A GENERALIZED THEORY OF TWO-PHOTON FLUORESCENCE MEASUREMENT INCLUDING FLUCTUATIONS AND HIGHER-ORDER CORRELATIONS*

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Abstract

The theory of measurement of mode-locked pulses by means of two photon fluorescence (TPF) is generalized to include simultaneously the effects of amplitude and phase fluctuations of the contributing modes and to include the two photon interactions between photons of different frequency representing higher order correlation measurements. The experimental investigation of the two-time third order intensity correlation measured by the interaction of the original mode-locked pulse and its delayed second harmonic is theoretically analyzed. The unusually high contrast ratio is explained in terms of the second harmonic power conversion coefficient and the total fractional fluctuations. The above analysis is generalized to study TPF displays produced by interaction of the fundamental laser pulse and its n-th harmonic. These displays are shown to recover asymptotically the temporal characteristics of the original laser beam.

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I. Introduction

Recently a considerable amount of attention has been devoted to the study of mode-locked laser radiation[1-2], in particular to the determination of its temporal characteristics. One of the useful techniques for investigating such radiation has been the intensity correlation measurement by means of two photon fluorescence (TPF) or second harmonic generation (SHG)[3]. The resolution limit of the techniques is sufficient to measure pulse durations in the picosecond region. The measurement method and the associated problem of interpreting the correlation data have been well treated theoretically by many workers in the field[4-7]. However, interpretation of the TPF trace still poses considerable ambiguities and in addition the technique per se has a fundamental limitation in that it cannot detect the phase information inherent in the laser radiation. That is to say it cannot detect the asymmetric time dependence of the intensity of mode-locked laser pulses, if any exists, as pointed out by Weber[8]. It was with precisely this limitation in mind that Weber[8-9] proposed several feasible experimental schemes by which to measure the third order intensity correlations. In fact, a special subset of such quantities, namely the two-time third order correlations, was demonstrated experimentally by Rentzepis and Duguay[10]. They obtained, by judicious choice of the dye solution, a TPF trace with an unusually high contrast ratio, which resulted mainly from the cross correlation between fundamental photons and their second harmonics. The significance of these kinds of experiments was discussed by Blount and Klauder[11], who pointed out that the second and third order intensity correlations are sufficient to yield the exact nature of input laser beams. Their analysis, and for that matter most of the existing theories of measurement, are useful only if the mode-locked laser radiation is reproducible, i.e. free of fluctuations.

This is often not the case in practice. Inherently present in the Q-switched mode-locked laser radiation are the phase as well as the amplitude fluctuations for all modes due to the mode competition, temperature fluctuations, the host medium induced frequency chirping[12-13], etc. As is well known, these fluctuations are of great importance in the nonlinear interaction of radiation with matter in general[14] and also play an important role in determining the nature of mode-locked laser beams as discussed in detail by Grütter, Weber, and Dändliker[5,15]. Thus, the effect of fluctuations on the intensity correlation measurement warrants an extensive treatment.

In addition, a detailed analysis of correlations higher than the second order is relevant in view of the associated experiments either performed or proposed. From a purely theoretical point of view, the importance of experiments involving higher order intensity correlations can be seen from the following intuitive argument[16,17]. Consider a mode-locked laser intensity, $I(t)$ having its maximum amplitude at time $t=0$. Typically, an experiment investigating the $(n+1)$-th order correlations consists of measuring the time integrated values of the quantity $I^n(t)I(t+\tau)$. As we go to higher order, i.e. as n is increased, the time dependence of $I^n(t)$ approaches that of a delta function. This is a quite general result valid for a broad class of fields including fluctuations, provided the field has a dominant maximum. Thus it becomes obvious that the time (τ) dependence of the measured correlations duplicates that of $I(t)$ asymptotically, which shows that by measuring higher order correlations the temporal characteristics of the laser radiation can be determined directly.

The purpose of this paper is to extend the existing theories of the intensity correlation measurement of mode-locked laser beams by taking into account the effects of phase as well as amplitude fluctuations and by generalizing our analysis to those multiple photon absorption processes susceptible to experimental verification, namely the cross correlation between fundamental photons and their harmonics. In Section II we introduce a general mode of description for the TPF experiments that will allow us to discuss correlations higher than the second order. In addition the two-time joint probability distribution function for the field is used to calculate the effect of fluctuations on the time (spatial) dependence of the usual second-order two-photon induced fluorescence and on the signal to background ratio for the fluorescence. In Sec. III the third-order intensity correlation function, as measured by Rentzepis and Duguay, is analyzed in great detail. In Sec. IV the whole analysis is generalized to n-th order correlation measurements and the main conclusions of this work are drawn.

II. General Mode of Description

The optical electric field is written, in the usual manner, as

$$E\,(1) = E^{(-)}\,(1) + E^{(+)}(1) \tag{1}$$

$$= \sum_k [\alpha_k e_k\,(1) + \text{c.c.}]\ ; \quad e_k(1) = i(\hbar\omega_k / 2V)^{1/2} \exp -i(\omega_k t_1 - k z_1)$$

The field is considered as a stochastic process, so that the complex mode amplitudes, $\alpha_k = a_k e^{i\varphi_k}$, are determined by the mode distribution of the field, $P(\{\alpha_k\},t)$. We may thus introduce the variance of the k-th mode as one of the fundamental characteristics of the field, viz.,

$$\sigma_k^{\,2} = \langle |\alpha_k - \langle\alpha_k\rangle|^2\rangle \tag{2}$$

Equivalently, we may define the phase and amplitude variations of the k-th mode as

$$(\Delta a_k)^2 = \langle (a_k - \langle a_k\rangle)^2\rangle \tag{2a}$$

$$(\Delta\varphi_k)^2 = \langle(\varphi_k - \langle\varphi_k\rangle)^2\rangle \quad (2b)$$

where the angular brackets denote the expectation value of the quantities. From the outset, we take the phase of each mode to be locked at a mean value, which can be put equal to zero without any loss of generality. However each phase is allowed to fluctuate around this zero mean value, thereby taking into account the temperature fluctuations in the medium, the vibration of resonator mirrors, etc.

A simple experimental configuration for measuring TPF is shown in Fig. 1. Mode-locked laser radiation is split in half by a mirror and impinges collinearly with its image on a suitable dye cell. The resultant field inside the dye at position z is then given by $E(z,t) - E(z, t+\tau)$; τ is the time delay between the two beams and is given by the optical path difference between the two and the minus sign is due to 180°phase difference. The general situation we shall wish to describe arises when the two pulses incident on the dye solution are superpositions of the fundamental beam and its n-th harmonic.

The time integrated fluorescence induced by the incident field can be expressed as

$$\mathcal{F} = \int_{-\infty}^{\infty} dt \sum_{k_1 \ldots k_4} \eta(k_1, \ldots, k_4) \langle\alpha_{k_1}\alpha_{k_2}\alpha^*_{k_3}\alpha^*_{k_4}\rangle \underset{\sim}{e}_{k_1}(1)\underset{\sim}{e}_{k_2}(1)\underset{\sim}{e}^*_{k_3}(1)\underset{\sim}{e}^*_{k_4}(1) \quad (3)$$

Here η denotes the cross-section for two-photon absorption at the frequencies involved. Since the incident field consists, in general, of a superposition of the fundamental beam with its n-th harmonic, the frequencies involved may be widely different, so that η cannot, in general, be taken constant, (as it is for the conventional TPF where the incident beam is just the superposition of the fundamental beam with a delayed replica of itself.) The time dependence of the incident pulse is reflected in the fluorescence trace via the time delay between the two pulses at a given point in the dye solution. In particular, for the case when one pulse is the fundamental and the other is its n-th harmonic, the fluorescence, $\mathcal{F}$, is proportional to the time integral of the two-time (n+1)-th order intensity correlation function of the field, viz., $\langle E^{-}(t+\tau)[E^{-}(t)E^{+}(t)]^{n} E^{+}(t+\tau)\rangle$.

We consider now the simplest type of two-photon fluorescence experiment, where the mode-locked laser beam interacts with itself in a fluorescent dye. Our purpose is to obtain an explicit expression for the time dependence of the fluorescence trace as a function of fluctuations. We can then compare this time dependence to the assumed time dependence of the incident pulse and determine the effects of fluctuations. In addition we shall later compare the expression to similar expressions we shall derive for the more general case of the fluorescence trace resulting from the interaction of a pulse with its n-th harmonic.

The problem of the effect of fluctuations of a mode-locked laser beam on non-linear optics experiments of this general nature have been considered in detail, by Grütter, Weber, and Dändliker[5,15]. In particular, it is shown in ref. [5] that the effect of random phase fluctuations is to reduce the height and change the shape of the central maximum of the second order intensity correlation function of the field; in ref. [15] the effects of phase and amplitude fluctuations on the normalized second moment of the intensity distribution function for the field are considered and it is shown that its value degrades as fluctuations increase. We shall now phrase these results in the language of the TPF experiment in order to provide a basis for comparison with our results for higher order correlations in later sections. If one inserts the total field in the dye solution, $E(z,t)-E(z,t+\tau)$ into Eq. (3) and evaluates the resulting expectation values using the two-time probability distribution function for the field (see Appendix I), the result is

$$\mathcal{F}(\tau) = C \int dt \; \{ |\langle E(t)\rangle|^4 + |\langle E(t+\tau)\rangle|^4 + 4(\sigma^2(t)\,|\langle E(t)\rangle|^2 + \sigma^2(t+\tau)\,|\langle E(t+\tau)\rangle|^2)$$

$$+ 4[\,|\langle E(t)\rangle|^2 |\langle E(t+\tau)\rangle|^2 + \sigma^2(t)\,|\langle E(t+\tau)\rangle|^2 + \sigma^2(t+\tau)\,|\langle E(t)\rangle|^2$$

$$+ \sigma(t)\sigma(t+\tau)(a\langle E(t)\rangle \; \langle E^*(t+\tau)\rangle + c.c.)]\} \qquad (4)$$

Eq. (4) is a general expression for TPF, including the effects of the phase and amplitude fluctuations, which are contained in $\sigma^2(t)$ and $\sigma^2(t+\tau)$. (Note that for the ideal case of $\sigma^2(t) = \sigma^2(t+\tau) = 0$, the above gives $\mathcal{F}(\tau{=}0)/\mathcal{F}(\tau{=}\infty) = 3:1$.)

The discussion of Eq. (4) becomes much more explicit if we specify at this point the characteristics of the input field, i.e., if we modelize the field. We assume:

$$\langle E(t)\rangle = \mathcal{E}_o \exp - [i\,\omega_o t + \tfrac{1}{2}(\Delta\Omega t)^2] \qquad (5)$$

This is consistent with the model of mode-locked laser radiation based on the regenerative pulse generator, first considered by Cutler[20] and recently elaborated by DeMaria et al[1] and Kuizinga and Siegman[21]. They showed that the self-consistency requirement at a steady state for mode-locked laser pulses can be satisfied by an optical field[22] having an average Gaussian spectral profile.

We further assume that the distribution function describing this steady state optical pulse is factorizable into functions of amplitudes $\{a_k\}$ and phases $\{\varphi_k\}$. Then the k-th mode variance can be written as

$$\sigma_k{}^2 = \langle a_k\rangle^2 f_k \qquad (6)$$

where

$$f_k = [(\Delta a_k)^2/\langle a_k\rangle^2] + [1-\langle\cos\varphi_k\rangle^2 - \langle\sin\varphi_k\rangle^2] \qquad (6a)$$

$$\simeq [(\Delta a_k)^2/\langle a_k\rangle^2] + (\Delta\varphi_k)^2 \text{ ; for small } \Delta\varphi_k \text{ and } \langle\varphi_k\rangle = 0 \ . \qquad (6b)$$

represents the total normalized variance resulting from amplitude as well as phase fluctuations. Although the amplitude contribution is rather small for strong optical fields, the phase fluctuations can be substantial due to frequency chirping as shown by Svelto[13]. If we additionally assume that the normalized variance f_k has a uniform value f over the entire field spectrum, we get

$$\sigma^2(t) = \sigma^2(t+\tau) = \mathcal{E}_o{}^2 f \qquad (7)$$

Upon inserting Eq. (5) and Eq. (7) into Eq. (4) and carrying our the time integration, we obtain

$$\mathcal{F}(\tau) = \mathcal{F}_o[1 + 2e^{-\frac{1}{2}(\Delta\Omega\tau)^2} + 8\sqrt{2}\ f\ (1 + \tfrac{1}{2}e^{-\frac{1}{4}(\Delta\Omega\tau)^2})] \qquad (8)$$

Note that the time difference, τ, can easily be transcribed into a spatial argument of the fluorescence pattern.

It is clear that the results derived earlier[5,15] regarding the effects of fluctuations on nonlinear interactions of a light beam with itself are implicit in Eq. (8). The presence of fluctuations introduces a new term in the fluorescence expression having a different time scale. Hence the time dependence of the TPF pattern undergoes a rather drastic change from a Gaussian to a non-Gaussian nature. (See. Fig 3) If we compute the contrast ratio as a function of fluctuations,

$$\mathcal{R} \equiv \mathcal{F}(\tau=0)\ /\ \mathcal{F}(\tau=\infty)$$

$$= [3 + 12\sqrt{2}\ f]\ /\ [1 + 8\sqrt{2}\ f] \qquad (9)$$

we see that the presence of fluctuations causes the contrast ratio to degrade from a value of 3:1 for $f = 0$, to a value of 1.5:1 for $f = \infty$. This corresponds to the transition from perfectly mode-locked radiation to a Gaussian or thermal field with the phases randomly distributed in the interval $(0, 2\pi)$. We have plotted in Fig. 2 the contrast ratio versus the total fractional fluctuation f. Recently, Picard and Schweitzer[7] and Harrach[4] showed that the ratio depends in a complicated but interesting way on the degree of mode-locking in the incoming laser pulses. In their analysis, oscillating modes were divided into two portions, namely the central group having a common phase and the two outer wings having random phases. The degree of mode locking was defined as the ratio of the width of central group to the total bandwidth. The measured values of the ratio that lie between the two limits 1.5 and 3 could thus be explained in terms of this degree of mode-locking. Eq. (9) indicates that the ratio depends significantly on the fluctuations as well, rendering the interpretation of experimental results more complicated. A few comments are due at this point. The assumed phase fluctuation $(\Delta\varphi)^2$ provides one way of describing imperfectly mode-locked laser radiation. Although this model is compatible with the analysis of Cutler[20] and of Kuizenga and Siegman[21], it is not yet clear which better approximates the actual laser pulses: the degree of mode-locking, the domain model or the fractional fluctuation f. It should be emphasized however that the uniform f is introduced in our analysis only for the mathematical simplicity and is not an essential assumption. Thus, by properly adjusting the k-th mode variance, σ_k^2, we can not only incorporate the other two models for laser pulses in the description of TPF but also can include the effect of fluctuations at the same time.

III. TPF of Third Order Correlations

In this section we consider the two-time, third order intensity correlation functions of the field. Measurements of third order intensity correlations by means of a third harmonic generation is treated in detail by Pike and Hercher[25]. The third order correlations that are considered in this section arise from the interaction between the fundamental laser field and its second harmonic and were first demonstrated experimentally by Rentzepis and Duguay[10]. Subsequently, Weber proposed several schemes to measure the two and three time third order intensity correlation functions, so that the phase information of the field could be recovered. We now analyze in detail the type of experiment performed by Rentzepis and Duguay and indicate precisely what information can be obtained from such an experiment.

The experimental scheme is as depicted in Fig. 4. A laser pulse, say 1.06μm Nd:glass, is passed through a nonlinear material like KDP and frequency upconverted to .53μm. The radiation field emerging from the crystal then consists of a superposition of the fundamental frequency pulse and its in-phase second harmonic. These two pulses are next separated in time (or distance) by passing the beam through another material such as bromobenzene. This resultant beam is then used in the normal TPF configuration to produce a fluorescence display. By choosing a dye solution with transition frequency greater than twice the fundamental 1.06μm frequency, Rentzepis and Duguay confined the two photon absorption processes to those involving two second harmonic photons or one second harmonic and one fundamental photon. This has an interesting effect on the TPF display as will become clear later on.

We proceed with a theoretical consideration of second harmonic generation (SHG), including the effect fundamental beam fluctuations have on the second harmonic beam. We then impose the Gaussian model on the incoming fundamental beam, calculate the second harmonic explicitly and derive an expression for the TPF display caused by the fundamental-second harmonic interaction in the dye solution.

The c-number second harmonic complex mode amplitude α_p with frequency ω_p is given in terms of the fundamental modes by[24]

$$\alpha_p = \gamma \sum_n \alpha_n \alpha_{p-n} S(p,n) \; ; \; \gamma = i\pi \left(\frac{\hbar\omega_o}{V} \right)^{\frac{1}{2}} \frac{\omega_p dL}{cn(\omega_p)} \qquad (10)$$

where d and n represent the effective nonlinear coefficient and the index of refraction of the medium respectively. The gain profile factor

$$S(p,n) = [e^{i\,\Delta k(p,n)L} - 1] / \, i[\Delta k(p,n)L] \qquad (11)$$

is given in terms of the crystal length L and the phase detuning factor $\Delta k(p,n)$,

$$\Delta k(p,n) = k(\omega_p) - k(\omega_n) - k(\omega_p - \omega_n) \qquad (12)$$

and describes the net effective interaction among these three modes during the transit time. To first order in the index of refraction, it can be easily shown that $S(p,n)$ is independent of ω_n and thus can be taken out of the summation in (10).

We now specify the fundamental beam to be of the Gaussian form with uniform normalized fluctuation, i.e., a common fractional amplitude variance $(\Delta a)^2/\langle a\rangle^2$ and the same phase variance $(\Delta\varphi)^2$. (See Eq. (7).) Then the expectation value of the second harmonic amplitude is given by

$$\begin{aligned}\langle a_p\rangle &\simeq \gamma[2\pi(\Delta\Omega)^3]^{-1} \int_{-\infty}^{\infty} d\omega \exp -\{[(\omega-\omega_o)^2/2(\Delta\Omega)^2] + [(\omega_p-\omega-\omega_o)^2/2(\Delta\Omega)^2]\}\, S(p,n) \\ &= \gamma[2\sqrt{\pi}(\Delta\Omega)^2]^{-1} \exp -[(\omega_p-2\omega_o)^2/2(\sqrt{2}\Delta\Omega)^2]\{[e^{i\kappa L(\omega_p-2\omega_o)}-1]/[i\kappa L(\omega_p-2\omega_o)]\}\end{aligned} \qquad (13)$$

where we have replaced the discrete summation over modes by an integral. Note that the resulting harmonic spectral profile is also a Gaussian, aside from the factor S, with half width twice that of the fundamental. Taking the inverse Fourier transform of Eq. (13) we obtain the time domain representation of the second harmonic pulse as

$$\langle E_h(t)\rangle = (2\pi)^{-1} \int_{-\infty}^{\infty} d\omega_p \, e^{-i\omega_p t} \langle a_p \rangle$$

$$= \mathcal{E}_h \, e^{-2i\omega_o t} \{\mathrm{erf}\,(\Delta\Omega\, t) - \mathrm{erf}\,[\Delta\Omega\,(t-\varkappa L)]\} \tag{14}$$

The resolution problem associated with the second harmonic generation and the effect of crystal length in its generation was considered explicitly by Mathieu and Keller[26]. There are two obvious limiting cases for the above time-dependence of the second harmonic pulse[24]. (See Fig. 5) The one we are interested in is $(\Delta\Omega\varkappa L) \ll 1$. In this limit we can expand $\mathrm{erf}[\Delta\Omega(t-\varkappa L)]$ in a Taylor series centered at $\Delta\Omega t$ and obtain

$$\langle E_h(t)\rangle = \mathcal{E}_h e^{-2i\omega_o t} (2/\sqrt{\pi})\, e^{-(\Delta\Omega t)^2} \sum_{m=0}^{\infty} (\Delta\Omega\varkappa L)^{m+1} H_m(\Delta\Omega t) \tag{15}$$

where H_m denotes the m-th order Hermite polynomial. In this limit the harmonic pulse shape resumes a Gaussian form with half-width equal to one half that of the fundamental pulse. Physically, this means that the gain profile is broad enough to convert the entire fundamental spectrum into its second harmonic. In the process, the width of the harmonic profile is doubled, so the time domain representation is halved. The cross correlation between the fundamental and the second harmonic will now yield the maximum information on the original mode-locked laser radiation.

To obtain the variance of the second harmonic mode, σ_p^2 we begin by expressing it in terms of $(\Delta a_p)^2$ and $(\Delta\varphi_p)^2$, viz.

$$\sigma_p^{\,2} = \langle |\alpha_p - \langle\alpha_p\rangle|^2\rangle$$

$$= (\Delta a_p)^2 + \langle a_p\rangle^2 \,[1 - \langle\cos\varphi_p\rangle^2 - \langle\sin\varphi_p\rangle^2] \tag{16}$$

Now, the amplitude and phase fluctuations can be expressed in terms of those of fundamental modes from Eq. (10), which can be rewritten as

$$a_p \exp(-i\varphi_p) = \gamma\, S(p) \sum_n a_n\, a_{p-n} \exp - i(\varphi_n + \varphi_{p-n}) \tag{17}$$

Since the phase fluctuations in the fundamental modes are modelized to have a common value $(\Delta\varphi)^2$, it follows immediately from Eq. (17) that

$$(\Delta\varphi_p)^2 = 2(\Delta\varphi)^2 \tag{18}$$

Also, the expectation values of a_p and $a_p^{\,2}$ follow readily from Eq. (17):

$$\langle a_p\rangle = \gamma\, S(p) \sum_n \langle a_n\, a_{p-n}\rangle$$

$$= \gamma\, S(p) \sum_n \langle a_n\rangle\langle a_{p-n}\rangle + (\Delta a_{p/2})^2 \tag{19}$$

$$\langle a_p^{\,2}\rangle = \gamma^2 S^2(p)\,[\sum_{nn'} \langle a_n\rangle\langle a_{p-n}\rangle\langle a_{n'}\rangle\langle a_{p-n'}\rangle$$

$$+ 2\sum_n \langle a_n\rangle\langle a_{p-n}\rangle\,(\Delta a_{p/2})^2$$

$$+ 4\sum_n (\langle a_n^{\,2}\rangle\langle a_{p-n}^{\,2}\rangle - \langle a_n\rangle^2\langle a_{p-n}\rangle^2)]$$

$$+ (\sigma^4) \tag{20}$$

where we have used the factorizability of the $P(\{\alpha_n\},t)$ function to obtain Eqs. (19) and (20). Thus, using the relation, $\langle a_n^{\,2}\rangle = \langle a_n\rangle^2 + (\Delta a_n)^2$, we obtain from Eqs. (19) and (20)

$$(\Delta a_p)^2 = 4\gamma^2 S^2(p) \sum_n [(\Delta a_n)^2 \langle a_{p-n}\rangle^2 + (\Delta a_{p-n})^2 \langle a_n \rangle^2] \tag{21}$$

Now, using Eqs. (21), (19) and (16), and replacing the discrete summation over modes in (21) with an integral, we obtain

$$\sigma_p^2 = \langle a_p \rangle^2 f' \tag{22}$$

with

$$f' = 2\{(2/\pi)^{\frac{1}{2}} [(\Delta a)^2/\langle a \rangle^2] + [1-\langle \cos \varphi \rangle^2 - \langle \sin \varphi \rangle^2]\}$$

$$\simeq 2\{(2/\pi)^{\frac{1}{2}} [(\Delta a)^2/\langle a \rangle^2] + (\Delta\varphi)^2\} \tag{23}$$

which can be compared with Eq. (6). The effect of second harmonic generation is then to increase the magnitude of the fluctuations by about 50%.

We now proceed to calculate the TPF display produced by the cross correlation of the fundamental with the narrow second harmonic. The total radiation field entering the TPF dye is

$$E(t) = E_f^+(t) + E_h^+(t) + E_f^-(t) + E_h^-(t)$$

$$= \sum_k [\alpha_k \underset{\sim}{e}_k(1) + \alpha_k^* \underset{\sim}{e}_k^*(1)] + \sum_p [\alpha_p \underset{\sim}{e}_p(1) + \alpha_p^* \underset{\sim}{e}_p^*(1)] \tag{24}$$

The determination of the TPF display begins from Eq. (3). Upon inserting Eq. (24) in Eq. (3) a total of 256 terms ensue. Fortunately a substantial reduction occurs due to the rapid oscillatory nature of many terms. A further reduction results if we choose, following Rentzepis and Duguay, a dye solution whose energy gap is wide enough to exclude absorption of two photons in the fundamental frequency range. So putting $\eta(\omega_o,\omega_o) = 0$, $\eta(\omega_o,2\omega_o) = 1$, $\eta(2\omega_o,2\omega_o) = 1$ in Eq. (3) we obtain

$$\mathcal{F}(\tau) = C\int_{-\infty}^{\infty} dt\{\langle|E_h(t)|^4\rangle+\langle|E_h(t+\tau)|^4\rangle+4[\langle|E_f(t)|^2|E_h(t)|^2\rangle+\langle|E_f(t+\tau)|^2|E_h(t+\tau)|^2\rangle$$

$$+\langle|E_f(t)|^2|E_h(t+\tau)|^2\rangle+\langle|E_f(t+\tau)|^2|E_h(t)|^2\rangle+\langle|E_h(t)|^2|E_h(t+\tau)|^2\rangle]\} \quad (25)$$

An additional simplification of Eq. (25) results if the fundamental and second harmonic pulses are separated from one another by a time delay, τ_o, greater than the individual pulse widths. This was the case in the experiment of Rentzepis and Duguay. Then the contribution from the third and fourth terms in Eq. (25) is proportional to $e^{-(\Delta\Omega\tau_o)^2}$ and hence can be neglected.

Each term appearing in Eq. (25) represents only a portion of the entire radiation field inside the dye solution, and the ensemble average can not be obtained by using the corresponding $\mathcal{W}$-function. Instead, the average values should be calculated directly from the field distribution function $P(\{\alpha_k\},t)$. Since the details of calculations are of no importance to us at this point, we relegate it to Appendix II and merely quote the results here. For the uniform normalized amplitude variance and the common phase fluctuation we assumed for the input laser beam we can obtain

$$\langle |E_h(t)|^4\rangle = |\langle E_h(t)\rangle|^4 + 8f'\mathcal{E}_h{}^2|\langle E_h(t)\rangle|^2 + (\sigma^4) \tag{26}$$

$$\langle |E_f(t)|^2|E_h(t+\tau)|^2\rangle = |\langle E_f(t)\rangle|^2|\langle E_h(t+\tau)\rangle|^2 + 4f\mathcal{E}_o{}^2|\langle E_h(t+\tau)\rangle|^2 + 8f'\mathcal{E}_h{}^2|\langle E_f(t)\rangle|^2 + (\sigma^4) \tag{27}$$

$$\langle |E_h(t)|^2|E_h(t+\tau)|^2\rangle = |\langle E_h(t)\rangle|^2|\langle E_h(t+\tau)\rangle|^2 + 2f'\mathcal{E}_o{}^2[|E_h(t)\rangle|^2 + |\langle E_h(t+\tau)\rangle|^2] + 2f'\mathcal{E}_h{}^2[e^{-2i\omega_o\tau}\langle E_h(t)\rangle\langle E_h^*(t+\tau)\rangle + c.c.] + (\sigma^4) \tag{28}$$

Upon inserting Eqs. (26), (27), (28) and (15) in Eq. (25) and carrying out the time integration we obtain up to the first power of $\Delta\Omega\kappa L$

$$\mathcal{F}(\tau) = \mathcal{F}_o[(2/\sqrt{3})e^{-\frac{2}{3}(\Delta\Omega\tau)^2} + \beta(\tfrac{1}{2}e^{-(\Delta\Omega\tau)^2} + 2\sqrt{2}fe^{-\frac{1}{2}(\Delta\Omega\tau)^2}) + \tfrac{1}{4}\beta + 2\sqrt{2}f(\beta+2) + 2f'(8+\sqrt{2})], \tag{29}$$

where β is the power conversion coefficient for second harmonic generation and is defined by

$$\frac{4}{\pi}(\Delta\Omega\kappa L)^2\,\mathcal{E}_h{}^2 = \beta\,\mathcal{E}_o{}^2\,. \tag{30}$$

We have derived in Eq. (29) the expression for the TPF display caused by the two-time third order intensity correlation function of the type measured experimentally by Rentzepis and Duguay. Since the power conversion factor, β, and the total normalized fluctuations, f and f', are small in most cases, it is apparent that the signal to background ratio for this third order correlation is better than that attained by conventional TPF. A quantitative estimation for the fluctuations can be obtained by applying Eq. (29) to the experimental results of Rentzepis and Duguay. They report a second harmonic conversion coefficient $\beta \simeq .001$ and the contrast ratio of approximately 10. Substituting these values in Eq. (29) we obtain $f \simeq f' \simeq (\Delta\varphi)^2 = .005$, since the fractional amplitude fluctuation can be neglected for strong radiation fields. For purposes of comparison, let us neglect the fluctuation and insert $\beta = .001$ in Eq. (29). Then we obtain easily the value for the contrast ratio, $R \simeq 5 \times 10^4$. Thus, normalized fluctuations of the order of one half percent cause the contrast ratio to decrease by several orders of magnitude. We note, however, that even with the presence of fluctuations, the signal to background ratio is considerably higher for this third order correlation technique than for the conventional TPF.

The role played by the fluctuations is essentially the same for this type of TPF, as for the conventional TPF, i.e., it increases the background fluorescence and obscures the signal portion of the display. However, by judicious choice of the experimental situation, one can cause the fluctuations to be suppressed to a great extent. In the TPF configuration for the field consisting of the superposition of fundamental and second harmonic pulses one can select only the fundamental pulse in the one arm by means of polarization, interference filter, or spectral displacement prism. (See Fig. 4) This together with the dye solution with transition frequency greater than the twice fundamental frequencies can virtually confine the TPF display to the cross correlation between the fundamental and the second harmonic beams, i.e. $\eta(\omega_o,\omega_o) = \eta(2\omega_o,2\omega_o) = 0$, $\eta(\omega_o,2\omega_o) = 1$. In this case Eq. (29) reduces to

$$\mathcal{F}(\tau) = \mathcal{F}_o[(1/3)\, e^{-\frac{2}{3}(\Delta\Omega\tau)^2} + 2\sqrt{2}\, f + 8f'] \tag{31}$$

It is quite evident from Eq. (31) that the background fluorescence is even more reduced than above and that the time dependence of the display reflects the Gaussian nature of the input beam, although the time scale is different by the factor 2/3. (See Fig. 6) These facts clearly point the way to considering cross correlations between the fundamental and even higher harmonics in hopes of recovering the time dependence of the original mode-locked laser emission directly in the time domain.

V. Generalizations and Conclusions

In this section we generalize our analysis to calculate the TPF display caused by the interaction of the fundamental laser pulse with its n-th harmonic. In view of the success of nonlinear optics researchers in growing crystals with increasingly large nonlinear susceptibilities, and given the present ready availability of extremely intense mode-locked lasers, one can envision the possibility of generating harmonic short pulses of sufficient intensity higher than the second order. In such an event the higher order cross correlations would become quite practical.

The amplitude of the n-th harmonic mode with frequency ω_{p_n} can be constructed from the fundamental modes via a straightforward generalization of Eq. (10):

$$\alpha_{p_n} = \gamma_n \sum_{k_1 \ldots k_{n-1}} \alpha_{k_1}\alpha_{k_2} \cdots \alpha_{k_{n-1}}\alpha_{p-(k_1+\ldots k_{n-1})}, \tag{32}$$

where γ_n is the corresponding conversion coefficient for n-th harmonic generation and we have confined ourselves, for simplicity, to the case where the phase detuning, Δk, is zero, i.e. S = 1.

We can similarly easily generalize our earlier result for the fluctuations of the second harmonic to obtain (See Eq. (23))

$$\sigma_{p_n}^{2} = \langle a_{p_n} \rangle^2 f_n \tag{33}$$

where the fractional fluctuation is given by

$$f_n = n[(\Delta\varphi)^2 + \frac{1}{n} \langle a_{p_n} \rangle^{-2} \sum_{\pi(\{k\},\{k'\})} \sum_{\substack{k_1,\ldots k_{n-1} \\ k'_2 \ldots k'_{n-1}}} (\Delta a_{k_1})^2 \langle a_{k_2} \rangle \ldots \langle a_{k_{n-1}} \rangle \langle a_{p-(k_1+\ldots+k_{n-1})} \rangle$$

$$\langle a_{k'_2} \rangle \ldots \langle a_{p-(k_1+k'_2+\ldots+k'_{n-1})} \rangle] \tag{34}$$

Here, $\pi(\{k\},\{k'\})$ denotes sum over all possible pair combinations between one member in group $\{k\}$ and another in $\{k'\}$. Using once more the Gaussian model for the incoming laser pulse, we can calculate the expectation value of the spectral profile of the n-th harmonic:

$$\langle a_{p_n} \rangle = \int dk_1 \ldots dk_{n-1} \langle a_{k_1} \rangle \ldots \langle a_{k_{n-1}} \rangle \langle a_{p_n-(k_1+\ldots+k_{n-1})} \rangle$$

$$= [2\pi n(\Delta\Omega)^2]^{-\frac{1}{2}} \exp-[(\omega_{p_n} - n\omega_o)^2/2(\sqrt{n}\,\Delta\Omega)^2] \tag{35}$$

Taking the inverse Fourier Transform of Eq. (35) we find the time dependence of the n-th harmonic to be

$$\langle E_h^{(n)}(t) \rangle = \mathcal{E}_h^{(n)} e^{-ni\omega_o t} e^{-\frac{1}{2}n(\Delta\Omega t)^2} \tag{36}$$

And now making the same assumptions that were used to arrive at Eq. (31), we easily calculate the fluorescence display caused by the interaction of the fundamental and its n-th harmonic to be

$$\mathcal{F}(\tau) = \mathcal{F}_o\{(n+1)^{-\frac{1}{2}} \exp-[n(\Delta\Omega\tau)^2/(n+1)] + A(f_n + f\, n^{-\frac{1}{2}})\} \qquad (37)$$

The above equation clearly shows that the time dependence of the TPF display asymptotically approaches that of the original laser beam, which in this case is a Gaussian pulse. (See Fig. 7) In addition, it is clear that the background noise terms are due solely to fluctuations present in the original beam.

In conclusion, we have analyzed in this paper the experiments for measuring the intensity correlations of a mode-locked laser radiation. In doing so we have generalized the TPF scheme to incorporate the two-time n-th order correlations. In the whole analysis the effects of fluctuations present in the laser radiation are taken into account in an explicit and consistent way. We have shown that the fluctuations affect the contrast ratio of the ordinary TPF significantly and that they complicate the time dependence of the signal portion of the pattern. Finally we have pointed out the interesting theoretical possibility that by measuring the TPF trace caused by the cross correlation between the fundamental and harmonic beams in a dye solution properly chosen one can practically recover the temporal characteristics of the input laser pulses. This argument holds true no matter what the original shape of the laser pulse is, since ideally the n-th order harmonic approaches a delta function as n grows large. In practice however, the experimental realization of this analysis is limited by the difficulties encountered in generating higher order harmonic pulses with ultrashort duration. This puts rather stringent requirements on the material properties as was discussed in Section V.

Appendix I

In this appendix we derive Eq. (4) in the text. The total field in the dye is given by $E(z,t) - E(z,t+\tau)$. Thus, as shown in great detail elsewhere[18], we may obtain the expression for TPF from Eqs. (1) and (3) as

$$\mathcal{F}(\tau) = C\int_{-\infty}^{\infty} dt\ [\langle |E(t)|^4\rangle + \langle |E(t+\tau)|^4\rangle + 4\langle E^-(t)E^-(t+\tau)E^+(t+\tau)E^+(t)\rangle] \qquad \text{(I-1)}$$

The ensemble average of the quantities appearing in Eq (I-1) can be compactly described, including explicitly the effect of fluctuations by means of the one-time and two-time quasiprobability distribution function of the field. The two-time joint probability function[19] for the field is defined in terms of the probability function for the mode amplitudes, $P(\{\alpha_k\},t)$ by

$$W(\mathcal{E}_1,\mathcal{E}_2) = \int d^2\{\alpha_k\}P(\{\alpha_k\},t)\,\delta^2[\mathcal{E}_1 - \sum_k \alpha_k \underset{\sim}{e}_k(1)]\,\delta^2[\mathcal{E}_2 - \sum_k \alpha_k \underset{\sim}{e}_k(2)] \qquad \text{(I-2)}$$

with

$$\mathcal{E}_i = \sum_k \alpha_k \underset{\sim}{e}_k(i) \qquad \text{(I-3)}$$

denoting the total complex field amplitude at t_i and z_i. The expectation value of a measurable quantity $\mathfrak{M}(\mathcal{E}_1,\mathcal{E}_2)$ can then be given in terms of $W(\mathcal{E}_1,\mathcal{E}_2)$ by

$$\langle \mathfrak{M}(\mathcal{E}_1,\mathcal{E}_2)\rangle = \int d^2\mathcal{E}_1\ d^2\mathcal{E}_2\ \mathfrak{M}(\mathcal{E}_1\mathcal{E}_2)\ W(\mathcal{E}_1,\mathcal{E}_2) \qquad \text{(I-4)}$$

For fields consisting of many modes, which is the case for mode-locked laser pulses, we can invoke an argument similar to the central limit theorem and construct, to a good approximation, the two-time field distribution function in terms of its first two moments in the rotating wave approximation, viz.

$$W(\mathcal{E}_1,\mathcal{E}_2) = \mathfrak{N}^{-1} \exp - [|\xi_1|^2 + |\xi_2|^2 - a\xi_1\xi_2^* - a^*\xi_1^*\xi_2] \qquad \text{(I-5)}$$

Here the dimensionless variable

$$\xi_i = [\mathcal{E}_i - \langle E(z,t_i)\rangle]/[\Lambda\ \sigma(t_i)]\ ;\ i = 1,2\ . \tag{I-6}$$

represents the departure of the fluctuating total field amplitude $\mathcal{E}_i$ from its mean value at t_i, scaled over the total effective variance at time t_i of the field

$$\sigma^2(t_i) = \sum_k |\underset{\sim}{e}_k(t_i)|^2 \langle|\alpha_k(t_i) - \langle\alpha_k(t_i)\rangle|^2\rangle \tag{I-7}$$

$$= \sum_k |\underset{\sim}{e}_k(t_i)|^2\ \sigma_k^{\ 2}(t_i)$$

and the parameter

$$\Lambda^2 = 1 - |a|^2 \tag{I-8}$$

with

$$a = [\sum_k \underset{\sim}{e}_k^*(t_1)\underset{\sim}{e}_k(t_2)\ \sigma_k(t_1)\ \sigma_k(t_2)]/\sigma(t_1)\ \sigma(t_2) \tag{I-9}$$

representing the degree of correlation of the field between t_1 and t_2. η is the normalization constant and is given by $\eta = [\pi\Lambda\sigma(t_1)\ \sigma(t_2)]^2$.

By combining Eqs. (I-1), (I-4), and (I-5) one can easily obtain Eq. (4) in the text.

Appendix II

The ensemble average of some functions of fundamental or second harmonic pulses can be calculated directly from the mode distribution function $P(\{\alpha_k\},t)$. To show this we derive Eq. (26) in detail here. The harmonic pulse, $E_h(t)$, can be expressed in terms of its modes as

$$E_h(t) = \sum_k \alpha_k \underset{\sim}{e}_k(1) \tag{II-1}$$

Thus, we have

$$|E_h(t)|^4 = \sum_{k_1,\ldots k_4} \alpha_{k_1}\alpha_{k_2}\alpha^*_{k_3}\alpha^*_{k_4} \underset{\sim}{e}_{k_1}(1)\, \underset{\sim}{e}_{k_2}(1)\, \underset{\sim}{e}^*_{k_3}(1)\, \underset{\sim}{e}^*_{k_4}(1) \tag{II-2}$$

By using the $P(\{\alpha_k\},t)$ - function we can take the average value of Eq. (II-2) and obtain

$$\langle |E_h(t)|^4\rangle = \sum_{k_1,\ldots k_4} \langle \alpha_{k_1}\alpha_{k_2}\alpha^*_{k_3}\alpha^*_{k_4}\rangle \underset{\sim}{e}_{k_1}(1)\, \underset{\sim}{e}_{k_2}(1)\, \underset{\sim}{e}^*_{k_3}(1)\, \underset{\sim}{e}^*_{k_4}(1) \tag{II-3}$$

As in Section II (See Eqs. (19),(20)) the average values of the right hand side of Eq. (II-3) can be written as

$$\begin{aligned}
&\sum_{k_1,\ldots k_4} \langle \alpha_{k_1}\alpha_{k_2}\alpha^*_{k_3}\alpha^*_{k_4}\rangle \\
&= \sum_{k_1,\ldots k_4} \langle \alpha_{k_1}\rangle\langle \alpha_{k_2}\rangle\langle \alpha^*_{k_3}\rangle\langle \alpha^*_{k_4}\rangle \\
&+ \sum_{k_1k_3k_4} \{[\langle \alpha^2_{k_1}\rangle - \langle \alpha_{k_1}\rangle^2]\, \langle \alpha^*_{k_3}\rangle\langle \alpha^*_{k_4}\rangle + c.c.\} \\
&+ \sum_{k_1k_2k_4} [\langle |\alpha_{k_1}|^2\rangle - |\langle \alpha_{k_1}\rangle|^2]\, \langle \alpha_{k_2}\rangle\langle \alpha^*_{k_4}\rangle \\
&+ \mathcal{O}(\sigma^4)
\end{aligned} \tag{II-4}$$

Upon inserting Eqs. (II-4) and (II-1) in Eq. (II-3) and making use of the rotating wave approximation we obtain

$$\langle|E_h(t)|^2\rangle = |\langle E_h(t)\rangle|^2 + 4 \sum_{k_1} \sigma_{k_1}^2 |\langle E_h(t)\rangle|^2 \qquad \text{(II-5)}$$

By replacing $\sigma_{k_1}^2$ with the normalized fluctuation f' (see Eq. (23)) Eq. (26) follows from Eq. (II-5). Eqs. (27) and (28) can be derived in a similar way.

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stochastic nature of the mode-locked laser radiation. The role of the $W(\mathcal{E}_1\mathcal{E}_2)$ function is particularly suited for such a description. The derivation of the quasi probability distribution function using the central limit theorem and characteristic function follows from the detailed exposition by Glauber. [see for example Glauber, lecture notes, from Quantum Optics and Electronics ed. C. DeWitt, A. Blandin, C. Cohen - Tannoudji, Gordon and Breach 1964]. Since we are interested only in intensity correlations rotating wave approximation leads to results identical to those obtained from more general derivation involving time dependent variances.

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23. This can be compared with a more general equation derived in ref. 15. It should be noted that the third term in eq. 5 of this reference should read $n \exp [-(\frac{2n-1}{4n}) x^2]$.

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Figure Captions

1. An experimental configuration for observation of two photon fluorescence. Incident beam is split in two halves which travel through the fluorescent dye collinearly. The dye fluoresces when two pulses overlap in the dye. The fluorescence intensity dependent on the product of the two beam intensities.

2. Conventional TPF contrast ratio R is plotted as a function of fractional fluctuation f. R approaches 3 and 1.5 at either extremes when f = 0 and ∞ respectively.

3. Normalized integrated fluorescence intensity is plotted as a function of delay between the pulses for different values of total fractional fluctuations f. Signal to background ratio is a function of f. The explicit temporal behavior and the width of the TPF trace can be compared with the actual pulse shape I(t).

4. An experimental configuration to measure third order intensity correlation with TPF of the fundamental pulse and its second harmonic. The same configuration can be used to measure higher order intensity correlation by using appropriate harmonic generation crystal and frequency selective element.

5. Plots of normalized temporal behaviour of second harmonic intensity as function of the total dispersion over the fundamental spectral bandwidth ($\kappa \Delta \Omega L$). The second harmonic pulse width can be compared with the original fundamental pulse. The second harmonic pulse becomes wider as dispersion increases resulting in imperfect phase matching over the fundamental bandwidth.

6. Plots of normalized integrated fluorescence intensity representing a third order correlation for difference fractional fluctuations.

7. Plot of pulse width normalized with original pulse width and signal to background ratio as a function of order of correlation. The contrast ratio, $\mathcal{F}(o)/\mathcal{F}(\infty) = \{[\frac{1}{\sqrt{n+1}} + 4f(n + n^{-\frac{1}{2}})]/4f(n+n^{-\frac{1}{2}})\}$ follows from eq. 49, where f_n is approximated to be nf. For $f = .005$ the ratio is in agreement with the experimental observation of Rentzepis and Duguay. Signal to background ratio decreases for higher order correlation due to the increase in fluctuations of the harmonic.

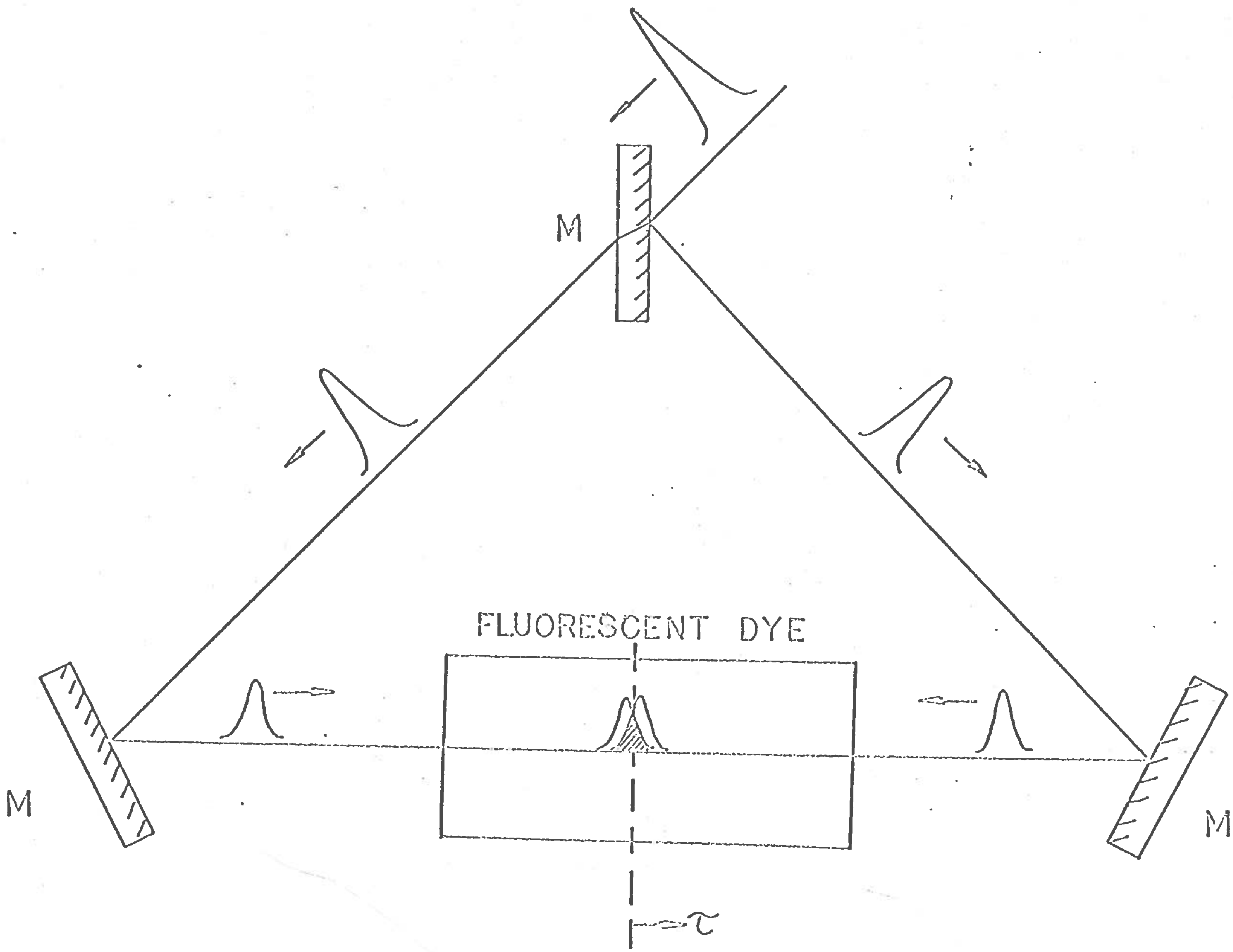


FIG.1

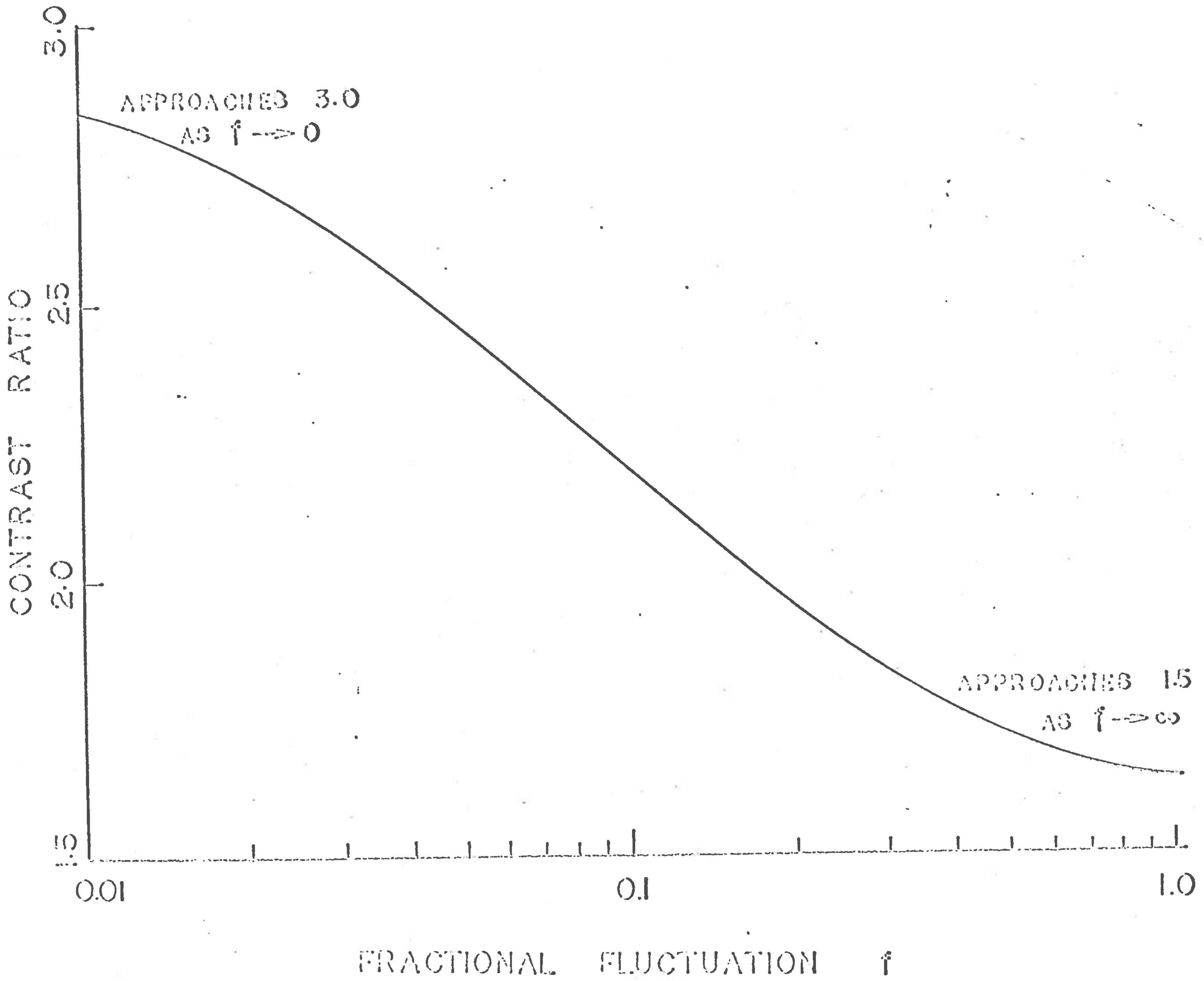
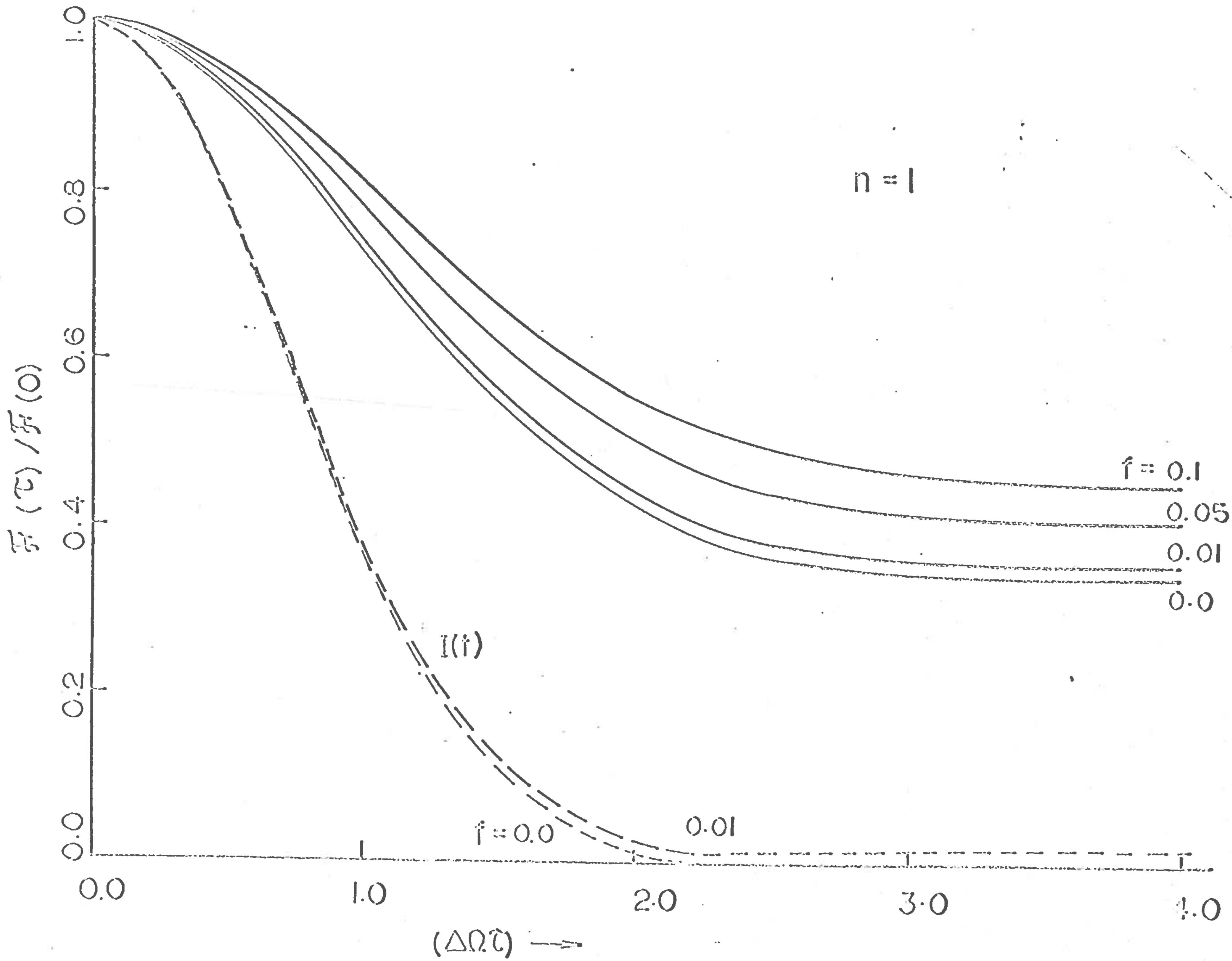
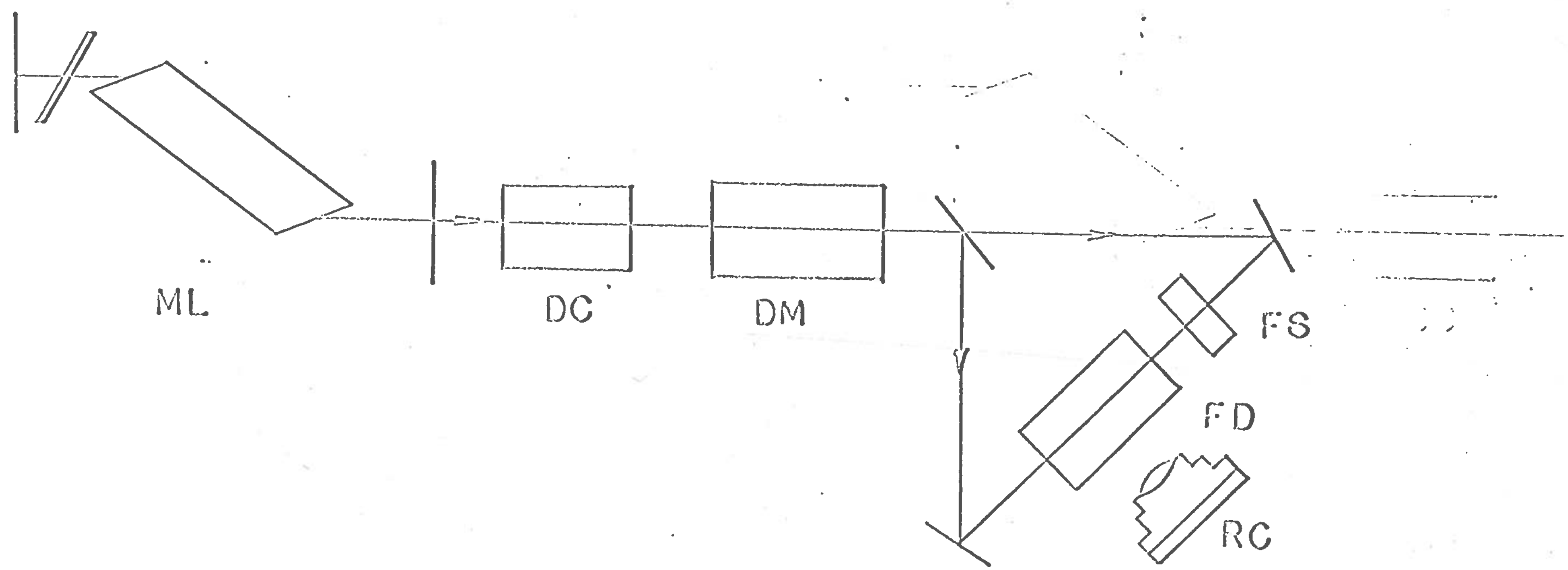

CONTRAST RATIO
3.0
2.5
2.0
1.5
APPROACHES 3.0
AS f --> 0
APPROACHES 1.5
AS f --> ∞
0.01
0.1
1.0
FRACTIONAL FLUCTUATION f

FIG. 2

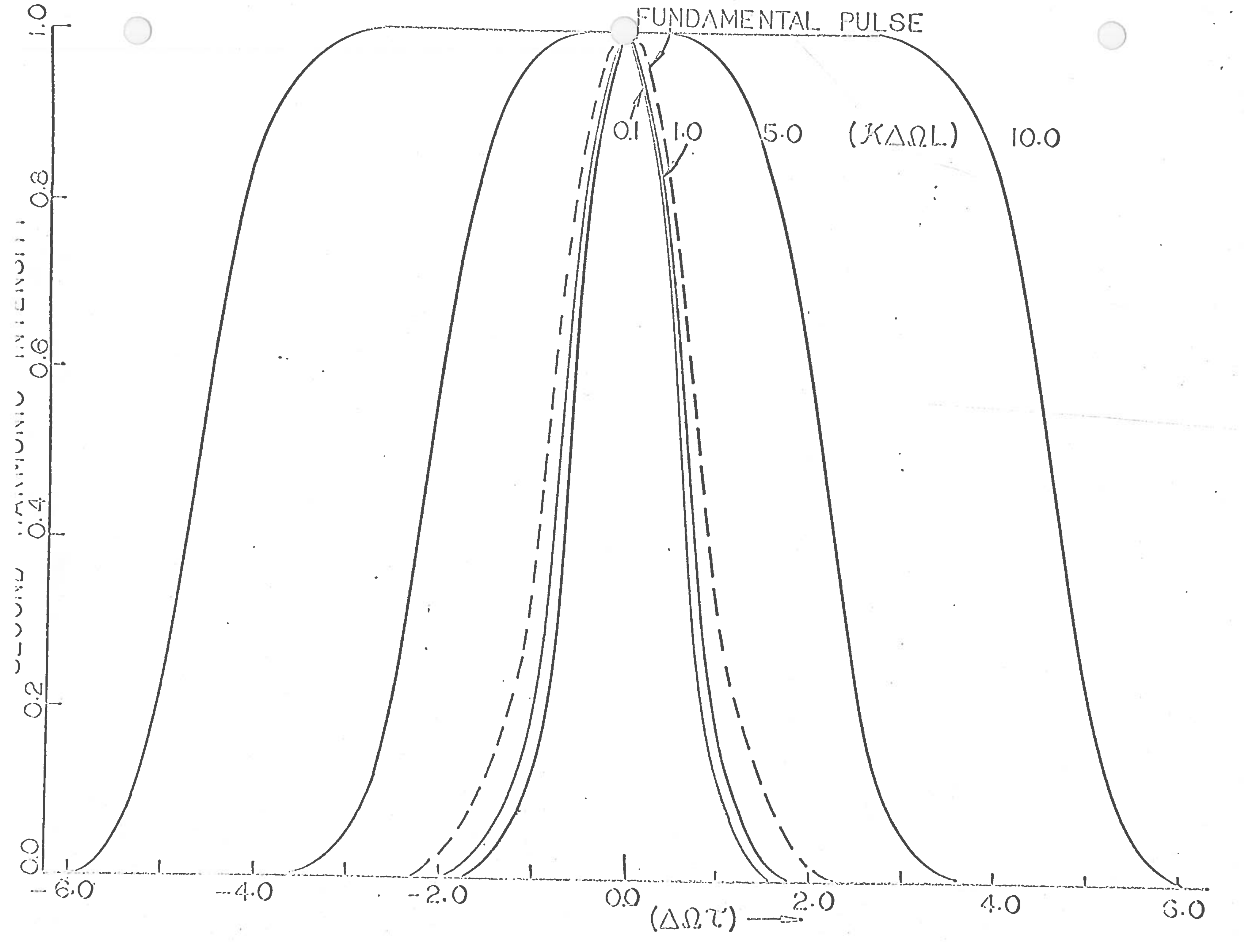


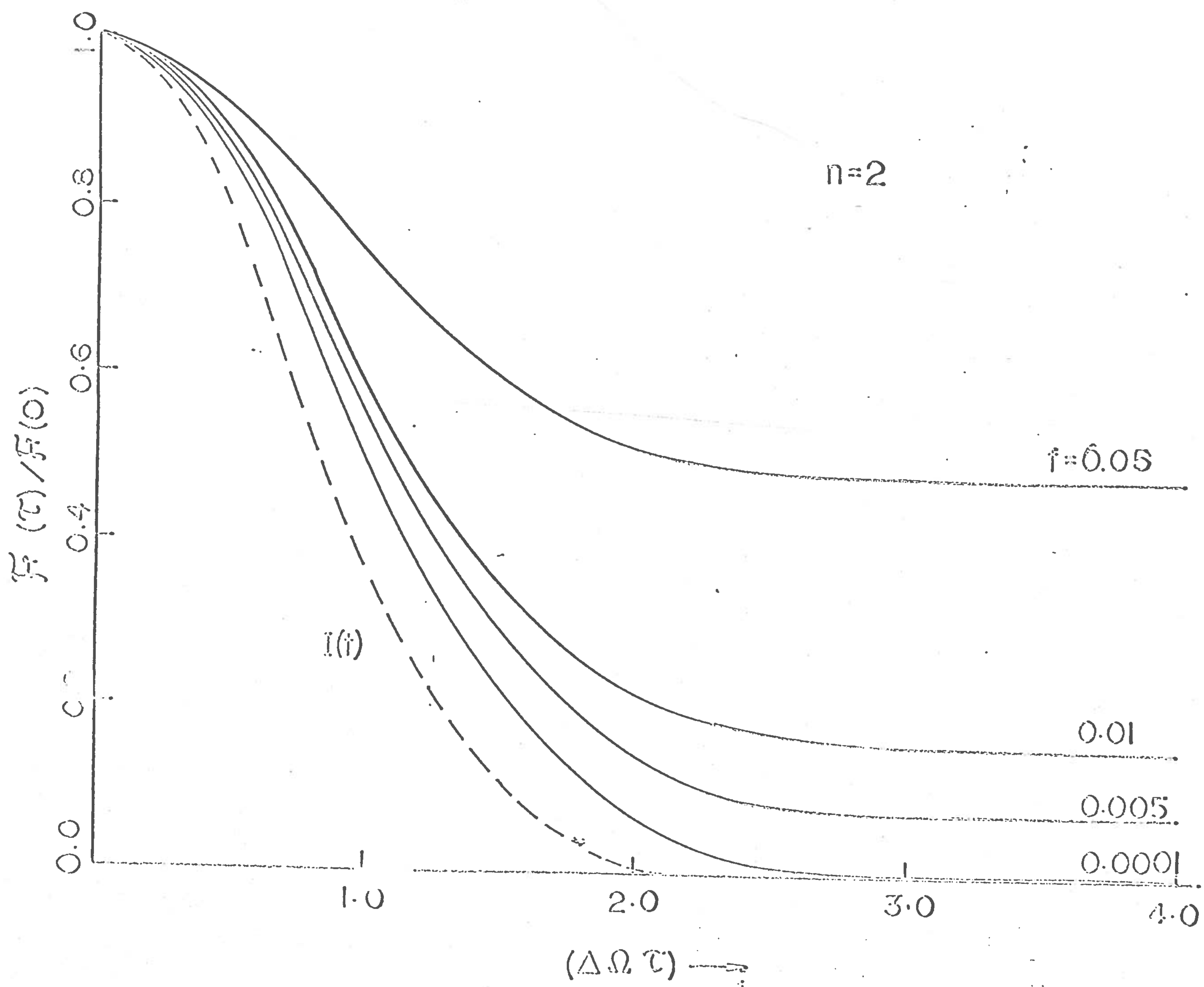
n = 1
1.0
0.8
0.6
0.4
0.2
0.0
F (τ) / F (0)
f = 0.1
0.05
0.01
0.0
I(t)
f = 0.0
0.01
0.0
1.0
2.0
3.0
4.0
(ΔΩτ) →



DC -- doubling crystal   DM -- dispersive medium   FD - fluorescent dye
FS -- frequency selective element   ML -- mode locked laser
RC -- recording camera

FIG. 4


FUNDAMENTAL PULSE
0.1
1.0
5.0
(ҞΔΩL)
10.0
1.0
0.8
0.6
0.4
0.2
0.0
-6.0
-4.0
-2.0
0.0
2.0
4.0
6.0
(ΔΩτ)



n=2
f=0.05
0.01
0.005
0.0001
I(t)
1.0
0.8
0.6
0.4
0.0
1.0
2.0
3.0
4.0
(ΔΩ τ) →
F (τ)/F(0)

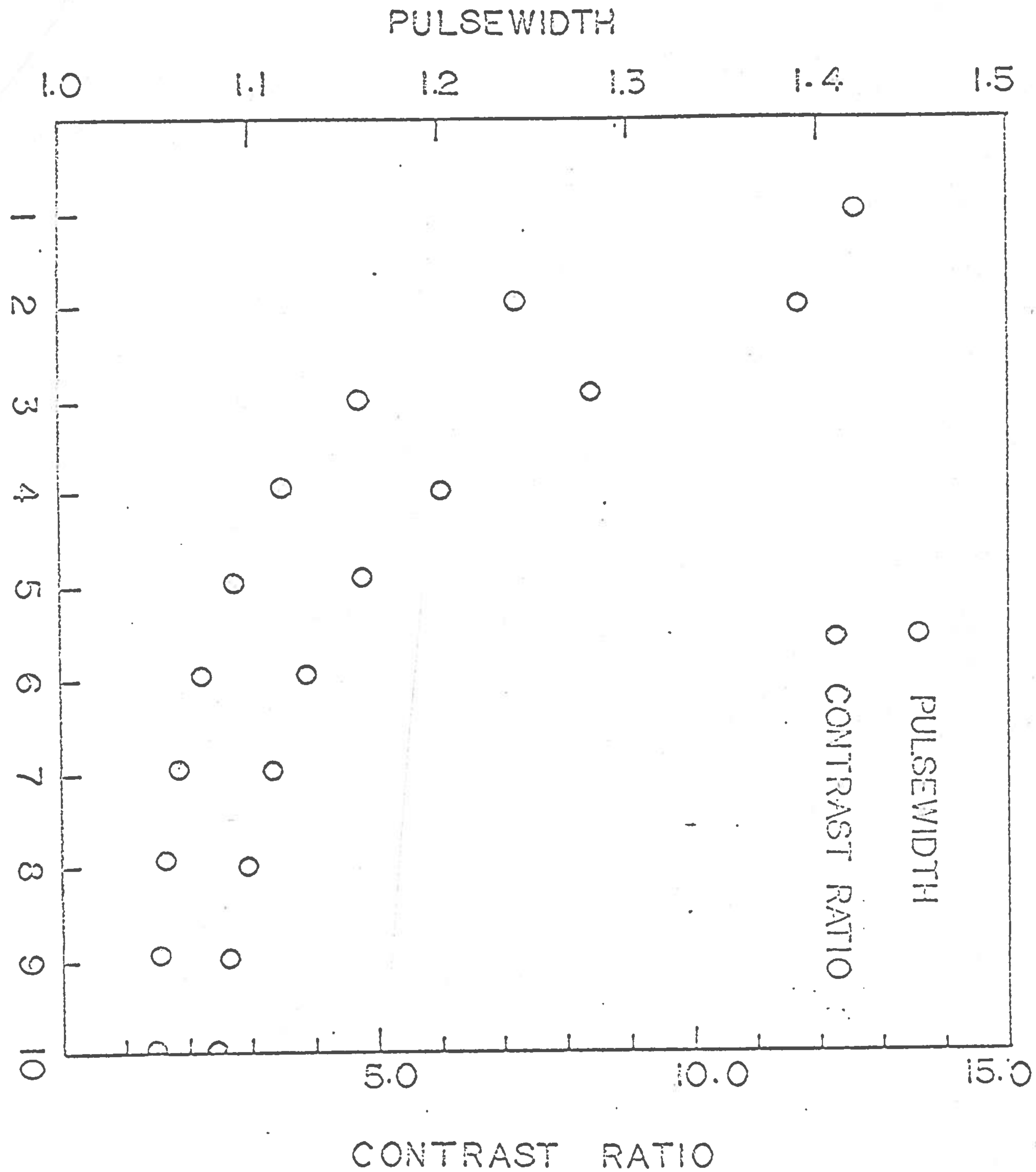

PULSEWIDTH
1.0
1.1
1.2
1.3
1.4
1.5
1
2
3
4
5
6
7
8
9
10
n
PULSEWIDTH
CONTRAST RATIO
5.0
10.0
15.0
CONTRAST RATIO

FIG.7