Freezing phase scheme for fast adaptive control and its application to characterization of femtosecond coherent optical pulses reflected from semiconductor saturable absorber mirrors

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We report on the development of a freezing phase scheme for complete-field characterization and adaptive coherent control with a femtosecond pulse shaper. The operational principle is based on a concept that the highest peak intensity will correspond to a frozen phase state of all spectral components involved in a coherent optical pulse. Our experimental and theoretical results reveal this new scheme to be fast and immune to the noise and laser power fluctuation. The freezing phase method has been used to investigate three types of semiconductor saturable absorber Bragg reflector (SBR). The optical pulses reflected from the SBR can be distorted in the spectral phase by a minor structural change of the SBR devices and can be clearly resolved with our method. The technique is useful for a variety of applications that require complete-field characterization and adaptive coherent control on the same setup. © 2005 Optical Society of America

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1. INTRODUCTION

In the quest to control and steer the quantum states of a complex system, an attractive scheme is adaptive laserpulse control. Several algorithms have been developed to tailor a coherent optical field to prepare specific products on the basis of fitness information. The concept appears to be universal and much progress has been made.^{1–11}

By using the broadband property of a ultrashort laser pulse, Dudovich *et al.* successfully combined a coherent control technique with single-pulse nonlinear Raman scattering for selective imaging of molecules in a condensed phase.² Similar coherent control enhancement was also observed in a two-photon fluorescence process.^{3,4} Along with these adaptive laser-pulse control studies,¹⁻¹¹ the question remains as to whether the optimal laser field contains a set of rational rules that govern the dynamics. A recent study suggests that the answer could be in the affirmative.⁵ Therefore the purpose of a femtosecond coherent control study is not only to control the evolution of a complex system but also to deduce the detailed dynamic mechanism from the optimal laser field used.

Coherent control technology with femtosecond optical pulses is usually implemented with a pulse shaper.⁶⁻¹⁸ The only approach of pulse shaping with conserved pulse energy is to adjust the spectral phase for the desired pulse shape.⁶⁻¹¹ In this study we develop a new freezing phase algorithm (FPA) for adaptive coherent control and optical field characterization with a single apparatus.⁴

This paper is organized as follows. In Section 2 we first present the theoretical background of the new freezing phase concept. Our experimental setup is depicted in Section 3. To facilitate the implementation of the freezing phase concept, we offer some simulation results in Subsection 4.A. In Subsection 4.B experimental results of three saturable Bragg reflectors (SBRs) are presented to verify the functionalities of our method. Conclusions are also presented.

2. THEORETICAL BACKGROUND

To illustrate the FPA, we express a coherent optical field in terms of its spectral components:

$$E(t) = \sum_{n=0}^{N} A_n \exp[j(\omega_0 + 2\pi n \Delta \nu_F)t + \phi_n].$$
 (1)

Here ω_0 is the optical carrier frequency and $\Delta \nu_F$, ϕ_n , and A_n denote the frequency span, phase constant, and amplitude of the spectral components, respectively.

In a typical 4-*f* pulse-shaping setup, the pulse spectrum is angularly dispersed with a grating.¹⁹ Each pixel *m* of a one-dimensional pixelated spatial light modulator (SLM) is assumed to accommodate spectral components from K_m^{0} to K_m^{t} . We can impose a phase retardation pattern $\{\Phi_m\}$ on the *M*-pixel SLM and transform the input field into a shaped output:

$$E(t) = \sum_{m=1}^{M} \sum_{k=K_m^{0}}^{K_m^{t}} B_k \exp[i(\omega_0 + 2\pi k\Delta\nu_F)t + \phi_k + \Phi_m].$$
(2)

Equation (2) indicates that the amplitude of the shaped pulse E(t) can reach its peak E_p at $t_p = m/\Delta \nu_F$ (where m

$$E_{p} = \exp(i\omega_{0}t_{p}) \sum_{m=1}^{M} \sum_{k=K_{m}^{0}}^{K_{m}^{t}} B_{k} \exp[i(\phi_{k} + \Phi_{m})].$$
(3)

We arbitrarily choose pixel j from the SLM and use it as a phase modulation component by varying its phase retardance. The remaining M-1 pixels serve as the reference, and by defining

$$C_{j} \exp(i\theta_{j}) = \sum_{m=1}^{j-1} \sum_{k=K_{m}^{0}}^{K_{m}^{t}} B_{k} \exp[i(\phi_{k} + \Phi_{m})] + \sum_{m=j+1}^{M} \sum_{k=K_{m}^{0}}^{K_{m}^{t}} B_{k} \exp[i(\phi_{k} + \Phi_{m})], \qquad (4)$$

$$D_j \exp(i\vartheta_j) = \sum_{k=K_j^0}^{K_j^t} B_k \exp[i(\phi_k + \Phi_j)],$$
(5)

we can express the resulting peak intensity I_p of the shaped output pulse as

$$I_{p} = |E_{p}|^{2} = C_{j}^{2} + D_{j}^{2} + 2C_{j}D_{j}\cos(\theta_{j} - \vartheta_{j}).$$
(6)

The first two terms represent the spectral intensities of the reference and the phase modulation component. The third term denotes interference between the two components. Note that I_p can achieve a maximum with $\theta_j = \vartheta_j$. By successively adjusting every spectral component, we can guide all spectral components toward a frozen phase state, where the shortest pulse can be produced.

To simplify further, we use four phase components to represent the spectral phase profile of a coherent optical pulse: $E(t) = \sum_{n=1}^{4} \nu_n \exp(i\phi_n)$. Figure 1(a) presents a phase-distorted pulse in the complex field plane. To freeze the spectral phases, we first pick up a spectral component $\nu_4 \exp(i\phi_4)$ and align it to the summed direction of the components $\nu_1 \exp(i\phi_1)$, $\nu_2 \exp(i\phi_2)$ and $\nu_3 \exp(i\phi_3)$ [see Fig. 1(b)]. The same procedure is repeated for $\nu_3 \exp(i\phi_3)$, $\nu_2 \exp(i\phi_2)$, and $\nu_1 \exp(i\phi_1)$ as depicted in Figs. 1(c)–1(e).



Fig. 1. Schematic diagrams showing successive freezing steps of a phase-distorted coherent optical pulse. For simplicity, the spectral phase profile of the optical pulse is represented with four spectral phase components.



Fig. 2. Schematic of an adaptive pulse-shaping setup used in this study. The apparatus is comprised of an all-reflective 4-*f* pulse shaper. Here Gr1 and Gr2 denote a pair of gratings; CM-1 and CM-2 are two concave reflectors with a 10-cm focal length; SLM is a one-dimensional pixelated spatial light modulator; BBO is a 3-mm-thick type-I β -Ba₂BO₄ second-harmonic crystal; and PD is a photodiode detector.



Fig. 3. Flow chart showing the procedure of the freezing phase scheme for a theoretical simulation or an experimental study.

In Fig. 1(f) fine-tuning is performed again on $\nu_4 \exp(i\phi_4)$ to achieve the frozen phase state.

3. EXPERIMENT

Figure 2 shows the schematic of an adaptive pulseshaping apparatus used in this study. The laser system is a Cr⁴⁺:forsterite laser (from Avesta Project, Ltd.) pumped by a diode-pumped Yb fiber laser (IPG Photonics, Ltd.). A typical output of the Cr⁴⁺:forsterite laser was 280 mW of average power at a repetition rate of 76 MHz with a 7.5 -W pump. The central wavelength is 1.252 μ m, and a typical full width at half-maximum (FWHM) bandwidth was ${\approx}42$ nm, corresponding to 50-fs pulse duration.

The pulse is tailored by a pulse shaper consisting of a pair of gratings Gr1 and Gr2 (600 grooves/mm), two concave reflectors CM-1 and CM-2 with f=10 cm of focal length, and a liquid-crystal SLM (Cambridge Research and Instrumentation Inc., Woburn, Massachusetts). The pixelated SLM consists of 128 97- μ m-wide pixels with a $3-\mu m$ gap between adjacent pixels. After it is reassembled by the output grating Gr2, the shaped pulse is focused onto a sample under test. The phase distortion in the reflected pulse can be precompensated by the SLM, which yields an optical pulse with constant phase in front of a 3-mm-thick type-I β -Ba₂BO₄ second-harmonic generation (SHG) crystal and generates a maximum secondharmonic (SH) signal. We effectively combine the β -Ba₂BO₄ SHG with a photodiode to offer a functionality of constant-phase detection. The photodiode signal is sent to a computer to deduce the desired compensating phase pattern with our FPA. We neglect the dispersion of the SHG crystal and the preceding optics at this point.

4. RESULTS

A. Simulation

We first perform a series of model simulations to find out an efficient freezing phase procedure. The input coherent optical pulse is assumed to have a Gaussian profile with 50 fs of FWHM pulse duration and 1.252 μ m of central wavelength. The coherent pulse is distorted with a phase profile of $15[(i-64)/64]^2+7[(i-64)/64]^3-7[(i-40)/64]^4$ $-8[(i-60)/64]^5$, where $i=1,2,\ldots,M$ denotes the position index of the SLM pixels.

We start our simulation by dividing the SLM pixels into two groups: We arbitrarily pick up one pixel to serve as a phase modulation and the remaining M-1 pixels form a reference group. We vary the phase of the modulation pixel from 0 to 2π to produce a SH intensity variation with a SHG crystal. For coherent control applications, two to three phase adjustments are usually sufficient to yield the information about the correct phase retardation of the modulation pixel for the frozen phase state. We set the modulation pixel to this phase retardation, and the procedure is repeated by scanning the phase modulation pixel over the SLM until the phase retardations of all pixels have been properly adjusted. Figure 3 is a flow chart showing the freezing phase procedure.

1. Freezing Phase Procedure with a Left-to-Right Scan of the Spatial Light Modulator

The intensity profile of the input pulse is presented with filled circles in Fig. 4(b), and the distorted spectral phase profile is shown in Fig. 4(c) with open circles. The freezing phase procedure was implemented by scanning a phase modulation pixel over the SLM from the left to the right side to compensate for the distorted phase profile. Figure





Fig. 4. (a) Time course of the maximum SH signal obtained during the freezing phase procedure with left-to-right scans of spatial light modulation pixels. Three time courses with the first, second, and third scans are presented from bottom to top. (b) The intensity profiles of a transform-limited pulse (TLP); input pulse with distorted phase (DP); and shaped pulses after first, second, and third freezing phase scans. (c) The spectral phase profile of the input pulse (open circles), compensating phase profile (crosses), and error phase profiles after the first (thin solid curve), second (short-dashed curve), and third (thick solid curve) freezing phase scans.



4(a) shows the time course of the maximum SH signal obtained during the left-to-right freezing phase scans. The xaxis indicates the pixel position index or the corresponding wavelength in a 4-f pulse shaper apparatus. The SH signal plotted along the y axis is a spectrally integrated signal. This is the maximum SH signal that can be obtained when the phase retardation of the selected pixel (i.e., the phase modulation pixel) is adjusted. In Fig. 4(a) the time courses during the first, second, and third scans are presented from bottom to top. The spectrally integrated SH signal had been normalized to that from a transform-limited pulse.

We first note that significant SH signal variation always occurs at the SLM pixels lying inside the input pulse spectrum (i.e., $1.20-1.29 \ \mu m$). The SH intensity reaches 75% of the transform-limited value after the first scan and 98% after the second scan. The corresponding pulse profiles after the first (open squares), second (open triangles), and third freezing scan (thick solid curve) are presented in Fig. 4(b). The pulse spectrum (dashed curve), the distorted phase (open circles), the compensating phase (crosses), and the error phase, which can be obtained from a summation of the distorted phase and the compensating phase, are shown in Fig. 4(c). After the first scan, the root-mean-squared (rms) deviation of the error phase from a linear function decreases to 1.995. The rms deviation can be decreased to 0.640 by the second freezing scan and to 0.095 by the third scan. We point out that a



Fig. 5. (a) Time courses of the maximum SH signal during the freezing phase process with center-to-two-sides scans of spatial light modulation pixels. The first and second scan are presented from bottom to top. (b) The intensity profiles of a transform-limited pulse (TLP), input pulse with distorted phase (DP), and the phase-compensated pulse after the first and second freezing phase scans. (c) The spectral phase profile of the input pulse (open circles), compensating phase profile (crosses), and error phase profiles after the first (thin solid curve) and second (short-dashed curve) freezing phase scans.

linear phase term in the frequency domain simply results in a shift [see Fig. 4(b)] of the entire pulse in the time domain. Thus, from the time invariance principle, our phase retrieval method does not miss any information about optical pulse characteristics.

2. Freezing Procedure with a Center-to-Two-Sides Scan of the Spatial Light Modulator

As shown in Subsection 4.A.1, the SH signal was found to change significantly with phase retardation of the phase modulation pixel lying within the input pulse spectrum. To use this finding effectively, we design the following new scan scheme: The freezing starts from the pixel corresponding to the input spectral peak and proceeds helically toward both sides of the SLM. Figure 5 presents the simulation result. We found that this scan scheme is indeed more efficient than the first scheme. The SH intensity can increase to 92% of the transform-limited value after the first scan, and the rms deviation of the error phase decreases to 1.356. With just two scans, the rms deviation is reduced to as small as 0.374.

3. Freezing Procedure with a Cascading Thinning-Out Scheme

Noise influence from a variety of noise sources is one of the major concerns in a coherent control application. To devise an efficient freezing procedure with high noise immunity, a special thinning-out scheme can be implemented in our freezing phase process. Here we used a pixel-grouping method similar to that reported by Mizoguchi *et al.*¹¹ The pixel numbers of the reference group and the phase modulation group vary from 64:64 (2s), 96:32 (4s), 112:16 (8s), 120:8 (16s), 124:4 (32s), 126:2 (64s), and 127:1 (128s) with ns denoting the total number of pixel segments.

The freezing process starts with a 2s thinning-out scheme and ends with 128s. Except for scheme 2s, which has only one configuration, there are *n* different arrangements for the scheme ns. We cycle the pixel segments through the SLM during each freezing stage. From the simulation result shown in Fig. 6(a), we found that the phase compensation almost completes at the stage 64s. The resulting pulse profiles and phase patterns after each phase compensation stage are presented in Figs. 6(b) and 6(c). Although the rms deviation of the error phase appears to be larger than that with the center-to-two-sides scan, this method was found to be superior in noise immunity, which is depicted in detail in Subsection 4.A.4.

4. Noise Influences on Various Freezing Phase Procedures

To properly assess the noise influences on these freezing phase procedures, we divide the noise source into two terms. The first is a multiplicative noise, which could originate from the intensity fluctuation of coherent pulses. The second one is an additive noise, which mainly comes from thermal noise in detection and feedback electronics. We assume each of the noise sources to be 5% of the maximum optical signal detected. The noises degrade the quality of the SH signal detection and therefore lead to an incorrect phase determination.

The results are shown in Fig. 7. Among the three above-mentioned freezing phase methods, phase freezing with the cascading thinning-out scheme appears to be the fastest method to yield the optimum solution. The slight decrease in SH intensity after 90 freezing steps is mainly caused by erroneous phase determination. This is supported by the observation that the signal level after the thinning-out scheme of 128s is close to the noise level (10%). Further phase freezing steps beyond this point cannot improve the result. Therefore in a real application we stop the freezing procedure in time based on the noise level encountered.

B. Experimental Results and Discussion

1. Phase Retrieving from Phase-Sensitive Second-Harmonic Patterns

The measured SHG signal generated with an optical pulse reflected from a gold-coated mirror is shown in Fig. 8(a). For reference, the spectrum of the mode-locked laser pulse is also plotted along the y axis to show the corresponding wavelengths of the SLM pixels. The experimental result reported here confirms the prediction of Eq. (6) that the SHG modulation sensitively depends on the am-





Fig. 6. (a) Time course of the maximum SH intensity during the freezing phase procedure with a cascading thinning-out scheme. Six time courses with a 2s to 64s thinning-out scheme are presented from bottom to top. (b) The intensity profiles of a transform-limited pulse (TLP), phase-distorted input pulse (DP), and phase-compensated pulse after the first, second, and third freezing phase scans. (c) The spectral phase profile of the input pulse (open circles), compensating phase profile (crosses), and error phase profiles after 8s (short-dashed curve), 32s (dotted curve), and 128s (thick solid curve) freezing phase scans.



Fig. 7. Time courses of the SH signal as a function of the number of freezing steps with the cascading thinning-out scheme (solid curve), center-to-two-sides scan scheme (long-dashed curve), and left-to-right scan scheme (short-dashed curve).

plitude of the spectral components chosen. Once the pixels lie outside the input pulse spectrum, the SH intensity modulation is no longer observable.

We can define the spectral phase sensitivity of the SHG to be the difference between the maximum and the minimum of the SH signal pattern shown in Fig. 8(a) when the phase retardation of the phase modulation group is varied from 0 to 2π . Figure 8(b) presents a direct comparison of the deduced spectral phase sensitivity plot of SHG and the Fourier-transformed infrared (FTIR) spectroscopy of the optical pulse. An excellent agreement was found, indicating that our adaptive phase compensation scheme can yield not only the spectral phase profile but also the spectral amplitude of a coherent optical pulse.

2. Complete-Field Characterization of Semiconductor Saturable Absorber Mirrors with an Adaptively Controlled Pulse Shaper

InAs quantum dots (QDs) have important applications for ultrafast optical shaping at 1.3 μ m. Under conditions of strong excitation, the absorption is saturated because initial states of the pump transition are depleted whereas the final states are partially occupied. Within 50–300 fs of excitation, the carriers in each band thermalize, and this leads to a partial recovery of the absorption. The faster time constant is more effective in shaping subpicosecond pulses.²⁰

A SBR usually consists of a highly reflective Bragg mirror and quantum well or QDs embedded in it. By proper choice of the position of the saturable semiconductor layer it is possible to change the effective field that bleaches its optical absorption and therefore the saturation fluence of the device. The main limitation of the SBR is most probably the strong wavelength dependence of the group delay introduced by the structure, which might be a problem for the generation of very short (<40-fs) pulses or to have a large tuning range with fixed pulse parameters.²¹ It is therefore important to characterize the complete-field profile of a femtosecond optical pulse reflected from a variety of SBR structures to reveal the underlying pulse distortion processes. Here we employ our newly developed adaptive control apparatus on three types of SBR sample.

The first SBR device is comprised of two coupled Ga_{0.47}In_{0.53}As quantum wells (QWs) that are embedded in an Al_{0.48}In_{0.52}As quarter-wave layer on a distributed Bragg reflector (DBR) stack (hereafter abbreviated as d-QW). The DBR stack is formed with 25 pairs of GaAs/AlAs designed to yield a Bragg wavelength at λ_B = 1.23 μ m. The other is a self-assembled InAs QD layer embedded in a quarter-wave- (λ /4)-thick or half-wave-(λ /2)-thick GaAs layer on a DBR stack. The DBR structures of the two devices are identical and contain a stack of 21-periods of 97 nm/112 nm GaAs/Al_{0.92}Ga_{0.08}As to yield high reflection at 1.3 μ m. The schematic device structures and the corresponding field distribution at a wavelength of 1.25 μ m are depicted in Fig. 9.

We first employ a SHG frequency-resolved optical gating (FROG) technique²² to characterize the femtosecond pulses reflected from the three SBR devices. The result for the QD $\lambda/2$ structures is shown in Fig. 10(a). The experimental SHG FROG trace was retrieved with an error of 0.0025. The retrieved spectral phase profiles for the three SBR devices, which are presented in Fig. 10(c), overlap with each other near the central region but the phase profiles of QD $\lambda/4$ and QD $\lambda/2$ differ significantly



Fig. 8. (a) Measured SHG signal with an optical pulse reflected from a gold-coated mirror is plotted as a function of the phase retardation of the phase modulation group of three consecutive pixels and their corresponding wavelength in a 4-*f* pulse-shaper apparatus. (b) Spectral phase sensitivity plot of SHG deduced from the FPA (open circles) and the optical pulse spectrum measured with FTIR spectroscopy (solid curve).



Fig. 9. Calculated distribution of field strength (white color for zero and black color for maximum strength) is presented along with the device structures of saturable absorber Bragg reflectors with InAs QD or d-QW embedded in a (a) $\lambda/4$ -thick layer or (b) $\lambda/2$ -thick layer.

at the short-wavelength side of the pulse spectra. As shown in Fig. 9, the field strength experienced by the InAs QDs in QD $\lambda/2$ is smaller, and therefore we expect to observe a weaker pulse-shaping effect and therefore a larger phase distortion in QD $\lambda/2$.

After performing the SHG FROG analysis, we move on to diagnose the complete-field characteristics by using our apparatus with the freezing phase scheme. The results are summarized in Fig. 11. We first use the SHG spectral phase sensitivity (see Fig. 8) to determine the spectral profile. Figure 11(a) presents a direct comparison of the spectral profiles of an optical pulse reflected from the d-QW sample deduced with the FPA and that measured with FTIR spectroscopy. An excellent agreement was found. We then present the measured phase profiles with the FPA for the three SBR devices in Fig. 11(b). The global features of the measured spectral phase profiles are similar to that obtained with the SHG FROG technique. The deviation was observed to occur at a region with a small spectral amplitude where retrieval with the FROG algorithm is usually less reliable. The slight shift of the QD $\lambda/2$ phase profile (short-dashed curve) from those of d-QW (solid curve) and QD $\lambda/4$ (long-dashed curve) also occurs in the spectra measured with FTIR spectroscopy. Note that the device structure of QD $\lambda/2$ is similar to QD $\lambda/4$ except for a twice thicker QDs layer embedded in QD $\lambda/2$. The clearly distinguishable differences in the spectral phase profiles ensure that our new complete-field characterization scheme is sensitive and accurate enough to reveal influences on femtosecond optical pulses from subtle structural changes in the SBR. Furthermore, unlike SHG FROG where pulse characteristics are retrieved with a sophisticated mathematical procedure, our method is a direct approach.

As explained in the above paragraph, the main limitation of the SBR in ultrashort laser application is the strong wavelength dependence of the group delay introduced by the device structure. With the measured spectral phase profiles we can deduce the group delay caused by the SBR devices. To properly remove any influences from laser optics, the phase profile of an optical pulse reflected from a gold mirror placed at the same position of the SBR devices was also measured. We deduced the group-delay times of the SBR devices by first taking a difference between the spectral phase profiles of the SBR and the gold mirror and then differentiating the phase difference profiles with angular frequency. The results are presented in Fig. 11(c), which shows that the d-QW SBR exhibits much weaker wavelength-dependent group delay within the entire spectral range of the optical pulse. Indeed this device had been confirmed experimentally to be able to generate femtosecond laser pulses with a pulse duration less than 60 fs at 1.25 μ m. As expected, among the three SBR structures, the QD $\lambda/2$ shows the largest group-delay variation in the spectral range, especially for the spectral components longer than 1.27 μ m. The larger group-delay variation caused by QD $\lambda/2$ can originate from the fact that a smaller field strength is experienced by the InAs QDs and therefore has a weaker pulseshaping effect.

3. Comparison with a Genetic Algorithm

For coherent control applications, time to achieve optimum control is often the major concern. In Fig. 12 we present a comparison of the measured SHG intensities with the freezing phase scheme and with a genetic algorithm.¹⁴⁻¹⁶ To ensure that the comparison is taken on a similar base, the genetic algorithm is implemented with a SLM by a 3s thinning-out scheme.¹¹

The genetic algorithm is implemented with a population size of 200, which comprises 90 of the fittest individuals from the previous generation, 10 randomly generated individuals, 60 from cross operation, and 40 from mutation of the 90 fittest parents. It was shown that the total number of SLM adjustments with our FPA can be as low



Fig. 10. (a) Measured and (b) retrieved SHG FROG patterns of a femtosecond optical field at 1.25 μ m reflected from the QD $\lambda/2$ SBR and (c) the retrieved spectral phase profiles from d-QW (solid curve), QD $\lambda/4$ (long-dashed curve), and QD $\lambda/2$ (short-dashed curve) SHG FROG traces.



as 700 to yield a phase resolution of 2°. Therefore adaptive coherent optimization with our freezing phase scheme can be greatly shortened. Furthermore, the quality of the optimization with our method is also slightly better than that with a genetic algorithm. The genetic algorithm was found to be more sensitive to the fluctuation of laser power. When the laser power decreases, the genetic algorithm tends to judge all individuals after this power reduction to be poorer than that of the previous generation. This erroneous judgment does not happen with the FPA.



Fig. 12. Measured SHG is plotted as a function of the number of phase adjustments with the FPA (solid curve) and a genetic algorithm (GA, dashed curve).



Fig. 11. (a) Spectral profiles of optical pulses reflected from d-QW (open circles) deduced with the FPA. For comparison, the pulse spectral profiles from d-QW (solid curve), QD $\lambda/4$ (long-dashed curve), and QD $\lambda/2$ (short-dashed curve) measured with FTIR spectroscopy are also presented. (b) Spectral phase profiles of optical pulses reflected from a gold mirror (thin solid curve), d-QW (thick solid curve), InAs QD $\lambda/4$ (long-dashed curve), and InAs QD $\lambda/2$ (short-dashed curve) determined with the freezing phase scheme are shown. (c) The deduced group-delay time of the three SBR devices are plotted over the pulse spectral range.

In summary, we have developed a new method for phase compensation with a femtosecond pulse shaper. The operational principle is based on the fact that the highest peak intensity corresponds to a complete frozen phase state of all spectral components. Our experimental and theoretical results reveal several advantages. This freezing phase scheme was employed to analyze three types of semiconductor saturable absorber Bragg reflectors (SBRs), and the influence on phase distortion from subtle structural change in a SBR can be revealed clearly. Our results show the new scheme to be faster and more accurate for complete-field characterization and adaptive control of coherent pulses. We believe that our technique will be useful for various applications that require complete-field characterization and adaptive coherent control on the same setup.

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REFERENCES

- 1. R. S. Judson and H. Rabitz, "Teaching lasers to control molecules," Phys. Rev. Lett. **68**, 1500–1503 (1992).
- 2. N. Dudovich, B. Orion, and Y. Silberberg, "Single-pulse coherently controlled nonlinear Raman spectroscopy and

microscopy," Nature (London) 418, 512-515 (2002).

- 3. T. Brixner, N. H. Damrauer, P. Niklaus, and G. Gerber, "Photoselective adaptive femtosecond quantum control in the liquid phase," Nature (London) **414**, 57–60 (2001).
- I. Pastirk, J. M. Dela Cruz, K. A. Walowicz, V. V. Lozovoy, and M. Dantus, "Selective two-photon microscopy with shaped femtosecond pulses," Opt. Express 11, 1695–1701 (2003), http://www.opticsexpress.org.
- C. Daniel, J. Full, L. González, C. Lupulescu, J. Manz, A. Merli, S. Vajda, and L. Wöste, "Deciphering the reaction dynamics underlying optical control laser fields," Science 299, 536–539 (2003).
- A. Assion, T. Baumert, M. Bergt, T. Brixner, B. Kiefer, V. Seyfried, M. Strehle, and G. Gerber, "Control of chemical reactions by feedback-optimized phase-shaped femtosecond laser pulses," Science 282, 919–922 (1998).
- W. S. Warren, H. Rabitz, and M. Dahleh, "Coherent control of quantum dynamics: the dream is alive," Science 259, 1581-1589 (1993).
- T. C. Weinacht, J. L. White, and P. H. Bucksbaum, "Toward strong field mode-selective chemistry," J. Phys. Chem. A 103, 10166–10168 (1999).
- R. J. Levis, G. M. Menkir, and H. Rabitz, "Selective bond dissociation and rearrangement with optimally tailored strong-field laser pulses," Science 292, 709–713 (2001).
- strong-field laser pulses," Science 292, 709-713 (2001).
 C. J. Bardeen, V. V. Yakovlev, K. R. Wilson, S. D. Carpenter, P. M. Weber, and W. S. Warren, "Feedback quantum control of molecular electronic population transfer," Chem. Phys. Lett. 280, 151-158 (1997).
- 11. R. Mizoguchi, K. Onda, S. S. Kano, and A. Wada, "Thinning out in optimized pulse shaping method using genetic algorithm," Rev. Sci. Instrum. **74**, 2670–2674 (2003).
- D. Yelin, D. Meshulach, and Y. Silberberg, "Adaptive femtosecond pulse compression," Opt. Lett. 22, 1793-1795

(1997).

- D. Meshulach, D. Yelin, and Y. Silberberg, "Adaptive realtime femtosecond pulse shaping," J. Opt. Soc. Am. B 15, 1615–1619 (1998).
- T. Baumert, T. Brixner, V. Seyfried, M. Strehle, and G. Gerber, "Femtosecond pulse shaping by an evolutionary algorithm," Appl. Phys. B: Lasers Opt. 65, 779–782 (1997).
 E. Zeek, R. Bartels, M. M. Murnane, H. C. Kapteyn, and S.
- E. Zeek, R. Bartels, M. M. Murnane, H. C. Kapteyn, and S. Backus, "Adaptive pulse compression for transform-limited 15-fs high-energy pulse generation," Opt. Lett. 25, 587–589 (2000).
- A. Efimov, M. D. Moores, N. M. Beach, J. L. Krause, and D. H. Reitze, "Adaptive control of pulse phase in a chirpedpulse amplifier," Opt. Lett. 23, 1915–1917 (1998).
- J. Kunde, B. Baumann, S. Arlt, F. Morier-Genoud, U. Siegner, and U. Keller, "Adaptive feedback control of ultrafast semiconductor nonlinearities," Appl. Phys. Lett. 77, 924–926 (2000).
- M. M. Wefers and K. A. Nelson, "Analysis of programmable ultrashort waveform generation using liquid-crystal spatial light modulators," J. Opt. Soc. Am. B 12, 1343–1362 (1995).
- A. M. Weiner, "Femtosecond pulse shaping using spatial light modulators," Rev. Sci. Instrum. 71, 1929–1960 (2000).
- U. Keller, "Recent developments in compact ultrafast lasers," Nature (London) 424, 831–838 (2003).
- D. Kopt, G. Zhang, R. Fluck, M. Moser, and U. Keller, "All-in-one dispersion-compensating saturable absorber mirror for compact femtosecond laser sources," Opt. Lett. 21, 486–488 (1996).
- R. Trebino, ed., Frequency-Resolved Optical Gating: The Measurement of Ultrashort Laser Pulses (Kluwer Academic, Boston, Mass., 2002).