Statistical Fluctuations in Nonlinear Optical Processes*

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The influence of partial coherence in a laser beam on nonlinear optical interactions is discussed. The presence of several different modes with random amplitude and phases is shown to lead to fluctuations in second-harmonic production and to an error in the measurement of the corresponding nonlinear constant. Experiments have been carried out with a "Q-switched" ruby, which demonstrate the role of partial spatial coherence in creating fluctuations in second-harmonic production. Mixing and higher harmonics generation are also considered. Limitations in momentum space necessitate a separate discussion of the creation of light beats at small difference frequencies.

I. INTRODUCTION

SINCE the first observation of optical harmonics by Franken *et al.*,[1] there has been much interest in nonlinear interactions involving optical waves.[2] Theoretical treatments have been given[3–5] providing a quantitative analysis of experimental situations. Unfortunately, these treatments fail to account for some disturbing experimental features, among which one can single out: the lack of one-to-one correspondence between the amplitude of fundamental and harmonic pulses,[6,7] the discrepancies between various measured values of the same nonlinear constants, and the null result of some down conversion experiments. The discrepancy between theory and experiment disappears if the assumption is dropped that an optical maser is an ideally coherent source giving rise to a field which can be represented by a completely determined function of time and space. High-power lasers usually operate in more than one single mode and the lack of spatial and temporal coherence in their output has been clearly demonstrated. Solid-state lasers have been known to yield a number of different frequencies. Early measurements of the spatial coherence of a ruby laser[8] have given coherence areas substantially smaller than the cross section of the crystal. Clark *et al.*[9] have taken time-resolved pictures of the front face of a ruby laser and shown that the spatial distribution of the intensity can be different for different spikes. Correspondingly, time resolved spectroscopy performed by Ridgway *et al.*[10] shows a change in the frequencies of oscillation from spike to spike and sometimes within a spike.

The representation of a laser as a coherent source is inadequate. Accordingly, the complex field of an optical maser will be written in the form

$$\mathbf{E}(\mathbf{r},t)=\sum_{n,j,k,\sigma}\mathbf{a}_{njk\sigma}u_{njk}(\mathbf{r})\exp(-2\pi i\nu_{njk}t),$$

where the a_{njk} are random coefficients.

The ν_{njk} are the frequencies of the optical modes specified by the "quantum" numbers n, j, k, and the polarization variable σ. The wave functions $u_{njk}(\mathbf{r})$ can be constructed from the knowledge of the spatial and temporal dependence of the field on the front face of the laser. This field is expanded as a sum over a set of orthogonal modes. These modes might coincide with the normal modes defined by Fox and Li[11] or Boyd and Gordon.[12] In any case they determine the corresponding $u_{njk}(\mathbf{r})$ over all space. Due to the very small divergence of the beam these u_{njk} can be written as $u_{jk}(\mathbf{r}_0)\exp(ik_{njk}z)$, where z designates the coordinate along the direction of propagation, $\mathbf{r}_0$ the position vector in the cross section plane, and k_{njk} is the wave number of the plane wave in the z direction at the frequency ν_{njk}. The $u_{jk}(\mathbf{r}_0)$ are now independent of n and the expression for the field can be rewritten for one sense of polarization:

$$E(\mathbf{r},t)=\sum_{n,j}a_{nj}\mathbf{u}_j(\mathbf{r}_0)\exp(ik_{nj}z-2\pi\nu_{nj}t). \qquad (1)$$

To simplify the notation only one index j has been used to label the transverse modes. Although the a_{nj}'s are random they are not necessarily statistically independent, and the number of modes involved in the representation of the field is not automatically equal to the number of independent parameters. Even modes with different eigenfrequencies may have statistically correlated amplitudes. The coupling between various laser modes by the nonlinearity of the medium is an example of this situation. In many other cases, however, it can be justified to consider the a's belonging to differ-

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2 P. A. Franken and J. F. Ward, Rev. Mod. Phys. **35**, 23 (1963).
3 N. Bloembergen, Proc. I.E.E.E. **51**, 124 (1963).
4 J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, Phys. Rev. **127**, 1918 (1962).
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7 J. A. Armstrong and J. Ducuing, Bull. Am. Phys. Soc. **8**, 233 (1963); see also, T. Tamai and M. Achiwa, J. Phys. Soc. Japan **18**, 917 (1963).
8 D. F. Nelson and R. J. Collins, J. Appl. Phys. **32**, 739 (1961).
9 G. L. Clark, R. F. Wuerker, and C. M. York, J. Opt. Soc. Am. **52**, 878 (1962).
10 S. L. Ridgway, G. L. Clark, and C. M. York, J. Opt. Soc. Am. **53**, 700 (1963).
11 A. G. Fox and T. Li, Bell System Tech. J. **40**, 453 (1961).
12 G. D. Boyd and J. P. Gordon, Bell System Tech. J. **40**, 489 (1961).

ent modes as independent. One example is the case of a ruby laser where different modes predominantly make use of different ions.

When Eq. (1) is used for the field E of the source, the fields and intensities at a given point in space resulting from the nonlinear interaction will themselves be random functions whose statistical properties will depend on the properties of the a's. In real experiments the measured quantities will not be functions of the field at a single point in space and time, but rather suitable averages over space and time of such functions. A simple example of this is the output current of a phototube. Even after such averages are taken, the randomness of the a's will give rise to fluctuations in the measurements and to some possibly large errors in the estimates of the nonlinear constants.[13,14] In Sec. II second-harmonic generation is considered, and it is shown that the presence of several incoherent modes in the laser beam leads to a discrepancy between the real and the measured nonlinear constant, and introduces a certain amount of randomness in the ratio between the square of the fundamental intensity and the second-harmonic intensity. Experimental results with a single-pulse ruby laser are described in Sec. III which demonstrate the part played by the spatial modes in the mechanism of fluctuations of second-harmonic production. The theory of fluctuations in higher order nonlinear processes is briefly discussed in Sec. IV. Section V is devoted to down conversion and related experiments. The case of microwave generation is given particular attention, and it is shown how a reduction factor, analogous to the one introduced by Forrester *et al.*[15] for the generation of beats in a photocathode, should be taken into account in this experiment.

II. MODE EFFECTS IN SECOND-HARMONIC GENERATION

Consider a laser beam of small angular spread incident on a nonlinear dielectric slab. Take the fundamental field and the induced harmonic polarization to be linearly polarized. The latter can then be expressed by means of Eq. (1) as,

$$P_s^{2\nu}(\mathbf{r}_0,z,t)=\chi^{\mathrm{NL}}\sum_{n,n',j,j'} a_{nj}a_{n'j'}u_j(\mathbf{r}_0)u_{j'}(\mathbf{r}_0)\times\exp i[(k_{nj}{}^d+k_{n'j}{}^d)z-2\pi(\nu_{nj}+\nu_{n'j})t].$$

Here $k_{nj}{}^d$ is the wave number in the dielectric, corresponding to frequency ν_{nj} close to the average laser frequency ν, and χ^{NL} the appropriate nonlinear constant.[3] This polarization is going to radiate and to give rise to a field at frequency 2ν. We assume that both angular and frequency spreads are small enough, that dispersion will affect equally the radiation of all terms in the expression of $P_s(2\nu)$. This excludes *a priori* the consideration of experiments in a "matched" direction.[16,17] Then the linearly polarized harmonic field in free space can be written

$$E^{(2\nu)}(\mathbf{r}_0,z,t)=\mathfrak{X}\sum_{n,n',j,j'} a_{nj}a_{n'j'}u_j(\mathbf{r}_0)u_{j'}(\mathbf{r}_0)\times\exp i[k(\nu_{nj}+\nu_{n'j'})z-2\pi(\nu_{nj}+\nu_{n'j'})t], \quad (2)$$

where $\mathfrak{X}$ is the product of χ^{NL} and a factor depending on the length l of the slab in the z direction and on the dispersion of the dielectric,

$$\mathfrak{X}=\chi^{\mathrm{NL}}\frac{4\pi}{n_1{}^2}(k_{2\nu}l)\frac{\sin(\Delta kl/2)}{(\Delta kl/2)},$$

where $\Delta k=k_{2\nu}-2k_\nu$.

A. Measured Quantities

In order to perform a measurement, the fundamental and the harmonic thus generated will be detected separately and compared. The detection will generally be done by a photocathode normal to the beam. The instantaneous current emitted by such a photosensitive surface will be proportional to the integral of the square of the amplitude of the optical field over the cross section of the beam.

$$I_{1,2}(t)=\eta_{1,2}\int_s|E_{1,2}(\mathbf{r}_0,t)|^2d\mathbf{r}_0.$$

In fact as there is a spread in the transit time of the photoelectrons, and as the associated circuitry has reactive properties, the detected current $I_{1,2}$ will be different from this.

$$I_{1,2}=\eta_{1,2}\int dt'h(t-t')\int_s|E_{1,2}(\mathbf{r}_0,t')|^2d\mathbf{r}_0,$$

where $h(t-t')$ expresses the action of a linear filter. We will make the simplifying assumption that this action can be represented by an integration over a suitable time interval T. In this case

$$I_1=\frac{\eta_1}{T}\int_{t-T/2}^{t+T/2}dt'\int_s d\mathbf{r}_0|E_1(\mathbf{r}_0,t')|^2, \quad (3)$$

$$I_2=\frac{\eta_2}{T}\mathfrak{X}^2\int_{t-T/2}^{t+T/2}dt'\int_s d\mathbf{r}_0|E_1(\mathbf{r}_0,t')|^4, \quad (4)$$

[13] J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, Bull. Am. Phys. Soc. 8, 233 (1963).
[14] N. Bloembergen, Proceedings of the Brooklyn Symposium on Optical Masers (to be published).
[15] A. T. Forrester, R. A. Gudmundsen, and P. O. Johnson, Phys. Rev. 99, 1961 (1955).
[16] J. A. Giordmaine, Phys. Rev. Letters 8, 19 (1962).
[17] P. D. Maker, R. W. Terhune, M. Nisenoff, and C. M. Savage, Phys. Rev. Letters 8, 21 (1962).

$$I_1=\frac{\eta_1}{T}\int_{t-T/2}^{t+T/2}dt'\int d\mathbf{r}_0 \times\sum_{n,n',jj'} a_{nj}a_{n'j'}{}^*u_j(\mathbf{r}_0)u_{j'}{}^*(\mathbf{r}_0)e^{2\pi i\Delta\nu t'}, \quad (3a)$$

$$I_2=\frac{\eta_2\mathfrak{X}^2}{T}\int_{t-T/2}^{t+T/2}dt'\int d\mathbf{r}_0 \times\sum_{\substack{n,n',n'',n'''\\ jj'j''j'''}} a_{nj}a_{n'j'}a_{n''j''}{}^*a_{n'''j'''}{}^*e^{2\pi i\Delta\nu t'} \times u_j(\mathbf{r}_0)u_{j'}(\mathbf{r}_0)u_{j''}{}^*(\mathbf{r}_0)u_{j'''}{}^*(\mathbf{r}_0). \quad (4a)$$

In practice, the time interval T, which will be called the time constant of the detecting circuit, will be short enough that any time dependence of the a_{nj}'s can be neglected when performing the integral.

Nevertheless, in most situations it will be larger than the period of any beat between various frequency components, so that for our purpose

$$\frac{1}{T}\int_{t-T/2}^{t+T/2}e^{2\pi i\Delta\nu t'}dt'=\delta(\Delta\nu).$$

Using this property and the orthonormality of the modes over the cross section of the beam, we obtain for I_1 and I_2,

$$I_1=\eta_1\sum_{n,j}|a_{nj}|^2, \quad (5)$$

$$I_2=\eta_2\mathfrak{X}^2\sum a_{nj}a_{n'j'}a_{n''j''}{}^*a_{n'''j'''}{}^*C_{j,j',j'',j'''} \times\delta(\nu_{nj}+\nu_{n'j'}-\nu_{n''j''}-\nu_{n'''j'''}), \quad (6)$$

where

$$C_{jj'j''j'''}=\int_s u_j(\mathbf{r}_0)u_{j'}(\mathbf{r}_0)u_{j''}{}^*(\mathbf{r}_0)u_{j'''}{}^*(\mathbf{r}_0)d\mathbf{r}_0.$$

B. Relation between Measured Quantities

Equations (5) and (6) describe the random relationship between I_1^2 and I_2. When the fundamental field consists of a homogeneous plane wave, the ratio $\eta_1^2I_2/\eta_2I_1^2$ is minimum and equal to $\mathfrak{X}^2$. In an experiment performed with a less than ideal beam this ratio will be taken as defining the measured nonlinearity $\mathfrak{X}_m$. From the expressions (3) and (4) for I_1 and I_2, it is immediately seen that $\mathfrak{X}_m>\mathfrak{X}_1$. This can easily be understood by considering harmonic generation by an interference pattern. As the effect is quadratic the maxima will overcompensate the minima. In the same way a succession of light pulses will be more efficient in generating second harmonic than a constant light beam having the same average intensity.

The lack of spatial and temporal coherence of the laser beam will give rise to fluctuations in the measured value of the nonlinearity. Introduce normalized variables

$$\mathfrak{X}=\left(\frac{\chi_m}{\chi}\right)^2=\left(\frac{\mathfrak{X}_m}{\mathfrak{X}}\right)^2 \quad\text{and}\quad \frac{a_{nj}}{I_1^{1/2}}=\alpha_{nj}.$$

The α_{nj}'s satisfy $\sum_{n,j}|\alpha_{nj}|^2=1$ and their statistical properties can be determined from those of the a_{nj}'s.

$$\mathfrak{X}=\sum\alpha_{nj}\alpha_{n'j'}\alpha_{n''j''}{}^*\alpha_{n'''j'''}{}^* \times\delta(\nu_{nj}+\nu_{n'j'}-\nu_{n''j''}-\nu_{n'''j'''})C_{jj'j''j'''}. \quad (7)$$

This expression can be split into a group of phase-independent terms A and a group of phase-dependent terms B

$$A=\sum|\alpha_{nj}|^4C_{jjjj}+2\sum{}'|\alpha_{nj}|^2|\alpha_{n'j'}|^2C_{jj'jj'}.$$

Here *and throughout the text* the $'$ on $\sum$ indicates the exclusion of terms for which *two* sets of indices (n,j) are identical.

$$B=\sum{}''\alpha_{nj}\alpha_{n'j'}\alpha_{n''j''}{}^*\alpha_{n'''j'''}{}^* \times\delta(\nu_{nj}+\nu_{n'j'}-\nu_{n''j''}-\nu_{n'''j'''})C_{jj'j''j'''}.$$

Here *and throughout the text* the $''$ on $\sum$ indicates the exclusion of terms such that the product $\alpha_{nj}\alpha_{n'j'}\alpha_{n''j''}{}^*\times\alpha_{n'''j'''}{}^*$ is identically real. When the phases of the a_{nj}'s and hence of the α_{nj}'s are assumed to be random and uniformly distributed over $(0,2\pi)$, the following properties hold: $\langle B\rangle=0\langle AB\rangle=0$. Then

$$\langle\mathfrak{X}\rangle=\sum\langle|\alpha_{nj}|^4\rangle C_{jjjj}+2\sum{}'\langle|\alpha_{nj}|^2\rangle\langle|\alpha_{n'j'}|^2\rangle C_{jj'jj'}, \quad (8)$$

$$\langle\mathfrak{X}^2\rangle-\langle\mathfrak{X}\rangle^2=\langle A^2\rangle-\langle A\rangle^2+\langle B^2\rangle.$$

The last term $\langle B^2\rangle$ represents the fluctuations due to the random phases; it can be considered as an interference effect and disappears when the modes do not overlap ($C_{jj'j''j'''}=0$). The origin of this term can be found in the fact that a given harmonic mode can in general be created by the interaction of several different pairs of fundamental modes. For instance all pairs n, n' such that $\nu_n+\nu_{n'}$ has a given value will give the same harmonic frequency and will be detected together. In the same way $C_{jj'j''j'''}$ can be different from 0 when $j\neq j''$, j''' and $j'\neq j''$, j'''. These points will be discussed further when particular cases are considered.

The expression $\langle A^2\rangle-\langle A\rangle^2$ represents amplitude fluctuations. In the case of nonoverlapping modes there is only one term,

$$\langle A^2\rangle-\langle A\rangle^2=\sum[\langle|\alpha_{nj}|^8\rangle-\langle|\alpha_{nj}|^4\rangle^2](C_{jjjj})^2.$$

The general expression for overlapping modes is more complicated and will not be reproduced. The general theory will now be applied to some simple cases of physical interest.

C. Second-Harmonic Generation with a Gas Laser

The output of a gas laser can be represented by a small number of modes. Second-harmonic power has been generated with a gas laser oscillating simultaneously in a single transverse spatial mode.[18–20] Due to the nonlinearity essential to the mechanism of the oscillator, the individual modes will have stabilized amplitudes and hence very small intensity fluctuations. In this approximation I_1 is a constant and the statistical properties of $\mathfrak{X}_m$ are those of I_2. In this case it is not really necessary to introduce normalized variables $\mathfrak{X}$ and α. The phases are random variables, but because of the same nonlinearity, they will not necessarily be independent. If independent uniformly distributed phases are nevertheless assumed, Eqs. (8) and (9) become, for the case of one spatial mode u_j,

$$\langle\mathfrak{X}\rangle = C\{\sum|\alpha_n|^4 + 2\sum{}'|\alpha_n|^2|\alpha_{n'}|^2\}$$

$$= \int|u_j(r_0)|^4 dr_0\{1+\sum{}'|\alpha_n|^2|\alpha_{n'}|^2\}, \qquad (8a)$$

$$\langle\mathfrak{X}^2\rangle - \langle\mathfrak{X}\rangle^2 = \left[\int|u_j(r_0)|^4 dr_0\right]^2\{4\sum{}'|\alpha_n|^4|\alpha_{n'}|^2|\alpha_{n''}|^2$$
$$\times\delta(2\nu_n-\nu_{n'}-\nu_{n''}) + \tfrac{4}{3}\sum{}'|\alpha_n|^2$$
$$\times|\alpha_{n'}|^2|\alpha_{n''}|^2|\alpha_{n'''}|^2$$
$$\times\delta(\nu_n+\nu_{n'}-\nu_{n''}-\nu_{n'''})\}. \qquad (9a)$$

The relation $\sum|\alpha_n|^2=1$ has been used in Eq. (8a). As $u_j(r)$ is known, $\int|u_j(r)|^4$ can be incorporated in χ and the relative error in the measurement is represented by the term $\sum{}'|\alpha_n|^2|\alpha_{n'}|^2$. If the α_n do not vary too strongly with n, the order of magnitude of this term is $N(N-1)/N^2$ and we can write

$$\mathfrak{X} = (\chi_m/\chi)^2 = 2 - N^{-1}. \qquad (10)$$

This relation has been derived by Ashkin *et al.*[18] The comments made earlier about phase fluctuations are here of special interest. As mentioned previously, their existence is due to the fact that any pair, n, n' for which $\nu_n+\nu_{n'}$ is equal to a given value, will generate the same harmonic frequency. An immediate consequence of this is that at least three different frequencies in the incident beam are necessary to observe these fluctuations. The Fourier coefficients of the harmonic field will then appear as sums of random variables and their amplitude will not be constant. Hence the detecting device measuring the sum of the square of these coefficients, will, in general, have a random output. In other words, the harmonic field will not exhibit the amplitude stabilization properties of the incident laser field. In this respect it is interesting to note that amplitude correlations of the type demonstrated by Brown and Twiss[21] for incoherent light should also be present in harmonic light generated by a multimode gas laser, although the fundamental light intensity of the amplitude stabilized laser oscillations does not show this effect.[22,23]

[18] A. Ashkin, G. D. Boyd, and J. M. Dziedzic, Phys. Rev. Letters 11, 14 (1963).
[19] N. I. Adams and P. B. Schoefer, Appl. Phys. Letters 2, 136 (1963).
[20] S. L. McCall and L. W. Davis, J. Appl. Phys. 34, 2921 (1963).

If the a_n do not vary too rapidly with n, it is possible to estimate the relative rms fluctuations in the measurement

$$\frac{(\langle\mathfrak{X}^2\rangle-\langle\mathfrak{X}\rangle^2)^{1/2}}{\langle\mathfrak{X}\rangle} = \left[\frac{8Q}{3}\frac{(8Q^2-3Q-2)}{(2Q+1)^2(4Q+1)^2}\right]^{1/2} \quad \text{for} \quad N=2Q+1 \quad \text{and}$$

$$\left[\frac{8Q}{3}\frac{(8Q^2-15Q+7)}{4Q^2(4Q-1)^2}\right]^{1/2} \quad \text{for} \quad N=2Q,$$

where N is the total number of modes. For $N=3$ and $N=4$ these numbers are, respectively, 18% and 25%. For large N, the relative root-mean-square fluctuations of $\mathfrak{X}$ are of order $(2/3N)^{1/2}$.

Experimentally, these fluctuations would be observed, if the time constant were small, but just larger than the largest beat period between modes.[24] The power available from a gas laser is so low that very long time constants are required to make the observation of harmonics possible. There it is a time average of I_2, rather than I_2 itself, which is measured. If the stochastic properties of the α_n's are stationary (and this is a reasonable assumption) this time average is equivalent to the statistical average we have computed. This will not be the case of the measured rms fluctuations which might be drastically reduced by the integration process [roughly a factor $(T/\tau)^{1/2}$, where τ is the correlation time of the α_n's]. If there is some amount of correlation between phases, Eqs. (8a) and (9a) do not, in general, apply. They will retain an approximate validity if the correlation is expressed by linear relations between the phases, as it will be if this correlation is due to any simple nonlinear process.

D. The Case of the Ruby Laser

Many different frequencies and spatial modes are present in the output of an ordinary ruby laser. If we

[21] R. Hanbury Brown and R. Q. Twiss, Nature 177, 28 (1956).
[22] R. J. Glauber, Proc. Third Conf. Quantum Electronics, Paris (to be published).
[23] L. Mandel in *Progress in Optics*, edited by E. Wolf (North-Holland Publishing Company, Amsterdam, 1963), Vol. II, pp. 181 ff.
[24] For time constants much shorter than the beat periods between modes, the replacement of a time integration by a Kronecker δ in Eqs. (5)–(7) is not allowed. The rms fluctuations measured instantaneously would not decrease with N. The problem for very short time constants would be similar to that of noise in nonlinear devices at radiofrequencies and to the problem of radar echoes from a rain cloud.

assume as before that the amplitude fluctuations are small the rms fluctuations in χ will be of order $CN^{-1/2}$, where N is the number of modes taking part in the interaction, C being a factor of order 1 depending on the structure of the modes. For N of order 10^3 or larger this will be very small. In fact, experimental observations[10,19] make clear that in this case not only the phases but the amplitudes of the various modes will fluctuate. The amplitudes will even fluctuate to the point that although a large number of modes might be available, only a few will oscillate in one spike. The order of magnitude of the fluctuations will then be given by $(n)^{-1/2}$, where n is the average number of modes going at the same time rather than the total number of modes. This would account for substantial fluctuations and thus correspond to the experimental situation, as reported in the following section. The average value of $\mathfrak{X}$ would not be affected by this particular mechanism, as long as the average is taken on a large number of trials, i.e., of individual spikes. Note that this might mean a very large number (as is the number of possible configurations) and that this requirement is not necessarily satisfied for ordinary measurements. If it is, we will have approximately

$$\langle\mathfrak{X}\rangle = \langle(\chi_m/\chi)^2\rangle \approx 2\,,$$

as given by Eq. (10) for large N.

When an ordinary ruby laser is used to generate second harmonic, the ratio of individual harmonic spikes to the square of the intensity of each spike at the fundamental frequency shows large fluctuations, as shown in Fig. 1. This can be accounted for by the above mechanism where both spatial and temporal coherence play a role.[13,14] When a large time constant is used in the detecting circuits, i.e., when the envelope of the pulses rather than the individual spikes are observed, it is very often found that a perfect square law does not hold for the correspondence I_2 versus I_1, but that the value of $\mathfrak{X}$ is still varying with time. The remark made above about the large number of spikes necessary to get really the average value of $\mathfrak{X}$ applies probably here. The time constant has a maximum value beyond which the pulse envelope will be distorted and any hope of recording the real correspondence will vanish. If T is made less than this value, the number of spikes on which the time average is performed might not be large enough and the nonstationarity of the α_n's will account for the variation of $\mathfrak{X}$ with time.

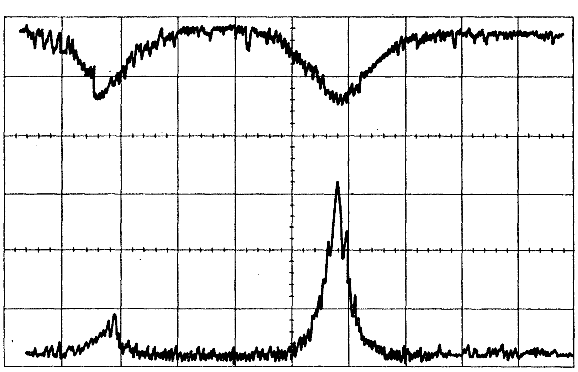

FIG. 1. Two laser pulses of about equal intensity at the fundamental frequency (top line) produce pulses of quite different relative intensity at the second-harmonic frequency (bottom line).

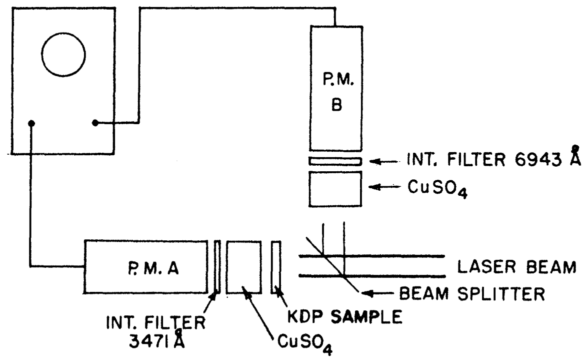

FIG. 2. Experimental arrangement to determine the ratio between fundamental intensity f and the second-harmonic intensity h in a giant pulse from a ruby laser.

III. EXPERIMENTS ON SECOND-HARMONIC GENERATION

Second-harmonic generation by individual spikes from a Q-switched ruby laser has been observed under fairly reproducible circumstances. In a first experiment the fundamental and harmonic intensity were compared for a large number of spikes. The time constants of the detecting devices (phototubes plus associated circuitry) were made as short as the available equipment permitted. This resulted in resolution times between 10 and 15 mμsec for both tubes. The laser beam was unfocused and the distances between the front face of the laser, the sample and the surface of the photocathode were short enough for the cross section of the beam to be constant. A diagram of the experimental arrangement is shown in Fig. 2. The results of a series of observations on more than 70 pulses are reproduced in Fig. 3. For a given value of the fundamental intensity, the corresponding harmonic intensity might vary from 1 to 3. For this experiment the rms fluctuations were estimated to be 40%. A standard ruby laser (not Q switched) gives similar results.

The theory developed in the preceding section provides an explanation for these observations. Nevertheless, it could be argued that some of the time variations of the envelope of the rather fast pulse (the duration is about 40 nsec) could happen in a time shorter than the response of our detecting circuit. If the time variations were different for the harmonic and the fundamental, this would give rise to different responses for the harmonic and the fundamental depending on the shape of the pulses.

To show that the observed fluctuations are due to the mechanism we have described, and not to imperfect

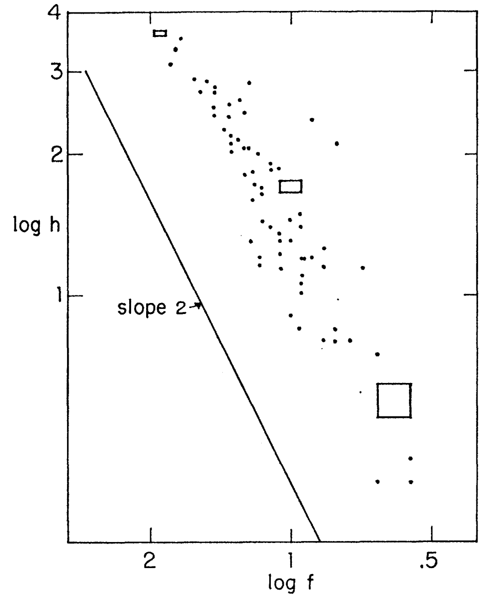

FIG. 3. A scatter diagram from 70 giant pulses, showing the random relationship between the fundamental and second-harmonic intensity.

experimental conditions, we carried out a series of experiments in which the fundamental beam was split in two approximately equal parts which in turn generated second harmonic in two identical crystals. The separate harmonic pulses were detected and their amplitudes compared. To avoid any spurious effects due to differences in the time constant of the two circuits, these quantities were made much longer (1 μsec) than the width of the observed pulses (40 mμsec). The peak amplitudes were thus very nearly proportional to the time integral of the real pulses. The experimental setup is shown in Fig. 4. When the beams were left unperturbed the correlation between pulse amplitudes was, as expected, very good. The remaining fluctuations could be ascribed to small variations in gain of the circuits and were accentuated by the rather long interval between

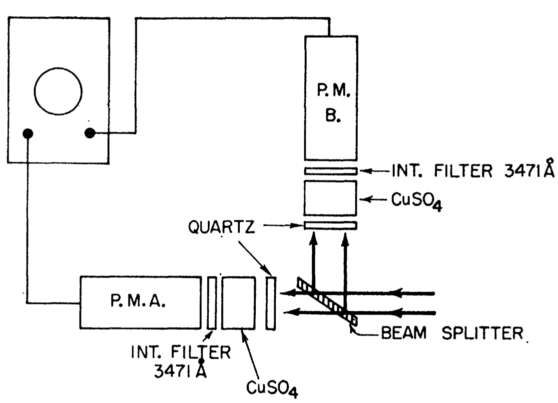

FIG. 4. Experimental arrangement to determine the relative yield of two second-harmonic generation processes with similar geometry.

pulses required for operation of the laser at an approximately constant temperature. In any case the rms fluctuations (4%) were found to be equal to the ones observed when splitting a single harmonic beam or the beam from a pulsed source of light.

According to our theory the correlation between the harmonic signals could then be spoiled by changing the relative phases and amplitudes (or alternatively the structure of the modes in one of the fundamental beams) but leaving the total intensity constant. This redistribution of the energy and of the phases was performed in three different and separate ways and led in all cases to a significant increase in the rms fluctuations.

In the first case one of the fundamental beams was superimposed on its mirror image (Fig. 5) by means of a prism with two reflecting sides. This beam consisted of two parts of nearly equal amplitude, one having suffered an odd number of reflections, the other an even number. This beam was thus replaced by half the sum of itself

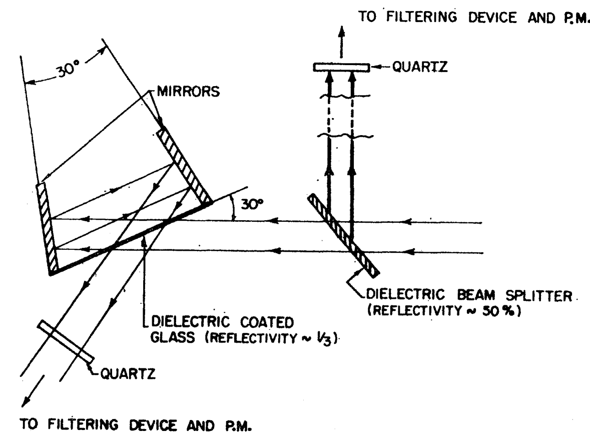

FIG. 5. The spatial similarity of the laser beams in the two quartz samples is destroyed by a glass prism, which modifies the geometry of the modes in one of the beams.

and its mirror image. This operation modified the expression for E_1 and E_2 and hence the dependence of I_2 on the a_n's in Eqs. (4) and (6), although it left I_1 unchanged. Substantial fluctuations were in fact observed. Figure 6 shows the corresponding scatter diagram together with the diagram for the unperturbed situation. This operation increased the rms fluctuations from 4 to 14%.

Another kind of perturbation was provided by a piece of frosted glass inserted in one of the beams. The glass was put very close to the nonlinear crystal to avoid the effects of the increase in cross section due to the acquired divergence of the beam. The resulting scattering provided a convenient way to mix linearly the modes of the fundamental beam, in a complicated although perfectly deterministic way; this in turn had the effect of changing the dependency of I_2 on the a_n's, I_1 was, as before, left constant. Here also a substantial increase in the fluctuations was observed, which went up with the amount of scattering provided by the frosted glass. The corresponding results are summarized in Fig. 7. Similar experiments were performed, in which one of the beams was reflected from the uneven surface of a rough silver mirror. The results were essentially the same as for the ground glass, the amount of fluctuations depending on the quality of the surface.

All these experiments consisted in the application of a linear perturbation, changing the relative values of the complex amplitudes, but not the total energy. If the fundamental beam had been spatially coherent, this would have modified the intensity of harmonic beam on the perturbed side in a constant way and hence would not have affected the correlation between the two harmonic signals. Thus these experiments clearly demonstrate the part played by partial spatial coherence in creating fluctuations. They show also the need to know the structure and the statistics of the laser beam before the nonlinearity could be measured with a reasonable accuracy.

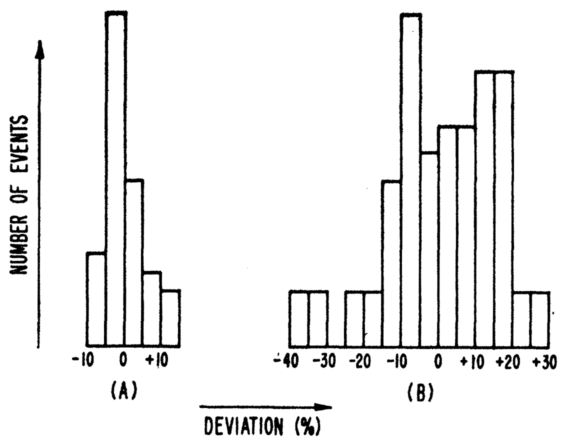

FIG. 6. A scatter diagram (A) of relative second-harmonic production with the arrangement of Fig. 4 is compared with the scatter diagram (B) with the arrangement of Fig. 5. The abscissa is proportional to the deviation from the average, the ordinate gives the number of pulses in a certain deviation interval. Comparison of these experimental results and those in Fig. 3 demonstrates the advantage of using a similar nonlinear process with a similar geometry as a reference for calibration of the incident laser pulse.

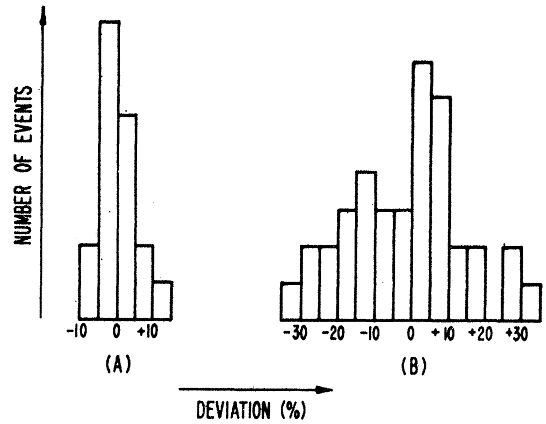

FIG. 7. The scatter diagram (A) obtained with the arrangement of Fig. 4 is compared with a corresponding diagram (B), when a frosted glass plate is inserted in front of one of the quartz plates.

IV. STOCHASTIC ASPECTS OF MIXING AND HIGHER ORDER PROCESSES

Partial coherence will also affect other nonlinear processes. Consider the situation in which two optical beams mix to give their sum or difference frequency. The case of a very small difference frequency will be treated separately in the next section. If $E_1(r)$, $E_2(r)$, $E_3(r)$ are the amplitudes of the linearly polarized fields at frequency ν_1, ν_2, ν_3, respectively, where $\nu_3=\nu_1\pm\nu_2$,

$$E_3(r)=\chi(\nu_3=\nu_1+\nu_2)E_1(r)E_2(r) \text{ for the sum},$$
$$E_3(r)=\chi(\nu_1=\nu_2+\nu_3)E_1(r)E_2^*(r) \text{ for the difference},$$

where χ is a suitable nonlinear constant. Using Eq. (1) for the fields E_1E_2, we find for the corresponding detected intensities:

$$\begin{aligned}
I_1&=\eta_1\sum_{n,j}|a_{nj}^{(1)}|^2,\\
I_2&=\eta_2\sum_{n,j}|a_{nj}^{(2)}|^2,\\
I_3&=\eta_3\mathcal{X}^2\Big\{\sum_{n,j,m,k}|a_{nj}^{(1)}|^2|a_{mk}^{(2)}|^2C_{jkjk}\\
&\quad+\sum a_{nj}^{(1)}a_{n'j'}^{(1)*}a_{mk}^{(2)}a_{m'k'}^{(2)*}\\
&\quad\times\delta(\nu_{nj}^{(1)}+\nu_{mk}^{(2)}-\nu_{n'j'}^{(1)}-\nu_{m'k'}^{(2)})C_{jkj'k'}\Big\},
\end{aligned}\tag{11}$$

where $\mathcal{X}$ depends on the geometry of the nonlinear medium. I_3 has two groups of terms: the first summation contains phase-independent terms, the second phase-dependent terms. Comparison of Eq. (11) with the corresponding expression for second harmonic, Eq. (6) shows that there are nearly twice as many phase-independent terms for second-harmonic generation than for mixing. A given product $a_{nj}a_{n'j'}^*$ occurs twice with the same coefficient in second-harmonic generation. This degeneracy affects the average value of the detected current. If the phases of the different modes are independent and uniformly distributed, only the phase-independent terms will remain after averaging

$$\langle I_3\rangle=\eta_3\mathcal{X}^2\sum_{n,j,m,k}\langle|a_{nj}^{(1)}|^2\rangle\langle|a_{mk}^{(2)}|^2\rangle C_{jjkk}.$$

If the modes are homogeneous, $(u_{nj}(x,y)=1)$ and all $C_{jjkk}=1$,

$$\langle I_3\rangle=\eta_3\mathcal{X}^2\langle I_1\rangle\langle I_2\rangle.$$

This should be compared to $\langle I_2\rangle=\eta_2\mathcal{X}^2(2N-1/N)(\langle I_1\rangle)^2$, obtained from second harmonic in the same situation.

The number of phase-dependent terms will be drastically reduced, if the two sets $\nu_{nj}^{(1)}$ and $\nu_{nj}^{(2)}$ are different. The condition $\nu_{nj}^{(1)}+\nu_{mk}^{(2)}=\nu_{n'j'}^{(1)}+\nu_{m'k'}^{(2)}$ will, in general, be satisfied only when $\nu_{nj}^{(1)}=\nu_{n'j'}^{(1)}$ and $\nu_{mk}^{(2)}=\nu_{m'k'}^{(2)}$. The beat of two gas lasers oscillating in a single transverse spatial mode[25] illustrates this comment. In this case the presence of phase-dependent terms in the detected second-harmonic intensity I_2 is caused by the even spacing of the longitudinal frequencies. Although this condition is still necessary here, it is no longer sufficient. The two sets $\nu_n^{(1)}$ and $\nu_n^{(2)}$ must have spacings equal to within the bandwidth of the detecting circuit. If the lengths of the two resonators are not the same and the time constant of the detector is long enough, this condition will not be fulfilled, and the expression for I_3 will have no phase-dependent term (apart from accidental coincidences).

$$I_3=\eta_3\mathfrak{X}^2C_{jjjj}\sum_{n,m}|a_n^{(1)}|^2|a_m^{(2)}|^2=CI_1I_2.$$

In this case there are no fluctuations. When the bandwidth of the detector is made larger than the difference between the two intermode spacings, fluctuations will appear. As the bandwidth is made larger, these fluctuations will become equal in the limit to fluctuations observed for equal intermode spacing. They will depend in a rather complicated way on the respective number of modes N_1 and N_2 in the two beams. For large numbers, the rms fluctuations will be of order $(N_>)^{-1/2}$, where $N_>$ designates the larger of N_1 and N_2.

The situation where two solid-state lasers beat to generate their sum frequency[26] is complicated. In this case most of the fluctuations will be due to the imperfect spatial and temporal overlapping of the spikes. These fluctuations which can be quite strong, will depend on the characteristics (in particular the distribution of energy over the cross section) of the lasers.

The case of a mercury line source beating with a solid-state laser[27] is simpler. In practice the coherence time and the coherence area of the mercury line will be much smaller than the corresponding quantities for the laser. The number of modes necessary to represent the field at the mercury line frequency $\nu_{\mathrm{Hg}}\pm\nu_L$, in the detection time T, and over the cross section of the beam, will be very large. This will ensure very small fluctuations at sum or difference frequencies $\nu_{\mathrm{Hg}}\pm\nu_L$. The equality $I_3=C'I_1I_2$ is closely satisfied.

High-Order Processes

The degeneracy which gives rise to the special features of second-harmonic generation will be stronger for higher harmonic processes. Consider, for instance, the generation of the sth harmonic. The amplitudes of the fields are connected by the relation,

$$E_s=\mathfrak{X}(\nu_s=s\nu_1)E_1{}^s,$$

where $\mathfrak{X}$ is a suitable nonlinear constant.[4] Using again expression (1) for the field E_1

$$E_s=\mathfrak{X}\sum_{\substack{p_1,p_2,\cdots p_N\\ p_1+\cdots+p_N=s}}a_{n'j'}{}^{p_1}\cdots a_{nNjN}{}^{p_N}$$
$$\times\frac{s!}{p_1!\cdots p_N!}u_{j_1}{}^{p_1}\cdots u_{jN}$$
$$\times\exp i[k(p_1\nu_{n'j'}+\cdots+p_N\nu_{nNjN})z$$
$$-2\pi(p_1\nu_{n'j'}+\cdots+p_N\nu_{nNjN})t].$$

The detected intensity I_s can again be split into phase-dependent and phase-independent terms. In the case of independent and uniformly distributed phases the phase-dependent terms average to zero. In this case,

$$\langle I_s\rangle=\eta_s\mathfrak{X}^2(\nu_s=s\nu_1)(s!)^2\sum_{\substack{p_1p_2\cdots p_N\\ p_1+\cdots+p_N=s}}|a_{n'j'}|^{2p_1}\cdots$$
$$\times|a_{nNjN}|^{2p_N}\left(\frac{1}{p_1!p_2!\cdots p_N!}\right)^2\Gamma_{j_1\cdots jNj_1\cdots jN},$$
$$(\langle I_1\rangle)^s=\eta_1{}^s s!\sum_{\substack{p_1p_2\cdots p_N\\ p_1+\cdots+p_N=s}}|a_{n'j'}|^{2p_1}\cdots|a_{nNjN}|^{2p_N}$$
$$\times\frac{1}{p_1!p_2!\cdots p_N!},$$

where

$$\Gamma_{j_1\cdots jNj_1\cdots jN}=\int|u_j(\mathbf{r}_0)|^2\cdots|u_{jN}(\mathbf{r}_0)|^2d\mathbf{r}_0.$$

In the case of a single mode,

$$I_s=\eta_s/\eta_1{}^s\mathfrak{X}^2(\nu_s=s\nu_1)I_1{}^s.$$

If the Γ's are not too different from one and the number of modes is very large so that the dominant contributions come from the terms where all $p_1=p_2=\cdots=p_N=1$, one finds

$$\langle I_s\rangle=\eta_s/\eta_1{}^s\mathfrak{X}^2(\nu_s=s\nu_1)s!\langle I_1\rangle^s.$$

In this case, multimoding results in an apparent relative increase $(s!)^{1/2}$ for the nonlinear constant. In a practical situation (for instance, a ruby laser) the Γ's might differ appreciably from unity, and the number of modes going at the same time is not necessarily large, so that the relation does not necessarily hold.

Fluctuations around the mean sth-harmonic production can also be derived from the stochastic properties of the fundamental modes. The phase fluctuations will be subject to the same limitation as for second-harmonic generation: i.e., even spacing of the frequencies. If this condition is satisfied, the number of phase-dependent terms will be of order of N^{2s-1}, the number of phase-independent terms being of order N^s, the rms fluctuations due to phase will be of order $N^{-1/2}$.

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V. PARTIAL COHERENCE AND DOWN CONVERSION

The beat between two laser beams, or between two modes in the same beam, to generate power at a small difference frequency needs a separate discussion, because the number of modes available in wave-vector space is limited at low frequencies. This includes in particular microwave generation and rectification, i.e., the creation of a static polarization.

Consider first the influence of temporal coherence. Assume that the output of each laser consists of N frequency modes (or lines), with incoherent phases. Each frequency will beat with the components of the other beam. The bandwidth of the system detecting the beat will determine how many of these components will be effective in beating with the given one. Thus the number of efficient pairs might be less than N^2 and the energy generated at the difference frequency smaller than in the case of coherent beams. If the bandwidth of the detector is less than the spacing between lines, as is the case in microwave generation, only one component will correspond to a given line in the other beam. Thus, if the frequencies are equally spaced, the number of pairs will be N, and the energy generated N times smaller than for coherent beams. Note that this applies equally well to the beats between different components of the same beam, but does not hold for the creation of a static polarization where each component beats with itself. There is no reduction due to incoherence in the latter situation.

The influence of spatial incoherence on the beat of two monochromatic fields is similar in nature. A pair of components from the two beams cannot contribute to power generation at the difference frequency, if the difference in transverse components of the wave vectors is larger than the absolute value of the wave vector at the difference frequency, or than the inverse of the transverse dimension of the sample. This imposes roughly, the condition

$$(k_{1x}-k_{2x})^2+(k_{1y}-k_{2y})^2<k_3^2 \text{ or } a^{-2}.$$

More precisely, at a point $\mathbf{r}$ of the nonlinear dielectric the two incident fields E_1 and E_2 at frequencies ν_1 and ν_2 will interact to give rise to a polarization,

$$P_s^{\nu_3}=\chi E_{\nu_1}E_{\nu_2}^*,$$

where χ is the appropriate nonlinearity constant.[3] This polarization will in turn reradiate and give rise to a field E_{ν_3}. If we call V the finite domain where the interaction takes place, the field at a point $\mathbf{R}$, remote from V will be expressed as

$$E_{\nu_3}(\mathbf{R})=\frac{4\pi^2}{c^2}\nu_3^2\frac{e^{ik_3R}}{R}\int P_s(\mathbf{r})e^{-i\mathbf{k}_3\cdot\mathbf{r}}d\mathbf{r},$$

where k_3 is a vector of modulus $2\pi(\epsilon_3)^{1/2}\nu_3c^{-1}$ in the direction of $\mathbf{R}$. For the sake of simplicity we have assumed the dielectric constants of the nonlinear medium and the surrounding medium to be equal, their value at frequency ν_3 being ϵ_3.

The total energy radiated by the nonlinear polarization will be given by

$$W=16\pi^4\frac{\nu_3^4}{c^3}\frac{1}{R^2}\int\int P_m(\mathbf{r}')P_m^*(\mathbf{r}'')\times\frac{\sin k_3|\mathbf{r}''-\mathbf{r}'|}{k_3|\mathbf{r}''-\mathbf{r}'|}d\mathbf{r}'d\mathbf{r}''. \quad (13)$$

In the case of interest, $k_3\ll k_2$, k_1, the directions of the two optical beams will necessarily nearly coincide and their cross sections will overlap completely. If a is the common radius of these cross sections and l the length of the volume in the direction of the optical beams, we will assume that k_3a, $k_3l\lesssim 1$. In this case $\sin k_3|r''-r'|/(k_3|r''-r'|)$ is practically 1 for $\mathbf{r}'$ and $\mathbf{r}''$ in V and Eq. (13) reduces to:

$$W\approx 16\pi^4\nu_3^4c^{-3}R^{-2}\left|\int P(\mathbf{r})d\mathbf{r}\right|^2. \quad (14)$$

In the case of monochromatic beams, E_1 and E_2 can be written by means of Eq. (1) as

$$E_1=\sum_{j=1}^{J_1}a_ju_j(\mathbf{r}_0)\exp(ik_1z),$$

$$E_2=\sum_{j=1}^{J_2}b_ju_j(\mathbf{r}_0)\exp(ik_2z).$$

The same set of orthonormal modes have been chosen to represent the two beams. Statistical independence of the a_j will be assumed. More specifically, the amplitude fluctuations of the a_j will be neglected and their phases are assumed to have a uniform distribution over $(0,2\pi)$. No special assumptions are then necessary for the b_j.

The orthogonality of the modes over the cross section gives

$$P_s=\chi\sum_{j,j'}a_jb_{j'}^*u_ju_{j'}^*,$$

$$\int P_sd\mathbf{r}_0=\chi\sum_j a_jb_j^*.$$

Equation (14) can then be written in the form

$$W_3=16\pi^4\chi^2\frac{\nu_3^4}{c^3}\frac{1}{R^2}\{\sum_j|a_j|^2|b_j|^2+\sum_{j\neq j'}a_ja_{j'}^*b_j^*b_{j'}\}. \quad (15)$$

Thus the generated power depends on the relative phases of the different modes. This will result in fluctuations of the kind mentioned previously. Note that they will be extremely large as $(\Delta W/\langle W\rangle)^2\approx 1-J^{-1}$, where J is the larger number of modes necessary to represent any of the beams. The average power generated is given by the first term on the right-hand side of Eq. (15), it will be J times smaller than for coherent beams, since

$$a_j^2\sim I_1/J,\quad b_j^2\sim I_2/J.$$

The reduction factor is determined only by the less coherent beam, and the use of one coherent beam will not improve the efficiency of the process.

When the restriction $k_3l \lesssim 1$ is lifted the above conclusions still hold. The expression for the radiated energy takes the form

$$W = 16\pi^4\chi^2 \frac{\nu_3^4}{c^3} \frac{1}{R^2} \{\sum_n a_n b_n^*\}^2 F(l),$$

where

$$F(l) = 1/k_m\{\mathrm{Si}[2l(k_3+k_1-k_2)]+\mathrm{Si}[2l(k_3+k_2-k_1)]\}$$

describes the influence of k_1, k_2, k_3 and the length of the rod. For large k_3l, the energy will be radiated directionally in a cone having the direction of propagation of the optical beams for its axis.

The same kind of considerations would apply equally well to the case where the nonlinear dielectric is placed inside a wave guide or a microwave cavity.[28] There the generated power would be proportional to $[\int d\mathbf{r}\mathbf{P}_s(\mathbf{r})\cdot\mathbf{E}_3(\mathbf{r})]^2$, where $E_3(\mathbf{r})$ is the normalized field of the excited mode. When the fundamental mode of a rectangular guide has its axis along the optical beams, $P_s(\mathbf{r})$ is exactly integrated along the cross section and once more W takes the form

$$W = \{\sum_n a_n b_n^*\}^2 G(l, k_3, k_1-k_2),$$

with the same result for the average energy and its rms fluctuations.

The results obtained in these various cases are quite general; they apply to all experimental situations where the beats of two optical beams generate a difference frequency ν_3 by a quadratic process, such that $k_3a<1$. The reduction of the efficiency due to the lack of spatial coherence had in fact been pointed out by Forrester *et al.*[15] for the case of photoelectric beats. The maximum efficiency in microwave generation will be obtained when two spatially coherent sources are used. If an experiment with only partially coherent or incoherent beams is not properly analyzed, a value of the nonlinear constant much smaller than the real value would result. This is probably the reason why most of the experiments performed so far have been given an apparent discrepancy in the relation[4]

$$\chi_{xyz}(\nu_3=\nu_1-\nu_2) = \chi_{yzx}(\nu_1=\nu_2+\nu_3),$$

equating the previously defined nonlinear constant to the linear electro-optic coefficient. This statement finds supplementary strength in the fact that this relation has been verified by the experiments of Bass *et al.*[29] for the static polarization produced by a laser beam in a nonlinear dielectric. Here we are dealing with the beat of a beam with itself, the a's and b's are the same. In this case the last term in Eq. (15) does not average to zero. In fact the term between curly brackets becomes $\{\sum|a_n|^2\}^2$. There is no reduction factor coming from the lack of spatial or temporal coherence in the case of rectification. This fact is obviously used every time the intensity of an incoherent light beam is measured by the direct photoelectric current.

When the wave number k_3 becomes larger than a^{-1}, the beats between neighboring modes will start making a contribution to the radiated energy. When k_3 becomes larger than $\theta k_{1,2}$, where θ is the widest aperture of the incident beams, all beats will contribute to the radiation. When this condition is largely satisfied, the beat frequency will be radiated in a narrow beam, similar to the incident waves. The radiated energy can be computed as in the case of second harmonic. There is no reduction factor due to the spatial incoherence of the source in this case, which applies, e.g., to the generation of far-infrared radiation by beating two ruby lasers. It is also the case for microwave modulation of light. The light waves at the sum and difference frequencies will have wave vectors, obtained by adding or subtracting the microwave vector k_m to the wave vectors of the incident light beam. Hence in this respect microwave modulation of light and optical-beat generation of microwave beats are not equivalent situations. In the former phase space does not impose restrictions on the waves to be created, i.e., on the final states, whereas in the latter case it does.

VI. CONCLUSION

The stochastic properties of nonlinear optical experiments give information about the coherence functions of the light field. Conversely, a precise absolute determination of nonlinear optical susceptibilities is possible only, if the correlation between the various excited optical modes is known. Phase correlations between different laser modes can, in principle, be determined. The interpretation of experimental results depends critically on the relative magnitude of the frequency spacing between modes and the bandwidth of the photoelectric detector circuit. Relative values of nonlinear susceptibilities should be measured by calibrating the laser beam or beams with a known material undergoing the same nonlinear process. Care should be exercised that the geometries of the laser beams in sample and monitor are the same.

The classical description of the electromagnetic field which has been used in this paper should give an adequate description of the fields produced by a relatively small number of highly excited laser modes. It would be of theoretical interest to extend Glauber's quantum mechanical description of the coherence properties of the light field to nonlinear interactions.

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