Convolution and Fourier Transform: from Gaussian and Lorentzian Functions to q-Gaussian Tsallis Functions

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Convolution and Fourier Transform: from Gaussian and Lorentzian Functions to q-Gaussian Tsallis Functions

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Abstract - A discussion is here proposed regarding the Voigt function, that is the convolution of Gaussian and Lorentzian functions, and the Lévy and q-Gaussian Tsallis distributions. The Voigt and q-Gaussian functions can be used as line shapes in Raman spectroscopy for fitting spectra. Using the convolution theorem, we can obtain the relaxations which are producing the Voigt line shape. To determine the relaxation governing the q-Gaussian line shape, we need to use the Lévy symmetric distribution, since the direct Fourier transform of the q-Gaussian is a very complicated function. According to the work by Deng, 2010, the q-Gaussian functions are mimicking the Lévy functions in an excellent manner. Being the Fourier transform of the Lévy function a stretched exponential relaxation, we can argue that the same mechanism is producing the q-Gaussian line shape. Moreover, using the convolution theorem for the q-Gaussians, we can further generalize the relaxation mechanism.

Keywords - Tsallis q-Gaussian Distribution, Voigt Distribution, pseudo-Voigt Distribution, Lévy Distribution, Convolution Theorem

Introduction

In Raman spectroscopy, Gaussian and Lorentzian function seem being the most popular for fitting Raman spectra. Besides these two functions, linear combinations (pseudo-Voigt distributions) or convolutions of them (Voigt distributions) are used too (Meier, 2005). The Voigtian convolution function turns out into a bell shape with a Gaussian kernel and wings which are asymptotically of the Lorentzian form. As determined by Cope and Lovett, 1987, the asymptotic solution of Voigtian expansion has the leading term equal to $a_0 \,/\, \pi x^2$). In some literature, the line wings are also mentioned as "tails". The use of the Voigt or pseudo-Voigt functions is suggested (Meier, 2005) because the spectral bands are characterized by intermediate profiles between Lorentzian and Gaussian outlines (Kirillov, 2004). However, other functions exist which have an intermediate character; they are the q-Gaussian functions.

q-Gaussians, also known as "Tsallis functions", are probability distributions derived from the Tsallis statistics (Tsallis, 1988, 1995, Hanel et al., 2009). The q-Gaussians are based on a generalized form of the exponential function (see discussion in Sparavigna, 2022), characterized by a continuous parameter q in the range 1 < q < 3. As given by Umarov et al., 2008, the q-Gaussian function is based on function $f(x) = Ce_q(-\beta x^2)$, where $e_q(.)$ is the q-exponential function and C a constant. The q-exponential has expression: $\exp_q(u) = [1 + (1-q)u]^{1/(1-q)}$, then function $f(x)$ possesses a bell-shaped profile. In the case that we have the peak at position x_o, the q-Gaussian is given as:

$$\text{q-Gaussian} = C\exp_q(-\beta(x-x_o)^2) = C[1-(1-q)\beta(x-x_o)^2]^{1/(1-q)}$$

For q equal to 2, the q-Gaussian is the Cauchy-Lorentzian distribution (Naudts, 2009). For q close to 1, the q-Gaussian is a Gaussian. Consequently, for the q-parameter between 1 and 2, the shape of the q-Gaussian function is intermediate between the Gaussian and the Lorentzian profiles.

Here our main aim is that of finding the relaxation mechanism which is producing the q-Gaussian line shape. We start from the Voigt function and its Fourier transform and discuss its physics using the convolution theorem. Then we will consider the Fourier transform of the q-Gaussian. Let us note that an expression for the Fourier transform of the q-Gaussian has been given by Rodrigues and Giraldi, 2016, but it is very complicated and therefore it is very difficult to extract the relaxation mechanism from it. For this reason, we will use the Lévy distribution and its Fourier transform.

The convolution

According to Townsend, 2008, the "Voigt function looks like Gaussian for small *x* (i.e., near line center), and like Lorentzian for large *x* (i.e., out in line wings)". We can also appreciate this fact by observing the pseudo-Voigt function, which is generally used for approximating the Voigt function. Being the pseudo-Voigt the linear combination of Gaussian and Lorentzian functions, the wings must be necessarily Lorentzian and the kernel Gaussian-like.

If we use Voigt functions or pseudo-Voigt functions for fitting spectra, the wings of the Raman lines will be always described by a Lorentzian behavior. It is reasonable to ask ourselves: is this always the experimental case? That is, are we always observing Lorentzian wings for the Raman bands? To answer this question, we started investigation in ChemRxiv0.

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We observed that a generalization of Voigt and pseudo-Voigt functions obtained by means of q-Gaussians can help us in describing the leading term of the line wings, that is, to measure the wing power law, besides telling whether it is Lorentzian or not. The q-Gaussians are therefore the solution of the question. Here we consider further investigation about the relaxation mechanisms, using the convolution theorem.

The Voigt profile is a convolution of a Lorentz distribution L and a Gaussian distribution G given by:

$$V(k;\sigma,\gamma) = \{G*L\}(k) = \int_{-\infty}^{\infty} G(k';\sigma)L(k-k';\gamma)dk'$$

where k, in spectroscopy, is representing the shift from the line center k_o, and

$$G(k;\sigma) = \frac{e^{-k^2/(2\sigma^2)}}{\sigma\sqrt{2\pi}},\ L(k;\gamma) = \frac{\gamma}{\pi(k^2+\gamma^2)}.$$

The convolution theorem states that the Fourier transform of a convolution of two functions is the pointwise product of their Fourier transforms. Let us consider two functions $g(x), h(x)$ and their Fourier transforms $G(k), H(k)$. We have the convolution as:

$$r(x) = \{g*h\}(x) = \int_{-\infty}^{\infty} q(\xi)h(x-\xi)d\xi$$

According to the convolution theorem:
$\mathcal{F}\{r(x)\}(k) = \mathcal{F}\{\{g*h\}(x)\}(k) = G(k)\cdot H(k)$,
$\mathcal{F}, \mathcal{F}^{-1}$ indicate the Fourier transform and anti-transform. Also:
$r(x) = \{g*h\}(x) = \mathcal{F}^{-1}\{G(k)\cdot H(k)\}(x)$,

Consequently, the Fourier transform of the Voigt function is the pointwise product of the Fourier transforms of Gaussian and Lorentzian functions:
$\mathcal{F}\{G*L\}(x) = \mathcal{F}\{G(k)\}(x)\cdot\mathcal{F}\{L(k))(x)$
Let us add that the Fourier transform of a Gaussian is a Gaussian, that is, in the Wolfram formalism[1] :

$$\mathcal{F}\{e^{-ax^2}\}(k) = \sqrt{\frac{\pi}{a}}e^{\pi^2k^2/a},$$

and in the case of the Lorentzian function[2]:

$$\mathcal{F}\{\frac{1}{\pi}\frac{\frac{1}{2}\Gamma}{(x-x_o)^2+(\Gamma/2)^2}\}(k) = e^{-2\pi ikx_o-\Gamma\pi|k|}.$$

This is the characteristic function of the Cauchy distribution. We can put the center at zero, so that:

$$\mathcal{F}\{\frac{\Gamma}{2\pi}\frac{1}{x^2+(\Gamma/2)^2}\}(k) = e^{-\Gamma\pi|k|}$$

Let us note that methods for the fast computation of Voigt Function are based on the Fourier transform too (Schreier, 1992, Mendenhall, 2007, see please also the discussion by Vogman, 2010).

If we consider the Fourier transform of the Voigt function, from the frequency domain to the time domain, we have a function of time as:
$f(t) = e^{-a|t|-bt^2} = e^{-a|t|}e^{-bt^2}$ (1)

Then we have two mechanisms, which are giving the Voigt convolution line shape, and we will consider them further in the following section.

Besides being involved in Raman spectroscopy, the Voigt convolution is used for the spin resonance lines too. In solids, these lines "are broadened by a number of mechanisms" (Stoneham, 1972). "Some of these mechanisms give a Gaussian lineshape, such as dipolar broadening in concentrated crystals (Van Vleck 1948) and strain broadening by dislocations (Stoneham 1966, 1969). Other mechanisms lead to a Lorentzian lineshape, such as the relaxation broadening due to the finite lifetime of a state". Stoneham is also telling that "If the mechanisms which lead to Lorentzian and Gaussian broadening are *independent*, the lineshape is just the convolution of a Gaussian and a Lorentzian". As we have seen using the convolution theorem, from the Voigt function we arrive to the product of two distributions. The product is suitable for independent distributions.

The convolution of two Lorentzian functions is a Lorentzian function, and the convolution of two Gaussians is a Gaussian. Applying the convolution theorem, we have the two functions of time:
$f(t) = e^{-a|t|}$ (2)
$f(t) = e^{-bt^2}$ (3)
Function (1) is representing one of the possible "intermediate" time functions between (2) and (3). For $t \to 0$, the exponential function prevails. For $t \to \infty$, the Gaussian decay prevails. In the frequency domain we have the opposite behavior, so that the Voigt function has a Lorentzian tail.

Let us consider the stretched exponential too:
$f(t) = e^{-|ct|^\alpha}$ (4)
And here it is necessary to consider the Lévy distribution,

As reported in Hristov, 2023, "the Lévy distribution with a Lévy index α is a distribution having a characteristic function" which is given by (4). The Gaussian distribution appears when α = 2, while for α = 1, we find the Lorentzian distribution. Therefore, the Lévy distribution is another function with a behavior intermediate between Gaussian and Lorentzian line

[1]https://mathworld.wolfram.com/FourierTransformGaussian.html

[2]https://mathworld.wolfram.com/FourierTransformLorentzianFunction.html

shapes, like the q-Gaussian Tsallis function. In Deng, 2010, we can find the relationship between them, that is, between Lévy and Tsallis functions.

The Lévy distribution, symmetric and with center at location zero, - Deng is giving - has the characteristic function being (4). The distribution is therefore (Deng, 2010):

$$f(x;\alpha,c) = \frac{1}{2\pi}\int_{-\infty}^{\infty} e^{-|ct|^{\alpha}} e^{-itx} dt$$

Deng is writing the q-Gaussian in the form:

$$qG(x;q,T) = C(1-(1-q)\frac{x^2}{T^{2/(3-q)}})^{1/(1-q)}$$

Comparing Lévy and Tsallis distributions, Deng investigated "the relationship between the parameters (α,c) and (q,T)". "Results show that they are usually totally different, except for two special cases of normal and Cauchy distributions. However, they can be approximated to each other through linking equation in (21) or (30) [of the Deng's article] depending on whether or not the kurtosis parameter is $\alpha < 0.3490$. When $\alpha \geq 1$, [let us remember that $\alpha = 1$ means Lorentzian and $\alpha = 2$ means Gaussian] the match quality between the Lévy and Tsallis distributions is either perfect or excellent. When $\alpha < 1$, the quality deteriorates a bit. When $\alpha < 0.3490$, except on the narrow region near origin where the two have a significant difference, the two match very well on the heavy tails".

This means that, in the case of matching, that is for $\alpha > 1$, the q-Gaussian functions are mimicking perfectly the Lévy functions; therefore, the characteristic function, which is the Fourier transform of the Tsallis distribution, can be seen as well-approximated by the stretched exponential of the Lévy distribution.

Spectroscopy and convolution

For what is regarding the relaxation mechanisms, let us further consider the words by Orazio Svelto, 1970, about the homogeneous broadening of the photon emission. In the case that we have a dipole damped oscillator model, we can observe the spectral line of the spontaneous emission with a "natural" or "intrinsic" broadening, coming from a relaxation mechanism given by (2). This homogeneous broadening produces a line profile (in the frequency domain) described by a Lorentzian function. Orazio Svelto is also mentioning the photon-phonon interaction as generating homogeneous broadening and therefore a Lorentzian line shape too. An inhomogeneous broadening (such as those caused by the Doppler effect and thermal effect, with relaxation (3)) is giving a Gaussian line shape. However, the most observed case is that of an intermediate profile, given by the convolution of the resonance relative probability and the broadening function, because natural band can be modified by different mechanisms (Svelto, 1970).

We have mentioned the natural broadening giving a Lorentzian profile, the thermal broadening introducing a Gaussian profile, and the general intermediate profiles as the most common cases. A consequence is that the Voigt profile is generally used to simulate the intermediate case. "Alternatively, [we can] suppose that the line [Lorentzian] is scanned by a spectrophotometer with a Gaussian sensitivity function" (Tatum, 2022). Then, in this experimental framework, we have the convolution of the line with the instrumental function profile. Let us remember that "the general expression that takes account of all the instrumentally induced distortion of the true band shape can be called the instrument function" (Seshadri and Jones, 1963). It is also known as the "instrumental transfer function" (Merlen et al., 2017).

As told by S.G. Rautian, 1958, each monochromatic component of the true radiation is replaced by the instrumental function, which is given by the convolution of the radiation with the "distortions [produced] both in the optical and recording parts of the apparatus" (Rautian, 1958). In Rautian, 1958, we can find several different instrumental functions that can be convoluted with the true radiation. And the true radiation can be a convolution of different broadening mechanisms. The Voigt convolution is based on Lorentzian and Gaussian profiles because the analysis starts from a Lorentzian damping model (natural radiation) with a weight which is a Gaussian one. Different approaches are existing (Kirillov, 2004), so that the true radiation line can be assumed different from a Lorentzian function; moreover, the weight function can be different from a pure Gaussian function.

In Merlen et al., 2017, researchers are telling that "If we do not take into account the instrumental transfer function that can be negligible in many cases (...), the total intensity of one phonon mode with a wavevector q_0 and a frequency $\omega(q_0)$, in a perfect crystal, is spread on a symmetric profile which is Lorentzian". Merlen et al. are also discussing the presence of asymmetric peaks in the framework of the approach by Richter et al., 1981. In the case of investigating the first order region of the Raman spectra of carbonaceous materials, for its fitting procedure, Merlen et al. suggest the use of Lorentzian and Gaussian functions for symmetric profiles and of the Breit-Wigner-Fano (BWF) line shape for asymmetric peaks (in particular they tell for the case of "One band: The G band is fitted by a Lorentzian if symmetric, and by a BWF if not symmetric"). We have discussed the G and G' bands of graphite in ChemRxiv1 and we

used q-Gaussians. We used q-Gaussians too for other carbonaceous materials (biochar and nanotubes) in SSRN1. The behavior of the peaks we considered is q-Gaussian, that is, not Lorentzian or Gaussian but q-Gaussian. For what is regarding the BWF asymmetric line shape, a discussion is proposed in ChemRxiv2, regarding the Raman LO mode band in Silicon Carbide.

For the spectra previously considered (Sparavigna, 2023), we have shown that q-Gaussians are able of fitting the bands in a successful manner for graphite, ChemRxiv1, anatase ChemRxiv3, SERS spectra, ChemRxiv4. But we have also seen that in the far-wing region, in the case of diamond (SSRN2), a linear combination of q-Gaussians can provide the best result. It seems that two different relaxation mechanisms are required in this case.

Convolution of q-Gaussian functions

The fact that the q-Gaussians are mimicking the Lévy functions tells us that we can well approximate the Fourier transform with a stretched exponential. Literature tells that the stretched exponential relaxation is able of fitting many relaxation processes in disordered and quenched electronic and molecular systems (Phillips, 1996). Consequently we can tell that, if the Raman band is described by a q-Gaussian, the relaxation can be approximated by the function (4). However, as previously told, in some cases it is better to use a linear combination of q-Gaussians. In fact, we used this combination with the aim of generalizing the pseudo-Voigt functions, which are linear combinations of Lorentzian and Gaussian functions.

Can a linear combination of q-Gaussians be used to represent a convolution of q-Gaussians? This is a question which is requiring further investigations. To conclude the proposed discussion, let us return to the convolution theorem and to the Fourier transform of the Voigt function, which is the pointwise product of the Fourier transforms of Gaussian and Lorentzian functions:

$$\mathcal{F}\{G*L\}(x) = \mathcal{F}\{G(k)\}(x) \cdot \mathcal{F}\{L(k)\}(x)$$

We can write, in the same manner, the convolution of a q-Gaussian (qG) with a Lorentzian function:

$$\mathcal{F}\{qG*L\}(x) = \mathcal{F}\{qG(k)\}(x) \cdot \mathcal{F}\{L(k)\}(x)$$

This corresponds to the product (in the time domain, that is consider x representing the time, and according to Lévy function):

$$f(t) = e^{-|ct|^{\alpha}} e^{-a|t|}$$

In the same manner we can consider the q-Gaussian convoluted with a Gaussian function:

$$\mathcal{F}\{qG*G\}(x) = \mathcal{F}\{qG(k)\}(x) \cdot \mathcal{F}\{G(k)\}(x)$$

Again, according to Lévy functions, in time domain:

$$f(t) = e^{-|ct|^{\alpha}} e^{-bt^2}$$

Between these two cases we have the convolution of two q-Gaussians (consider please the "q" indices as different in general):

$$\mathcal{F}\{qG*qG\}(x) = \mathcal{F}\{qG(k)\}(x) \cdot \mathcal{F}\{qG(k)\}(x)$$

so that:

$$f(t) = e^{-|ct|^{\alpha}} e^{-|dt|^{\beta}}$$

This last expression is generalizing that related to the Voigt convolution.

In the investigation of the Raman spectra, besides the q-Gaussians, we have used a linear combination of them to approximate their convolution, in analogy with the case of the Voigt convolution. The two q-Gaussians are therefore appearing as a consequence of two different broadening mechanisms, related to two different time relaxations.

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