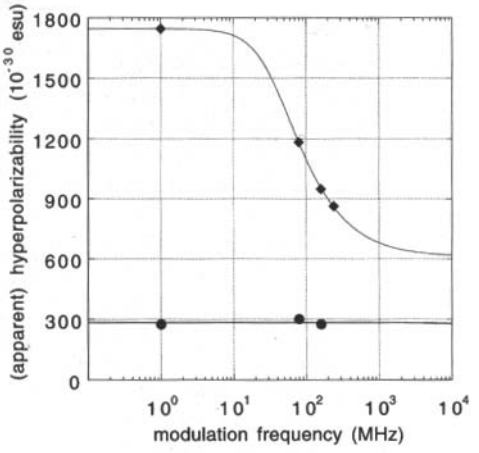

Clays Figure 1. First hyperpolarizability values for the free chromophore (round dots, 280 ± 20 × 10^{-30} esu) and for the same chromophore, embedded in an amylose matrix (squares). Note the demodulation for the embedded chromophore, and the resulting limiting, fluorescence-free, inherent β value (605 ± 10 × 10^{-30} esu).

The technique takes advantage of the inherent difference in temporal characteristics between the immediate (nonlinear) scattering and the time-delayed (multiphoton) fluorescence. We have implemented the Fourier transform in the frequency-domain of this principle. For a certain modulation frequency, the time-delayed fluorescence experiences a phase-shift and a demodulation (reduction in amplitude). The higher the modulation frequency, the larger the phase-shift (90° maximum) and the smaller the amplitude of the fluorescence. The immediate scattering, however, does not experience any shift, nor a reduction in amplitude. For high enough modulation frequencies, the only cause of signal is hyper-Rayleigh scattering, since the amplitude of the fluorescence is completely "demodulated" (essentially zero, exactly zero in the limit of infinitely high frequency).

The high modulation frequencies necessary for demodulation are obtained as the harmonics of a mode-locked Ti:sapphire laser producing 100 fs pulses at 80 MHz. The femtoseconde pulse also ensures high enough peak-power to observe an incoherent second-order nonlinear optical signal.

The experimental setup, with phase-locked signal generators for cross-correlation, was tested with a well-characterized non-fluorescent octopolar and ionic chromophore (crystal violet, CV), with and without a centrosymmetric fluorophore with known fluorescence lifetime (9, 10-diphenylanthracene, DPA).[1] The β values for CV without DPA are constant for all modulation frequencies (DC, fundamental, second-, and third-harmonic), as expected for the nonfluorescent CV. The apparent β values for CV when DPA is added are decreasing functions of the modulation frequencies. The limiting value at the third harmonic is identical to the fluorescence-free value. This demonstrates the capability of high-frequency-demodulation for fluorescence suppression.

This new technique is applied for the comparative study of an ionic chromophore, free in solution (non-fluorescent) or embedded in an amylose helix (restriction-induced fluorescence).[2] Figure 1 shows the modulation frequency dependence for the free and embedded chromophore. We find that the β value for the complex is twice that for the free dye, confirming amylose inclusion as a viable strategy in second-order nonlinear optics, not only at the bulk, but also at the molecular level.

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Backward Second-harmonic Generation in Periodically-poled $LiNbO_3$

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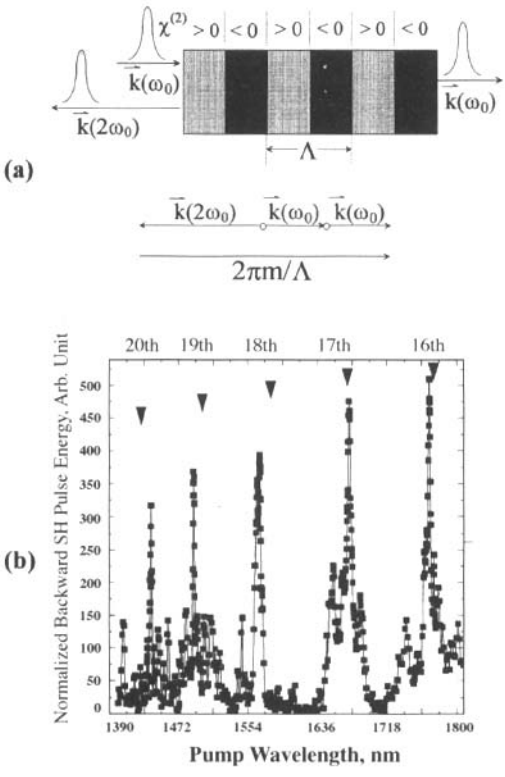
In the nonlinear optical processes that generate second-harmonic waves, some of the waves propagate in the direction opposite to the fundamental waves and are thus known as backward second-harmonic (BSH) waves. Because of the extremely large phase-mismatch that occurs in such a process, the BSH effect is negligible. Conventional phase-matching techniques using either angle or temperature tuning cannot compensate for large phase-mismatching. Thus, the backward process has been virtually ignored by most of the nonlinear optics community. While there were some early theoretical studies,[1] it is only recently that the backward parametric processes have been analyzed theoretically in detail.[2] In general, the backward nonlinear processes are important since they can lead to mirrorless optical parametric oscillators (OPOs) and other novel effects.[2] Meanwhile, recent progress in a quasi-phase matching (QPM) technique[3] has proven that QPM is a powerful technique that allows a large degree of phase-matching capability.[4] Using such a technique, we have experimentally demonstrated, for the first time, the BSH generation in QPM $LiNbO_3$.[5]

For BSH generation, the phase-matching condition is $2k_\omega + k_{2\omega} - k_g = 0$, where k_ω and $k_{2\omega}$ are the propagation constants for fundamental and second-harmonic waves in the medium, respectively; and k_g is defined as $k_g = 2\pi m/\Lambda$ with m as an integer and Λ is the grating period of the domains. For $LiNbO_3$, the grating period has to be approximately 0.178 μm to achieve first-order phase matching for fundamental wavelength of 1.55 μm. Because of the technical difficulty of fabricating a submicron grating period, the first-order phase-matching could not be observed. Thus, we used higher-order gratings ($m > 1$) to observe BSH.

We used a 6-mm long z-cut $LiNbO_3$ QPM sample with a 3.3 μm period. The experiments were performed with both femtosecond and nanosecond tunable optical parametric oscillators or amplifiers. Figure 1 (page 30) shows the output BSH energy per pulse vs. fundamental wavelength, where 20^{th} to 16^{th} order phase-matching at the fundamental wavelengths ranging from 1440–1760 nm can be seen. Arrows indicate phase-matching wavelengths predicted theoretically, which are in good agreement with the experimental results. We determined the maximum conversion efficiency to be approximately 0.02% and 0.3% using 200-fs and 9-ns pulses, respectively. Because the fundamental and backward second-harmonic beams



Gu Figure 1. (a) Schematic of a backward SHG configuration. (b) Wavelength scan showing phase-matching locations for backward SHG. Arrows indicate theoretically-predicted phase-matching wavelengths.

propagate in opposite directions, the effective interaction length for the process is determined by the pulse duration of the pump. Therefore, for the ultrashort laser pulse, second-harmonic conversion efficiency is reduced significantly.

We recently observed BSH generation in a periodically-segmented ion-exchanged potassium titanyl phosphate waveguide. Due to a much shorter period of index gratings, we achieved QPM at much lower orders. Most recently we observed BSH generation in periodically-poled potassium titanyl phosphate waveguide. Because of the presence of the waveguide, we achieved a much higher conversion efficiency by using much lower pump energy per pulse.

In conclusion, we have demonstrated, for the first time to our knowledge, BSH generation in periodically-poled bulk $LiNbO_3$, which we believe is a significant first step toward realizing mirrorless OPOs and other unique backward parametric configurations.

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Extremely Nonlinear Methyl-red Doped Nematic Liquid Crystal Film

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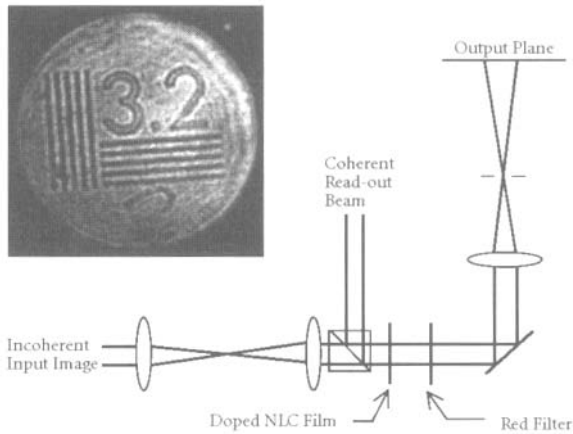
To date, liquid crystals remain an important electro-optics material because of their broadband birefringence. They possess a sizable birefringence, $\Delta n \sim 0.3$, spanning the visible to IR spectral regime.[1] Because of their easy susceptibility to optical fields, they are also well known for their nonlinear optical properties associated with laser induced director axis reorientation effects.[2]

A recent discovery[3] has lead to dramatic improvement in both the magnitude of the nonlinear index coefficient and the conditions under which the effect can be generated. A typical nematic liquid crystal, Pentyl Cyano Biphenyl [5CB] with traces of the laser dye methyl-red dissolved in it at a concentration of about 0.5% was used and it was shown that these films are extremely photosensitive and easily yield photocharges and photovoltaic effects from the photo-excited dye dopant. These photo induced DC space charge fields cause the highly birefringent liquid crystalline axis to reorient, giving rise to a large refractive index change for extraordinarily polarized light. In general, the response time of the nematic liquid crystal director axis reorientation is on the order of milliseconds.[1]

In a grating diffraction experiment, a nonlinear index change coefficient $n_2 = 6$ cm^2/W was observed.[3] Similar values of n_2 are obtained at other Argon laser lines within the absorption band of the Methyl-red dye. Recent studies show that the effect can be enhanced by an AC field, with an effective $n_2 > 10$ cm^2/W, equivalent to a nonlinear third-order susceptibility $\chi^{(3)} > 300$ esu. This is perhaps the most nonlinear optical material known to date.

Such extremely nonlinear film allows the observation of various nonlinear optical processes at unprecedented low optical power. In particular, self-defocusing and optical limiting of cw laser at nanowatt threshold was demonstrated,[4] as was incoherent-coherent image conversion at μW/cm^2 light intensity level. Figure 1 depicts the setup used to demonstrate incoherent-coherent image conversion. A slight modification of the setup could also be used for visible to IR [or other wavelength] image conversion applications. The polarized green image bearing beam creates a spatially distributed phase shift on the NLC film, which is read by a coherent (red) He-Ne laser. Visible images can be generated with an input incoherent light intensity as low as 90 μW/cm^2. This is comparable to the in-



Khoo Figure 1. Experimental setup for incoherent to coherent image conversion. Inset is a photograph of the coherent image at He-Ne laser wavelength.