FREQUENCY BANDWIDTHS IN NONDEGENERATE N-WAVE-MIXING INTERACTIONS AND INDUCED-GRATING GEOMETRIES

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We calculate frequency bandwidths for variable-frequency N-wave-mixing interactions and show that the most important parameter for bandwidth determinations is the angle between the two beams (most often one input and one output beam) whose frequencies are varying. Geometries in which the two variable-frequency beams copropagate attain the largest bandwidths, while counterpropagating variable-frequency beams produce the narrowest bandwidths. When more than two beams vary in frequency, additional angles are necessary to characterize the interaction; we calculate bandwidths for the planar case and discuss autocorrelators employing second harmonic generation. Finally, we briefly discuss the merits of a few broadband geometries.

1. Introduction

Variable-frequency N-wave-mixing interactions and induced-grating methods form the basis of a rich variety of techniques and devices. Saturation spectroscopy [1,2], polarization spectroscopy [3,4], coherent anti-Stokes Raman spectroscopy [5], and the tunable-laser-induced-grating technique [6–8] are examples of such processes for the case $N = 4$. Pulselength-measurement techniques based on second-harmonic-generation autocorrelators [9], when applied to picosecond tunable dye lasers, are variable-frequency three-wave-mixing processes.

In utilizing such interactions, it is usually important to attain a large frequency bandwidth, that is, to maintain phase-matching despite large variations in two or more of the input or output beam frequencies [8]. Researchers have employed a wide variety of geometries for N-wave-mixing interactions and induced-grating experiments, and various authors have treated the bandwidth problem in the past, usually reporting the discovery of a particularly good (broadband) or bad (narrowband) geometry [10–15]. (Of course, if one is interested in optical frequency filters [11], the good-bad dichotomy reverses.) It is well-known, for example, that the four-wave-mixing phase conjugate geometry is a particularly narrowband geometry [10,16].

Although such specific results can be found in the literature, a general treatment of the geometry-dependent bandwidth issue does not appear to exist. Consequently, it is the purpose of this paper to present such a treatment – which will be seen to be quite simple. We also give an intuitive picture for these results. We begin with two sections treating the case in which only two frequencies vary and then include a section on the more complicated situation involving three variable frequencies. A discussion of broadband wave-mixing geometries completes the analysis.

2. Interactions producing a variable-frequency output beam

We first consider the two-variable-frequency case in which one input frequency and the output frequency vary. Our treatment will suffice for all N-wave-mixing (and induced-grating) interactions, although it was first presented to explain the narrowband nature of the four-wave-mixing phase-conjugate process, in which the input and the counterpropagating output beam vary in frequency.

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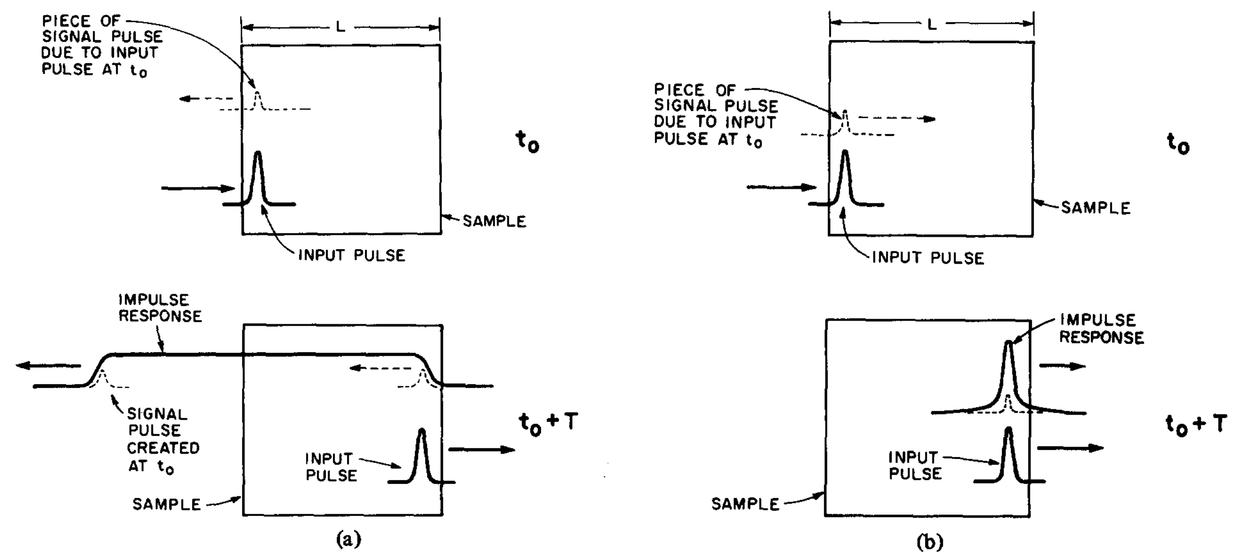

Fig. 1. (a) Illustration of the finite time response, and hence narrow bandwidth, of an interaction involving counterpropagating output and variable-frequency input beams. The box corresponds to the interaction region. The upper pulse in each box is in part of the output pulse, created by the interaction, while the lower pulse in each box is the input pulse; their vertical separation is for the purposes of illustration only. (b) Illustration of the instantaneous time response and hence, very large bandwidth, of an interaction involving copropagating output and variable-frequency input beams. See comments in the caption of (a).

Following Siegman et al. [16], suppose that the wave-mixing process has an interaction length L, and a corresponding transit time $T = L/c$ along the output-wave direction (fig. 1a). Let the fixed-frequency beams be cw waves and the variable-frequency input beam be a short pulse (with pulse-length much less than T). The wave-mixing process will create output radiation only at points in time and space when all input beams coincide. As a result, when the input pulse ***counterpropagates*** with respect to the about-to-be-created output wave, the short pulse will shed a continuous "tail" of induced output wave. The output wave begins to appear at the instant that the input pulse first arrives at the entrance to the interaction region, but radiation created later cannot "catch up" with radiation created earlier: the younger radiation experiences later birth, but in addition, its point of creation is ***behind*** the older output radiation. As a result, the approximate pulse-shape of the output wave will be a square wave of duration $2T$ or $2L/c$. The time response of the interaction is thus not instantaneous.

The frequency response of the interaction, if one considers this as a linear system with input field, is then the Fourier transform of the above impulse response. Because the impulse response is broadened by an amount on the order of $2T$ in time, the frequency response of the interaction with respect to the variable-frequency input beam will be finite, that is, the geometry will have a narrow bandwidth. In other words, frequency scans broad compared to $1/2T$, a large phase-mismatch will occur (or equivalently, the Bragg angle for the interaction will vary from its original value or cease to exist).

Now consider a geometry in which the output beam and variable-frequency input beam ***copropagate*** (fig. 1b). We can calculate the impulse response for this interaction in an analogous manner; it is easy to see that the impulse response for such a geometry is a delta-function. Because the output beam and input beam copropagate, the output beam will consist of an identical short pulse copropagating – both in space and in time – with the short input pulse. The frequency response of this system will be large, and hence, despite a large frequency scan, very little or no phase mismatch will result.

We have neglected dispersion in this argument. The presence of dispersion will of course weaken this argument somewhat: if the two pulses have different group velocities, they will separate as they propagate, and the impulse response will actually have nonzero width. However, its width will in general still be much less than that of the counterpropagating-beam case.

Thus, wave-mixing geometries in which the output beam and the variable-frequency input beam *counter*propagate will be narrowband, and, conversely, geometries in which these beams *co*propagate will be broadband. In particular, four-wave-mixing phase conjugators (in which these beams counterpropagate) have narrow bandwidths, as is well-known. What is not so well-known, however, is that such phase conjugators have narrow bandwidth *because* these beams counterpropagate.

3. Interactions involving two arbitrary variable-frequency beams

The argument in the preceding section demonstrates that wave-mixing geometries in which one input frequency and the output frequency vary will have a maximally broad bandwidth if the variable-frequency beams copropagate. We now extend this result to *N*-wave-mixing geometries in which *any* two frequencies vary. We also work more rigorously, deriving an analytical expression for the interaction bandwidth.

Consider a general *N*-wave mixing process for which

$$\sum_{i=1}^{N} \mu_i \omega_i = 0, \qquad (1)$$

where $\mu_i = \pm 1$, and ω_i is a (positive-valued) input or output frequency. We make no distinction between input and output beams since they enter equivalently into the analysis. If the *k*-vectors of the interaction are labeled $\boldsymbol{k}_i$ for $i = 1 \dots N$, the phase-mismatch will be

$$\Delta \boldsymbol{k} = \sum_{i=1}^{N} \mu_i \boldsymbol{k}_i. \qquad (2)$$

Now suppose that initially the process is phasematched ($\Delta \boldsymbol{k} = 0$), but that two frequencies are varied while all beam directions remain fixed. Necessarily, eq. (1) will remain satisfied, so that if ω_i is incremented by $\delta\omega_i$ and ω_j by $\delta\omega_j$, we have

$$\mu_i \delta\omega_i + \mu_j \delta\omega_j = 0. \qquad (3)$$

If Ω is the angle between the two variable-frequency beams, the squared magnitude of the phase-mismatch due to the variation in frequencies will be

$$|\Delta \boldsymbol{k}|^2 = (\delta\omega_i^2/c^2)(n_i^2 + n_j^2 - 2n_i n_j \cos\Omega) \qquad (4)$$

neglecting dispersion. Setting $|\Delta \boldsymbol{k}|^2$ equal to $(\pi/L)^2$, where L is the interaction length, allows simple solution for the actual bandwidth, $\Delta\omega_i$

$$\Delta\omega_i = (2\pi c/L)\,[(n_i - n_j)^2 + 4n_i n_j \sin^2(\Omega/2)]^{-1/2}, \qquad (5)$$

which is plotted in fig. 2. We see that the bandwidth will be maximized when $\Omega = 0$, that is, when the variable-frequency beams *copropagate*. The optimal case, $\Delta\omega_i \to \infty$, occurs when these beams copropagate and their refractive indices are equal: $n_i = n_j$. When the refractive indices are unequal, the bandwidth will decrease for all geometries, but copropagating beams will continue to maximize it. In addition, note that the bandwidth is independent of μ_i and μ_j. Finally, for degenerate processes involving m_i photons from the beam of frequency ω_i and *k*-vector $\boldsymbol{k}_i$, the bandwidth will be reduced by a factor of m_i; all other conclusions remain valid.

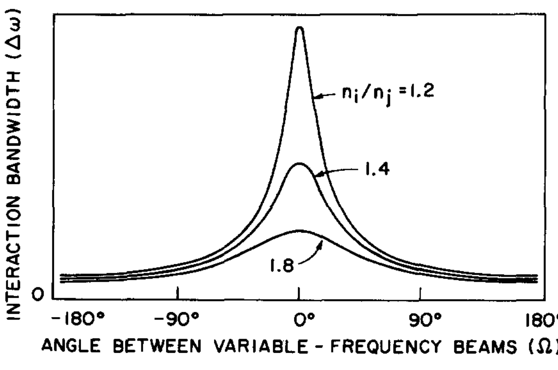

Fig. 2. The interaction bandwidth for an *N*-wave-mixing interaction in which two beam frequencies vary versus the angle between these two beams (Ω). The minimum bandwidth, occurring when $\Omega = \pm\pi$, is given by: $\Delta\omega_{\min} = 2\pi c/[(n_i + n_j)L]$, while the maximum bandwidth attainable by a process, occurring for copropagating variable-frequency beams ($\Omega = 0$) is given by $\Delta\omega_{\max} = 2\pi c/(|n_i - n_j|L)$, or if $n_i = n_j$, $\Delta\omega_{\max} = 2\pi c/(|\omega_i n_i' - \omega_j n_j'|L)$. When three frequencies vary, copropagation may not yield the maximal bandwidth.

Dispersive effects generally play only a very small role in the determination of the bandwidth since index-mismatch effects due to the changes in frequency usually predominate. Actual calculation of the geometry bandwidth will, however, occasionally require the additional dispersive terms, since choosing a large-bandwidth geometry often forces cancellation or near-cancellation of the refractive-index terms in eq. (5), with the dispersive terms then dominating. We can include dispersion effects in this analysis by replacing $n_\eta + \omega_\eta n'_\eta$, where $n'_\eta = \partial n/\partial\omega|_{\omega=\omega_\eta}$ and $\eta = i, j$. Copropagating beams will continue to maximize the bandwidth provided that $|\omega_\eta n'_\eta| < n_\eta$.

4. Interactions with three variable-frequency beams

We can generalize eq. (4) to N-wave-mixing processes in which *three* frequencies (ω_i, ω_j and ω_l) vary, obtaining

$$|\Delta k|^2 = (\delta\omega_i^2/c^2)[n_i^2 + n_l^2 - 2n_i n_l \cos\Omega_{il}]$$
$$+ (\delta\omega_j^2/c^2)[n_j^2 + n_l^2 - 2n_j n_l \cos(\Omega_{ij} - \Omega_{il})]$$
$$+ 2\mu_i\mu_j(\delta\omega_i\delta\omega_j/c^2)[n_l^2 + n_i n_j \cos\Omega_{ij}$$
$$- n_i n_l \cos\Omega_{il} - n_j n_l \cos(\Omega_{ij} - \Omega_{il})], \tag{6}$$

where Ω_{ij} is the angle between $\boldsymbol{k}_i$ and $\boldsymbol{k}_j$, and now, Ω_{il} is the angle between $\boldsymbol{k}_i$ and $\boldsymbol{k}_l$ measured in the same direction as Ω_{ij}. We assume a coplanar arrangement. Notice that now μ_i and μ_j appear and may help to determine the optimal values of Ω_{ij} and Ω_{il} for a particular process. The bandwidth for a particular interaction will depend on the relation between $\delta\omega_i$ and $\delta\omega_j$, and is easily calculated from eq. (6) upon appropriate substitution.

Application of this result to specific wave-mixing processes is straightforward, although computationally messy. For example, consider a second-harmonic-generation (SHG) autocorrelator (e.g., for ultrashort-pulsewidth measurement of dye-laser pulses), for which it is desirable to minimize $|\Delta k|$, and hence the amount of realignment necessary, as the input frequency is varied. Since these devices generally employ a non-collinear-input-beam geometry, we require the use of eq. (6). Using $\delta\omega_i = \delta\omega_j = \delta\omega_l/2$ and $\mu_i = \mu_j = -\mu_l$, we find

$$\cos\Omega_{il} = (n_i^2 - n_j^2 + 4n_l^2)/4n_i n_l, \tag{7}$$

$$\sin\Omega_{ij} = (2n_l/n_j)\sin\Omega_{il}, \tag{8}$$

for the beam angles that optimize the autocorrelator bandwidth. Since $n_l = (n_i + n_j)/2$ for an initially phase-matched interaction, we find that $\Omega_{ij} = \Omega_{il} = 0$, and again, copropagating beams optimize the bandwidth. In practice, most commercial autocorrelators [9] employ non-zero values of Ω_{ij} and Ω_{il} in order to obtain background-free operation, thus sacrificing some bandwidth.

In a higher-order process with one or more fixed-frequency beams in addition to the variable-frequency beams considered above for the autocorrelator, we obtain a bandwidth-optimizing beam geometry with *noncollinear* variable-frequency beams. Additional beam(s) allow a more general index-matching equation, which we will now take to be: $n_l = (n_i + n_j - \delta)/2$, where δ incorporates all fixed-frequency-beam refractive indices. It is then easy to show that, for small values of δ, the optimal beam angles, Ω_{ij} and Ω_{il} will be

$$\Omega_{il} \approx [(n_j/n_i n_l)\delta]^{1/2} \quad \text{if } \delta \geq 0,$$
$$\approx 0 \quad \text{if } \delta < 0, \tag{9}$$

$$\Omega_{ij} \approx 2[(n_l/n_i n_j)\delta]^{1/2} \quad \text{if } \delta \geq 0,$$
$$\approx 0 \quad \text{if } \delta < 0, \tag{10}$$

working to first order in δ. Thus, collinear propagation of the variable-frequency beams is not the universal solution to the problem of N-wave-mixing bandwidth maximization when more than two beams vary in frequency.

5. Discussion and conclusions

While it is well known that phase conjugation by four-wave mixing is an inherently narrowband process ‡, the set of broadband four-wave-mixing geometries is not so well-known. In this section, we discuss a few geometries that achieve broad bandwidth and, hence, are appropriate for variable-frequency non-

‡For footnote see next page.

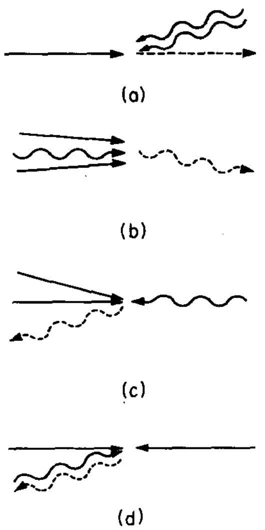

Fig. 3. (a) The infinite-bandwidth geometry of saturation and polarization spectroscopy. The wiggly lines denote variable-frequency beams, and the dashed line represents the output beam. (b) The large-bandwidth geometry involving near-copropagating of all beams. The wiggly lines denote variable-frequency beams, and the dashed line represents the output beam. (c) Another large-bandwidth geometry. The wiggly lines denote variable-frequency beams, and the dashed line represents the output beam. In this case, two beams can be made to counterpropagate [7,8], thus simplifying alignment considerably. See text for additional advantages and disadvantages of this geometry. (d) The (narrow-bandwidth) geometry of phase conjugation by four-wave mixing. The wiggly lines denote variable-frequency beams, and the dashed line represents the output beam. Observe that the variable-frequency beams *counterpropagate.*

linear-optical techniques. We also consider the practical problems of the presence of backgrounds, such as polarizer leakage and small-angle scattered light.

The usual geometry for saturation and polarization spectroscopy [1–4] (fig. 3a) employs two variable-frequency beams combined in the same beam emanating from the same laser. The output beam remains at a fixed frequency. Like phase conjugation, these interactions, when viewed as four-wave-mixing processes, are automatically phase-matched. Unlike phase conjugation, however, these processes achieve ***infinite*** bandwidth due to the necessary exact copropagation and the same wavelengths and indices of refraction of the variable-frequency beams. This conclusion is, of course, independent of dispersion effects. On the negative side, however, this geometry always possesses a background of at least polarizer leakage.

The use of a fixed-frequency output beam in the above geometry requires the variation of two input frequencies by exactly the same amounts. This is easy if the beams are one and the same, but prohibitively difficult otherwise. Consequently, most variable-frequency wave-mixing experiments control a single input-beam frequency allowing the output-beam frequency to vary as a result. A popular geometry of this sort is the collinear-beam geometry, which attains very long interaction lengths, but possesses a beam separation problem when the wavelengths involved are approximately equal. *Near*-copropagation of all beams (fig. 3b) is a useful alternative arrangement that allows easier beam separation [18]. Still, a problem exists in the form of small-angle-scattered-light background from all of the input beams, which can be reduced by using larger angles between the beams, but at a cost in frequency bandwidth.

As a partial solution to this trade-off, we have employed a broadband four-wave-mixing geometry (fig. 3c) in which the two variable-frequency beams approximately copropagate, but the other two beams emanate from other angles. Such a geometry can significantly reduce the scattered light in a given beam direction, reducing the number of beams producing scattered light to a maximum of one. In earlier experiments [7] we allowed the other two (fixed-frequency) beams to nearly counterpropagate with the variable-frequency beams to maintain a long interaction length and ease of alignment. (Polarization- and wavelength-filtering helped to reduce the scattered-light background from the one beam that remained in a nearly copropagating direction with the output beam.) This type of four-wave-mixing geometry is useful, and we obtained good results with it, but it should be mentioned here that, unlike the other geometries mentioned in this section, this arrangement suffers from its own drawbacks. It has severe constraints regarding the

‡ "Forward-going phase conjugation" [17] employs the variable-frequency beams in a nearly copropagating geometry, and hence, achieves the desired large bandwidth. Such an arrangement has its drawbacks, however: mainly a small *angular* bandwidth [17] in contrast to the 2π angular bandwidth of the usual "backward-going" phase conjugation. As a result, researchers generally shun forward-going phase conjugation, since angular bandwidth is more important in the phase-conjugation process than frequency bandwidth.

allowable wavelengths in the interaction: often, the wavelengths involved cause phasematching and a long interaction length to be incompatible due to the required Bragg angle, and, for some wavelengths, phasematching is impossible. Ref. [8] provides a more complete analysis of this problem and of four-wave mixing geometries, in general.

We must conclude that there is no wave-mixing geometry that is ideal for all applications requiring broad bandwidth. Choice of appropriate geometry will always require thoughtful analysis of the material parameters and sources of experimental noise, in particular. In any case, an understanding of the significant role played by the relevant beam angles of the geometry in determining the frequency bandwidth of the interaction is certainly helpful.

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