A global non-analytical fit method for the complex lineshape analysis of Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$

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Abstract. The implementation of global non-analytical fit is exemplified by its application to the spectral analysis of the complex lineshape of Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ at ($\pi$,0). It deals with properly a multi-level globality in the fitting parameters and efficiently the non-analytical evaluation of the fitting function, thus exhibiting a potential applicability to a wide range of systematic analysis task.

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Angle-resolved photoemission spectroscopy (ARPES) has greatly advanced our microscopic understanding of the high-temperature superconductivity by presenting high-precision measurements of various dependence[1]. Among them, for example, the doping and temperature ($T$) dependent measurement on Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (Bi2212) revealed the existence of a coherent component in the lowest-lying binding energy position which is closely related to the superconducting transition[2]: the momentum and photo energy ($h\nu$) dependent investigations identified the long-sought bilayer band splitting effect in the very overdoped (OD) Bi2212[3] and argued for its persistence across a large doping range of the sample[4]. Due to the spectroscopic nature of ARPES, the intrinsic single-particle spectral function is masked by the matrix element effect[5] from a direct detection of the multi-component lineshape, for example, the bilayer-split peak-dip-hump lineshape of the OD65 Bi2212 (with a $T_c=65$ K). In Ref.[6], a combined analysis of the $h\nu$ and $T$ dependence was achieved for a reliable extraction of the intrinsic spectral information. To naturally account for the physical constraints imposed by the intimate connection of the two dependence of data, a global fitting program was developed, to our knowledge, for the first time in the spectral analysis of complex lineshape with simultaneous consideration on various dependence.

In this paper, we revisit this systematic spectral analysis on the 35 energy distribution curves (EDC’s) [Fig. 1(a)-(c)] by detailing the implementation of the global fitting procedure. It deals with properly a multi-level globality (defined below) in the fitting parameters and efficiently the non-analytical evaluation of the fitting function, thus exhibiting a potential applicability to a wide range of systematic analysis task.

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Figure 1. (color) Results of the global non-analytical fit on the $T$-dependent (a) (see also [d] for an enlargement of the $T=30$ K curves) and $h\nu$-dependent ARPES spectra at $(\pi,0)$ of (Pb)-OD65 in the superconducting state at $T=10$ K (b) and normal state at $T=80$ K (c). The AB hump lies lower in energy than the BB hump. The width difference of the peak is due to resolution variations from 10 to 18 meV at different $h\nu$’s.

For clarity, we recap the fitting function used for the lineshape modelling in Ref. [6], with only a slight modification described below.

$$I(\omega, T, h\nu) = I_0(T, h\nu) \cdot \left[ \sum_{\alpha} \left( J_{\alpha b}(\omega, T, h\nu) \cdot f(\omega, T) \right) \otimes R(\omega, \Gamma'(h\nu)) + B(\omega, T) \right] + I_1(T, h\nu),$$

where $f$ is the Fermi function, $R$ the $h\nu$-dependent resolution Gaussian, $B$ the $T$-dependent empirical background function[6], $I_0$ and $I_1$ the EDC-specific linear intensity coefficients. The summation is over the spectral intensity of anti-bonding band (AB) hump, bonding band (BB) hump, AB peak and BB peak, given by, respectively,

$$J_{\alpha b}(\omega, T, h\nu) = M_{\alpha b}(h\nu) \cdot C_{\alpha b}(T) \cdot A_{\alpha b}(\omega, T, \alpha, \varepsilon_{\alpha b}),$$

$$J_{bb}(\omega, T, h\nu) = M_{bb}(h\nu) \cdot C_{bb}(T) \cdot A_{bb}(\omega, T, \alpha, \varepsilon_{bb}),$$

$$J_{ap}(\omega, T, h\nu) = M_{ap}(h\nu) \cdot (1 - C_a(T)) \cdot A_p(\omega, \Gamma_a(T), \varepsilon_{ap}),$$

$$J_{bp}(\omega, T, h\nu) = M_{bp}(h\nu) \cdot (1 - C_b(T)) \cdot A_p(\omega, \Gamma_b(T), \varepsilon_{bp}).$$
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, where $M$ is the (squared) matrix element dependent only on $h\nu$, $C$ the remaining spectral weight in the hump with its $T$ dependence to be fitted, $A_h$ and $A_p$ the self-normalized spectral functions for the hump and the peak, respectively, given by

$$A_h(\omega, T, \alpha, \omega_0) = \frac{\xi |\Sigma'\prime(\omega, T)|}{(\omega - \sqrt{\omega_0^2 + \Delta_{uc}(T)^2})^2 + \Sigma'\prime(\omega, T)^2},$$

$$A_p(\omega, \Gamma(T), \omega_0) = \frac{2\sqrt{\ln 2}}{\sqrt{\pi} \Gamma(T)} \exp\left[-\left\frac{\omega - \omega_0}{\Gamma(T)/2}\right]^2\right].$$

We modified the empirical form of the imaginary part of self-energy into $\Sigma''(\omega, T) = |\sqrt{(\alpha\omega)^2 + (\beta T)^2 + \zeta}|$, where lifetime broadening associated with the impurity level of samples is reflected by $\zeta$ and that with doping level by $\alpha$ and $\beta$. They are assumed to be independent of $h\nu$ and $T$. $\xi$ is a self-normalization factor given by $\int_{-\infty}^{\infty} A_h(\omega) d\omega = 1$. $\Gamma(T)$ is a $T$-dependent linewidth of the peak. For the bilayer-split bands in the superconducting state, a realistic SCG opening introduces an effective $T$-dependent energy shift from their $T$-independent renormalized band energy positions ($\varepsilon_{ah}$ and $\varepsilon_{bh}$) while the energy position of the peak ($\varepsilon_{ap}$ or $\varepsilon_{bp}$) is subjected to the fit.

In order to retrieve reliable quantitative information from a multi-dimensional fit to a set of interconnected EDC’s, many physical constraints are involved and strictly followed. For example, in the the $h\nu$-dependent set, all functions of $T$ ($C$, for example) are shared variants to all EDC’s at the same $T$ while the functions of $h\nu$ (the matrix elements, for example) are locally specified, and in the $T$-dependent set, vice versa. In other words, for a given fitting parameter which is subjected to a constrain, it is required to have the same value for a specific group of EDC’s (forming a so-called EDC’s subset for this parameter). Thus, the total number of its possible values across the whole EDC set is smaller than the set size, 35. We define this number the globality level ($Globality$ for this fitting parameter. Particularly, the parameters with $Globality = 1$ are in the top globality level, for example, $\varepsilon_h$, $\alpha$ and those kept fixed during the fit, such as the parameters for the BB peak component which is not considered in the three-component fit exemplified below; the lowest globality level ($Globality = 35$) is owned by those local parameters with their values specific to each EDC, such as $I_0$.

Technically, our fitting function contains some non-analytical features, convolution by the resolution function and integration in the self-normalization of spectral function. The conventional point-by-point evaluation of the fitting function in the fitting iteration fails to guarantee an efficient optimization to such a large data set. We developed our fitting program in WaveMetrics Igor Pro 4.0[7]. After implanting the fast algorithm for non-analytical fitting[8] in the two-level global fit procedure by Igor (where only the top and the lowest globality levels are concerned), we further establish its multi-level global fitting capability. We illustrate below the 5-step implementation of our global non-analytical fit based on the 3-component model (neglecting the $J_{bp}$ term in the first equation) on the $h\nu$ and $T$ dependent lineshapes of (Pb-)OD65[6].

Step 1 : Include in the fit the 9 normal state (@80K) then the 9 superconducting state (@10K) $h\nu$-dependent EDC’s on Pb-OD65 (in an ascending order sorted by the
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Table 1. List of fitting parameters

| Para No. | 0  | 1  | 2  | 3  | 4  |
|----------|----|----|----|----|----|
| Parameters | $I_0$ | $M_{ab}$ | $M_{bh}$ | $M_{ap}$ | $M_{bp}$ |
| $Globality^b$ | 35 | 10$(2 \times 9 + 17 \times 1)$ | 10$(2 \times 9 + 17 \times 1)$ | 10$(2 \times 9 + 17 \times 1)$ | 1 |
| Fixation | × | × | × | × | arb. $^a$ |
| Para No. | 5  | 6  | 7  | 8  | 9  | 10  | 11 |
| Parameters | $C_a$ | $C_b$ | $\varepsilon_{ab}$ | $\varepsilon_{bh}$ | $\varepsilon_{ap}$ | $\varepsilon_{bp}$ |
| $Globality$ | 17$(10 \times 2 + 1 \times 15)$ | 1 | 1 | 1 | 17$(10 \times 2 + 1 \times 15)$ | 1 | 1 |
| Fixation | × | 1 | × | × | × | arb. | × |
| Para No. | 12 | 13 | 14 | 15 | 16 | 17 |
| Parameters | $\Gamma_a$ | $\Gamma_b$ | $\delta E_F^d$ | $I_1$ | $\beta$ | $\zeta$ |
| $Globality$ | 2$(18 \times 1 + 17 \times 1)$ $^c$ | 1 | 1 | 35 | 1 | 2$(18 \times 1 + 17 \times 1)$ $^c$ |
| Fixation | × | arb. | 0 | × | × | × |

$^a$ arbitrary value;  
$^b$ the number without (or outside) the bracket: $Globality$ (number of subsets); expression inside the bracket: size of each subset (number of EDC’s contained) $\times$ number of subsets of the same size;  
$^c$ to assume that the quality difference between Pb-OD65 and OD65 used in the two dependent investigations should be reflected only in the quasi-particle lifetime;  
$^d$ a slight correction in energy for $E_F$ of each EDC to account for the uncertainty in $E_F$ alignment.

$h\nu$ values, respectively). Append to the EDC’s set the 17 $T$-dependent EDC’s on OD65 using He-I (in a descending order sorted by the $T$ values). The EDC’s are labelled from 0 to 34 and concatenated sequentially in X and Y scale, respectively, to form a huge "EDC".

Step 2 : Discriminate the fitting parameters of the top globality level from the otherwise (of intermediate globality or the lowest globality). Then fix those parameters if they are required to be invariable during the fit. We summarize in Table 1 the properties of all the fitting parameters involved.

Step 3 : Build a global fit parameters set, which includes sequentially, the 9 parameters with $Globality = 1$, the remaining 9 parameters with $Globality > 1$ ($9 \times 35 = 315$, totally) grouped in the number order of the EDC in the EDC concatenation to which they belong. Hence, we end up with another index to locate the parameter in a global manner, called GL Para No. hereafter, from which we can set up a bidirectional mapping to the original Para No. (in Table 1) for a specific EDC.

Step 4 : The global fit problem on a set of EDC’s has been converted into a fit on the EDC concatenation with the global fit parameters set. Having in mind the unique calling pattern of the fitting function in Igor curve fitting routine$^8$ and the time-consuming non-analytical evaluation in the fitting function (see Ref. $^9$), a direct fit is practically unfeasible, which asks for a function calling time up to $(M+1) \times N$ ($M$ and $N$ denote the total GL Para No. and point numbers of the EDC concatenation, here 324 and $\sim 5400$, respectively) for a single iteration! The introduction of a fast algorithm greatly reduces the function calling time down to $(M+1) \times L$ ($L$ denotes the total EDC number included in the concatenation, here 35) at the expense of a size increase in the RAM which is used for the storage of the trial fit curves$^8$. 


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Figure 2. Truth tables as black-and-white plots (black: True, white: False), where the true pertinence of elements in the global fit parameters set to each EDC is recovered for a) a two-level global fit or b) a multi-level global fit in the example problem. The arrows identified by the Para No. locate the corresponding features of the fitting parameters with intermediate globality levels. For example, the three arrows from "1" indicate $M_{ah}$ has two independent values for the $h\nu$ and $T$ dependence subset, respectively, one for the EDC pairs taken at the same $h\nu$ and the other for all the EDC’s taken with He-I (the so-called parameter degeneracy of $M_{ah}$). A horizontal line segment with unit length stands for a parameter with Globality = 35 while a full-scale line segment for the one with Globality = 1. Note that the patterns may vary dramatically with different orders the EDC and parameters of the sets would be arranged in.

We can further cut down the time to $(K + 1) \times L$ ($K$ denotes the total Para No., here 18) by exploiting the fact that in a global fit a large number of the global fit parameters are actually not related to the function evaluation for a given EDC, which needs to be reduced from the EDC concatenation during each iteration of the fit. This is because only $K$ out of $M$ parameters in the global fit parameters set comes from a single EDC.

To pick out properly those relevant parameters which really contribute to the function update on a given EDC, a truth table helps, which is convenient to build at the end of Step 3. In Fig. 2(a), we show this binary table as a black-and-white plot. The bottom black wall and black bricks in the diagonal clearly indicate the order in which the global fit parameters set is organized and, especially, the EDC-specific grouping nature of the Globality > 1 parameters following the Globality = 1 ones (reminiscent of Step 3).

Step 5 : The global fit involves only two levels in Step 4. The task of this step is to achieve a multi-level globality defined in Table 1. This only requires our making modifications on the truth table accordingly. Fig. 2(b) shows a truth table modified to achieve the wanted globality configuration. In contrast to Fig. 2(a), the introduction of intermediate globality levels to the global fit parameters set results in the parameters degeneracies for certain EDC’s as well as, in return, some "void"
parameters, which seemingly have no effect on any EDC in the concatenation. To prevent Igor curve fit routine from "a singular matrix error" report, we simply fix them to arbitrary values during the fit. This modification doesn’t alter the conservation in the column sum of truth values in the table (i.e., the total Para No., \( K \)), and thus neither the iteration efficiency.

In Fig. 1 the fit curves collapse well onto the experimental EDC’s, reminiscent of Figs. 2-3 in Ref. 6. Pay attention to the great contrast in the overall quality (or details) of the fit between the ones shown in Fig. 2 of Ref. 9 and ours, though on data sets generally twice larger in size. This suggests the local fit scheme with manual adjustments on the global fitting parameters is unlikely to yield a reliable result in a truly global sense. In contrast, our fully-automatic fitting routine can yield a robust, efficient and physically-constrained global fit for a large data set, which is frequently encountered in the spectral analysis of ARPES.

Note that the algorithm proposed here is not limited to the application of the ARPES lineshape analysis only, but of a great applicability to a wide range of problems where a multi-level global analysis or/and non-analytical nonlinear fit is involved. For example, the analysis of hysteresis curves in the magnetism research can be a good case in point for its potential application. The angle-dependent hysteresis measurement contains the information of magnetic anisotropy which is currently retrieved only qualitatively due to the lack of a global fit on the angle-dependent curves set. Furthermore, in most cases, the single-domain coherent rotation model in the textbook is not sufficient to include the necessary physics to reproduce the hysteresis curve in calculation. By using spin dynamics as the non-analytical evaluation kernel in the global fit, the roles of collaborative spin motion and domain formation which are requisite for the hysteresis can be investigated quantitatively. Generally, our algorithm provides a useful and universal guideline to serve the advanced systematic analysis in experimental physics.

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