

Toward reliable laser-induced breakdown spectroscopy analysis under practical physical constraints

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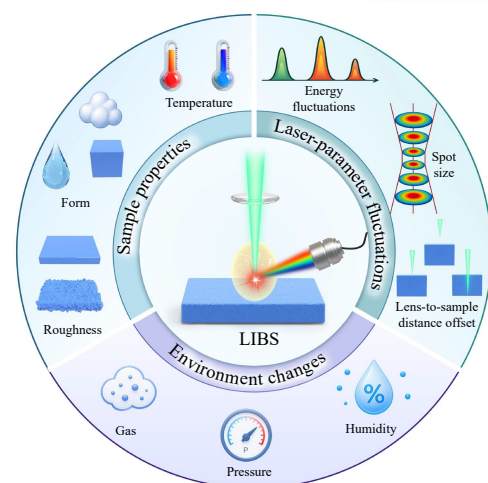
Received August 27, 2026; accepted September 4, 2026

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ABSTRACT

Laser-induced breakdown spectroscopy (LIBS) serving as a representative in-situ elemental technique, is widely recognized for its fast response and multi-element detection capability. Nevertheless, its performance is still prone to being affected by versatile realistic physical constraints. Variations in laser parameters, sample properties, matrix composition, ambient gas and pressure can all perturb laser ablation efficiency and plasma's spatiotemporal evolution, leading to signal fluctuations and aggravated self-absorption and matrix effects. These issues consequently compromise the repeatability of spectral signals, the accuracy of analytical results as well as the robustness of analytical methods. This perspective discusses two complementary strategies toward reliable LIBS analysis. At the hardware level, spatial confinement, energy injection and light-field control are leveraged to steer plasma evolution and upgrade raw spectral quality; and at the software level, spectral pre-processing, knowledge-driven algorithms, data-driven models and knowledge-data hybrid frameworks are employed to minimize analytical errors and boost model generalization. The LIBS reliability improvement is expected to shift from single optimization to hardware-software synergetic optimization and adaptive parameter adjustment, opening up new avenues for robust LIBS analysis under real-world physical constraints.

Keywords LIBS, plasma modulation, data-driven modeling, physical constraints



Due to its ability of rapid, multi-element and in-situ analysis, laser-induced breakdown spectroscopy (LIBS) has shown considerable applications in industrial-quality control, geological exploration and even in the extreme-environment monitoring [1, 2]. As illustrated in the middle of Fig. 1, this technique uses high-energy laser pulses to ablate samples along with the

generation of a laser-induced plasma. By collecting the characteristic spectra from atoms and ions we can obtain the elemental information. In general, qualitative LIBS typically relies on the measured spectral-line positions, intensities, or full-spectrum features to identify elements and classify materials. While quantitative analysis depends on standard samples to establish a calibration



relationship between spectral features (e.g., spectral-line intensity or peak area) and the elemental concentration. Besides, some calibration-free and data-driven approaches have also been developed recently [3, 4].

Nevertheless, LIBS still faces many practical constraints, covering from spectral information collecting to reliable analytical results, as shown in Fig. 2. The analysis reliability mainly presents the repeatability of spectral signals, the accuracy of analytical results and the robustness of analytical methods under fluctuated experimental conditions. Since the plasma intrinsically suffers from the highly transient and spatially nonuniform nature, its evolution is readily influenced by the practical physical constraints coming from laser parameter fluctuations, variations of samples and matrix composition, changes in ambient gas and pressure [5, 6]. These factors can alter the laser ablation efficiency and the plasma's spatiotemporal evolution, which in turn cause fluctuations in spectral intensity and baseline, thereby reducing the signal repeatability. Besides, they may also exacerbate self-absorption and matrix effects, distorting the relationship between spectral features and analytical targets (e.g., elemental concentrations and identities) and thus leading to a loss of analytical accuracy [7, 8]. Moreover, broadening and distortion of spectra would complicate the elemental identification in complex samples. When the external conditions change, this original relationship will move degrading the robustness of analytical methods [9, 10]. In present days the major challenges toward reliable LIBS analysis therefore come from practical physical constraints and the induced signal quality problems.

To mitigate the impact of physical constraints on LIBS, one typical method is the hardware control which utilizes the ways of active modulation for laser-ablation and plasma evolution, in order to improve the spectra stability and controllability [11]. Based on their different mechanisms these strategies can be broadly classified into spatial confinement, energy injection, and light-field modulation, as well as the hybrid modulation, as shown in Fig. 1(A). Spatial confinement usually employs cavities or magnetic fields to restrict the plasma expansion which can regulate its temperature, density and spatial distribution and improve the raw spectrum quality [13]. Energy injection may take the form of double-pulse excitation, which enhances energy coupling between the laser and the sample hence improving the signal intensity and stability [14, 15]. Light-field modulation focuses on optimizing the conditions of the incident lasers, e.g., via the way of beam shaping to improve the uniformity of energy distribution and ablation, thereby enhancing the spectrum repeatability [12]. Remarkably, one may choose the hybrid modulation by combining different approaches, such as using the microwave-assisted coupling in a cavity that significantly prolongs the plasma lifetime while simultaneously reducing the signal fluctuations [16, 17]. Recent papers in *Frontiers of Physics* have intensively focused on the role of these methods in improving the stability and controllability of

spectral signals. Xin *et al.* [15] used remote dual-pulse excitation to analyze magnesium alloy melts. They improved the signals by optimizing the ablation efficiency and plasma's spatiotemporal evolution processes. Zhao *et al.* [12] systematically reviewed versatile light-field modulation methods, importantly focusing on the effects of beam shaping and polarization control techniques. Khumaeni *et al.* [17] discovered the importance of microwave-assisted coupling which explicitly prolonged the plasma lifetime and enhanced the emission signals. Overall, an active plasma modulation from the hardware-control layer can significantly improve the quality of raw LIBS spectra by improving the energy coupling and ablation uniformity, as well as suppressing the excessive plasma expansion. However, its effectiveness remains limited under practical physical constraints since the alleviation for self-absorption and matrix effects is often insufficient and further corrections with spectral mathematical modeling etc. are required. Hardware-based approaches alone, therefore, can not ensure the reliability of real LIBS analysis.

In parallel, utilizing ways of spectral pre-processing and modeling from the software level can further enhance the LIBS reliability [8, 10]. As displayed by Fig. 1(B), the main strategies include spectral pre-processing, knowledge-driven modeling, data-driven modeling, and knowledge-data hybrid modeling. Here, spectral pre-processing methods such as normalization, background subtraction, self-absorption correction can reduce signal fluctuations, baseline drifts and spectral shape distortions in the raw LIBS spectra [18, 19]. The calibration-free knowledge-driven methods are able to accurately estimate the elemental concentration from the plasma temperature and electron density depending on the assumptions of local thermodynamic equilibrium and optical thinness, which hence reduces the reliance on calibration standards [19, 20]. The data-driven methods can well compensate for the deviations caused by self-absorption and matrix effects, through the learning of relationship between spectral features and analytical targets under different experimental conditions, thus enhancing the analytical accuracy [9, 10, 21]. Most importantly, the newly-developed knowledge-data hybrid modeling incorporates the physical knowledge into data-driven models. When combined with the way of transfer learning it can iteratively update the old model by using a small amount of data from new conditions significantly improving its generalization [22, 23]. Recent researches published in *Frontiers of Physics* highlight the significant advances in this area uncovering the software-based methods in improving actual LIBS reliability. Guo *et al.* [8] systematically reviewed data-processing methods in LIBS discussing the critical role of spectral pre-processing for signal stabilization and ways for compensating the self-absorption and matrix effects. Ni *et al.* [24] proposed an image-assisted normalization method which effectively reduced the signal variations caused by laser-energy fluctuations and sample hetero-

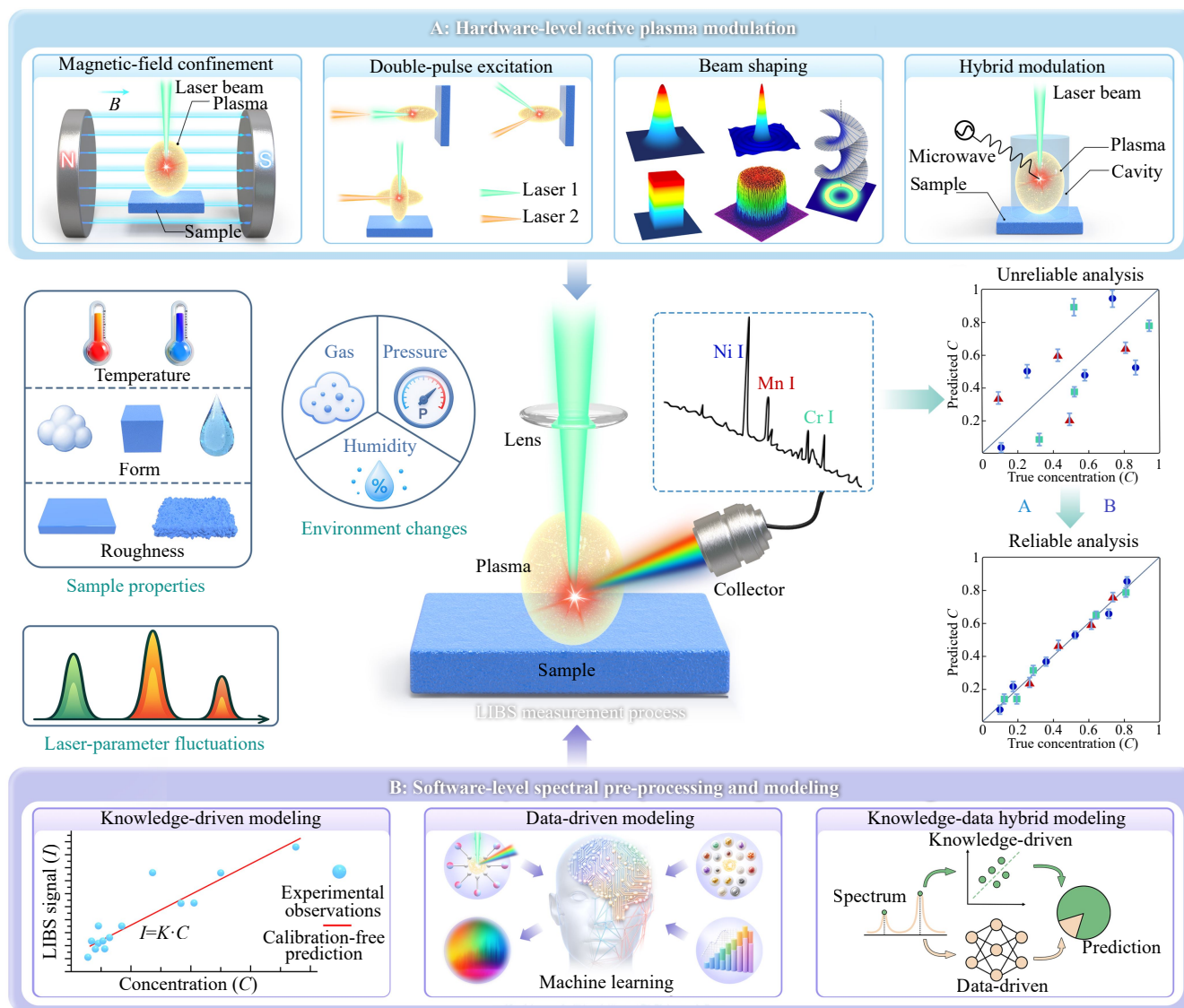


Fig. 1 Hardware-level (A) and software-level (B) routes for improving the reliability of LIBS analysis under practical physical constraints. Reproduced from Refs. [10–12].

geneity resulting in an improved analytical reliability. Hao *et al.* [10] deeply discussed the potential applications of machine learning in LIBS, presented the particular strength of hybrid modeling and transfer learning for mitigating the matrix effects, signal fluctuations and improving the model generalization. It is worth noting that, although the above software-level approaches could improve the final results the ultimate LIBS performance still relies on the raw spectra quality detected experimentally. If the experimental conditions reveal substantial fluctuations, the robustness of subsequent pre-processing and modeling may be reduced considerably. As a result, only software-based approaches remains incapable of ensuring the reliability of LIBS [25].

To conclude, practical physical constraints will affect the LIBS reliability by altering the efficiency of laser ablation and the uniformity of plasma spatiotemporal evolution, which gives rise to signal quality issues and ultimately leads to reduced spectral repeatability, limited analytical accuracy, and weakened methodological robustness. To address these issues, existing studies have proposed the corresponding solutions from two layers of hardware control and software analysis (see Fig. 2) [8, 10–12, 26]. The former improves raw spectral quality by using spatial confinement, energy injection, light-field modulation and so on. And the latter can in turn use the way of spectral pre-processing and modeling for enhancing the model generalization across versatile

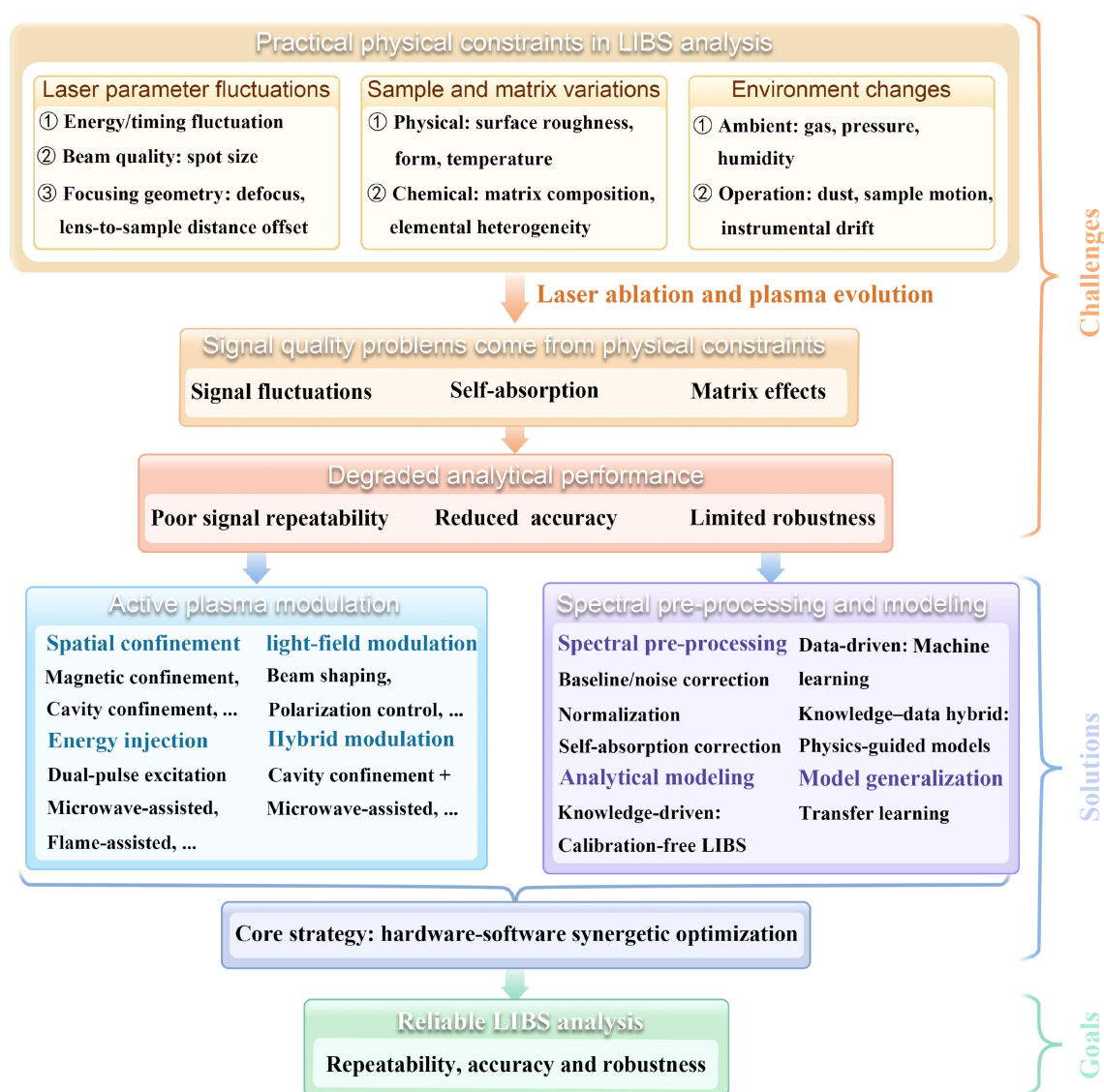


Fig. 2 Challenges and solutions framework for reliable LIBS analysis in which the core strategy is using hardware-software synergetic optimization.

scenarios. Nevertheless, neither hardware nor software can singly struggle to simultaneously balance signal stability, analytical accuracy, and method robustness. In the future, the improvement of LIBS reliability is expected to evolve toward hardware-software synergetic optimization accompanied by real-time feedback of spectra and plasma information, which can in turn adaptively tune lasers or environment parameters [25, 27, 28]. This approach will offer a more promising direction for achieving more stable and reliable LIBS outcomes under realistic physical constraints.

Declarations The authors declare that they have no competing interests and there are no conflicts.

Acknowledgements We acknowledge financial support from the National Natural Science Foundation of China under Grant Nos.

12674604, 12174106, 11474094, and 12474070, the Natural Science Foundation of Chongqing under Grant No. CSTB2024NSCQMSX1117, the Shanghai Science and Technology Innovation Project under Grant No. 24LZ1400600, and the Science and Technology Commission of Shanghai Municipality under Grant No. 18ZR1412800.

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