Plasma diagnostics using multi-heterodyne interference of optical frequency comb

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Contents of this presentation

- **Background & overview of my research**
- **Introduction of optical frequency comb**
  - Brief history, Basic properties, Generation method, Frequency range
- **Applications of optical frequency comb**
  - Applied spectroscopies using frequency combs including for plasma diagnostics
- **Frequency-comb interference spectroscopy**
  - Principles and measurement of argon metastable atoms in RF plasma
- **Concluding remarks**
LAPD-2013 is the biennial Laser Aided Plasma Diagnostic conference. Originally inaugurated at Kyushu University, Kyushu, Japan in 1983, it has been organized by various plasma research groups around the world since.
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I was born in Kumamoto city in September, 1983!!
Kyoto University [~2012, B.S. M.S. Ph.D]
(Prof. K. Tachibana and Prof. O. Sakai)
- Small-scale plasmas (Microplasmas) generated at high and atmospheric pressure

- Diagnostics and control of plasma jet
- Development of laser spectroscopy using optical frequency comb

Ph.D. thesis:
"Spectroscopic Study of Dielectric Barrier Discharge at Atmospheric Pressure"
The University of Tokyo [2012-, JSPS PD]
(Prof. K. Terashima)
- Microplasmas generated in high-density fluids
  - Supercritical fluids (CO$_2$, Xe, H$_2$O, etc.)
  - Low temperature (He > 5 K, Cryoplasmas)

Diagnostics of microplasmas
- Development of electron density measurement by laser interferometry
  (Poster)
Contents of this presentation

- Background & overview of my research

- **Introduction of optical frequency comb**
  - What is the optical frequency comb??
    - Generation and properties
  - Brief history and recent development
    - Expansion of wavelength range

- Applications of optical frequency comb

- Frequency-comb interference spectroscopy

- Concluding remarks
Optical frequency comb

- **What is optical frequency comb**
  - Coherent laser light (source)
  - Its spectrum consists of a series of discrete, equally spaced elements
  - Wide frequency band

Illustration of enlarged spectrum of frequency comb in THz~GHz order
What is **optical frequency comb**

- Coherent laser light (source)
- Its spectrum consists of a series of discrete, equally spaced elements
- Wide frequency band
- Developed energetically around 2000

  - Novel Prize in Physics 2005 shared by Prof. John L. Hall (NIST) and Prof. Theodor W. Hänsch (Max Planck Inst.)

- Extremely precise ruler for light frequency (frequency metrology, atomic clock)
Optical frequency comb generation

Simplest setup of frequency comb generation
- Fabre-Perot cavity
- Electro-optic modulator

Same interval frequency

Input CW single frequency laser beam

EOM driver

Modulation frequency: $f_m$

Mirror

Electro-optic modulator (EOM)

Output laser beam

Optical frequency comb generation

Simplest setup of frequency comb generation

- Fabre-Perot cavity
- Electro-optic modulator

EOM driver

Modulation frequency: \( f_m \)

Input CW single frequency laser beam

Electro-optic modulator (EOM)

Output laser beam

Light intensity

Laser frequency

Optical frequency comb generation

Simplest setup of frequency comb generation

- Fabre-Perot cavity
- Electro-optic modulator

Electro-optic modulator is a phase modulation device

It generates side bands in the laser beam with the differential frequency at $f_m$

Optical frequency comb generation

Simplest setup of frequency comb generation
- Fabre-Perot cavity
- Electro-optic modulator

EOM driver
Modulation frequency: $f_m$

Mirror
Electro-optic modulator (EOM)
Output laser beam

Input CW single frequency laser beam

Light intensity vs. Laser frequency

Laser beam is trapped in Fabre-Perot cavity and transmit through the EOM many times

Frequency band become wider inside the cavity

Optical frequency comb generation

Simplest setup of frequency comb generation

- Fabre-Perot cavity
- Electro-optic modulator

Electro-optic modulator (EOM)

Modulation frequency: $f_m$

Same interval frequency

Input CW single frequency laser beam

Mirror

EOM driver

Output laser beam

Laser beam is output from the cavity with many frequency comb elements with a interval frequency $f_m$ ranging a very wide frequency band

Optical frequency comb generation

Pulse chain of Modelock laser
- Ti:Sapphire Laser
- Fiber laser

Fourier transformation

Optical frequency comb
- Offset frequency: $f_0$
- Interval frequency: $f_{\text{rep}}$

Example of comb structure in Ti:Sa fs laser

- We measured beat signal generated in Ti:Sa fs laser using spectrum analyzer
- Our optical frequency laser source
  - Ti:Sapphire fs modelock laser (Spectra Physics)
- Peak interval of beat signal spectra
  - $81 \text{ MHz} = f_{\text{rep}}$

Power spectrum of beat signal generated by frequency comb measured in spectrum analyzer
Example of wide spectrum of optical frequency comb

- Comparing with diode laser spectrum
- Detected range: 810 ±10 nm
- From \( f_{\text{rep}} \) and \( \lambda \) range
  - Number of combs in Ti:Sa fs laser is > 100,000!!

Overall spectrum of frequency comb (fs modelock laser) compared with diode-laser spectrum
Wavelength (Frequency) of frequency combs

- Near infrared (IR) – visible range
  - Mode-locked Ti:Sapphire femtosecond (fs) laser
  - Mode-locked Yb-fiber (Er-fiber) laser
    - Broadband frequency comb generation by photonic crystal fiber

- Far IR $\rightarrow$ Terahertz (THz) range
  - [ OPO ] [ Photoconductive emitter excited by fs laser ]

- Visible $\rightarrow$ Vacuum ultraviolet (VUV) range
  - [ SHG ] [ High harmonic generation in Kr or Xe gases ]
Contents of this presentation

- Background & overview of my research
- Introduction of optical frequency comb

- Applications of optical frequency comb including plasma diagnostics
  - Brief introduction of major applications
  - Broadband cavity ring-down spectroscopy
    - Prof. N. Sadeghi’s work on plasma diagnostics

- Frequency-comb interference spectroscopy
- Concluding remarks
Applications of optical frequency comb

- **Accurate measurement of laser wavelength**
  - Frequency standards for optical clock, frequency metrology
    - D. J. Jones, J. L. Hall *et al.*, Science, **288** (2000) 635.

- **Molecular spectroscopy in IR range (FT-IR, dispersion)**

- **Dual-comb spectroscopy**

- **Broadband cavity enhanced absorption spectroscopy**
Cavity enhanced absorption spectroscopy by single-frequency laser

- High sensitivity of target species
- Need to prepare many laser source for each target species

Broadband absorption spectroscopy by incoherent light sources (lamp…)

- Measure many species by one light source
- Cannot form resonance cavity mode (lower sensitivity than ICLAS)

Broadband cavity enhanced absorption spectroscopy by optical frequency comb

- Measure many of target species by one laser source with very high sensitivity

**Broadband Cavity Enhanced Absorption**

**Optical frequency comb**

Comb’s frequency

\[ f = nf_{\text{rep}} + f_0 \]

(\(f_{\text{rep}}\): interval frequency)

Decided by laser source

**Fabre-Perot Cavity**

Interval of resonance mode frequencies

\[ \Delta f = \frac{c}{2L} \]

(\(c\): speed of light, \(L\): cavity length)

Tuned by piezo actuator (Cavity length tuning)

When the relationship between \(f_{\text{rep}}\) and \(\Delta f\) becomes

\[ mf_{\text{rep}} = \Delta f \]

Comb elements can be stored inside the cavity and experience absorption many times
Broadband cavity enhanced absorption spectroscopy for plasma diagnostics

Ti:Sapphire laser frequency was doubled by BBO crystal (~800 nm → ~400 nm)

DC discharge plasma was generated inside the cavity (Excited species generation)

Figure 1. Experimental set-up for absorption measurements in the glow discharge. BBO, frequency doubling crystal; HR, dichroic mirror for filtering the red; M$_{1,2}$, high reflector mirrors for 370–420 nm; PTZ, piezoelectric actuator; PM, photomultiplier.

**Argon metastable atoms**

Two electronic transitions

**Nitrogen ions**

Rotational distributions

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**Figure 4.** Absorption spectrum of argon glow discharge in the 394 nm range with $R = 99.992\%$ mirrors recorded at the third order of the spectrograph.

**Figure 8.** Absorption spectra of $\text{N}_2^+(B^2\Sigma_u^+; 0 \leftarrow X^2\Sigma_g^+; 0)$ band recorded at the end of the discharge zone ($z = 4$) and at the maximum of the SLA ($z = 18 \text{ cm}$) of a nitrogen microwave plasma.

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- Background & overview of my research
- Introduction of optical frequency comb
- Applications of optical frequency comb

- Frequency-comb interference spectroscopy
  - Brief introduction of research background
  - Principles of FCIS method
  - Absorption profile measurement (Experiments) of argon metastable atoms in small RF plasma

- Concluding remarks
The reason why I was interested in frequency comb was that Prof. K. Tachibana said that he met and did experiments with Prof. J. L. Hall at JILA, Colorado.

Atomic absorption spectroscopy in plasmas
- Densities of excited species, gas temperature & density, etc…
- Requirements of frequency scan of single-frequency laser or meter-scale large spectrometer for getting fine absorption profile when wide-band light source (lamp…) is used
- Mode hop of diode-laser frequency (for high-pressure plasmas)

Novel spectroscopy using interference between optical frequency comb and single-frequency laser
- Frequency-Comb Interference Spectroscopy (FCIS)
1. Frequency-comb laser including absorption frequency of target species transmits through tested plasma source

2. Interference between transmitted frequency comb (FC) and single-frequency (SF) laser set at absorption wavelength

3. Measurement of GHz-range spectrum of beat signals generated by the interference

There is no need to…

scan laser-beam frequency

use large-scale spectrometer

in this frequency-comb interference spectroscopy
Basic concept of FCIS method

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Measurement scheme of FCIS method

1. Target species absorb frequency-comb (FC) elements corresponding to their transition frequencies (Green → Red lines)

2. Absorbed FC light is interfered with single-frequency (SF) laser (Blue sold line)

3. Measure beat signal generated by the interference which shows absorption spectrum with the origin at SF laser frequency

- Single-frequency laser beam
- No target species
- Absorption spectrum

Laser & detected power vs Laser frequency
- Measurement frequency
- Broadenings of atomic absorption spectrum
  - $1 \text{ GHz (Low pressure)} \sim 10 \text{ GHz (AP)}$
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- Broadenings of atomic absorption spectrum → 1 GHz (Low pressure) ~ 10 GHz (AP)
Theoretical description

Laser intensity

\[ I_{IF}(t) = \left| E_{FC}(t) + E_{SF}(t) \right|^2 = \left| \sum_{m=-\infty}^{\infty} E_{FC0}^{(m)} \cos[2\pi (f_A + mf_{rep}) t] + E_{SF0} \cos[2\pi f_{SF} t] \right|^2 \]

Theoretical description

\[ I_{IF}(t) = \left| E_{FC}(t) + E_{SF}(t) \right|^2 = \left| \sum_{m=-\infty}^{\infty} E_{FC0}^{(m)} \cos[2\pi(f_A + mf_{rep})t] + E_{SF0} \cos[2\pi f_{SF} t] \right|^2 \]

\[ = \sum_{m=-\infty}^{\infty} \left( E_{FC0}^{(m)} \cos[2\pi(f_A + mf_{rep})t] \right)^2 + \left( E_{SF0} \cos[2\pi f_{SF} t] \right)^2 + \]

\[ 2 \sum_{i,j=-\infty}^{\infty} E_{FC0}^{(i)} E_{FC0}^{(j)} \cos[2\pi(f_A + mf_{rep})t] \cos[2\pi(f_A + mf_{rep})t] + \]

\[ 2 E_{SF0} \cos[2\pi f_{SF} t] \sum_{m=-\infty}^{\infty} E_{FC0}^{(m)} \cos[2\pi(f_A + nf_{rep})t] \]

\[ = \frac{1}{2} \sum_{m=-\infty}^{\infty} \left( E_{FC0}^{(m)} \right)^2 \left\{ \cos[4\pi f_A + mf_{rep} t] + 1 \right\} + \frac{1}{2} \left( E_{SF0} \right)^2 \left\{ \cos[4\pi f_{SF} t] + 1 \right\} + \]

\[ \sum_{i,j=-\infty}^{\infty} E_{FC0}^{(i)} E_{FC0}^{(j)} \left\{ \cos[2\pi(2f_A + (i + j)f_{rep} t)] + \cos[2\pi(i - j)f_{rep} t] \right\} + \]

\[ E_{SF0} \sum_{m=-\infty}^{\infty} E_{FC0}^{(m)} \left\{ \cos[2\pi(f_A + mf_{rep} + f_{SF} t)] + \cos[2\pi(f_A + mf_{rep} - f_{SF} t)] \right\} \]

Theoretical description

Time variation of light intensity which can be detected in high-speed detector

\[ I_{DET}(t) = \left[ \frac{1}{2} \sum_{m=-\infty}^{\infty} (E_{FC0}^{(m)})^2 + \frac{1}{2} (E_{SF0})^2 \right] + \sum_{i,j=-\infty}^{\infty} E_{FC0}^{(i)} E_{FC0}^{(j)} \cos[2\pi (i - j) f_{rep} t] + \]
\[ E_{SF0} \sum_{m=-\infty}^{\infty} E_{FC0}^{(m)} \cos[2\pi \{ (f_A - f_{SF}) + m f_{rep} \} t] \]

\[ \frac{1}{2} \sum_{m=-\infty}^{\infty} (E_{FC0}^{(m)})^2 + \frac{1}{2} (E_{SF0})^2 \rightarrow \infty \text{ at } f_{BEAT} = 0 \text{ DC component} \]

\[ \sum_{i,j=-\infty}^{\infty} E_{FC0}^{(i)} E_{FC0}^{(j)} \rightarrow \infty \text{ at } f_{BEAT} = |mf_{rep}|, i-j = m \text{ Interference between comb elements} \]

\[ E_{SF0} E_{FC0}^{(m)} \text{ at } f_{BEAT} = |(f_A - f_{SF}) + m f_{rep}| \text{ Interference between frequency comb and SF laser (absorption profile info.)} \]

Optical frequency comb: Ti:Sapphire modelock laser (fs laser)
Single-frequency laser: External-cavity diode laser

Diode-laser wavelength was set at 811.53 nm (Ar 2p_9-1s_5 transition).
RF discharge in Ar flow

- RF power: 13.56 MHz
  1~5 W
- Ar gas flow: 100 mL/min
  20 Torr
- Glass tube: Inner 3 mmφ
  Outer 6 mmφ
- Electrodes: Width 5 mm
  Distance 5 mm
Experimental setup of FCIS measurement

Fast photo detector: **Bandwidth >40 GHz**
Microwave spectrum analyzer: **Band width ~ 100 GHz**
Beat signal of interference inside single-mode fiber
Results (Output signal of spectrum analyzer)

Output of spectrum analyzer (400~500 MHz)

Interference of comb elements:
81 MHz intervals

Interference of FC and SF lasers:
2 peaks between combs’ interference (Higher and lower frequency sides from SF-laser frequency)

1. Record peak intensities of FC&SF interference beat signals
2. Label larger and smaller peaks comparing their peak intensities
3. Plot intensities at center between comb elements’ interference
Results (Data analysis of absorption spectra)

Smaller and larger peak Intensities of FC&SF interference beat signals

Automatic identification of smaller and larger peaks from one spectrum

Smaller signal at -88 dBm indicates there is only one peak between combs’ peaks (-88 dBm is noise level)

This smaller and larger peak spectra correspond to spectra of higher and lower frequency from SF-laser’s frequency
Results (Data analysis of absorption spectra)

Smaller and larger peak

Turn back smaller data points to negative frequency
Results (Ar$^m$ 1s$_5$ atomic absorption spectra)

Comparison of FCIS result with diode-laser absorption method

Diode-laser LAS was performed by external-cavity diode laser changing the laser axis to transmit small plasma cell

Good agreement between two measurement methods

Results (Measurement of frequency shift)

Measurement with small shift of diode-laser frequency

Shift of DL frequency (0.002 nm → 0.9 GHz)

Shift of frequency origin in the beat signal

When the absorption profile shifts, the FCIS method can detect below 0.5 GHz shift without any change of measurement conditions at this time

Doppler shift ~500 m/s

Summary of FCIS plasma diagnostics

- **Frequency-comb interference spectroscopy (FCIS method)**
  - Combination of fs modelock laser and diode laser gave us interference beat-signal spectra between optical frequency comb and single-frequency laser
  - Confirm ~10-GHz band measurement of beat spectra (measurable broadenings of absorption spectra)

- **Application of FCIS to plasma diagnostic**
  - Absorption spectra of Ar metastable atoms in small RF discharge at 20 Torr
  - Spectrum shape of FCIS method was similar to result of diode-laser absorption spectroscopy
  - Potentials to measure small shift in absorption spectrum
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Concluding remarks

- **Optical frequency comb**
  - Characteristics of wide frequency band keeping good coherence consisting of comb elements with a same interval frequency
  - Potentials to further innovation in applied spectroscopy including plasma spectroscopic diagnostics

- **Applications of frequency comb to spectroscopy**
  - Replacement of incoherent broadband light sources for more accurate spectroscopic measurement
  - Novel spectroscopy realized by utilizing its features of broadband (NIR ~ Visible) and good coherence

- **For plasma diagnostics**
  - Broadband cavity enhanced absorption spectroscopy
  - Frequency-comb interference spectroscopy
Development and experimental evaluation

Keiichiro URABE and Osamu SAKAI:
“Absorption spectroscopy using interference between optical frequency comb and single-wavelength laser”,

Theoretical discussion and shift measurement

Keiichiro URABE and Osamu SAKAI:
“Multi-heterodyne interference spectroscopy using probing optical frequency comb and reference single-frequency laser”,
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