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Assessment of an MCMC algorithm convergence for Bayesian estimation of the particle size distribution from multiangle dynamic light scattering measurements

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Abstract. Recovering the particle size distribution (PSD) from dynamic light scattering (DLS) measurements is known to be a highly ill-posed inverse problem. In a former study, we proposed a new Bayesian inference method applied directly to the multiangle DLS measurements to improve the estimation of multimodal PSDs. The *posterior* probability density of interest is sampled using a MCMC Metropolis-within-Gibbs algorithm. In this work, we experimentally examined the convergence of the used MCMC strategy using the simulation method recently proposed by Chauveau and Vandekerkhove (2013). This method is based on the evolution in time (iterations) of the Kullback-Leibler divergence between the target *posterior* density and the successive densities of the algorithm of interest. The convergence of the used MCMC algorithm was examined when processing simulated and experimental data.

Keywords: Particle Size Distribution, Dynamic Light Scattering, Inverse Problem, Bayesian Inference, MCMC algorithm, Entropy, Kullback divergence.
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INTRODUCTION

Particle size distribution (PSD) is an important physical characteristic of several particulate systems such as dry powders, colloidal suspensions, aerosols and emulsions. These systems are prevalent in many fields in industry and academia. Many physical parameters of particulate systems are highly size-dependent, for example, stability of emulsions and suspensions, abrasiveness of dry powders, color and finish of colloidal paints and paper coatings are all size-dependent [1].

Dynamic Light Scattering (DLS) is one of the most popular techniques used for measuring the PSD of a sample composed of particles dispersed in a liquid. The particle sizes can range from a few nanometers to several micrometers [2]. A typical DLS experiment consists in illuminating particles dispersed in a liquid by using a narrow monochromatic laser beam. At a given scattering angle θ, the intensity of scattered light is measured by a photon detector. The scattered intensity fluctuates in time due to the random Brownian motion of the dispersed particles. Analysis of these fluctuations yields information about the particle size. The analysis is done by using the time autocorrelation function (ACF) of the measured intensity signal. This time ACF is computed using a digital correlator.

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The PSD is retrieved by inverting the normalized electric field time ACF, $g_\theta^{(1)}(\tau)$. $g_\theta^{(1)}(\tau)$ is related to the measured normalized intensity time ACF, $g_\theta^{(2)}(\tau)$, by [2]

$$g_\theta^{(2)}(\tau) = 1 + \beta |g_\theta^{(1)}(\tau)|^2, \tag{1}$$

where τ is the time delay and $\beta(< 1)$ is an instrumental factor.

For a polydisperse sample i.e. particles with different sizes, the electric field time ACF, $g_\theta^{(1)}(\tau)$, is related to the number-weighted PSD, $f(D)$, by [1]

$$g_\theta^{(1)}(\tau) = \frac{1}{I_\theta} \int_0^\infty f(D) C_I(\theta, D) \exp(-\frac{\Gamma_{0,\theta}}{D}\tau) dD, \tag{2}$$

where I_θ is a proportionality constant ensuring $g_\theta^{(1)}(0) = 1$. $C_I(\theta, D)$ represents the fraction of light intensity scattered at θ by a particle of hydrodynamic diameter D. $C_I(\theta, D)$ is calculated through the Mie theory [3]. $\Gamma_{0,\theta} = \frac{16\pi n^2 \sin^2(\theta/2) k_B T}{3\lambda_0^2 \eta}$ with k_B, T and λ_0 are the Boltzmann constant, the absolute temperature and the wavelength of laser light in vacuum, respectively. n is the medium refractive index and η is the viscosity.

Retrieving the PSD from DLS measurements (time ACF) involves inversion of the integral equation 2. This problem is known to be an ill-posed inverse problem, since the solution may not be the one that provides the best fit to the data. Moreover a small amount of noise present in data can lead to large variations in the estimated PSD.

Several analysis methods have been proposed to retrieve the PSD such as the method of cumulants [4], non-negative least squares [5], regularized non-negative least squares [6] (CONTIN is the most widely used routine), maximum entropy [7] and truncated singular value decomposition [8] as well as stochastic optimization methods (neural network [9], particle swarm optimization [10]). In general, these methods work well for monomodal PSDs. However, they are very noise sensitive and the reproducibility of the results is not ensured. Their main drawback is the poor capacity to discriminate peaks of multimodal PSDs. The best results are obtained for populations with comparable intensity contributions and spaced at least by a factor 2 in diameter. Multiangle DLS (MDLS), which consists in processing the whole DLS data acquired at different angles, is a promising method to overcome these problems. Former studies have demonstrated that MDLS provides more robust, reproducible and accurate PSD estimate than single-angle DLS, particularly for polydisperse and/or multimodal samples [11, 12, 13].

Methods based on Bayesian inference have been investigated in [14, 15, 16, 17] to solve the DLS inverse problem. In [17], we have proposed a new Bayesian inference method applied directly to the MDLS measurements to improve multimodal PSDs estimation. The derived *posterior* probability density function (pdf) is simulated using an MCMC Metropolis-within-Gibbs algorithm. The results show an improvement in the estimation of multimodal PSDs compared to the method proposed in [16].

Since the used MCMC algorithm is associated with unknown rate of convergence, the aim of the present work is to assess the convergence of the MCMC strategy used in [17]. To this end, we propose to use the simulation method recently proposed by the authors of [18] and which helps to assess the MCMC algorithms efficiency. This simulation method is based on the evolution in time (iterations) of the Kullback divergence between the target *posterior* density and the algorithm successive densities. The Kullback divergence estimation requires an estimate of the entropy of the algorithm successive densities.

The paper is organized as follow. Section 2 recalls the inversion method proposed in [17]. In section 3, we give a brief description of the used simulation method to assess the MCMC algorithm convergence. Results obtained from simulated and experimental data are presented in section 4. Finally, conclusions are drawn in section 5.

PROPOSED BAYESIAN INVERSION METHOD

In [17], a multiangle DLS analysis method is proposed to estimate the PSD from the measured intensity ACFs. MDLS data are acquired at different angles $\{\theta_r,\ r=1,\dots,R\}$. For each angle θ_r, the intensity ACF is measured for different values of the time delay, $\{\tau_m,\ m=1,\dots,M_r\}$ with M_r is the total number of points at θ_r. An additive noise model is proposed to model the measured intensity ACFs, $\tilde{g}^{(2)}_{\theta_r}(\tau_m)$, as follow

$$\begin{aligned}
\tilde{g}^{(2)}_{\theta_1}(\tau_m) &= g^{(2)}_{\theta_1}(\tau_m)+w_1(j), \quad m=1,\cdots,M_1 \ , \\
&\vdots \\
\tilde{g}^{(2)}_{\theta_r}(\tau_m) &= g^{(2)}_{\theta_r}(\tau_m)+w_r(j), \quad m=1,\cdots,M_r \ , \\
&\vdots \\
\tilde{g}^{(2)}_{\theta_R}(\tau_m) &= g^{(2)}_{\theta_R}(\tau_m)+w_R(j), \quad m=1,\cdots,M_R \ ,
\end{aligned} \tag{3}$$

where $g^{(2)}_{\theta_r}(\tau_m)$ is the noise-free intensity ACF at θ_r. The white noises $w_r(m)$ are assumed to be independent, normally distributed with zero-mean and variances σ_r^2 at the angle θ_r.

The PSD is estimated for a discrete set of diameters $\{D_i,\ i=1,\dots,N\}$ from a fixed size range, associated with $\{f(D_i), i=1,\dots,N\}$. After digitizing (2) inserted into (1), the noise-free intensity ACF $g^{(2)}_{\theta_r}(\tau_m)$ is related to the PSD $\{f(D_i), i=1,\dots,N\}$ by

$$g^{(2)}_{\theta_r}(\tau_m) = 1+\beta_r\left(\frac{1}{I_{\theta_r}}\sum_{i=1}^{N} f(D_i)C_I(\theta_r,D_i)\exp\left(-\frac{\Gamma_{0,\theta_r}}{D}\tau_m\right)\Delta D_i\right)^2 \tag{4}$$

Let us consider the vector $\tilde{\mathbf{g}}^{(2)} = \left[\tilde{\mathbf{g}}_1^{(2)^T},\dots,\tilde{\mathbf{g}}_R^{(2)^T}\right]^T$ where $\tilde{\mathbf{g}}_r^{(2)} = [\tilde{g}^{(2)}_{\theta_r}(\tau_1),\dots,\tilde{g}^{(2)}_{\theta_r}(\tau_{M_r})]^T$ for $r=1,\dots,R$. Let us also define the vector $\mathbf{f}=[f(D_1),\dots,f(D_N)]^T$.

A Bayesian inference method is proposed to estimate the PSD $\mathbf{f}$ from the measured intensity ACFs $\tilde{\mathbf{g}}^{(2)}$. By assuming the independence between the angular measurements and taking into account the assumption of independent white Gaussian noise, the joint *posterior* pdf of variables $\mathbf{f}$ and σ_r^2 can be written using the Bayes theorem as follow

$$p\left(\mathbf{f},\sigma_1^2,\dots,\sigma_R^2|\tilde{\mathbf{g}}^{(2)}\right) = \frac{p(\mathbf{f})\prod_{r=1}^{R} p\left(\tilde{\mathbf{g}}_r^{(2)}|\mathbf{f},\sigma_r^2\right)p(\sigma_r^2)}{p\left(\tilde{\mathbf{g}}^{(2)}\right)}, \tag{5}$$

where $p(\mathbf{f})$ is the *prior* pdf that expresses *prior* information about the PSD, $p(\sigma_r^2)$ is the *prior* pdf of the noise variance at θ_r and $p\left(\tilde{\mathbf{g}}^{(2)}\right)$ is a normalizing constant. The likelihood function at the angle θ_r, $p\left(\tilde{\mathbf{g}}_r^{(2)}|\mathbf{f},\sigma_r^2\right)$, is given by

$$p(\tilde{\mathbf{g}}_r^{(2)}|\mathbf{f},\sigma_r^2) = \frac{1}{(2\pi)^{\frac{M_r}{2}}\sigma_r^{M_r}}\exp(-\frac{\chi_r(\mathbf{f})}{2\sigma_r^2}), \tag{6}$$

with $\chi_r(\mathbf{f}) = \sum_{m=1}^{M_r}(\tilde{\mathrm{g}}_{\theta_r}^{(2)}(\tau_m) - \mathrm{g}_{\theta_r,\mathbf{f}}^{(2)}(\tau_m))^2$.

The used *prior* pdfs are the non-negativity and smoothness *prior* for the PSD according to the physical nature of the problem and the Jeffrey's *prior* for the noise variances. After marginalization, we get the following *posterior* pdf of interest

$$p(\mathbf{f}|\tilde{\mathbf{g}}^{(2)}) \propto \begin{cases} \exp\left(-\|\mathbf{L}_2\mathbf{f}\|_2^2\right)\prod_{r=1}^{R}[\chi_r(\mathbf{f})]^{-\frac{M_r}{2}}, & \text{if } \mathbf{f}\geq 0 \\ 0, & otherwise, \end{cases} \tag{7}$$

where the square matrix $\mathbf{L}_2$ represents the discrete operator of the second derivative.

The derived *posterior* pdf (7) is highly multivariate and known up to a multiplicative constant. A random walk Markov chain Monte Carlo (MCMC) Metropolis-within-Gibbs algorithm [19] is used in order to sample this *posterior* pdf. The used proposal distribution for each element of $\mathbf{f}$ is selected as a Gaussian distribution with zero mean and standard deviation tuned to have an acceptance rate close to 50% [20].

To make inference about the PSD from the generated Markov chain $\{\mathbf{f}^{(L_0)},\cdots,\mathbf{f}^{(L)}\}$, the Minimum Mean Square Error (MMSE) estimator is used. The estimated PSD is given by

$$\hat{\mathbf{f}} = \frac{1}{L-L_0+1}\sum_{l=L_0}^{L}\mathbf{f}^{(l)}, \tag{8}$$

where L_0 is the burn-in period and L is the length of the generated chain.

Some issues not discussed in [17] are: does the stationary distribution of the generated Markov chain converge to the target *posterior* distribution?, and how to choose the parameters L_0 and L?. In the next section we give a brief description of a simulation method that helps to assess MCMC algorithms efficiency. This method id only based on Monte Carlo simulations.

SIMULATION METHOD FOR MCMC EVALUATION

The simulation-based method to estimate MCMC efficiency proposed in [18] is grounded on the evolution of the Kullback-Leiber divergence between the target *posterior* pdf ,$p(\mathbf{f}|\tilde{\mathbf{g}}^{(2)})$, and the marginal density of the MCMC algorithm, p^t, at time (iteration) t. The choice of this criterion is justified by the fact that the Kullback distance is a natural measure of the algorithm quality and has strong connections with ergodicity of Markov chains and rates of convergence. The Kullback divergence is defined by

$$\kappa(p^t, p(\mathbf{f}|\tilde{\mathbf{g}}^{(2)})) = \mathscr{H}(p^t) - \int p^t \log\left(p(\mathbf{f}|\tilde{\mathbf{g}}^{(2)})\right), \tag{9}$$

where $\mathscr{H}(p^t) = E_{p^t}[\log p^t]$ is the entropy of the MCMC marginal density, p^t, at time t.

The estimation of the Kullback divergence requires an estimate of the entropy of the algorithm successive densities. In this method, the entropy estimation is based on the simulation of K parallel (iid) Markov chains $(\mathbf{f}_k^0, \cdots, \mathbf{f}_k^t \sim p^t, \cdots)$ for $k = 1, \ldots, K$, started from a diffuse initial distribution p^0. At time t, $(\mathbf{f}_1^t, \cdots, \mathbf{f}_K^t)$ forms a K-samples iid $\sim p^t$. Based on the sample $(\mathbf{f}_1^t, \cdots, \mathbf{f}_K^t)$, they used the nearest neighbor entropy estimate [18] defined by

$$\hat{\mathscr{H}_K}(p^t) = \frac{1}{K}\sum_{k=1}^{K} \log(\rho_k^N) + \log(K-1) + \log\left(\frac{\pi^{N/2}}{\Gamma(N/2+1)}\right) + 0.5772, \tag{10}$$

where $\rho_k = \min\{d(\mathbf{f}_k^t, \mathbf{f}_l^t), l \in \{1, \cdots, K\}, l \neq k\}$ is the Euclidean distance from the kth point to its nearest neighbor in the sample $(\mathbf{f}_1^t, \cdots, \mathbf{f}_k^t, \cdots, \mathbf{f}_K^t)$. N is the problem dimension (total number of the PSD points).

An estimation of $\int p^t log\left(p(\mathbf{f}|\tilde{\mathbf{g}}^{(2)})\right)$ is given by a Monte Carlo integration

$$E_{\hat{p}_K^t}[\log\left(p(\mathbf{f}|\tilde{\mathbf{g}}^{(2)})\right)] = \frac{1}{K}\sum_{k=1}^{K} log\left(p(\mathbf{f}_k^t|\tilde{\mathbf{g}}^{(2)})\right). \tag{11}$$

For a converging MCMC algorithm, the sequence of marginals p^t theoretically satisfies $\kappa(p^t, p(\mathbf{f}|\tilde{\mathbf{g}}^{(2)})) \to 0$ as $t \to \infty$. However, since the *posterior* pdf of interest is known up to a multiplicative constant, $p(\mathbf{f}|\tilde{\mathbf{g}}^{(2)}) = Cst\,\phi(\mathbf{f}|\tilde{\mathbf{g}}^{(2)})$, where

$$\phi(\mathbf{f}|\tilde{\mathbf{g}}^{(2)}) = \begin{cases} \exp\left(-\|\mathbf{L}_2\mathbf{f}\|_2^2\right) \prod_{r=1}^{R} [\chi_r(\mathbf{f})]^{-\frac{M_r}{2}}, & \text{if } \mathbf{f} \geq 0 \\ 0, & otherwise, \end{cases} \tag{12}$$

the only quantity that can be estimated is $\kappa(p^t, \phi(\mathbf{f}|\tilde{\mathbf{g}}^{(2)}))$. For a converging MCMC, $\hat{\kappa}_K(p^t, \phi(\mathbf{f}|\tilde{\mathbf{g}}^{(2)})) \to \log(Cst) + bias_K(p^t, N)$ as $t \to \infty$ where $bias_K(p^t, N)$ is the bias of the entropy estimation. Thus, without knowing the normalization constant and even if the Kullback divergence stabilizes in time no conclusion about the convergence can be drawn. To handle this problem, the authors of [18] suggest to manipulate the difference of the Kullback divergences of the studied MCMC algorithm and a benchmark MCMC with marginals known to converge. The difference of the divergences stabilizes near 0 if the studied MCMC converges. In the absence of the benchmark MCMC, we propose to take into account the divergence stabilization criterion and the accuracy of the PSD estimation to judge the MCMC convergence.

RESULTS ON SIMULATED AND EXPERIMENTAL MDLS DATA

In this section, we present some obtained results of the PSD estimation and the evolution in time of the Kullback divergence $\hat{\kappa}_K(p^t,\phi(\mathbf{f}|\tilde{\mathbf{g}}^{(2)}))$ for simulated and experimental data. The studied samples are spherical latex particles with refractive index 1.59, dispersed in pure water (refractive index 1.33 and viscosity $\eta = 0.89$ mPa.s). We used a vertically-polarized laser of wavelength $\lambda_0 = 638$ nm. The temperature was stabilized at 298.15 K. For each example presented in this section, 500 iid Markov chains are generated for the estimation of the Kullback divergence.

The inversion method [17] is first tested on a simulated example of monomodal PSD (example 1). The studied PSD is Gaussian with a mean diameter of 500 nm and a standard deviation of 10 nm. The noise-free time ACFs $g^{(2)}_{\theta_r}(\tau_m)$ were simulated from the corresponding PSD using (4) ($\Delta D = 1$ nm) for the scattering angles 60°, 90°, 120° and 150°. The noisy time ACFs $\tilde{g}^{(2)}_{\theta_r}(\tau_m)$ were simulated by adding a white Gaussian noise (3) with $\sigma_r = 0.001$ for all the scattering angles. For the estimation procedure, a set of discrete diameters is fixed in the interval $[400, 600]$ nm with a regular step of $\Delta D = 10$ nm ($N = 21$). The true PSD compared to the estimated one by the Bayesian method [17] are presented on the Figure 1(a). The estimated PSD is very close to the true PSD. The result of the estimation of the Kullback divergence, $\hat{\kappa}_K(p^t,\phi(\mathbf{f}|\tilde{\mathbf{g}}^{(2)}))$, is shown in Figure 1(b). The Kullback divergence stabilizes quickly after $t = 1000$ iterations.

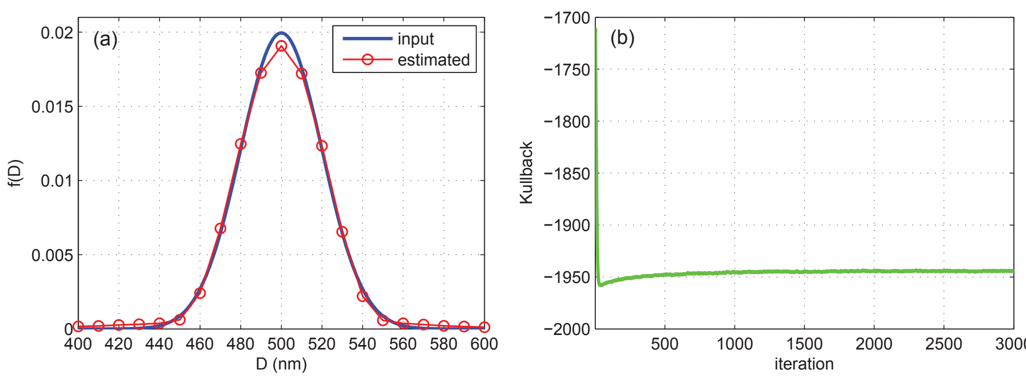

FIGURE 1. Simulated monomodal PSD (example 1). (a) The true PSD compared to the estimated one by the inversion method [17]. (b) Evolution of the Kullback divergence $\hat{\kappa}_K(p^t,\phi(\mathbf{f}|\tilde{\mathbf{g}}^{(2)}))$. $N = 21$ and $K = 500$.

The inversion method is also tested on a simulated example of multimodal PSD (example 2). The simulated PSD is a bimodal distribution sum of two Johnson's functions of equal quotients and expressed as follow

$$f(D) = \sum_{i=1}^{2} \frac{0.5\sigma_i}{\sqrt{2\pi}(D_{max} - D_{min})} \exp(-0.5[u_i + \sigma_i \log(\frac{D - D_{min}}{D_{max} - D})]^2), \quad (13)$$

with $u_1 = 3.4$, $\sigma_1 = 2.1$, u_2=-2.4, $\sigma_2 = 2.0$, $D_{min} = 200$ nm and $D_{max} = 900$ nm. The noisy time ACFs $\tilde{g}^{(2)}_{\theta_r}(\tau_m)$ were simulated for the scattering angles 60°, 70°, 80°, 90°, 100°, 120° and 140° following the same procedure as the previous example. The discrete diameters are fixed in the selected range $[100, 1000]$ nm with a regular step of $\Delta D = 10$ nm ($N = 91$). The true PSD compared to the estimated one by the proposed method in [17] are shown in Figure 2(a). The proposed Bayesian method successfully estimated the full PSD with separation of the two peaks of the distribution. Figure 2(b) shows the estimation results of the Kullback divergence $\hat{\kappa}_K(p^t, \phi(\mathbf{f}|\tilde{\mathbf{g}}^{(2)}))$. As it can be noticed, the Kullback divergence stabilizes after $t = 5000$ iterations.

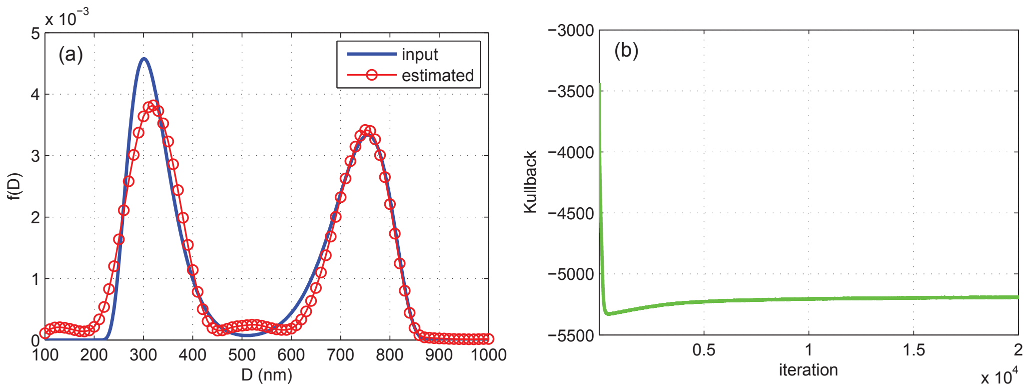

FIGURE 2. Simulated bimodal PSD (example 2). (a) The true PSD compared to the estimated one by the inversion method [17]. (b) Evolution of the Kullback divergence $\hat{\kappa}_K(p^t, \phi(\mathbf{f}|\tilde{\mathbf{g}}^{(2)}))$. $N = 91$ and $K = 500$.

The experimental MDLS data were acquired using the Nano DS equipment from CILAS. The studied sample was a trimodal mixture of polysterene latex spheres represented by a combination of 3 distributions with nominal diameters (standard deviations) are 400 (7) nm, 600 (10) nm and 1020 (10) nm respectively. The intensity ACFs were acquired at 13 angles between 60° and 120°, with a step of 5°. The estimated PSD is shown on Figure 3(a). The result shows that the three populations are well recovered and the peaks of the estimated PSD seem to be close to the expected ones. The result of the estimation of the Kullback divergence, $\hat{\kappa}_K(p^t, \phi(\mathbf{f}|\tilde{\mathbf{g}}^{(2)}))$, is shown in Figure 3(b). The Kullback divergence stabilizes very quickly after $t = 1000$ iterations.

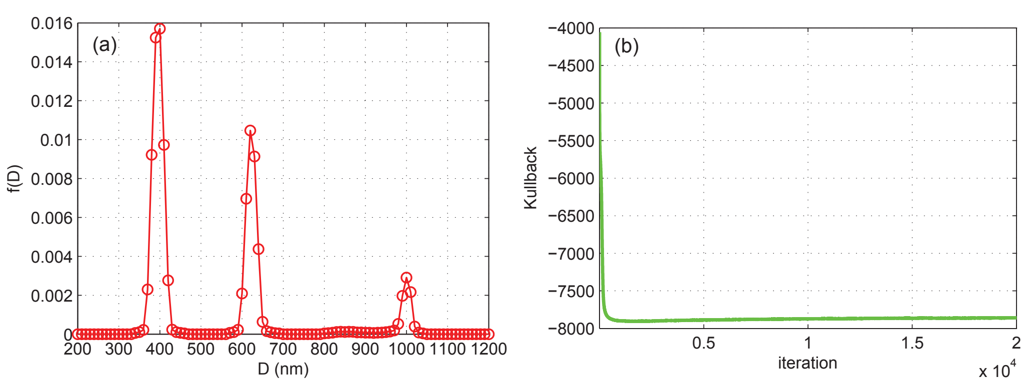

FIGURE 3. Experimental trimodal PSD. (a) Estimated PSD by the inversion method [17]. (b) Evolution of the Kullback divergence $\hat{\kappa}_K(p^t, \phi(\mathbf{f}|\tilde{\mathbf{g}}^{(2)}))$. $N = 101$ and $K = 500$.

CONCLUSION

The convergence of the MCMC algorithm used in [17] for the PSD inversion from multiangle DLS data is examined. The used criterion is the stabilization of the Kullback divergence, between the target *posterior* density and the MCMC algorithm successive densities, supported by the accuracy of the PSD estimation. According to the good PSD estimation results obtained for simulated and experimental data, and the fast stabilization of the Kullback divergence, we can experimentally conclude that the used MCMC algorithm is converging.

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