ISTITUTO NAZIONALE DI RICERCA METROLOGICA
Repository Istituzionale

Perspectives and recent advances in super-resolution spectroscopy: Stochastic and disordered-based approaches

Availability:
This version is available at: 11696/74519 since: 2022-08-08T10:11:32Z

(Article begins on next page)

27 July 2024

Perspectives and recent advances in super-resolution spectroscopy: Stochastic and disordered-based approaches (F)

View Online   Export Citation   CrossMark

A. Boschetti,[1,2] (iD) L. Pattelli,[2,3] (iD) R. Torre,[1,2] (iD) and D. S. Wiersma[1,2,3,a)] (iD)

AFFILIATIONS

[1]Department of Physics and Astronomy, University of Florence, 50019 Sesto Fiorentino, Italy
[2]European Laboratory for Non-Linear Spectroscopy (LENS), University of Florence, 50019 Sesto Fiorentino, Italy
[3]Istituto Nazionale di Ricerca Metrologica INRiM, 10135 Torino, Italy

[a)]Author to whom correspondence should be addressed: wiersma@lens.unifi.it

ABSTRACT

Spectroscopic applications are characterized by the constant effort to combine high spectral resolution with large bandwidth. A trade-off typically exists between these two aspects, but the recent development of super-resolved spectroscopy techniques is bringing new opportunities into this field. This is particularly relevant for all applications where compact and cost-effective instruments are needed such as in sensing, quality control, environmental monitoring, or biometric authentication, to name a few. These unconventional approaches exploit several strategies for spectral investigation, taking advantage of concepts such as sparse sampling, artificial intelligence, or post-processing reconstruction algorithms. In this Perspective, we discuss the main strengths and weaknesses of these methods, tracing promising future directions for their further development and widespread adoption.

Published under an exclusive license by AIP Publishing. https://doi.org/10.1063/5.0096519

I. INTRODUCTION

Several compact spectrometers are currently available on the market that are based on traditional designs. Their reduced footprint intrinsically limits their spectral resolution with typical spectral responses ranging from few nanometers to ∼10 nanometers for the smallest models. Significant progress has been made in the past years in the field of integrated spectrometers with a few notable examples leveraged on reconstructive spectroscopy[1–4] through regularization functions or compressed sensing.[5–8] In most cases, however, compressed sensing has been used to reduce the amount of data required for a full spectral reconstruction without sacrificing the resolution, rather than to enhance it or reveal finer spectral features beyond the intrinsic resolution of the apparatus. Several excellent reviews and Perspectives are already available in the literature for the interested reader, discussing original approaches to spectroscopy, spectral reconstruction strategies, and high resolution spectrometers.[9–12]

In this Perspective, our main focus is on "super-resolution" spectroscopy techniques that allow us to reach a spectral resolution that is beyond that expected for a given system based, e.g., on its footprint, dispersion, decorrelation, or detection properties. The observed resolution enhancement is typically achieved by either exploiting the optical properties of a light source in some unconventional ways or through post-processing reconstruction algorithms or both. We will look with particular interest at strategies based on statistics and disorder.

II. RECONSTRUCTIVE SPECTROSCOPY
A. Speckled pattern-based spectroscopy

The interference of multiply scattered laser light by a disordered medium results in a speckled pattern, which is highly sensitive to the wavelength of the incident radiation in most situations. In this respect, speckle-based spectroscopy applications are distinct from all other approaches in that they translate the spectral reconstruction problem into a different domain, namely, that of pattern recognition. In recent years, this strategy inspired several applications targeted at the realization of low-cost, alignment-free, on-chip wavemeters and spectrometers.[13–15] Random media used for speckle spectroscopy include single[16] or multimode fibers,[17] spiral waveguides,[18] integrating spheres,[19] and photonic amorphous structures up to pieces of mother-of-pearl.[20]

In this paper, we introduce briefly the working principle of speckle-based spectroscopy and then discuss the latest developments

aimed at further enhancing the resolution beyond that associated with the typical frequency decorrelation of speckle patterns.

Without loss of generality, let us consider as an example a speckle spectrometer based on a multimode optical fiber (MMF). Just as in common disordered systems, light propagating through a MMF is scrambled into multiple modes that interfere with each other along the fiber length, resulting in a fully developed speckle pattern at the end, whose spatial modulations are eventually recorded by a multi-pixel detector. For each input wavelength, a unique and distinct speckle pattern is obtained, providing a deterministic mapping between input frequencies and a spatial intensity pattern "fingerprint" at the fiber output [see Fig. 1(a)].
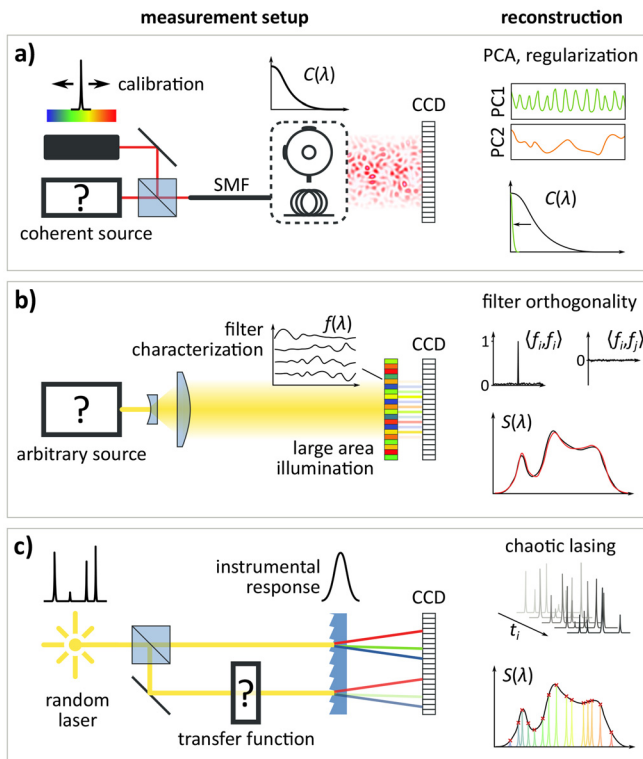

FIG. 1. Comparison of different spectral super-resolved reconstruction techniques. (a) Speckle-spectroscopy relies on a spatial-to-spectral mapping for the characterization of coherent light sources. It uses a tunable laser for calibration, a mode-scrambling device (such as a scattering medium or a multi-mode fiber), and a matrix detector. Input light is fed into the scattering system via a polarization-maintaining single mode fiber (SMF) for stability. Principal component analysis (PCA) is then used to extract spectral information below the speckle decorrelation limit $C(\lambda)$. (b) Filter-array reconstruction spectroscopy is used to characterize arbitrary light sources based on the transmittance through a set of random transmittance filters, which have been previously resolved with high resolution. Provided that the filter responses are highly uncorrelated in the spectral domain, illuminating them allows us to achieve a resolution, which is larger than that obtained with a comparable array of bandpass filters. (c) The STORS technique is used to characterize a transfer function exploiting random laser illumination, using a low-resolution dispersive element and a matrix detector. Collecting the response to enough narrow and sparse excitation peaks allows reconstructing the unknown spectrum overcoming the resolution limit of the dispersive element. A reference beam is used to compensate for intensity fluctuations.

For this reason, using a speckle-based spectrometer requires an *a priori* calibration step where a tunable laser source is used to feed known wavelengths into the optical system and reconstruct the transmission matrix T that relates the spectral components to the pixel intensities at the detector D. The calibration laser source is, thus, scanned over $\lambda_1, \ldots, \lambda_N$ wavelength steps, and the intensity level of $1, \ldots, M$ pixels are recorded at each stage. This results in a $M \times N$ T-matrix, where the i-th column contains all the intensities measured at λ_i:

$$D(x_1, \ldots, x_M) = T(\lambda, x)S(\lambda_1, \ldots, \lambda_N).$$

After this necessary calibration step, a generic spectrum S can be, in principle, reconstructed from the measured D-matrix and by inverting the T matrix: $S = T^{-1}D$. In practice, however, this simple inversion is numerically unstable in the presence of noise and is, therefore, combined with a non-linear optimization process seeking a response S that minimizes the euclidean L_2 norm $\|D - TS\|_{L2}$. Following this approach, the minimum measurable wavelength shift $\Delta\lambda$ corresponds to the FWHM of the spectral correlation function of speckle intensity $C(d\lambda)$ for input light λ: $C(d\lambda, x) = \frac{\langle D(\lambda,x)D(\lambda+d\lambda,x)\rangle}{\langle D(\lambda,x)\rangle\langle D(\lambda+d\lambda,x)\rangle} - 1$, where the averaging operation is performed for each pixel x at all wavelengths.

The number of distinguishable spectral channels is related to the number of modes supported by the fiber with a resolution of the order of 1 pm at 1500 nm being achievable using a 100 m step-index MMF optical fiber.[21] The resolving power and bandwidth scale linearly with fiber length and the number of supported modes, respectively. Speckle-based spectrometers do not have fixed free spectral ranges—their working bandwidth is determined by the range over which the calibration is performed—unlike more conventional spectrometers that can tune their bandwidth by rotating the dispersive element that, in this case, cannot be rotated.

The main breakthrough of these devices lies in the fact that multiple scattering from a small size random medium can reach high spectral sensitivities: a resolution of 0.6 nm over a bandwidth of 25 nm was demonstrated for an integrated random photonic structure large only a few tens of micrometers,[22] a result which would be impossible to obtain using a traditional dispersive micro-spectrometer design.

The main drawbacks of speckle-based spectrometers are represented by the need of tunable laser sources for their calibration, and the fact that their resolving power grows with increasing optical path length, which in turn makes the whole system particularly sensitive to environmental fluctuations, vibrations, and temperature changes. This poses a practical limit to the stability of these devices, which require frequent recalibrations to make sure that the observed intensity pattern fluctuations are due to the frequency content of the input signal rather than external factors. Similarly, the polarization and spatial incoupling of the input signal into the speckle-generating system must remain as stable as possible, e.g., by using first a polarization-maintaining single-mode fiber, which, however, limits the available signal.

Conceptually, this approach requires some basic *a priori* knowledge of the measured signal, as this defines the range over which the calibration laser must be scanned to perform the required characterization of the transfer matrix of the system. The final performance of this class of devices is eventually largely determined by the quality of the reconstruction algorithms implemented in the post-processing

step. Significant improvements have been recently demonstrated thanks to the introduction of well-established compressed sensing approaches, principal component analysis (PCA), or machine learning, which are particularly apt to the pattern recognition tasks associated with this technique.

In the context of speckle-based spectroscopy, assuming a certain optical path length, the super-resolution barrier is represented by the inherent decorrelation of the speckle patterns with frequency. A broadband enhancement of the spectral resolution can be achieved by favoring evanescent coupling between different windings of spiraled waveguide coils, as an additional mechanism to further scramble the propagating modes together.[18] Even more remarkably, in a recent demonstration by Bruce et al., the speckle correlation limit was overcome by eight orders of magnitude down to the attometer scale[23] by resorting to a PCA decomposition, which proved to be an optimal basis on which to measure speckle modulations. First, a training set of normalized speckles in the spectral range of interest is used to build the covariance matrix, whose principal components represent the basis of the dataset on which the measured speckle of the unknown spectrum is projected. To obtain a sufficiently large spectral range, the technique needs to be integrated with the transmission matrix method. Even more interesting perhaps is the fact that in the PCA, basis different components seem to be linked to the characteristic variations associated with different environmental factors such as temperature or external variations, which allows for potentially interesting speckle-based multi-sensing applications. Moreover, this allows it to decouple to some extent the resolution from the fiber length. Indeed, it has been observed that by extending the fiber over 50 m, the resolution obtained with PCA reconstruction remains constant, allowing the use of shorter fibers, less sensitive to external parameters.

The combined use of PCA and transmission-matrix characterization is also the core of the wave-meter implemented by Metzger et al.,[19] which allows achieving sub-femtometer resolution over a broad bandwidth spanning over the visible and near infrared range. In their proof of concept, light is injected through a single mode-fiber into an integrating sphere. A CMOS camera records the speckle produced by the interference of the scattered waves inside the sphere. As an application, the authors demonstrate the stabilization of a narrow laser line with relevant applications in metrology and atomic interferometric measurements. Similar applications can be envisioned for the spectrometer demonstrated in the work of Coluccelli et al.,[24] consisting of a multi-mode fiber and a camera for speckle acquisition. The system is coupled to an optical frequency comb light source and can be used for high-resolution spectroscopy experiments, also thanks to the careful thermal and mechanical insulation measures that have been adopted that increase stability, robustness, and compactness of the device.

Even in their super-resolved version, one advantage of speckle spectrometry applications is their measurement speed. By using a fast camera, the exposure time can be reduced to microseconds enabling an acquisition rate into the tens of kHz.[23] For laser line stabilization applications, as reported in Ref. 19, the update rate of the control loop using PCA to detect speckle variation is limited to 200 Hz. However, the actual measurement duration should account also for the time spent during the repeated frequency calibration step, which requires a full frequency sweep over the spectral range of interest using a tunable laser, and the corresponding acquisition time to collect a set of speckle

pattern images for each frequency. This wavelength calibration must typically be performed at regular intervals every few minutes to compensate for environmental fluctuations and drifts in order to avoid systematic errors in the frequency identification or artifacts in the spectral response of the instrument.

Coupling a trained deep learning model with speckle spectrometry holds promise to improve the precision of laser line stabilization by rejecting more efficiently the environmental and instrumental noise,[25,26] while only one example has been reported to date on the use of neural networks to actually improve the resolution of the state-of-the-art PCA approach, as well as the spectral bandwidth.[27]

B. Super-resolved spectroscopy based on optimization algorithms

Compressed sensing is a very broad processing technique used to reconstruct a signal through a limited number of measurements. It relies on the assumption that the target signal has a sparse representation in some predetermined domains (e.g., in terms of its Fourier or its wavelet components), i.e., that it can be represented with high fidelity by combining only a few elements with non-zero amplitude taken from a suitable basis. When this is the case, a most likely signal shape can be guessed numerically based on a limited set of measurements even below the Nyquist sampling theorem.

Whenever a signal is sparse under some representation, then its information content can be compressed. Typical examples include natural images, vector fields, and audio signals for which several compression schemes have been developed. On the contrary, typically incompressible signals include TV static, uncorrelated (white) noise, or signals that are already compressed. Exploiting the sparse representability of a signal, it is often possible to perform just a small fraction of random measurements to infer what combination of few non-zero elements in the sparse representation are consistent with the measurements and, hence, with the complete dataset. Recently, compressed sensing has been applied to spectroscopy showing promising results. The basic idea is to measure a spectrum with a reduced number of measurements and to retrieve it from the compressed measurements using reconstruction algorithms. Some examples are based on etalon arrays, Fabry–Pérot resonators, 2D-thin film filters, and nanophotonic structures.[8,20,28–30] In mathematical terms, the measurement column vector y with L components is represented using the relation

$$y = Mx, \qquad (1)$$

where x is a column vector of N components representing the spectrum of the illumination source and M is the sensing matrix of the optical structure (e.g., a matrix of transmittance filters, where each row represents a transmission spectrum). Because the length of the measurement vector is smaller than the length of the spectrum vector ($L < N$), the system is inevitably underdetermined with infinite possible solutions. In the transformed domain (transformation matrix P), y and, therefore, x can be represented as sparse vectors, $x = Pa$, where a is the transformed vector with few non-zero components in the transformation domain, from which it follows that

$$y = MPa. \qquad (2)$$

In the works of Donoho and Càndes,[31,32] it is shown that for most underdetermined systems, the sparsest solution a compatible with y

can be recovered by solving a convex optimization problem, namely, by finding the solution that minimizes the L_1 norm of the vector a. Then by finding the sparsest a vector components that minimize the L_1 norm and verify Eq. (2), it is possible to retrieve the complete dataset of y. A second necessary requirement is that matrix M needs to be composed of uncorrelated and random measurements with respect to the transformation basis to obtain an incoherent sampling covering a broad range in the transform domain (e.g., a periodic sampling in direct space corresponds in the Fourier-transform domain only to sense a single frequency).

We should stress that the concept of sparsity can be used with different meanings, depending on the application. In compressed sensing, sparsity is a purely mathematical concept implying that most elements of a vector in the transformed basis are zero. This property is only connected to the possibility of compressing a physical signal, not of increasing its resolution. If anything, compressed sensing leads to a partial loss of information and, therefore, of resolution, while on the other hand, it helps reduce a signal's complexity and memory footprint.

In the more general field of digital signal processing, a complementary approach to compressed sensing is used to achieve spectral super-resolution reconstruction by solving an under-determined system of equations instead of using compression. This kind of reconstructive spectroscopy is based on filter arrays and is very performant in terms of scalability and high spectral resolution. Moreover, due to its deterministic nature, robustness against perturbations, and one-time calibration requirement, filter-array spectroscopy represents one of the fastest methods for spectral reconstruction. In the early implementations of these techniques, non-negative least squares algorithms were used to estimate and restore the target spectrum using low-quality and low-cost filter arrays.[33]

A resolution improvement based on these principles has been demonstrated by Oliver et al.[34,35] In this case, the spectral reconstruction is achieved by projecting the target spectrum onto a random basis of M non-ideal broadband spectral filters. Using a regularization algorithm, a highly resolved spectral signal is reconstructed based on the assumption the signal was sparse in the observation domain. As in compressed sensing, the number of M filters can be reduced significantly, even though, in this case, the super-resolution effect can only be obtained via an oversampling of the unknown spectrum in contrast with the typical scope of compressed sensing approaches [see Fig. 1(b)].

When discussing super-resolved reconstructive spectroscopy, a mandatory mention is in order for hyperspectral imaging applications. This field, which deals with spectral augmentation strategies for images, shares some affine traits with multi-filter array reconstruction. A typical example is that of predicting the hyperspectral content of an image starting from a single RGB shot,[36] which is analogous to a (multi-pixel) reconstruction of a three-filter array (the RGB channels) into a few tens of output channels distributed across the visible spectrum. Considerable research efforts have been devoted to the development of spectral super-resolution strategies for hyper-spectral imaging due to the relevant applications in remote sensing applications. By exploiting the inherent sparsity of natural images in terms of their spectral content (analogously to the spatial sparsity, but in the spectral domain), low-cost and fast demonstrations were reported based on learning the relationship between RGB and hyperspectral images.[37,38]

Deep learning approaches have also been proposed[39,40] for this task, which have reached state-of-the-art performance by leveraging also the spatial information contained in the image and using the surrounding context information provided by the scene to infer more precise spectral information.[41,42]

As such, however, it seems not possible to directly translate these methods to the domain of general spectroscopic applications, where there is no spatial information and illumination can comprise extremely narrow spectral features, which are typically absent under natural conditions.

III. STOCHASTIC OPTICAL RECONSTRUCTION SPECTROSCOPY (STORS)

The concept of stochastic sparse sampling at the basis of the work of Boschetti et al.[43] allows one to reconstruct a spectrum with resolution surpassing that imposed by the response function of the detection apparatus. In the frequency domain, this is obtained by exploiting the emission characteristics of a pulsed random laser in a chaotic regime. In this regime, the emission of a random laser is characterized by few separated narrow peaks at random and uncorrelated frequencies from shot to shot, spanning over the gain bandwidth of the active medium. A target transfer function can, therefore, be stochastically probed—over a continuous frequency range—in a transmittance experiment. By collecting a large number of random peak frequencies and amplitudes, it is possible to reconstruct the transmission function irrespectively of the spectral width or shape of the instrument response function of the detection apparatus [see Fig. 1(c)]. The reconstruction is possible since a random sub-sampling of a dense space is repeated sequentially, allowing to over-determine a transfer function at each frequency free of any resolution constraint, provided that the random laser peaks in each emission spectrum are well separated in frequency. In its simplest implementation, this approach allows reconstructing arbitrary spectral features with no a priori information, independently from the input light polarization, at a resolution that is not limited by the detection apparatus.

Input light polarization is not critical as it would not change the results of a spectral reconstruction. In the case of bi-refringent, optically active, or chiral materials where a more complete characterization may be required in terms of polarization, the input light can be simply filtered after the chaotic laser source without affecting the general working principle of the technique.

The time needed for a complete spectral reconstruction, given the purely statistical nature of this method, depends on the amount of narrow peaks generated by the chaotic light source per unitary frequency range. In the case of an optically pumped random laser, the sparse frequency sampling regime required for the reconstruction is achieved by pumping the light source slightly above threshold and by adjusting the pumping geometry, gain level, and concentration of scatterers. To make a quantitative example, assuming a target spectral range and a spectral resolution of 10 and 0.01 nm, respectively, at least 1000 sampling random laser peaks are needed as a lower limit for the reconstruction. Assuming a broad instrumental response of ∼1 nm, the random laser peak spectral density should be lower than 0.5 peak/nm. Using a more conservative value of 0.2 peaks/nm, even in the best case of not overlapping peaks or repeated occurrences (i.e., uniformly distributed lasing peak frequencies), a minimum of 500 random laser spectra would be required. At a pump laser repetition rate

of 10 Hz, this statistics is reached in ~1 min of acquisition time. The subsequent numerical postprocessing for the spectral reconstruction can be neglected on this timescale.

Due to its stochastic sampling nature, compressed sensing approaches represent a straightforward option to speed up the measurement, which would significantly reduce the size of the statistical ensemble required for accurate reconstruction of the transfer function—and therefore, the measurement duration.

The main advantages of the technique reside in its independence on the spectral response of the instrument and the robustness endowed by the chaotic emission of optically pumped random laser sources, which are characterized by cheap fabrication costs and high robustness. It is interesting to note that random frequency instabilities observed in traditional laser sources can also be used, in principle, to perform a STORS reconstruction. This is particularly relevant for applications with solid-state laser sources and could avoid expensive fabrication steps to stabilize their cavity resonances.

On the downside, a few open problems are still hindering its practical applications. Namely, the useful bandwidth depends on the width of the gain curve of the active medium. In this respect, stochastic spectral reconstruction approaches can serve a complementary role in wavelength ranges where stable and narrow tunable laser sources are not available. Additionally, alternative pumping schemes (e.g., electric, CW optical pumping) would be desirable to avoid the need of optical pulsed pumping of the random laser. While current electrically pumped random laser sources still are hampered by a lower degree of randomness and sparsity in their emission lines, important progress has recently been made in this direction.[44–46]

It is worth noting that, in STORS, the concept of "sparsity" has a more physical meaning, which refers to the need of sampling only a few well-separated points during each measurement in order to surpass the instrument resolution. In a way, this approach is deeply different from compressed sensing, indeed several redundant measurements are performed at each position while collecting the statistical ensemble needed to reconstruct the signal. In this respect, the two

approaches can be seen as complementary. Moreover, compressed sensing can help optimize the performance of STORS by minimizing the number of independent measurements needed to retrieve the spectrum—similarly to what has been already demonstrated in stochastic microscopy applications.[47] It is important to realize that super-resolution techniques are enabled by an oversampling of the signal in some domain, while all compression techniques, as the term suggests, will typically degrade the resolution or at best maintain it. In other words, a spectrum is reconstructed with increased resolution if it is overdetermined in some domain.

IV. PRACTICAL LIMITATIONS

As concerns reconstructive spectroscopy approaches, their main limitation lies in the need of *a priori* information about the target spectrum for correct reconstructions. Even in this case, these techniques are often prone to artifacts and reconstruction of spurious features due to numerical instabilities in the underlying underdetermined optimization problem. This stands in contrast with direct measurement methods such as STORS, where no prior information is required. This makes STORS applicable to the reconstruction of arbitrary transfer functions including both narrow and broadband features, even if the spectral content of an emitting light source cannot be extracted (see also Fig. 2). The same is not granted for reconstructive spectroscopy (based on either speckle, random encoders, or filter matrices), for which the successful reconstruction of a spectrum relies on the strong assumption that the signal is sparse in the direct domain of observation where the spectral reconstruction is carried out. This makes reconstructive approaches less suitable to reconstruct continuous spectral profiles—as multiple interference contributions at different frequencies sum their intensities incoherently—thus reducing the useful speckle contrast. Speckle spectrometers work properly when the signal contains few frequencies such as few sparse peaks. This is why these devices work well as wave-meters for laser lines stabilization, as monitors of known signals, and to measure shifts from a reference frequency up to attometer-accuracy (comb-speckle), rather than for
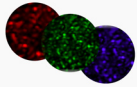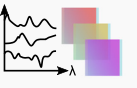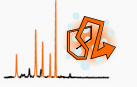
	prior calibration	a priori assumptions	compatible illumination	compatible application
speckle spectrometry	pattern database	sparse signal (L_1 regularization)	coherent, polarized, limited bandwidth (speckle contrast)	source characterization
filter-array reconstruction spectroscopy	filter database	sparse signal (L_1 regularization)	arbitrary light source (spatially homogeneous)	source, source + filter characterization
stochastic reconstruction spectroscopy	none	none	isolated narrow peaks	arbitrary transfer function

FIG. 2. Overview of recent spectral super-resolution techniques on the basis of four main criteria: (1) the need for *a priori* calibration using a high precision tunable laser and (2) the need for *a priori* knowledge about the spectral features of the collected light. This is a major requirement for techniques that rely on regularization algorithms for retrieving spectra, (3) the requirements on the illumination sources, and (4) the spectral information that can be retrieved from different targets, e.g., laser line wavelengths with speckle spectrometry or transmission functions using STORS.

general spectroscopic characterization or investigation, since some knowledge of the spectral function must be supposed. Similar considerations apply for reconstructive spectroscopy, as exemplified by the fact that small spurious features are often inadvertently introduced by the reconstruction process. Another limitation of filter-based spectroscopic instruments is related to the design of broadband random filters compatible with fabrication constraints and yet exhibiting highly uncorrelated spectral responses. Song *et al.* demonstrated a deep-learning approach to optimize the spectral responses of the filters taking into account practical fabrication and optical constraints to maximize the encoding capacity of a set of broadband stochastic filters, enhancing the reconstruction accuracy and reducing the sensitivity to fabrication errors.[48]

V. FUTURE DEVELOPMENTS

Although speckle spectroscopy is not suitable for spectroscopic investigation of transfer functions, it has great prospects of evolving toward top-level wavemeter devices, allowing the measurements of light source emission frequencies with high precision. A further development in this direction could be that of understanding and handling the spectral information encoded in the phase singularity points—the vortices—easily obtainable from CCD speckle images. These have shown to be highly sensitive to speckle fluctuation and could be used to improve this technology even more in the future.[49]

Stochastic optical reconstruction spectroscopy and generative spectroscopy based on filter matrices or encoders can be extended to spectroscopic investigations where small instruments and fast discrete-wavelength measurements are required—such as in quality control of food and water, gas monitoring, or waste sorting. The operation of stochastic optical reconstruction spectroscopy was recently demonstrated using an optically pumped random laser, which highlighted the ease of fabrication and how the narrow emission lines of a random laser could be used for reconstruction. At the moment, however, these spectral features depend on an optical pumping scheme, which is inadequate to be used in a compact and cost-effective device. The development of a miniaturized, high-resolution spectrophotometer is strongly relying on the progress made with the light sources to reach the resolution required for different applications. Electrically pumped random lasers or conventional laser sources without cavity stabilization, such as unprocessed diode lasers and distributed feedback lasers, represent good candidates, as they may exhibit spectral drifts and mode jumps enabling sparse sampling in the wavelength range of interest. Despite the numerous advances made in the last few years in the development of electrically pumped random lasers, however, we are still far from obtaining the emission performances, chaotic behavior, and narrow linewidths of the optically pumped sources needed for high resolution spectroscopy.

An attractive possibility in the near future is to explore the limit of the maximum resolution enhancement obtainable in STORS while relaxing the sparsity constraint on the probing peaks. Indeed, in its current formulation, this reconstruction technique works only if the sampling peaks are sparse enough compared to the instrumental response in order to obtain a convolution-free spectral reconstruction. Based on recent developments in super-resolution stochastic reconstruction microscopy, deep neural networks can be used to generate super-resolved images starting from a set of frames with denser distributions of point emitters, thus accelerating the overall reconstruction

process.[50] A similar approach could be applied to the spectral domain by creating a training dataset with denser spectra with possibly overlapping peaks of known frequency in order to reduce the amount of acquisitions during a spectral reconstruction measurement.

An important application not strictly related to super-resolution reconstruction is represented by spectral classification that often requires separating spectra that exhibit subtle (yet known) differences such as in the case of contamination detection or plastic identification and sorting for recycling purposes. By combining STORS and machine learning, a limited number of random laser shots may be sufficient to distinguish a consistent number of different objects, especially for spectra with few distinguishing features like polymers in the near-infrared region. A similar idea works also for reconstructive spectroscopy based on filters, where few filters with tuned spectral responses can perform a sparse sampling of the transmission spectra, classifying objects as a function of their main spectral fingerprints and the number of filters used. In these cases, rather than an oversampling of the data for improving resolution, a down-sampling or a "manual" compression at the experimental level is operated, reducing the amount of data needed for the identification. The power of sparse sampling approaches relies on the twofold ability of increasing resolution on one side, and performing spectral compression when irrelevant or redundant information is present. The latter approach can work very well if combined with feature selection machine learning algorithms based on the choice of the only spectral components useful to the recognition, speeding up the classification as well as reducing the memory requirements.

AUTHOR DECLARATIONS
Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

A. Boschetti: Conceptualization (equal); Visualization (equal); Writing – original draft (lead); Writing – review and editing (equal). **L. Pattelli:** Conceptualization (equal); Visualization (equal); Writing – original draft (supporting); Writing – review and editing (lead). **R. Torre:** Funding acquisition (equal); Supervision (supporting); Writing – review and editing (supporting). **D. S. Wiersma:** Conceptualization (equal); Funding acquisition (equal); Supervision (lead); Writing – original draft (supporting); Writing – review and editing (supporting).

DATA AVAILABILITY

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

REFERENCES

[1] J. Bao and M. G. Bawendi, "A colloidal quantum dot spectrometer," Nature **523**(7558), 67–70 (2015).

[2] Z. Yang, T. Albrow-Owen, H. Cui, J. Alexander-Webber, F. Gu, X. Wang, T.-C. Wu, M. Zhuge, C. Williams, P. Wang, A. V. Zayats, W. C. Dai, S. Hofmann, M. Overend, L. Tong, Q. Yang, Z. Sun, and T. Hasan, "Single-nanowire spectrometers," Science **365**(6457), 1017–1020 (2019).

[3] W. Hadibrata, H. Noh, H. Wei, S. Krishnaswamy, and K. Aydin, "Compact, high-resolution inverse-designed on-chip spectrometer based on tailored disorder modes," Laser Photonics Rev. **15**(9), 2000556 (2021).

[4] N. H. Wan, F. Meng, T. Schröder, R. J. Shiue, E. H. Chen, and D. Englund, "High-resolution optical spectroscopy using multimode interference in a compact tapered fibre," Nat. Commun. **6**(1), 7762 (2015).

[5] G. C. Valley, G. A. Sefler, and T. J. Shaw, "Multimode waveguide speckle patterns for compressive sensing," Opt. Lett. **41**(11), 2529–2532 (2016).

[6] Z. Zhang, Y. Li, Y. Wang, Z. Yu, X. Sun, and H. K. Tsang, "Compact high resolution speckle spectrometer by using linear coherent integrated network on silicon nitride platform at 776 nm," Laser Photonics Rev. **15**(11), 2100039 (2021).

[7] C. Kim, D. Park, and H. N. Lee, "Compressive sensing spectroscopy using a residual convolutional neural network," Sensors **20**(3), 594 (2020).

[8] Y. Oiknine, I. August, D. G. Blumberg, and A. Stern, "Compressive sensing resonator spectroscopy," Opt. Lett. **42**(1), 25–28 (2017).

[9] J. Wang, B. Zheng, and X. Wang, "Strategies for high performance and scalable on-chip spectrometers," J. Phys. **3**(1), 012006 (2020).

[10] Y. Wan, X. Fan, and Z. He, "Review on speckle-based spectrum analyzer," Photonic Sens. **11**(2), 187–202 (2021).

[11] H. Cao, "Perspective on speckle spectrometers," J. Opt. **19**(6), 060402 (2017).

[12] H. Cao and Y. Eliezer, "Harnessing disorder for photonic device applications," Appl. Phys. Rev. **9**(1), 011309 (2022).

[13] S. F. Liew, B. Redding, M. A. Choma, H. D. Tagare, and H. Cao, "Broadband multimode fiber spectrometer," Opt. Lett. **41**(9), 2029–2032 (2016).

[14] G. J. Hornig, T. R. Harrison, L. Bu, S. Azmayesh-Fard, and R. G. Decorby, "Wavemeter based on dispersion and speckle in a tapered hollow waveguide," OSA Continuum **2**(2), 495–506 (2019).

[15] D. Yi, Y. Zhang, X. Wu, and H. K. Tsang, "Integrated multimode waveguide with photonic lantern for speckle spectroscopy," IEEE J. Quantum Electron. **57**(1), 1–8 (2021).

[16] Y. Wan, X. Fan, S. Wang, Z. Zhang, S. Zhao, and Z. He, "Wavemeter capable of simultaneously achieving ultra-high resolution and broad bandwidth by using Rayleigh speckle from single mode fiber," J. Lightwave Technol. **39**(7), 2223–2229 (2021).

[17] B. Redding and H. Cao, "Using a multimode fiber as a high-resolution, low-loss spectrometer," Opt. Lett. **37**(16), 3384–3386 (2012).

[18] B. Redding, S. F. Liew, Y. Bromberg, R. Sarma, and H. Cao, "Evanescently coupled multimode spiral spectrometer," Optica **3**(9), 956–962 (2016).

[19] N. K. Metzger, R. Spesyvtsev, G. D. Bruce, B. Miller, G. T. Maker, G. Malcolm, M. Mazilu, and K. Dholakia, "Harnessing speckle for a sub-femtometre resolved broadband wavemeter and laser stabilization," Nat. Commun. **8**(1), 15610 (2017).

[20] Y. Kwak, S. M. Park, Z. Ku, A. Urbas, and Y. L. Kim, "A pearl spectrometer," Nano Lett. **21**(2), 921–930 (2021).

[21] B. Redding, M. Alam, M. Seifert, and H. Cao, "High-resolution and broadband all-fiber spectrometers," Optica **1**(3), 175–180 (2014).

[22] B. Redding, S. F. Liew, R. Sarma, and H. Cao, "Compact spectrometer based on a disordered photonic chip," Nat. Photonics **7**(9), 746–751 (2013).

[23] G. D. Bruce, L. O'Donnell, M. Chen, and K. Dholakia, "Overcoming the speckle correlation limit to achieve a fiber wavemeter with attometer resolution," Opt. Lett. **44**(6), 1367–1370 (2019).

[24] N. Coluccelli, M. Cassinerio, B. Redding, H. Cao, P. Laporta, and G. Galzerano, "The optical frequency comb fibre spectrometer," Nat. Commun. **7**(1), 12995 (2016).

[25] U. Kürüm, P. R. Wiecha, R. French, and O. L. Muskens, "Deep learning enabled real time speckle recognition and hyperspectral imaging using a multimode fiber array," Opt. Express **27**(15), 20965–20979 (2019).

[26] E. Edrei, N. Cohen, E. Gerstel, S. Gamzu-Letova, N. Mazurski, and U. Levy, "Chip-scale atomic wave-meter enabled by machine learning," Sci. Adv. **8**(15), eabn3391 (2022).

[27] R. K. Gupta, G. D. Bruce, S. J. Powis, and K. Dholakia, "Deep learning enabled laser speckle wavemeter with a high dynamic range," Laser Photonics Rev. **14**(9), 2000120 (2020).

[28] E. Huang, Q. Ma, and Z. Liu, "Etalon array reconstructive spectrometry," Sci. Rep. **7**(1), 40693 (2017).

[29] C. Kim, W. B. Lee, S. K. Lee, Y. T. Lee, and H. N. Lee, "Fabrication of 2D thin-film filter-array for compressive sensing spectroscopy," Opt. Lasers Eng. **115**, 53–58 (2019).

[30] Z. Wang and Z. Yu, "Spectral analysis based on compressive sensing in nanophotonic structures," Opt. Express **22**(21), 25608–25614 (2014).

[31] D. L. Donoho, "For most large underdetermined systems of linear equations, the minimal ℓ1 solution is also the sparsest solution," Commun. Pure Appl. Math. **59**(6), 797–829 (2006).

[32] E. Candes and J. Romberg, "Sparsity and incoherence in compressive sampling," Inverse Probl. **23**(3), 969 (2007).

[33] C. C. Chang and H. N. Lee, "On the estimation of target spectrum for filter-array based spectrometers," Opt. Express **16**(2), 1056–1061 (2008).

[34] J. Oliver, W. Lee, S. Park, and H. N. Lee, "Improving resolution of miniature spectrometers by exploiting sparse nature of signals," Opt. Express **20**(3), 2613–2625 (2012).

[35] J. Oliver, W. B. Lee, and H. N. Lee, "Filters with random transmittance for improving resolution in filter-array-based spectrometers," Opt. Express **21**(4), 3969–3989 (2013).

[36] R. M. Nguyen, D. K. Prasad, and M. S. Brown, "Training-based spectral reconstruction from a single RGB image," in European Conference on Computer Vision (Springer, Cham, 2014), pp. 186–201.

[37] B. Arad and O. Ben-Shahar, "Sparse recovery of hyperspectral signal from natural RGB images," in European Conference on Computer Vision (Springer, Cham, 2016), pp. 19–34.

[38] J. Aeschbacher, J. Wu, and R. Timofte, "In defense of shallow learned spectral reconstruction from RGB images," in Proceedings of the IEEE International Conference on Computer Vision Workshops (IEEE, 2017), pp. 471–479.

[39] S. Galliani, C. Lanaras, D. Marmanis, E. Baltsavias, and K. Schindler, "Learned spectral super-resolution," arXiv:1703.09470 (2017).

[40] Z. Shi, C. Chen, Z. Xiong, D. Liu, and F. Wu, "HSCNN+: Advanced CNN-based hyperspectral recovery from RGB images," in Proceedings of the IEEE Conference on Computer Vision and Pattern Recognition Workshops (IEEE, 2018), pp. 939–947.

[41] J. Li, C. Wu, R. Song, Y. Li, and F. Liu, "Adaptive weighted attention network with camera spectral sensitivity prior for spectral reconstruction from RGB images," in Proceedings of the IEEE/CVF Conference on Computer Vision and Pattern Recognition Workshops (IEEE, 2020), pp. 462–463.

[42] J. He, Q. Yuan, J. Li, and L. Zhang, "PoNet: A universal physical optimization-based spectral super-resolution network for arbitrary multispectral images," Inf. Fusion **80**, 205–225 (2022).

[43] A. Boschetti, A. Taschin, P. Bartolini, A. K. Tiwari, L. Pattelli, R. Torre, and D. S. Wiersma, "Spectral super-resolution spectroscopy using a random laser," Nat. Photonics **14**(3), 177–182 (2020).

[44] W. Margulis, A. Das, J. P. von der Weid, and A. S. Gomes, "Hybrid electronically addressable random fiber laser," Opt. Express **28**(16), 23388–23396 (2020).

[45] T. L. Shen, H. W. Hu, W. J. Lin, Y. M. Liao, T. P. Chen, Y. K. Liao, T. Y. Lin, and Y. F. Chen, "Coherent Förster resonance energy transfer: A new paradigm for electrically driven quantum dot random lasers," Sci. Adv. **6**(41), eaba1705 (2020).

[46] A. Consoli, N. Caselli, and C. López, "Electrically driven random lasing from a modified Fabry–Pérot laser diode," Nat. Photonics **16**(3), 219–225 (2022).

[47] L. Zhu, W. Zhang, D. Elnatan, and B. Huang, "Faster STORM using compressed sensing," Nat. Methods **9**(7), 721–723 (2012).

[48] H. Song, Y. Ma, Y. Han, W. Shen, W. Zhang, Y. Li, X. Liu, Y. Peng, and X. Hao, "Deep-learned broadband encoding stochastic filters for computational spectroscopic instruments," Adv. Theory Simul. **4**(3), 2000299 (2021).

[49] A. Dávila and J. A. Rayas, "Single-shot phase detection in a speckle wavemeter for the measurement of femtometric wavelength change," Opt. Lasers Eng. **125**, 105856 (2020).

[50] E. Nehme, L. E. Weiss, T. Michaeli, and Y. Shechtman, "Deep-STORM: Super-resolution single-molecule microscopy by deep learning," Optica **5**(4), 458–464 (2018).