Enhanced resolution in argon and neon spectra using a Super-Resolution algorithm

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Abstract – This paper presents the principles and application of a super-resolution (SR) technique, based on a $L^1$-norm minimization procedure. In particular, the technique is applied to improve low-resolution resolution spectra as obtained from the optogalvanic effect in neon and argon discharges over the 413–423 nm wavelength range. By applying the super-resolution algorithm to the experimental data, a surprising 70-fold reduction of the linewidth is achieved allowing to resolve prior indistinguishable peaks. It is found that further improvements on the resolution are limited by the signal-to-noise ratio of the original spectra. The importance of a suitable mathematical representation of the experiment and the discussion on other applications of this super-resolution technique in spectroscopy are also presented.

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Introduction. – In atomic and molecular spectroscopy it is desirable to resolve relevant spectroscopic features present in the spectra by achieving the highest possible resolution. Unfortunately, the light sources and instruments employed in experiments induce limits in the final resolution of the measurements. To tackle this limitation experimentalists traditionally resort to line-stabilized, high-resolution light sources along with Doppler free spectroscopy techniques [1] or supersonic sample cooling [2]. These techniques present, however, the inconvenience of involving relatively elaborated experimental arrangements. Here, we present a novel mathematical technique aimed at helping experimentalists to improve the spectral resolution of their data. This technique, called Super-Resolution (SR) is based on the mathematical tools developed within the scope of Compressed Sensing (CS) [3,4]. The basic idea behind CS is that, under the a priori knowledge that a signal or an image has a sparse representation (see below), this can be reconstructed from far fewer data/measurements as compared to those required by the well-known Shannon/Nyquist sampling theory [5]. Here, the term sparsity means that there exists a basis in the space of functions (e.g. a Fourier basis or wavelets) where the signal can be represented by a small number of elements. In spectroscopy, a spectrum of a single element or molecule can be represented as a linear combination of single peaks of a similar shape centered at their specific transition wavelength values. In this case, the spectrum can be represented by a small number of parameters, namely the locations of the transitions, their corresponding widths, and their amplitudes. These parameters constitute the sparse representation in this case.

In a general context, CS can be regarded as an optimal sampling and interpolation algorithm for sparse signals in the appropriate base.

Along the ideas of CS, some new versions of algorithmic or computational SR algorithms have been recently developed. These SR algorithms have been successfully applied to many practical cases, including medical imaging [6], satellite imaging, and video applications (see, for instance, sect. 1.2 in [7] and the references therein).

In the context of the present work, SR algorithms are applied to the particular case of recovering high-resolution features, such as peak centers, from experimental spectra. We show here that this kind of mathematical problem can be treated by the recently developed mathematical theory of SR by Candès and Fernandez-Granda [8,9].
The aim of this paper is to show that SR algorithms can also be a valuable tool in spectroscopy by allowing to resolve peaks that cannot be separated from the original data due to instrumental limits in spectral resolution [10,11]. In particular, we show here that the SR algorithm can dramatically improve the resolution of measurements, in particular allowing a surprising effective 70-fold reduction in the linewidth of the original spectral measurements. This improvement allows to separate apart real structures that are not resolved in the original experimental data. In particular, in order to test and quantitatively show the power of the SR technique, we present here experimental optogalvanic spectra of well-known neon and argon atomic transitions. Upon the application of the SR technique we can quantify dramatic improvements in the experimental data. In particular, in order to test and quantitatively show the power of the SR technique, we present here experimental optogalvanic spectra of well-known neon and argon atomic transitions. Upon the application of the SR technique we can quantify dramatic improvements in the resolved features. The optogalvanic effect (OGE) consist of a change in the total current of a plasma when interacting with laser light. The change is resonant when the laser wavelength corresponds to an atomic or molecular transition of the gas species present in the plasma [12]. Although the specific example shown here is devoted to the OGE effect, we believe SR algorithms can be applied in other spectroscopic areas, including photoionization, photoabsoption, Raman or others. To the best of our knowledge this is the first application of SR algorithms based on CS ideas aimed at improving the resolution of spectroscopic measurements.

Super-Resolution algorithms. – In any imaging system, the resolution limit is measured by the shortest separation between two features, before they mingle into one. The limitations of the instrument used and, on a more fundamental level, the diffraction limit set the constraints to the maximum experimentally achievable resolution in any optical system. There are currently several instances and many different approaches to SR in imaging, and we invite the reader to read the exciting applications of optical SR currently being developed (e.g. [13] and references therein). In an analogous manner as in optics, in any type of optical spectroscopy, the spectral resolution is limited by the linewidth of the light source and quantified by the minimum separation in wavelength required to clearly separate apart two different transitions. This has been traditionally considered the ultimate limit of resolution in spectroscopy. Since the applications of SR techniques to spectroscopy are scarce and not widespread in the experimental community, we will give a basic introduction in the context of optogalvanic spectroscopy.

Let us denote by \( y \) a one-dimensional acquired signal as obtained from a source \( x \) through an optical or any other measuring device. In our particular case \( y \) corresponds to the optogalvanic signal and \( x \) corresponds to the original spectrum as produced by the physical interaction of the laser with the plasma. Let us assume, as it happens in our optogalvanic case, that the values of \( y \) and \( x \) both depend on the wavelength \( \lambda \). A mathematical model of such a process can be described by the following convolution model:

\[
y(\lambda) = \int_0^\infty G(\lambda - s)x(s)ds + w(\lambda).
\]

Here, \( G \) represents, mathematically, the effect of the measurement equipment/process upon the original physical data, \( s \) is an integration variable and \( w \) is a noise term that can be deterministic or stochastic and such that

\[
\|w\|_{L^1} = \int_0^\infty |w(\lambda)|d\lambda < \varepsilon,
\]

but otherwise arbitrary. Here, \( \|w\|_{L^1} \) denotes the continuum version of the vector norm, colloquially known as the taxicab norm. Notice that \( \varepsilon \geq 0 \) is a constant measuring the noise level of the acquired signal. In the reciprocal space (1) becomes

\[
\hat{y}(\nu) = \left\{ \begin{array}{ll}
\hat{G}(\nu)\hat{x}(\nu) + \hat{w}(k), & \text{for } k \in [-\nu_c, \nu_c], \\
\hat{w}(\nu), & \text{otherwise},
\end{array} \right.
\]

where \( \hat{G} \) denotes the Fourier transform of the function \( G \) and \( \nu = 1/\lambda \) is the usual definition of wavelength per unit distance. In this mathematical representation the instrument \( G \) can be seen as a low-pass filter that limits the bandwidth of an otherwise ideal and spectrally narrow source signal. In (3) \( \nu_c > 0 \) is the cutoff number of wavelength per unit distance of such a filter. For the moment, and for simplicity in the exposition, we discard the noise term of (2) by letting \( \varepsilon = 0 \).

Formally, the aim of the SR algorithms is to find an estimate \( \hat{x}_{\text{est}} \) of the source \( x \) such that

\[
\left\{ \begin{array}{ll}
(i) \ G \ast \hat{x}_{\text{est}} = \hat{G} \ast \hat{x} \text{ on } [-\nu_c, \nu_c], \\
(ii) \ \hat{x}_{\text{est}} \text{ is supported in } [-\nu_{\text{hi}}, \nu_{\text{hi}}],
\end{array} \right.
\]

where \( \nu_{\text{hi}} > \nu_c \) and \( \hat{x}_{\text{est}} \) denotes the Fourier transform of the estimate \( x_{\text{est}} \). In simpler terms, the aim of SR is to use the frequency-limited information as produced by the instrument (or light source) to make an extrapolation (in this case \( x_{\text{est}} \)) to higher frequencies in reciprocal space. This extrapolation would allow, upon an inverse Fourier transformation, to obtain a far richer spectrum, i.e. the “super-resolved” spectrum as compared to that obtained originally from \( y \), which corresponds to the measured spectrum. A quantitative measure of the improvement in our signal can be defined in terms the SR Factor \( \text{SRF} = \nu_{\text{hi}}/\nu_c \). An alternative way to measure the effect of the SR algorithm on the data is to define the percentage improvement of the signal (PEIS) as \((1 - \text{SRF})^{-1} \times 100 \). It is well known that the problem given as in (4) is hopefully ill posed, namely the solution \( x_{\text{est}} \) may not exist or may not be unique [14]. This means that, based on a truncated Fourier transform, one may find a very large set of extrapolations that faithfully represent the original data within the interval \([-\nu_c, \nu_c]\), but which are wildly different outside this interval. These many solutions would not
necessarily have any physical meaning even if they satisfied (4). Remarkably, in the case where the source $x$ is composed of a “well-separated” train of point-like sources, problem (4) becomes tractable. This kind of sources can be modeled by

$$x = \sum_{j \in T} a_j \delta_{\lambda_j},$$

where $\lambda_j$ and $a_j$ are the locations (in the spectrum) and intensities of the point sources, respectively, $\delta_\tau$ is the Dirac delta function\(^1\) and $T$ is a given set of numeration indexes.

As mentioned in the introduction, the above model can be used as a simple model for atomic or molecular transitions. In these, their linewidths, even when Doppler broadened, are very narrow as compared to the spectral resolution of the laser used in the experiment. We emphasize again that the models and method in this paper will be only useful to determine the locations and relative intensities of the spectral lines. The present analysis is not valid when the linewidth of the laser is small as compared to the Doppler broadening of the transitions. For such a case a more realistic modeling of the transition linewidths on the source should be proposed, but that goes beyond the scope of this work.

Next, we define the minimum separation of the point sources as $\Delta = \min_{j,j \in T} |\lambda_j - \lambda_l|$, where $\lambda_i, \lambda_j$ denote again their locations on the spectral line. The main result of the SR theory states that, if the point sources are sufficiently apart from each other, namely

$$\Delta \geq 2/\nu_c,$$

then there exists an efficient algorithm that recovers exactly the original source. In other words, problem (4) has a unique solution $x_{\text{est}} = x$. This solution can be computed by solving a convex minimization problem [9] that is, finding the minimum of a convex function under constraints (e.g. (7) below). Notice that there is no bound on $\nu_c$. So, in principle, at least mathematically, the improvement on the resolution may be infinite (namely, $\text{SRF} = +\infty$), and the location of the point sources can be determined with infinite precision. As we will see later on, the presence of noise imposes a natural constraint on $\text{SRF}$, hence on the improvement that can be obtained by the estimate $x_{\text{est}}$.

Before stating the SR algorithm in detail, we need another ingredient. In the digital signal processing, as in any digital data acquisition experiment, there is a finite size in the wavelength step that is acquired, so sources and signals can be both regarded as discrete finite vectors $x = (a_1, \ldots, a_N)$ and $y = (y_1, \ldots, y_N)$ measured at a fixed grid of equally spaced wavelengths $\lambda$.

In the case of continuous signals, the filter resolution is defined by $\nu_c$, that is, the width of the low-pass filter $G$ (see fig. 1) and it is related to the physical properties of the experimental setting. In the experiments we also have another loss of resolution due to the finite size of

\(^1\)In mathematical rigor $\delta_\tau$ is a Dirac measure.

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Super-Resolution algorithm for the argon and neon spectra

![Fig. 1: (Colour online) Convolution of a train of point sources with a Gaussian filter. The graphs on the right show: the results for a continuous signal (top); the same signal after sampling on a discrete grid (bottom). The basic features of the continuous signal can be recovered by an interpolation of the discrete one. Hence, regarding the filter $G$, both graphs have the same cutoff wavelength per unit distance $\nu_c$.](image)

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sampling, i.e. the wavelength step. Notice that the latter loss of resolution may change, for example, by reducing the scanning step of the laser used in the experiment, but this does not affect the resolution of the filter $G$. The aim of SR algorithms is to reverse (or deconvolve) the effect of the filter $G$ on the source signal. Once the sampling step size is smaller than a certain threshold, its effect on the SR algorithms is negligible. This important difference, combined with the unboundedness of the $\nu_c$ and the separated point source nature of the source signal, explain why, in our case, the improvement in resolution seems to be independent of the finite sampling step size (see fig. 1).

Now, again discarding noise, eq. (1) becomes

$$y_\ell = \sum_{j=1}^N G(\lambda_\ell - s_j) a_j, \quad |\lambda_\ell| \leq \nu_c^{-1},$$

for $\ell \in \{0, \ldots, N-1\}$, or in a more compact notation we write $y = Gx$, where $G$ is an $N \times N$ matrix whose entries are given by $G_{ij} = G(\lambda_\ell - s_j)$. Hence, the SR problem (4) is reduced to finding out the entries $a_j$ of the vector $x$. As proposed in [9], to solve this problem it is enough to solve a convex minimization program, namely

$$\begin{cases}
\min_{\tilde{x}} \|\tilde{x}\|_1, \\
\text{subject to } G\tilde{x} = y,
\end{cases}$$

where $\|\tilde{x}\|_1 = \sum_{j=1}^N |a_j|$. The main result in [9] is that the problem (7) has a unique solution $x_{\text{est}}$ that coincides with the original $x$ over the original range of inverse space, but has a much larger span of harmonics beyond the interval defined by the original cutoff wave number. This is a surprising result and problem (7) is at the core of the mathematical theory of SR. The main feature of the $L^1$-minimization, that “explains why” the solution of (7) recovers the original signal, that is, it is an efficient way to detect the support of the sparsest signals (that is, signals that contain the least number of peaks) among all possible signals that produce the same observable data $y$ via the filter $G$. The role of the minimum separation condition (6)
is to make the solution of problem (7) unique and hence it must coincide with the original source signal. Another important feature of problem (7) is that it is a convex optimization problem under linear constraints. This is a standard problem in non-linear programming that even some spreadsheet software is able to solve. In the presence of noise, that is for \( \varepsilon > 0 \), the reconstruction given by (7) will not be exact [8], but should instead satisfy

\[
\| G(x_{\text{est}} - x) \|_{L^1} \leq C_0 (\text{SRF})^2 \varepsilon,
\]

where \( C_0 > 0 \) is a positive constant. Therefore, the quality (or accuracy) of the estimated vector \( x_{\text{est}} \) degrades linearly with the noise and quadratically with the SRF. Hence, there is a tradeoff between the noise level and the SRF that limits the performance of the SR algorithm. Is important to emphasize that, in order to apply this algorithm successfully, a good control on the signal-to-noise ratio is required in the original experimental data.

**Filter model in optogalvanic spectroscopy.** – The last ingredient that is needed to apply the above SR algorithm (7) to any experimental setting is to identify filter \( G \) and estimate its parameters. In general, the shape of the optogalvanic peaks does not present a unique profile condition and it may depend, for example, on the current of the plasma [15]. Nevertheless, it is known that the upper part of each peak can be well approximated at first order by a Gaussian profile, and the lower part by a Lorentzian. Since the aim of our algorithm is to identify the position and relative strength of the peaks and these positions are well correlated with the upper part of the peaks, we propose, in a heuristic way, the following Gaussian form of the filter

\[
\hat{G} (\nu) = B \exp \left( -\frac{\nu^2}{a^2} \right), \quad \text{for } |\nu| < \nu_c,
\]

where \( B \) is a normalization constant and \( a \) corresponds to the effective broadening of the experimental peaks. Note that it is convenient to write \( G \) in terms of its Fourier transform since in this case it is a diagonal operator (recalling that the Fourier transform of a Gaussian is also a Gaussian). What remains is simply to estimate the parameters \( \nu_c, a \) and \( B \) that more closely model the experiment. Notice that \( \nu_c \) is the cutoff frequency and it is an important limit since it represents the frequency (in Fourier or reciprocal space) where the noise-to-signal ratio is small enough to consider the signal consisting of pure noise.

In order to estimate the filter, we propose here a simple but effective approach: given the above functional form for the filter, we proceeded to obtain the filter parameters using high-resolution experimental data \( x_{\text{know}} \) from a gas species. In our present case these correspond to neon optical transitions as reported by NIST [16] and its corresponding spectrum as measured by our apparatus \( y_{\text{fit}} \). The procedure to obtain the parameters of the filter is started by, first, computing the Fourier transform of the measured spectrum and the NIST reference spectrum. Second, since in Fourier space the action of filter is multiplicative (see (3)), the filter can be thus be obtained as the quotient of the above quantities. That is

\[
\hat{G} (\nu) = \frac{y_{\text{fit}} (\nu)}{x_{\text{know}} (\nu)}, \quad \text{for } \nu \in [-\nu_c, \nu_c],
\]

where for \( \nu \notin [-\nu_c, \nu_c] \) the fraction \( \frac{y_{\text{fit}} (\nu)}{x_{\text{know}} (\nu)} \) only contains the wave number information coming from the noise term (see fig. 2). To explain our fitting method to the experimental data of neon, in fig. 2 we show a plot of the discrete cosine transform \(^2\log |y_{\text{fit}}/x_{\text{know}}| = \log \hat{G}\) for some of our neon and NIST data on a particular interval. First, starting at \( \nu = 0 \) there is a noisy signal mounted on a smooth decreasing curve that after a certain point \( \nu_c \) can be regarded as pure noise. The point where the signal behavior changes becomes the estimated cutoff frequency \( \nu_c \).

Once \( \nu_c \) is known, the parameters \( a \) and \( B \) can be determined by a linear regression procedure for \( \log (|y_{\text{fit}}/x_{\text{know}}|) \) on the interval \( (0, \nu_c) \). To have a more robust estimation, the determination of \( \nu_c \) and the linear regression is repeated on intervals around different peaks and then the reported values are given by their average value. This approach to estimate the parameters is effective and good enough for the current application.

**Experiment.** – The experimental arrangement employed is shown in fig. 3. It consists of three main parts: the light source, the plasma discharge, and the detection part. The electrodes, of planar geometry and with a separation gap of 7 mm, were made of copper. The argon and neon used was of research grade purity. The vacuum system consists of a Pfeiffer turbomolecular pump of 100 l/s that allows us to reach a base pressure of \( 10^{-5} \) torr. Pressure was measured by using a MKS baratron capacitive manometer of type 626 and a Varian Eyesys mini-BA vacuum gauge. We use a DC power supply HP6516 with a voltage range of \((0-3000) \) volts.

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\(^2\)Fourier and cosine transforms are equivalent in this case, here we use the latter because it has the advantage of letting us work with real signals only.
To excite the plasma we used an Ekspla Optical Parametric Oscillator (OPO) pumped by a Nd:YAG pulsed laser. For the optogalvanic effect the voltage variations were recorded using a ballast resistor of 3.9 Kohm. This voltage was amplified by a SR280 Stanford Research Systems boxcar averager. The power of the laser was measured using an Ophir pyroelectric sensor PE10-S. These measurements were sent to a National Instruments USB-6009 Data Acquisition System (DAQ) and collected in a computer using a LabVIEW automatization program.

The laser resolution in the visible range reported here was 0.1 nm and its wavelength was varied in an automated way. For each wavelength we collected several voltage values and the values reported here are an average of at least 20 of these.

Results and discussion. – The argon discharge in fig. 4 was obtained with a step wavelength variation of 0.1 nm. For the neon discharge in fig. 5 the wavelength variation was in steps of 0.1 nm for the (600–650) nm wavelength range and 0.2 nm for the (650–700) nm range.

For the filter parameters estimation we used four different wavelengths ranges (windows) for the neon experiment and one for the argon experiment. The latter was used to double-check that the main features of the filter were the same for both experiments. The high-resolution experimental signal data used to calculate the filter parameters corresponded to the transitions reported in the NIST atomic database for neon [16]. This was used as the known source signal \( x_{\text{know}} \). The windows were chosen in such a way that some clearly defined peaks of the source signal \( x_{\text{know}} \) were contained inside a single peak in the measured signal \( y_{\text{fit}} \). In table 1 we show the neon windows considered, the values of the cutoff wave number \( \nu_c \) and filter parameter \( a \). The mean values for this experiment are \( a = 6.87 \times 10^8 \text{ cm}^{-1} \) and \( \nu_c = 14.37 \times 10^8 \text{ cm}^{-1} \). The considered window for argon was 414.5–417.5 nm and the obtained parameters are \( a = 6.70 \times 10^8 \text{ cm}^{-1} \) and \( \nu_c = 14.7 \times 10^8 \text{ cm}^{-1} \), these values are within 2% and 5% correspondence with the neon mean values, respectively.

As explained in the previous sections, the resolution of the estimated signal \( x_{\text{est}} \) is given by \( \nu_{\text{hi}} = 1/\lambda_{\text{hi}} \), and the SR algorithm does not prescribe any bound on \( \nu_{\text{hi}} \) (or on the smallness of \( \lambda_{\text{hi}} \)), therefore, in principle, it can be chosen at will. In practice, this is not possible due to the signal-to-noise ratio and the numerical stability in the solution of (7). The latter problem is a consequence of the fact that, for small \( \lambda_{\text{hi}} \) it is necessary to solve a large linear system of \( \sim 1/\lambda_{\text{hi}} \) equations that may be numerical ill conditioned. The numerical scheme of the SR algorithm and the adjustment of the filter parameters was performed as follows: First, all signals, \( x_{\text{know}}, y_{\text{fit}} \) and \( y \) were linearly interpolated onto a fix grid equally spaced of step size \( \lambda_{\text{hi}} = 10^{-4} \text{ nm} \), which yields \( \nu_{\text{hi}} = 10^{12} \text{ cm}^{-1} \). Second, the filter parameters were estimated by the above procedure, and third, the minimum in problem (7) was found to obtain \( x_{\text{est}} \). There is a final consideration to be made regarding a source of error that originates from the experimental offset in the measured intensity of the peaks produced by the boxcar amplifier. This error is assimilated in the model of the noise term \( w \) in (1) as an additive constant. By adjusting this offset, by means of another
additive constant, the performance of the SR algorithm improves dramatically. In the presented experiments this value was set so as to lower the signal by 1.6% with respect to the highest peak (near 420 nm). The numerical implementation was based on the useful Matlab code from [17]. In fig. 6 we show a comparison between the argon optogalvanic spectra before and after applying the SR algorithm. The peaks in the region 419–421 nm previously unresolved are now distinguishable due to the SR algorithm. The physical reality of these separated peaks was verified by comparing the data obtained by the SR algorithm to the data base. We point out that the identification of the appropriate filter $G$ that models accurately the experiment and the estimation of its parameters are two of the main issues to be addressed in order to correctly use of SR algorithms. We believe that the SR algorithm could have a major impact on the analysis of experimental spectroscopic data. This is because it makes possible to obtain high-resolution spectral features without resorting to expensive and sophisticated experimental infrastructures.

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