Nonlinear Spectrometer Using a White-Light Continuum Z scan

Mihaela Balu, Joel Hales, David J. Hagan and Eric W. Van Stryland

Applications such as three-dimensional fluorescence imaging and microfabrication using nonlinear absorption, and all-optical switching exploiting optically induced changes in refractive index, require nonlinear optical properties of materials to be known over broad wavelength ranges. There are different methods for measuring the spectral dependencies of these optical nonlinearities, but they are often time-consuming.

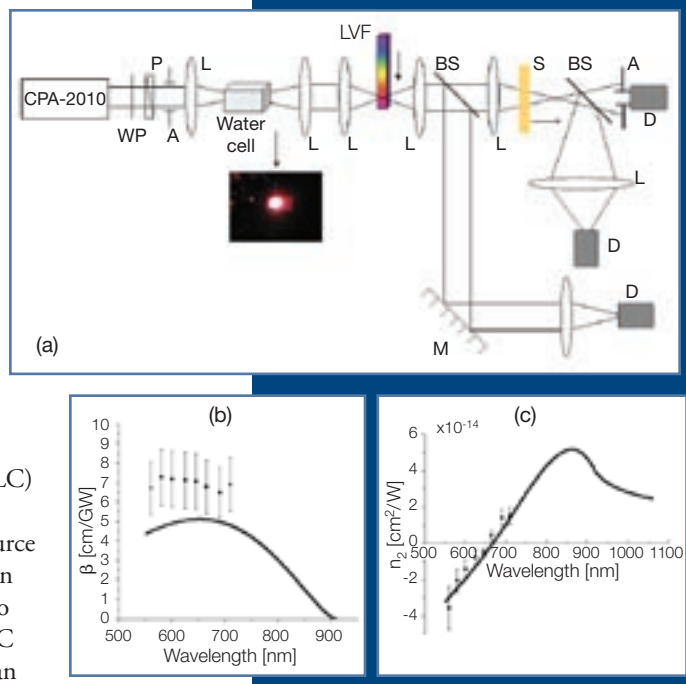
Among the methods, the single-wavelength *Z* scan is a simple and sensitive technique for measuring nonlinear absorption (NLA) and nonlinear refraction (NLR).[1] With a single wavelength, the method provides information about the frequency-degenerate NLA and NLR. In this method, the transmittance of a beam, usually of Gaussian spatial profile, focused through a sample is measured as a function of the sample position *Z* with respect to the beam waist. If all the energy transmitted by the sample is collected, the sign and the magnitude of the NLA are extracted from the measurement.

The transmission measured through an aperture placed in the beam in front of the detector provides the sign and the magnitude of the NLR once the NLA has been obtained. This is a rapid method to determine nonlinear coefficients for a particular wavelength, but obtaining a broad spectral dependence of the nonlinearities requires considerable time for wavelength tuning, beam characterization and realignment.

We introduce a faster technique for measuring the spectral dependence of NLA and NLR using a broadband white-light continuum (WLC) source in place of the single-wavelength source conventionally used in the *Z* scan; we refer to this method as a WLC *Z* scan.[2] The WLC can be generated by several methods.

Here, we use femtosecond, regeneratively amplified, Ti:sapphire laser pulses focused into a cell filled with deionized water. We spectrally separate the WLC wavelengths using a commercially available linear variable frequency (LVF) bandpass thin-film filter. This acts as a bandpass filter with a spatially varying center wavelength. The filter needs to be used prior to the sample to prevent frequency nondegenerate nonlinearities from occurring.[3] To measure the NLA and the NLR, we perform *Z* scans in which the LVF is translated through the beam for each *Z* position. The experimental setup is shown in the figure, part (a). We use computer-controlled motors attached to both stages for the LVF and the sample holder. For a 1 kHz laser pulse-repetition rate, averaging 500 pulses per data point, a single experiment takes approximately 8 min to provide the spectrum of NLA and the dispersion of NLR from 560 to 710 nm.

To confirm the validity of this method, we test this new procedure on a well-characterized semiconductor—ZnSe.[1,4,5] The values of the two-photon absorption coefficient β and its associated NLR index n_2, obtained from fits (see Ref. 1) at different wavelengths, are presented in (b) and (c) along with the theoretical predictions of Refs. 4 and 5. As predicted by the theory in Ref. 5, n_2 changes sign around 0.7 of the bandgap energy. To our knowledge, this is the first time the sign change of n_2 is demonstrated by tuning the wavelength. The agreement between experiment and theory confirms the value of this method for obtaining degenerate nonlinearities. ◭



(a) WLC *Z*-scan experimental setup. CPA-2010-Clark MXR femtosecond laser source: WP, waveplate; P, polarizer; L, lens; LVF, linear variable filter from Ocean Optics; M, mirror; BS, beamsplitter; S, sample; A, aperture; D, detector. (b) 2PA coefficient β and (c) nonlinear refractive index n_2, obtained from theory (solid lines) and from the fits of the experimental data (dots).

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Optical-to-Terahertz Conversion for Plasmon–Polariton Surface Spectroscopy

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Surface plasmon polaritons (SPPs) guided by metal surfaces are widely used for optical spectroscopy. In the visible and near-infrared ranges, SPPs of metal surfaces are well localized near the surface and extend only a fraction of wavelength from it (roughly 640 nm for Au at the wavelength 800 nm).

This localization provides high sensitivity of the SPP propagation wavenumber to the properties of very thin layers deposited on the metal surface. Thus, angular resolution of SPP excitation resonance allows one to determine the concentration of various molecules inside the layers and to perform time-dependent monitoring of biospecific reactions.[1]

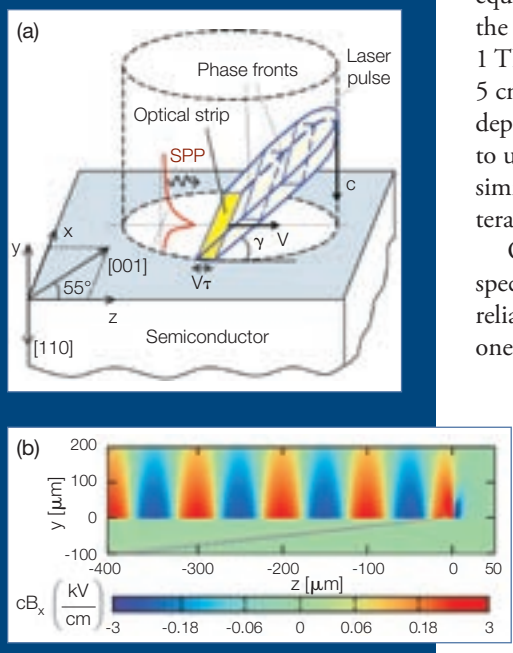
Many molecules, including key biological constituents such as proteins, ribonucleic acids and deoxyribonucleic acids, have resonances in the terahertz range. Therefore, there is keen interest in performing terahertz spectroscopy, in particular for the identification and characterization of biomolecules. However, SPPs guided by metals become inadequate for terahertz spectroscopy, because the SPPs lose their localization (e.g., a 1 THz SPP extends from Au surface for 5 cm), and thus their sensitivity to surface deposits deteriorates. It is more natural to use SPPs on semiconductors that have similar electromagnetic properties in the terahertz range to metals in the optical.

One of the challenges in terahertz spectroscopy is the need for small and reliable terahertz sources. Furthermore, one needs to couple the bulk radiation to SPPs. We proposed a scheme that can potentially solve these two problems and allows a direct excitation of terahertz SPPs at semiconductor surfaces using optical pulses.[2]

This scheme is based on two ideas and is shown in the figure, part (a). First, we use an optical pulse with a tilted intensity front to create an optical spot on the surface that moves slower than the speed of light.[3] Second, optical rectification in the semiconductor creates a pulse of nonlinear polarization that essentially follows the envelope of the spot. Because of its slow velocity, the pulse of moving nonlinear polarization will efficiently excite the SPP with the same phase velocity. The frequency of the excited SPP lies in the terahertz range and can be controlled by the doping concentration and the tilt angle of the optical pulse. The excitation efficiency depends on the crystallographic orientation of the surface, and the optimal orientation for GaAs is shown in (a).

We analyzed the kinematic properties of the excited SPP and its excitation efficiencies.[2] A typical distribution of the magnetic field of the terahertz radiation excited behind the moving spot is shown in (b). The pronounced oscillations at $y > 0$ are the SPP that is localized within about 100 μm in the air. Its penetration into the semiconductor is small, because the SPP frequency (3 THz) is chosen to be significantly smaller than the plasma frequency (7.7 THz) to avoid large absorption losses. The faster field oscillations at $y < 0$ describe the Cherenkov radiation of bulk waves propagating into the semiconductor.

In conclusion, we proposed and analyzed a phase-matched scheme to directly convert optical energy into terahertz SPPs that propagate along semiconductor surfaces. The scheme can be used to perform surface terahertz spectroscopy. ◭



(a) Excitation scheme for terahertz SPP. A normally incident laser pulse with duration τ and a tilted intensity front produces a striplike optical spot that moves with velocity $V = c \cot\gamma$ and has a width $V\tau$. (b) Snapshot of the magnetic field $B_x(y, z)$ for GaAs, $\tau = 17$ fs, $\gamma = 45.2°$, and peak laser intensity 2 GW/cm^2.

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Two-Beam Second-Harmonic Generation

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Second-harmonic generation (SHG) is widely used for probing planar surfaces noninvasively. The reason is that SHG is forbidden in the bulk of a centrosymmetric medium but allowed at its surface, where the inversion symmetry is broken. This rule, however, is strictly true only for electric–dipole interactions. For example, a bulk SHG signal can arise through quadrupole interactions, often leading to an unwanted background in planar-surface studies.

On the other hand, quadrupole contributions also play an important role in the SHG response of the surface. When properly exploited, such contributions enable surface studies in material systems that cannot be investigated by traditional SHG techniques.

One such system is composites of silicon nanocrystals embedded in fused silica. Their sharply curved nanointerfaces give rise to unusual bonding structures that are believed to be responsible for the unusually efficient light emission observed from this silicon system. However, conventional SHG from such closed, randomly oriented interfaces is extremely inefficient, because they respond centrosymmetrically at optical wavelengths. Consequently, nanointerfacial SHG becomes quadrupolar.

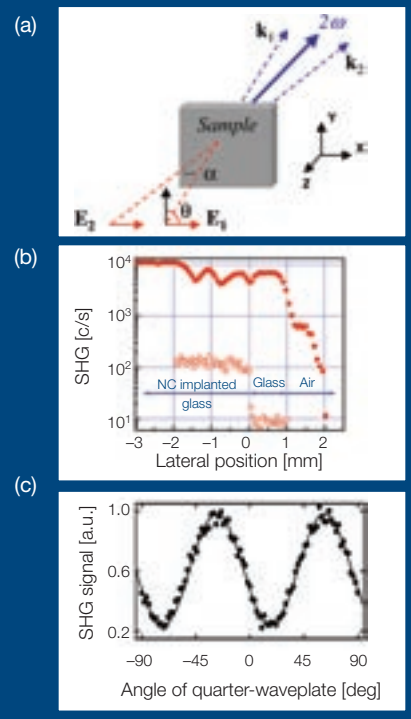
Recently, we showed that such SHG is enhanced dramatically by using two intersecting and orthogonally polarized input beams [see part (a) of the figure], whereas dipolar SHG from the planar substrate surfaces is suppressed [part (b)].[1,2] Background SHG from the bulk substrate is easily separated from the nanocrystal signal, which is robust, sensitive to nanointerface chemistry and useful for spectroscopy of the poorly understood nanointerface structure.[1]

The two-beam SHG configuration also opens the door to new ways of addressing another long-standing problem in surface nonlinear optics: separation of surface and bulk contributions. We showed that one can distinguish part of the bulk response of isotropic materials from effective surface contributions by inspecting the dependence of a two-beam SHG signal on the polarization of one input beam [see part (c) of the figure].[3,4]

The dependence is dictated by the symmetry of the material and is nearly independent of its linear optical properties. Unlike methods relying on coherence length, the new method does not require a comparison of signals measured in different geometries. The measured bulk component can then be used to estimate the strength of surfacelike, indistinguishable bulk contributions.[3,4]

Intriguingly, we find that, even when the nonlinear response is of bulk origin, the strongest signals are obtained when the input beams intersect at the surface of the nonlinear medium.[2,5] In contrast, the signal is strongly suppressed when the beams cross deep in the bulk of the material. We showed that the effect can be generalized to all phase-mismatched nonlinear optical processes in which the strength of the interaction is turned on and off gradually.[5]

Our results emphasize the importance of detailed polarization measurements in establishing the origin of the nonlinear response of surfaces and interfaces. On the other hand, the results also indicate that two-beam SHG could complement third-harmonic microscopy as a probe of highly centrosymmetric biological samples by enabling use of shorter wavelength sources for which samples are transparent to second-harmonic, but not third-harmonic, light.[2]



(a) Two-beam SHG geometry. (b) Lateral scan of single-beam SHG (circles) and two-beam SHG (solid dots) across the boundary between Si nanocrystals implanted and unimplanted fused silica sample. (c) Dependence of the transmitted two-beam SHG signal from an air-glass interface on the polarization of one input beam.

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Photorefractive Polymers with Superior Performance

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Photorefractive polymers offer several advantages over their inorganic counterparts, including their flexibility, large area, wavelength tunability, ease of fabrication and low cost.[1] The photorefractive effect refers to the modulation of a refractive index owing to the spatial distribution of charges and the electro-optic effect.

Optical information can be recorded, erased and rewritten using a low-power laser beam. Currently, photorefractive materials are potential candidates to explore applications in beam cleanup and amplification, real-time image processing, optical phase conjugation, pattern recognition, 3D displays and novelty filters.

A drawback has been that high-voltage (5–10 kV) poling is required for the operation of photorefractive polymeric devices. We have now developed devices that can be poled at voltages that are an order of magnitude lower by carefully engineering the material composition using high-performance nonlinear chromophores for thinner devices.

Four-wave-mixing experiments were performed to investigate the photorefractive characteristics of the devices. The measurements were done at 633 nm using a standard cw laser with a total writing power of 1 W/cm^2. For a thickness of 20 μm, the device showed more than 90 percent diffraction efficiency with a dominant fast response time of 27 ms at an applied voltage of approximately 1 kV. A useful diffraction efficiency of roughly 10 percent can be observed, even at a voltage as little as 450 V. This demonstrates a major step forward in the possible applications of photorefractive devices. Part (a) in the figure shows diffraction efficiency as a function of poling voltage.

We have also been able to extend the operating wavelength to the near infrared. By using a new sensitizer dye, we have demonstrated a first-time proof of photorefractivity at 975 nm in an all-organic composite with 90 percent diffraction efficiency and video-rate response time using a low-power cw laser.

This is a significant advance in the development of an all-organic photorefractive device for near-infrared imaging and optical communication.[2] Furthermore, we made devices to operate at 1550 nm, which is suitable for fiber-optic applications.

Using two-photon absorption (TPA) processes, we demonstrated more than 40 percent diffraction efficiency, maintaining near-video-rate response time. This approach provides the inherent advantage of nondestructive readout with cw laser light. The instantaneous optical powers reached by cw reading beams are much less than those of the pulsed writing beams used to initiate charge generation through TPA.[3]

As an application of these devices, we demonstrated beam cleanup and aberration correction in a free-space communication application. We demonstrated high-quality aberration corrections using an oil-filled phase plate, which generates atmospheric-like wavefront aberrations.[4] Part (b) of the figure shows the result of correction of a severely aberrated image by our photorefractive polymers.

In summary, we have successfully reduced the applied poling voltage required without sacrificing the diffraction efficiency and response time in photorefractive polymers and have demonstrated beam cleanup applications. The operating wavelength was also extended from the visible to 1550 nm. ◭

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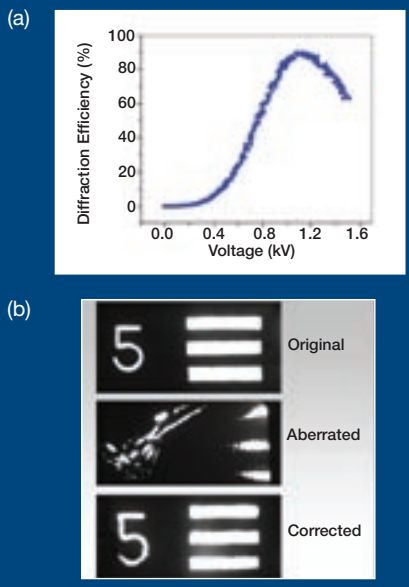

(a) Diffraction efficiency as a function of poling voltage. (b) Correction of a severely aberated image by a photo-refractive polymer.

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