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Studies of Low Temperature Thermoluminescence of GAGG:Ce and LuAG:Pr Scintillator Crystals Using the $T_{\text{max}}$-$T_{\text{stop}}$ Method

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HIGHLIGHTS

▶ Thermoluminescence of GAGG:Ce and LuAG:Pr has been studied by the $T_{\text{max}}$-$T_{\text{stop}}$ method.

▶ Glow curves have been analyzed in order to derive trap parameters.

▶ Inclusion of quasi-continuous trap distributions has resulted in high quality fits.

ARTICLE INFO

ABSTRACT

Low temperature thermoluminescence of GAGG:Ce and LuAG:Pr scintillator crystals has been studied by means of the $T_{\text{max}}$-$T_{\text{stop}}$ method. It is shown that the glow curves of both materials are superpositions of discrete glow peaks and broad quasi-continuous Gaussian distributions of trapping levels. A model function has been built and trap parameters have been evaluated.

Keywords: LuAG:Pr

GAGG:Ce

Thermoluminescence

$T_{\text{max}}$-$T_{\text{stop}}$ method

Trap distribution

1. Introduction

Rare-earth activated wide-bandgap insulator crystals have always attracted attention as potential fast and efficient scintillator materials for manifold applications. Oxides and fluorides activated with cerium entered the market in the eighties of the 20th century [1], while the interest in praseodymium ions emerged more than 20 years later. In particular, Pr-activated lutetium aluminum garnet (Lu$_3$Al$_5$O$_{12}$:Pr, LuAG:Pr) was...
suggested as a promising scintillator in 2005 [2], characterized by a high density of 6.7 g/cm$^3$, a perfect energy resolution of 4.6% (at 662 keV), and a fast decay constant of 20 ns [3,4,5], with a prompt implementation in the field of medical diagnostics [6,7]. Interestingly, further studies on intrinsic defects occurring in the Lu$_3$Al$_5$O$_{12}$ host (and generally in rare-earth oxide garnets) motivated the search for new materials in the frame of so-called “bandgap engineering” [8] and resulted with a development of a family of Gd$_3$(Al$_x$Ga$_{1-x}$)O$_{12}$:Ce scintillators, of which Gd$_3$Al$_2$Ga$_3$O$_{12}$:Ce (hereby referred to as GAGG:Ce) with a high light output of 42000 ph/MeV and a reasonably fast decay time of 50 ns being the most prospective one [9].

Although the scintillation yield of GAGG:Ce is more than twice higher than that of LuAG:Pr (42000 ph/MeV vs. 20000 ph/MeV), in both cases there seems to be plenty of room for improvement, which is related to the fact that the yields of both materials observed at room temperature are much lower than at other temperatures (180 K for GAGG:Ce, 450 K for LuAG:Pr). In LuAG:Pr this feature has been attributed to the presence of shallow electron traps, which affect the energy transfer from the LuAG host to the Pr$^{3+}$ ions [10]. In GAGG:Ce the distinct yield drop above 180 K has also been investigated [11], but its origin is still unclear. Nevertheless, the participation of traps in the GAGG-to-Ce$^{3+}$ transfer is also very possible. Both LuAG:Pr and GAGG:Ce display intense thermoluminescence (TL) signal following X-ray irradiation at 10 K, which confirms the existence of various traps in these materials [11,12,13]. Though the complex structure of the glow curves turns their quantitative analysis into an elaborate and relatively difficult task, any efforts to derive reliable trap parameters are desired and should contribute to the state of knowledge on the processes of energy transfer and radiative recombination in oxide garnet scintillators.

In this paper we present the results of detailed low temperature TL studies on GAGG:Ce and LuAG:Pr. Besides a standard glow curve recording we have introduced a procedure of gradual thermal bleaching of traps following the McKeever’s $T_{\text{max}}$-$T_{\text{stop}}$ method [14]. Our results point out that the glow curves of either GAGG:Ce or LuAG:Pr are certainly not formed by plain convolutions of discrete peaks. Instead, the curves are dominated by broad bands, on which a few weaker discrete peaks are imposed. In such a case an approach generally used by most of scientists, i.e. a deconvolution of a glow curve into several first-order (or ever general-order) peaks, makes no sense. Although it is usually possible to find thereby a set of trap parameters (activation energies $E$ and frequency factors $s$) that lets the experimental curve be well reproduced by the fitted one, such parameters are simply of no physical value. In particular, broad bands analysed with the well-respected Randall-Wilkins model for first-order peaks [15] often yield extremely low values of the frequency factor (even below 100 s$^{-1}$ [12,16]). Nikl et al. [17,18] ascribed the incidence of such bands to a so-called thermally assisted tunneling. However, as suggested by Medlin [19], McKeever [14], and Chen [20], these bands may also originate from quasi-continuous distributions of trapping levels. We show that the performed $T_{\text{max}}$-$T_{\text{stop}}$ measurements on GAGG:Ce and LuAG:Pr indicate that indeed in both materials we deal with such distributions, whereas a fitting procedure incorporating a superposition of distributions and discrete peaks provides a credible set of trap parameters.
2. Materials and Experiment

The 1 mm thick samples of GAGG:Ce and LuAG:Pr used in the current research have been studied before and selected results concerning their low temperature TL have already been presented [11,12]. The Lu$_3$Al$_5$O$_{12}$:2.5%Ce and Gd$_3$Al$_2$Ga$_3$O$_{12}$:0.5%Ce crystals have been grown at Furukawa Co. Ltd. and at Tohoku University, respectively.

A custom set-up consisting of an Inel XRG3500 X-ray generator (Cu-anode tube, 45 kV / 10 mA), an Acton Research Corporation SpectraPro-150 monochromator (set to zero-order), a Hamamatsu R928 photomultiplier, and an APD Cryogenics Inc. closed-cycle helium cooler with a Lake Shore 330 programmable temperature controller, has been employed to investigate low temperature TL. During a standard TL readout the sample is first exposed to X-rays for 10 min at 10 K. Then, after the X-ray shutter has been closed and the remaining afterglow has disappeared (at least to some extent), the sample is heated up to 300 K at a constant rate of about 0.14 K/s and the collected signal forms a typical glow curve.

The utilized $T_{\text{max}}-T_{\text{stop}}$ procedure is similar to the one originally proposed by McKeever [14]. A single run consists of two steps. First, after a 10 min X-ray irradiation, a partial glow curve ranging from 10 K to a particular temperature denoted as $T_{\text{stop}}$ is measured and, simultaneously, most of the carriers are removed from the traps peaking below $T_{\text{stop}}$. When $T_{\text{stop}}$ is reached, the heater is turned off and the sample is cooled down back to 10 K. Then, without any irradiation, a complete glow curve is recorded in the entire range, i.e. from 10 to 300 K. Such a procedure is repeated several times, each time with an increased $T_{\text{stop}}$ value, gradually bleaching the remaining structure of the glow curve; an example of a two-step run of such an experiment on GAGG:Ce is presented in Fig. 1 (first step: 0-2600 s, second step: 2600-3500 s). We note that an utter $T_{\text{max}}-T_{\text{stop}}$ study in case of LuAG:Pr has involved the values of $T_{\text{stop}}$ between 45 and 275 K, while for GAGG:Ce proceeding with $T_{\text{stop}} > 125$ K has been suspended since the peaks at 180 K and above are resolved clearly enough.

3. Experimental Results

As a starting point in the data processing, each second-step curve (Figs. 2 and 3) is analyzed in order to derive the value of $T_{\text{max}}$ corresponding to the first local maximum observed above 10 K. In this way a set of pairs of values ($T_{\text{stop}}$, $T_{\text{max}}$) is obtained for each crystal (hence the $T_{\text{max}}-T_{\text{stop}}$ name of the method). In his paper McKeever [14] discusses three typical cases that can be met with this particular method. Some examples important for the current work are reprinted and explained in Fig. 4.

Subsequently, the ($T_{\text{stop}}$, $T_{\text{max}}$) pairs are used to build so-called $T_{\text{max}}-T_{\text{stop}}$ plots (Figs. 5 and 6). According to McKeever [14], for GAGG:Ce one can anticipate the existence of discrete traps peaking at about 35, 45 and 85 K (and some more at and above 180 K), as well as a quasi-continuous trap distribution producing a broad band centered around 80 K. Similarly, in LuAG:Pr there are most probably a few peaks associated with discrete traps, overlapping with a very broad distribution-related band. We note that the slopes of lines...
fitted to the \((T_{\text{stop}}; T_{\text{max}})\) points in the thermal ranges corresponding to the expected distributions are close to unity both for GAGG:Ce \((\alpha = 0.98 \pm 0.02)\) and for LuAG:Pr \((\alpha = 1.04 \pm 0.06)\), which strongly supports the interpretation in which the incidence of quasi-continuous distributions of traps is assumed [14,16].

4. Theoretical Considerations

Since the experimental results indicate the existence of several traps producing discrete glow peaks and, furthermore, the peak positions are independent of the irradiation dose (proving that the kinetics is of the first order), we start with the standard equation proposed by Randall and Wilkins [15]:

\[
I(T) = n_0 s \exp\left(-\frac{E}{kT}\right) \exp\left[-\frac{s}{\beta} \int_{T_0}^{T} \exp\left(-\frac{E}{k\theta}\right) d\theta\right]
\]  

\((E – \) the trap depth, \(s – \) the frequency factor, \(k – \) the Boltzmann constant, \(n_0 – \) the initial trap concentration, \(\beta – \) the heating rate). To remove the inconvenience of the analytically unsolvable integral we incorporate a continued fraction solution given by Paterson [21]. To do this we approximate \(T_0\) with 0, transform the expression and substitute \(\theta\) with a dummy variable:

\[
I(T) = n_0 s \exp\left(-\frac{E}{kT}\right) \exp\left[-\frac{sE}{\beta k} \int_{0}^{\infty} e^{-\beta u^2} du\right]
\]  

\[
I(T) = n_0 s \exp\left(-\frac{E}{kT}\right) \exp\left[-\frac{sE}{\beta k} \exp(\varphi)\right]
\]  

where:

\[
\varphi(x) = \frac{\exp(-x)}{x} \left(1 + \frac{1}{x + 2} + \frac{2}{(x + 4) + 6} + \frac{n(n + 1)}{x + 2(n + 1)\cdots}\right)
\]  

in which:

\(x = E / kT\)

while \(n\) is a positive integer corresponding to the number of terms used in the \(\varphi(x)\) function approximation. For our calculations we have chosen \(n = 20\), since higher values have had no meaningful impact on the fit quality, only increasing thereby the calculation time.

Our second task in composing the model that would properly reproduce the observed glow curves is to include a quasi-continuous distribution of trapping levels. Based on our \(T_{\text{max}}; T_{\text{stop}}\) measurements a large part of the TL signal is gradually bleached after each increase of the \(T_{\text{stop}}\) value. According to McKeever [14] and Chen [20] this suggests an unknown continuous distribution of traps that are emptied with each
consecutive temperature increase. We use this information presuming that the observed total intensity is a convolution of a glow curve function $F$ and a function $A$ that describes the amplitude of each curvelet:

$$I(T) = \int_{E_2}^{E_1} \int_{E_2}^{E_1} A(E, s) F(E, s, T) dEds$$

(6)

To simplify the model and to avoid integrating over frequency factors $s$ we follow the postulate of Chen [20] that they are constant for each of the traps building a particular distribution. Since due to some computational limitations the integral in Eq. (6) must be replaced with a summation, we eventually arrive at the model function $M$ that combines the discrete peaks with a continuous distribution:

$$M(T, n_0, E, s, A) = \sum_{j=1}^{n} n_0 s \exp \left( -\frac{E_j}{kT} \right) \exp \left[ \left( \frac{sE_j}{kT} \right) \phi(T, E_j) \right] +$$

$$+ \sum_{j=1}^{n} A_j s \exp \left( -\frac{E_j}{kT} \right) \exp \left[ \left( \frac{sE_j}{kT} \right) \phi(T, E_j) \right] \Delta E$$

(7)

$n$ – the number of discrete first order peaks, $E'$ – the energy values ranging from $E_0$ to $E_\nu$ with a step of $\Delta E$, $\nu$ – the number of objects in the $\{E'\}$ set corresponding to the number of trapping levels building the distribution, $A_j$ – an unknown set of amplitudes characterizing the trap distribution). The first sum in the $M$ function characterizes the discrete first-order glow peaks, whilst the second one describes the continuous distribution of trapping levels. The $A_j$ parameters are related to the envelope of the histogram built by the traps forming the distribution.

In our first approach to fit the data it has turned out that for both GAGG:Ce and LuAG:Pr the envelope of the histogram can be very well approximated with a superposition of two Gaussian functions. Therefore in the final step we modify the model function $M$ by restricting the $A_j$ values of the individual glow peaks, tying them to Gaussian distributions:

$$M(T, n_0, E, s, A) = \sum_{j=1}^{n} n_0 s \exp \left( -\frac{E_j}{kT} \right) \exp \left[ \left( \frac{sE_j}{kT} \right) \phi(T, E_j) \right] +$$

$$+ \sum_{j=1}^{n} \sum_{l=1}^{2} G(n_{0,j}, E_{0,j}, \sigma_j) \exp \left( -\frac{E_j}{kT} \right) \exp \left[ \left( \frac{sE_j}{kT} \right) \phi(T, E_j) \right] \Delta E$$

(8)

where $G$ is a Gaussian function similar to the one proposed by Medlin [19]. This substitution allows us to reduce the number of variable parameters describing the quasi-continuous distribution to 6, since we represent it with two overlapping Gaussian distributions of instead of a sum of plenty of glow peaks with different population parameters corresponding to each energy level $E_j$.

In order to assess the quality of the fits we employ the FOM (figure of merit) parameter proposed by Bos et al. [22] and Chen [20], which can be expressed as:
FOM = \sum_{i=1}^{N} \frac{|I_i - M(T_i)|}{A} \cdot 100\% \tag{9}

in which \( i \) numbers the successive datapoints, \( I_i \) are the experimental values of TL intensity at temperatures \( T_i \), \( M(T_i) \) – the values of the fitted function at \( T_i \), and \( A \) – the integral of the fitted glow curve. The values of FOM not exceeding a few percent usually indicate good fits (the lower the value, the better the fit) [22].

As an alternative parameter for the fit quality evaluation we utilize the adjusted coefficient of determination \( \bar{R}^2 \) defined as:

\[
\bar{R}^2 = 1 - \frac{\sum_{i=1}^{N} (I_i - M(T_i))^2}{\sum_{i=1}^{N} I_i^2 \frac{N}{N - P}} \tag{9}
\]

\((N – the total number of datapoints, \( P – the number of model parameters). The closer to unity the value of \( \bar{R}^2 \), the better the fit.

5. Calculations

For nonlinear regression of the curves we have used the restricted Marquardt-Levenberg method [23,24]. A fitting program employing Eq. (8) has been written in Python 2.7.6 [25] with NumPy extension [26].

The numbers of discrete first order peaks have been determined based on the data from the \( T_{\text{max}} - T_{\text{stop}} \) procedure. In this way we have pinpointed the discrete traps peaking at around 35, 45, 85, 180, 240, and 255 K (GAGG:Ce) or at 85, 170 and 260 K (LuAG:Pr). Preliminary input values for fitting have been either taken from previous works [11,12] or chosen by trial and error. Concerning the selection of the frequency factor for distributions we point out that Chen [20] has proposed to start with an arbitrary value between \( 10^{10} \) and \( 10^{12} \) s\(^{-1}\). The values that can be found in literature for various scintillators are generally consistent with this suggestion: \( 2.4 \cdot 10^{11} \) s\(^{-1}\) for YAG:Ce [27], \( 3 \cdot 10^{12} \) s\(^{-1}\) for LuAG:Ce [28], or \( 7 \cdot 10^{11} \) s\(^{-1}\) for YAP:Ce [29]. In the current work we have performed calculations with the following values of \( s \): \( 10^{10} \), \( 10^{11} \), \( 10^{12} \), \( 10^{13} \), and \( 10^{14} \) s\(^{-1}\). For both GAGG:Ce and LuAG:Pr the value of \( 10^{12} \) s\(^{-1}\) provides the best fit quality and the lowest errors reflected by the values of FOM and \( \bar{R}^2 \).

The final results of the fitting procedure are presented in Figs. 7 and 8. In each figure, besides the total fitted glow curve, its components (discrete peaks and distributions) are clearly indicated. The distributions themselves are shown in Figs. 9 and 10, wherein the energy range of the distribution spans from 0.01 to 0.37 eV for GAGG:Ce, and from 0.01 to 0.80 eV for LuAG:Pr, using 150 glow peaks corresponding to the energy values separated by \( \Delta E = 0.00213 \) eV and \( \Delta E = 0.00526 \) eV, respectively (the bars represent the \( n_{oi} \) values corresponding to the energy \( E_j \)). The values of all the trap parameters derived from the fitting procedure are listed in Tables 1 and 2. We note that the values of FOM and \( \bar{R}^2 \) let us qualify the fits as “technically” good. From the scientific point of view it is important that the broad bands yielding doubtful,
very low values of the frequency factor when fitted as discrete peaks have been replaced with quasi-continuous distributions, the existence of which has been confirmed by the $T_{\text{max}}-T_{\text{stop}}$ method.

6. Conclusions

The performed studies allow us to gain an insight into the distribution of traps in the energy structure of GAGG:Ce and LuAG:Pr. We have suspected that the materials contain sets of discrete traps superposed with wide quasi-continuous distributions of trapping levels. This hypothesis has been proved correct by the $T_{\text{max}}-T_{\text{stop}}$ method. Our further calculations suggest that indeed the glow curves of both materials, besides comprising a few “standard” glow peaks obeying the first order kinetics, are formed by broad bands that can be well described by double Gaussian distributions.

The incidence of quasi-continuous distributions and their double Gaussian structure, however, seems to be the only similarity between GAGG:Ce and LuAG:Pr. The former scintillator exhibits most of its TL at much lower temperatures (below 150 K) compared to the latter, which is confirmed by the values of FWHM of the distribution bands (68 and 80 meV for GAGG:Ce vs. 80 and 216 meV for LuAG:Pr). The wider of the distributions in LuAG:Pr covers almost the entire low temperature region up to room temperature. Actually it is reflected in Fig. 3, wherein the last recorded spectrum is bleached at 270 K, i.e. above the maximum of the 3rd discrete peak, and we still observe some residual emission. Concerning the depths of the traps (both discrete and those forming the distributions) it can also be noticed that traps in GAGG:Ce are generally shallower than in LuAG:Pr. We remark that these observations are fully consistent with the predictions of Dorenbos [30], according to whom the replacement of lutetium with yttrium or gadolinium would lower the bottom of the conduction band, but should not affect the number of traps, thus traps should be indeed shallower in GAGG:RE than in LuAG:RE (RE = Ce, Pr, …).

We also note that, as already mentioned, by including the quasi-continuous distributions of trapping levels one arrives at acceptable values (from the physical point of view) of the frequency factors of the discrete traps. If one insisted on fitting the glow curves of GAGG:Ce or LuAG:Ce with an assumption that they were composed of separate first order (or even general order) glow peaks, one would either have to accept very low, non-physical values of frequency factors [12] or to increase the numbers of peaks significantly in order to emulate the observed wide structures. In this respect the $T_{\text{max}}-T_{\text{stop}}$ method, although tedious and time-consuming, provides a valuable tool for low temperature TL studies and the reliability of results is well worth the amount of time spent on performing the complete measurements.
Acknowledgements

Thermoluminescence measurements have been performed at the National Laboratory for Quantum Technologies (NLTK) supported by the EU from the European Regional Development Fund.
Figures

(for reviewer's convince, original files are attached separately)

Fig. 1. Typical $T_{\text{max}}$-$T_{\text{stop}}$ measurement procedure (in this particular case $T_{\text{stop}} = 75$ K).

Fig. 2. Glow curves of consecutive TL measurements with increasing $T_{\text{stop}}$ for GAGG:Ce.
Fig. 3. Glow curves of consecutive TL measurements with increasing $T_{\text{stop}}$ for LuAG:Pr.

Fig. 4. Schematic glow curves and corresponding $T_{\text{max}}$-$T_{\text{stop}}$ plots as proposed by McKeever [14]: a) a single first order glow peak gives rise to a straight line with a slope equal to zero; b) a series of well separated glow peaks produces a “staircase” (the more rounded the corners of the “staircase” are, the more likely the glow peaks obey the second order of kinetics); c) a quasi-continuous distribution of glow peaks results in a straight line with a slope close to 1.
Fig. 5. The $T_{\text{max}}$-$T_{\text{stop}}$ plot for GAGG:Ce. Each point represents the location of the first local maximum in the corresponding glow curve.

Fig. 6. The $T_{\text{max}}$-$T_{\text{stop}}$ plot for LuAG:Pr. Each point represents the location of the first local maximum in the corresponding glow curve.
Fig. 7. The glow curve of GAGG:Ce (experiment and fits; the corresponding fit parameters are singled out in Table 1).

Fig. 8. The glow curve of LuAG:Pr (experiment and fits; the corresponding fit parameters are singled out in Table 2).
Fig. 9. The population of quasi-continuous trapping levels of GAGG:Ce.

Fig. 10. The population of quasi-continuous trapping levels of LuAG:Pr.
### Tables

**Table 1.**

Parameters of traps detected in GAGG:Ce, derived from fitting involving discrete traps and a double Gaussian distribution of trapping levels \( n_0 \) – initial concentration; \( E \) - trap depth; \( \sigma \) - distribution halfwidth; \( s \) - frequency factor.

<table>
<thead>
<tr>
<th>#</th>
<th>peak/distribution</th>
<th>( n_0 ) (arb. units)</th>
<th>( E ) (eV)</th>
<th>( \sigma ) (eV)</th>
<th>( s ) (s(^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>peak (1st order)</td>
<td>( 1.62 \times 10^5 )</td>
<td>0.055</td>
<td>-</td>
<td>( 8.16 \times 10^6 )</td>
</tr>
<tr>
<td>2</td>
<td>peak (1st order)</td>
<td>( 3.52 \times 10^5 )</td>
<td>0.052</td>
<td>-</td>
<td>( 6.69 \times 10^4 )</td>
</tr>
<tr>
<td>3</td>
<td>peak (1st order)</td>
<td>( 2.59 \times 10^5 )</td>
<td>0.103</td>
<td>-</td>
<td>( 7.20 \times 10^5 )</td>
</tr>
<tr>
<td>4</td>
<td>distribution</td>
<td>( 5.97 \times 10^6 )</td>
<td>0.156</td>
<td>0.0380</td>
<td>( 10^{12} )</td>
</tr>
<tr>
<td>5</td>
<td>distribution</td>
<td>( 1.14 \times 10^7 )</td>
<td>0.230</td>
<td>0.0287</td>
<td>( 10^{12} )</td>
</tr>
<tr>
<td>6</td>
<td>peak (1st order)</td>
<td>( 1.63 \times 10^5 )</td>
<td>0.168</td>
<td>-</td>
<td>( 3.39 \times 10^2 )</td>
</tr>
<tr>
<td>7</td>
<td>peak (1st order)</td>
<td>( 5.62 \times 10^4 )</td>
<td>0.511</td>
<td>-</td>
<td>( 1.37 \times 10^9 )</td>
</tr>
<tr>
<td>8</td>
<td>peak (1st order)</td>
<td>( 3.23 \times 10^5 )</td>
<td>0.309</td>
<td>-</td>
<td>( 1.14 \times 10^4 )</td>
</tr>
</tbody>
</table>

\[ \text{FOM} = 4.186\%, \quad R^2 = 0.9997 \]

**Table 2.**

Parameters of traps detected in LuAG:Pr, derived from fitting involving discrete traps and a double Gaussian distribution of trapping levels \( n_0 \) – initial concentration; \( E \) - trap depth; \( \sigma \) - distribution halfwidth; \( s \) - frequency factor.

<table>
<thead>
<tr>
<th>#</th>
<th>peak/distribution</th>
<th>( n_0 ) (arb. units)</th>
<th>( E ) (eV)</th>
<th>( \sigma ) (eV)</th>
<th>( s ) (s(^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>peak (1st order)</td>
<td>( 9.61 \times 10^4 )</td>
<td>0.141</td>
<td>-</td>
<td>( 7.87 \times 10^7 )</td>
</tr>
<tr>
<td>2</td>
<td>peak (1st order)</td>
<td>( 2.34 \times 10^6 )</td>
<td>0.387</td>
<td>-</td>
<td>( 7.67 \times 10^9 )</td>
</tr>
<tr>
<td>3</td>
<td>peak (1st order)</td>
<td>( 9.43 \times 10^5 )</td>
<td>0.773</td>
<td>-</td>
<td>( 2.23 \times 10^{13} )</td>
</tr>
<tr>
<td>4</td>
<td>distribution</td>
<td>( 6.49 \times 10^6 )</td>
<td>0.512</td>
<td>0.0301</td>
<td>( 10^{12} )</td>
</tr>
<tr>
<td>5</td>
<td>distribution</td>
<td>( 6.66 \times 10^6 )</td>
<td>0.446</td>
<td>0.111</td>
<td>( 10^{12} )</td>
</tr>
</tbody>
</table>

\[ \text{FOM} = 4.288\%, \quad R^2 = 0.9997 \]
References

[25] https://www.python.org/download/releases/2.7.6/