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Supporting Information

for

Exploring combinations of spatial laser beam shaping for advancing nanoparticle synthesis by ablation in liquids

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Data processing details

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Details of size distribution fitting procedure and calculated parameters

This Supporting Information describes the statistical analysis and fitting procedure used for the nanoparticle size distributions. Particle diameters were extracted from SEM images using ImageJ, yielding for each sample a list of individual particle sizes, hereafter referred to as “data” throughout this section. The analysis was performed in two steps. First, the median, median absolute deviation (MAD), and robust coefficient of variation (RCV) were calculated directly from the experimental particle-size data. Second, the particle-size distributions were fitted with lognormal functions to determine the fitted median and mode. A single lognormal distribution was used for HEA and FeNi NPs, whereas a two-component lognormal model was used for the bimodal Au NP distributions to determine the characteristics and relative fractions of the two populations.

- *Direct statistical analysis of data*

The statistical parameters were calculated directly from the raw particle-size data using Python. The commands used are summarized below.

Parameter	**Python command**
Median (m)	numpy.median(data)
Median absolute deviation (MAD)	scipy.stats.median_abs_deviation(data)
Mean(d)	numpy.mean(data)
Standard deviation (std)	numpy.std(data)

The median is less sensitive to outliers than the mean and was therefore considered as the most representative descriptor of the nanoparticle diameter. This is particularly relevant for lognormal distributions where the mean can be strongly influenced by sporadic measured nanopaticles located in the tails of the distribution. The width of the distribution was then characterized using the median absolute deviation (MAD). For comparison between samples, the robust coefficient of variation (RCV) was calculated as:

$$RCV = \frac{MAD}{m}$$

Although some studies use polydispersity index (std²/mean²), the RCV was preferred because it is consistent with the use of the median. The MAD values reported in Table 3 and Figures 2–4 of the manuscript, and the RCV values reported in Figures 2–4 and 9, were calculated directly from the experimental particle-size data. This approach facilitates comparison between samples regardless of the fitting model used.

- *Selection of the fitting model*

Based on the calculated parameters, theoretical Gaussian distributions and lognormal distributions were calculated using their analytical expressions:

$$\text{Gaussian}: G(x) = \frac{1}{Std \times \sqrt{2\pi}} e^{-\frac{1}{2} \times \left(\frac{x-d}{std}\right)^2}$$

$$\text{Lognormal}: N(x) = \frac{1}{x\sigma\sqrt{2\pi}} e^{\left(-\frac{1}{2}\left(\frac{\ln x - \mu}{\sigma}\right)^2\right)}$$

where *x* is the nanoparticle diameter, μ and σ are the mean and the standard deviation.

Histograms were constructed from the experimental data and normalized by the total bin area in order to obtain probability density values. The resulting histograms were compared with the calculated Gaussian and log-normal distributions (Figure S1).

The log-normal model showed better agreement with the experimental diameter distributions than the Gaussian model. Consequently, only log-normal fitting results are discussed in the following sections. Additionally, the log-normal distribution obtained from histogram fitting was compared with the distribution calculated directly from the raw data to confirm the suitability of the model for describing the experimental data.

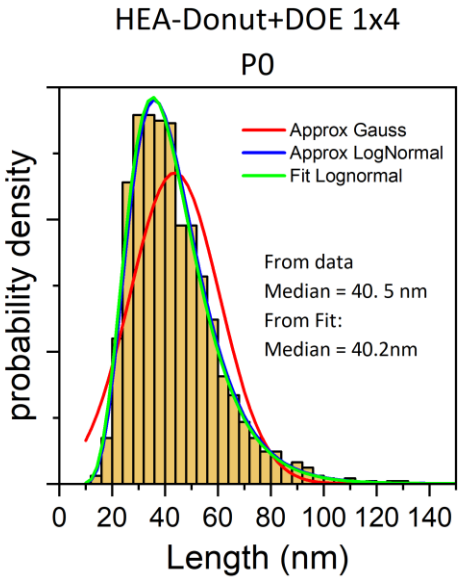


Figure S1: Selected NP size distribution histogram and comparison with calculated Gaussian and lognormal distributions and the fitted lognormal model.

- *Lognormal fitting procedure*

In the case of Au NPs, the fit of the histogram with a single lognormal distribution differs from the distribution calculated from the raw data. This is attributed to the presence of a bimodal distribution. Therefore the fitting procedure was extend to a bimodal lognormal model:

Bimodal Log-Normal Distribution

$$f(x) = \frac{C}{x\sigma\sqrt{2\pi}}\exp\left(-\frac{1}{2}\left(\frac{\ln x - \mu}{\sigma}\right)^2\right) + \frac{(1-C)}{x\sigma_2\sqrt{2\pi}}\exp\left(-\frac{1}{2}\left(\frac{\ln x - (\mu + \Delta\mu)}{\sigma_2}\right)^2\right)$$

Parameters:

C — mixing weight ($0 \leq C \leq 1$)

μ — mean of the log of the first component

$\Delta\mu$ — shift in mean for the second component ($\Delta\mu > 0$)

σ — standard deviation of the first component

σ_2 — standard deviation of the second component

For a unimodal distribution C = 1 (fixed). Once the fitting is completed, the median is calculated using:

$$m_{fit} = e^{\mu_{fit}}$$

The fitted median values reported in Table 3 and Figure 8 of the main manuscript were obtained from the log-normal fits, whereas MAD and RCV (Figure 9) were calculated directly from the experimental particle-size data. For the bimodal Au NP distributions, the reported fitted median values correspond to the dominant small-particle population, while the mixing parameter C describes its relative fraction. Figure S2 summarizes the obtained statistical data and their use throughout the article.

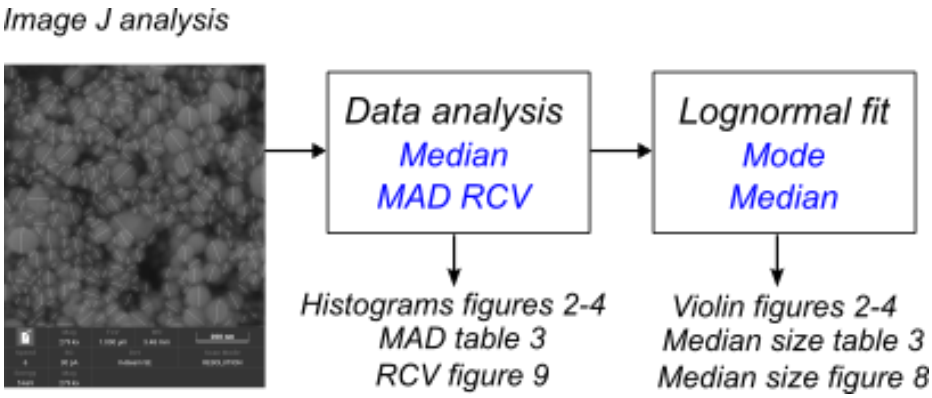


Figure S2: Summary of the statistical analysis and reported values.