Sensitive Measurement of Photon Lifetime and True Reflectances in an Optical Cavity by a Phase Shift Method

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FOR THE COMMANDER

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# SENSITIVE MEASUREMENT OF PHOTON LIFETIME AND TRUE REFLECTANCES IN AN OPTICAL CAVITY BY A PHASE SHIFT METHOD

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- Photon lifetime
- Laser

## Abstract
A simplified method for measuring the effective photon lifetime in an optical resonator was developed. The technique requires the passage of a modulated continuous-wave laser beam through the resonator and the measurement of the resultant shift in the phase of the transmitted intensity. The method not only permits a quick and precise measurement of the mirror reflectances, but also permits these measurements to be in situ. Such an "on the spot" evaluation...
Capability should be extremely useful in applications ranging from the investigation of new laser systems to the development of improved optical coatings. The method is also sensitive to the effects of absorption, scattering, and transmission from elements in the cavity. Cavity losses smaller than 100 ppm were detected.
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I. INTRODUCTION

The precision measurement of optical reflectances or transmittances can be very difficult when either of these quantities approaches unity. It is, however, in this limit of high reflectance or high transmittance that such measurements can be of great importance, particularly with regard to the design and construction of low-gain laser systems, where the critical parameters are often the difference between a mirror reflectance or window transmittance and unity. In this report, a sensitive, simple laboratory method is presented that can be used to make measurements to a precision of better than 100 ppm, thus permitting the testing of mirror reflectivity as high as 99.99% or the determination of single pass gain or loss coefficients, or window transmission losses, to $10^{-4}$.

The technique described herein can be used to measure the effective photon lifetime in an optical resonator. The effective photon lifetime is defined as the characteristic time $\tau$ for the photon energy to be dissipated within the optical resonator to $e^{-1}$ (or 0.34) of its original value. The photon is launched into the carefully aligned resonator on the optical axis. The dissipation of photon energy within the resonator is by absorption, scattering, or transmission at the resonator mirror surface coatings or in the medium between. Thus, the technique is a sensitive and useful method for measuring the scattering, absorption, and reflectivity or transmission, or both, of any solid, liquid, or gaseous material that is a part of, or is introduced into, this resonator.

The photon lifetime is determined from the phase shift in the amplitude-modulated photon flux intensity, which has passed through the optical resonator. A fundamental arrangement of the basic components is shown in Fig. 1. A continuous photon source is passed through a piezo-optical birefringence modulator or photoelastic modulator (PEM), e.g., by Morvue, located at position 1, which produces a time-varying, linearly
Figure 1. Photon Lifetime Measurement Method
polarized photon beam with a \( \sin^2 (2\pi ft \pm \phi_0) \) modulation at \( f = 50 \text{ kHz} \). This modulated beam is then passed through the optical resonator, which consists of two mirrors. The light beam that emerges from the resonator is also modulated but is shifted in phase by \( \alpha \), where the angle \( \alpha \) is related to the time the photon spends in the cavity \( \tau \) by the simple expression
\[
\tan(\alpha) = 4\pi f\tau
\] (1)

This lifetime \( \tau \) corresponds to some number \( n \) of round trips that the photons make within the optical resonator. The time \( \tau' \) to make one round trip is equal to twice the distance \( L \) separating the first and last mirror divided by the speed of light through the medium between mirrors \( c \). Therefore
\[
\tau = n\tau' = \frac{2nL}{c}
\] (2)

For large mirror reflectance \( R \), this lifetime is related to the overall loss of the optical resonator per pass \( 1 - \mathcal{R} \) by the standard relationship
\[
\mathcal{R} = \exp \left( \frac{-\tau'}{\tau} \right) = \exp \left( \frac{-1}{n} \right)
\] (3)

and for the particular resonator shown in Fig. 1
\[
\mathcal{R} = R_1 R_2 \leq 1
\] (4)

The derivation of Eq. (1) is straightforward from the differential equation for flux intensity \( I \) at a given wavelength
\[
\frac{dI}{dt} + \frac{I}{\tau} = k_1 \cos (4\pi ft) + k_2
\] (5)
in which the identity \( \sin^2(x) = 1/2 - 1/2 \cos(2x) \) is used. The constants \( k_1 \) and \( k_2 \) are of no consequence since they do not enter into the solution for the phase shift. Equation (5) is completely analogous to the equation describing an RC circuit with an impressed voltage varying as \( E_0 \cos(4\pi ft) \). For small \( n \) (1 < \( n < 100 \)), a more fundamental deviation involving summations rather than integrations is required. This more complicated analysis, given in the Appendix, yields an even simpler expression for \( \mathcal{R} \):

\[
\mathcal{R} = \frac{n}{(n + 1)} \quad (3a)
\]

It can readily be verified that the expansions of Eqs. (3) and (3a), respectively, differ only in second-order corrections, which are very small for \( n \gg 1 \). Since Eq. (3a) is simpler, it will be used throughout this discussion.
II. EXPERIMENT AND RESULTS

The experimental apparatus is shown in Fig. 1. The phase shift $\phi$ can be determined by means of standard lock-in amplifier techniques in several ways. First, the phases resulting from the modulator being placed in position 1 can be measured and then moved to position 2. The difference between these two measured values is the phase shift $\phi$ resulting from the optical resonator. Second, the modulator can be left in position 1 and a measurement made before and after removing or changing one of the optical elements in the resonator. Again, the difference will yield the change in the lifetime associated with the change in that particular optical element. The third technique requires either two lock-in amplifiers and a ratiometer or a two-channel lock-in amplifier and a ratiometer or servo-loop. (Princeton Applied Research model 5204 is excellent for this application because it has two channels and a ratiometer in one unit.) With the modulator in position 2, the lock-in(s) is adjusted to the appropriate phase to null out signals on both channels A and B. Then, channel B is rotated by 90 deg and the modulator moved to position 1. In this arrangement, the signal in channel A is proportional to the sine of the phase shift $\phi$, whereas that in channel B is proportional to the cosine of the phase shift $\phi$. The outputs from these two channels are then fed into the ratiometer, which reads directly

$$\frac{A}{B} = \frac{\sin(\phi)}{\cos(\phi)} = \tan(\phi)$$  \hspace{1cm} (6)

This technique is convenient for making in situ measurements of optical resonators. The detector gain can be servoed to hold the "in phase" output of the lock-in constant, thereby causing the quadrature channel to read $\tan(\phi)$ directly.
The main application of the setup in Fig. 1 is the measurement of mirror reflectances to very high precision. In this configuration, the single-pass reflectance is

$$ R = R_1 R_2 $$

By the measurement of three $\alpha$'s for the mirrors with reflectances $R_1$, $R_2$, and $R_3$ in the combinations $R_1 R_2$, $R_2 R_3$, and $R_1 R_3$, $R_1$ and $R_2$ can easily be determined. It is also useful to plot $\tan(\alpha)$ versus $L$ and use the slope of the line to determine the value of $R$. Such plots should be linear and extrapolate to the origin (Fig. 2). We have used this technique to measure the reflectivity of mirrors ranging from 95% reflecting to 99.98% reflecting with a precision uncertainty of less than 100 ppm (Fig. 2 and Table I).

Although, in principle, the photon source may be an incoherent, nearly monochromatic, highly collimated beam or a continuous-wave (cw) laser beam of reasonable beam quality, mode matching as well as a spatially coherent source are necessary in order to obtain a linear phase-frequency plot (Fig. 3). The nonlinear phase-frequency response occurs when more than a single mode of the resonant cavity is excited by the laser because the differing longitudinal modes have different photon lifetimes.

In addition, for high-reflectivity mirrors, a laser source is also necessary in order to provide sufficient light on the detector to keep statistical noise low so that good measurement can be made. There is also a certain amount of noise introduced into the measurement as a result of the thermal agitation of the cavity under test mainly because of length variations caused by acoustic forces; however, with a reasonable time constant, this effect is averaged out in the lock-in amplifier.
Figure 2. Tan (\(\alpha\)) vs Intracavity Distance
Table 1. Results of Tests on Several Mirrors of Differing Characteristics

<table>
<thead>
<tr>
<th>Mirror Set</th>
<th>$R_1$</th>
<th>$R_2$</th>
<th>$R_3$</th>
<th>Mirror Radii of Curvature</th>
<th>Wavelength, Å</th>
</tr>
</thead>
<tbody>
<tr>
<td>I</td>
<td>0.9994</td>
<td>0.9997</td>
<td>0.9997</td>
<td>2M, ∞</td>
<td>8742</td>
</tr>
<tr>
<td>II</td>
<td>0.9981</td>
<td>0.9992</td>
<td>0.9989</td>
<td>6M, ∞</td>
<td>6328</td>
</tr>
<tr>
<td>III</td>
<td>0.9957</td>
<td>0.9992</td>
<td>0.9991</td>
<td>0.9987</td>
<td>6328</td>
</tr>
<tr>
<td>IV</td>
<td>0.9983</td>
<td>0.9992</td>
<td>0.9991</td>
<td>6M, 6M</td>
<td>6328</td>
</tr>
<tr>
<td>V</td>
<td>0.9905</td>
<td>0.9952</td>
<td>0.9952</td>
<td>2M, 2M</td>
<td>5288</td>
</tr>
<tr>
<td>VI</td>
<td>0.9925</td>
<td>0.9962</td>
<td>0.9962</td>
<td>2M, ∞</td>
<td>5288</td>
</tr>
</tbody>
</table>
Figure 3. Tan (α) vs Intracavity Distance
By the removal of $R_2$ in Fig. 1, it is obvious that the transmittance $T$ of $R_1$ can be measured by moving $R_1$ in and out of the beam and taking the ratio of the two intensities. In general, calibrated attenuation of the detector will be necessary.

A second configuration (Fig. 4), yields an overall single-pass reflectance

$$W = R_1 R_3^2 R_2$$

(7)

and permits the measurement of any mirror reflectance $R_3$ for a specified polarization when the reflectances of $R_1$ and $R_2$ have been determined. The apparatus can then be packaged and standardized for measuring reflectances. Furthermore, the measurement can be performed for a range of angles of incidence $\theta/2$. The lower curve in Fig. 2 was obtained with the use of this three-mirror configuration with $\theta = 13$ deg. The results of tests on several mirrors of differing characteristics are given in Table I. The self-consistency indicates that the method is accurate within 100 ppm.
Figure 4. Three-Mirror Configuration for Measurement of Photon Lifetime
III. DISCUSSION

The main advantage of this technique in the determination of reflectances and transmittances is that it can be used for in situ laser cavities as a diagnostic and gain measurement, a very significant factor because the reflectances of ultralow-loss mirrors can easily be altered as a result of handling or transport procedures. Further, once a laser cavity is assembled about a potential gain medium, an in situ cavity finesse experiment can be very valuable with respect to predicting the laser threshold condition, particularly since subtle losses in the gain medium such as those that might be caused by turbulent scattering can be evaluated with this method. The method is also useful for cavity tuning and alignment as the phase shift will decrease with detuning because of losses caused by beam walk-off and vignetting on apertures.

Many other applications are envisioned, including very sensitive gas-phase absorption studies and the study of optically transparent materials under high electric or magnetic fields or both. Similar phase-shift methods have also been routinely used with laser-induced fluorescence to measure excited-state lifetimes in atoms and molecules.3

The most important application, however, is the use of the method in studying high reflectances and transmittances or very weak absorptances, where the conventional methods fail. Therefore, the phase-shift method can best be applied to an evaluation of low-loss dielectric coatings and Brewster windows for laser applications. The variability of the phase shift with cavity length and modulation frequency, as well as the photon lifetime in general, always makes it practical to arrange an experimental apparatus to permit a significant and easy-to-measure phase shift. The method is quantitatively accurate inasmuch as the only measurements are length, frequency, and phase, and the results are subject to verification by both the linearity of the phase-length plot and self-consistency checks.
APPENDIX

The analysis is based on the important assumption that an integration over intensities can be performed rather than the standard summation over amplitudes. The use of intensities is justified by the fact that both the probe laser cavity and the test optical cavities are moving and causing the coherent lengths of the light to be greatly reduced. This variation results in random fluctuation in the output on the microsecond time scale that is the result of constructive and destructive interference. Since the phase-sensitive measurement is integrated on the second time scale, however, this variation averages out. Consequently, only intensities are of concern.

Let the incoming light to the optical cavity be given by intensity

\[ I(t) = I_0 \sin(4\pi ft + \phi_0) \]  \hspace{1cm} (A-1)

which is equivalent to the \( \cos(4\pi ft) \) used in Eq. (5). The light that is transmitted at time \( t \) will be the sum of the intensities from different internal reflectors each reduced by the round-trip reflectivity losses \( \mathcal{R} \), i.e.,

\[ I_{out}(t) = TI_0 \sum_{k=1}^{\infty} \mathcal{R}^k \sin[4\pi f(t - k\tau') + \phi_0] \]  \hspace{1cm} (A-2)

where \( \tau' \) represents the round-trip time, and \( T \) is the transmittance after passage through the mirror substrates. It is a reasonably straight-forward exercise to perform the summation of Eq. (A-2) with the following identities \(^5\)

\[ \sum_{k=0}^{\infty} \mathcal{R}^k \sin[(k + 1)\theta] = \frac{\sin(\theta)}{1 - 2\mathcal{R}\cos(\theta) + \mathcal{R}^2} \]  \hspace{1cm} (A-3)
and

$$\sum_{k=0}^{\infty} R^k \cos[(k+1)\theta] = \frac{\cos(\theta) - R}{1 - 2R\cos(\theta) + R^2} \quad (A-4)$$

for $R < 1$. By substituting and rearranging, we obtain

$$I_{out} = T I_0 \frac{\sin(\omega t) - R\sin(\omega t + \omega \tau')}{1 - 2\omega R\cos(\omega \tau') + R^2} \quad (A-5)$$

where $\omega = 4\pi f$ and $\phi_0 = 0$ for convenience. We now use the trigonometric theorem, which states that

$$S = a \sin(\omega t + \alpha) + b \sin(\omega t + \beta) = r \sin(\omega t + \phi) \quad (A-6)$$

where

$$r = (A^2 + B^2)^{1/2}, \quad \tan(\phi) = \frac{B}{A}$$

and

$$A = a \cos(\alpha) + b \cos(\beta) \quad \text{and} \quad B = a \sin(\alpha) + b \sin(\beta)$$

The final expression for the transmitted intensity simplifies to

$$I_{out} = T I_0 \frac{\sin(\omega t + \phi)}{(1 - 2\omega R\cos(\omega \tau') + R^2)^{1/2}} \quad (A-7)$$
where

$$\tan(\phi) = \frac{-\mathcal{H}\sin(\omega T')} {1 - \mathcal{H}\cos(\omega T')} \approx \frac{-\omega T'}{1 - \mathcal{H}} \equiv -\eta \omega T'$$  \hspace{1cm} (A-8)

Equation (A-8) approximates to the simpler form because, for all cases of interest, $\omega T' \ll 1$, and, consequently, $\sin(\omega T') \approx \omega T'$ and $\cos(\omega T') \approx 1$.

Rearrangement of Eq. (A-8) results in the simple expression of Eq. (3a).
REFERENCES

LABORATORY OPERATIONS

The Laboratory Operations of The Aerospace Corporation is conducting experimental and theoretical investigations necessary for the evaluation and application of scientific advances to new military concepts and systems. Versatility and flexibility have been developed to a high degree by the laboratory personnel in dealing with the many problems encountered in the nation's rapidly developing space and missile systems. Expertise in the latest scientific developments is vital to the accomplishment of tasks related to these problems. The laboratories that contribute to this research are:

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