Method of measurements with random perturbation: Application in photoemission experiments

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We report on an application of a simultaneous perturbation stochastic approximation (SPSA) algorithm to filtering systematic noise (SN) with nonzero mean value in photoemission data. In our analysis, we have used a series of 50 single-scan photoemission spectra of W(110) surface where different SNs were added. It was found that the SPSA-evaluated spectrum is in good agreement with the spectrum measured without SN. On the basis of our results, a wide application of SPSA algorithm for evaluation of experimental data is anticipated. © *2008 American Institute of Physics.* [DOI: 10.1063/1.2890515]

Very often, measurements of physical quantities are considerably hindered by observation with noises of unknown nature. This problem appears in many industrial as well as scientific applications, such as pattern recognition, product quality improvement, control of heavy ion beams, probing of electronic structure of solids and molecules by spectroscopic techniques, etc. Traditionally, experimental noises of different origins are assumed to be independent from each other and characterized by zero mean values. These assumptions are frequently hard to justify. However, without them, the validity of many algorithms is questionable in applications. For example, it is known that the standard "least-squares method" or the "maximum likelihood method"[1] give wrong estimates if the noise in the registration channel has an "unknown-but-bounded" deterministic nature or it is a "dependent" sequence from the probabilistic point of view.

Let us consider photoemission (PE) measurement[2,3] where systematic noise (SN) of unknown origin appears at some particular kinetic energies of photoelectrons. Such noise may introduce additional spectral features that cannot be expected from electronic structure of sample. If this noise is not zero mean, it cannot be eliminated by simple increase of number of scans in PE experiment. One of the effective ways to deal with such noise is of using new mathematic algorithms.

Mathematical algorithms for search and optimization play a severe role in finding best options to solve many problems not only in physics but also in engineering, business, medicine, as well as in other natural and social sciences. In the case of prior information ambiguity, recursive algorithms are most effective among many other approaches. Such algorithms start with an initial "guess" of a solution, and this assessment is updated on an iteration basis with the purpose of improving the measured (observable) objective function of sample.

One particular optimization algorithm that has attracted considerable international attention in the recent past is the simultaneous perturbation stochastic approximation (SPSA) method (see, for review, Refs. 4 and 5). Likewise, simulated annealing or genetic algorithms, SPSA uses only objective function measurements, that is of decisive importance in experiments where direct probing of the gradient of the objective function is rather difficult or not possible at all. In difference to other methods, SPSA is especially efficient for high-dimensional problems in terms of providing a good solution for a relatively small number of measurements of the objective function. Recently, this algorithm was successfully applied to a number of tasks such as queuing systems[6] or control of heavy ion beams.[7]

In the present study, we demonstrate, for the first time, application of the SPSA algorithm for analysis of PE spectra, which include a noise with nonzero mean value. PE spectra of W(110) surface were collected upon excitation with two *independent* photon sources: the first one was used to probe the electronic structure of W(110) and second one was used as a source of the noise. It was shown that the spectrum obtained after application of the SPSA algorithm to a series of 50 PE single-scan spectra is in good agreement with the spectrum measured without SN. On the basis of these results, we conclude that exploiting of SPSA can be useful for analysis of experimental spectroscopic data. We can expect a wide application of this method for filtering systematic noises that can appear in many kinds of measurements or experiments.

Photoemission spectra[2,3] were taken from a W(110) single crystal kept at room temperature. Experiments were performed in the setup based on the hemispherical energy analyzer (SPECS PHOIBOS 150).[8] The overall-system energy resolution accounting for the thermal broadening was set to 150 meV and electrons were collected in angle-integrated mode around the surface normal. The base pressure was in the range of 1×10^{-10} mbar. Prior to experiment, the W(110) crystal was carefully cleaned by repeated cycles of heating up to 1300 °C in oxygen ambient pressure of 5×10^{-8} mbar for 15 min each and subsequent flashing up to 2300 °C. After such procedure, the crystal was kept in vacuum for 24 h in order to passivate the surface of the crystal via absorption of residual gases in the experimental chamber. This step was necessary in order to stabilize the

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crystal surface in the long-term surface-sensitive PE experiment.

In order to generate studied PE signal (objective function), we have used the He IIα resonance line ($h\nu$ =40.8 eV) as an excitation light source. The photocurrent emitted in this process can be expressed as

$$j(E_{\mathrm{kin}}) = I \times \mathrm{DOS}(E_{\mathrm{kin}}) \times d\sigma(E_{\mathrm{kin}})/d\Omega, \quad (1)$$

where I is the intensity of the light source (He IIα), DOS denotes the electronic density of states of the W(110) surface, $d\sigma(E_{\mathrm{kin}})/d\Omega$ is the cross section of the photoemission process, $E_{\mathrm{kin}}=h\nu-W-E_B$ stands for the kinetic energy of the photoelectron (W is the work function of the material, E_B is the binding energy of the electron in the solid). Since $d\sigma(E_{\mathrm{kin}})/d\Omega$ is practically constant in the small energy range, the total photocurrent can be written as

$$j(E_{\mathrm{kin}}) \approx I \times \mathrm{const} \times \mathrm{DOS}(E_{\mathrm{kin}}). \quad (2)$$

It is proportional to the density of states and to the intensity of incoming radiation.

Usually, the task of PE experiment is to derive the dependence

$$\bar{\theta}(E_{\mathrm{kin}}) = \mathrm{const} \times \mathrm{DOS}(E_{\mathrm{kin}}) \approx j(E_{\mathrm{kin}})/I$$

from the measured photocurrent as a function of E_{kin}.

In a large variety of PE experiments,[2,3] this aim can be achieved by smoothing of experimental spectra. As a rule, several kinetic energy scans (e.g., k scans) are performed. From one scan to another, the probed parameter $\theta(E_{\mathrm{kin}})$ may vary around its mean value $\bar{\theta}(E_{\mathrm{kin}})$ in accordance with the distribution function. These parameter variations may be caused by even slightly changed experimental conditions (e.g., temperature drift, etc.).

$$\theta_n(E_{\mathrm{kin}}) = \bar{\theta}(E_{\mathrm{kin}}) + w_n(E_{\mathrm{kin}}), \quad n = 1,2,\ldots,k,$$

where $w_n(E_{\mathrm{kin}})$ is a stochastic perturbation with a zero mean value.

If we assume that the light intensity may change in a controlled way being $I_1, I_2, \ldots, I_k$ for scans numbers $n=1,2,\ldots,k$, respectively, Eq. (2) can be rewritten as

$$j_n(E_{\mathrm{kin}}) = I_n \theta_n(E_{\mathrm{kin}}), \quad n = 1,2,\ldots,k. \quad (3)$$

If $I=I_1=I_2=\cdots=I_k$, the typical approach to determine the value $\bar{\theta}(E_{\mathrm{kin}})$ is averaging the experimental data

$$\bar{\theta}(E_{\mathrm{kin}}) \approx \frac{1}{kI}\sum_{n=1}^{k} j_n(E_{\mathrm{kin}}).$$

In the general case, least-squares analysis can be applied

$$\bar{\theta}(E_{\mathrm{kin}}) \approx \frac{\Sigma_{n=1}^{k} j_n(E_{\mathrm{kin}}) I_n}{\Sigma_{n=1}^{k} I_n^2}. \quad (4)$$

Given that the systematic noises cannot be abolished, unknown experimental systematic errors $N_n(E_{\mathrm{kin}})$ should be accounted for in Eq. (2),

$$j_n(E_{\mathrm{kin}}) = I_n \theta_n(E_{\mathrm{kin}}) + N_n(E_{\mathrm{kin}}), \quad n = 1,2,\ldots,k. \quad (5)$$

In case that $N_n(E_{\mathrm{kin}})$ is SN with a nonzero mean value, use of the traditional least-squares technique (4) leads to the biased (shifted) estimated parameter value

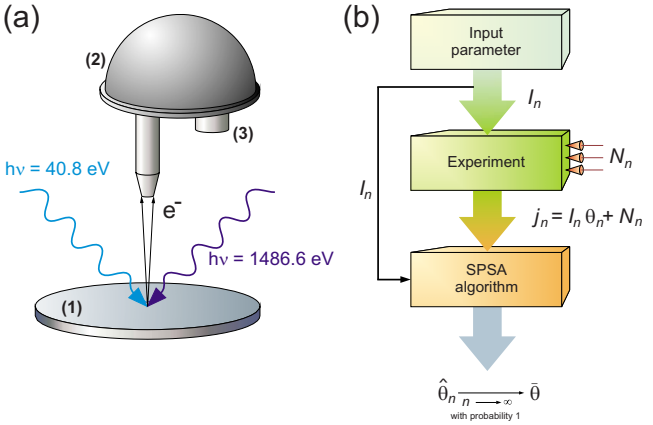

FIG. 1. (Color online) (a) Scheme of the PE experiment, where sample (1) is illuminated by two light sources, He IIα and Al $K\alpha$, electrons are analyzed by photoelectron spectrometer (2) and detector (3) registers the photocurrent signal $j_n(E_{\mathrm{kin}})$. (b) Layout of the present study using the SPSA algorithm for eliminating noises of unknown nature.

$$\bar{\theta}(E_{\mathrm{kin}}) + \frac{\Sigma_{n=1}^{k} N_n(E_{\mathrm{kin}}) I_n}{\Sigma_{n=1}^{k} I_n^2}.$$

Therefore, a very high degree of experimental precautions is required to screen systematic noises that may appear upon acquisition of the experimental data.

In order to justify the application of our novel technique of data evaluation that allows filtering of systematic noises with nonzero mean values, the described above PE experimental conditions were reconsidered. In addition to the He IIα excharge lamp, generating the studied PE signal, a radiation from a Röntgen tube (Al $K\alpha$, $h\nu$=1486.6 eV) was used for excitation of secondary photoelectrons, those E_{kin} dependence of photocurrent does not reflect the DOS(E_{kin}) of the W(110) single crystal.

The layout of the experiment is shown in Fig. 1(a). The valence-band PE spectra excited with the He IIα radiation

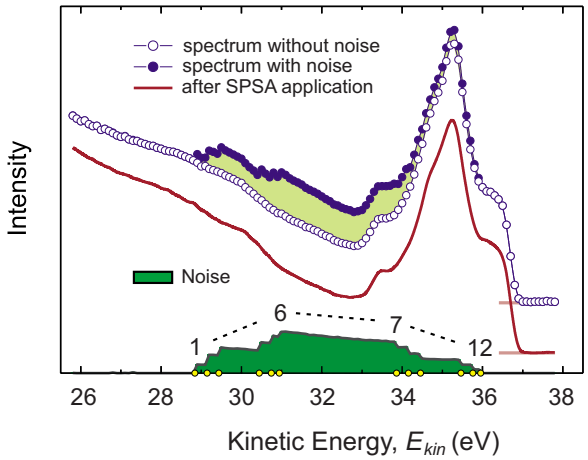

FIG. 2. (Color online) Experimental PE spectra of the valence band of W(110) obtained with He IIα radiation without (open circles) and with (filled circles) systematic noise. Spectrum obtained after application of the SPSA algorithm to a series of 50 experimental single-scan spectra is shown by thick line. The shaded area in the bottom is systematic noise measured separately. 12 control points used for demonstration of the convergence dynamic of the algorithm are marked with labels (1)–(12) on the kinetic energy axis.

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were collected in the range of 25.8–37.8 eV kinetic energy of emitted photoelectrons (Fig. 2, open circles).

For acquisition of each individual single-scan PE spectrum ($n=1,2,\ldots,50$), intensity of radiation (I_n) was randomly selected and then measured by a special detector.

In every single-scan experiment, the systematic noise $N(E_{\mathrm{kin}})$ was introduced by switching on the x-ray source when measuring in the range of 28.8–35.9 eV kinetic energies (shaded area in the bottom of Fig. 2). In such way, generated noise represents secondary electrons in the x-ray spectrum of the W(110) surface. We emphasize that this source of electrons is independent from the first one caused by the He IIα radiation. A resulted single-scan spectrum is shown by filled circles in Fig. 2.

Let us assume, that the observable variables I_n (He IIα intensity, probing signals) are random and independent from each other. They are characterized by a mean value $\bar{I}$ and a positive dispersion $\sigma_n^2>0$. We will consider values $\Delta_n=I_n-\bar{I}$ as probing excitations (perturbations) introduced by experimenters in a way that they do not correlate with $N_n(E_{\mathrm{kin}})$. The latter assumption is of crucial importance to justify the reported SPSA-based approach of data evaluation. This assumption is valid if I_n is randomly selected, while N_n is unknown but bounded deterministic function.

The main issue of the novel algorithm reported in Refs. 9 and 10 is to estimate mean value $\bar{\theta}(E_{\mathrm{kin}})$ of $\mathrm{const}\times\mathrm{DOS}(E_{\mathrm{kin}})$ via measuring individual photoemission spectra $j_n(E_{\mathrm{kin}})$ with known probing random excitations I_n. Intuitively, the underlaying mathematics can be understood by considering Eq. (5), which can be partially differentiated with respect to variable I_n. If I_n and N_n are independent from each other values, the partial derivative of $j_n(E_{\mathrm{kin}})$ over I_n equals to $\theta_n(E_{\mathrm{kin}})$.

On the basis of the discussion presented earlier, the task of filtering of systematic noise can be reformulated as the problem to build up a sequence of estimations of a mean parameter value, $\{\hat{\theta}_n(E_{\mathrm{kin}})\}$, in a model of linear regression. It was shown[10] that, in this case, one can effectively use a SPSA-type algorithm or a randomized least-squares method.

Figure 1(b) demonstrates an application of the recurrent SPSA approach,[10]

$$\hat{\theta}_n(E_{\mathrm{kin}})=\hat{\theta}_{n-1}(E_{\mathrm{kin}})-\frac{\Delta_n}{\sigma_n^2 n}[I_n\hat{\theta}_{n-1}(E_{\mathrm{kin}})-j_n(E_{\mathrm{kin}})], \quad (6)$$

where $n=1,2,\ldots,k$, $\hat{\theta}_0(E_{\mathrm{kin}})=0$, for evaluation of the described experimental photoemission data.

If the estimation sequence $\{\hat{\theta}_n(E_{\mathrm{kin}})\}$ leads to some particular value $\tilde{\theta}(E_{\mathrm{kin}})\geqslant 0$, we can assert that this final result approximates the real value of $\bar{\theta}(E_{\mathrm{kin}})$ (for details, see Ref. 10). In the experiment, however, we can follow only finite number of measurements. Supposing that after some measurements, the sequence of $\{\hat{\theta}_n(E_{\mathrm{kin}})\}$ becomes stable, we can assume with high probability that this estimation value is in good agreement with the real value $\bar{\theta}(E_{\mathrm{kin}})$.

For our analysis, we have used 50 PE spectra where different SNs were added. Figure 3 shows the evolution of

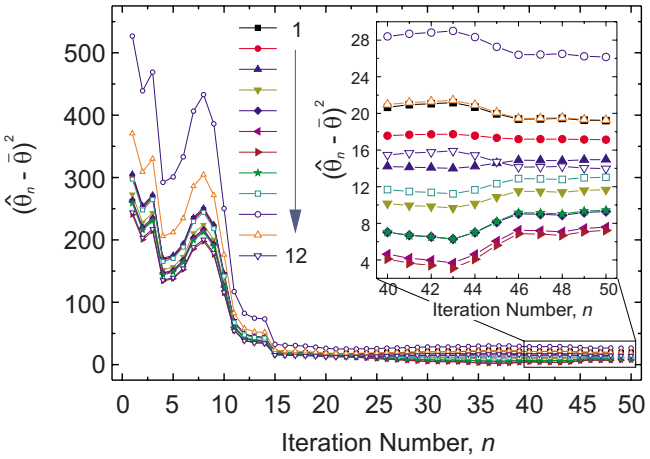

FIG. 3. (Color online) Convergence of the SPSA algorithm: squared errors $[\hat{\theta}_n(E_{\mathrm{kin}})-\bar{\theta}(E_{\mathrm{kin}})]^2$ of the 12 control points marked in Fig. 2.

estimated values $\hat{\theta}_n(E_{\mathrm{kin}})$ with a limit value, which has to be proportional to the DOS. In order to prove the SPSA algorithm, we chose 12 control points in PE spectra at particular kinetic energies (points are marked on kinetic energy axis in Fig. 2). The sequence of the squared estimation errors for these points in PE spectra during step-by-step application of the algorithm is plotted in Fig. 3 with corresponding zoom for the last ten steps (see inset). One can see that in spite of the overestimated large initial guess values, the algorithm process is almost stabilized at around 20th iteration. From iteration 46th, we have the most stable estimations consequence. Figure 2 shows the result of the application of 50 steps of the SPSA algorithm (bottom spectrum). The evaluated spectrum is in good agreement with the one directly measured in the experiment, when the x-ray source was not activated (no SN noise).

In conclusion, we have demonstrated that the application of the SPSA algorithm is an effective way of SN attenuation in the linear regression case. As an example, we applied this algorithm to filtering the systematic noise in the PE spectra. It was found that set of 50 single-scan spectra is already enough to eliminate the systematic error. On the basis of these results, we conclude that application of SPSA can be useful for analysis of a large variety of experimental data.

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