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**Authors:** Shinichi Watanabe, Kana Sumihara, Sho Okubo, Makoto Okano, Hajime Inaba

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Supplemental Document

Ultra-precise determination of thicknesses and refractive indices of optically thick dispersive materials by dual-comb spectroscopy

Kana A. Sumihara\textsuperscript{1}, Sho Okubo\textsuperscript{2}, Makoto Okano\textsuperscript{1}, Hajime Inaba\textsuperscript{2} and Shinichi Watanabe\textsuperscript{1,*}

\textsuperscript{1} Department of Physics, Faculty of Science and Technology, Keio University, 3-14-1 Hiyoshi, Kohoku-ku, Yokohama, Kanagawa 223-8522, Japan.

\textsuperscript{2} National Metrology Institute of Japan (NMIJ), National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba, Ibaraki 305-8563, Japan.

*Corresponding author (e-mail: watanabe@phys.keio.ac.jp)

Table of Contents

1. Criterion for the minimum of the evaluation functions

2. Details of the contributions to the measurement uncertainty
1. Criterion for the minimum of the evaluation functions

In this section, we discuss the criterion for the minimum of the sum of the evaluation functions [Eqs. (2) and (3) in main text], which is required for a precise determination of the geometrical thickness \(d\) and the complex refractive index \(N(\omega) = n(\omega) + ik(\omega)\) of the sample. In addition, we explain why the local minima of \(E(M,d)\) appear at an interval of \(\lambda_0/2n_{\text{air}}\) when plotted as a function of \(d\) as shown in Fig. 5(b).

In order to provide a clear description of the problem, we assume that the imaginary part of \(N(\omega), k(\omega),\) is negligible (in the actual analysis of the experimental data, however, we do not neglect the imaginary part). By using this assumption, \(n(\omega)\) can be straightforwardly determined by solving Eq. (10),

\[
n(\omega) = \frac{c}{\omega d} \left( 2\pi M \pm \frac{n_{\text{air}}d}{c} \omega + \theta(\omega) \right), \quad (S1)
\]

where,

\[
\theta(\omega) = \text{Arg} \left(1 - r_{\text{sa}}(\omega)^2 \exp \left( i \frac{2n(\omega)d}{c} \omega \right) \right). \quad (S2)
\]

In our experiment, \(\omega/2\pi \approx 194\) THz and \(d \approx 0.5\) mm. Hence, the exponent on the right-hand side of Eq. (S2) is much larger than \(2\pi\), and thus we rewrite it as

\[
\frac{2n(\omega)d}{c} \omega = \alpha(\omega) + 2\pi K, \quad (S3)
\]

where \(\alpha(\omega)\) is an apparent unwrapped angle, and \(K\) is an integer chosen to satisfy the condition \(0 \leq \alpha(\omega_0) < 2\pi\) at a certain frequency \(\omega_0\). Therefore, \(\Delta \phi(\omega)\) can be written as

\[
- \Delta \phi(\omega) = \text{Arg} \left(1 - r_{\text{sa}}(\omega)^2 \exp(i \alpha(\omega)) \right) - \frac{\alpha(\omega)}{2} + \frac{n_{\text{air}}d}{c} \omega + 2\pi \left( M - \frac{K}{2} \right). \quad (S4)
\]

The graphical representation of Eq. (S2) in the complex plane is depicted in Fig. S1. Since \(\alpha(\omega)\) monotonically increases with \(\omega\), \(\theta(\omega)\) oscillates as a function of \(\omega\). From Eq. (S4), it is found that the oscillation of \(\theta(\omega)\) is the origin of the experimentally observed oscillating behavior of \(\Delta \phi(\omega)\) as a function of \(\omega\) [see Fig. 4(c)] [1]. Thus, by choosing \(M\) and \(d\) in such a way that a synchronized oscillation of \(\Delta \phi(\omega)\) and \(\theta(\omega)\) with the same period and phase is realized, we can minimize \(E(M,d)\), i.e., we can minimize the amplitude of the oscillating component in \(n(\omega)\).
Next, we explain the reason why the local minima of $E(M,d)$ appear at an interval of $\lambda_0/2n_{\text{air}}$ when plotted as a function of $d$. To reveal the $d$ dependence of $E(M,d)$, we replace $d$ with $d_m = d_0 + \frac{m\lambda_0}{2n_{\text{air}}}$ in Eq. (S4), where $m$ is an integer that identifies the position of the local minimum counted from the correct value of $d$, $d_0$. By substituting $d_m = d_0 + \frac{m\lambda_0}{2n_{\text{air}}}$ into Eq. (S4), we obtain

$$-\Delta\phi(\omega) = \text{Arg}\left\{1 - r_{sa}(\omega)^2 \exp\left(i\alpha'(\omega)\right)\right\} - \frac{\alpha'(\omega)}{2} + \frac{n_{\text{air}}d_0}{c}\omega + \frac{m\lambda_0\omega}{2c} + 2\pi\left(M - \frac{K}{2}\right), \quad \text{(S5)}$$

where $\alpha'(\omega)$ is

$$\alpha'(\omega) = \frac{2\omega}{c}d_0\omega - 2\pi K. \quad \text{(S6)}$$

Here, we assume $\frac{\omega\lambda_0}{c} \sim 2\pi$, because the value of $\omega$ is considered to be almost constant under our experimental conditions ($\omega/2\pi$ lies in the range of about 193–198 THz). Under this assumption, and by defining $\alpha''(\omega) = \alpha'(\omega) - 2m\pi$, Eq. (S5) can be rewritten as

$$-\Delta\phi(\omega) = \text{Arg}\left\{1 - r_{sa}(\omega)^2 \exp\left(i\alpha''(\omega)\right)\right\} - \frac{\alpha''(\omega)}{2} + \frac{n_{\text{air}}d_0}{c}\omega + 2\pi\left(M - \frac{K}{2}\right). \quad \text{(S7)}$$

Equation (S7) is equivalent to Eq. (S4). This means that, when $d_0$ minimizes the oscillation of $n(\omega)$ in the vicinity of $d_0$, $d_m$ also locally minimizes the oscillating component in $n(\omega)$ in its vicinity. Thus, the local minima of the evaluation function appear at an interval of $\lambda_0/2n_{\text{air}}$.

Although each $d_m$ corresponds to a local minimum of $E(M,d)$, a different $d_m$ results in a different $n(\omega)$ because of the difference in $\alpha''(\omega)$. When $n(\omega)$ changes, the value of $r_{sa}(\omega)$ slightly changes, and this causes a change in the degree of the agreement between the spectral oscillations of

![Graphical representation of Eq. (S2). $\theta(\omega)$ is plotted in the complex plane for a given $\omega$.](image)
Δφ(ω) and θ(ω). Because the spectral oscillations of Δφ(ω) and θ(ω) show the best agreement for the correct value of d, it is possible to choose the appropriate value of d among the various possible values \(d_m\).

2. Details of the contributions to the measurement uncertainty

| Category | Uncertainty factor | Uncertainty due to \(x_i\) \((k = 1)\) | Conversion factor, \(\partial u(x_i)/\partial x_i\) | Uncertainty due to \(u(x_i)\) \((k = 1)\) |
|----------|--------------------|----------------------------------|---------------------------------------------|----------------------------------|
| Interferometer | Wavelength (optical frequency) of the laser source \(x_1\) | 2.0 × 10^{-13} | \(l_s/n_s\) | negligible |
| Refractive index of air | Temperature measurement of air \(x_2\) | 0.3 K | \((l_s/n_s)(dn_a/dT)\) | 0.04 nm |
| | Pressure measurement of air \(x_3\) | 2 kPa | \((l_s/n_s)(dn_a/dP)\) | 0.79 nm |
| | Pressure measurement of air \(x_4\) | 10% | \((l_s/n_s)(dn_a/dp)\) | 0.02 nm |
| | Temperature fluctuation of air \(x_5\) | 0.02 K | \((L_s - L_2)/n_2\)(dn_a/dT) | 1.58 nm |
| | Pressure fluctuation of air \(x_6\) | 0.003 kPa | \((L_s - L_2)/n_2\)(dn_a/dP) | 0.68 nm |
| | Humidity fluctuation of air \(x_7\) | 0.3% | \((L_s - L_2)/n_2\)(dn_a/dP) | 0.27 nm |
| Sample | Measured temperature of the sample \(x_8\) | 0.3 K | \(dl_s/dT\) | negligible |
| | Alignment (deviation from normal incidence) (θ) \(x_9\) | 0.005 rad | \(l_s(1/n_s-1/\sqrt{n_s^2-n_a^2\sin^2\theta})\) | 0.16 nm |

Total \((k = 1)\) 1.9 nm

Relative \((k = 1)\) \(3.7 \times 10^{-6}\)

- The refractive index of air \((n_a)\) was calculated using Ciddor’s formula. At a temperature of 23 °C, a humidity of 40%, an air pressure of 1013.25 hPa and a CO₂ concentration of 450 ppm, the atmospheric refractive index at a wavelength of 1550 nm is 1.000 265 440 (22).
- Sample thickness \((l_s)\): ∼520 μm
- Refractive index of the sample: ∼3.5
- Measurement time: ∼ 36000 s for the whole measurement process, where in total ∼ 4800 s were spent to measure the interferograms with the sample (∼0.87 s × 5500)
- Optical frequency (vacuum wavelength) used for the measurement: ≈193.414 THz (≈1550.00 nm)
- We assumed that the length difference between the reference path and the sample path \((L_s = 0.3\ m)\) determines the amount of the fluctuations of air.
- \(k\) is the coverage factor.
- *Uncertainty of the refractive index of air at the measurement position when the sample has been removed
- **Fluctuation of the optical path length due to environmental fluctuations in the sample path
- ***Function of the uncertainty factor \(x_9\), not the conversion factor
TABLE 1(b) Contributions to the uncertainty of the value of $n$ measured in this work

| Category                      | Uncertainty factor, $x_i$ | Uncertainty of $x_i$ ($k = 1$) | Conversion factor, $\partial u(x_j)/\partial x_i$ | Uncertainty due to $x_i$, $u(x_i)$ ($k = 1$) |
|-------------------------------|---------------------------|--------------------------------|-----------------------------------------------|-----------------------------------------------|
| Interferometer                | Wavelength (optical frequency) of the laser source $x_1$ | $2.0 \times 10^{-13}$ | 1 | $2.0 \times 10^{-13}$ |
| Refractive index of air       | * Temperature measurement of air $x_2$ | 0.3 K | $dn_a/dT$ | $2.7 \times 10^{-7}$ |
|                               | * Pressure measurement of air $x_3$ | 2 kPa | $dn_a/\rho$ | $5.3 \times 10^{-6}$ |
|                               | * Humidity measurement of air $x_4$ | 10 % | $dn_a/\rho$ | $1.0 \times 10^{-7}$ |
|                               | * Temperature fluctuation of air $x_5$ | $0.02$ K | $((L_s/L_a) - 1) dn_a/\rho$ | $1.1 \times 10^{-5}$ |
|                               | * Pressure fluctuation of air $x_6$ | $0.003$ kPa | $((L_s/L_a) - 1) dn_a/\rho$ | $4.5 \times 10^{-6}$ |
|                               | * Humidity fluctuation of air $x_7$ | $0.3$ % | $((L_s/L_a) - 1) dn_a/\rho$ | $1.8 \times 10^{-6}$ |
| Sample                        | Measured temperature of the sample $x_8$ | 0.3 K | $dn_a/dT$ | $5.6 \times 10^{-5}$ |
| Alignment (deviation from normal incidence) $(\theta)$ $x_9$ | 0.005 rad | 0 | 0 | |

Total ($k = 1$) | $5.8 \times 10^{-5}$ |
Relative ($k = 1$) | $1.7 \times 10^{-5}$ |

TABLE 2(a) Expected contributions to the uncertainty of $d$ in the case of smaller uncertainties for air and the sample temperature

| Category                      | Uncertainty factor | Uncertainty of $x_i$ ($k = 1$) | Conversion factor, $\partial u(x_j)/\partial x_i$ | Uncertainty due to $x_i$, $u(x_i)$ ($k = 1$) |
|-------------------------------|-------------------|--------------------------------|-----------------------------------------------|-----------------------------------------------|
| Interferometer                | Wavelength (optical frequency) of the laser source $x_1$ | $2.0 \times 10^{-13}$ | $l_s$ | negligible |
| Refractive index of air       | * Temperature measurement of air $x_2$ | $0.02$ K | $l_s dn_a/\rho$ | negligible |
|                               | * Pressure measurement of air $x_3$ | $0.1$ kPa | $l_s dn_a/\rho$ | $0.04$ nm |
|                               | * Humidity measurement of air $x_4$ | $1$ % | $l_s dn_a/\rho$ | negligible |
|                               | * Temperature fluctuation of air $x_5$ | $0.02$ K | $l_s dn_a/\rho$ | $1.58$ nm |
|                               | * Pressure fluctuation of air $x_6$ | $0.003$ kPa | $l_s dn_a/\rho$ | $0.68$ nm |
|                               | * Humidity fluctuation of air $x_7$ | $0.3$ % | $l_s dn_a/\rho$ | $0.27$ nm |
| Sample                        | Measured temperature of the sample $x_8$ | $0.02$ K | $d_l/\rho$ | negligible |
| Alignment (deviation from normal incidence) $(\theta)$ $x_9$ | $0.005$ rad | $l_s(1 - n_s/\sqrt{n_s^2 - n_a^2 \sin^2 \theta})$ | *** | $0.16$ nm |

Total ($k = 1$) | $1.7$ nm |
Relative ($k = 1$) | $3.4 \times 10^{-6}$ |
TABLE 2(b) Expected contributions to the uncertainty of n in the case of smaller uncertainties for air and the sample temperature

| Category                   | Uncertainty factor, \(x_i\) | Uncertainty of \(x_i\) \((k = 1)\) | Conversion factor, \(\partial u(x_i)/\partial x_i\) | Uncertainty due to \(x_i, u(x_i)\) \((k = 1)\) |
|---------------------------|-------------------------------|------------------------------------|-------------------------------------------------|-------------------------------------------------|
| Interferometer            | Wavelength (optical frequency) of the laser source \(x_1\) | \(2.0 \times 10^{-13}\) \(n_s\) | \(2.0 \times 10^{-13}\) | \(2.0 \times 10^{-13}\) |
| Refractive index of air   | Temperature measurement of air \(x_2\) | \(0.02\) K \(dn_a/dT\) | \(1.8 \times 10^{-8}\) | \(1.8 \times 10^{-8}\) |
|                          | Pressure measurement of air \(x_3\) | \(0.1\) kPa \(dn_a/dP\) | \(2.6 \times 10^{-7}\) | \(2.6 \times 10^{-7}\) |
|                          | Humidity measurement of air \(x_4\) | \(1\) % \(dn_a/dP\) | \(1.0 \times 10^{-8}\) | \(1.0 \times 10^{-8}\) |
|                          | Temperature fluctuation of air \(x_5\) | \(0.02\) K \((L_s/L_a - 1)(dn_a/dT)\) | \(1.1 \times 10^{-5}\) | \(1.1 \times 10^{-5}\) |
|                          | Pressure fluctuation of air \(x_6\) | \(0.003\) kPa \((L_s/L_a - 1)(dn_a/dP)\) | \(4.5 \times 10^{-6}\) | \(4.5 \times 10^{-6}\) |
|                          | Humidity fluctuation of air \(x_7\) | \(0.3\) % \((L_s/L_a - 1)(dn_a/dP)\) | \(1.8 \times 10^{-6}\) | \(1.8 \times 10^{-6}\) |
| Sample                   | Measured temperature of the sample \(x_8\) | \(0.02\) K \(dn_s/dT\) | \(3.7 \times 10^{-6}\) | \(3.7 \times 10^{-6}\) |
|                          | Alignment (deviation from normal incidence) \((\theta) x_9\) | \(0.005\) rad \(0\) | \(0\) | \(0\) |

Total \((k = 1)\) \(1.2 \times 10^{-5}\)

Relative \((k = 1)\) \(3.5 \times 10^{-6}\)

References

1. L. Duvillaret, F. Garet, and J. L. Coutaz, "Highly precise determination of optical constants and sample thickness in terahertz time-domain spectroscopy," Appl. Optics 38, 409-415 (1999).