1
Introduction

> In our highly complex and ever changing world it is reassuring to know that certain physical quantities can be measured and predicted with very high precision. Precision measurements have always appealed to me as one of the most beautiful aspects of physics. With better measuring tools, one can look where no one has looked before. More then once, seemingly minute differences between measurement and theory have led to major advances in fundamental knowledge. The birth of modern science itself is intimately linked to the art of accurate measurements.
>
> T.W. Hänsch. Nobel Price lecture 2005 [1]

1.1
Motivation

The demand for custom-made products with higher machining precision has always pushed the development of new methods. The down-sizing of technologies into the micro- and nanometer scale prompts the need for a new generation of processing tools. Ultra-fast laser radiation can fulfill many of the corresponding requirements. This new generation of radiation processing tools demands the development of focusing methods, beam delivering and new process handling and surveillance equipment. The complete chain from the laser source as a tool to the work piece has to be designed from the viewpoint of micro-nanoscaling.

The unique properties of ultra-fast laser radiation enable new technologies for engineering due to its ability to deposit optical energy in precisely localized volumes and as a no-contact tool. The deposition of optical energy occurs without inducing stress into the material. The necessity for ultra-fine machining and manipulation with high throughput has moved scientists to develop new laser sources with key properties like ultrashort pulse duration, ultra-high repetition rates, or both simultaneously. High-repetition rate systems like fiber, slab or disk-lasers for high throughput are expected to become industrially applicable soon.

The involved processes themselves like melting, evaporation, and plasma-formation, can be in general quite complicated due to the simultaneous presence

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of multiple phenomena. Most of the phenomena are on an ultra-fast time scale, demanding also new diagnostic techniques for imaging. Many of these processes are still little understood and have to be investigated for a deeper understanding. Here ultra-fast optical metrology using femtosecond laser radiation is the key technology needed to enable the observation, detection and highlighting of the processes on time-scales $\ll$ 1 ns, or measurement of process variables during micro- and nanostructuring.

For the transfer from the laboratory into the production the new ultra-fast application methods, with the processes involved during ultra-fast machining, have to be controlled adopting ultra-fast optical metrology implying the following:

1. Understanding of the laser-induced processes like melting, evaporation or plasma-formation in advance
2. Detectability and manipulability of process parameters during machining.

In production technology today ultra-fast laser radiation is not widely adopted, because complex processes are expected. By using ultra-fast laser radiation additional processes are given to those in laser engineering with conventional laser radiation. But many of these processes are negligible; some examples are:

- Time-scales for absorption of the ultra-fast laser radiation in the dense matter and in an evolving plasma-plume are separated, in contrast to nanosecond laser radiation. The interaction of an ultra-fast laser pulse with the generated plasma can be omitted.
- Time-scale for absorption is much smaller than the relaxation time of matter for phase change from solid to vapor. This implies that after absorption of ultra-fast laser radiation, matter is excited independent of the material properties.
- Matter is instantaneously[1)] excited into a plasma state due to the high intensities of the focused laser radiation. The new machining tool "ultra-fast laser radiation" enables nearly melt-free ablation by the ultrashort pulse duration in the femtosecond regime.
- Typical limitations of machining by wavelength-dependent absorption of laser radiation is mostly overruled by multi-photon processes using ultra-fast laser radiation. Ultra-hard matter, like diamond or tungsten carbide, are difficult to machine using conventional milling techniques, whereas using ultra-fast laser radiation enables machining of usually non-machinable matter. The scales of the created features using femtosecond laser radiation are beyond the resolution of conventional techniques as is well-shown for laser drilling of metals in Figure 1.1.

Ultra-fast laser radiation as an operative tool has advanced now to an engineering level and, as a consequence, ultra-fast engineering technology is even advanced

1) transition time $< 10^{-14}$ s

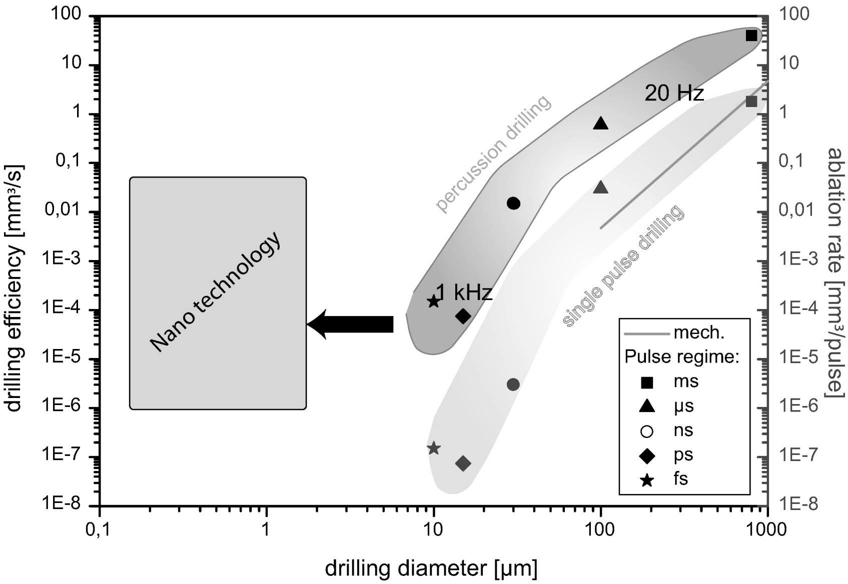

Fig. 1.1 Drilling efficiency and ablation rate versus diameter for mechanical and laser drilling by single pulse and percussion drilling with different pulse durations (data from [2]).

enough to become a new field within the mechanical engineering sciences. To transfer this technology to industry, the new field of ultra-fast optical metrology presented here will document the induced processes by ultra-fast laser radiation and the technologies necessary to detect these processes in order to control them. The physics of ultra-fast laser radiation-matter interaction with the processes of absorption, polarization, evaporation and ionization will be systematically ordered, elucidating key properties of ultra-fast laser radiation adopted in ultra-fast optical metrology.

Ultra-fast optical metrology using pump and probe techniques, utilizing laser radiation simultaneously to initiate and to detect a process, opens the detection with temporal resolutions up to 10 fs and spatial resolutions of 20 nm and below. The methodology and the applications of this new field of research is presented: the choice and the characterization of the probe radiation are fundamental issues for a successful measurement.

The transfer to high throughput ultra-fast engineering is feasible with the enhanced understanding of the involved processes gained by ultra-fast optical metrology. The processing can be controlled by new technologies for ultra-precise manipulation using ultra-fast optical metrology. As a consequence, ultra-fast optical metrology today is ready for applications in ultra-fast engineering technology.

1.2
Definition of Optical Pump and Probe

Optical pump and probe technique is a measurement technique using laser radiation. The laser radiation exhibits at least two functions:

1. Pumping by processing matter, exhibiting, for example, excitation, melt, ablation, evaporation and ionization,
2. Probing to monitor these processes.

In the case of optical pump and probe techniques the radiation beams are extracted from one laser source or from two laser sources. Using one laser source, the radiation is divided into at least two beams, and one beam is delayed temporally to the other in order to monitor using the investigated process (Figure 1.2). The temporal resolution is given by the pulse duration of the laser radiation.

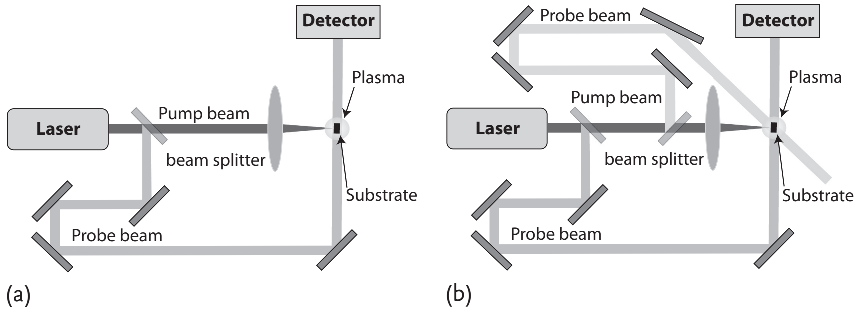

Fig. 1.2 Scheme of optical pump and probe metrology using one probe (a) and two probe beams (b).

Ultra-fast radiation adopted in a pump and probe set-up enables one to investigate laser-induced micro- and nanostructuring with > 10 as temporal and 20 nm spatial resolution due to its high temporal resolution, on the one hand, and on the other hand due to the unique properties of this radiation-like broad spectral distribution and small coherence.

1.3
Guideline

The following chapters will give an insight into the optical pump and probe metrology for ultra-fast engineering. The working tool – ultra-fast laser radiation – is described by its market position (Section 1.4), and deals with a resume on the history of high-speed metrology before the invention of the laser, where in the nineteenth century pump and probe metrology became popular. Following this, the history of laser ultra-fast metrology will be sketched.

In the second chapter an overview of the ultra-fast laser source will be given. The sources (Section 2.1), the properties of the focused laser radiation (Section 2.2), and the tools to manipulate and move the beam spatially (Section 2.3), are described concluding with some outstanding challenges in ultra-fast metrology (Section 2.4), and especially the optical pump and probe techniques (Section 2.5).

The third chapter is dedicated to some fundamentals of laser radiation-matter interaction. The interaction of laser radiation at intensities $I > 10^{12}\ \mathrm{W/cm^2}$ will be discussed, also non-linear processes (Section 3.1), like laser-induced multi-photon absorption (Section 3.2). The plasma generated at these intensities gets a dominant position in the laser-induced process, due to its extreme energetic properties (Section 3.3).

The ultra-fast laser radiation is a fundamental tool of optical pump and probe metrology and will be discussed under this point of view in the fourth chapter. The properties of ultra-fast laser radiation and its propagation through matter, which is important for pump and probe metrology, will be described (Section 4.1). The conditioning of the laser radiation is the scope of Section 4.2; in the case of coherent processes, the generation of quantum states is meant. Probe radiation is often used as an illumination tool, and for nanotechnology applications the imaging is conditional to the resolution limit of the optical system (Section 4.3). The pump and probe technique enables one to detect at different time-steps a process by delaying the probe radiation relative to the exciting pump radiation. Methods and limits of temporal delaying are described in Section 4.4.

The fifth chapter describes the methodology of optical pump and probe in practice, showing also limits of this metrology. The chapter is subdivided into non-imaging and imaging detection. A selection of non-imaging detection methods like spectroscopy (Section 5.1) will be given, furthermore a selection of imaging detection methods describing some actual used set-ups for imaging techniques (Section 5.2) will be presented.

A selection of applications for optical pump and probe metrology for engineering is given in the sixth chapter for drilling and structuring metals (Sections 6.1 and 6.2) and for marking and welding glasses (Sections 6.3 and 6.4).

The seventh chapter describes the perspectives of this new field of research and forecasts optical pump and probe metrology for the future, showing potential applications by using new laser sources (Section 7.1) and also new detectors in combination with improved pump and probe methods (Section 7.2).

1.4
Matrix of Laser Effects and Applications

Ultra-fast laser applications are derived from given applications. As proposed by Sucha [3] and shown in Figure 1.3, the properties of ultrashort laser radiation, such as “high-speed”, “high-Power”, “bandwidth”, “structured spectral coherence” and “short coherence length”, drive different phenomena, like ablation, THz imaging,

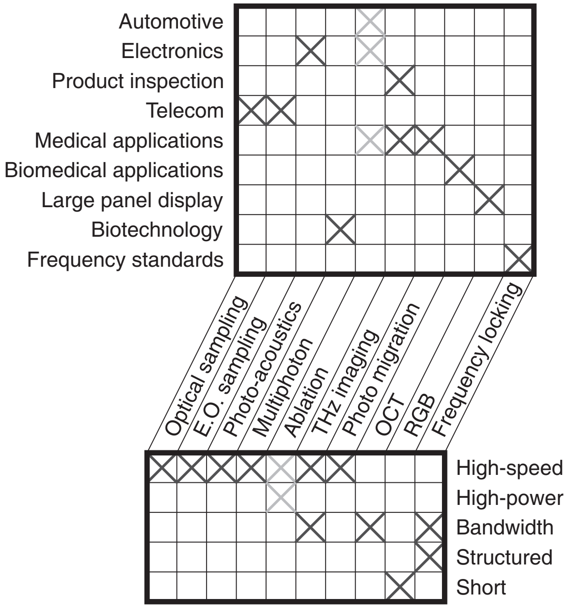

Fig. 1.3 Matrix of laser properties, techniques and applications (according to [3]).

OCT (Optical Coherence Tomography) and frequency conversion for the RGB laser (Red-Green-Blue). From these phenomena the markets are derived. The resulting routes from the properties to the phenomena to the market are many-fold, demonstrating that ultra-fast technology is becoming increasingly market-present in different areas.

From this matrix and the markets it can be deduced that ultra-fast engineering – mainly depicted by the phenomena "ablation" – is connected by two properties of ultra-fast laser radiation: small pulse duration and large peak power. Ablation includes cutting, joining and milling, being the main market for applications with classical laser sources, like cw-CO_2, and cw-Nd:YAG lasers. For ultra-fast mechanical engineering, the property "ablation" emerges with nearly melt-free ablation. This enables new approaches for micro- and nanotechnology applications.

1.5 Historical Survey of Optical Ultra-fast Metrology

1.5.1 Metrology Techniques Before the Advent of Laser

Time-resolved detection of optical emission in former times was mainly limited by the mechanical shutter restricting the temporal resolution to about 1 ms. High-speed metrology has been used in photography since the nineteenth century by using flash-bulbs having microsecond time resolution. High-speed photography was developed in 1834 using a mechanical streak camera. Mechanical streak cameras use a rotating mirror or moving slit system to deflect the light beam. They are limited in their maximum scan speed by mechanical properties, and thus the temporal resolution is limited to about 1 μs [4]. The first practical application of high-speed photography was Eedweard Muybridge's investigation on whether horses' feet were actually all off the ground at once during a trot. Muybridge had successfully photographed a horse in fast motion using a series of twenty-four cameras.

Schlieren photography was developed in 1864 by adapting Foucault's knife-edge test for telescope mirrors to the analysis of fluid flow and propagating shock waves [5]. In 1867 August Töpler combined this set-up using a light spark with about 1 μs emission duration and was able to detect sound waves in air.

A type of Kerr-shutter was invented in 1899 called the Abraham–Lemoine shutter with temporal resolutions of about 10 ns [6, 7]. Two polarizing filters were mounted at 90° to block all incoming light. A Kerr-cell, which changes the polarization of the passing radiation when energized, was placed between the filters and used as a shutter, energized for a very short time by, for example, a spark, and allowed a detector like a photographic plate connected to an imaging system or a spectrometer to be properly exposed.

In 1930 stroboscopes were used for the first time to study synchronous motors [8–10]. Also the dynamics of high-velocity particles like bullets were measured in 1960. Temporal resolutions up to 100 ns could be achieved by flashing. The rapatronic camera, developed in 1940, utilized two polarizing filters and a Kerr-cell to overcome the mechanical limitation of a camera's shutter speed with shutter times of about 10 ns and has been adopted for photography of ab initio nuclear experiments [11, 12].

The improvement of electronics in 1950 enabled the development of electro-optical streak cameras based on a evacuated tube with a photon-sensitive cathode emitting electrons after irradiation and an electron-detecting phosphor screen [13, 14]. The electrons are accelerated in an electric field toward the positively charged phosphor screen. Temporally resolved information with > 200 fs resolution is given by streaking spatially the electrons orthogonally to their propagation direction applying a second, step-like high-voltage electric field. The streaked electron beam induces an optical emission at the phosphor screen, which can be detected by conventional photography.

1.5.2 Ultra-fast Pump and Probe Metrology

The advent of the laser in 1960 [15] created many new fields of research in physics, new applications in mechanical engineering, and pushed optical metrology. Using dye lasers, invented in 1964 [16], a broad spectrum of wavelengths becomes accessible for time-resolved spectroscopy. The dye laser has been brought into the ultra-fast picosecond regime by introducing mode-locking [17]. Ultra-precise spectroscopy is achieved less by the short pulse duration than by small spectral line widths of the laser sources being ideally in the cw-mode (and consequently not described in this book).

Ultra-fast time-resolved measurements are well established in physics, chemistry, and physical chemistry, where fundamental time-scales of selected chemical reactions become accessible for predicting the kinetics of chemical reactions. The photosynthesis of hemoglobin (Hb) has been investigated by transient absorption spectroscopy which could be described as a fast photo dissociation for HbO_2 and a slow dissociation for HbCO [18]. Temporal shaping of the ultra-fast laser radiation enabled chemically induced reactions and control by cooling the vibration modes of HBr molecules [19, 20]. The dissociation of NaI into its constituents has been measured and modeled [21]; in solid-state physics and electrical engineering, carrier dynamics and transport have been probed on picosecond time scales, being directly relevant to the operation of modern high-speed devices [22].

Also, ultra-fast investigations adopting optical pump and probe metrology on an atomic scale and attosecond time scale have been performed using ultra-fast laser radiation. For example, high harmonics X-ray attosecond probe radiation has been used on a Kr atom, which was ionized in the inner shell, generating a hole. The hole recombines with an electron from the outer shell emitting an additional electron, called an Auger-electron. A second laser pulse probes the Kr atom after generation of the electron hole. A life time of 8 fs for the electron-hole has been measured [23–25].

Many investigations have been made by investigating laser-induced processes in condensed matter. Electronic units made from semiconductors like gallium arsenide have a dominant position for information and communication technology. The miniaturization of these elements is not finished today. The processes to generate the circuits on these materials are becoming more and more difficult because the resolution limit for processing techniques using UV radiation is today limited to 60 nm. The detection of the processes during the generation of features with dimensions < 50 nm improves understanding [26]. Pump and probe photo-emission spectroscopy of metals like Au and Ta, has been used to detect the electron dynamics of solids at the surface [27, 28]. For crystalline silicon the thermalization times after irradiation with ultra-fast laser radiation have been measured for the electrons to about 100 fs and respectively for the phonons to about 50 ps [29]. Also, after excitation of GaAs with femtosecond laser radiation using spectral broad probe radiation, the complex dielectric function has been investigated on measuring the reflectivity. The complex dielectric function changes in time from semiconductor to metallic behavior [30].

Imaging using laser radiation as an illumination source has been done since the invention of the laser itself. Some of the latest results are given here. Shadowgraphy of ablation plumes during percussion drilling of metals has been achieved, demonstrating the complex expansion and dynamics of plasma [31] and of the vapor by using fluorescent emission photography [32]. The laser-induced modifications in glasses have been detected by time-resolved interferometry [33] and holography, as well as speckle interferometry, have been adopted using solid state, gas and excimer lasers for different scientific fields.

The experiments described were mostly achieved in the focus of scientific research. In order to emphasize ultra-fast metrology for mechanical engineering, substantial definitions and solution for ultra-fast metrology are given in the following chapters.