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Compact optical setup for forward-box degenerate four-wave mixing measurement

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We have devised compact optics for the forward-box configuration in degenerate four-wave mixing spectroscopy. The core of the optical setup is the successive use of a pair of polarization-based beam splitters that divide the laser beam into four parallel beams. This setup is compact, easy to assemble, and adaptable for laser light over a wide spectral range. Selection of the combination of beam polarization produces two types of transient grating: either a polarization grating or an intensity grating. © *1999 American Institute of Physics.* [S0034-6748(99)02711-2]

Degenerate four-wave mixing (DFWM) is widely used for applications such as measuring dynamic photoexcitation and determining its rate constant as an essential laser spectroscopy technique.[1,2] The orthodox optical setup for DFWM is a forward-box configuration.[3]

The usual optics of the forward-box configuration consists of multiple beam splitters and mirrors separating a laser beam wave front into three waves. After being delayed appropriately, these waves are combined at one spot. Use of a partially reflecting mirror as a beam splitter causes power loss in rear surface reflection and often creates stray light. Many optical components are required to realize the forward-box configuration and optical components are very difficult to coordinate because optics must be three dimensional and are complicated. If the optical setup becomes larger, beam quality deteriorates over long distances and the DFWM signal becomes difficult to find. Setting up a mask filter to eliminate stray light requires observation of the beam path in the dark from the sample to the sensor. Tunable lasers with ultrashort pulses are currently used in laser spectroscopy.[4] As the laser pulse duration becomes shorter, beam energy density becomes higher and partially reflecting mirrors adaptable to such pulses become extremely difficult to fabricate over a wide spectral range.

We devised a compact optical DFWM setup (Fig. 1) whose major innovation is the use of calcite crystals as displacers to divide the laser beam into two parallel beams that are mutually cross polarized. The length of the calcite crystal determines the distance between displaced beams. Because calcite dispersion is small, the distance changes slightly over a wide spectral range. This change does not affect measurement, because the size of holes in the mask is slightly smaller than that of the laser beams. A calcite crystal 30 mm long typically separates a beam to a distance of 3.3 mm and is transparent from 350 to 2300 nm and semitransparent up to 215 nm. The amplitude ratio of horizontal and vertical polarization components of input light determines the power ratio of the two output beams. This is controlled by adjusting the rotation angle of a half-wave or quarter-wave plate. Successive use of two combinations of wave plate and calcite crystal produces four parallel beams having a square spatial configuration. The intensity ratio of any three of the four beams is controlled individually by adjusting the rotation angles of the two wave plates.

According to our measurement in the visible region using a displacer 30 mm long, the extraordinary ray has an advanced phase of 3.3 mm compared to the ordinary ray (Fig. 1, inset). Simple geometrical calculation using the data of refractive indices 1.6584 for the ordinary ray and 1.4864 for the extraordinary ray yields a phase difference of 3.2924 mm.

The four parallel beams are focused onto one spot after passing through optical delays. Where the DFWM signal appears is determined easily by selecting one beam as a monitor. During measurement, the monitor beam is steered off from the mixing and guided to a light intensity sensor. The DFWM signal appears at the same position where the monitor beam was pointed. The holes of the mask that are placed after the focusing lens have a 1.0 or 0.8 mm diameter, but the resulting diffraction is negligible. Furthermore the diffracted light can be removed by using masks, because diffracted light and DFWM signal light propagate in different directions.

Of the four linearly polarized beams, two are horizon-

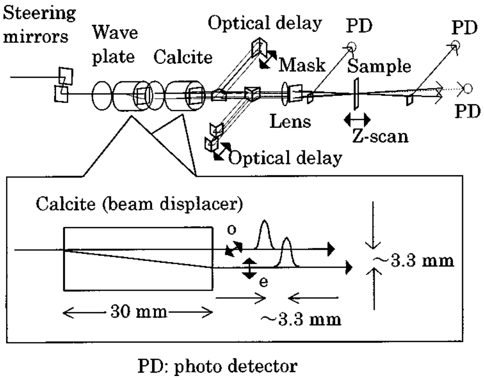

FIG. 1. Compact optical DFWM setup. The inset shows the beam displacement and phase difference between extraordinary and ordinary rays in a 30-mm-long calcite crystal.

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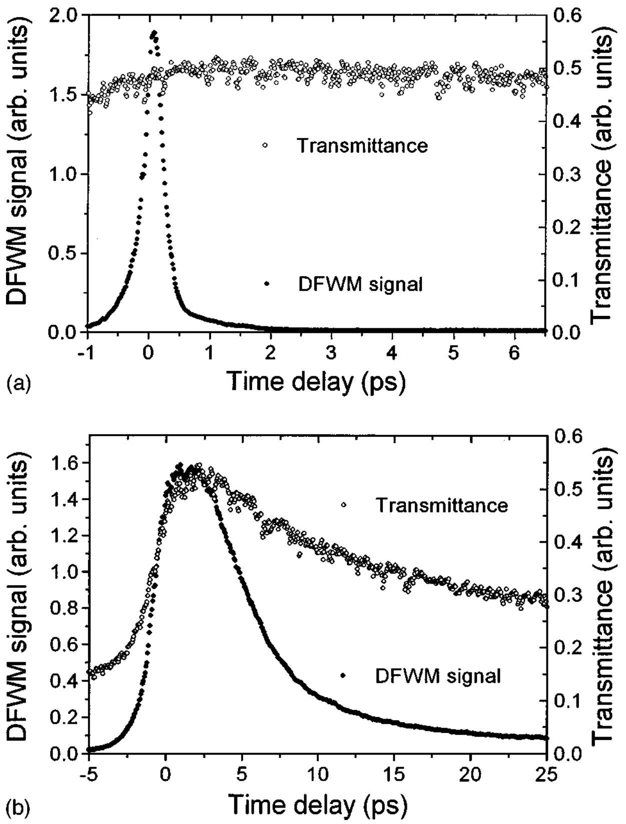

FIG. 2. Time delay plots of DFWM signal and transmitted light of probe beam measured using 613 nm 150 fs laser pulses. The sample is Toshiba sharp-cut filter glass at 630 nm: (a) scanning *T*1 mode; (b) scanning *T*2 mode.

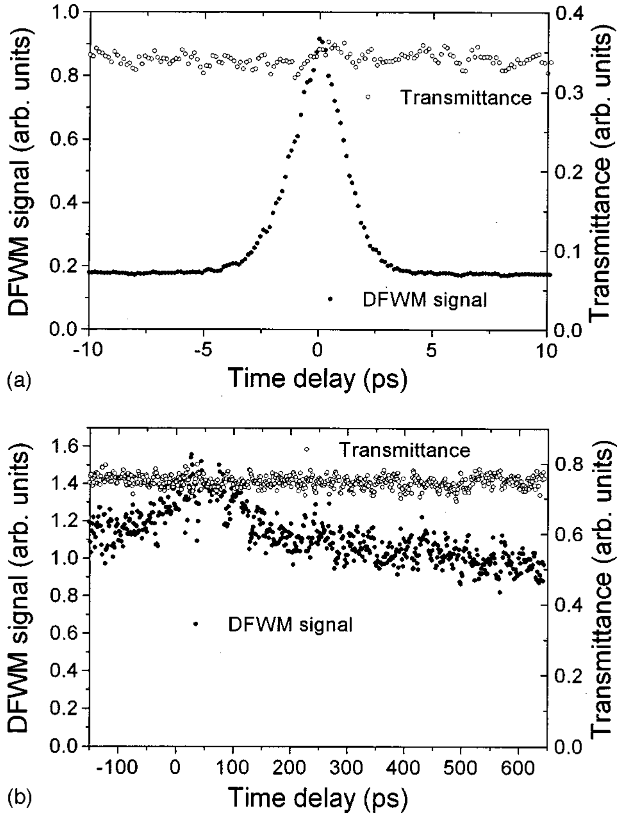

FIG. 3. Same plots as in Fig. 2 but measured using 613 nm 8 ns laser pulses: (a) scanning *T*1 mode; (b) scanning *T*2 mode.

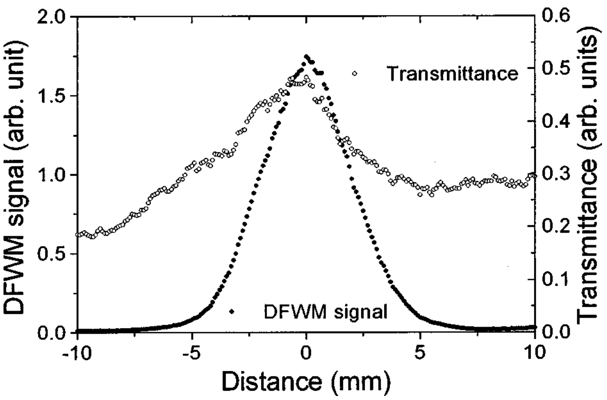

FIG. 4. *Z*-scan plots of DFWM signal and transmitted light of probe beam measured using 613 nm 150 fs laser pulses.

tally polarized and two are vertically polarized. If two beams with different polarization are selected for pumping, net polarization is modulated spatially, forming a polarization grating. If two beams of the same polarization are used, the net intensity is modulated spatially, forming an intensity grating. In these cases, the longitudinal relaxation time (*T*1) and transverse relaxation time (*T*2) are measurable.

In examples of measurement (Figs. 2 and 3), the sample glass is a Toshiba sharp-cut filter at 630 nm 3 mm thick. Figures 2(a) and 2(b) are measured using 613 nm optical parametric amplifier light pumped by 400 nm second harmonic generation light of a 150 fs pulse Ti:sapphire laser. Figures 3(a) and 3(b) are measured using 613 nm optical parametric oscillator light pumped by 355 nm third harmonic generation light of an 8 ns pulse Nd:YAG laser. Figure 3(a) is scanned by the *T*1 mode and Fig. 3(b) by the *T*2 mode. A long duration of 8 ns leads the long plain part at the foot of the delay curve in Fig. 3(a) and the slow delay curve in Fig. 3(b). The relaxation time of Fig. 3(a) is longer than that of Fig. 2(a) due to the long coherent length of the 8 ns pulse laser.

Whether the sample is located at the common beam focus is determined from target translation along the nearly collinear axis of beam propagation and the peak detection of refracted light intensity. In this so-called *Z*-scan measurement (Fig. 4), incoming light beyond the focus point indicates the noise level. Scattered light from the sample and stray light from the surfaces of optical components cause noise. Minimizing the number of optical components and adequately setting the mask filter reduce noise considerably. DFWM signals from a transparent high refractive index glass 1.0 mm thick having nonlinearity on the order of 10^{-12} esu, for example, are easily detected by a conventional photodiode such as a Hamamatsu S2281-01 with a C2719 amplifier.

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