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A method for improving wavelet threshold denoising in laser-induced breakdown spectroscopy

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ABSTRACT

The wavelet threshold denoising method is an effective noise suppression approach for noisy laser-induced breakdown spectroscopy signal. In this paper, firstly, the noise sources of LIBS system are summarized. Secondly, wavelet multi-resolution analysis and wavelet threshold denoising method are introduced briefly. As one of the major factors influencing the denoising results in the process of wavelet threshold denoising, the optimal decomposition level selection is studied. Based on the entropy analysis of noisy LIBS signal and noise, a method of choosing optimal decomposition level is presented. Thirdly, the performance of the proposed method is verified by analyzing some synthetic signals. Not only the denoising results of the synthetic signals are analyzed, but also the ultimate denoising capacity of the wavelet threshold denoising method with the optimal decomposition level is explored. Finally, the experimental data analysis implies that the fluctuation of the noisy LIBS signals can be decreased and the weak LIBS signals can be recovered. The optimal decomposition level is able to improve the performance of the denoising results obtained by wavelet threshold denoising with non-optimal wavelet functions. The signal to noise ratios of the elements are improved and the limit of detection values are reduced by more than 50% by using the proposed method.

1. Introduction

Laser-induced breakdown spectroscopy (LIBS) has been demonstrated its potential in elemental analysis of various samples [1,2]. LIBS offers significant advantages compared to other techniques, since LIBS has multi-element capability, low sample requirements and high speed of acquisition allowing real-time measurements [3]. The major goal of LIBS is the extraction of useful information, concerning the elemental qualitative or quantitative composition of samples, from deterministic and static signals [4]. The processes in LIBS, including ablation, atomization, excitation, ionization and recombination are quite complex and difficult to reproduce. As a result, each LIBS signal usually exhibits unpredictable variations, which leads to usually poor precision and sensitivity. The improvements in the precision and sensitivity of LIBS can be achieved by enhancing the signal strength or reducing the background, while an effective denoising method is often required in order to extract useful information from raw data [5,6].

In recent years, several techniques are available to minimize the noise and increase the signal to noise ratio (***SNR***). The hardware method such as the double pulse LIBS technique [7], the use of high voltage and fast discharge circuit [8], the configuration of optimum detection gating conditions and lasers energy [9,10], and the optimization of optical camera has been used to improve ***SNR*** [11]. The drawbacks of the hardware for ***SNR*** enhancement are poor portability, long design period and high cost [12,13]. Correspondingly, the software method is a flexible and convenient way for removing noise from contaminated spectroscopic signals. For example, the smoothing method is to create an approximating function that attempts to capture important patterns in the data, while leaving out noise. However, distortions in magnitude result in limited applicability [14]. Fourier transform filter denoising is accomplished by removing Fourier components with frequencies higher than a cutoff frequency. Fourier transform assumes the signal is stationary, but LIBS signal is always non-stationary [15]. Compared with traditional Fourier transform, wavelet transform is a time-frequency localization analysis method. For the excellent characters of wavelet analyses, the wavelet transform denoising has become an important tool to reduce the noise [16–18].

Currently there are threshold denoising method, coefficient correlation denoising method and modulus maxima denoising method in wavelet transform denoising [19–21]. In these methods above, the wavelet threshold denoising method (***WTD***) is the simplest in realization and it requires the least computational time. Thus ***WTD*** has been used widely. Yuan et al. applied ***WTD*** for LIBS signal denoising and the optimal decomposition level was determined by root mean square error of prediction [22]. Wiens et al. used ***WTD*** for removing noise and obtained the satisfying denoising results, but they gave no details about the optimal decomposition level selection method [23].

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Zhang et al. utilized ***WTD*** with double thresholds correction scheme for noise suppression, and the optimal decomposition level was obtained by the white noise testing method [24]. When the decomposition level (***DL***) is too small, the power compression of the useful signal is not obvious so that the result of noise reduction is not ideal; when ***DL*** is too excessive, the obvious power compression of the useful signal causes the loss of useful information and results in the decreased of the ***SNR*** and the increased of the mean square error (***MSE***). Therefore, it is necessary to determine the optimal ***DL***.

In order to select the optimal ***DL*** and effectively achieve better noise reduction of LIBS spectra, one objective is to put forward a scheme of choosing optimal ***DL***, and another objective is to analyze the noise suppression performance by ***WTD***. The proposed method of choosing optimal ***DL*** is based on the entropy analysis of the wavelet coefficients. In Section 2, the noise sources of LIBS system and ***WTD*** method are briefly introduced, and then the noise and the noisy LIBS signal are analyzed by using entropy. In Section 3, the method of choosing optimal ***DL*** based on entropy analysis is presented. In Section 4, the synthetic signals are analyzed to verify the performance of the proposed method, and then the ultimate denoising capacity of ***WTD*** with the optimal ***DL*** is studied. Section 5 gives a brief description of the LIBS apparatus and experiment parameters. Next, we verify the performance of the improved method by analyzing the observed LIBS signals, and further discuss its applicability in Section 6. Finally, this study is summarized.

2. Theory and method

In this section, we show a representation of a LIBS signal receiving system, indicating the various types of noise source which may arise. Next, we provide a brief overview of wavelet transform and some rigorous mathematical treatment on this issue can be found in the proper references provided along each of the parts within this section.

2.1. Summary of noise in LIBS system

As shown in Fig. 1(a), a typical LIBS signal receiving system has three main parts, namely optical system, detector system and electronic system. In the initial optical system, a fundamental noise source is photon shot noise, which results from the random nature of photon emission [25]. Photon shot noise is random, with a Poisson distribution of the number of detected photons [26]. Although some factors, such as the integration time and quantum efficiency of the detector, influence photon shot noise, its intrinsically random nature makes photon shot noise impossible to avoid. Noise in photodetector arises from contributions made by both the dark current and the flicker noise [27]. The ideal photodetector does not produce current in the absence of light. However, the random generation of electrons and holes in the depletion region of the photodetector is swept by the high electric field, and this effect is measured by dark current [28]. Flicker noise is the noise produced when a junction diode is operated at the onset of avalanche breakdown. Because of the attenuation of the flicker noise in the most of avalanche diodes, the flick noise would be negligible [29]. Johnson noise (thermal noise) is also unavoidable, and generated by the random thermal motion of charge carriers (usually electrons), inside an electrical conductor, which happens regardless of any applied voltage [30]. Thermal noise is approximately white noise, meaning that its power spectral density is nearly equal throughout the frequency spectrum. The amplitude of the signal has very nearly a Gaussian probability density function.

The useful LIBS signal can be defined as the amount of light incident upon the detector per unit time, and the noise can be seen as the "disturbance" on the useful LIBS signal that hinders an accurate measurement. Examining the sources of noise in LIBS system and understanding which contributions have the greatest impact on the uncertainty of LIBS system measurement are useful to optimize the quality of results. Fig. 1(b) shows the noise sources of a noisy LIBS signal. According to the different mechanism of noises, the noise suppression approaches have their respective features. Photon shot noise is an inherent noise and can be reduced by increasing the light intensity or the exposure time [31]; dark current noise can be minimized by cooling the photodetector [32]; Johnson noise can be removed by electrical hardware design [33]. These methods mentioned above are inconvenient in practical applications, so it is necessary to study simple and effective ways for LIBS noise suppression.

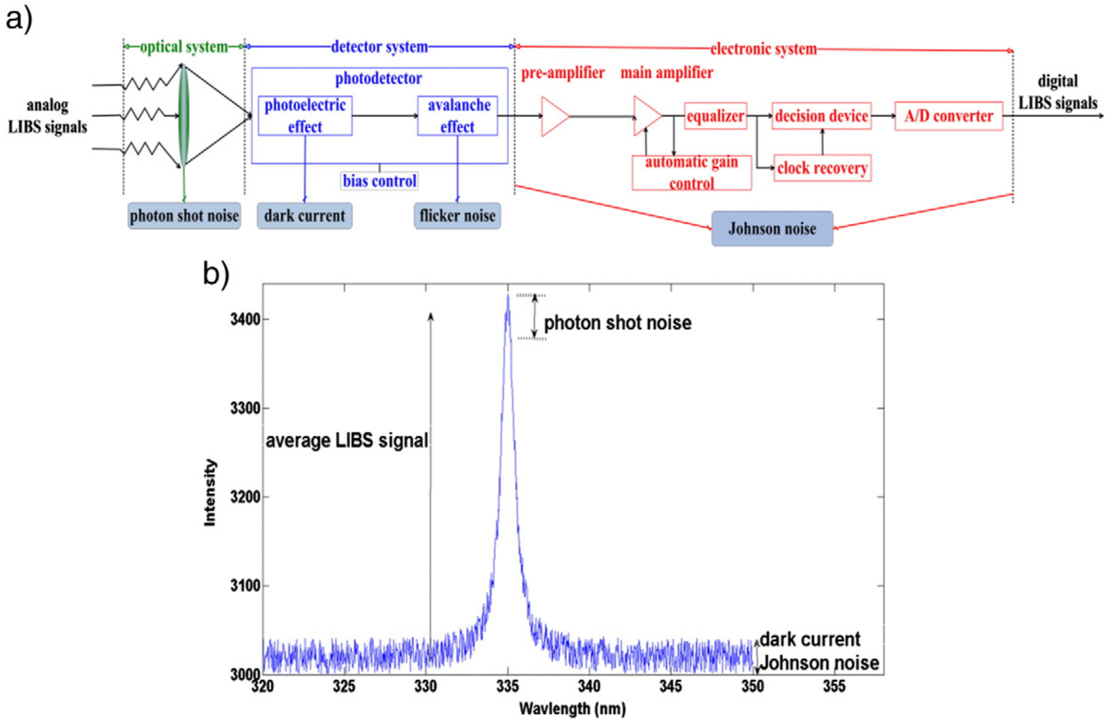

Fig. 1. (a) The noise classification in a LIBS signal receiving system; (b) the noise sources of a noisy LIBS signal.