang, *J. Mater. Chem. C* **11**(22), 7183–7210 (2023).
- Y. Fang, G. Chen, M. Bick, and J. Chen, *Chem. Soc. Rev.* **50**(17), 9357–9374 (2021).
- P. C. Hsu and X. Li, *Science* **370**(6518), 784–785 (2020).
- H. Zhai, D. Fan, and Q. Li, *Nano Energy* **100**, 107435 (2022).
- L. Lei, S. Shi, D. Wang, S. Meng, J. G. Dai, S. Fu, and J. Hu, *ACS Nano* **17**(3), 1803–1830 (2023).
- H. Fu, L. Zhang, Y. Dong, C. Zhang, and W. Li, *Mater. Chem. Front.* **7**(12), 2337–2358 (2023).
- Y. Sun, H. Chang, J. Hu, Y. Wang, Y. Weng, C. Zhang, S. Niu, L. Cao, Z. Chen, N. Guo, J. Liu, J. Chi, G. Li, and L. Xiao, *Adv. Opt. Mater.* **9**(3), 2001216 (2020).
- H. Gong, J. Ai, W. Li, J. Zhu, Q. Zhang, J. Liu, Y. Jin, and H. Wang, *ACS Appl. Mater. Interfaces* **13**(42), 50319–50328 (2021).
- G. Fu, H. Gong, J. Xu, B. Zhuang, B. Rong, Q. Zhang, X. Chen, J. Liu, and H. Wang, *J. Mater. Chem. A* **12**(11), 6351–6358 (2024).
- B. Wang, G. Xu, S. Song, Z. Ren, X. Li, J. Li, X. Xu, W. Zhou, H. Li, L. Zhang, and Y. Li, *Electrochim. Acta* **390**, 138891 (2021).
- Q. Fan, H. Fan, H. Han, Z. Bai, X. Wu, C. Hou, Q. Zhang, Y. Li, K. Li, and H. Wang, *Adv. Funct. Mater.* **34**(16), 2310858 (2023).
- B. Wang, G. Xu, S. Song, Z. Ren, D. Liu, L. Hao, F. Xing, H. Wei, L. Zhang, and Y. Li, *Chem. Eng. J.* **445**, 136819 (2022).
- M. S. Ergoktas, G. Bakan, E. Kovalska, L. W. Le Fevre, R. P. Fields, P. Steiner, X. Yu, O. Salihoglu, S. Balci, V. I. Fal'ko, K. Novoselov, R. A. W. Dryfe, and C. Kocabas, *Nat. Photonics* **15**(7), 493–498 (2021).
- L. Zhang, B. Wang, X. Li, G. Xu, S. Dou, X. Zhang, X. Chen, J. Zhao, K. Zhang, and Y. Li, *J. Mater. Chem. C* **7**(32), 9878–9891 (2019).
- G. Xu, L. Zhang, B. Wang, X. Chen, S. Dou, M. Pan, F. Ren, X. Li, and Y. Li, *Sol. Energy Mater. Sol. Cells* **208**, 110356 (2020).
- S. Wu, C. Jia, X. Fu, X. Weng, J. Zhang, and L. Deng, *Electrochim. Acta* **88**, 322–329 (2013).

- <sup>19</sup>Y. Tian, X. Zhang, S. Dou, L. Zhang, H. Zhang, H. Lv, L. Wang, J. Zhao, and Y. Li, *Sol. Energy Mater. Sol. Cells* **170**, 120–126 (2017).
- <sup>20</sup>R. Brooke, E. Mitraka, S. Sardar, M. Sandberg, A. Sawatdee, M. Berggren, X. Crispin, and M. P. Jonsson, *J. Mater. Chem. C* **5**(23), 5824–5830 (2017).
- <sup>21</sup>X.-G. Li, H.-Y. Wang, and M.-R. Huang, *Macromolecules* **40**(5), 1489–1496 (2007).
- <sup>22</sup>Y. Wang, C. Zhu, R. Pfattner, H. Yan, L. Jin, S. Chen, F. Molina-Lopez, F. Lissel, J. Liu, N. I. Rabiah, Z. Chen, J. W. Chung, C. Linder, M. F. Toney, B. Murmann, and Z. Bao, *Sci. Adv.* **3**(3), e1602076 (2017).
- <sup>23</sup>B. D. Paulsen, K. Tybrandt, E. Stavrinidou, and J. Rivnay, *Nat. Mater.* **19**(1), 13–26 (2020).
- <sup>24</sup>N. A. Kukhta, A. Marks, and C. K. Luscombe, *Chem. Rev.* **122**(4), 4325–4355 (2022).
- <sup>25</sup>S. T. M. Tan, A. Gumyusenge, T. J. Quill, G. S. LeCroy, G. E. Bonacchini, I. Denti, and A. Salleo, *Adv. Mater.* **34**(21), e2110406 (2022).
- <sup>26</sup>D. Venkateshvaran, M. Nikolka, A. Sadhanala, V. Lemauro, M. Zelazny, M. Kepa, M. Hurhangee, A. J. Kronemeijer, V. Pecunia, I. Nasrallah, I. Romanov, K. Broch, I. McCulloch, D. Emin, Y. Olivier, J. Cornil, D. Beljonne, and H. Sirringhaus, *Nature* **515**(7527), 384–388 (2014).
- <sup>27</sup>F. Paquin, J. Rivnay, A. Salleo, N. Stingelin, and C. Silva-Acuña, *J. Mater. Chem. C* **3**(41), 10715–10722 (2015).
- <sup>28</sup>A. Ugur, F. Katmis, M. Li, L. Wu, Y. Zhu, K. K. Varanasi, and K. K. Gleason, *Adv. Mater.* **27**(31), 4604–4610 (2015).
- <sup>29</sup>I. Yavuz, B. N. Martin, J. Park, and K. N. Houk, *J. Am. Chem. Soc.* **137**(8), 2856–2866 (2015).
- <sup>30</sup>S. Kee, N. Kim, B. S. Kim, S. Park, Y. H. Jang, S. H. Lee, J. Kim, J. Kim, S. Kwon, and K. Lee, *Adv. Mater.* **28**(39), 8625–8631 (2016).
- <sup>31</sup>S. Dongmin Kang and G. Jeffrey Snyder, *Nat. Mater.* **16**(2), 252–257 (2017).
- <sup>32</sup>C. Liu, K. Huang, W.-T. Park, M. Li, T. Yang, X. Liu, L. Liang, T. Minari, and Y.-Y. Noh, *Mater. Horiz.* **4**(4), 608–618 (2017).
- <sup>33</sup>Y. Yao, H. Dong, F. Liu, T. P. Russell, and W. Hu, *Adv. Mater.* **29**(29), 1701251 (2017).
- <sup>34</sup>S. T. Keene, J. E. M. Laulainen, R. Pandya, M. Moser, C. Schnedermann, P. A. Midgley, I. McCulloch, A. Rao, and G. G. Malliaras, *Nat. Mater.* **22**(9), 1121–1127 (2023).
- <sup>35</sup>R. Noriega, J. Rivnay, K. Vandewal, F. P. Koch, N. Stingelin, P. Smith, M. F. Toney, and A. Salleo, *Nat. Mater.* **12**(11), 1038–1044 (2013).
- <sup>36</sup>I. E. Jacobs and A. J. Moulé, *Adv. Mater.* **29**(42), 1703063 (2017).
- <sup>37</sup>J. Liu, L. Qiu, R. Alessandri, X. Qiu, G. Portale, J. Dong, W. Talsma, G. Ye, A. A. Sengrhan, P. C. T. Souza, M. A. Loi, R. C. Chiechi, S. J. Marrink, J. C. Hummelen, and L. J. A. Koster, *Adv. Mater.* **30**(7), 1704630 (2018).
- <sup>38</sup>Y. Yang, Z. Liu, G. Zhang, X. Zhang, and D. Zhang, *Adv. Mater.* **31**(46), e1903104 (2019).
- <sup>39</sup>M. Seifrid, G. N. M. Reddy, B. F. Chmelka, and G. C. Bazan, *Nat. Rev. Mater.* **5**(12), 910–930 (2020).
- <sup>40</sup>I. E. Jacobs, G. D'Avino, V. Lemauro, Y. Lin, Y. Huang, C. Chen, T. F. Harrelson, W. Wood, L. J. Spalek, T. Mustafa, C. A. O'Keefe, X. Ren, D. Simatos, D. Tjhe, M. Statz, J. W. Strzalka, J. K. Lee, I. McCulloch, S. Fratini, D. Beljonne, and H. Sirringhaus, *J. Am. Chem. Soc.* **144**(7), 3005–3019 (2022).
- <sup>41</sup>Z. Peng, L. Ye, and H. Ade, *Mater. Horiz.* **9**(2), 577–606 (2022).
- <sup>42</sup>D. E. Shen, C. L. Goins, A. L. Jones, A. M. Österholm, and J. R. Reynolds, *ACS Mater. Lett.* **6**(2), 528–534 (2024).
- <sup>43</sup>R. Wu, D. Meli, J. Strzalka, S. Narayanan, Q. Zhang, B. D. Paulsen, J. Rivnay, and C. J. Takacs, *Nat. Mater.* **23**, 648–655 (2024).
- <sup>44</sup>S. Chen, P. Kühne, V. Stanishchev, S. Knight, R. Brooke, I. Petsagkourakis, X. Crispin, M. Schubert, V. Darakchieva, and M. P. Jonsson, *J. Mater. Chem. C* **7**(15), 4350–4362 (2019).
- <sup>45</sup>K. Lee, A. J. Heeger, and Y. Cao, *Phys. Rev. B* **48**(20), 14884–14891 (1993).
- <sup>46</sup>A. J. Heeger, *Phys. Scr.* **T102**(1), 30–35 (2002).
- <sup>47</sup>M. Schubert, C. Bundesmann, H. V. Wenckstern, G. Jakopic, A. Haase, N. K. Persson, F. Zhang, H. Arwin, and O. Inganäs, *Appl. Phys. Lett.* **84**(8), 1311–1313 (2004).
- <sup>48</sup>K. Lee, R. Menon, C. O. Yoon, and A. J. Heeger, *Phys. Rev. B* **52**(7), 4779–4787 (1995).
- <sup>49</sup>A. Redondo, E. A. Ticianelli, and S. Gottesfeld, *Mol. Cryst. Liq. Cryst. Incorporating Nonlinear Opt.* **160**(1), 185–203 (1988).
- <sup>50</sup>S. Rossi, O. Olsson, S. Chen, R. Shanker, D. Banerjee, A. Dahlin, and M. P. Jonsson, *Adv. Mater.* **33**(49), e2105004 (2021).
- <sup>51</sup>C. Dingler, R. Walter, B. Gompf, and S. Ludwigs, *Macromolecules* **55**(5), 1600–1608 (2022).
- <sup>52</sup>C. Cobet, K. Oppelt, K. Hingerl, H. Neugebauer, G. Knör, N. S. Sariciftci, and J. Gasiorowski, *J. Phys. Chem. C* **122**(42), 24309–24320 (2018).
- <sup>53</sup>J. Chen, Y. He, and L. Li, *Thin Solid Films* **762**, 139565 (2022).
- <sup>54</sup>P. Chandrasekhar, B. J. Zay, D. Lawrence, E. Caldwell, R. Sheth, R. Stephan, and J. Cornwell, *J. Appl. Polym. Sci.* **131**(19), 40850 (2014).
- <sup>55</sup>P. Chandrasekhar, B. J. Zay, G. C. Birur, S. Rawal, E. A. Pierson, L. Kauder, and T. Swanson, *Adv. Funct. Mater.* **12**(2), 95–103 (2002).
- <sup>56</sup>H. Fan, K. Li, X. Liu, K. Xu, Y. Su, C. Hou, Q. Zhang, Y. Li, and H. Wang, *ACS Appl. Mater. Interfaces* **12**(25), 28451–28460 (2020).
- <sup>57</sup>L. Zhang, G. Xia, X. Li, G. Xu, B. Wang, D. Li, A. Gavriluk, J. Zhao, and Y. Li, *Synth. Met.* **248**, 88–93 (2019).
- <sup>58</sup>M. S. Ergoktas, G. Bakan, P. Steiner, C. Bartlam, Y. Malevich, E. Ozden-Yenigun, G. He, N. Karim, P. Cataldi, M. A. Bissett, I. A. Kinloch, K. S. Novoselov, and C. Kocabas, *Nano Lett.* **20**(7), 5346–5352 (2020).
- <sup>59</sup>F. Sun, H. Jiang, H. Wang, Y. Zhong, Y. Xu, Y. Xing, M. Yu, L. W. Feng, Z. Tang, J. Liu, H. Sun, H. Wang, G. Wang, and M. Zhu, *Chem. Rev.* **123**(8), 4693–4763 (2023).
- <sup>60</sup>Y. Peng and Y. Cui, *Joule* **4**(4), 724–742 (2020).
- <sup>61</sup>R. Hu, Y. Liu, S. Shin, S. Huang, X. Ren, W. Shu, J. Cheng, G. Tao, W. Xu, R. Chen, and X. Luo, *Adv. Energy Mater.* **10**(17), 1903921 (2020).
- <sup>62</sup>L. Balhorn, Q. MacPherson, K. C. Bustillo, C. J. Takacs, A. J. Spakowitz, and A. Salleo, *Proc. Natl. Acad. Sci. U. S. A.* **119**(46), e2204346119 (2022).
- <sup>63</sup>O. Panova, C. Ophus, C. J. Takacs, K. C. Bustillo, L. Balhorn, A. Salleo, N. Balsara, and A. M. Minor, *Nat. Mater.* **18**(8), 860–865 (2019).
- <sup>64</sup>B. D. Paulsen, R. Wu, C. J. Takacs, H. G. Steinruck, J. Strzalka, Q. Zhang, M. F. Toney, and J. Rivnay, *Adv. Mater.* **32**(40), e2003404 (2020).
- <sup>65</sup>B. D. Paulsen, A. Giovannitti, R. Wu, J. Strzalka, Q. Zhang, J. Rivnay, and C. J. Takacs, *Small* **17**(42), e2103213 (2021).
- <sup>66</sup>T. J. Quill, G. LeCroy, D. M. Halat, R. Sheelamantula, A. Marks, L. S. Grundy, I. McCulloch, J. A. Reimer, N. P. Balsara, A. Giovannitti, A. Salleo, and C. J. Takacs, *Nat. Mater.* **22**(3), 362–368 (2023).
- <sup>67</sup>Y. Ding, M. A. Invernale, and G. A. Sotzing, *ACS Appl. Mater. Interfaces* **2**(6), 1588–1593 (2010).
- <sup>68</sup>P. B. Catrysse, A. Y. Song, and S. Fan, *ACS Photonics* **3**(12), 2420–2426 (2016).
- <sup>69</sup>K. Fu, Z. Yang, Y. Pei, Y. Wang, B. Xu, Y. Wang, B. Yang, and L. Hu, *Adv. Fiber Mater.* **1**(1), 61–70 (2019).
- <sup>70</sup>L. Lao, D. Shou, Y. S. Wu, and J. T. Fan, *Sci. Adv.* **6**(14), eaaz0013 (2020).
- <sup>71</sup>M. Alberghini, S. Hong, L. M. Lozano, V. Korolovych, Y. Huang, F. Signorato, S. H. Zandavi, C. Fucetola, I. Uluturk, M. Y. Tolstorukov, G. Chen, P. Asinari, R. M. Osgood, M. Fasano, and S. V. Boriskina, *Nat. Sustainable* **4**(8), 715–724 (2021).
- <sup>72</sup>D. Miao, X. Wang, J. Yu, and B. Ding, *Adv. Funct. Mater.* **31**(14), 2008705 (2021).
- <sup>73</sup>S. Zeng, S. Pian, M. Su, Z. Wang, M. Wu, X. Liu, M. Chen, Y. Xiang, J. Wu, M. Zhang, Q. Cen, Y. Tang, X. Zhou, Z. Huang, R. Wang, A. Tunuhe, X. Sun, Z. Xia, M. Tian, M. Chen, X. Ma, L. Yang, J. Zhou, H. Zhou, Q. Yang, X. Li, Y. Ma, and G. Tao, *Science* **373**(6555), 692–696 (2021).
- <sup>74</sup>S. Zhong, L. Yi, J. Zhang, T. Xu, L. Xu, X. Zhang, T. Zuo, and Y. Cai, *Chem. Eng. J.* **407**, 127104 (2021).
- <sup>75</sup>N. Kobayashi, H. Izumi, and Y. Morimoto, *J. Occup. Health* **59**(5), 394–407 (2017).
- <sup>76</sup>G. Petroffé, L. Beouch, S. Cantin, P.-H. Aubert, C. Plesse, J.-P. Duden, F. Vidal, and C. Chevrot, *Sol. Energy Mater. Sol. Cells* **177**, 23–31 (2018).
- <sup>77</sup>J. Gao, J. Zhou, M. Yuan, S. Yu, W. Ma, Z. Hu, H. Xiang, and M. Zhu, *ACS Appl. Mater. Interfaces* **16**(23), 30421–30429 (2024).
- <sup>78</sup>C. Kuang, S. Chen, M. Liao, A. Rahmanudin, D. Banerjee, J. Edberg, K. Tybrandt, D. Zhao, and M. P. Jonsson, *npj Flexible Electron.* **8**(1), 55 (2024).
- <sup>79</sup>P. Topart and P. Hourquebie, *Thin Solid Films* **352**(1–2), 243–248 (1999).
- <sup>80</sup>F. Wang, Y. Zhang, C. Tian, C. Girit, A. Zettl, M. Crommie, and Y. R. Shen, *Science* **320**(5873), 206–209 (2008).
- <sup>81</sup>X. Yu, G. Bakan, H. Guo, M. S. Ergoktas, P. Steiner, and C. Kocabas, *ACS Nano* **17**(12), 11583–11592 (2023).

- <sup>82</sup>G. Shi, H. Fan, W. Wang, C. Hou, Q. Zhang, Y. Li, H. Xiao, G. Dai, K. Li, and H. Wang, *Mater. Today Chem.* **39**, 102166 (2024).
- <sup>83</sup>W. Yang, S. Lin, W. Gong, R. Lin, C. Jiang, X. Yang, Y. Hu, J. Wang, X. Xiao, K. Li, Y. Li, Q. Zhang, J. S. Ho, Y. Liu, C. Hou, and H. Wang, *Science* **384**(6691), 74–81 (2024).
- <sup>84</sup>Y. Zhou, J. Fang, H. Wang, H. Zhou, G. Yan, Y. Zhao, L. Dai, and T. Lin, *Adv. Electron. Mater.* **4**(5), 1800104 (2018).
- <sup>85</sup>J. Mandal, M. Jia, A. Overvig, Y. Fu, E. Che, N. Yu, and Y. Yang, *Joule* **3**(12), 3088–3099 (2019).
- <sup>86</sup>T. G. Yun, M. Park, D. H. Kim, D. Kim, J. Y. Cheong, J. G. Bae, S. M. Han, and I. D. Kim, *ACS Nano* **13**(3), 3141–3150 (2019).
- <sup>87</sup>A. Aili, T. Jiang, J. Chen, Y. Wen, R. Yang, X. Yin, and G. Tan, *Next Energy* **3**, 100121 (2024).
- <sup>88</sup>X. Zhang, F. Wang, H. Guo, F. Sun, X. Li, C. Zhang, C. Yu, and X. Qin, *Adv. Sci.* **11**(10), e2305228 (2024).
- <sup>89</sup>X. Li, W. Guo, and P. C. Hsu, *Adv. Mater.* **36**(12), e2209825 (2024).
- <sup>90</sup>B. Fang, J. Yan, D. Chang, J. Piao, K. M. Ma, Q. Gu, P. Gao, Y. Chai, and X. Tao, *Nat. Commun.* **13**(1), 2101 (2022).
- <sup>91</sup>Z. Zhang, K. Guo, Y. Li, X. Li, G. Guan, H. Li, Y. Luo, F. Zhao, Q. Zhang, B. Wei, Q. Pei, and H. Peng, *Nat. Photonics* **9**(4), 233–238 (2015).
- <sup>92</sup>Z. Zhang, X. Chen, P. Chen, G. Guan, L. Qiu, H. Lin, Z. Yang, W. Bai, Y. Luo, and H. Peng, *Adv. Mater.* **26**(3), 466–470 (2014).
- <sup>93</sup>D. Chen, K. Jiang, T. Huang, and G. Shen, *Adv. Mater.* **32**(5), e1901806 (2020).
- <sup>94</sup>T. Qin, S. Peng, J. Hao, Y. Wen, Z. Wang, X. Wang, D. He, J. Zhang, J. Hou, and G. Cao, *Adv. Energy Mater.* **7**(20), 1700409 (2017).
- <sup>95</sup>B. Saravanakumar, S. S. Jayaseelan, M. K. Seo, H. Y. Kim, and B. S. Kim, *Nanoscale* **9**(47), 18819–18834 (2017).
- <sup>96</sup>J. Yu, W. Lu, J. P. Smith, K. S. Booksh, L. Meng, Y. Huang, Q. Li, J. H. Byun, Y. Oh, Y. Yan, and T. W. Chou, *Adv. Energy Mater.* **7**(3), 1600976 (2016).
- <sup>97</sup>V. Coropceanu, J. Cornil, D. A. da Silva Filho, Y. Olivier, R. Silbey, and J. L. Bredas, *Chem. Rev.* **107**(4), 926–952 (2007).
- <sup>98</sup>A. G. MacDiarmid and A. J. Epstein, *Faraday Discuss. Chem. Soc.* **88**, 317 (1989).
- <sup>99</sup>R. P. McCall, J. M. Ginder, M. G. Roe, G. E. Asturias, E. M. Scherr, A. G. MacDiarmid, and A. J. Epstein, *Phys. Rev. B* **39**(14), 10174–10178 (1989).
- <sup>100</sup>K. Lee, S. Cho, S. H. Park, A. J. Heeger, C. W. Lee, and S. H. Lee, *Nature* **441**(7089), 65–68 (2006).
- <sup>101</sup>J. Hwang, D. B. Tanner, I. Schwendeman, and J. R. Reynolds, *Phys. Rev. B* **67**(11), 115205 (2003).
- <sup>102</sup>R. Wu, M. Matta, B. D. Paulsen, and J. Rivnay, *Chem. Rev.* **122**(4), 4493–4551 (2022).
- <sup>103</sup>W. Bao, J. Wan, X. Han, X. Cai, H. Zhu, D. Kim, D. Ma, Y. Xu, J. N. Munday, H. D. Drew, M. S. Fuhrer, and L. Hu, *Nat. Commun.* **5**, 4224 (2014).
- <sup>104</sup>A. M. Dimiev, G. Ceriotti, N. Behabtu, D. Zakhidov, M. Pasquali, R. Saito, and J. M. Tour, *ACS Nano* **7**(3), 2773–2780 (2013).
- <sup>105</sup>N. F. Mott and M. Kaveh, *Adv. Phys.* **34**(3), 329–401 (1985).
- <sup>106</sup>S. R. Jackson, R. L. Kingsford, G. W. Collins, and C. G. Bischak, *Chem. Mater.* **35**(14), 5392–5400 (2023).
- <sup>107</sup>X. Yan, M. Xiong, X. Y. Deng, K. K. Liu, J. T. Li, X. Q. Wang, S. Zhang, N. Prine, Z. Zhang, W. Huang, Y. Wang, J. Y. Wang, X. Gu, S. K. So, J. Zhu, and T. Lei, *Nat. Commun.* **12**(1), 5723 (2021).
- <sup>108</sup>J. Hynynen, D. Kiefer, L. Yu, R. Kroon, R. Munir, A. Amassian, M. Kemerink, and C. Muller, *Macromolecules* **50**(20), 8140–8148 (2017).
- <sup>109</sup>K. Gu, C. R. Snyder, J. Onorato, C. K. Luscombe, A. W. Bosse, and Y. L. Loo, *ACS Macro Lett.* **7**(11), 1333–1338 (2018).
- <sup>110</sup>S. Fratini, M. Nikolka, A. Salleo, G. Schweicher, and H. Sirringhaus, *Nat. Mater.* **19**(5), 491–502 (2020).
- <sup>111</sup>T. J. Quill, G. LeCroy, A. Marks, S. A. Hesse, Q. Thiburce, I. McCulloch, C. J. Tassone, C. J. Takacs, A. Giovannitti, and A. Salleo, *Adv. Mater.* **36**(15), e2310157 (2024).
- <sup>112</sup>A. D. Easley, T. Ma, C. I. Eneh, J. Yun, R. M. Thakur, and J. L. Lutkenhaus, *J. Polym. Sci.* **60**(7), 1090–1107 (2022).
- <sup>113</sup>M. Moser, T. C. Hidalgo, J. Sargailis, J. Gladisch, S. Ghosh, R. Sheelamantula, Q. Thiburce, A. Giovannitti, A. Salleo, N. Gasparini, A. Wadsworth, I. Zozoulenko, M. Berggren, E. Stavrinidou, S. Inal, and I. McCulloch, *Adv. Mater.* **32**(37), e2002748 (2020).
- <sup>114</sup>J. Sargailis, A. Savva, V. Druet, B. D. Paulsen, R. Wu, A. Hamidi-Sakr, D. Ohayon, G. Nikiforidis, X. Chen, I. McCulloch, J. Rivnay, and S. Inal, *Adv. Funct. Mater.* **31**(21), 2010165 (2021).
- <sup>115</sup>J. Guo, S. E. Chen, R. Giridharagopal, C. G. Bischak, J. W. Onorato, K. Yan, Z. Shen, C. Z. Li, C. K. Luscombe, and D. S. Ginger, *Nat. Mater.* **23**(5), 656–663 (2024).
- <sup>116</sup>G. Tzamalidis, N. A. Zaidi, C. C. Homes, and A. P. Monkman, *Phys. Rev. B* **66**(8), 085202 (2002).
- <sup>117</sup>S. Stafstrom, J. L. Bredas, A. J. Epstein, H. S. Woo, D. B. Tanner, W. S. Huang, and A. G. MacDiarmid, *Phys. Rev. Lett.* **59**(13), 1464–1467 (1987).
- <sup>118</sup>J. L. Bredas and G. B. Street, *Acc. Chem. Res.* **18**(10), 309–315 (1985).
- <sup>119</sup>L. A. A. Pettersson, S. Ghosh, and O. Inganäs, *Org. Electron.* **3**(3–4), 143–148 (2002).
- <sup>120</sup>A. Bile, D. Ceneda, V. E. S. Maryam, D. Scirè, G. Buscarino, M. Mosca, D. P. Adorno, R. Macaluso, R. L. Voti, C. Sibilia, T. G. Folland, K. Aydin, M. Centini, and M. C. Larciprete, *Opt. Mater.* **154**, 115732 (2024).
- <sup>121</sup>C. Wan, Z. Zhang, D. Woolf, C. M. Hessel, J. Rensberg, J. M. Hensley, Y. Xiao, A. Shahsafi, J. Salman, S. Richter, Y. Sun, M. M. Qazilbash, R. Schmidt-Grund, C. Ronning, S. Ramanathan, and M. A. Kats, *Ann. Phys.* **531**(10), 1900188 (2019).
- <sup>122</sup>P. Hu, P. Hu, T. D. Vu, M. Li, S. Wang, Y. Ke, X. Zeng, L. Mai, and Y. Long, *Chem. Rev.* **123**(8), 4353–4415 (2023).
- <sup>123</sup>K. Aryana, H. J. Kim, M. R. Islam, N. Hong, C.-C. Popescu, S. Makarem, T. Gu, J. Hu, and P. E. Hopkins, *Opt. Mater. Express* **13**(11), 2997–3415 (2023).
- <sup>124</sup>R. L. Voti, K. Agharahimli, M. Misano, M. C. Larciprete, G. Leahu, F. A. Bovino, C. Sibilia, T. Cesca, G. Mattei, F. V. Lupo, and R. Macaluso, *Int. J. Therm. Sci.* **197**, 108832 (2024).
- <sup>125</sup>M. N. Julian, C. Williams, S. Borg, S. Bartram, and H. J. Kim, *Optica* **7**(7), 746 (2020).
- <sup>126</sup>S. Abdollahramezani, O. Hemmatyar, M. Taghinejad, H. Taghinejad, A. Krasnok, A. A. Eftekhari, C. Teichrib, S. Deshmukh, M. A. El-Sayed, E. Pop, M. Wuttig, A. Alù, W. Cai, and A. Adibi, *Nat. Commun.* **13**(1), 1696 (2022).
- <sup>127</sup>Y. Zhang, C. Fowler, J. Liang, B. Azhar, M. Y. Shalaginov, S. Deckoff-Jones, S. An, J. B. Chou, C. M. Roberts, V. Liberman, M. Kang, C. Ríos, K. A. Richardson, C. Rivero-Baleine, T. Gu, H. Zhang, and J. Hu, *Nat. Nanotechnol.* **16**(6), 661–666 (2021).
- <sup>128</sup>J. Long, S. Taylor, and L. Wang, *ACS Photonics* **7**(8), 2219–2227 (2020).
- <sup>129</sup>J. King, C. Wan, T. J. Park, S. Deshpande, Z. Zhang, S. Ramanathan, and M. A. Kats, *Nat. Photonics* **18**(1), 74–80 (2024).
- <sup>130</sup>Q. Li, T.-H. Chen, and P.-C. Hsu, *Next Energy* **3**, 100108 (2024).
- <sup>131</sup>M. C. Larciprete, D. Ceneda, D. Scirè, M. Mosca, D. Persano Adorno, S. A. Dereshgi, R. Macaluso, R. L. Voti, C. Sibilia, T. Cesca, G. Mattei, K. Aydin, and M. Centini, *APL Mater.* **11**(9), 091107 (2023).
- <sup>132</sup>M. C. Larciprete, M. Centini, S. Paoloni, I. Fratoddi, S. A. Dereshgi, K. Tang, J. Wu, and K. Aydin, *Sci. Rep.* **10**(1), 11544 (2020).
- <sup>133</sup>M. A. Kats, D. Sharma, J. Lin, P. Genevet, R. Blanchard, Z. Yang, M. M. Qazilbash, D. N. Basov, S. Ramanathan, and F. Capasso, *Appl. Phys. Lett.* **101**(22), 221101 (2012).
- <sup>134</sup>H. Liu, J. Lu, and X. R. Wang, *Nanotechnology* **29**(2), 024002 (2018).
- <sup>135</sup>Y. Qu, Q. Li, L. Cai, M. Pan, P. Ghosh, K. Du, and M. Qiu, *Light* **7**(1), 26 (2018).
- <sup>136</sup>K. Tang, K. Dong, J. Li, M. P. Gordon, F. G. Reichertz, H. Kim, Y. Rho, Q. Wang, C. Y. Lin, C. P. Grigoropoulos, A. Javey, J. J. Urban, J. Yao, R. Levinson, and J. Wu, *Science* **374**(6574), 1504–1509 (2021).
- <sup>137</sup>S. Wang, T. Jiang, Y. Meng, R. Yang, G. Tan, and Y. Long, *Science* **374**(6574), 1501–1504 (2021).
- <sup>138</sup>Z. Liu, J. Sun, J. Mao, H. Zhu, W. Ren, J. Zhou, Z. Wang, D. J. Singh, J. Sui, C.-W. Chu, and Z. Ren, *Proc. Natl. Acad. Sci. U. S. A.* **115**(21), 5332–5337 (2018).
- <sup>139</sup>Z. Shao, X. Cao, H. Luo, and P. Jin, *NPG Asia Mater.* **10**(7), 581–605 (2018).
- <sup>140</sup>C. C. Nadell, B. Huang, J. M. Malof, and W. J. Padilla, *Opt. Express* **27**(20), 27523–27535 (2019).
- <sup>141</sup>W. Ma, Z. Liu, Z. A. Kudyshev, A. Boltasseva, W. Cai, and Y. Liu, *Nat. Photonics* **15**(2), 77–90 (2021).
- <sup>142</sup>J. Jiang, M. Chen, and J. A. Fan, *Nat. Rev. Mater.* **6**(8), 679–700 (2020).
- <sup>143</sup>S. Doshi, D. Ludescher, J. Karst, M. Floess, J. Carlstrom, B. Li, N. Mintz Hemed, Y. S. Duh, N. A. Melosh, M. Hentschel, M. Brongersma, and H. Giessen, *Nanophotonics* **13**(12), 2271–2280 (2024).
- <sup>144</sup>W. Lu, L. de S. Menezes, A. Tittl, H. Ren, and S. A. Maier, *Nanophotonics* **13**(1), 39–49 (2024).

- <sup>145</sup>S. Chen, E. S. H. Kang, M. Shiran Chaharsoughi, V. Stanishev, P. Kuhne, H. Sun, C. Wang, M. Fahlman, S. Fabiano, V. Darakchieva, and M. P. Jonsson, *Nat. Nanotechnol.* **15**(1), 35–40 (2020).
- <sup>146</sup>R. Kaissner, J. Li, W. Lu, X. Li, F. Neubrech, J. Wang, and N. Liu, *Sci. Adv.* **7**(19), eabd9450 (2021).
- <sup>147</sup>J. Karst, M. Floess, M. Ubl, C. Dinger, C. Malacrida, T. Steinle, S. Ludwigs, M. Hentschel, and H. Giessen, *Science* **374**(6567), 612–616 (2021).
- <sup>148</sup>J. F. Chang, M. C. Gwinner, M. Caironi, T. Sakanoue, and H. Sirringhaus, *Adv. Funct. Mater.* **20**(17), 2825–2832 (2010).
- <sup>149</sup>Y. Q. Zheng, Y. Liu, D. Zhong, S. Nikzad, S. Liu, Z. Yu, D. Liu, H. C. Wu, C. Zhu, J. Li, H. Tran, J. B. Tok, and Z. Bao, *Science* **373**(6550), 88–94 (2021).
- <sup>150</sup>J. Y. Lee, B. K. Ju, and K. H. Cho, *Nanomaterials* **12**(9), 1611 (2022).
- <sup>151</sup>S. So, T. Badloe, J. Noh, J. Bravo-Abad, and J. Rho, *Nanophotonics* **9**(5), 1041–1057 (2020).