without Li^+ at the same temperature and n_i is the intrinsic carrier concentration. This effect could still be important in the Si-doped material if the Li^+ concentration is large compared to n_e. This latter condition is probably well satisfied at 900°C as measurement[6] indicates the total Li concentration $\sim 2-3 \times 10^{19}$ cm^{-3}. Therefore, the first method of enhancement would be if V_{As}^- is an ionized acceptor.[8] The second method would be if V^- were V_{Ga}^-. The enhancement of V_{Ga}^- would decrease V_{Ga} via

$$V_{Ga} + e^- \rightleftarrows V_{Ga}^-$$

which would increase V_{As} by the equilibrium *eqn*,

$$[V_{Ga}]\,[V_{As}] = K.$$

The suggestion of V_{Ga} acceptors is in accord with previous experimental work.[6,8] The use of V_{As} to promote Si site transfer was also proposed by Queisser[3] to explain the photoluminescence of heat-treated, Si-doped GaAs.

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SECOND HARMONIC GENERATION FROM SHORT PULSES*

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The second harmonic pulse shape generated from ultrashort pulses is calculated. In long dispersive crystals the pulse width may be much broader than the incident pulse and the power conversion ratio may saturate at a low value. These effects should be prominent in $LiNbO_3$ with 4×10^{-13} sec pulses.

The second harmonic power generated from short pulses which contain a number of frequency components such as those from a mode-locked laser is calculated. Because of the frequency dependence of the phase-matching angle in physical crystals, the complete spectrum of ultrashort pulses cannot be simultaneously phase matched. This results in a broadening of the second harmonic pulse width in time which is proportional to the length of the crystal. Furthermore, the peak power of the second harmonic which can be generated by a fixed incident intensity is limited, even in a very long crystal. We shall show that these effects become important, particularly in $LiNbO_3$ with mode-locked pulses from a Nd^{+++} glass laser. Since the modes of such a laser can easily range in wavelength over a 100-Å-wide band, the mode-locked pulse theoretically may be 4×10^{-13} sec long.

Consider the electric field of a mode-locked laser pulse consisting of N modes of equal amplitude E_0 and equal frequency spacing δ, all with phase zero. The field of the nth mode is $E_n = E_0 e^{i(\omega_n t - k_n z)}$, where $\omega_n = \omega_0 + n\delta$ and $k_n = k(\omega_n)$. The sum frequency field generated from mixing E_n and E_m in a crystal of length L is[2]

$$E_{nmi}(L) = -\frac{i(\omega_{n+m})}{cn_2} 2\pi\, d_{ijk}\, E_{nj} E_{mk} \frac{e^{i\Delta k_{nm} L} - 1}{i\Delta k_{nm}}\, e^{i(\omega_{n+m} t - k_{n+m} L)}, \quad (1)$$

where $\omega_{n+m} = \omega_n + \omega_m$, $k_{n+m} = k(\omega_{n+m})$ and $\Delta k_{nm} = k_{n+m} - k_n - k_m$. d_{ijk} is the nonlinear coefficient of the crystal, with indices ijk indicating the crystalline axes and the components of the various electric fields among these axes. n_2 is the refractive index near the second harmonic frequency. The total

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second harmonic intensity, averaged over an optical cycle, is

$$I_2(L) = \frac{n_2 c}{8\pi} \left| \sum_{n,m=-N/2}^{N/2} E_{nmi}(L) \right|^2 \sin^2 \theta_{2i} , \qquad (2)$$

where θ_{2i} is the angle between the i crystal direction and $\mathbf{k}(2\omega)$.

If the second harmonic is an extraordinary ray and the fundamental is an ordinary ray, and if the crystal is exactly phase matched at the central frequency ω_0, then

$$\Delta k_{nm} = \frac{\omega\delta}{c} [2a_2^e - a_0^o] (n+m) \equiv (n+m)\kappa . \qquad (3)$$

For a frequency width $N\delta \ll \omega_0$, the refractive index for the ordinary ray at the fundamental frequency ω_m is given by $n_m = n_0 + ma_0^o\delta$ and for the extraordinary ray at the frequency ω_{n+m} by $n_{n+m} = n_2^e + (n+m)a_2^e\delta$, where n_2^e and a_2^e are calculated at the phase-matching angle so that $n_2^e = n_0$. In this case the intensity becomes

$$I_2(L) = \frac{2\pi\omega_0^2}{n_0 c} d_{ijk}^2 E_{0j}^2 E_{0k}^2 \sin^2 \theta_{2i} |S|^2 , \qquad (4)$$

where

$$S = \sum_{n,m=-N/2}^{N/2} \frac{e^{i(n+m)\kappa L} - 1}{i\kappa(n+m)} e^{i(n+m)\phi}$$

with $\phi = \delta[t - (n_0^o + a_2^e\omega_0) L/c]$.

Performing the summation over the discrete modes n and m, we find a rather simple form for S under the following reasonable assumptions. There are a large number of modes ($N \gg 1$), the fractional frequency spread is small ($N\delta/\omega_0 \ll 1$), the phase mismatch between adjacent modes is small ($\kappa L \ll \pi$), and the time of observation is shorter than the time between pulses in the pulse train of the mode-locked laser ($\phi \leq 1$). Furthermore, we displace the time origin to $t' = t - (n_0^o + 2a_2^e\omega_0)L/c$, and define unitless parameters $M = N\kappa L/2\pi$ and $T = t'/T_p$, with T_p the fundamental pulse width $= 2\pi/N\delta$. In this case

$$S = S(2\pi(T-M)) - S(2\pi T) , \qquad (5)$$

where

$$S(x) = \frac{2N}{\kappa} \left[Si(x) - \frac{1-\cos x}{x} \right]$$

$Si(x)$ is the tabulated function

$$\int^x \frac{\sin u}{u} du .$$

This can also be written in the form

$$S = \frac{2N}{\kappa} \int_{\pi T}^{\pi(T-M)} \frac{\sin^2 u}{u^2} du . \qquad (6)$$

Let us examine this function for various limits. If the crystal length is small, M is small; then (6) can be written as $|S| = N^2 L \sin^2 \pi T/(\pi T)^2$, which is the same result one would derive for a monochromatic wave of amplitude NE_0 and results in second harmonic power generation proportional to L^2 as expected.[2] In this case the second harmonic pulse width is a constant slightly less than the initial pulse width.

The limit of S for large M is most easily seen from Eq. (5). $S(x)$ is an odd, monotonic function of x so that S is symmetrical around $T = M/2$, where $S = 2S(\pi M)$. For large x, $\mathrm{Si}(x) \to \pi/2$, while $(1 - \cos x)/x \to 0$, so that the peak value of S for large M is $2\pi N/\kappa$, independent of length. This means that for sufficiently long crystals, the peak power of second harmonic saturates, and the pulse merely broadens in time. The saturation value for the power conversion ratio is given by

$$\frac{I_2}{I_0} = \frac{(8\pi)^3}{n_0^3 c} \frac{d_{\mathrm{eff}}^2 I_0 4\pi^2}{\Delta\omega^2 (2a_2^e - a_0^o)^2} , \qquad (7)$$

where $\Delta\omega$ is the cull spectral width and I_0 is the fundamental light intensity. d_{eff} is an effective nonlinear coefficient, $d_{\mathrm{eff}} = d_{ijk} \sin \theta_{2i} \cos \theta_{0j} \cos \theta_{0k}$, where θ_{0j} and θ_{0k} are the angles between j and k and $\mathbf{E}_0$, respectively. This expression holds, of course, only when $I_2/I_0 < 1$. The depletion of laser power due to production of second harmonic of comparable intensity has been neglected in Eq. (1). It is also clear from Eq. (5) that the quarter-power point occurs when $T = M$, ($S = S(2\pi M) = N\pi/\kappa$) for large values of M. Thus, for sufficiently long crystals, the second harmonic pulse width is proportional to M, i.e., to the crystal length. Numerical examples will be given below.

Computer plots of second harmonic power as a function of time are shown in Fig. 1. Note that the pulse becomes essentially square for large values of the crystal length. It is interesting to note that second harmonic generation in long crystals may be a practical way of generating a square light pulse of variable width. Figure 2 shows the peak power and pulse width as a function of crystal length. The predicted limiting behavior of these

functions is clearly evident. The transition region occurs near $M = 1$. This corresponds to the crystal length for which the rays at the edge of the spectrum suffer a 2π phase mismatch in ΔkL when the center of the spectrum is entirely phase matched.

The above analysis is of practical importance when 100 Å bandwidth at 1.06 μ in a glass:Nd laser is locked together. The crystal length for which $M = 1$ is calculated from $L(M = 1) \approx 2\pi c/\omega\Delta\omega(2a_2^e - a_0^o)$. $2a_2^e$ is computed at the phase-matching angle from

$$a_2^e(\theta_m) = \left(\frac{n_0^o}{n_2^o}\right)^3 a_2^o \cos^2\theta_m + \left(\frac{n_0^o}{n_2^e}\right)^3 a_2^e \sin^2\theta_m \qquad (8)$$

and refractive index data.[3] In $LiNbO_3$, $L(M = 1) \approx .06$ cm for a pulse of width 4×10^{-13} sec (100 Å bandwidth). The second harmonic pulse width will broaden when the crystal is significantly longer than this. Here the phase-matching angle is essentially 90° at room temperature; however, $L(M = 1)$ does not depend strongly on phase-matching angle in $LiNbO_3$, since $a_2^e \simeq a_0^o$ over a large temperature range. In KDP and ADP the corresponding crystal lengths are a hundred times longer, since $2a_2^e(\theta_m)$ and a_0^o are of the same order of magnitude. $L(M = 1) = 5$ cm in KDP and 3 cm in ADP for the above pulse.

The maximum power conversion ratio (7) is less in $LiNbO_3$ than in KDP and ADP, since the larger nonlinear coefficient in $LiNbO_3$ does not sufficiently

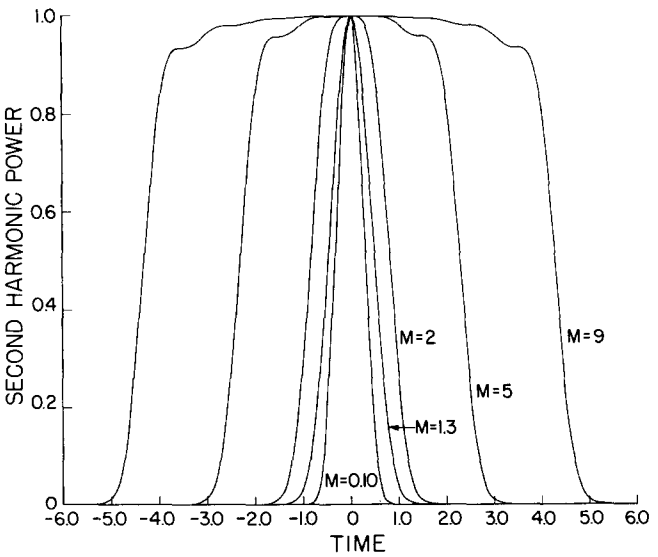

Fig. 1. Second harmonic power as a function of time for varying crystal lengths (M), normalized to the same peak power.

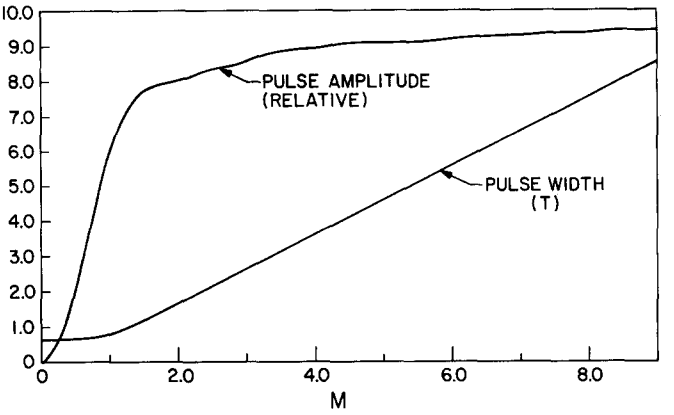

Fig. 2. Second harmonic peak power and pulse width as a function of crystal length (M).

compensate for its higher dispersion. The maximum power conversion ratio to second harmonic is $I_2/I_0 = 10^{-3}\ I_0$ in $LiNbO_3$, $10^{-2}\ I_0$ in ADP, and $4 \times 10^{-2}\ I_0$ in KDP, and $4 \times 10^{-2}\ I_0$ in KDP for a 100 Å bandwidth pulse. I_0 is the fundamental light intensity expressed in MW/cm². It should not be difficult, then, to convert a sizeable fraction of the incident light pulse to second harmonic in KDP and ADP, since these materials can easily support intensities the order of 100 MW/cm². In $LiNbO_3$, however, damage will probably appear in the crystals at the 1000 MW/cm² intensity level required to convert a large fraction of the fundamental to second harmonic.

This analysis indicates that for 4×10^{-13} sec light pulses, crystals of $LiNbO_3$ no thicker than half a millimeter must be used to avoid generating a second harmonic pulse which is substantially broader than the fundamental light pulse. Furthermore, it will probably not be possible to get large conversion ratios in $LiNbO_3$. In KDP and ADP, however, pulse-broadening and power-limiting do not seem to be serious problems in the generation of second harmonic. That the dispersion of crystals does not greatly alter the characteristics of the fundamental light pulses has already been shown.[4]

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