Gauss-Newton based kurtosis blind deconvolution of spectroscopic data

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The spectroscopic data recorded by dispersion spectrophotometer are usually degraded by the response function of the instrument. To improve the resolving power, double or triple cascade spectrophotometer and narrow slits have been employed, but the total flux of the radiation decreases accordingly, resulting in a lower signal-to-noise ratio (SNR) and a longer measuring time. However, the spectral resolution can be improved by mathematically removing the effect of the instrument response function. Based on the Shalvi-Weinstein criterion, a Gauss-Newton based kurtosis blind deconvolution algorithm for spectroscopic data is proposed. Experiments with some real measured Raman spectroscopic data show that this algorithm has excellent deconvolution capability.

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The spectra recorded by dispersion spectrometer are usually degraded by the response function of the instrument. The major factors of the instrument response function limiting the resolving power are the slit width and the diffractive limit of the dispersion grating. To improve the resolving power, double or triple cascade spectrometer, and narrow slits have been employed[1], but the total flux of the radiation available decreases accordingly, resulting in a low signal-to-noise ratio (SNR) and a longer measure time. Hence, there is a trade-off between the resolving power, the SNR, and the measure time. However, spectral resolution can be significantly improved by mathematically removing the effect of the instrument response function. If the instrument response function is a prior knowledge, it is named deconvolution. The most popular method is Wiener filtering[2]. In practical applications, the instrument response function is often unknown in advance, even if it can be measured accurately, it may change along with the time. So the blind deconvolution is a more significant work by which the true spectra can be estimated from the measured data. Senga *et al.*[3] developed a blind deconvolution method based on homomorphic filtering and applied it to the blind deconvolution of infrared molecular absorption spectra successfully. However, this method requires calculating the logarithm of the Fourier spectrum, which is time consuming, and is limited to the case where the spectral slit function is triangle. The Jansson algorithm[4] starts from an initial guess of the true spectra, and estimates them iteratively. But the choice of the convolution kernel is often not true. Yuan *et al.*[5,6] proposed a high-order statistical algorithm of blind deconvolution for spectroscopic data.

In this letter, based on the Shalvi-Weinstein criterion[7], a kurtosis Gauss-Newton based algorithm is proposed for spectroscopic data blind deconvolution. The real measured spectroscopic data experimentally demonstrate the feasibility of this method. Most spectroscopic data measured by spectrophotometer can be mathematically modeled as a convolution of the true spectrum and a unit-impulse response function[4] (convolution kernel), i.e.,

$$\mathbf{x} = \mathbf{f} * \mathbf{w}, \tag{1}$$

where $*$ denotes the convolution operation, and $\mathbf{w} = (w(1), w(2), \cdots, w(N))^{\mathrm{T}}$ is the true spectrum, $\mathbf{x} = (x(1), x(2), \cdots, x(N))^{\mathrm{T}}$ is the measured spectroscopic data, $\mathbf{f} = (f_0, f_1, f_2, \cdots)^{\mathrm{T}}$ is the convolution kernel.

The true spectrum can be restored by the inverse operation of the convolution in principle

$$\mathbf{y} = \mathbf{g} * \mathbf{x}, \tag{2}$$

where $\mathbf{y} = (y(1), y(2), \cdots, y(N))^{T}$ is the deconvoluted data, and $\mathbf{g} = (g_0, g_1, \cdots, g_Q)^{T}$ is the unit impulse response function (deconvolution kernel) of the deconvolution operation with finite length $Q+1$.

For the ideal deconvolution, the deconvoluted data $\mathbf{y}$ should satisfy

$$y(n) = w(n). \tag{3}$$

But for high-order statistical blind deconvolution algorithm (for real value data), only following equation can be achieved approximately[7]

$$y(n) = \pm w(n-m), \tag{4}$$

where m is the spectral-shift parameter which means that, compared with the true spectrum, the estimated spectrum may have spectral shift m. This shift and the sign ambiguity can be corrected by the correlation operation.

So the cascades of the convolution and deconvolution should satisfy

$$\mathbf{h} = \mathbf{g} * \mathbf{f} = \delta, \tag{5}$$

or in matrix form

$$\mathbf{h} = \mathbf{F}\mathbf{g} = \delta, \tag{6}$$

where $\mathbf{h} = (h_0, h_1, h_2, \cdots)^{T}$, δ denotes the Kronecker delta function, and $\mathbf{F}$ is a Teoplitz matrix with $Q+1$ columns and possibly an infinite number of rows whose elements are

$$\mathbf{F}_{ij} = f_{i-j}, \quad 0 \le j \le Q. \tag{7}$$

If **f** is known in advance, **g** is known accordingly, the operation is named deconvolution which can be resolved by Wiener filtering method[2]. Unfortunately, in most practical applications, **f** is not known exactly, **w** should be estimated from the measured data **x**, it is hereby named blind deconvolution.

Because all the spectroscopic data are real value, the normalized kurtosis based Shalvi-Weinstein criterion of blind deconvolution[7] is $\max|k_y(4,2)|$, where

$$k_y(4,2)=\frac{\mu_y(4)}{\mu_y^2(2)}-3 \tag{8}$$

is the kurtosis of the estimated data **y**, and $\mu_y(k)$ is the kth central moment of **y**, which can be obtained by

$$\mu_y(k)=E\left\{[\mathbf{y}-m_y]^k\right\},\quad \text{for}\quad k=2,4, \tag{9}$$

where $E\{\cdot\}$ denotes the expectation operation, $m_y=E\{\mathbf{y}\}$ is the mean of **y**. To maximize the kurtosis amplitude $|k_y(4,2)|$, the necessary condition is that the gradient of $|k_y(4,2)|$ with respect to **g** is equal to zero vector, namely

$$\mu_y(2)\nabla_{\mathbf{g}}[\mu_y(4)]=2\mu_y(4)\nabla_{\mathbf{g}}[\mu_y(2)]. \tag{10}$$

Gauss-Newton algorithm[8] is a well known optimization algorithm. However, for the spectroscopic data blind deconvolution, it has special formulation. In the following, we will provide the Gauss-Newton algorithm for the application. Only at the optimal point, these equations are satisfied, we hereby define a nonzero error vector

$$\mathbf{e}(\mathbf{g})=\mu_y(2)\nabla_{\mathbf{g}}[\mu_y(4)]-2\mu_y(4)\nabla_{\mathbf{g}}[\mu_y(2)], \tag{11}$$

then perturb the deconvolution kernel **g** to **g**+**d** in a manner to drive the perturbed error vector $\mathbf{e}(\mathbf{g}+\mathbf{d})$ closer to zero vector than $\mathbf{e}(\mathbf{g})$. By linear approximation, we obtain

$$\mathbf{e}(\mathbf{g}+\mathbf{d})\approx\mathbf{e}(\mathbf{g})+\mathbf{M}_{\mathbf{g}}\mathbf{d}, \tag{12}$$

where the $(Q+1)\times(Q+1)$ matrix $\mathbf{M}_{\mathbf{g}}$ is given by

$$\mathbf{M}_{\mathbf{g}}=\mu_y(2)\mathbf{H}_{\mathbf{g}}(4)-2\mu_y(4)\mathbf{H}_{\mathbf{g}}(2)$$
$$+\nabla_{\mathbf{g}}[\mu_y(4)]\nabla_{\mathbf{g}}[\mu_y(2)]^T-2\nabla_{\mathbf{g}}[\mu_y(2)]\nabla_{\mathbf{g}}[\mu_y(4)]^T, \tag{13}$$

and the $(Q+1)\times(Q+1)$ matrix $\mathbf{H}_{\mathbf{g}}$ is

$$\mathbf{H}_{\mathbf{g}}(k)_{ij}=k(k-1)E\{(\mathbf{y}-m_y)^{k-2}$$
$$[x(n-i)-E\{x(n-i)\}][x(n-j)-E\{x(n-j)\}]\}. \tag{14}$$

The perturbation vector **d** is selected so as make the error vector equal zero vector, which is obtained by solving Eq. (12)

$$\mathbf{d}^0=-\mathbf{M}_{\mathbf{g}}^{-1}\mathbf{e}(\mathbf{g}). \tag{15}$$

Before applying the algorithm above-proposed to the real spectra, proper length $Q+1$ should be decided. Sufficiently long $Q+1$ can result in sufficient deconvolution. However, too long $Q+1$ may degrade the accuracy of the central moments in Eq. (9). We set $Q+1=\Delta_n$ simply, where Δ_n is the full-width at half-maximum (FWHM) of the narrowest spectral line. **g** is

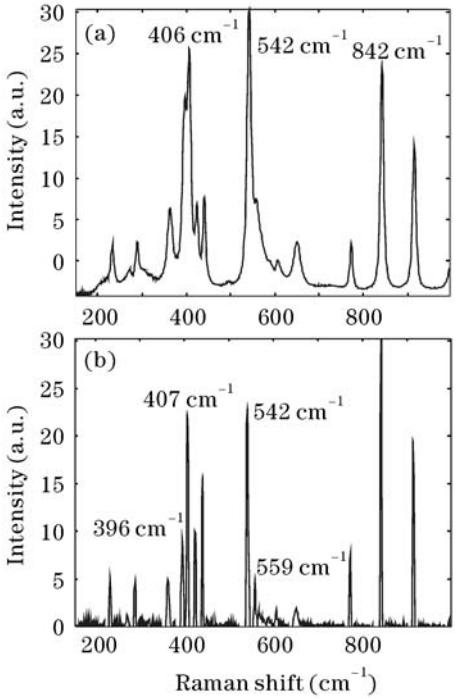

Fig. 1. (a) A set of Raman spectroscopic data of glucopyranose from 153 to 999 cm^{-1} with length $N=847$; (b) the estimated spectrum of (a).

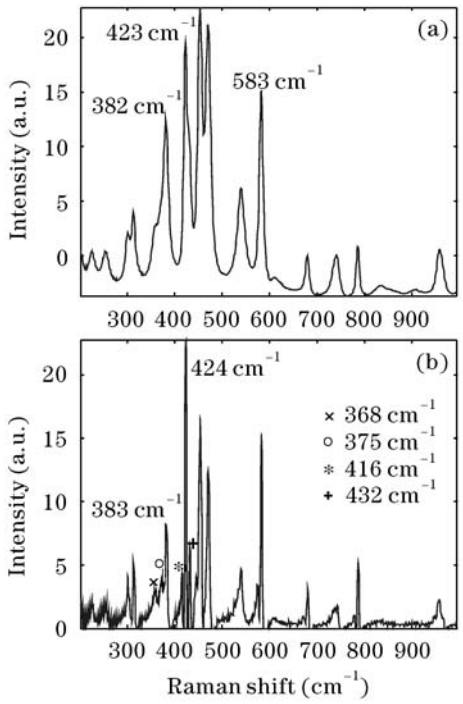

Fig. 2. (a) A set of Raman spectroscopic data of D-glucuronic from 613.7 to 4800 cm^{-1} with length $N=794$; (b) the estimated spectrum of (a).

initialized randomly with the constraint $\|\mathbf{g}\| = 1$.

To demonstrate the feasibility of this algorithm, we apply it to two real measured Raman spectra. Figure 1(a) is a set of Raman spectroscopic data of glucopyranose from 153 to 999 cm^{-1} with length $N = 847$, Fig. 1(b) is the estimated true spectrum. Here, $Q = \Delta_{842} - 1 = 6$. It is clearly displayed that the spectral resolution is improved remarkably. The peak at 406 cm^{-1} is split into two peaks at 396 and 407 cm^{-1} respectively. The peak at 542 cm^{-1} is split into two peaks at 542 and 559 cm^{-1} respectively. Figure 2(a) is a set of Raman spectroscopic data of D-glucuronic from 613.7 to 4800 cm^{-1} with length $N = 794$, Fig. 2(b) is the estimated spectrum. Here $Q = \Delta_{583} - 1 = 8$. The peak at 382 cm^{-1} is split into three peaks at 368, 375, and 383 cm^{-1} respectively. The peak at 423 cm^{-1} is split into three peaks at 416, 424, and 432 cm^{-1} respectively. From the experiments abovementioned, excellent decnvolution results are achieved.

Based on the Shalvi-Weinstein criterion, a normalized kurtosis based algorithm for spectroscopic data blind deconvolution is proposed. With the experiments on some real measured Raman spectroscopic data, this algorithm has excellent deconvolution capability. The spectral resolution can be improved considerably. The length of the deconvolution kernel $Q + 1$ influences the deconvolution results. Sufficiently long $Q + 1$ can result in sufficient deconvolution. However, too long $Q+1$ may degrade the accuracy of the central moments. We simply set $Q + 1$ be equal to the FWHM of the narrowest spectral line in above-mentioned experiments. However, because the dimensions of matrices $\mathbf{M_g}$ and $\mathbf{H_g}$ are $(Q + 1) \times (Q + 1)$, for the deconvolution kernel with longer length $Q + 1$, it can be very computationally demanding.

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