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SPECTRAL DISTRIBUTION OF AN ULTRAFAST SUPERCONTINUUM LASER SOURCE

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Supercontinuum generation, the production of intense fast broadband pulses by passing picosecond and subpicosecond laser pulses through nonlinear media was first observed in 1970 by Alfano and Shapiro [1]. The shape, fine structure and extent of the spectrum produced are functions of the nonlinear index of refraction of the medium, the shape, wavelength, duration, intensity and phase modulation of the pump laser pulse, and the interaction length of the pulse in the medium. Typically, the broadened spectrum consists of larger frequency extent towards the blue than the red by factors of approximately 1.6–2. The coherent and ultrafast wide frequency band of this supercontinuum has been mainly used as a spectral tool for time-resolved absorption spectroscopy [2,3] and nonlinear optical effects [2,4]. Recently, new uses in engineering applications [5], such as ranging, imaging, atmospheric remote sensing and optical fiber characterization, have been proposed. In this letter we investigate in a consistent manner the spectral distribution of this supercontinuum, in particular, the intensity distribution, the asymmetry in the Stokes and anti-Stokes region and the extent of the spectrum.

Maxwell's equation in a nonlinear medium is given by

$$\nabla^2 E - (n^2/c^2)\partial^2 E/\partial t^2 = (2nn_2/c^2)\frac{\partial^2}{\partial t^2}\langle E\cdot E\rangle E\,, \qquad (1)$$

where n is the linear index of refraction and n_2 is the nonlinear index of refraction. In this equation we have neglected the dispersion and the imaginary part of n and n_2.

Assuming the experimental regime whereby one component of E is present and its transverse variation is neglected, Maxwell's equation reduces to

$$\partial^2 E/\partial z^2 - (1/v_g^2)\,\partial^2 E/\partial t^2 = (nn_2/c^2)\frac{\partial^2}{\partial t^2}|E|^2 E\,. \qquad (2)$$

Writing the electric field $E(z, t) = E_0\Phi(z, t)$, and defining the new variables Z, T: $T = t/\tau$ and $Z = z/v_g\tau$, eq. (2) reduces to

$$(\partial^2/\partial Z^2 - \partial^2/\partial T^2)\,\Phi = \epsilon\frac{\partial^2}{\partial T^2}|\Phi|^2\Phi\,, \qquad (3)$$

where τ is the characteristic time associated with the pulse and $\epsilon = n_2|E_0|^2/n$. Typical values for ϵ in the region of interest are in the range 10^{-4}–10^{-2}. In the following, eq. (3) is solved by the perturbation technique referred to as the method of multiple scales [6].

The functional dependence of Φ on Z, T and ϵ is not disjoint. To first order in ϵ, Φ depends on the combinations ϵT and ϵZ as well as on the individual T, Z and ϵ. Carrying the perturbation to higher orders, Φ depends additionally on $\epsilon^2 T$, $\epsilon^2 Z$, $\epsilon^3 T$, $\epsilon^3 Z$ .... Hence, it is convenient to write $\Phi(Z, T; \epsilon)$ as

$$\Phi(Z, T; \epsilon) = \hat{\Phi}(Z_0, T_0, Z_1, T_1, Z_2, T_2, \ldots; \epsilon) , \quad (4)$$

where

$$T_0 = T , \quad T_1 = \epsilon T , \quad T_2 = \epsilon^2 T , \ldots ,$$

$$Z_0 = Z , \quad Z_1 = \epsilon Z , \quad Z_2 = \epsilon^2 Z , \ldots ,$$

and where the number of independent variables has been increased. The T_n and Z_n represent different time and distance scales. Thus, instead of determining Φ as a function of T and Z, we determine Φ as a function of $T_0, T_1, T_2 \ldots$ and $Z_0, Z_1, Z_2 \ldots$ and seek a uniform expansion solution to Φ in the form:

$$\begin{aligned}\Phi = &\Phi_0(T_0, Z_0, T_1, Z_1, T_2, Z_2, \ldots) \\ &+ \epsilon\Phi_1(T_0, Z_0, T_1, Z_1, T_2, Z_2, \ldots) \\ &+ \epsilon^2\Phi_2(T_0, Z_0, T_1, Z_1, T_2, Z_2, \ldots) + \ldots . \end{aligned} \quad (5)$$

Using the chain rule for derivatives:

$$\begin{aligned}\partial/\partial T &= \partial/\partial T_0 + \epsilon\,\partial/\partial T_1 + \epsilon^2 \partial/\partial T_2 + \ldots , \\ \partial^2/\partial T^2 &= \partial^2/\partial T_0^2 + 2\epsilon\,\partial^2/\partial T_0 \partial T_1 \\ &\quad + \epsilon^2(2\,\partial^2/\partial T_0\,\partial T_2 + \partial^2/\partial T_1^2) + \ldots , \end{aligned} \quad (6)$$

with similar expressions for $\partial/\partial Z$ and $\partial^2/\partial Z^2$.

Using expressions (5) and (6) in the differential equation (3), and equating the respective coefficients of ϵ^n, we have for the ϵ^0, ϵ, ϵ^2 terms

$$\left(\frac{\partial^2}{\partial Z_0^2} - \frac{\partial^2}{\partial T_0^2}\right)\Phi_0 = 0 , \quad (7a)$$

$$\left(\frac{\partial^2}{\partial Z_0^2} - \frac{\partial^2}{\partial T_0^2}\right)\Phi_1 + \left(2\,\frac{\partial^2}{\partial Z_1 \partial Z_0} - 2\,\frac{\partial^2}{\partial T_1 \partial T_0}\right)\Phi_0 = \frac{\partial^2}{\partial T_0^2}\,|\Phi_0|^2\Phi_0 , \quad (7b)$$

$$\begin{aligned}&\left(\frac{\partial^2}{\partial Z_0^2} - \frac{\partial^2}{\partial T_0^2}\right)\Phi_2 + \left(2\,\frac{\partial^2}{\partial Z_1 \partial Z_0} - 2\,\frac{\partial^2}{\partial T_1 \partial T_0}\right)\Phi_1 \\ &+ \left(\frac{\partial^2}{\partial Z_1^2} - \frac{\partial^2}{\partial T_1^2} + 2\,\frac{\partial^2}{\partial Z_0 \partial Z_2} - 2\,\frac{\partial^2}{\partial T_0 \partial T_2}\right)\Phi_0 \\ &= 2\,\frac{\partial^2}{\partial T_1 \partial T_0}\,|\Phi_0|^2\Phi_0 + \frac{\partial^2}{\partial T_0^2}\,|\Phi_1|^2\Phi_1 . \end{aligned} \quad (7c)$$

If the incoming pulse is described by:

$$\Phi_{in} = f(Z_0 - T_0)\exp(\mathrm{i}KZ_0 - \mathrm{i}WT_0) , \quad (8)$$

where $W = (\omega\tau)$, τ is the pulse width, and $f(Z_0 - T_0)$ is the pulse form function (gaussian, hyperbolic secant, etc.), we take Φ_0 in the form:

$$\Phi_0 = A(Z_0, T_0, Z_1, T_1, \ldots) f(Z_0 - T_0)\exp(\mathrm{i}KZ_0 - \mathrm{i}WT_0). \quad (9)$$

In the following, the specific form of A is computed to first order in ϵ which means to values of $\epsilon Z \sim O(1)$, the solution is valid. (In the original variables ϵZ is $n_2|E_0|^2 z/c\tau$.) Introducing the new variables:

$$U_1 = Z_1 - T_1 , \quad U_1 = \epsilon U , \quad V_1 = Z_1 , \quad V_1 = \epsilon V = \epsilon Z , \quad (10)$$

and writing $A(Z_1, T_1)$ in polar coordinates,

$$A = a\,\mathrm{e}^{\mathrm{i}\beta} , \quad (11)$$

the equations determining A can be deduced directly from [7b]. The equations for a and β are given by:

$$\begin{aligned}\partial a/\partial V_1 &= \tfrac{1}{2}a^3 \operatorname{Re} G(U_0) , \\ \partial\beta/\partial V_1 &= \tfrac{1}{2}a^2 \operatorname{Im} G(U_0) , \end{aligned} \quad (12)$$

where the function $G(U_0)$ is given by:

$$G(U) = \frac{-W^2 f^3 + 6\mathrm{i}Wf^2 f' + 6f(f')^2 + 3f^2 f''}{f' + \mathrm{i}Wf} , \quad (13)$$

and where the derivatives are with respect to the variable U_0. For the gaussian form function $f(U_0) = \mathrm{e}^{-U_0^2}$, the real and imaginary parts of $G(U_0)$ are respectively:

$$\begin{aligned}R(U_0) &= \operatorname{Re} G(U_0) = [\exp(-2U_0^2)/(4U_0^2 + W^2)] \\ &\quad \times (12U_0 - 10U_0 W^2 - 72U_0^3) , \\ I(U_0) &= \operatorname{Im} G(U_0) = [\exp(-2U_0^2)/(4U_0^2 + W^2)] \\ &\quad \times (W^3 - 12U_0^2 W + 6W) . \end{aligned} \quad (14)$$

The solutions of eq. (12) are:

$$\begin{aligned}a &= 1/[1 - R(U_0)V_1]^{1/2} , \\ \beta &= -[I(U_0)/2R(U_0)]\,\ln[1 - R(U_0)V_1] . \end{aligned} \quad (15)$$

These solutions differ from those in the SPM literature in that a is not a constant and β is not symmetric in U_0. They reduce to those for $V \ll 1$, and $\omega\tau \gg 1$, in that case:

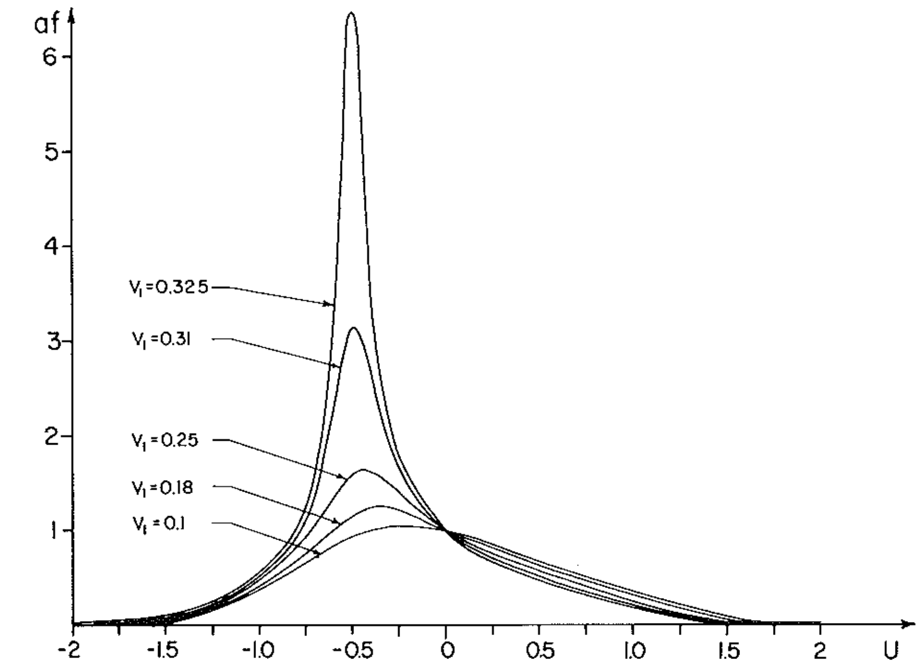

Fig. 1. The magnitude of the electric field as function of U for different V_1's.

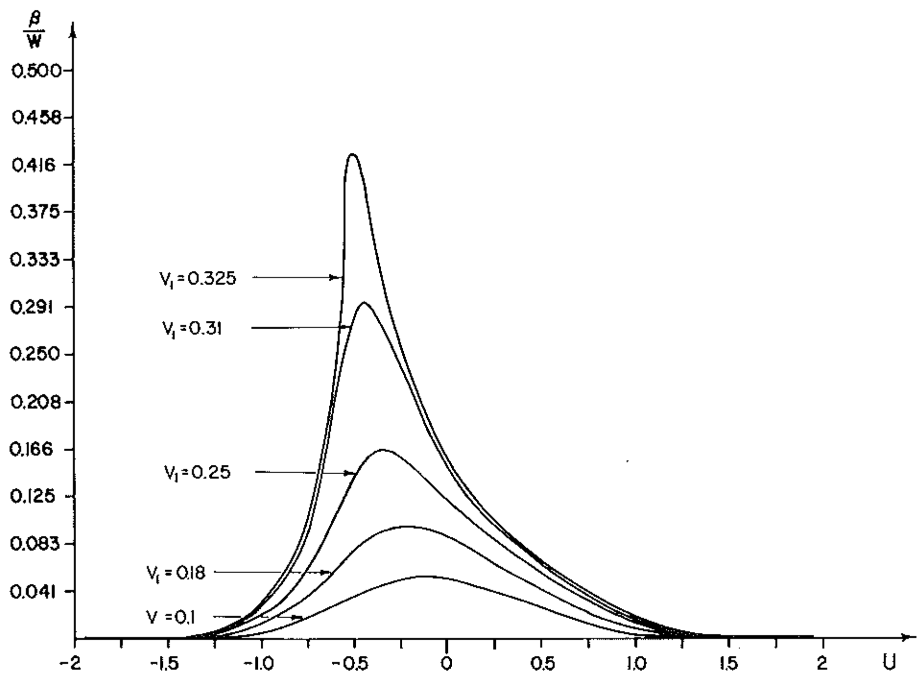

Fig. 2. The phase function β as function of U for different V_1's.

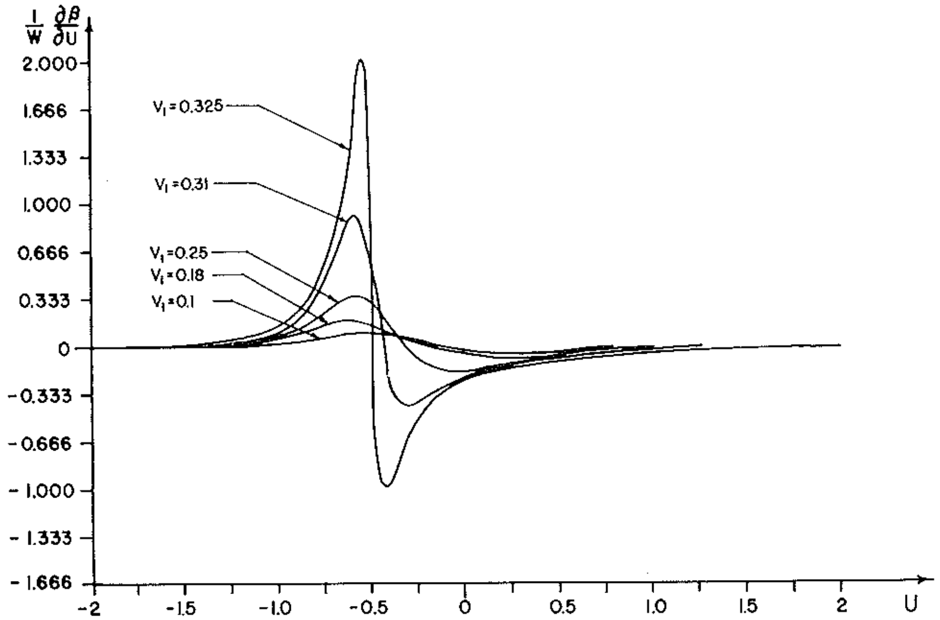

Fig. 3. The derivative of the phase function as function of U for different V_1's.

$a \sim 1$

$$\beta \sim \tfrac{1}{2} I(U_0)\, V_1 \sim \tfrac{1}{2} W \exp(-2U_0^2)\, V_1 \,. \tag{16}$$

In figs. 1, 2 and 3; fa, β and $\partial\beta/\partial U$ are respectively plotted as functions of U for different values of V_1, and $W \gg 1$. fa is the magnitude of the electric field, and β is the phase mod(WU). The salient features of figs. 1 and 2 are that, at high V_1, the curves become skewed and distorted from the form function, in particular at later times ($U<0$).

The spectral distribution for the outgoing signal is given by:

$$S(\omega', z) \propto |\tilde{\mathscr{E}}(\omega', z)|^2 \,, \tag{17}$$

where

$$\tilde{\mathscr{E}}(\omega', z) = (1/2\pi)\ \mathrm{Re} \int \mathrm{e}^{\mathrm{i}\omega' t} E(t, z)\, \mathrm{d}t \,. \tag{18}$$

In the U–V coordinates, $W = \omega\tau$ and $W' = \omega'\tau$,

$$\tilde{\mathscr{E}}(W', V) = (\tau/2\pi)\ \mathrm{Re} \int_{-\infty}^{\infty} \mathrm{d}U \exp[\mathrm{i}(W - W')U] \times f(U)\, a(U, V) \exp[\mathrm{i}\beta(U, V)] \,. \tag{19}$$

The extent of the spectral distribution can be estimated through:

$$W' - W|_{\text{anti-Stokes}} \approx \max(\partial\beta/\partial U) \,, \tag{20a}$$

$$W' - W|_{\text{Stokes}} \approx \min(\partial\beta/\partial U) \,. \tag{20b}$$

The asymmetry between the Stokes and anti-Stokes frequency extent increases as the value of V_1 increases. (see fig. 3.) Table 1 summarizes the approximate frequency extents for some values of V_1, and compares them with the traditional SPM results. It is worth noting that this asymmetry results without recourse to

Table 1
The approximate frequency extents on the Stokes and anti-Stokes sides for selected V_1 in the present theory and as compared with the traditional SPM result.

V_1	$\lvert\Delta\omega/\omega\rvert^{\mathrm{S,aS}}_{\mathrm{SPM}}$	$\Delta\omega^{\mathrm{S}}_{\max}/\omega$	$\Delta\omega^{\mathrm{aS}}_{\max}/\omega$
0.1	0.060	0.054	0.074
0.18	0.109	0.097	0.171
0.25	0.151	0.156	0.347
0.31	0.188	0.404	0.911
0.325	0.197	0.995	2.03

contributions from plasmas, response time of the nonlinearity, etc. The values calculated at $V_1 = 0.18$ are in excellent agreement with the frequency extent observed in solids and liquids in ref. [1].

In comparing the spectral distribution deduced from the above with experimental results, the sample thickness should be chosen such that it is much smaller than the self-focusing length [7]. Filaments will introduce an uncertainty in the value of E_0 and consequently V_1.

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