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An attempt to apply the Wigner-Ville distribution in analysis of a rhodamine 6G photoacoustic pulsed signal

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Abstract

The non-stationary signal analysis methods based on Joint Time-Frequency Representations appears to be a valuable tool for pulsed PA signal investigation. The thermal deactivation of metastable states (dimers) of Rhodamine 6G in PVA using pulsed photoacoustic spectroscopy was studied. Analysis of Rhodamine 6G photoacoustic pulsed signals involving Wigner-Ville Distribution indicate monomer-dimer forms energy transfer.

The Joint Time-Frequency Representations (JTFRs), especially Wigner-Ville Distribution (WVD) are the most promising techniques of spectral analysis of nonstationary signals. Cohen's class of bilinear time-frequency signal representations has been defined [1] as

$$C_{x,y}(t, f, \Phi) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} e^{j\xi(\mu-\tau)} \Phi(\xi, \tau) \left[x\left(\mu + \frac{\tau}{2}\right) y^*\left(\mu - \frac{\tau}{2}\right) \right] e^{-j2\pi f\tau} d\xi d\mu d\tau \quad (1)$$

with $x(t)$, $y(t)$ - analyzed time signals, and $\Phi(\xi, \tau)$ - kernel of distribution. Denoting in (1) $R_{x,y}(t, \tau)$ as a local time-dependent cross-correlation function

$$R_{x,y}(t, \tau) = x\left(t + \frac{\tau}{2}\right) y^*\left(t - \frac{\tau}{2}\right) \quad (2)$$

and $w(t, \tau)$ as two-dimensional smoothing function, depending only on the distributions kernel

$$w(t, \tau) = \int_{-\infty}^{\infty} \Phi(\xi, \tau) e^{j\xi\tau} d\xi \quad (3)$$

and then setting smoothed local cross-correlation function $R_{x,y}^{AV}(t, \tau)$ as

$$R_{x,y}^{AV}(t, \tau) = \int_{-\infty}^{\infty} w(u-t, \tau) R_{x,y}(u, \tau) du \quad (4)$$

we obtain finally

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$$C_{x,y}(t, f, \Phi) = \frac{1}{2\pi} \int R_{x,y}^{AV}(t, \tau) e^{-j2\pi f\tau} d\tau \quad (5)$$

Bilinear joint time-frequency representation of Cohen's class can be then derived as a Fourier transform of smoothed local cross-correlation, defined in (4) as time-domain convolution of local cross-correlation $R_{x,y}$ with weighing function $w(t, \tau)$.

The general form of time-frequency representation for discrete-time band-limited signal and finite time of observation may be expressed as follows [2]:

$$R_{x,y}(n, k) = x(n+k) y^*(n-k), \quad (6)$$

$$R_{x,y}^{AV}(n, k) = \sum_{n1=n-(M-1)}^{n+M-1} w(n-n1, k) R_{x,y}(n, k), \quad (7)$$

$$C_{x,y}(n, f, \Phi) = 2 \sum_{k=-N+1}^{N-1} R_{x,y}^{AV}(n, k) e^{-j2\pi fk} \quad (8)$$

where $w(m, k)$ with $|k| \leq N-1$ and $|m| \leq M-1$ denotes discrete smoothing function, N is observation time period, M is smoothing order and n, f are time and frequency indexes respectively.

As was shown in [2] choosing of appropriate mask $w(m, k)$ results in computation of different form of time-frequency distribution. In this paper, the Smoothed Pseudo Wigner-Ville Distribution (SPWVD) was used, with mask

$$w(m, k) = g_M(m) |h_N(k)|^2 \quad (9)$$

where $g_M(m)$ denotes normalized symmetrical smoothing window ($M > 1$), $h_n(k)$ - time-domain window. The cross-JTFR is here replaced by auto-JTFR ($y(n)=x(n)$).

The main objective of the study was to determine directly, via pulsed photoacoustic spectroscopy, the deactivation of metastable states - dimers of Rhodamine 6G in a poly-vinyl-alcohol (PVA) matrix. In the studies, Rhodamine samples 20 μm thick were used. They were deposited on microscopy glasses, at two different concentrations: the lower one at 1×10^{-3} M (monomers only), and the higher one, 1×10^{-1} M, including both monomer and dimer forms.

The samples were excited by a Rhodamine 6G based dye laser using a pulse lasting 10 ns and 65 μJ energy. A photoacoustic chamber, applying microphone detection and digital signal recording, was used, including the application of synchronous analysis for improving the signal-to-noise ratio.

Due to their nonstationarity PAS signals should be analyzed using time-frequency signal representation and main goal of our work is here to determine precisely frequency variations in time domain. We expect a more exact signal description from time-frequency distribution, than in case of time or spectral signal representation only. We hope that the used analysis tool should then not only represent correctly variations of signal spectrum, but also preserve the instantaneous energy of signal.

Usually spectrograms are employed for this purpose, where spectral slices for segments of constant length moving stepwise over the signal are calculated. For every signal segment the magnitude spectrum

Fig. 1. The time and frequency PA response for a Rhodamine 6G in PVA at concentration of 1×10^{-3} M (monomers)

Fig. 2. As in Fig. 1 but at concentration of 1×10^{-1} M (monomers and dimers)

is computed involving FFT procedure. The resolution in frequency domain depends here on segment length. The techniques for frequency resolution increase (Zoom-FFT, chirp-Z) need stretching of signal observation time, which is not adequate to our goal.

The Wigner-Ville Distribution provides the most precise signal frequency analysis without decreasing the time domain resolution, exploring also new areas in time-frequency plane, invisible by means of spectrograms. The trade-off between time and frequency resolution, inherent in spectrogram analysis, doesn't hold for WVD. We have here free choice of analysis resolution to meet our needs most closely. The WVD shows not only significantly better resolution, but also satisfies boundary conditions i.e. its time average is equal to spectral energy density and frequency average is equal to the signal power. The WVD vanishes also for those time points, where signal vanishes.

During our experiments, in order to observe signal time fluctuation thoroughly, short Kaiser-type time window with 80 dB sidelobe attenuation and 33 samples length has been used. Window shift of 3 samples has been applied. Simultaneously, in order to keep frequency resolution sufficiently good, the FFT computes the spectrum for 512 frequency points. Smoothing window has been chosen also very short (3 points) in order to save the details during short impulse evaluation.

The time and frequency responses acquired as a result of WVD analysis are presented in Figs 1 and 2. In Fig. 1 at a concentration of 1×10^{-3} M, when almost only a monomer form of the pigment is present in the sample, one strong signal is observed. Its shape follows an exponential decrease dependence, and its width is related directly to the deactivation characteristic time. Fig. 2 presents analyses of the signal that originate from the sample (at 1×10^{-1} M), in which both monomer and dimer forms of Rhodamine 6G are present. The signal analysis of this system is a more complicated and the picture consists of several maxima of differentiated shapes. The maximum related to the monomer form is narrower which means a decrease of the deactivation time of this form. A decrease of the said time and the occurrence of other maxima point to the energy transfer from the monomer to dimer forms of the pigment, and reflect a longer time of deactivation of the dimer form [4].

The photoacoustic signal amplitude depends of course on the optical density of the samples, what was taken into account in calculations. The time and frequency responses acquired as a result of WVD analysis (Fig. 1,2) indicate that at the higher concentration, when a dimer form dominates, the energy is transferred between monomers and dimers. Further research will deal with the characteristic times of the thermal deactivation the two forms from WVD analyses.

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