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Elimination of the Coherent Artifact in a Pump-Probe Experiment by Directly Detecting the Background-Free Diffraction Signal *

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The influence of the coherent artifact in a semiconductor Ga-doped ZnO film on femtosecond pump-probe measurement is studied. The coherent artifact mixed into the pump-probe signal can be directly inspected by detecting the background-free first-order diffraction signal induced by the interference between the pump and probe pulses. Experimental results show that by varying the polarization angle or adjusting the relative intensity between the pump and probe pulses, the coherent artifact can be eliminated from the pump-probe measurement.

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The degenerate pump-probe technique has been applied to time-resolved measurements over the past decades.[1−9] Observing ground-state bleaching and excited-state absorption or stimulated emission through transient absorption experiments is widely applied in investigations of the carrier dynamic process of organic materials, semiconductors and other materials.[10,11] During the degenerate pump-probe process, however, the coherent artifact can be easily formed around zero delay time between the pump and probe pulses.[12,13] This phenomenon is explained by the diffraction from a transient grating induced by interference between the pump and probe beams.[14−17] When the pump and probe pulses are temporally overlapped, the interference between the pump and probe pulses leads to a transient refractive-index grating due to the nonlinear effect in non-resonance materials.[18−20] In a resonance material, the interference pattern of two coherent pulses can generate a periodic spatial distribution of the carrier concentration.[21−23] This spatial periodicity of the free electrons and holes causes a similar periodic perturbation in the refractive index.[24]

The two kinds of grating described above lead to a diffraction of the pump beam into the probe beam.[25,26] Indeed, this coherent artifact can be found in most time-resolved degenerate pump-probe experiments. Experimentally, the diffraction signal of the pump beam mixed into the probe beam is overlapped with the pump-probe trace, which disturbs the analysis of relaxation dynamics to determine the amplitude of the signal and relaxation.[18,27−30]

In this Letter, the influence of the coherent artifact on femtosecond pump-probe measurement is studied in semiconductor Ga-doped ZnO film. The coherent artifact mixed into the pump-probe signal can be directly inspected by detecting the background-free first-order diffraction signal induced by the interference between the pump and probe pulses. Experimental results show that by varying the polarization angle or adjusting the relative intensity between the pump and probe pulses, the coherent artifact can be eliminated from the pump-probe measurement.

The ZnO:Ga film was fabricated by radiofrequency (rf, 13.56 MHz) magnetron sputtering technique. The target for the rf magnetron sputtering process is prepared by cold pressing and subsequent sintering of a mixture of ZnO and Ga_2O_3 powders (both 5N) with a 95:5 molar ratio of Zn:Ga in a oxygen atmosphere at 1050℃ for 12 h. The process chamber was a custom-built rf magnetron sputtering chamber with a base pressure of 6.67×10^{-5} Pa. A ZnO:Ga target was used for sputtering with a flux ratio of Ar:O_2=3:1, the rf power was 150 W. The distance between the substrate and target was 12 cm. The deposition rate was about 22 nm/min (the rf power was 150 W). The double smoothed sapphire substrate was degreased both in acetone and in methanol, each for 10 min at room temperature, then etched in HF for 1–2 s, followed by rinsing in deionized water and drying with a 5N nitrogen. After a thermal cleaning of the sapphire substrate at 600℃, a 100-nm-thick ZnO:Ga buffer layer was deposited at the rf power of 100 W on a double smoothed and cleaned sapphire substrate. The buffer layer sputtering was carried out at room temperature. The chamber deposition pressure was 1 Pa. Then a

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2-μm-thick ZnO:Ga film was deposited on the buffer layer using the rf magnetron sputtering technique at a substrate temperature 350℃. The ZnO:Ga film was annealed at 600℃ under a 1 Pa nitrogen pressure for 40 min and then cooled down under it.

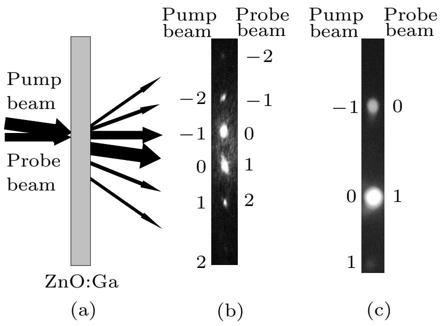

Fig. 1. (a) The diffraction geometry. (b) CCD image of the diffraction pattern of transmitted light with pump power at 7 mW and probe power at 7 mW. (c) CCD image of the diffraction pattern of transmitted light with pump power at 7 mW and probe power at 1 mW. The minus first-order pump and zero-order probe fall together, and so on, as indicated. The first-order pump and zero-order probe are detected by PMT.

The multi-pass amplified Ti:sapphire laser emitting 30 fs, 800 nm laser pulses at a repetition rate of 1 kHz was split into two beams. One was served as the probe beam and focused into a ZnO:Ga film with thickness 2 μm by a 10-cm focal-length lens. The other (pump beam) passed through a time-delay device and a $\lambda/2$ plate to control the path length and polarization of the pump beam, respectively. Both beams were focused by lenses before passing through the ZnO:Ga film. The centers of the pump and the probe beams spatially overlapped at the ZnO:Ga film at an angle of 12°. The probe beam was focused into the photomultiplier tube (PMT) while the first diffracted order of the pump beam was collected into another PMT. An infrared filter was used to remove the luminescence at about 395 nm on the ZnO:Ga film mixed in the detected beam. As the time-delay device moves, the two temporal signals can be simultaneously detected.

The pump and probe pulses form the transient grating induced by two-photon absorption of two interfering optical beams in the ZnO:Ga film.[31] The pump and probe beams were simultaneously diffracted by this transient grating. The diffraction geometry is shown in Fig. 1(a). Figure 1(b) shows the diffraction patterns imaged by a CCD with the power of both pump and probe beams fixed at 7 mW. From Fig. 1(b), the multi-order diffraction patterns can be clearly observed. The multi-order diffraction patterns are distributed symmetrically on both sides of the pump and probe beams in space. The minus first-order diffraction of the pump beam, which is mixed into the probe beam, is located in the mirror direction of the first order diffraction. The detection on the first-order diffraction of the pump beam, which is different from the transmitted direction of the probe beam, may be more direct and convenient to present the existence of the coherent artifact. When the probe power decreases down to 1 mW, only the first-order diffraction of the pump beam can be seen, as shown in Fig. 1(c). The minus first-order diffraction of the pump beam is always mixed into the probe beam.

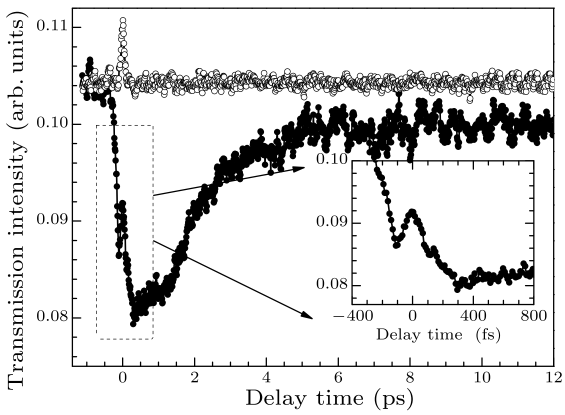

Fig. 2. Time-resolved profile of the first-order diffraction of the pump beam (open circles) and the probe transmission (closed circles). The pump power and probe power are set at 7 mW and 1 mW, respectively. The inset shows the enlarged map of the trace in the dashed rectangle.

To temporally investigate the relationship between the coherent artifact and the first-order diffraction of the pump beam, the time-resolved temporal profile of the first-order diffraction of the pump beam and the probe transmission were measured, in which the pump power and probe power were set at 7 mW and 1 mW, respectively. The results are shown in Fig. 2. We can see the dramatic variation of the transmission around zero delay time, which can be attributed to the strong coherent artifact. The inset shows the enlarged map of the trace in the dashed rectangle. The full width at half maximum (FWHM) of the coherent artifact signal is 110 fs, which is the same as that of the incident pulse autocorrelation. Namely, the coherent artifact can be observed only in the temporal overlapping region of pump and probe pulses. The rising part of the pump-probe trace consists of three processes. The process within the first 1 ps corresponds to the free carrier absorption, which implies the cooling of the hot carriers to a quasi-thermal equilibrium with time constant 1 ps. The slow decay process with the time constant more than 9 ps appears after 3 ps, which is attributed to spontaneous emission. The process from 1 to 3 ps represents the electron-hole plasma (EHP) radiation recombination, in which the carrier concentration should be higher than the Mott transition density of about 10^{18} cm^{-3}.[32] When the carrier concentration decreases below the Mott transition density, the slow decay process related to the spontaneous emission will be dominant.

The result indicated by open circles in Fig. 2 shows the time-resolved profile of the first-order diffraction of the pump beam. The first-order diffraction of the pump beam temporally presents near the zero time delay, the FWHM is also 110 fs. The first-order diffraction of the pump beam disappears while the pump and probe pulses are slightly separated temporally.

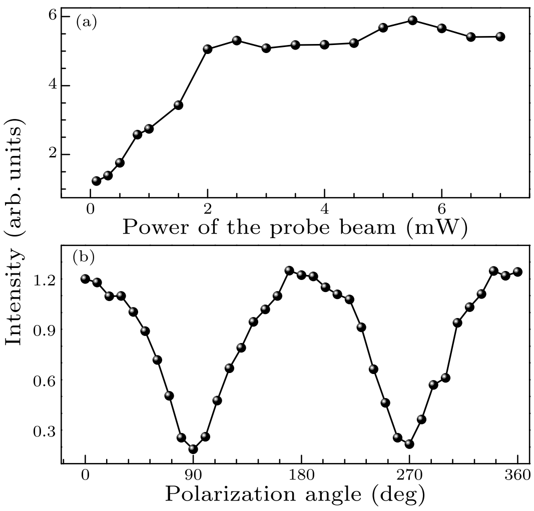

Fig. 3. (a) Dependence of the intensity of the first-order diffraction of the pump beam on the power of the probe beam. The pump power is set at 7 mW. (b) Dependence of the intensity of the first-order diffraction of the pump beam on the polarization angle between the pump beam and the probe beam.

The coherent artifact appears simultaneously with the first-order diffraction of the pump beam at zero time delay. In a degenerate pump-probe configuration, the first-order diffraction of the pump beam is the mirror component of the minus first-order diffraction of the pump beam. They both have the same frequency but different directions. The influence of the coherent artifact can be discriminated by directly inspecting the background-free first-order diffraction of the pump beam.

Since the transient grating is induced by two-photon absorption of two interfering optical beams, the interference of the pump and probe pulses can be weakened drastically by decreasing the probe power. Figure 3(a) shows the dependence of the intensity of the first-order diffraction of the pump beam on the power of the probe beam, in which the polarization angle between the pump and probe beams is set at 0°. It indicates that the diffraction intensity increases with increasing probe power when the probe power is below 2 mW. The diffraction intensity approaches its saturation value when the probe power is increased to 2 mW. The experimental results described above indicate that the coherent artifact in the pump-probe measurements can be suppressed by a reduction of the power of the probe beam.

Further, we also measured the dependence of intensity of the first-order diffraction of the pump beam on the polarization angle between the pump and probe beams, and the results are shown in Fig. 3(b). We can see that the dependence of the diffraction intensity on the polarization angle displays a periodic change. The change period of the diffraction intensity is π. The diffraction intensity presents strongest as the polarization angle of the pump and probe beams is set at 0° or 180°, and the diffraction intensity becomes weakest as the polarization angle of the pump and probe beams is set at 90° or 270°. The diffraction intensity can be eliminated by setting the polarization angle of the pump and probe beams to be orthogonal.

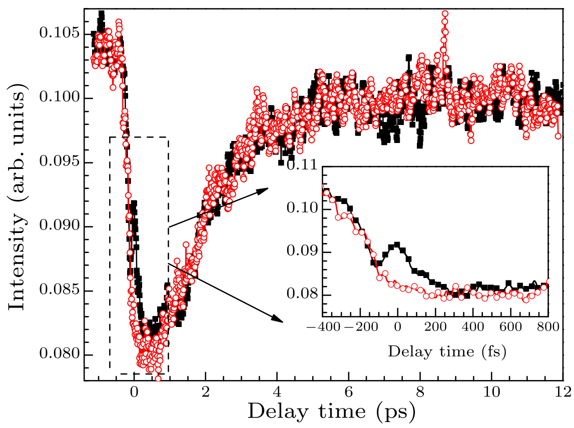

Fig. 4. Time-resolved profile of probe transmission with different probe powers. The pump power is set at 7 mW, and the probe power is set at 1 mW (closed circles) and 0.2 mW (open circles). The inset shows an enlarged region of the data in the dashed rectangle.

Figure 4 shows the time-resolved profile of the probe transmission. The polarization angle between the pump and probe beams is set at 0° and the pump power is set at 7 mW. The probe power is set at 1 mW and 0.2 mW, respectively. From the inset in Fig. 4, we can see that the coherent artifact for the probe power of 0.2 mW is eliminated. Generally, the coherent artifact can be eliminated by setting the orthogonal polarization between the pump beam and probe beam according to the above description. However, orthogonal polarization configuration can not be used in pump-probe measurements such as the optical Kerr shutter.[18] Therefore, for optical Kerr shutter probe-pump experiments, the coherent artifact can be eliminated by reducing the probe-to-pump power ratio.

In summary, the influence of the coherent artifact on femtosecond pump-probe measurement has been studied in semiconductor Ga-doped ZnO film. The coherent artifact mixed into the pump-probe signal can be directly inspected by detecting the background-free first-order diffraction signal induced by the interference between the pump and probe pulses. Experimental results show that by varying the polarization angle or adjusting the relative intensity between the pump and probe pulses, the coherent artifact can be eliminated from the pump-probe measurement.

References

[1] Wundke K, Pötting S, Auxier J, Schülzgen A, Peyghambarian N and Borrelli N F 2000 *Appl. Phys. Lett.* **76** 10

[2] Liu X, Liu W, Yin J, Qu J, Lin Z and Niu H 2011 *Chin. Phys. Lett.* **28** 034202

[3] Li F, Meng F, Feng W, Wang Sh, Tian H and Gong Q 2010 *Chin. Phys. Lett.* **27** 068202

[4] Elim H I, Ji W, Ma G H, Lim K Y, Sow C H and Huan C H A 2004 *Appl. Phys. Lett.* **85** 1799

[5] Tsai T -R, Chang C -F and Gwo S 2007 *Appl. Phys. Lett.* **90** 252111

[6] Gopinath J T, Thoen E R, Koontz E M, Grein M E, Kolodziejski L A, Ippen E P and Donnelly J P 2001 *Appl. Phys. Lett.* **78** 3409

[7] Sanchez S, De Matos C and Pugnet M 2001 *Appl. Phys. Lett.* **78** 3779

[8] Ye H, Wicks G W and Fauchet P M 2000 *Appl. Phys. Lett.* **77** 1185

[9] Sun C -K, Vallée F, Keller S, Bowers J E and DenBaars S P 1997 *Appl. Phys. Lett.* **70** 2004

[10] Lioudakis E, Othonos A, Dimakis E, Iliopouios E and Georgakilas A 2006 *Appl. Phys. Lett.* **88** 121128

[11] Si J and Hirao K 2007 *Appl. Phys. Lett.* **91** 091105

[12] Luo C W, Wang Y T, Chen F W, Shih H C and Kobayashi T 2009 *Opt. Express* **17** 11321

[13] Borri P, Romstad F, Langbein W, Kelly A E, M ørk J and Hvam J M 2000 *Opt. Express* **7** 107

[14] Vandeny Z and Tauc J 1981 *Opt. Commun.* **39** 396

[15] Palfrey S L and Heinz T F 1985 *J. Opt. Soc. Am.* B **2** 674

[16] Sanchez F 1992 *J. Opt. Soc. Am.* B **9** 2196

[17] Cundiff S T 2008 *Opt. Express* **16** 4639

[18] Yan L H, Yue J J, Si J H and Hou X 2008 *Opt. Express* **16** 12069

[19] Dogariu A, Xia T, Hagan D J, Said A A, Van Stryland E W and Bloembergen N 1997 *J. Opt. Soc. Am.* B **14** 796

[20] Qu S, Zhao Ch, Zhao Q, Qiu J, Zhu C and Hirao K 2004 *Opt. Lett.* **29** 2058

[21] Ravn J N 1992 *IEEE J. Quantum Electron.* **28** 315

[22] Kalt H, Lyssenko V G, Renner R and Klingshirn C 1985 *J. Opt. Soc. Am.* B **2** 1188

[23] Divakara Rao K and Sharma K K 1995 *J. Opt. Soc. Am.* B **12** 658

[24] Si J, Qiu J, Zhai J, Shen Y and Hirao K 2002 *Appl. Phys. Lett.* **80** 359

[25] Schneider Th, Wolfframm D, Mitzner R and Reif J 1999 *Appl. Phys.* B **68** 749

[26] Dean D R and Collins R J 1973 *J. Appl. Phys.* **44** 5455

[27] Wang C -R, Luo T and Lu Q -B 2008 *Phys. Chem. Chem. Phys.* **10** 4463

[28] Chudoba C, Nibbering E T J and Elsaesser T 1998 *Phys. Rev. Lett.* **81** 3010

[29] Conrad U, Gdde J, Jhnke V and Matthias E 1999 *Appl. Phys.* B **68** 511

[30] Lebedev M V, Misochko O V, Dekorsy T and Georgiev N 2005 *J. Exp. Theor. Phys.* **100** 272

[31] Liu H, Zhang H, Si J, Zhang J, Yan L, Wei X, Wen X and Hou X 2010 *Opt. Commun.* **283** 5203

[32] Takeda J, Jinnouchi H, Kurita S, Chen Y F and Yao T 2002 *Phys. Status Solidi* B **229** 877