Eliminate coherence spike in reflection-type pump-probe measurements

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Abstract: The coherence spike of femtosecond laser pulses in the reflection-type pump-probe measurements has been systematically studied in the semiconductor (100) InP. By varying the setup of the pump-probe measuring system, *i.e.* the polarizations of pump and probe pulses, the incident angles of pump and probe beams, and the interval of delay time between pump and probe pulses, the dramatic changes in the strength of coherence spike could be clear observed. Furthermore, the proposed methods to remove the coherence spike from the transient reflectivity curves have been demonstrated in the time-domain measurements.

OCIS codes: (300.6500) Spectroscopy, time-resolved; (320.7100) Ultrafast measurement; (260.7120) Ultrafast phenomena

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1. Introduction

The pump-probe technique has been applied to the time-resolved measurements over past decades. In order to understand the reaction mechanisms or the electronic structure of the excited states in media, measuring the characteristics of lifetime is widely adopted by scientists. Moreover, this pump-probe technique is also of increasing interest in solid state physics, where it is used for studying metals, semiconductors, superconductors, and other materials. However, this technique gives rise to the coherence interference, *i.e.* the so-called coherence spike or coherence artifact, around zero delay time between pump and probe pulses. In 1981, Vardeny and Tauc [1] first proposed that the coherence artifact mostly refers to a pump polarization coupling term appearing when pump and probe overlap, which was also confirmed by the spectral hole burning of Cruz *et al*. [2]. Then, Eichler *et al*. further explained it by diffraction from a transient grating induced by interference of the pump and probe beams [3]. They provided very important basic principles for the generation of the coherence artifact. Indeed, this coherence artifact could be found in most time-resolved spectroscopy (the transient reflectivity change $\Delta R/R$ or the transient transmissivity change $\Delta T/T$)[4]-[7] and disturbs the analysis of relaxation dynamics to determine the amplitude of signal near zero delay and relaxation time from the trace of $\Delta R/R$ or $\Delta T/T$. For instance, Wang *et al*. [4] revealed that the controversy in lifetime of p-like excited state of the hydrated electron is due to the existence of a coherence spike at zero delay time in pump-probe spectroscopic kinetics traces. After removing this spike effect, they could obtain the intrinsic lifetimes of the two incompletely relaxed states in bulk water are 180 ± 30 and 545 ± 30 fs. Besides, the reflection-type pump-probe measurements for the numerous solid state materials also suffer from this serious problem. Therefore, how to unambiguously distinguish the true pump-probe signal of materials from the annoying coherence spike or obtain the coherence-spike-free pump-probe signal is indeed the key issue in the time-resolved femtosecond spectroscopy. In this paper, we report the systematical studies for the origin of the coherence spike in reflection-type pump-probe measurements and further demonstrate the effective methods for removing it.

2. Experiments

The experiments were performed with a femtosecond Ti:sapphire laser of Micra-10 (Coherent) pumped at 532 nm from an Nd:YAG laser (Verdi, Coherent). A beamsplitter reflected 50% of light in a pump channel, whereas the remnant was transmitted and served as a probe. Both pump and probe beams passed through two acousto-optic modulators (AOM). However, only one in the pump beam was driven by the RF driver and modulated the pump beam at 87 KHz. After travelling through a delay stage, a half-wave ($\lambda/2$) plate, and a polarizer, the pump beam was focused by a 200-mm lens on the surface of a sample with 125 μm in diameter. The $\lambda/2$ plate and polarizer allowed us to adjust the intensity and polarization (electric field, **E**) of pump beam (both needed to intensity control). On the other hand, the probe beam only passed through the $\lambda/2$ plate and the polarizer after the AOM and focus on the surface of the sample with 84 μm in diameter by the 150 mm lens. The powers of pump and probe beams were 32 mW and 3 mW, respectively. The best spatial overlap of pump and probe beams on the sample was realized by monitoring with a CCD camera. The reflection of the probe beam was received by the photodiode. However, since the variation due to the reflectivity change of the samples was very small, typically between 10^{-5} and 10^{-7}, and it was very difficult to detect by the photodiode directly under the noisy background such as laser noise, electric noise, and

#110045 - $15.00 USD Received 13 Apr 2009; revised 22 May 2009; accepted 10 Jun 2009; published 22 Jun 2009

mechanical vibration. Usually the lock-in technique is used to reduce such background noise. In order to eliminate the high-level noise in the audio frequency, the small signal was modulated at 87 KHz where the noise was smaller and the gain of the narrow-band amplifier was maxima. The narrower bandwidth could be adjusted by the longer time constant of the phase detector. However, too long time constant would obscure the signal. Typical value used in this experiment was 1 s. As mentioned above, the AOM modulation was commonly applied to the pump pulse train to detect the signal due merely to change in the reflected probe intensity induced by the pump pulses. Then, the probe pulse train was also modulated by the reflectivity change of the samples on a constant intensity of the probe pulse train, *i.e.* a AC signal $\Delta I(t)$ was added to a DC signal $I_0(t)$. This signal was detected by the photodiode and sent to the lock-in amplifier, which was phase-locked to the AOM. The lock-in amplifier only extracted the AC signal $\Delta I(t)$, of which frequency was exactly equal to the modulation frequency in-phase with the AOM. By varying the delay time (t) between pump and probe pulses, $\Delta I(t)$ would change as a function of delay time. Therefore, the temporal evolution of the reflectivity change (ΔR) could be measured in the reflection-type pump-probe measurements.

3. Results and discussion

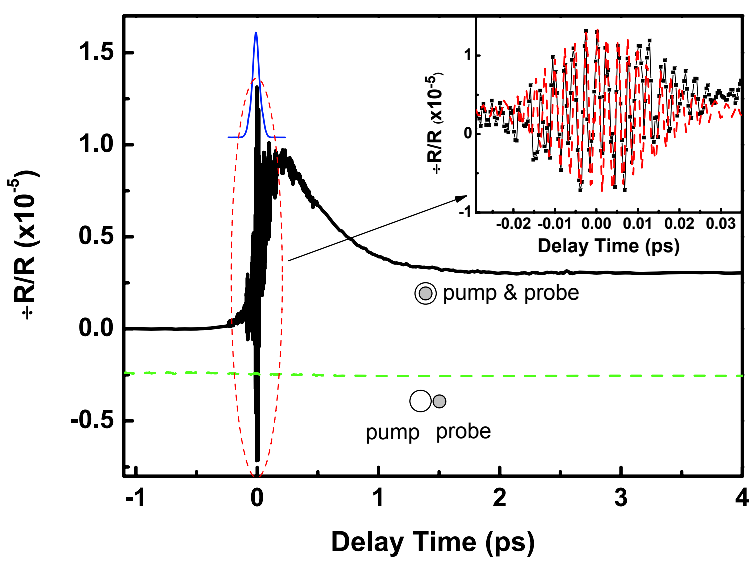

Fig. 1. The transient reflectivity change of InP. The thin line represents the second-order autocorrelation trace. The dashed line indicates that the transient reflectivity change under the separation of pump and probe spots on the surface of samples. The inset shows the coherence spike on an enlarged scale. The dashed line in the inset is the first-order interferometric autocorrelation trace. All of the polarization configurations are pump $\perp$ (the polarization of pump pulses is perpendicular to the incident plan) and probe $\|$ (the polarization of probe pulses is parallel to the incident plane). All of the measurements were performed at $\theta_{pump} = 1^{\circ}$, $\theta_{probe} = 7^{\circ}$, and the interval of delay time $\Delta t = 0.33$ fs.

In order to investigate the coherence spike in the reflection-type pump-probe measurements, we chose the popular semiconductor (100) InP as a test sample, which has been well studied in

#110045 - $15.00 USD Received 13 Apr 2009; revised 22 May 2009; accepted 10 Jun 2009; published 22 Jun 2009

ultrafast dynamics [8]. The typical $\Delta R/R(t)$ of InP is shown in Fig. 1. After the excitation of a pump pulse, the $\Delta R/R$ rapidly grows and then decays to a thermal equilibrium state. Besides, the most dramatic variation is the strong coherence interference (spike) around zero delay time marked by the dashed circle in Fig. 1. This coherence spike appears as the second-order (intensity) autocorrelation trace of pump and probe beams. Namely, this can only be observed at the temporal overlapping region of pump and probe pulses. Through the precisely delay-time scanning with the resolution of 0.33 fs, a periodic oscillation could be clearly observed at zero delay time as shown in the inset of Fig. 1. Comparing with the result of the first-order interferometric autocorrelation curve (the dashed line in the inset of Fig. 1), which was directly measured at the position of the samples, the periodic oscillation are due to the interference between pump and probe pulses. However, under the standard pump-probe setup, e.g. the configuration in Fig. 2(a), the detector only receives the probe pulses and no pump pulses. There are two possibilities of interfering pump pulses. One is due to the scattering from the surface of samples. The other is due to the diffraction from the transient grating in materials. The coherence spike disappears while the pump and probe spots are slightly separated in space as shown by the dashed line in Fig. 1. This implies that the coherence spike cannot be simply explained by the scattering due to the surface roughness.

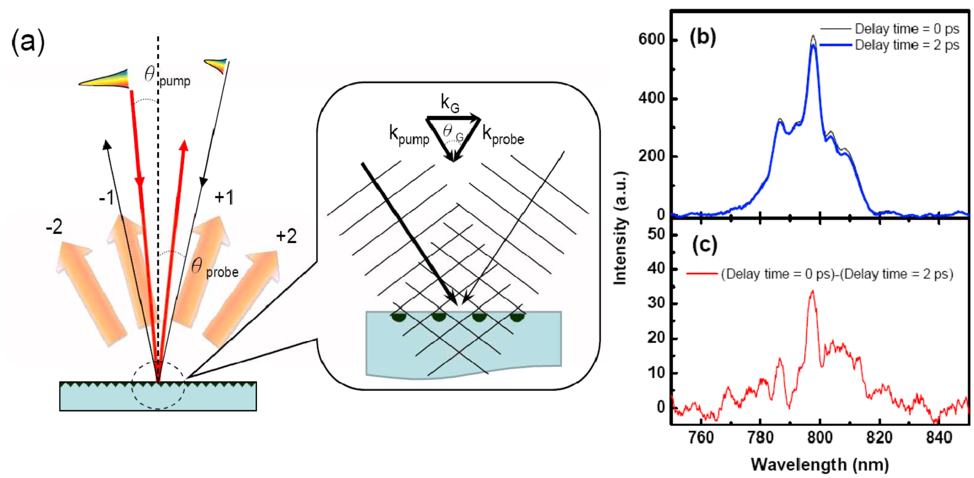

Fig. 2. (a) Schematics of a reflection-type pump-probe measurement and the generation of a transient grating in materials. The thick lines represent the pump beam. The thin lines represent the probe beam. The thick arrows represent the diffracted light at various orders ($m = 0, +1, -1, \ldots$). $\theta_G = \theta_{pump} + \theta_{probe}$ (b) The spectra measured at $\theta = 8^\circ$ (from the surface normal of samples). (c) The spectrum obtained by subtracting the spectrum with thick line from the spectrum with thin line in (b).

Figure 2(a) sketches the generation of a transient grating in the reflection-type pump-probe experiments. Both pump and probe pulses with different propagation direction overlap on the surface of a sample. If the delay time between pump and probe pulses is around zero, they produce an interference pattern on the sample. The modulation of interference pattern causes a periodical change in the refractive index (caused by the bleaching of the interband absoption [9]) [3]. The transient grating vector is given by (1)

$$\mathbf{k}_G = \mathbf{k}_{pump} - \mathbf{k}_{probe} \quad (1)$$

where $\mathbf{k}_{pump}$ and $\mathbf{k}_{probe}$ are the propagation vectors of pump and probe pulses, respectively.

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According to the grating equation (2):

$$d(\sin\theta_m - \sin\theta_{pump}) = m\lambda \tag{2}$$

where $d = 2\pi$ / k_G is the period of the transient grating, θ_{pump} is the incident angle of a pump beam, m is the order of interference, and θ_m is the diffraction angle of the m-th order. The pump pulse could be diffracted by this transient grating as shown with the thick arrows in Fig. 2(a). At $\theta = 8^\circ$ (from the surface normal of the sample) around $m = +1$, the spectra of the scattering light have been measured at the delay time = 0 ps and 2 ps. Fig. 2(c) shows the difference between the thin line with delay time 0 ps and the thick line with delay time 2 ps in Fig. 2(b). This additional spectrum around the diffractive angle with $m = +1$ indicates that the existence of the transient grating at zero delay time. Once the pump pulse is diffracted into the detector, the interference between pump pulse and probe pulse will be observed. For instance, the optical path of diffracted light with $m = -1$ is just collinear with the optical path of the probe pulses and then lead to the coherence interference around the zero delay time. This implies that the coherence spike is unambiguously caused by the diffracted light of pump pulses due to the transient grating of the samples, which is consistent with the theoretical results of Eichler *et al*. in transmission-type pump-probe experiments [3].

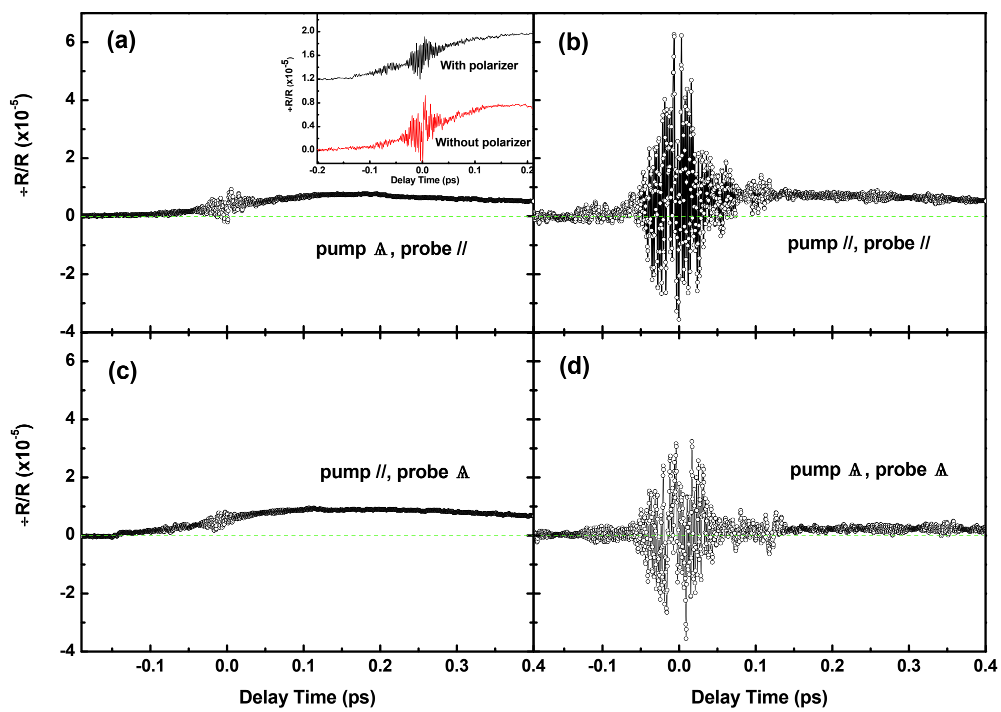

Fig. 3. The coherence spike in $\Delta R/R$ under four polarization configurations (a) pump $\perp$ (the polarization of pump pulses was perpendicular to the incident plan) and probe $\|$ (the polarization of probe pulses is parallel to the incident plane), (b) pump $\|$ and probe $\|$, (c) pump $\|$ and probe $\perp$, (d) pump $\perp$ and probe $\perp$. The inset of (a) is that the measurements were performed with and without the polarizer in front of the detector. All of the measurements were performed at $\theta_{pump} = 1^\circ$, $\theta_{probe} = 7^\circ$, and the interval of delay time $\Delta t = 0.33$ fs.

In principle, the interference is expected not to take place between the pump and probe

#110045 - $15.00 USD Received 13 Apr 2009; revised 22 May 2009; accepted 10 Jun 2009; published 22 Jun 2009

pulses with perpendicular polarization. However, the coherence spike was still observed in Fig. 1 which was performed with pump $\perp$ (the polarization of pump pulses is perpendicular to the incident plane) and probe $\parallel$ (the polarization of probe pulses is parallel with the incident plane). This maybe simply assigned to the imperfect linear polarization of light even the polarizer with extinction ratio $> 10^4 : 1$ we used in this experiment. Additionally, we added one polarizer in front of the detector to exclude the pump pulse of which polarization is perpendicular to the polarization of the probe pulse or eliminate the coherence spike. As you can see in the inset of Fig. 3(a), unexpectedly, the coherence spike was still observed. When the polarizations of both pump and probe pulses are set to be parallel with the incident plane (pump $\parallel$, probe $\parallel$), the extremely large coherence spike appear around zero delay time as shown in Fig. 3(b). Similarly, this situation also happens in the case of the polarizations of both pump and probe pulses set to be perpendicular to the incident plane (pump$\perp$, probe$\perp$ in Fig. 3(d)), but it is smaller than that in Fig. 3(b). This is because of the weaker modulation strength in the interference pattern due to the polarization of pump and probe pulses which are parallel to the incident plane in Fig. 3(d). Thus, the coherence spike in the case of pump $\parallel$ probe (Fig. 3(b) and 3(d)) is much larger than that in the case of pump $\perp$ probe (Fig. 3(a) and 3(c)).

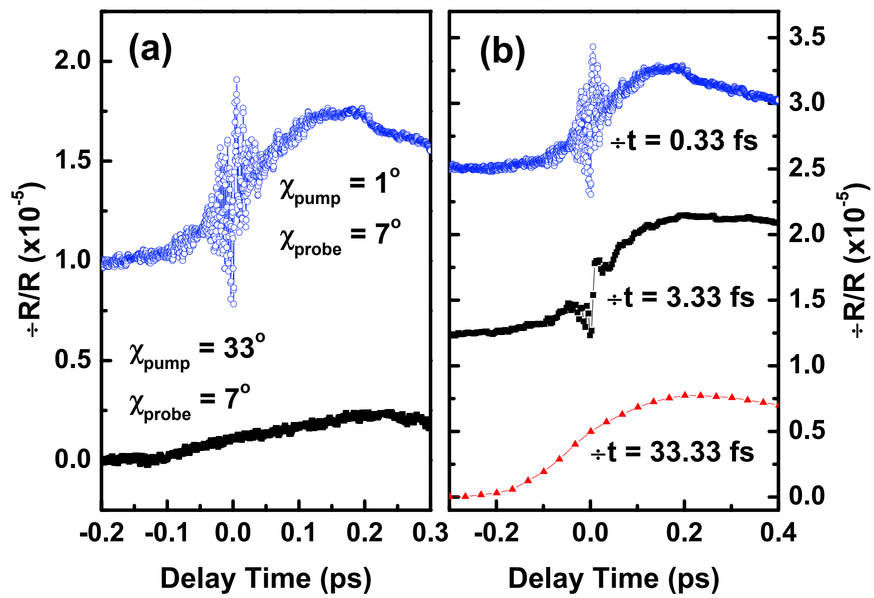

Fig. 4. (a) The transient reflectivity changes with various incident angles of pump and probe beams. All of the measurements were performed at the interval of delay time $\Delta t = 0.33$ fs. (b) The transient reflectivity changes with various intervals of delay time Δt. All of the polarization configurations are pump $\perp$ and probe $\parallel$.

Could we eliminate the coherence spike in this reflection-type pump-probe measurement? According to the above analysis, the coherence spike is mainly induced by the transient grating due to the interference between pump and probe pulses. Therefore, one possible way is to reduce the transient grating in the sample. For instance, the coherence spike completely disappears with $\theta_{pump} = 33^\circ$ and $\theta_{probe} = 7^\circ$ in Fig. 4(a). When the incident angles of pump and probe beams are increased, the size of the transient grating vector $\mathbf{k}_G$ will increase according to Eq. (1). Then, the period (d) of transient grating becomes shorter to increase the diffraction angle θ_m. Further, the grating equation (2) will be failed to be satisfied with a larger incident

#110045 - $15.00 USD   Received 13 Apr 2009; revised 22 May 2009; accepted 10 Jun 2009; published 22 Jun 2009

angle of the pump beam. Thus, the pump pulses cannot be diffracted by the transient grating established by the interference between pump and probe pulses and no coherence spike in $\Delta R/R$. In some experiments small θ_{pump} and θ_{probe} are needed to be set for the angle-resolved ultrafast spectroscopy in anisotropic materials such as high-T_c superconductor, $YBa_2Cu_3O_{7-\delta}$, by varying the polarizations of pump and probe pulses [10]-[12]. If the θ_{pump} and θ_{probe} must be fixed in small angle, the coherent spike in $\Delta R/R$ could be removed by increasing the interval of delay time. As shown in the Fig. 4(b), the coherence spike gradually vanishes with increasing the interval of delay time Δt. For $\Delta t = 33.3$ fs, there is no coherence spike in $\Delta R/R$.

Generally, the coherence spike is not welcome in the pump-probe measurements. On the other hand, however, this coherence spike can give us the information for the characteristics of pulses we used. In the inset of Fig. 1, the oscillation of coherence spike is almost equal to the results of standard first-order autocorrelation measurements. This means that the characteristics of pulses, *i.e.* the coherent length or bandwidth can be directly estimated from the coherence spike.

4. Summary

In summary, we have demonstrated the origin of coherence spike in the reflection-type pump-probe measurement. The strength of coherence spike changes with various setups in the pump-probe measuring system, such as the polarization of pump and probe pulses, incident angle of pump and probe beams, and the interval of the delay time. Moreover, two effective methods to eliminate the coherence spike in the reflection-type pump-probe measurement have been suggested and demonstrated. One is to utilize the experimental configuration with the large incident angles of pump and probe beams which causes grating equation fail to be satisfied. The other way is to increase the interval of delay time Δt during measurements.

Acknowledgments

This work was supported by the National Science Council of Taiwan, R.O.C. under grant: NSC95-2112-M-009-011-MY3, NSC96-2923-M-009-001-MY3, and by the Grant MOE ATU Program at NCTU.

#110045 - $15.00 USD Received 13 Apr 2009; revised 22 May 2009; accepted 10 Jun 2009; published 22 Jun 2009