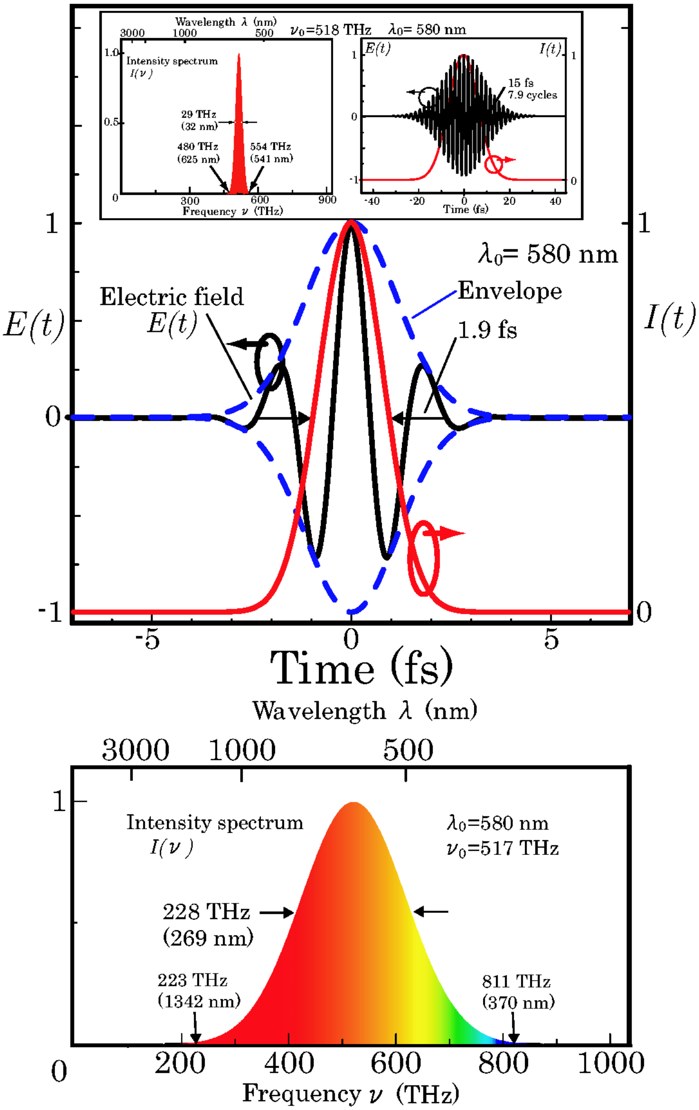
Preface

Extreme technology has always opened new exciting fields in science and technology. This book is mainly concerned with extreme technologies in the ultrashort time scale (around sub-ten femtoseconds ; 10^{-14}–10^{-15} s) and in the ultrasmall space scale (around sub-nanometers ; $\sim 10^{-10}$ m). Unfortunately, until recent years both technologies developed separately. This book is the first attempt to describe recent advances in femtosecond technology and the fusion of this to nanometer technology. That is, the purpose of this book is to review contributions we have made to the fields of ultrafast optics as well as optical scanning tunneling microscopy (STM) in recent years (1996-2004). Also, in the introductions of several chapters, historical progresses from various sights in this interdisciplinary field are summarized briefly with tables.

Ultrashort optical pulse technology in the near-infrared, the visible and the ultraviolet region is now in a time scale into the few femtosecond range in the optical-mono-cycle region. The full-width at half-maximum (FWHM; T_{du}) of the temporal intensity profile in the mono-cycle pulse equals the single cycle period T_{per} of the electric field, $T_{\mathrm{du}} = T_{\mathrm{per}}$ (Fig. 1). For example, the mono-cycle pulse of a Gaussian profile with a 580 nm center wavelength has a $T_{\mathrm{du}} = 1.9$ fs duration and a $\Delta\nu_{\mathrm{B}} = 228$ THz FWHM bandwidth (the corresponding wavelength bandwidth of $\Delta\lambda_{\mathrm{B}} = 269$ nm) with a spectral broadening from 370 to 1342 nm, according to a relationship of $T_{\mathrm{du}} \times \Delta\nu_{\mathrm{B}} = k$. Here, k is a constant depending on the temporal intensity profile. This equation suggests that with the decrease in pulse duration the spectral bandwidth rapidly increases.

"Few-to-Mono Cycle Photonics" means technology and science are necessary for the realization and application of the few-to-mono cycle pulse in the optical frequency region. In this book, among these widely and rapidly developing fields, four basic technologies of the ultrabroadband pulse generation, the ultrabroadband chirp or phase compensation, the phase and amplitude characterization of the ultrashort pulse, and the feedback field control of the ultrabroadband, ultrashort pulse are dealt with. In addition, the theory of the ultrashort pulse nonlinear prorogation beyond the slowly-varying-envelope approximation is developed. In particular, the generation of the shortest pulse with a 2.8 fs duration, a 1.5 cycle and a 460–1060 nm spectral broadening in the near-infrared and visible region, and the computer-controlled feedback

Monocycle optical pulse



$$\Delta \nu_{B} = k / T_{du} \rightarrow \text{ultra-large } \Delta \nu_{B}$$

Fig. 1. Monocycle Gaussian pulse with a center wavelength of 580 nm and a duration of 1.9 fs. The inset shows a 15-fs Gaussian pulse with the same center wavelength, for the comparison

manipulation that combines spectral-phase characterization and compensation should be noted. However, the carrier-envelope phase technology, which is currently developing rapidly, is hardly described.

Ultrashort optical pulse technology, which is based on sophisticated laser technology, has the following significant, unique capabilities: to clarify ultrafast phenomena in all fields of natural science and engineering at the highest time resolution; to control ultrafast time-sequential phenomena; to produce an ultra-high peak electric field; and to generate, transmit and process an ultra-high density information signal. In addition, since time is one of essential parameters to describe temporal dynamic phenomena in any disciplines, this fastest technology (among the human-developed ones) is called for across all the fields and disciplines in natural science and engineering. However, this optical technology has the drawback of relatively low spatial resolution ($\gtrsim$ μm) because of the electro-magnetic wave with a finite wavelength.

On the other hand, STM has the highest spatial resolution of sub-nm, which enables us to observe spatial dynamics at single-atomic and single-molecular levels in real space. There have been a lot of studies using STM, which is related to various phenomena that occur on conductive surfaces, such as thin film growth, molecular adsorption, chemical reaction, electron standing wave, charge density wave, Kondo effect, thermo-dynamics of voltex at surface of high-T_c superconductors. For current researchers, nanoscale science and technology is one of the most attractive and important fields, and realizing new functional devices with nanoscale elements is one of their main goals. In these cases, interactions between optical and electronic systems play essential roles. When the scale of specimens was larger, photo-assisted spectroscopy provided a very helpful way to investigate such structures in materials. For example, photoelectron spectroscopy, photo-scattering spectroscopies and reflection methods have revealed various physical properties of materials until now. However, since the device size is already as small as a few tens of nanometers, these conventional optical methods are not applicable because of the spatial resolutions limited by light source wavelengths, which are generally more than 100 nm as mentioned before. At this moment, only STM related technology is a promising candidate for the investigation of the characteristics of nanoscale structures. Since tunneling current is used as the probe, electronic structures are picked up. Therefore, when STM is combined with the optical system, the analysis of the transient response of photo-induced electronic structures is expected at the ultimate spatial resolution. Therefore, the combination of optical systems with STM is considered to be a very promising technique. However, STM has the inevitable disadvantage of very low time resolution ($\sim$ sub ms) because of the slow response time of the highly sensitive, integrated detector for the very low tunneling current.

To overcome the problems of both technologies and to utilize both features, a new technology and science is required. That is “optical STM”. Optical STM means technology and science of the femtosecond-time resolved

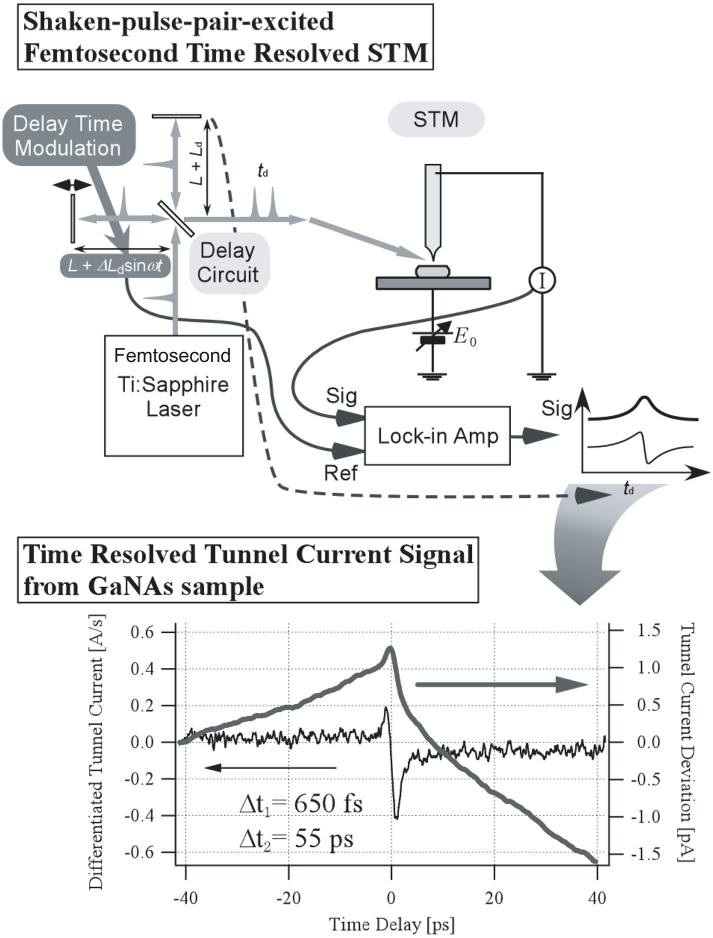


Fig. 2. Femtosecond-time-resolved STM and its application

STM (FTR-STM) and the STM-level phenomena controlled by femtosecond optical pulses, tunable laser excitation and laser excitation power including nonlinear optical phenomena at the atomic level. One example of the principles for FTR-STM is shown in Fig. 2. Its upper part is the schematic of the FTR-STM system, and the lower part is an example result measured for a GaAs sample. Relaxation of the photoinduced current in the band structure is picked up as the two components in the picosecond range. The vales are close to those obtained by the conventional optical pump probe technique, however, since the probe is the tunneling current, the spatial resolution is atomic scale in this case. That is, the controlled delay time between two femtosecond optical pulses for excitation is employed to get highly temporal resolution. The integrated tunneling current of a tip at a fixed position for each pulse-delay time is employed to get highly spatial resolution, This principle is similar to the conventional pump (a pulse pair) and probe techniques in ultrafast optics. That is, two sequential photon energies of two optical pulses with delay time play the role of the pump to induce or change the tunneling current. And, the observed signal of the integrated tunneling current plays the role of the probe to get information on the temporal surface phenomena at the atomic level. As a result, the probe signal as a function of the delay time provides nonlinear-optically induced dynamics at the spatiotemporal extreme level. Thus, this spatiotemporal-extreme frontier technology has a possibility to open a new field by clarifying and manipulating ultrafast dynamic phenomena at the atomic level, which have not been revealed so far by conventional techniques because of measurements of the temporally coarsened and spatially averaged information in addition to the statisical treatment of a single element.

Accordingly, this book consists of two parts. The first part of few-cycle photonics is organized into six chapters. The second part of optical STM is organized into four chapters.

In Chap. 1, Karasawa, Mizuta and Fang discuss theoretically nonlinear propagation of ultrashort, ultrabroadband optical pulses exceeding the conventional approximation of the slowly-varying envelope in an electric field by various methods of numerical computer analysis.

In Chap. 2, Yamashita, Karasawa, Adachi and Fang review the experiments leading to the generation of ultrabroadband optical pulses with a near or over one-octave bandwidth and a well-behaved spectral phase by unconventional methods including an induced phase modulation technique.

In Chap. 3, Yamashita, Morita and Karasawa focus experimentally and theoretically on the active chirp compensation for ultrabroadband pulses using a spatial light modulator (SLM) technique.

In Chap. 4, Morita, Yamane and Zhang cover the phase and amplitude characterization of the electric field in few-cycle pulses with some techniques.

In Chap. 5, Yamashita, Yamane, Zhang, Adachi and Morita detail experimentally and theoretically the feedback control that combines spectral-phase characterization and compensation for optical pulse generation in the few-to-mono cycle region in the case of various kinds of fiber outputs.

In Chap. 6, Morita and Toda discuss experimentally and theoretically wavelength-multiplex electric-field manipulation of ultrabroadband pulses and its application to the vibration motion control of molecules.

In Chap. 7, Shigekawa and Takeuchi introduce the fundamentals of laser combined STM after brief explanation of the STM bases.

In Chap. 8, Shigekawa and Takeuchi review light-modulated scanning tunneling spectroscopy for visualization of nano-scale band structure in semiconductors.

In Chap. 9, Futaba focuses on the control experiment of semiconductor surface phenomena by femtosecond optical pulse-pair excitation at the atomic level.

In Chap. 10, Shigekawa and Takeuchi discuss the development of FTR-STM using a method of two optical pulse excitation and its application.

In Chap. 11, Yamashita, Shigekawa and Morita describe briefly near-future subjects and directions in few-cycle photonics and optical STM.

Many works introduced here have been carried out by many Hokkaido University and Tsukuba University postdoctoral fellows, visiting associates, graduate (and undergraduate) students. And, most of those works were done under the support of Japan Science and Technology Corporation (JST) as a project of "Optical-wave technology in the cycle-time region and its application to single-atomic and single-molecular dynamic phenomena" for 1997 to 2002.

Finally, the editors wish to thank all of the authors who have contributed to the excellent individual chapters and sections of this book. One of the editors (M. Yamashita) greatly acknowledges Mr. Naoya Nakagawa and Ms. Noriko Yoshida for their secretarial assistance.

Sapporo,
October 2004

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