Time Resolved Pump-Probe Spectroscopy

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Abstract

In this paper, the working principle of femtosecond pump-probe spectroscopy is introduced. A experimental setup to study the damage mechanism of some dielectric films is shown and how to use the experimental data to explain the physical processes happened in the film is explained.

Introduction

A principle figure of the pump-probe spectroscopy is shown in Figure 1. The pulse train coming out a laser is split into two pulse trains. One is used as the pump beam, by which the sample is exited. The pump leaves some time evolving change in the sample. Another beam is used to probe the change. There is a time delay between the pump pulse train and the probe pulse train. With the change of the delay between the two pulsed train, we can get a spectrum of reflectivity, absorption, luminescence, or Raman scattering of the probe after the sample. In this way, the change caused by the pump is studied.

The time resolution of the pump probe spectroscopy has a direct relationship with the pulse duration used in the experiment. Femtosecond laser pulse can make the time resolution of this technique goes to femtosecond scale. This enables researchers to know many quite fast dynamic processes happened when the pump pulse passes through the sample.

From Boyd's book\(^1\), the response time of the nonlinear refractive index from electronic polarization is in femtosecond scale, and that from molecular orientation is in picosecond scale. These kind of physical process can be reached by femtosecond pump-probe spectroscopy.

In the past, researchers mainly use femtosecond pump-probe spectroscopy get the dynamic behavior of electrons when the sample is excited by the pump pulse\(^2\). In reference 3, the vibrational and orientational dynamics of the OH-stretching mode of HDO is studied. These processes happen in several picoseconds. Drescher etc using a few-femtosecond visible light pulse and a synchronized sub-femtosecond soft X-ray pulse measure the lifetime of some core-excited atoms with attosecond resolution\(^4\). Ueda etc apply this technique in exploring the dynamic behavior of electrons exitons in low dimensional semiconductor nano-structures. They get the carrier multiplicatin information near zero time delay\(^5\).

Experiment

The experimental setup is shown in Figure 2. The laser pulse comes from a Ti:Sapphire laser amplifier. The center wavelength of the laser is at about 800 nm. The pulse duration of the pulse is about 50fs. The power is about 200mW with a repetition rate at 1KHz. The power from the amplifier can go up to 2W. The translation stage in the pump arm is driven by a stepper motor to change the delay between the pump and probe pulses. The step motor can move 0.9 degree in one step, which corresponding to 1.25 micron meter. Because the movement of the translation stage causes a two times change of the optical path, the change of the delay between the pump pulse and the probe pulse is 8.3fs. The focal length of
the positive mirror in the pump arm is 250mm. The focal length of the positive lens in the probe arm is 100mm. As a result, the beam radius of the pump beam at the sample position is about 2.5 times of that of the probe beam, if the sample is at the focal point of the two beams. The beam radius of the pump and the probe are about 50μm and 20μm. This is to make sure the area covered by the probe beam is also illuminated by the pump beam. The sample here are some dielectric films (e.g., hafnia) on a glass plate.

In the setup, three same detectors are used. The pump reference serves as a reference to the transmission and reflectivity to cancel the linear change caused by the vibration of the incident pulse power.

![Figure 2: Experimental setup.](image)

**Theoretical Analysis**

A typical result curve is shown in Figure 3. The peaks around zero time delay comes from the overlap of the pump and the probe pulses in the film and the substrate. This kind of “coherence spike” is due to cross-phase modulation, four-wave mixing and pump-pulse mediated multi-photon absorption for the probe pulse. Figure 3 (b) shows the transmission signal from an uncoated substrate for different excitation levels. Because the substrate has negligible contribution to the reflection change, the “coherence spike” in the reflection can be attributed solely to the film.

This kind of spike can be seen for all films. Therefore, we use the coherence spike in the reflection to determine the position of the zero time delay between the pump and the probe beams. To avoid difficulties in the data interpretation caused by the coherence spike, we analyze the data only for delays greater than 60fs.

The excitation of the film by the pump pulse is controlled by the local intensity inside the film, which in turn is the result of interference effects. The pump pulse forms a standing wave along the propagation direction Z of the pump pulse. As a result, the dielectric function \( \Delta \epsilon(z) \) is also spatially modulated, leading to an amplitude and phase grating.

Based on the Drude theory of electron-hole plasmas, we ansatz that the change of the dielectric function is proportional to the conduction band electron density, \( N_{\text{CB}}(z) \), generated by the standing wave pump field. The dielectric function can be written by

\[
\Delta \epsilon(z) = \Delta \epsilon_{\text{max}} \times \frac{N_{\text{CB}}(z)}{N_{\text{CB, max}}}, \quad \text{where} \quad \Delta \epsilon_{\text{max}} = \Delta \epsilon_r + i \Delta \epsilon_i.
\]
Δε_{max} is unknown, and N_{CB}(z)/N_{CB,max} represents the spatial profile of the excited carrier density in the direction normal to the film.

Assuming the film is divided into many slices along the direction of Z, we can represent the dielectric constant at the i_{th} slice with ε(z_{i})=ε_{0}+Δε(z_{i}). For a certain Δε_{max}, R(λ) and T(λ) can be obtained by applying the optical matrix formalism to the air-slices-substrate system. Integration over the pulse spectrum yields the reflection and transmission data that can be compared that from the experiment.

Figure 3: (a) ΔR=R_{0} and ΔT=T_{0} data for the Ta_{2}O_{5} film, measured at 60% (solid line) and 40% (dashed line) of the single pulse breakdown threshold F_{th} = 0.56J/cm^{2}. (b) Transient transmission signal of an uncoated substrate for two different excitation fluences relative to the single pulse breakdown fluence of Ta_{2}O_{5}.

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Figure 4: The retrieved dielectric function for the Ta_{2}O_{5} film for delays <2 ps, at an excitation density of 60% and 40% of the single-shot breakdown threshold.

Then using a computer program, we can get the best values of Δε_{r} and Δε_{i} at a given delay that minimize the deviation between the experimental and calculated ΔR and ΔT data. A retrieved dielectric
function for Ta$_2$O$_5$ film is shown in Figure 4. Different mechanisms have variant effect on the change of the dielectric function. Once we know that, we can further analyze which kind of mechanism is primary in the experiment.

**Conclusion**

Femtosecond pump-probe spectroscopy is a good dynamic analysis technique in femtosecond scale. In this paper, we show how we can use pump-probe spectroscopy to study the main physical mechanisms in the near damage region of the dielectric films.

**References**