Rapid recognition of laser-induced breakdown spectrum in laser damage

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Abstract. In this paper, the technology of rapid target recognition based on laser-induced breakdown spectroscopy (LIBS) data of target in the process of laser damage was studied. In terms of LIBS data classification, this paper used support vector machine (SVM), which was more advantageous in nonlinear and small sample cases, as the core algorithm. In order to solve the problem of large amount of high-precision LIBS data, which led to slow recognition, a segmented weighted peak intensity algorithm was proposed. Compared with the original data, the length of the spectrum vector constructed by this method can be reduced by about 10 times. Under the condition of ensuring the recognition accuracy, the recognition time of LIBS samples can be reduced by an order of magnitude. At the same time, the method can balance the recognition time and accuracy by adjusting the segment length, which has good flexibility and practicability.

1. Introduction

With the rapid development of laser technology, laser weapons are going to the battlefield [1-3]. In the process of laser damage to targets, due to the different material composition of different targets, the damage efficiency of different targets under the laser irradiation are quite different [4]. In order to accurately judge the damage grade of the target and select more effective damage mode according to different targets, it needs a technology that can cooperate with laser weapons to carry out material identification of the target at a long distance. The LIBS is the spectrum radiated from the target surface when the high-energy laser acts on the target surface. The characteristic spectrum line in the plasma spectrum is formed by the energy level transition of atoms and ions in the plasma. According to the central wavelength of the characteristic spectrum line, we can determine what elements are contained in the plasma [5-7]. Because the spectrum of plasma excited by different materials is different and the excitation conditions only need high-energy laser, so it is very suitable for material identification as an auxiliary means of laser weapons.

LIBS have many attractive advantages, such as no sample processing, long-distance, fast analysis and being applicable to the three states of solid, liquid and gas at the same time. Since Brech and Cross first proposed laser generated plasma for spectral analysis [8] in 1962, it has been a research hotspot. The team of the Army Research Laboratory (ARL) applied the multivariate analysis method to the spectral data of a variety of samples, such as plastic products and organic explosives, to classify and identify these substances. Jennifer L. et al. established the effect of principal component analysis (PCA) and partial least squares discriminant analysis (PLS-DA) classification model on natural ore
classification [9]. Jagdish P. Singh of the University of Mississippi in the United States used hierarchical cluster analysis (HCA), PLS-DA and artificial neural network (ANN) to classify and identify the spectral data of biological tissues [10]. Narahara Chari of the Massachusetts Institute of Technology and others applied the technology to the identification and classification of drug samples [11], and compared the non-linear method support vector machine (SVM) with two other traditional linear methods of Soft independent modelling cluster analysis (SIMCA) and PLS-DA experimental results. The results show that in terms of model sensitivity, due to the burning of LIBS samples and self-absorption of spectral lines and other factors, the data will show certain nonlinearity. When SVM method is used, the recognition accuracy of test set samples is higher than SIMCA and PLS-DA. At present, most of the researches still use laser-induced breakdown spectroscopy technology for material classification and element recognition in the ideal environment in the laboratory. Because the laser weapon is in a complex outdoor environment and has high real-time requirements, the relevant LIBS classification and recognition technology has not been studied. Based on the complex background and strong noise interference in the actual use environment of laser weapons and the requirements of laser weapons for high real-time LIBS classification, this paper proposes a segmented weighted spectral peak intensity algorithm to construct the spectral vector, which can ensure the recognition accuracy and greatly reduce the time-consuming of recognition and analysis.

2. LIBS principle

Under the irradiation of laser beam, the surface of solid material is heated by absorbing photons, and damaged. Hot electrons escape from the surface to form free electrons. The melted sample is a dense plasma, which contains atoms, molecules, ions, electrons and clusters, and spreads along the normal direction of the solid surface at high speed to form a plasma plume [12,13]. For example, for a 15-50ns pulse width Q-switched ruby laser or a Nd: YAG laser, when the pressure is 760*133.322Pa, the typical breakdown threshold is 10MW / cm². Plasma is a highly ionized state of atoms and molecules, which is the fourth form of matter. In the high temperature and high ionization state of plasma, the emission spectrum of atoms has many new characteristics [14]. These new characteristics can be analysed by the energy level diagram of the plasma. Figure 1 is the energy level diagram of hydrogen like ions under the condition of plasma.

![Energy level diagram of hydrogen like ions.](image)

It can be seen from the diagram that, unlike the normal atom energy level diagram, there is a quasi-continuous region of energy level near the ionization limit of the normal atom. The higher the plasma temperature and the degree of ionization, the more the quasi continuous region expands to the ground state, so that there is no discrete energy level in the whole bound range of electrons. There are three
kinds of radiation in plasma: excitation radiation, bremsstrahlung and compound radiation. The transition of excited radiation (bound to bound) between the bound energy levels of an atom gives a discrete spectrum.

The continuous spectrum measured in the experiment is the sum of bremsstrahlung and compound radiation. When the electron temperature in the plasma is low, the compound radiation becomes significant, because the electron velocity is slow, and it is easy to capture when it meets the ion. The compound radiation power density is proportional to the fourth power of the atomic number. The emission spectrum of laser plasma has a strong continuous background. The transition of electrons in continuous region or between continuous and discrete energy levels constitutes a continuous spectrum. Because the range of continuous transition is very wide, it can be seen from ultraviolet to infrared. However, the influence of continuous background is related to many factors, especially the ambient pressure and plasma temperature.

3. LIBS data recognition algorithm

Support vector machine (SVM) [15], principal component analysis (PCA) [16], partial least squares discriminant analysis (PLS-DA) [17], soft independent modelling cluster analysis (SIMCA) and artificial neural network (ANN) are commonly used for spectral data classification. As mentioned in the introduction, due to the strong nonlinear characteristics of LIBS, the recognition efficiency of SVM is higher than that of PLS-DA and Si MCA: two linear algorithms. At the same time, due to the influence of matrix effect, the effect of PCA on LIBS data classification is not ideal. SVM and ANN are both widely used and high-precision algorithms in the field of LIBS classification. Because of the sparsity of SVM, the performance of SVM is better than ANN in the case of a small number of samples. At the same time, the operation of SVM is less than Ann, which can get the classification results more quickly. Considering the high real-time requirement of laser weapon target system, SVM is used as the core algorithm of LIBS data classification.

The basic model of SVM is to find the best separating hyperplane in the feature space to maximize the positive and negative sample intervals on the training set. SVM is a supervised learning algorithm to solve the binary classification problem. After introducing kernel function, SVM can also be used to solve the nonlinear problem.

![SVM schematic diagram](image)

**Figure 2.** SVM schematic diagram.

As shown in figure 2 is the SVM schematic diagram when the sample vector length is 2. Black and white dots represent the spectral data of two different substances. A straight line needs to be calculated to separate the two types of samples. Because there are a large number of sampling points in each spectrum, it can be regarded as a high-dimensional sample, which requires solving a hyperplane that can separate the two types of samples and the partition interval is required to be the maximum.

The support vector of hyperplane can be expressed as:

$$\omega^T x + b = 0$$  \hspace{1cm} (1)

The geometric interval from the sample point to the hyperplane ($\omega, b$) is:
In order to maximize the interval, the distance from the sample point to the hyperplane is required to be the minimum, that is to solve the following constraint problems:

\[
\begin{align*}
\min_{\omega, b} & \frac{1}{2} ||\omega||^2 \\
\text{s.t.} & \frac{|\omega^T x_i + b|}{||\omega||} \geq y_i, i = 1, ..., N
\end{align*}
\] (3)

The Lagrange multiplier method is used to obtain the Lagrange function \(L(\omega, b, \alpha)\), that is, to add the Lagrange multiplier \(\alpha_i \geq 0\) to each constraint, and transform the objective function into:

\[
\min_{\omega, b} \max_{\alpha} [L(\omega, b, \alpha)] = \min_{\omega, b} \max_{\alpha} \left\{ \frac{1}{2} ||\omega||^2 + \sum_{i=1}^{m} \alpha_i [1 - y_i (\omega^T x_i + b)] \right\}
\] (4)

The dual problem can be obtained by using Lagrange multiplier method. The original problem is transformed into:

\[
\min_{\omega, b} \max_{\alpha} [L(\omega, b, \alpha)] \rightarrow \max_{\alpha} \min_{\omega, b} [L(\omega, b, \alpha)]
\] (5)

The partial derivatives of \(\omega\) and \(b\) are obtained and their values are 0:

\[
\begin{align*}
\frac{\partial L}{\partial \omega} &= \omega - \sum_{i=1}^{m} \alpha_i x_i y_i = 0 \\
\frac{\partial L}{\partial b} &= -\sum_{i=1}^{m} \alpha_i y_i = 0
\end{align*}
\] (6)

It is found that \(\alpha = \sum_{i=1}^{m} \alpha_i x_i y_i\) is substituted into Lagrange function:

\[
L(w, b, \alpha) = \frac{1}{2} w^T w + \sum_{i=1}^{m} \alpha_i \left(1 - y_i (w^T x_i + b)\right)
\]

\[
= \frac{1}{2} \sum_{i=1}^{m} \alpha_i y_i x_i^T \sum_{j=1}^{m} \alpha_j y_j x_j + \sum_{i=1}^{m} \alpha_i \left(1 - y_i \left(\sum_{j=1}^{m} \alpha_j y_j x_j^T x_i + b\right)\right)
\]

\[
= \frac{1}{2} \sum_{i=1}^{m} \sum_{j=1}^{m} \alpha_i \alpha_j y_i y_j x_i^T x_j + \sum_{i=1}^{m} \alpha_i \left(1 - y_i \sum_{j=1}^{m} \alpha_j y_j x_j^T x_i + b\right)
\]

\[
= \sum_{i=1}^{m} \alpha_i - \frac{1}{2} \sum_{i=1}^{m} \sum_{j=1}^{m} \alpha_i \alpha_j y_i y_j x_i^T x_j
\] (7)

When the optimal solution of the original problem satisfies the KKT condition:

\[
\begin{align*}
\alpha_i & \geq 0 \\
y_i (f(x_i) - 1) & \geq 0 \\
\alpha_i (y_i (f(x_i) - 1)) & = 0
\end{align*}
\] (8)

The primal problem and the dual problem have the same solution. Assuming the optimal solution of the dual problem is \(\alpha^*\), the primal problem can be solved as follows:

\[
w^* = \sum_{i \in S} \alpha_i^* y_i x_i
\]

\[
b^* = \frac{1}{|S|} \sum_{s \in S} \left( y_s - \sum_{i=1}^{m} \alpha_i y_i x_i^T x_i \right)
\] (9)

The decision function corresponding to the original problem is:

\[
f(x) = \text{sign}(\omega^* x + b^*)
\] (10)
Equations 1 to 10 are the solution process of SVM algorithm, but the above derivation is based on the assumption that the sample data is linearly separable. However, in practice, the samples are often linear and indivisible, that is, nonlinear problem. In this case, the kernel function $\Phi$ can be introduced to project the sample into a higher-dimensional space to make it linearly separable [18], as shown in figure 3.

![Figure 3. SVM kernel function.](image)

Function $\Phi$ is a mapping from low-dimensional feature space to high-dimensional feature space. If there is function $K(x, z)$, for any low-dimensional feature vector $x$ and $Z$, there are:

$$K(x, z) = \Phi (x) \ast \Phi (z)$$  \hspace{1cm} (11)

Common kernel functions include linear function, polynomial function, radial basis function (RBF) and sigmoid function. Because the spectrum collected by high-precision spectrometer has a high dimension, this paper selects the polynomial function suitable for high-dimensional samples as the kernel function of SVM.

$$\begin{align*}
\text{Linear:} & \quad K(x, z) = x \ast z \\
\text{Polynomial:} & \quad K(x, z) = (y \ast z + r)^d \\
\text{RBF:} & \quad K(x, z) = e^{-\gamma \|x - z\|^2} \\
\text{Sigmoid:} & \quad K(x, z) = \tanh (y \ast z + r)
\end{align*}$$  \hspace{1cm} (12)

It can be seen from the above introduction that the original SVM is a classifier for two-classification, but there are three kinds of spectral samples in this experiment. At this time, the effect of three classification or even multi-classification can be achieved by combining the positive and negative samples of the input SVM model. The common SVM multi-classification modeling methods are divided into one to many modeling (OVA SVMs) and one to one modeling (OVO SVMs).

![Figure 4. SVM one versus many modeling.](image)

Suppose that there are $n$ types of spectral data, as shown in figure 4. One to many modelling means that one type of spectral data is taken as a positive sample while the other $N-1$ types of spectra are combined as a negative sample. In this method, $n$ SVM models are built. When forecasting, spectral data are respectively brought into the decision function of each model to get its type. One to one modelling refers to the establishment of a traditional SVM model for each two types of spectral data. If there are $n$ types of spectral data, then a total of $n \times (n-1)$ SVM models are established. The decision
function results of each model are recorded during prediction, and finally the spectral type is obtained. Because of the high real-time requirement of laser weapon system, this paper uses OVA SVMs with less calculation.

LIBS data of steel plate, carbon fibre and epoxy resin are collected through experiments as shown in figure 5. The wavelength range is from 200 to 900nm, with a total of 7638 sampling points. LIBS data is directly used as the spectrum vector to SVM, and 20 spectra of one type are taken as positive samples, and 20 spectra of the other two types are taken as negative samples to establish SVM model. 60 spectra of three types that are not involved in the modelling are taken for spectrum recognition experiments during recognition, and the recognition accuracy and average recognition time are shown in table 1.

| Target type | Recognition accuracy | Average time consuming (ms) |
|-------------|----------------------|-----------------------------|
| Steel plate | 96.6%                | 3939                        |
| Carbon fibre| 98.3%                | 3774                        |
| Epoxy resin | 100%                 | 4208                        |

From table 1, it can be seen that the sample recognition accuracy is high, in which the steel plate has two spectral sample recognition errors, the carbon fibre has one spectral sample recognition error and all the epoxy resin samples have correct recognition. It takes about 4 seconds to identify a spectral sample.

4. Spectral vector construction method

The high-precision spectrometer can collect more accurate and detailed LIBS peak information, but there will be a lot of useless information in the place where there is no peak. All these useless data will participate in the subsequent SVM modelling and Recognition calculation process, which wastes a lot of time. The recognition time of LIBS samples in about 4 seconds cannot meet the high real-time
requirements of laser weapon system. Therefore, this paper proposes a segmented weighted spectral peak intensity algorithm, which uses the spectral peak information to directly construct the spectral vector. The useless 0 value in the processed spectral data will not participate in the calculation.

4.1. Automatic peak finding and peak type matching

Firstly, it is necessary to extract the spectral peak information from LIBS data. In this paper, symmetrical zero area transformation method is used to automatically find the peak of the processed LIBS data. Symmetrical zero area transform peak seeking is a method of convolution transformation between the symmetrical "window" function with zero area and the spectral data measured by experiments, and threshold processing of the transformed data to obtain the position of the spectral peak. The symmetric zero area window function $C_j$ has the following properties:

$$
\sum_{j=-m}^{m} C_j = 0 \quad \forall \quad C_j = C_{-j}
$$

(13)

Where $W = 2m + 1$ is the window width of the transformation function, if $y$ is the experimental spectrum, the spectral data $y_i$ after symmetrical zero area transformation can be expressed as:

$$
y_i = \sum_{j=-m}^{m} C_j \ast y_{i+j}
$$

(14)

Because of the "0" area property and symmetry property of $C_j$, the convolution transformation of the linear basis and window of the signal will be zero, and the place where there are spectral peaks is not zero (greater than), and the place where the maximum transformation value is $i_{\text{peak}}$ is the spectral peak position. Generally, square wave function, Gauss function, second derivative of Gauss function, cosine square function, Lorentz function and other symmetric functions can be selected as transformation functions. In this paper, Gauss function is used as peak type function as shown in equation 15:

$$
f(j) = \exp \left(-4\ln2\left(\frac{j}{H_g}\right)^2\right)
$$

(15)

Hg is the half height and full width of Gauss line, and the transformation function can be constructed according to formula 16 to meet the condition of formula

$$
C(j) = f(j) - \frac{1}{W} \sum_{-m}^{m} f(j)
$$

(16)

$C(j)$ is the obtained symmetric zero area transformation function, $W = 2m + 1$ is the window width of the transformation function, $f(j)$ is the selected peak type function. Using the transformation function constructed above to transform the processed LIBS data with symmetrical zero area transformation can extract all peak information from the data. However, in the actual environment, the collected LIBS data may contain some large amplitude point noise or other peaks generated by physical mechanism. Symmetrical zero area transformation can only extract peak information from the data but does not have peak information In the mechanism of shape matching, all the peaks generated by plasma excitation radiation need to be screened out from the extracted peaks.

![Figure 6. Linear functions of different peaks.](image-url)
The natural line type of spectral lines is generally Lorentz line type, but the actual spectral line type will change due to some mechanisms [19]. For example, the Doppler broadening caused by the irregular motion of the light-emitting atoms (molecules) in the light source is Gauss type, and the broadening caused by the interaction (collision) between the atoms conducting light absorption (or emission) and the external gas molecules is Lorentz type. In addition, the line shape is also related to the spectrum type, the line diffusion function of the dispersion system of the detection instrument (generally Gauss line shape), the detection environment, etc. Therefore, Voigt function is generally used to describe the spectral line shape. The curve of three linear functions with the same half height and full width is shown in figure 6. Voigt function is the convolution of Lorentz function and Gauss function in form. Its accurate calculation is more complex, and its approximate form is often used in application. Its expression is as follows:

$$Voigt(j) = k \times \frac{Hl}{4j^2 + Hl^2} \times \frac{2}{\pi} \times (1 - k) \times \frac{\sqrt{4ln2}}{\sqrt{Hg}} \times exp\left(-4ln2\left(\frac{1}{Hg}\right)^2\right)$$

(17)

Among them, Hl and Hg are half height and full width of Lorentz line and Gauss line respectively, and K is the scale factor of line outline. In this paper, we use Voigt function fitting to screen the peak shape of the spectrum. We use Voigt function fitting to fit each peak extracted from LIBS data. The ratio of fitting error to Voigt linear area can be less than 0.1%. Then the parameter expression of spectral peak discrimination is:

$$S_{voigt} = \frac{h}{RMSE}$$

(18)

Where h is the peak height of the fitted Voigt function and RMSE is the root mean square error between the fitted peak data and the original data. The size of $S_{voigt}$ depends not only on the information of peak height, but also on the similarity of peak signal and Voigt function. Therefore, for the pseudo peak of non-Voigt line caused by high-frequency noise or other factors, its RMSE is relatively large. Under the condition of the same peak height h, the value of the discrimination parameter $S_{voigt}$ will also be reduced, which can effectively remove the pseudo peak and improve the discrimination rate.

4.2. Segmented weighted peak intensity algorithm

As shown in table 1, although the processed LIBS spectral data is directly used as the sample to establish SVM model, the accuracy is high, but the recognition process is time-consuming. This is because the amount of spectral data collected by high-precision spectrometer is too large. When using SVM for modelling or recognition, the dimension of each sample is 7638, so processing such high-dimensional sample data will inevitably consume a lot of Time. In this paper, based on the information of spectral peaks extracted and screened from LIBS data, a method of spectral vector construction with piecewise weighted spectral peak intensity is proposed.

First of all, the real effective information in LIBS data is the filtered spectral peak information, which mainly includes the spectral peak intensity and the position of the spectral peak. However, the positions and quantities of the spectral peaks contained in LIBS data of different materials are different. If all the spectral peaks are used to construct the spectral vector directly, the length of the spectral vector constructed by different materials will be different. SVM algorithm requires that all samples have the same dimension (vector length). So firstly, the spectral data is divided into several equal length spectral segments, which can ensure that no matter how many peaks there are in different kinds of spectral samples, the length of the final constructed spectral vector is equal. Then, in each segment, the weighted spectral peak intensity of the segment is calculated by using the spectral peak information in the segment as the final value representing a dimension of the segment in the spectral vector. Assuming the spectral data band range [wlstart, wlend] band, for example, each segment p (nm), the constructed spectral vector is:

$$SVMvector = [S_1, S_2, ..., S_{(\text{wlend} - \text{wlstart})/p}]$$

(19)

$$S_i = f(\text{peak}[xi,xi+p]) = \sum_{j=0}^{n} \text{Intensity} (\text{peak}[j]) \times \text{Location} (\text{peak}[j])$$

(20)
Where Si is the weighted peak intensity in the i-th segment. \( f(\text{peak}[xi, xi + p]) \) represents the spectral peak information in the i-th segment, including intensity information and location information, \( \text{Intensity}(\text{peak}[jj]) \) represents the intensity weight of the j-th peak in \([xi, xi + p]\), and \( \text{Location}(\text{peak}[jj]) \) represents the location weight of the j-th peak in \([xi, xi + p]\). Next, we discuss how to construct the spectral peak weight function. The physical mechanism of LIBS needs to be taken into account when constructing the weight function. Due to the instability of the plasma itself, the spectral peak intensity fluctuation in LIBS is a normal phenomenon, which has little reference significance for spectral identification. Different positions of the spectral peaks represent different elements, which has great reference significance for spectral identification. Therefore, the change of peak intensity should only change the peak weight slightly, while the change of peak position should change the peak weight greatly. In conclusion, the following peak intensity weight function and position weight function are proposed in this paper:

\[
\text{Intensity}(\text{peak}[jj]) = \sum_{k=-m/2}^{m/2} \ln (I(k) + 1)
\]

\[
\text{Location}(\text{peak}[jj]) = \sum_{k=-m/2}^{m/2} a * \text{Gauss} \left( \frac{W(k) - xi}{p}, \sigma, \mu \right)
\]

Table 2 shows the accuracy and time-consuming of the SVM based method using the same experimental data as table 1.

| Table 2. | accuracy and time consumption of spectral classification under different segment length, the first number in brackets is the recognition accuracy, and the second number is the average recognition time consumption (ms). |
|-----------|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| Segment (nm) | Steel plate | Carbon fibre | Epoxy resin |
| 1         | (98.3%, 537ms) | (100%, 592ms) | (100%, 694ms) |
| 2         | (98.3%, 492ms) | (98.3%, 501ms) | (100%, 589ms) |
| 3         | (98.3%, 329ms) | (98.3%, 437ms) | (100%, 502ms) |
| 4         | (98.3%, 245ms) | (98.3%, 351ms) | (100%, 421ms) |
| 5         | (98.3%, 163ms) | (98.3%, 219ms) | (100%, 376ms) |
| 8         | (96.6%, 122ms) | (98.3%, 161ms) | (98.3%, 258ms) |
| 10        | (95.0%, 96ms)  | (96.6%, 104ms) | (98.3%, 166ms) |

It can be seen from table 2 that the smaller the segment length is, the longer the average recognition time is, but the higher the accuracy is. When the segment length is 1nm, the recognition accuracy of carbon fibre is even higher than that when the spectral data is directly used as the spectral vector in table 2. Because the length of spectrum vector constructed by segmented weighted peaks is 700 even if the segmented length is 1nm, which is far less than the length 7638 of spectrum data as spectrum vector directly, the average recognition time is one order of magnitude less than that of spectrum data directly. When the segment length is less than 5, the segment weighted peak method can ensure that the recognition accuracy is not lower than that using LIBS data directly.

Because the segmented weighted spectral peak intensity algorithm uses the spectral peak information in LIBS data for weighting operation, its calculation amount is directly related to the number of spectral peaks. Because of the characteristics of segmented calculation, if the distribution of spectral peaks in the whole region is relatively average, each segmented weight value contains more information, which is more conducive to identification calculation. As can be seen from figure 5, LIBS of epoxy resin The data contains the largest number of peaks, and the distribution of peaks is relatively uniform. Therefore, compared with the results of the other two targets, the average time-consuming is the longest, and the recognition accuracy is higher.
5. Conclusion
In this paper the technology of LIBS data fast recognition was analysed, the specific steps of multi classification of LIBS data using SVM has been described. In order to solve the problem of time-consuming in the recognition and classification of LIBS data using SVM algorithm, this paper proposes the algorithm of segmented weighted peak intensity. Firstly, the peak information in LIBS data is extracted and the peak type is screened based on its physical mechanism. Secondly, the weighted intensity of the peak in each segment is calculated by the extracted effective peak information of LIBS data. Compared with using LIBS data as spectrum vector directly, the length and recognition time of each LIBS sample vector are reduced by an order of magnitude without reducing the recognition accuracy. At the same time, because all the effective peaks are used in this method, the recognition accuracy is high. When the segment length is less than 5nm, the accuracy is above 96%. The real-time performance of the system can also be improved by adjusting the segment length according to the requirement of accuracy. This method is very suitable for assisting laser weapons to identify and classify targets quickly and accurately.

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