Fully Automated Data Acquisition for Laser Production Cyber-Physical System

Yohei Kobayashi [®], Takashi Takahashi, Tomoharu Nakazato, Haruyuki Sakurai, Hiroharu Tamaru, Kenichi L. Ishikawa, Kazuyuki Sakaue, and Shuntaro Tani

(Invited Paper)

Abstract—The many tunable parameters involved in laser processing, such as wavelength, pulse duration, pulse energy, and scan speed, not to mention various other complicating factors on the material side, makes it practically impossible to reliably find an optimized parameter set to realize a specific processing target. Currently, an acceptable parameter set is mainly found by tapping the experience and intuition of skilled people within the present production system. However, such methods do not scale to the masscustomization needs of the coming super-smart society, and it has become critical to develop ways to transfer such human experience and intuition to a more scalable setting: namely, the cyber-space. A major challenge in developing a cyber-space solution has been augmenting the limited experimental and theoretical insights of the laser processing phenomenon to the specific problems at hand. Here, we focus on automated data acquisition systems coupled with artificial intelligence (AI) methods to overcome this technological gap. We propose ways to realize cyber-physical systems specializing in specific facets of laser production by showing experimental

Manuscript received February 1, 2021; revised April 13, 2021; accepted April 14, 2021. Date of publication April 22, 2021; date of current version May 26, 2021. This work was supported in part by the New Energy and Industrial Technology Development Organization (NEDO) project "Development of advanced laser processing with intelligence based on high-brightness and high-efficiency laser technologies (TACMI project)", Council for Science, Technology and Innovation (CSTI), Cross-ministerial Strategic Innovation Promotion Program (SIP), "Photonics and Quantum Technology for Society 5.0" (Funding agency: QST), MEXT Quantum Leap Flagship Program (MEXT Q-LEAP) Grant Number JPMXS0118067246, and in part by the Center of Innovation Program from Japan Science and Technology Agency. (*Corresponding author: Yohei Kobayashi.*)

Yohei Kobayashi is with the Institute for Solid State Physics, the University of Tokyo, Kashiwa 277-8581, Japan, and also with the Research Institute for Photon Science and Laser Technology, the University of Tokyo (UTripl), Bunkyo 113-0033, Japan. (e-mail: yohei@ issp.u-tokyo.ac.jp).

Takashi Takahashi, Tomoharu Nakazato, Haruyuki Sakurai, and Shuntaro Tani are with the Institute for Solid State Physics, the University of Tokyo, Kashiwa 277-8581, Japan (e-mail: taka-taka@issp.u-tokyo.ac.jp; nakazatot@issp.u-tokyo.ac.jp; h.sakurai@issp.u-tokyo.ac.jp; stani@issp.u-tokyo.ac.jp).

Hiroharu Tamaru is with the Graduate School of Science, the University of Tokyo, Bunkyo 113-0033, Japan, and also with the Research Institute for Photon Science and Laser Technology, the University of Tokyo (UTripl), Bunkyo 113-0033, Japan (e-mail: tamaru@psc.t.u-tokyo.ac.jp).

Kenichi L. Ishikawa is with the Department of Nuclear Engineering and Management and Photon Science Center, Graduate School of Engineering, The University of Tokyo, Bunkyo-ku 113-8656, Japan, and also with the Research Institute for Photon Science and Laser Technology, the University of Tokyo (UTripl), Bunkyo 113-0033, Japan (e-mail: ishiken@n.t.u-tokyo.ac.jp).

Kazuyuki Sakaue is with the Photon Science Center, Graduate School of Engineering,the University of Tokyo, Bunkyo 113-8656, Japan (e-mail: ksakaue@psc.t.u-tokyo.ac.jp).

Color versions of one or more figures in this article are available at https: //doi.org/10.1109/JSTQE.2021.3074516.

Digital Object Identifier 10.1109/JSTQE.2021.3074516

results from four kinds of automated data acquisition systems. We lastly discuss such methods in context as an important first step to creating an AI based cyber-physical simulator.

Index Terms—Cyber-physical systems, Fiber lasers, Laser ablation, Machine learning.

I. INTRODUCTION

THE WORLDWIDE market for laser manufacturing is growing at approximately twice the rate of the conventional machining market. Such growth reflects how the laser is a promising candidate to realize multi-functional processing, including cutting, drilling, welding, and polishing, all through the proper selection of laser parameters. Moreover, laser production systems have a high affinity to digital production since laser processing systems are almost universally compatible with external electrical control. Laser manufacturing is thus poised to become a key tool to realize the mass-customized production outlined in prominent high-tech future-society strategies, such as Industry 4.0 or Society 5.0 [1], [2].

Our concept for a cyber-physical system of the laser production workflow is depicted in Fig. 1. In the coming super-smart society (or Society 5.0), personalized manufacturing is believed to be a key element to realizing an inclusive society. Such mass customization can only be realized through a highly digitalized industrialization scheme, such as the Industry 4.0 scheme. Personalized orders are sent to a main server in cyber space, which then chooses which machines, and what processes/parameters should be applied for each step in the production chain.

While a cyber-physical laser processing system is certainly attractive, there are some notable obstacles to its implementation. Most critically, it is still difficult to reliably predict and control laser processing across a wide range of systems. There are too many combinations of processing parameters, such as wavelength, pulse duration, repetition rate, average power, fluence, intensity, scanning speed, and scanning pattern, to allow for an exhaustive study of all their effects. This is further complicated by the fact that an appropriate parameter set is also dependent on the specifics of the material. Consequently, processing parameters are instead often selected by the experience and intuition of capable craftsmen in a factory setting. While this may be a viable solution for standardized and streamlined production processes, it is quickly overwhelmed when one tries to up-scale

This work is licensed under a Creative Commons Attribution 4.0 License. For more information, see https://creativecommons.org/licenses/by/4.0/



Fig. 1. Laser-based fabrication chain assisted by cyber-physical system. MDG, Meister Data Generator; TGV, Through glass via.



Fig. 2. Physics involved in a laser drilling or cutting process. Spatial scale is from angstrom to mm, and time scale is from femtosecond to millisecond.

to mass-customized production, as manual finding of correct parameters is too time and labor intensive to address individually. More related to the current discussion, such intuition is hard to engrain into a cyber-physical system as envisioned in Fig. 1.

A promising approach to overcome this hurdle and allow for the effective scaling of laser manufacturing to mass-customized production is to transfer the experience and intuition of such craftsmen to cyber space. Here, personal experience would correspond to information in a database, while intuition would be modeled by a simulator. An ordering party could then simply request some item via 3-D CAD data, and the database and/or simulator could then work to find the optimum parameters to realize this order. The scalability of this system would depend on digital storage space and computation power, both of which have continued their decades-long trend of consistent increase.

The most straightforward approach to realize the above is to create a physics theory-based simulator. In fact, first-principles calculations and databases of the results of these calculations have become powerful tools in the field of materials synthesis, where such methods have been utilized to develop thermoelectric materials and lithium-ion battery materials. Unfortunately, this remains a difficult approach for laser processing as many of the underlying physical processes are still under debate. As an example, we illustrate the various complex processes involved in laser drilling or cutting. Fig. 2 shows a schematic of the pulsed-laser drilling process in time and space.

At first, a laser pulse hits the surface, and electrons are excited in the material; a semiconducting or dielectric material is shown in the figure. While this process is relatively well known in the perturbative regime as the light-matter interaction and electron excitation process have been studied intensively, nonperturbative interactions are not as well-known. For example, high-intensity optical light-matter interaction are still debated in the context of solid-state high-harmonic generation [3]. Additionally, there remains big differences in electron excitation probabilities predicted by relatively well-established theories, e.g., Keldysh equation, and experiment [4]. In the case of metals, energy transfer from light to electrons is treated as an inverse bremsstrahlung process; however, even here it remains that this is a simplified treatment ignoring the complicated feature of coherence of electrons in materials. Real-time first-principles computation [5]–[9] such as the time-dependent density-functional theory and the time-dependent density-matrix method is a powerful approach to accurately simulate highly nonlinear, nonequilibrium electron dynamics and evaluate the energy transfer from laser to the electronic system. It still remains, however, a challenge to take proper account of electron correlation.

Next, the general picture is that scattering processes redistributes energy within the various material subsystems. Electronelectron scattering redistributes the electron energy distribution to a thermal distribution in 10 to 100 femtoseconds, while electron-phonon scattering makes optical and acoustic phonons in a time scale of 100 femtoseconds to 100 picoseconds. Phononphonon scattering makes heat within nanoseconds, which then diffuses in accordance with classical heat-diffusion equations. In these processes, the scattering strength or the lifetime of each step are studied by experiments such as time-resolved reflectivity measurement, time-resolved photoemission spectroscopy, and ultra-short pulse x-ray diffractions [10]–[21]. The obtained time constants of these energy transfers still have large deviations in reported values.

At more macroscopic scales, the consequences of the aforementioned energy influx occur. First, phase explosion might occur, followed by more gradual melting, evaporation, or other modification of the material surface. For these processes, a molecular dynamics calculation is often used to investigate dynamics. The simulation range is often limited however, as the number of atoms for the calculation is typically limited to 10^8 by computational resources [22]–[24], which is much smaller than ablated atom numbers by one pulse (10^{13}). First principles calculation is another approach to understand laser processing, and it has been successful in explaining, for example, the fluence threshold for gentle ablation of copper [25]. However, the typical numbers of atoms in first principles calculation are much smaller than that of molecular dynamics calculations, and thus suffers from the same scale limitations.

Lastly, we must note that laser processing is most often conducted with multiple pulses. After the morphology change from previous pulses, subsequent pulses propagate through the complex shape of the hole, altering their spatial profiles. The scattering effects of laser-generated plasma and debris also may play a role.

Altogether, it is apparent that the laser processing contains multi-scale and complex physics with characteristic timescales ranging in the femtosecond to millisecond and in space from angstrom to mm scale. There is still much research required to realize a laser processing simulation only relying on physical theories to find optimized parameters.



Fig. 3. Schematic of the progress in how to find an optimized parameter set for some laser processing goal. The vertical axis shows the parameter finding speed or quality.

In lieu of a purely theoretical approach, an artificial intelligence (AI) approach, such as deep learning, could be effective in realizing a near-term practical simulator. A high-quality and large dataset could allow for the effective use of neural networks to simulate important trends in laser processing. Fig. 3 shows how the effectiveness of parameter finding methods may progress in ten or twenty years. A "physical" AI solution would play an important role in bridging the divide between a stagnating "experience and intuition" approach and a not-yet-mature purely theoretical approach. In fact, this kind of computer-based optimization approach has recently been the subject of intense research and development in a wide range of fields, including chemical and material synthesis [26]–[28]. By linking it with robotic automation, a shift to full automation of tasks that used to require manual labor has received much attention [29]–[31].

The success of such an AI-based approach is highly dependent on the quality and quantity of data available. An AI can only learn from dependencies which are present in the data. Furthermore, a sufficient number of data points, typically in the range of a few thousand to a few tens of thousands, are required to allow for AI approaches to derive non-data-specific conclusions. We note that such dataset characteristics have traditionally been difficult to realize with laser processing. The highly non-linear nature of the processing has made its control difficult, while its irreversible nature has made it difficult to gather a large amount of data. Moreover, the destructive nature of most laser processing methods has restricted both data quantity and quality. Nonetheless, while perhaps not reasonable before, developments in light source quality and automation technologies have made such large-scale data acquisition more feasible in recent years.

Fig. 4 shows our conceived generalized data acquisition loop. Here, needs or demands are shown coming in from the left. Laser parameters such as wavelength, pulse duration, fluence are chosen at first. The machining parameters, such as scanning speed pattern, are selected next. After processing, the obtained shape and quality are observed through some measurement equipment, like a 3-D microscopy or a scanning electron microscope (SEM). Measured data are extracted to a database, after which the next round of processing and measurement starts with a different set of parameters. After many round trips, the best parameter set is output. Assuming that one iteration takes a few tens of minutes



Fig. 4. Schematic of the data acquisition circulation for processing recipe finding. Automated circulation could enable the acquisition of enough data for DNN construction in order to realize an effective simulator.

under manual operation, one to ten circulation per day would be a realistic number of loops for human operation. It can easily be seen that for deep learning approaches, all automated circulation is critical to achieve the 1,000 to 10,000 circulations required.

Once a desired deep neural network (DNN) for some specific processing is constructed, the DNN functions as a simulator without any additional physical processing circulation. The DNN can give inferences for different parameters within seconds, and one could obtain promising processing parameters in realistic time. We note that this is also advantageous for theoretical studies. Although a DNN does not tell us directly the correct physics, numerous "right solutions" work as a high-quality cyber-experimental tool for theorists. Such AI knowledge could serve as a backbone for the cyber-physical feedback required in true cyber-physical systems, as envisioned in Fig. 1. The major obstacle here becomes the development of the automated, high-quality data acquisition systems required to drive this approach.

In this paper, we demonstrate automated, high-quality data acquisition systems for laser processing, where we show four kinds of different monitoring methods focusing on different aspects of laser processing. An automated compressor system coupled with plasma monitoring via photodiode allowed for the precise measurement of the pulse-duration dependent damage threshold with thousands of data points. An automated SEM measurement system was demonstrated to acquire high-quality and high-quantity SEM data, where we utilize this system to study percussion drilling at different laser parameters. An automated 3-D interferometric microscope measurement system was constructed to observe pulse-to-pulse surface morphology change during laser grooving. An in situ detection system of shot-by-shot surface scattering pattern from a processed surface by using a high-speed camera was coupled with a DNN to allow for in-line prediction of ablated hole depths. Finally, we discuss how such systems will be critical for creating a cyber-physical system for laser manufacturing.



Fig. 5. Schematic of the experimental setup to find the pulse-duration dependent damage threshold precisely.

II. EXPERIMENTS

In this section, four kinds of automated data acquisition systems, each using a photo diode, a scanning electron microscope, a 3-D microscope, and a high-speed camera, respectively, are described.

A. High-Precision and Automated Damage Threshold Measurement System

The laser induced damage threshold is an important parameter to characterize how easily materials are machined by the laser. Furthermore, the pulse-duration dependent threshold should also yield information about how the laser energy couples and dissipates within the material. However, the damage threshold is often difficult to measure precisely, as it is sensitive to changes in laser or material parameters. This is related to the fact that the energy transfer from light to electrons contains highly nonlinear processes. Correspondingly, small changes in experimental conditions are easily amplified, and the damage threshold may differ even for the "same" experiment. To increase the precision of the most likelihood value of the damage threshold, the number of measurements becomes critical.

To measure the damage threshold of materials precisely over a large range of pulse-durations, we constructed an automated threshold detection setup complete with mechanical pulse energy and pulse duration control. The setup is shown in Fig. 5. A home-made, 1-MHz repetition rate, Yb-doped fiber laser system was utilized as the light source. The pulse duration could be varied from 0.5 ps to 30 ps by an external compressor, and the fluence could be changed through an automated half waveplate and polarized beam splitter. More details of the experimental setup can be found in reference [32].

Laser pulses from this setup were illuminated onto a Cu plate. In order to detect if the material was damaged or not under a certain illumination pulse-duration/fluence combination, a photodiode was utilized to detect the plasma emission from the material surface. Fig. 6 shows the image of the damaged spots taken by a microscope (a) and the corresponding photodiode signal at each spot (b). When the material is damaged, it can be seen that a photoemission signal is observed with exact one-by-one correspondence. By using this photodiode observation, this system could be programmed to adaptively explore



Fig. 6. (a) Damage photo of Cu surface observed by optical microscope. Black dots correspond to the damaged points. (b) Plasma emission measured by a photodiode. Plasma emission are observed only when the material is damaged.

fluence ranges only near the damage threshold, thus obtaining numerous useful data around the threshold. The automated and adaptive nature of our setup allowed us to evaluate the threshold with a precision of around 1%. Note that this value should be distinguished from absolute accuracy, as this value could change in accordance with the surface roughness, purity, focus area, and pulse shape. With the help of this technique, 10,000 data could be acquired in 8 hours.

B. Automated SEM Measurements

To optimize processing parameters in a cyber-physical system with a machine learning approach, high-quality data is essential. Here, quantitative data with high reproducibility and traceability is referred to as "high-quality". While a scanning-electron microscope (SEM) plays a major role in qualitative evaluation of ablation features, the obtained data are undoubtedly high-quality in the above sense as well. SEM images are particularly useful in evaluating the qualities of laser processing not readily observed by optical microscopes, for example, evidencing the presence (or not) of thermal damage by observing melt-like features. Hence, quantitative evaluation of electron microscope images may become key to optimizing such qualities. Thanks to the recent advances in deep-learning technology, images can be routinely quantified into feature vectors, which can then be used for more specific tasks, such as image recognition, segmentation, and feature extraction. In exchange for such powerful analysis capabilities, deep learning requires a large amount of data. However, with a few exceptions in semiconductor fabrication plants, data acquisition using SEMs has relied on manual operation, and even skilled operators typically collect only a few hundred images per day.

Here, we have modified an electron microscope and developed a measurement system that can automatically acquire data at a maximum of 300 images per hour. We created a structured data format for describing the procedures of processing and measurement, thereby linking the operation of a laser processing machines to the electron microscope. In this way, the electron microscope automatically measures a laser-processed area. The measured image is tied to the processing parameters and automatically uploaded to a central database. An example of measurement for laser-drilling parameter search using this system is shown in Fig. 7. The pulse energy, repetition rate, focal position, and number of irradiation pulses are each varied



Fig. 7. A SEM picture for 10,000 processing. Automated laser processing and SEM system acquires this data in 5 hours.

in 10 conditions for a total of 10,000 different combinations of parameters. Conventionally, the measurement of electron microscopy images of such a large number of processing points has been limited to acquiring only a small portion of typical data. By using a fully automated electron microscopy measurement system, we are able to acquire all 10,000 data points in a form that is linked to the processing parameters, and that can be easily analyzed using deep learning.

C. Automated 3-D Microscope Measurement

The pulse-to-pulse morphology change is not only of great interest to the design of processing strategies, but also to fundamental studies on how intermediate states affect the final processing outcome. A 3-D microscope is a powerful tool to evaluate laser processed surfaces. The vertical resolution can be as good as 10 nm, which is comparable to the one-pulse gentle ablation rate using an ultrashort pulse. Here, again, the small working distances of microscopy systems make *in situ* processing impractical, while manual sample transfer takes non-trivial time in mounting/unmounting and alignment.

To overcome these difficulties, we have constructed an automated transfer system between a laser shooting stage and a 3-D microscope [33]. Lateral repeatability of the position here was as well as 0.1μ m, which was well below the spatial resolution of the microscope. Fig. 8 shows how the spatial shape of a Cu target changes with successive pulse irradiation. In this condition, the first pulse can be seen to not make a hole, but a bump. Only with subsequent pulses is a hole formed. Such differing actions of the first and second pulse were only visible due to the pulse-by-pulse nature of our measurement scheme. Here, roughly one data is obtained in one minute; correspondingly, a total of 1,000 data could be obtained in a day. We have demonstrated the fabrication of a deep-learning-based 3-D morphology simulator using such data [34].

D. High-Speed Camera Observation for In-Situ Monitoring With Help of DNN

Real-time monitoring of the laser manufacturing process is highly demanded in industry. Information regarding how deep a hole or groove is during processing could be used for feedback to realize a desired shape. A light scattering pattern from the



Fig. 8. A 3-D data of shot-to-shot morphology. This data could be used to investigate physics of the laser processing as well as the 3D simulator.



Fig. 9. In *situ* monitoring setup and typical result. (a) A schematic of the experimental setup. A scattered light is detected by a high-speed camera, and the scattered light patterns are used to make a DNN to learn how deep the hole or groove is. (b) Measured groove depth vs estimated depth by DNN.

processed surface, while without phase information and hence incomplete for complete surface reconstruction, contains plenty of information about the current state of the processing. By observing this pattern, it may be possible to predict features of the current processing.

Fig. 9(a) shows the schematic of the experimental setup. A 1-kHz femtosecond Ti:sapphire laser was used for the laser grooving, while a 1-kHz high-speed camera was used to monitor the scattered pattern from the material surface. Groove depths were studied through a supervised learning technique. Three successive patterns of the scattered light were utilized for the deep learning. In Fig. 9 (b), it can be seen that the DNN works to successfully predict ablated depths from scattered patterns. Provided with correct training data, similar DNNs could be constructed to work to predict the ablated volume and illuminated laser fluence at the same time [35].

In this application, one thousand data points could be obtained in one second with use of the high-speed camera. Hence, the data acquisition time with regards to different laser parameter was negligible compared to the learning time. Once implemented, the inference time of the constructed DNN is at the millisecond level, which is fast enough to make real-time feedback control realistic in future implementations. This technique could be deployed to a wide range of laser manufacturing machines for *in situ* monitoring. The framerates of low-cost and simple CCD or CMOS cameras have improved significantly in recent years, and such devices may suffice in some cases to provide an even more simplified implementation of our high-speed camera monitoring concept.

III. CYBER PHYSICAL SYSTEM

We lastly discuss our visions of how such automated cyberphysical systems could be incorporated into a future laser processing infrastructure.

As shown in Fig. 1, a smart laser-based fabrication chain has to calculate the optimal processing machines and processing parameters for each personalized order. Such decisions may be done by sifting through databases, or by relying on DNN simulators. Most importantly, if the main server has difficulty to find an appropriate parameter, advanced automation allows for the possibility of the AI to conduct its own experiments.

We have developed just such a fully automated laser processing, measurement, and evaluation system by combining the various instruments and measurement methods described in Section II. We name this device the Meister Data Generator (MDG). This conglomerate of highly automated processingand-measurement equipment can perform AI-based parameter optimization. Such optimization AIs, which we call "Agents", can operate the MDG to find optimized laser processing parameters on a trial-and-error basis, using various measurement instruments as guides. An AI Agent can each have its own strategy, for example, Bayes' optimization, to find an appropriate solution to its task at hand. The Agents can be considered as a cyber-analogue to the human craftsman. As individual craftsmen have their strengths and specialties, so do agents with different algorithmic strategies and approaches. Moreover, their access to both cyber and physical resources allow for various unique uses of both. Some agents may revert to simply studying past data, which, while faster than conducting actual experiments, may result in parameters with large ambiguities. Others may choose to conduct physical experiments with the MDG, which while accurate, will also be more costly. Such Agents' flexibility between cyber and physical spaces is realized by the agent operating environment, which works as a middle layer, in addition to a powerful data management system, which oversees the accumulation and remote analysis of experimental data with a single command.

Such MDGs could also be utilized to augment the functionality of local laser processing machines. It is of course costprohibitive to install advanced measuring equipment to each and every laser processing machine in the field. However, just as smartphones serve as gateways to vast amounts of information



Fig. 10. Upper: Concept of the Meister data generator MDG and smart laser machine. Lower: picture of MDG.

on the internet without locally storing or deriving it, such a "smart" laser machine may be developed to tap the expertise of centralized MDGs and databases to its advantage. This relationship is outlined in Fig. 10. Equipped with edge processing functionality, ideally a smart laser machine would be able to interpret a user's request, communicate its goals or parameters required for optimization to an MDG, and execute the returned solution. The user could then access the information from the MDG to infer quantitative and qualitative features of their results without possessing advanced measuring facilities. Additionally, the equipment of rudimentary sensors may allow for applications of seemingly advanced functionality, such as real-time feedback, utilizing know-how developed through MDGs. The utilization of two or more monitoring and measurement data would help to make a better simulator or feedback system, leading to a more reliable cyber-physical system.

IV. CONCLUSION

We have demonstrated automated data acquisition systems related to various problems in laser processing in order to obtain high-quality big datasets appropriate for deep learning analysis. Such strategies allow for the effective establishment of high-quality databases and simulators, important to advancing practical and theoretical aspects of laser processing. These processing data are important building blocks for the construction of a cyber-physical system of the laser processing. Additionally, the solutions provided by this system should play important roles in advancing our fundamental understanding of strong-field light-matter interaction.

REFERENCES

- [1] H. Lasi, P. fettke, H-G. Kemper, T. Feld, and M. Hoffman, "Industry 4.0," *Bus. Inf. Syst. Eng.*, vol. 6, pp. 239–242, 2014, https://doi.org/10.1007/s12599-014-0334-4
- [2] [Online]. Available: https://www8.cao.go.jp/cstp/english/society5_0/ index.html

- [3] S. Y. Kruchinin, F. Krausz, and V. S. Yakolev, "Colloquium: Strongfield phenomena in periodic systems," *Rev. Mod. Phys.*, vol. 90, 2018, Art. no. 021002.
- [4] L. V. Keldysh, "Ionization in the field of a strong electromagnetic wave," Sov. Phys. JETP, vol. 20, pp. 1307–1314, 1965.
- [5] M. Noda *et al.*, "SALMON: Scalable Ab-initio light–matter simulator for optics and nanoscience," *Comput. Phys. Commun.*, vol. 235, pp. 356–365, Feb. 2019.
- [6] N. Tancogne-Dejean *et al.*, "Octopus, a computational framework for exploring light-driven phenomena and quantum dynamics in extended and finite systems," *J. Chem. Phys.*, vol. 152, Mar. 2020, Art. no. 124119.
- [7] T. Otobe, "Wavelength dependence of the laser-excitation process on a silicon surface," *Phys. Rev. Appl.*, vol. 13, Feb. 2020, Art. no. 024062.
- [8] Y. Sanari et al., "Role of virtual band population for high harmonic generation in solids," Phys. Rev. B, vol. 102, Jul. 2020, Art. no. 041125(R).
- [9] K. L. Ishikawa, Y. Shinohara, T. Sato, and T. Otobe, "Strong-field electron dynamics in solids," 2020, *arXiv:2003.14090*.
- [10] G. C. Cho, W. Kütt, and H. Kurz, "Subpicosecond time-resolved coherentphonon oscillations in GaAs," *Phys. Rev. Lett.*, vol. 65, pp. 764, 1990.
- [11] K. Ishioka *et al.*, "Ultrafast electron-phonon decoupling in graphite," *Phys. Rev. B*, vol. 77, 2008, Art. no. 121402.
- [12] R. Ernstorfer *et al.*, "The formation of warm dense matter: Experimental evidence for electronic bond hardening in gold," *Science*, vol. 323, 2009, Art. no. 1033.
- [13] P. Maldonado *et al.*, "Tracking the ultrafast nonequilibrium energy flow between electronic and lattice degrees of freedom in crystalline nickel," *Phys. Rev. B*, vol. 101, 2020, Art. no. 100302.
- [14] I. Gierz *et al.*, "Phonon-Pump extreme-ultraviolet-photoemission probe in graphene: Anomalous heating of dirac carriers by lattice deformation," *Phys. Rev. Lett.*, vol. 114, 2015, Art. no. 125503.
- [15] M. Breusing, C. Ropers, and T. Elsaesser, "Ultrafast carrier dynamics in graphite," *Phys. Rev. Lett.*, vol. 102, 2009, Art. no. 086809.
- [16] P. A. George* *et al.*, "Ultrafast optical-pump terahertz-probe spectroscopy of the carrier relaxation and recombination dynamics in epitaxial graphene," *Nano Lett.*, vol. 8, no. 12, pp. 4248–4251, 2008 [Online]. Available: https://doi.org/10.1021/nl8019399
- [17] C. Jean *et al.*, "Direct observation of gigahertz coherent guided acoustic phonons in free-standing single copper nanowires," *J. Phys. Chem. Lett.*, vol. 5, pp. 4100–4104, 2014.
- [18] S. Sadasivam, M. K. Y. Chan, and P. Darancet, "Theory of thermal relaxation of electrons in semiconductors," 2017, arXiv:1709.00451v1 [cond-mat.mes-hall].
- [19] Y. Ishida, H. Masuda, H. Sakai, S. Ishiwata, and S. Shin, "Revealing the ultrafast light-to-matter energy conversion before heat diffusion in a layered dirac semimetal," *Phys. Rev. B*, vol. 93, 2016, Art. no. 100302(R).
- [20] M. E. Siemens *et al.*, "Quasi-ballistic thermal transport from nanoscale interfaces observed using ultrafast coherent soft X-ray beams," *Nature Mater*, vol. 9, pp. 26–30, 2010.
- [21] A. J. Minnich *et al.*, "Thermal conductivity spectroscopy technique to measure phonon mean free paths," *Phys. Rev. Lett.*, vol. 107, 2011, Art. no. 095901.
- [22] C Wu, and L. V. Zhigilei, "Microscopic mechanisms of laser spallation and ablation of metal targets from large-scale molecular dynamics simulations," *Appl. Phys. A*, vol. 114, no. 1, pp. 11–32, 2014.
- [23] C.-Yu Shih, M. V. Shugaev, C. Wu, and L. V. Zhigilei, "Generation of subsurface voids, incubation effect, and formation of nanoparticles in short pulse laser interactions with bulk metal targets in liquid: Molecular dynamics study," *J. Phys. Chem. C*, vol. 121, pp. 16549–16567, 2017.
- [24] H. Wu et al., "Experimental and computational study of the effect of 1 atm background gas on nanoparticle generation in femtosecond laser ablation of metals," *Appl. Surf. Sci.*, vol. 435 2018, Art. no. 1114.
- [25] Y. Tanaka, and S. Tsuneyuki, "Possible electronic entropy-driven mechanism for non-thermal ablation of metals," *Appl. Phys. Exp.*, vol. 11 2018, Art. no. 046701.
- [26] A. C. Vaucher *et al.*, "Automated extraction of chemical synthesis actions from experimental procedures," *Nat. Commun.*, vol. 11, 2020, Art. no. 3601.
- [27] P. S. Gromski, J. M. Granda, and L. Cronin, "Universal chemical synthesis and discovery with 'The chemputer'," Trends Chem., vol. no. 2, no. 1, pp. 4–12, 2020. [Online]. Available: https://www.sciencedirect. com/science/article/pii/S2589597419301868
- [28] J. Recatala-Gomez, A. Suwardi, I. Nandhakumar, A. Abutaha, and K. Hippalgaonkar, "Toward accelerated thermoelectric materials and process discovery," ACS Appl. Energy Mater., vol. 3, no. 3, pp. 2240–2257, 2020, doi: 10.1021/acsaem.9b0222. [Online]. Available: https://pubs.acs.org/ doi/10.1021/acsaem.9b022222020

- [29] B. Burger et al., "A mobile robotic chemist," Nature, vol. 583, pp. 237–241, 2020, doi: 10.1038/s41586-020-2442. [Online]. Available: https://www.nature.com/articles/s41586-020-2442-2020
- [30] S. Steiner *et al.*, "Organic synthesis in a modular robotic system driven by a chemical programming language," *Science11*, vol. 363, no. 6423, 2019, Art. no. eaav2211. [Online]. Available: https://science.sciencemag. org/content/363/6423/eaav2211
- [31] C. W. Coley *et al.*, "A robotic platform for flow synthesis of organic compounds informed by AI planning," *Sci. 09*, vol. 365, no. 6453, 2019, Art. no. eaax1566 [Online]. Available: https://science.sciencemag.org/ content/365/6453/eaax1566
- [32] T. Takahashi, S. Tani, R. Kuroda, and Y. Kobayashi, "Precision measurement of ablation thresholds with variable pulse duration laser," *Appl. Phys. A*, vol. 126 2020, Art. no. 582. [Online]. Available: https://doi.org/10. 1007/s00339-020-03754-5
- [33] S. Tani, and Y. Kobayashi, "Pulse-by-pulse depth profile measurement of femtosecond laser ablation on copper," *Appl. Phys. A*, vol. 124, pp. 265, 2018.
- [34] S. Tani, and Y. Kobayashi, Submitted.
- [35] S. Tani, Y. Aoyagi, and Y. Kobayashi, "Neural-network-assisted in situ processing monitoring by speckle pattern observation," *Opt. Exp.*, vol. 28, pp. 26180–26188, 2020.



Yohei Kobayashi was born in Sapporo, Hokkaido, Japan, in 1970. He received the B.S., M.S., and Ph.D. degrees in applied physics from the University of Tokyo, Tokyo, Japan, in 1993, 1995, and 1998, respectively. From 1998 to 2008, he was a Researcher with the National Institute of Advanced Industrial Science and Technology. In 2008, he was an Associate Professor with the Institute for Solid State Physics, The University of Tokyo. Since 2018, he has been a Professor with The University of Tokyo. Since 2017, he has been a Representative of TACMI consortium

(Technological Approaches toward Cool laser Manufacturing with Intelligence) with more than 70 corporations, which aims to make area of corporation related to a laser manufacturing. His research focuses on state-of-the-art laser development from 1-Hz linewidth lasers to a few femtosecond pulse lasers. He has studied precise laser control, which realize an optical frequency comb or parameter changeable high-power femtosecond lasers for laser processing. His current research focuses on a combination between laser technology and deep learning method to realize intelligent laser manufacturing.





He is currently working toward the Ph.D. degree with The University of Tokyo, Tokyo. His research focuses on laser technology.



Tomoharu Nakazato was born in Itabashi, Tokyo, Japan, in 1974. He received the B.S. degree in physics from the Tokyo University of Science, Tokyo, in 1998, and the M.S. degree in physics and the Ph.D. degree in chemistry from the Tokyo Institute of Technology, in 2000 and 2007, respectively. From 2007 to 2013, he was a Researcher with the Institute of Laser Engineering, Osaka University, Suita, Japan. From 2013 to 2018, he was a Researcher with the Research Institute for Science and Technology, The University of Tokyo, Tokyo. Since 2018, he has been a Researcher with The

University of Tokyo. His research focuses on development and applications of laser in ultraviolet region, which is an attractive light source for various fields such as, micro-processing and medicine. He is currently engaged in management of laser processing projects, as a whole in order to ensure the promotion of each research in the projects and translation of the results to society.



Haruyuki Sakurai was born in Tokyo, Japan. He received the B.S., M.S., and Ph.D. degrees in physics from the University of Tokyo, Tokyo, Japan, in 2014, 2016, and 2019, respectively.

From 2019 to 2020, he was a Project Researcher with the Institute for Photon Science and Technology, The University of Tokyo.

Since 2020, he has been a Project Research Associate with the Institute for Solid State Physics, also with The University of Tokyo. His research interests include laser processing, ranging from the fundamen-

tals of light-matter interaction, to application.



and free electron lasers.

Kazuyuki Sakaue was born in Saitama, Japan, in 1982. He received the B.S., M.S., and Ph.D. degrees in physics from Waseda University, Tokyo, Japan, in 2004, 2006, and 2009, respectively. From 2009 to 2018, he was a Researcher and Assistant Professor with Waseda University. In 2018, he was a Principal Researcher with Photon Science Center, the Graduate School of Engineering, The University of Tokyo, Tokyo. His research focuses on micro or nano-fabrication of material surface using advanced lasers or light sources, such as high-order harmonics



Hiroharu Tamaru was born in Kanagawa, Japan, in 1970. He received the B.S., M.S., and Ph.D. degrees in applied physics from the University of Tokyo (UTokyo), Tokyo, Japan, in 1993, 1995, and 2005, respectively.

From 1997 to 2008, he was a Research Associate with UTokyo. In 2008, he was a Project Lecturer with Photon Science Center, Graduate School of Engineering, UTokyo and in 2015, he was a Project Associate Professor with the Institute for Photon Science and Technology, Graduate School of Science, UTokyo.

Since 2017, he has been the Vice-Representative of TACMI consortium. His research interests include experimental and theoretical studies of photon-matter interactions at nanometric scales, and development of advanced methodologies for their detections. His current research interests include system designs for integration of laser processing systems, data acquisition systems, and networked database systems, to realize intelligent laser manufacturing.



Shuntaro Tani as born in Tochigi, Japan, in 1986. He received the B.S., M.S., and Ph.D. degrees in physics from Kyoto University, Kyoto, Japan, in 2008, 2010, and 2014, respectively. He is currently a Research Associate with The University of Tokyo, Tokyo, Japan. His research focuses on light-matter interaction under strong laser pulses, including spatiotemporal dynamics and deep-learning approach.



Kenichi L. Ishikawa received the B.Eng. and M.Eng. degrees in nuclear engineering from The University of Tokyo, Tokyo, Japan, in 1992 and 1995, respectively, and the Ph.D. (Dr. rer. nat.) degree in physics from RWTH Aachen University, Aachen, Germany, in 1998. He is currently a Professor with the Department of Nuclear Engineering and Management, Graduate School of Engineering, Research Institute for Photon Science and Laser Technology, The University of Tokyo. Since 2019, he has been a concurrently Guest Professor with Osaka University, Suita, Japan.

He was a Postdoctoral Researcher with CEA-Saclay from 1998 to 2000, a Special Postdoctoral Researcher with RIKEN from 2000 to 2002, an Associate Professor with The University of Tokyo from 2002 to 2008, a Senior Researcher with RIKEN from 2008 to 2009, and a Project Associate Professor with The University of Tokyo from 2009 to 2014. He is internationally renowned for his contributions to numerical calculation and theoretical modeling of laser-matter interaction, ranging from strong-field physics, ultrafast laser science, attosecond science to laser processing. He is currently participating several national projects of Japan to promote smart laser manufacturing and, in particular, leading a nation-wide network project for proposing the optimal processing parameters using artificial intelligence and simulation in cyberspace.