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Flexible and High-Performance Solution-Processable Single-Detector Organic Spectrometer

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Spectrometers are indispensable tools for civil and military-related optoelectronic applications. To meet the requirements of the revolutionary data/Al-driven era, next-generation spectrometers must not only be flexible with minimal sizes but exhibit high accuracy and resolution. In this study, a compact, high-performance, and flexible organic spectrometer is reported, fabricated using solution processing, which employs an optical cascade architecture by integrating organic electrochromic devices and photodetectors. This organic spectrometer can not only achieve a resolution of 0.56 nm, an accuracy of 0.14 nm, and a broad detection range from 400 to 1000 nm but also realize a vital absolute spectral irradiance measurement ranging from 10^{-8} to 10^{-4} W cm⁻² nm⁻¹. Additionally, its intrinsic flexibility and highly replaceable feasibility of bandgap-tunable organic materials enable their high applicability with excellent portability and adaptability in the upcoming data/Al-driven era or scenarios.

1. Introduction

Spectrometers are indispensable in the modern information society and play a crucial role in industrial processes, daily life, and scientific research, including environmental monitoring, biomedical diagnostics, and spectral imaging, by revealing light wavelength and intensity information.^[1–4] In particular, the upcoming data/AI-driven era has further amplified the urgent demand for portable/embeddable spectrometers in field applications, such as intelligent consumer electronics, wearable devices, remote industry control/automation, and defense

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applications, with lab(spectrometer)-onchip characteristics. Depending on the grating and interferometer, conventional benchtop spectrometers often face inherent limitations regarding bulky size owing to the long optical path, mechanically movable components,^[5,6] and external complex calibrated elements, such as the cosine corrector and integrating sphere,^[7,8] thereby restricting their widespread application in on-chip optoelectronics. The conventional approach based on typical benchtop spectrometers, which minimize the physical optical components or optical path lengths to the submillimeter scale, has inherent adverse effects associated with such scaling.^[9] Recently, a newly emerging approach using computational spectral reconstruction has been developed to overcome these scaling issues, with a high resolution in certain

cases.^[9,10] These pioneering computational spectrometers, which are based on the principle of wavelength multiplexing and matrix computational algorithms (with necessary statistical/approximation treatment for measuring errors), include diverse architectures such as filter/detector arrays or singlepixel spectral detectors using primarily inorganic quantum dot arrays,^[11] gradient single-nanowire alloys,^[12] and bandgap engineered 2D materials and van der Waals heterojunctions.^[13-15] The wavelength multiplexing principle lays the foundation for an unknown spectrum dynamic response to achieve a mathematically solvable solution.^[11,16] Ideally, the overall performance, footprint, flexibility, operational spectral range, and adaptability of the spectrometer to different application scenarios and operational stability under various environments, such as different temperatures and humidity, must be considered in practical applications in the information and data-driven age.^[9,10] Stateof-the-art computational spectrometers are typically fabricated using intrinsically rigid inorganic nanomaterials via an intricate photo/electron lithography process,^[12,14,17,18] posing significant challenges for the anticipated adoption in the increasingly widespread Internet of Things and 5G-related flexible electronics such as wearable and portable devices.^[19,20] Furthermore, the absolute spectral irradiance, which is a critical parameter for any measured spectrum, has not been adequately addressed in these state-of-the-art studies.[9,21]

Pioneering computational spectrometers rely on the dynamic wavelength multiplexing approach,^[9,10] and operate in three steps: 1) learning to encode a working calibrated matrix

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Figure 1. Schematic mechanisms of computational spectrometers. A) Incident spectrum-tuning includes two approaches: i) an equivalent set of miniaturized optical elements, such as a photonic chip,^[18] waveguide,^[23] multi-foci metalens,^[24] and pinhole;^[25] and ii) single but tunable optical element such as a metasurface structure with a liquid crystal.^[26] B) Detector response-tuning includes two approaches: i) compositionally gradient detectors with varying spectral response; and ii) a single detector approach achieved primarily by external modulation, such as gating voltages and bias. C) Schematic illustration of our organic spectrometer and its working mechanism: an integrated two-layered single-detector device with onsite light tuning on the top OECD layer and an in situ dynamic tunable response from the bottom OPD layer. The reconstructed spectrum is obtained by solving equations in the corresponding approach (A–C) in the right panels. *S* (λ , x) represents the modulated spectrum response dependent on wavelength (detector) and coordinates (optical elements) in Figure 1A, *R* (λ , *d*) represents the responsivity dependent on the wavelength and displacement (in space or bias) of devices in Figure 1B, and *r*(λ) and *T*(*V*) represent the responsivity of the OPD and transmittance of the OECD in Figure 1C, respectively.

using a series of known monochromatic lights, 2) sampling the unknown spectrum to generate a dynamic photoresponse matrix by tuning either the incident light or photodetector, and 3) reconstructing the unknown spectrum by combining and solving the two previous matrices using computational algorithms. The primary distinction between these computational spectrometers is the modulation approach, which involves tuning either the incident light or detector response.^[11,14,22] Thus, these spectrometers are categorized into two types (Figure 1A,B). Regardless of the modulation used (incident spectrum or detector response), the generated calibrated matrix should have no linear dependence between any two datasets in the matrix, and as much dissimilarity as possible is preferred.^[13,14]

For the incident spectrum-tuning category (Figure 1A), the calibrated matrix is obtained using a set of

miniaturized optical elements (working as filters or dispersers) (Figure 1A (i)), such as a photonic chip,^[18] waveguide,^[23] multifoci metalens,^[24] pinhole,^[25] filters array,^[11,22] and structurally colored nanowires,^[27] which are generally related to dedicated micro/nano-scale fabrication. Another approach is to use a single tunable optical element, such as a metasurface structure combined with a tuned liquid crystal, to achieve the required light tuning (Figure 1A (ii)).^[26] In most of these studies, various optical array structures are employed for spectral tuning, which complicates fabrication and limits the footprint.^[9]

Regarding spectrometers based on the modulation of the detector response (Figure 1B), this can first be achieved using an array of photodetectors (Figure 1B (i)), such as gradient single-nanowire alloys,^[12] and perovskite arrays.^[28,29] The dynamic and wavelength multiplexing spectral responses of this type of spectrometer are spatially distributed according to the components of the detector array, which require dedicated fabrication. Therefore, a more efficient method is to generate a dynamic and wavelength-multiplexing spectral response using only a single detector (Figure 1B (ii)). Recently, a novel strategy involving the convenient tuning of a single detector's response has become mainstream in this category (Figure 1B (ii)).^[14,30] This includes numerous tunable van der Waals junction-based diodes such as WSe₂/MoS₂,^[14] ReSe₂/SnS₂,^[31] and MoS₂/Black phosphorus.^[15] Despite these achievements, the properties of these 2D materials are often influenced by factors such as morphology, crystalline quality, surface defects, and doping level.^[32,33] Coupled with the required microscale fabrication, batch-to-batch or device-to-device variations pose significant challenges for these devices (Text S1 and Table S1, Supporting Information).^[34]

Ideally, a single-pixel spectrometer that does not require physical tuning (filtering or dispersing) of the light would be optimal. This could be achieved by engineering a wavelengthmultiplexed, dynamic light response matrix through the tuning of a single detector, thereby eliminating the need for arrays, filters, interferometers, or moving parts.^[9,10] Additionally, convenient fabrication without a dedicated nano/microscale process, the mechanical flexibility of the device, and the capability to meet highly diverse scenarios are of paramount importance.^[19]

This motivates us to investigate whether an integrated single-device architecture can simultaneously combine onsite and in situ wavelength-independent modulation at the same quality level as that of a normal-light interferometer/grating and high-performance detector, together with high sensitivity in the full spectrum range. It is preferable for a single-pixel spectrometer to also have a wide linear response range to light intensity. Achieving this would ensure the realization of a compact spectrometer with the same performance as the conventional spectrometer while offering portability and adaptability in the coming data/AI-driven era for various scenarios.^[20]

Over the past few decades, organic optoelectronic materials have driven significant technological innovations in fields such as organic photodetectors (OPDs),^[35] organic electrochromic devices (OECDs),^[36–38] and organic photovoltaics.^[39–41] In particular, OPDs offer unique advantages in light perception and detec-

tion, rendering them promising candidates as next-generation spectrometers owing to their intrinsic flexibility and broad spectral range (300–1100 nm).^[42–44] wide-range linear response (>100 dB) in terms of light intensity.^[45] solution-processed capability, and comparable or better optoelectronic performance.^[46] Initially, we fabricated an OPD-based single-detector spectrometer to obtain the required spectral response matrix by electrically tuning the OPD responsivity (Text S2, Supporting Information). This single-detector approach was unsuccessful because it was extremely difficult to realize the workable bias-modulated wavelength-dependent responsivity of OPDs. Therefore, alternative strategies need to be explored. The OECD which utilizes the electrochemical doping of organics/polymers, easily offers onsite bias-tunable spectral transmittance with a broad wavelength range of 300–2000 nm.^[47,48] Therefore, if these two types of organic-based devices (OECDs and OPDs) are used as new hybrid single-pixel devices (Figure 1C), a new type of intrinsic flexible organic spectrometer that can minimize the footprint and with the advantages of organic-based electronics could meet the increasing and challenging demands of the data-driven and Internet era.

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Herein, we designed and fabricated a highly compact and flexible organic spectrometer (FlexOS) with high performance that could measure absolute spectral irradiance based on an optical cascade architecture of the OPD (bottom layer) and OECD (top layer). In particular, we used a convenient and straightforward solution process, with tunable footprints ranging from microto millimeter scales (Figure 2A). The architecture and operation principle of our spectrometer are significantly different from previous ones, achieving a single-pixel device comprising two easily integrated cascade layers without any complicated array or movable structure (Figure 1C). The top layer of the OECD subunit functioned as a spectrum-tuning unit onsite, while the bottom OPD subunit simultaneously measured the tuned spectrum from the OECD in situ. In simple terms, no spatial optical elements or detector array structures were required, and all subunit devices collapsed into a single pixel (Figure S1, Supporting Information). By measuring the photocurrent of an organic spectrometer (org-spectrometer) generated by an unknown spectrum using the wavelength multiplexing principle and then computationally cross-referencing with a previously self-calibrated built responsivity matrix, any incoming unknown light spectrum, including monochromatic, narrowband, and broadband light, could be reconstructed with an accurate measurement of the absolute irradiance intensity (Figure 2B). Particularly, with PCE-10 and COTIC-4F as the OPD materials (Experimental Section, Figure S2, Supporting Information) for a broad visible-to-nearinfrared (Vis-NIR) light measurement, our org-spectrometer achieves a resolution and accuracy of 0.56 and 0.14 nm, respectively, for resolving peak monochromatic wavelengths, newly defined quantitative spectral mismatch degree (M value, see below) as low as 0.030, and absolute spectral irradiance from 10^{-8} to 10^{-4} W cm⁻² nm⁻¹ in the spectrum range of 400–1000 nm. Thus, owing to the high stability of corresponding flexible devices and tunable sizes of the FlexOS, we envision that our study will pave the way for the commercialization of highly compact and flexible computational spectrometers and boost their huge application potential in various fields in the information and 5G age.

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Figure 2. Org-spectrometers with an optical cascade architecture. A) Schematic diagram of our org-spectrometer with an optical cascade architecture: the top OECD layers modulate light onsite, while the bottom OPD layer measures the light in situ. B) Reconstruction of unknown light, including monochromatic, narrowband, and broadband lights, with absolute radiance measurements. C) Schematic diagram of the operating principle of the org-spectrometer based on OECDs and OPD: i) mechanism diagram of the spectrum tuning of OECDs and the light response of OPDs; ii) schematic of the energy-level diagrams of neutral, polaron states in electrochromic polymers; and iii) energy levels of the bulk heterojunction of PCE-10 and COTIC-4F. D) Schematic of the spectral responsivity of the org-spectrometer with various voltages applied on the OECDs.

2. Results and Discussion

2.1. Materials, Fabrication, and Architecture Design of the Org-Spectrometer

Our org-spectrometer comprised an optical cascade architecture with one bottom OPD for light detection and two top-stacked OECDs with independent bias modulation for on-site light modulation (Figure 2C). The detailed fabrication process is described in Experimental Section (Supporting Information). The rationale for this optimized architecture and build-up is thoroughly analyzed in Text S3 (Supporting Information) using a systematic mathematical analysis.

2.2. OECD Fabrication and Performance

The two OECDs for light modulation were constructed using indium tin oxid (ITO) /electrochromic polymer (ECP) /ionic liquid/PEDOT:PSS/ITO,^[49] where the electrochromic polymer served as the primary medium for modulating the spectral transmittance. Moreover, the ionic liquid and PEDOT:PSS layers served as the ion transport and ion storage layers, respectively (Figure 2C (i)). Two common electrochromic materials (ECP-purple and ECP-blue) were simultaneously used in the two independent OECDs for more dynamic and accurate wavelength multiplexing of light modulation (Text S3, Supporting Information). In the optimized devices, ECP-purple and ECP-blue, whose chemical structures and related properties are shown in Figures

S2,S3 (Supporting Information), were selected owing to their excellent tunable spectra, fast switching speed, and high stability over a broad spectral range of 300–2000 nm (Figure 2C (ii), Figure S4, Supporting Information).^[47,48]

2.3. OPD Fabrication and Performance

For the OPD, we adopted state-of-the-art bulk heterojunction (BHJ) photodiodes featuring a vertical structure of ITO/ZnO/BHJ/MoOx/Ag.^[50] In the optimized OPD, the donor PCE-10 and narrow-bandgap non-fullerene acceptor COTIC-4F (Experimental Section, Supporting Information, Figure 2C (iii)) were selected owing to their excellent performance in terms of low dark current ($I_{\text{dark}} = 5.6 \text{ pA}$, $I_{\text{rms}} = 4.1 \text{ pA}$ at V = 0 V), wide spectral response range (300-1100 nm), high responsivity $(R = 0.45 \text{ A W}^{-1}, 980 \text{ nm})$, and fast response (433 ns) (Figures \$5,\$6, Supporting Information). State-of-the-art OPDs generally exhibit a wide range of linear responses.^[43] For example, the OPD based on PCE-10:COTIC-4F exhibits nearly eight orders of magnitude of light intensity in the range of 152 dB (Figure S6, Supporting Information), which is essential for accurately measuring the absolute irradiance. Furthermore, its operational advantage with excellent performance under zero-voltage conditions significantly reduces the overall power consumption of the spectrometer (Text S4, Supporting Information).^[46]

Thus, by integrating the aforementioned high-performance OECDs (for light modulation) and OPDs (for light detection),



we developed a single-detector spectrometer with high resolution, accuracy, and measurable absolute irradiance over a broad spectral range (Experimental Section, Supporting Information). Combined with the superiority of the solution process and the intrinsic flexibility of organic electronics, flexible single-detector spectrometers with a tunable device footprint (from $2 \times 2 \mu m^2$ to $4 \times 4 \text{ mm}^2$) (Figure S7, Supporting Information) and similar performance as that on the rigid substrate were also realized. To accommodate various application requirements, such as a specific spectrum detection range, detection limit, and even operational temperature and humidity, the high feasibility of replaceable OPDs (Table S4, Supporting Information) and OECDs (Table S5, Supporting Information) render our spectrometer with excellent adaptability. For instance, to measure a particular spectrum with a narrow band, such as the visible and ultraviolet ranges, materials with high sensitivities to a particular spectral range can be selected to meet such requirements (Text \$5 and Table \$4, Supporting Information).^[43]

2.4. Working Mechanism of the Org-Spectrometer

In our org-spectrometer, the incident light is modulated using two sequentially independent electrochromic devices: ECPpurple- and ECP-blue-based OECDs. Under the control of the applied bias on the OECDs, the quantities of the neutral and polaronic states undergo corresponding reversible changes, with transmittance variations across a wide range of 300-1000 nm (Figure S4, Supporting Information).^[51] The visible spectral region is assigned to the neutral state absorption (π – π * transition) that covers 400-600/700 nm,^[48] while the polaronic state (P1-P2) contribution dominates above the isosbestic point (ECP-purple, 620 nm; ECP-blue, 703 nm) (Figure 2C (ii); Figure S4, Supporting Information). As needed, the OECD materials could be easily replaced for a specified spectral range (Table S5, Supporting Information). Thus, dynamic wavelength multiplexing modulation at the OPD layer is achieved onsite by tuning the neutral and polaronic states of the OECDs. These two states change in opposite directions,^[48] resulting in a dynamic wavelength multiplexing response from the OPD unit for incident spectrum measurements (Figure S8, Supporting Information). Moreover, the simultaneously dynamic wavelength multiplexing tuning of the incident spectrum from the OECD unit and the dynamic photoresponse from the OPD unit for this tuned spectrum generate the responsivity matrix R (λ , Vp, Vb) (Figure 2D). Notably, Vp and Vb represent bias on ECP-purple and ECP-blue-based OECDs, respectively. These variables serve as the foundation for constructing a mathematically solvable matrix with a system of linear equations for our single-detector reconstruction spectrometer (Text S6, Supporting Information). The cascade order of these two OECDs does not affect the responsivity of the org-spectrometer (Figure **S9**, Supporting Information).

Following the workflow diagram (Figure S10, Supporting Information), our spectrometer operates in three steps: 1) learning step (acquisition of responsivity matrix), 2) sampling step (measurement of photocurrent of the unknown spectrum), and 3) reconstructing step. In the first step, a learning responsivity matrix R (λ , Vp, Vb) was obtained as a function of both the light wavelength of a series of monochromatic light and the applied

voltages of the two OECDs. Completing the learning process for all $l \times m \times n$ steps (*l*, *m*, and *n* represent the displacements of λ , Vp, and Vb, respectively, corresponding to the wavelength and voltages of the two OECDs) generated the full 3D matrix (Figure S11, Supporting Information). For example, to demonstrate a full 400–1000 nm range incident light measurement, a 600 \times 11×16 (105600 sets of data) learning matrix is generated (Experimental Section, for the original dataset, see Figure S11, Supporting Information). Owing to the high responsivity of the OPD across the entire spectrum, our spectrometer maintained a peak responsivity of >0.15 A W⁻¹ even after modulation by the two OECDs (Figure S11, Supporting Information). Furthermore, the efficiency of the spectrometer is an important parameter for assessing the light loss caused by OECDs.^[52] The maximum spectrometer efficiency of our device is measured as >30% within the wavelength range of 400–1000 nm (Figure S11, Supporting Information).

In the second sampling step, the photocurrent of the orgspectrometer for unknown incident light is measured using the principle of wavelength multiplexing at 11×16 different displacement voltages applied to the OECD unit. This generates a 2D photocurrent matrix I (Vp, Vb), which could be automatically performed with I/V meter scanning (Experimental Section, Supporting Information). Finally, based on the responsivity matrix R (λ, Vp, Vb) generated from the learning step and the photocurrent matrix I (Vp, Vb) measured in the sampling step, the spectrum of the incident light is reconstructed. The solvability and performance of the spectrometer are dependent on the dissimilarity between any sets of spectral transmittances of the OECDs with different wavelengths of multiplexing tuned light and the diversity of the OPD response (Texts S2,S3, Supporting Information). Thus, the combination of finely tuned light from the OECD unit and wide linear response across the wavelength range of the OPD in our single-detector spectrometer guarantees a highly dynamic response matrix with maximum dissimilarities for our device.

For the reconstructing step, through normal theoretical derivation (Text S6, Supporting Information), the integral equation is as follows: $\int_{\lambda_i}^{\lambda_j} r(\lambda) \cdot T(\lambda, Vp, Vb) \cdot P(\lambda) = \int_{\lambda_i}^{\lambda_j} R(\lambda, Vp, Vb) \cdot$ $P(\lambda) = I(Vp, Vb)$, where $r(\lambda)$ and $T(\lambda, Vp, Vb)$ are the responsivity of the OPD and combined spectral transmittance of the two OECDs, respectively. Both the *r* (λ) and *T* (λ , *Vp*, *Vb*) generate the spectral matrix of the integrated spectrometer responsivity matrix: *R* (λ , *Vp*, *Vb*), which could be obtained in the first step. *P* (λ) represents the unknown spectral intensity within the range of the λ_i to λ_i ($\lambda_i < \lambda_i$). Integrating these two-layered OPD and OECD devices yields the dynamic wavelength-multiplexing photocurrent matrix *I* (*Vp*, *Vb*) of the modulated spectrum via the two OECDs. Subsequently, the unknown spectrum is reconstructed by computing the constrained least-squares solution using the wellknown Tikhonov regularization,^[53] which efficiently addresses measurement errors.^[54,55] Here, the regularization parameters are obtained using the generalized cross-validation method.^[55] By computationally cross-referencing with a self-calibrated and previously built matrix, the reconstructed spectrum with absolute irradiance could be obtained in any wavelength range or the entire spectrum owing to the large linear dynamic range (LDR) of the OPD, as mentioned above (Figure S10, Supporting Information).

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Figure 3. Characterization of the org-spectrometer. A) 3D-matrix contour plot of the spectral responsivity of the org-spectrometer dependent on V_p and V_b applied on the OECDs. The matrix included 176 different spectral responses, with 11 (V_p) × 16 (V_b) voltage values and a wavelength step of 1 nm for a series of known monochromatic light. B) Monochromatic light (FWHM of \approx 2 nm) reconstructed using the org-spectrometer (solid line) compared to commercial spectrometers (Ocean USB4000, dashed line). C) Performance metrics of the org-spectrometer in terms of $\Delta\lambda$, resolving power, and PSNR. D) Resolution of the org-spectrometer determined using the wavelength difference between the closest two peaks; i) two monochromatic lights of 740.9 and 741.3 nm; ii) complex spectrum measured using a commercial spectrometer; and iii) complex spectrum reconstructed using the org-spectrometer. E), F), and G) Quasi-monochromatic light (FWHM of \approx 4 nm), narrowband light (FWHM of \approx 30 nm), and broadband spectra reconstructed using the org-spectrometer, respectively. H) LDR of the OPD and its corresponding responsivities as a function of optical power ($\lambda =$ 980 nm). I) Spectra with absolute irradiance reconstructed using the org-spectrometer and a commercial spectrometer.

2.5. Performance of the Org-Spectrometer

The org-spectrometer, modulated by two OECDs under variables *Vp* and *Vb*, demonstrated tunable responsivity across the spectral range of 400–1000 nm when PCE-10 and COTIC-4F are used as the photoactive materials in the OPD (**Figure 3A**). We first evaluated the performance of the device by reconstructing monochromatic light (full width at half maximum, FWHM of ≈ 2 nm). For the monochromatic light within the 730–750 nm range (Figure 3B), the org-spectrometer exhibits an average accuracy $\Delta \lambda$ of 0.14 nm ($\Delta \lambda$: defined as the peak difference between the reconstructed and the reference spectrum), wavelength

resolving power of up to 6900, and peak signal-to-noise ratio (PSNR) of >30 dB (Figure 3C), demonstrating a high consistency with the reference spectrum obtained using a commercial spectrometer. Additionally, the spectrometer achieves a spectral resolution of 0.56 nm, defined as the minimum distinguishable separation between the adjacent spectral peaks, which surpasses existing state-of-the-art compact spectrometers (Table S7, Texts S7,S8, Supporting Information). Taking advantage of its full spectral response from 400–1000 nm, the org-spectrometer could reconstruct various quasi-monochromatic (FWHM of \approx 4 nm) and narrowband spectra (FWHM of \approx 30 nm) in the entire spectrum range. As shown in Figure 3E,F, the reconstructed

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Figure 4. Characterizations of the flexible org-spectrometer. A) Schematic of the FlexOS for possible future application in wearable devices. B) Spectral responsivity matrix of the FlexOS dependent on V_p and V_b applied in flexible OECDs fabricated on ITO/PET substrates. C) Spectra of quasi-monochromatic light (FWHM of \approx 4 nm), narrowband light (FWHM of \approx 30 nm), and broadband spectra (solid line) reconstructed using the FlexOS compared to commercial spectrometers (dashed line). D) Comparison of the figure-of-metrics of the state-of-the-art reconstructed spectrometers (Refs. [12,14,17,26]) and our spectrometers in the Vis–NIR region, including bandwidth, accuracy, M value, footprint, and flexible availability.

spectra obtained using our spectrometer are all highly consistent with the reference spectrum in terms of both shape and central peak positions (Figure 3E,F). To comprehensively and quantitatively evaluate the reliability and performance of our spectrometer beyond individual metrics, we introduced a spectral mismatch degree (M value) as an overall figure-of-merit to quantify the disparity between the reconstructed and reference spectra for the first time (Text \$9, Supporting Information). A smaller M value indicates better spectral reconstruction reliability and accuracy of the spectrometer. With this comprehensive performance evaluation of the M parameter, we found that our spectrometer generally outperformed state-of-the-art results reported in the literature,^[12,14,15] and exhibited comparable performance to bulky commercial spectrometers, as shown in Table S9 (Supporting Information). For example, the reconstructed quasimonochromatic and narrowband spectra yield average M values of 0.023 and 0.029, respectively, from our org-spectrometer, which are better than those in the literature and comparable to a widely used commercial spectrometer (Model: Ocean USB 4000, Text S9, Table S9, Supporting Information). Moreover, our orgspectrometer could measure both natural and complex spectra with the same reliability. For example, the reconstructed spectra for the indoor LED light and broadband infrared spectrum (400-1000 nm) show M values as low as 0.024 and 0.039 (Figure 3G), respectively, which are better than those of previously reported reconstructed spectrometers (Table \$9, Supporting Information).

Our strategy and device architecture is a straightforward and easy approach to fabricate devices with customized performance parameters, such as a specified spectrum detection range, resolution, and accuracy, as well as environmentally demanding parameters, by simply replacing the active organic materials in the OPD and OECD layers (Text S5, Tables S4 and S5, Supporting Information). As a demonstration, we fabricated spectrometers using different active materials (PM6:F-2F and PM6:CH17) in the OPD layer to measure the spectra at wavelengths 400–750 and 400–850 nm, respectively. The spectrometer using PM6:F-2F exhibited a higher accuracy ($\Delta \lambda = 1.4$ nm) compared to that using PM6:CH17 ($\Delta \lambda = 2.2$ nm), highlighting the versatility of this strategy for application-driven optimization (Figures S12–S14, Supporting Information).

2.6. Measurement of Absolute Spectral Irradiance

Absolute spectral irradiance, which measures the power of electromagnetic radiation received per unit area within a unit wavelength interval, is an important parameter for precise spectral analysis.^[56] The org-spectrometer has a large LDR of 152 dB when PCE-10:COTIC-4F is used as the OPD unit, achieving a robust response across a broad spectral range (Figure 3H). This capability enables the org-spectrometer to accurately measure the absolute spectral irradiance across the 400–1000 nm



range, as demonstrated in the reconstructed narrowband spectra (peak wavelength = 725 nm) with intensities ranging from 0.96 to 42.6 μ W cm⁻² nm⁻¹ (Figure 3I). Notably, the peak spectral intensity obtained has a minimum variation of 5.1% (Figure S15, Supporting Information) compared to that of a commercial spectrometer (Model: Ocean USB 4000). Similarly, the absolute light intensities of quasi-monochromatic and narrowband lights (10⁻⁸ to 10⁻⁴ W cm⁻² nm⁻¹) with different wavelengths are successfully reconstructed with a peak intensity variation of 1.2% compared to the reference spectrum obtained using a commercial spectrometer (Figure S16, Supporting Information). Notably, for absolute spectral irradiance measurements, our single-detector org-spectrometer exhibits significant superiority over commercial spectrometers because it achieves the measurement using a self- and in situ-calibrated responsivity matrix without any movable or spatially related mechanical/optical parts, which typically require calibration over time.^[6,7]

2.7. Flexible Org-Spectrometer

FlexOS represents a significant advancement in spectroscopic technology because it provides bendable configurations that offer adaptability, portability, and integration into wearable electronics (Figure 4A). Leveraging the solution-processable characteristics of the org-spectrometer provides inherent advantages for the fabrication of flexible spectrometers. Using the same active materials, such as PCE-10:COTIC-4F, we fabricated a flexible spectrometer on an ITO/ polyethylene terephthalate (PET) substrate, which combined the intrinsic flexibility and high performance of org-spectrometers (Experimental Section, Supporting Information). Similar to their rigid counterparts, the corresponding FlexOS demonstrated almost identical capabilities for accurate measurements across the spectral range of 400-1000 nm (Figure 4B). Following the same workflow for unknown spectra reconstruction (Figure S10, Supporting Information), the FlexOS exhibited a performance comparable to that of the rigid device regarding spectral accuracy (0.57 nm) and resolution (2.58 nm) (Figure S17, Supporting Information). In addition, the flexible spectrometer accurately reconstructs quasi-monochromatic, narrowband, and broadband spectra (Figure 4C) with a minimum M value of 0.031, which is close to that of the rigid spectrometer (0.024). To evaluate its suitability for wearable applications, we assessed its spectral-reconstruction capabilities under bending conditions (Experimental Section, Supporting Information). Compared to the reference spectrum, the reconstructed spectrum at the bending state with a 2.7 m⁻¹ curvature exhibited a minimum changed M value of 0.035 (vs 0.031 for the rigid one) (Figure S18, Supporting Information) due to negligible change in responsivity (Figure S19, Supporting Information), highlighting its capability to maintain spectral measurement fidelity under severe deformation. Moreover, after 300 bending and release cycles, the FlexOS exhibited a negligible change in the M value (0.037) (Figure S18-S20, Supporting Information). Figure 4D summarizes and compares the bandwidth, accuracy, M value, footprint, and flexibility of our org-spectrometer with those of other representative compact spectrometers, demonstrating that our singledetector FlexOS exhibits comprehensive superiority compared to previously reported computational spectrometers. The intrinsic biocompatibility of organic electronics for our organic spectrometer further expands their application potential to various bioelectronic applications.

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2.8. Stability of the Org-Spectrometer

Device stability plays a vital role in determining its practical applicability, encompassing the stability of both the OPDs and OECDs, as well as the integrated spectrometer (Text S10, Supporting Information). Within the org-spectrometer framework, the OPDs maintained >99% of their initial photoresponse after 2 00 000 on-off switching cycles (Figure S21, Supporting Information). Furthermore, they exhibited only a 5% variation in the initial dark current under zero bias after storage under ambient conditions for 1000 h (Figure S21, Supporting Information). The OECDs exhibited excellent optical stability, with 50.5% attenuation of the transmittance after 1000 on-off cycles (Figure S22, Supporting Information). Notably, the org-spectrometer precisely and reliably reconstructed the spectrum with an M value of 0.063 even after storage under ambient conditions for 20 weeks (Figure S23, Supporting Information). To assess its practical applications in various environments, we evaluated the performance of our device under harsh conditions. With operating temperatures ranging from 273 to 323 K (relative humidity, RH = 30%), all the reconstructed guasi-monochromatic spectra at 760 nm yielded M values 50.030 (Figure S23, Supporting Information). Furthermore, the org-spectrometer showed M values of 0.026, 0.033, and 0.042 in different atmospheres with RHs of 15%, 50%, and 70% (Temperature, T = 298 K), respectively, demonstrating the high consistency of the reconstructed results with the reference spectra (Figure S24, Supporting Information). The excellent operational stability of our devices under various vigorous operating environments demonstrates that they can cope with most civil application conditions (Text S10, Supporting Information). Additionally, our spectrometers show high batch-to-batch reproducibility owing to the convenient and straightforward solution processing technique (Figure S25, Supporting Information). In principle, owing to the easy fabrication and simple device structure, the performance of our orgspectrometer should rely only on the OPD and OECD active materials; thus, all devices with the same materials should perform the same regardless of the fabrication process, in contrast to numerous approaches, such as those using mechanical exfoliation and micro/nanoscale fabrication (Texts S1 and S5, Supporting Information).^[9]

2.9. Spectral Imaging

Spectral imaging is a powerful technology that combines spectroscopic and imaging capabilities.^[56,57] To demonstrate this capability, spatial scanning is performed using our spectrometer (**Figure 5**A). A portrait of Confucius was projected onto the org-spectrometer, which was then scanned at a step size of 4 mm. The photocurrents were measured at each mapping step by varying the V_{bias} of the OECDs, which generated a 3D photocurrent data cube (*X*, *Y*, *I*). Subsequently, this photocurrent data





Figure 5. Spectral imaging of the org-spectrometer. A) Schematic of the spectral imaging process using the org-spectrometer with a spatial scanning approach. The measured 2D spatial coordinate photocurrent matrix is used for reconstructing spectral cubes. B) Spectral images of the reconstructed portrait of Confucius at different wavelengths: i) 510, ii) 525, iii) 550, and iv) 650 nm. C) Spectral image of Confucius reconstructed using the org-spectrometer compared to those of commercial spectrometers at points A and B marked in the Figure 5C inset.

cube was computationally converted into a spectral cube (X, Y, λ) using the reconstruction algorithm (Figure 5A). Cross-sectional views of this cube in the X-Y plane yielded a series of singlewavelength spatial maps (Figure 5B). By applying the standardized color-matching functions of the International Commission on Illumination (CIE) to the spectral data cube, we produced a pseudo-colored image that closely resembled the original photograph (Figure 5C, Experimental Section, Supporting Information). The reconstructed spectra from different points on the portrait in Figure 5C were highly consistent with the reference spectra obtained using a commercial spectrometer (Figure 5D), further demonstrating the potential of the spectrometer for spectral imaging. However, there was an inherent trade-off between image resolution and the mapping step (both wavelength and spatial scanning steps). If the spatial scanning step is smaller than 4 mm in the spectral image reconstruction, the resolution could be further improved. Moreover, integrating a 2D OPD array such as a charge-coupled device and matching OECDs would enable an organic and flexible hyperspectral camera for spectral snapshot imaging without mechanical scanning or filter switching, meeting the growing demand for hyperspectral imaging in smartphones and cameras.^[57,58]

3. Conclusion

We designed and fabricated a single-pixel org-spectrometer by integrating OECDs and OPDs with an optical cascade architecture using a convenient solution process. In addition to the high performance comparable to that of commercial bulky spectrometers, including a resolution of 0.56 nm, an accuracy of 0.14 nm, a broad detection ranging from 400 to 1000 nm, and significant stability in various conditions, our org-spectrometer successfully realized the vital absolute spectral irradiance measurement under various light intensities from 10⁻⁸ to 10⁻⁴ W cm⁻² nm⁻¹, addressing the critical challenges faced by state-of-the-art computational spectrometers. The high performance of our spectrometer was also demonstrated in spectral imaging for the accurate reconstruction of a real image. Notably, the performance parameters could be tuned by simply replacing the active materials in the device. Moreover, the corresponding FlexOS exhibited almost the same performance as that of its rigid counterpart. In addition to its high mechanical stability, the FlexOS offers unprecedented advantages for wearable and implantable devices. Overall, the strategy using our optical cascade architecture with an integrated and tunable OECD and OPD, together with its high and

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robust performance, offers vast adaptive design opportunities for flexible and miniaturized spectrometers, rendering them with potential for various civil and military-related optoelectronic applications in the revolutionary data/AI-driven era.

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.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

flexible spectrometer, organic electrochromic device, organic photodetector

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