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A Sensitive Scheme to Observe Weak Photo-Refraction Effects in Some Nonlinear Optical Crystals Pumped by Ultrashort Optical Pulses *

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We present a sensitive scheme, for the first time to our knowledge, to observe photo-refraction (PR) effects in some nonlinear optical crystals, e.g. β-BBO, LBO and BIBO, pumped by an intense ultrashort laser pulse chain. These quite weak effects are "amplified" by sensitive cw intracavity loss modulation. Our results show that they are repeatable and are dependent on pumping power and wavelength, and their response time ranges from tens of seconds to several minutes. The recorded dynamical transitions between the self-focusing to the self-defocusing (or vice versa) induced by the PR effect may be critically important for us to give more insight into the stability of some cascade nonlinear frequency conversions, e.g. multi-stage optical parametric amplifiers.

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The PR effect has been found in many dielectric or semiconductor materials (e.g. $LiNbO_3$, $BaTiO_3$ and $KNbO_3$ etc.) in the past few decades.[1−3] One of its important properties is the dynamical refractive index variation transitions of optical materials.[2,4] Many interesting applications based on photorefractive (PR) effects, including optical amplification, laser beam clean-up and reconfigurable waveguides and so on, have been reported.[5−7] However, the PR effect also has some deleterious influence on the performances of some devices, such as both the phase- and group-velocity-matched scheme of ultrashort pulse SHG and the attosecond control of optical waveforms.[8,9]

The dynamical nonlinear lens due to PR effect may seriously affect the stability of multi-stage optical parametric chirped-pulse amplification (OPCPA) which has been increasingly gaining attention since it offers a particularly promising route toward compact ultrashort ultrahigh-peak-power laser systems.[10] In this kind of system, even if the PR effects in the used nonlinear optical crystals are so weak that their influences of the PR effect seem usually negligible, the accumulation of the PR effect from the cascade nonlinear frequency converters may be very important. The dynamical nonlinear lens caused by the PR effect can change not only the spatial properties of both the pumping and the signal but also the phase-matched condition between the pumping and the signal, thus degrading the stability of the system. So far there have been no detailed experimental reports on the PR phenomena under ultrashort pulse excitation in the nonlinear optical crystals, such as β-BBO, LBO and BIBO, which are typically used in OPCPA. In this Letter, we present, for the first time to our knowledge, the observation of the PR effects in a β-BBO, LBO and BIBO crystals due to multi-photon excitation with intense ultrashort pulses by use of an intracavity method to "magnify" the PR effect. Our results may help us to gain more insight into the roles of PR effects in OPCPA.

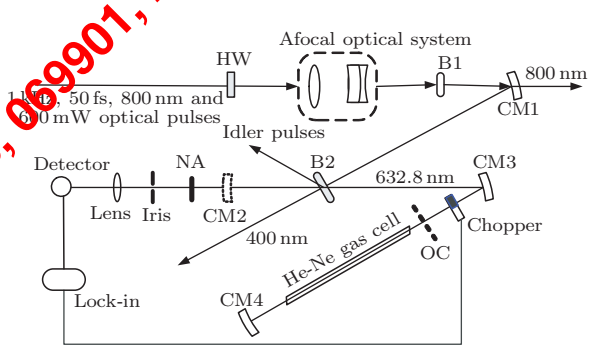


Fig. 1. Experimental setup. HW: half wave plate at 800 nm; CM1: concave mirror with 0° HR at 400 nm and AR at 800 nm; CM2, CM3 and CM4: mirrors with 0° HR at 632.8 nm; OC: output coupler of the He-Ne laser; NA: variable neutral attenuator; β-BBO crystal; B2: measured sample.

As we know, the output power of a laser oscillator is very sensitive to its configuration or intracavity loss, thus a small variation of the cavity configuration due to the week dynamical lens of some optical component inside the cavity will obviously modulate the intracavity loss and thus the output power. Figure 1 shows the schematic diagram of our setup for the observation of the PR effects of some nonlinear crystals pumped by intense 400 nm ultrashort light

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pulses which are the second harmonic (SH) of 50 fs, 600 mW, 1-kHz and 800 nm pulses from a Ti: sapphire regenerative amplifier. For the sake of high SH conversion efficiency, an afocal optical system is used to reduce the beam size from 12.0 to 4.0 mm. A half-wave plate (HW) is aligned so that the SH pulse chain is polarized horizontally. The material dispersion induced by the waveplate and the fused silica lens is pre-compensated by adjusting the grating pulse compressor of the Ti:sapphire laser system so that about 23% SH conversion efficiency is achieved with a 29.2°-cut 0.2-mm-thick type-I phase-matched β-BBO (B1) crystal. The 400 nm beam is reflected and focused by a concave dichronic mirror (CM1) to the measured sample B2, while the fundamental beam is transmitted through the CM1.

The He-Ne laser oscillator used here consists of a tube filled with gain gases, three concave mirrors coated in HR at 632.8 nm (CM2, CM3 and CM4). The curvature radii are 0.5 m, 0.5 m and 5.0 m, respectively, while the separation between CM2 and CM3, or CM3 and CM4 is 0.75 m or 1.825 m. The B2 crystal is situated at the beam waist position between CM2 and CM3 for maximal cw laser intensity. The pumping pulses and the cw beams are overlapped inside the crystal B2. After going through a variable neutral attenuator and an iris, the leaking beam from CM2 is focused into a detector which is connected to a lock-in amplifier.

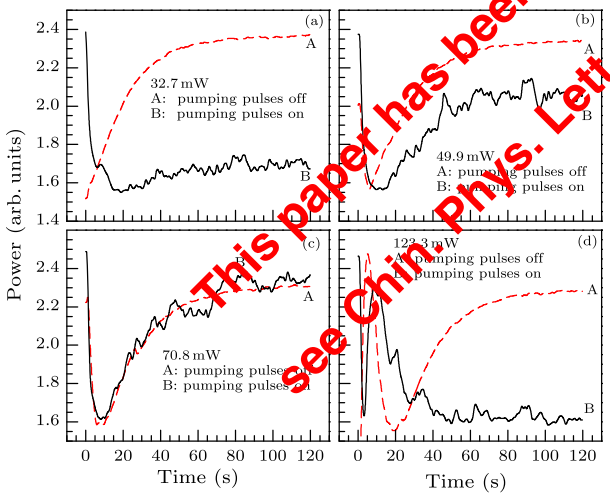


Fig. 2. The cw He-Ne laser power vs time delay after switching on (solid curves) or off (dashed curves) the broadband 400 nm pumping laser beam onto the β-BBO crystal with different pumping power.

First, the measured sample B2 is a 29.2°-cut 2-mm-thick type-I phase-matched β-BBO crystal. In order to reach a good spatial overlap between the pumping pulses and the cw He-Ne laser, we optimize the He-Ne laser for maximal output power and align the orientation of the B2 crystal to obtain the strong idler pulses around the wavelength of 1064 nm due to the phase-matching for optical parametric amplification. Once the broadband 400 nm pumping beam switches on, the 632.8 nm laser power becomes fluctuating. The fluctuation lasts from tens of seconds to several minutes, much longer than not only the interaction time of the two beams but also the interval of the pumping pulse train, and depends on the intensity of the pumping beam. Figure 2 presents the variation of the cw He-Ne laser power vs time delay after switching on (solid curves) or off (dashed curves) the broadband 400 nm pumping laser beam onto the B2 crystal with the different pumping powers. At a low level of pumping power (32.7 mW), as shown in Fig. 2(a), switching on the pumping beam makes the cw power decay monotonously with delay time. Surprisingly, it takes about 50 s for the cw laser power to reach a stable level without the pumping beam. Figure 2(b) shows some oscillating structures appear both in the solid and in the dashed curves for the pumping power up to 49.9 mW. From Figs. 2(b)–2(d), one can see that the higher the pumping power, the more the oscillating structures within the recording time of about 130 s, thereby the smaller the oscillating periods. Noticeably, the initiative value of the measured power just after switching off pumping pulses is approximately equal to those after switching on pumping pulses for a long enough time, so it is easy to understand that the tendencies of dashed curves also show a strong dependence on the pumping power before switching off. The cw power can recover after blocking the pumping pulses for a long enough time, which excludes that the observed phenomena are from the gradually laser induced damage on the BBO crystal.

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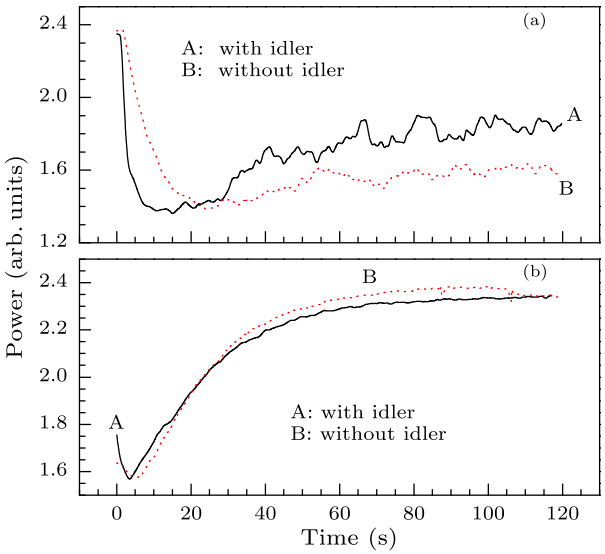


Fig. 3. Measured cw laser power vs time delay after switching on (a) and off (b) the broadband 400 nm, 38.0 mW pumping laser beam onto the β-BBO crystal in the presence/absence of the phase-matched condition.

The data in Fig. 2 were recorded under the type-I

phase-matched condition for optical parametric amplification. One can rotate the B2 crystal slightly so that the type-I phase-matched condition is not met anymore thus without appearance of the idler pulses. Figure 3 makes a comparison of the measured curves of the cw laser power vs time delay with and without the appearance of the idler pulses pumped by 38.0 mW pulse chain. In Fig. 3, one can see that the performance of the cw power under the phase-matched condition is similar to that without the phase-matched condition for both on- and off-states of pumping pulses. However, Fig. 3(a) shows that for the long time delay after switching on the pumping beam, the phase-matching results in higher output power of the cw beam, which is reasonable because the appearance of the idler beam means the occurrence of parametric gain inside the cavity. From Fig. 3, we believe that the phase-matched condition is not indispensable for the occurrence of the observed phenomena.

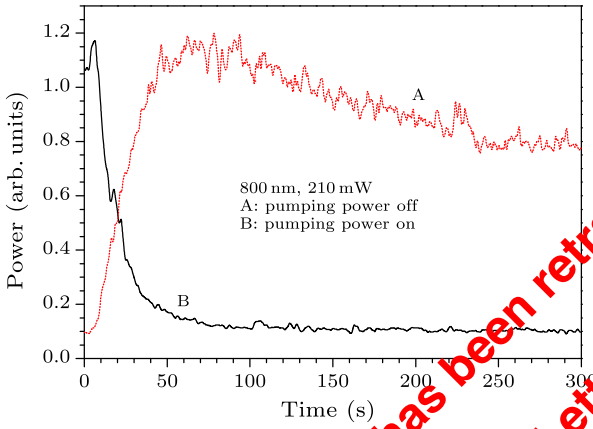


Fig. 4. The cw He-Ne laser power vs time delay after switching on (solid curve) and off (dashed curve) the incident 210 mW, 800 nm pulsed laser beam to the B2 crystal.

If using 800 nm pulses instead of 400 nm pulses as a pumping beam, our measurements show even when the pumping power rises to more than 200 mW, there is no oscillating structure in the curves of the output power vs time delay after switching on/off pumping pulses (see Fig. 4), which are similar to those in Fig. 2(a). However, the dynamical response time of the cw power on the pumping beam is longer under the radiation of 800 nm pulses than under the radiation of 400 nm pulses. These results imply that the modulation is more sensitive to the 400 nm laser pulses than to the 800 nm laser pulses.

The observed phenomena can also happen if using BIBO and LBO crystals instead of BBO crystal as measured sample B2, but fail in optical glass, such as BK7 and fused silica, which seems that the phenomena only occur when some materials with nonzero EO coefficients are used. As shown in Fig. 5, according to our experiments, for BIBO crystal, the cw power spends about 9 s after switching on the 40 mW pumping beam, and 7 s after switching off the pumping beam to reach a steady state. Similarly, the corresponding time delays for LBO are 5 s and 10 s, respectively.

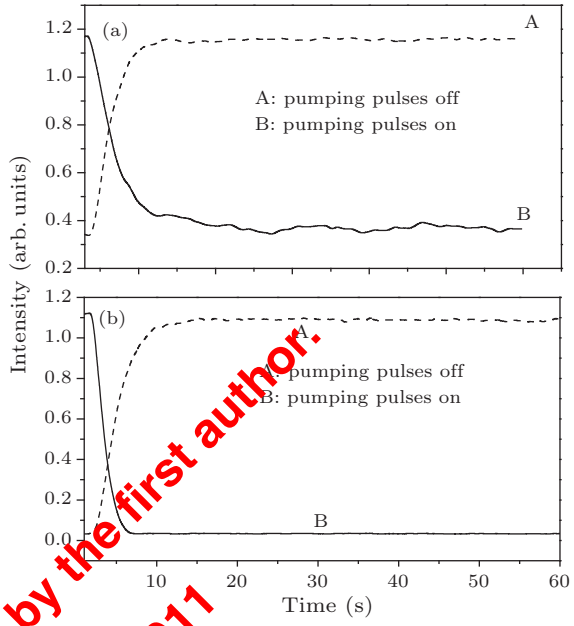


Fig. 5. The cw He-Ne laser power vs time delay after switching the broadband 400 nm, 40 mW pumping pulses on (solid curve) or off (dashed curve) to the BIBO (a) and LBO (b) crystals.

We can observe easily the slow variation phenomena by loss modulation of He-Ne intracavity, however the complicated intracavity losses, such as diffraction loss and Fresnel loss, make it difficult to directly characterize the phenomena. We compare the results measured by using the extracavity and intracavity methods under the same pumped condition. It is noticeable that the cw laser has quite different tendencies of time-dependent modulations from the intracavity probe under the same pumping and detection conditions. This discrepancy implies that the main contribution to the slow variation of the cw laser comes from the non-uniform spatial variation of the refractive index (dynamical lens effect) rather than the variation of Fresnel loss of the B2 crystal induced by 400 nm pulses laser. The non-uniform spatial variation of the refractive index changes the spatial focusing or defocusing of the cw laser beam thus changes laser intensity.

The dynamical observed lens-alike phenomena occur typically in some optical materials with large enough electro-optic coefficients. They have slow response times (tens of seconds or several minutes) with strong dependences on power, wavelength, and spatial intensity profile of the laser pulses, which are consistent with the characteristics of PR effects. Usually, the conventional PR effect[3] cannot be observed in these optical nonlinear crystals because of their large band gaps, e.g. about 6.43 eV[11] for BBO crystal.

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However, using intense ultrashort laser pulses as a pumping source allows us to observe the interband PR effect through two-photon[12] or multi-photon absorption. The intrinsic physics of the observed lens-like oscillating structures in the nonlinear optical crystals can be qualitatively understood as follows.[13] Immediately after the ultrafast pulses are turned on, the ultrafast laser pulse chain generates charge carriers through multiphoton excitation, and the generated photo-charge carriers start to redistribute themselves by drift and diffusion, thereby changing the electric field in the illuminated region of the crystal from the amplitude of the externally applied field (if any) and resulting in the initial buildup of the induced nonlinear lens. The dynamical lens brings about the transient focusing of the probing beam, which depends on the electric field and the electro-optic coefficient. At times exceeding several characteristic relaxation times, the induced lens starts to relax to its steady-state value, which is determined by the ratio of conductivities in the illuminated and dark regions of the crystal which depends strongly on the power and wavelength of the pulsed laser.

In summary, we have presented a sensitive detection for the very weak interband photorefractive effect in some nonlinear optical crystals (β-BBO, LBO and BIBO) due to multiphoton excitation by intense ultrashort laser pulses, which is based on intracavity loss modulation. As is expected, the reproducible effect has a strong dependence on the power, wavelength, and spatial intensity profile of the intense laser pulses and the electro-optic coefficient of the optical materials but without the requirement of phase-matching. For some optical applications, e.g. used as nonlinear gain materials for optical parametric chirped pulse amplification, this kind of interband PR effect will degrade the output performance, especially in multistage optical parametric amplification systems. The strong dependence on the wavelength of the pulsed laser beam implies that it is helpful for promoting the stability of the optical parametric amplification to choose pumping pulse with long wavelength or the nonlinear crystal with large energy band to minimize the interband PR effect. In spite of the sensitivity of the intracavity, it is difficult to characterize the PR effect quantitatively due to the diffraction loss of the cw He-Ne cavity in our setup. However, we are taking some efforts to simplify the design of the cw cavity thus to detect quantitatively the dynamical lens by the intracavity method.

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