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EVALUATION OF THERMAL AND ELASTIC PROPERTIES FOR SOLID SURFACES USING TRANSIENT REFLECTING GRATING METHOD

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ABSTRACT

Subnanosecond time-resolved transient reflecting grating (TRG) method has been developed and applied to various solid surfaces such as a DLC film on a multiplayer substrate and an Ar ion-implanted silicon. This method provides a depth profiling of thermal and acoustic properties by changing the observed depth which can be experimentally controlled, and has the least depth region of hundreds of nanometers. We propose a new microscopic method using the TRG method where the signal is measured at various delay times while scanning a sample. This gave a various kinds of images including thermal and elastic properties at local places in the surface and subsurface region. Further, for clarifying a fundamental mechanism of a heat generation, the TRG method has been improved to have a time resolution of hundreds of femtoseconds. In this time range, ultrafast carrier dynamics can be monitored and the energy transfer processes from carriers to phonons were detected clearly.

INTRODUCTION

Surfaces and subsurfaces of materials modified by means of ion implantation, chemical and physical vapor deposition, etc., sometimes show higher order structures such as gradient structures and multilayered structures. Their evaluation places some technical requirements on the measurement method being applied in manufacturing process, i.e., it should be noncontact, nondestructive, and *in situ*. Photothermal and related techniques can partially cover the requirements[1] and deduce thermal and elastic properties for thin films, multilayered films and surface-modified materials. In the latest device technology, device components get smaller and smaller to the size region of nanometers, and for example, thin films of several nanometers in thickness are practically used. Conventional photothermal methods cannot measure thermal or elastic properties for such thin films of nanometer thickness because the methods usually have the least depth resolution of several micrometers. Thus, measurement methods for evaluating thin films of nanometer thickness are required.

On the other hand, as the device size is reduced, the time of carrier transport between components also gets faster and faster to the time region of picoseconds. Ultrafast carrier transport generates heat in the nano-sized region, so that there are some local places with high temperature. To improve device

performance, the fundamental physical processes of such heat generation at a surface or interface must be clarified and
the temperature should be controlled. In order to clarify the origin of heat generation, the source process es, namely the photo-excited carrier dynamics should be studied.

For the last 10 years, we have developed new photothermal methods that allow sub-nanosecond time-resolved measurements and offer a nano-scale depth resolution. These methods are Transient reflecting grating (TRG) and Transient reflectivity (TR), and they were applied to various solid surfaces [2,3,4], solid-liquid interfaces [5,6,7,8], multi-layered films [9] and liquid surfaces [10]. In this paper, we will show some results obtained by applying the TRG method to thin films and multilayered films of nano-scale thickness. Also, a new type of microscope using the TRG method is proposed for investigating thermal and elastic properties at local places. Futher, the time resolution of the TRG method has been improved from sub-nanosecond to sub-picosecond, and the results are also shown about ultrafast carrier dynamics related to the fundamental processes of thermal generation.

Experimental

Schematic illustration of the principle for the TRG method is shown in Fig.1. In the TRG technique, two crossed pump pulses are incident at a solid surface and, as a result, the focused spot is irradiated with a pulse of an interference pattern. The complex refractive index at the spot changes due to a physical property change, or grating-patterned surface deformation occurs mainly due to a surface acoustic wave. After the pump pulses irradiation, a probe pulse is also incident there, and the complex refractive index change and the grating-patterned surface deformation are detected through the diffracted light of the probe light. A refractive index change due to photoexcited carriers is observed until several picoseconds, and that due to temperature rise and the following diffusion are observed from several picoseconds to nanoseconds. A surface deformation is detected in the time range of several nanoseconds.

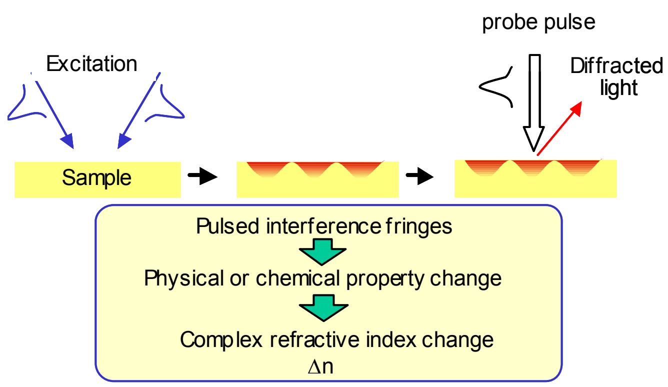

Figure 1 The principle of the transient reflecting grating method.

For measurement with a time resolution of sub-nanosecond, Nd-YAG laser (Pulse width :80 ps, Repetition frequency : 1 kHz, Wavelength : 1064 nm) was used as a light source. The wavelength was frequency-doubled to 532 nm. The pulse was separated into pump and probe pulses using a partial reflective mirror. The pump pulses were further divided into two pulses by a half mirror. The two pump pulses were crossed and irradiated onto the same spot of the sample surface, to coincide in time to form an interference pattern. The probe pulses were also incident at the spot after passing through a computer controlled optical delay line. The diffracted signal of the probe pulses were detected with a photomultiplier, and observed by a computer after averaging the signal with a box-car integrator.

In measurements with a time resolution of 200 fs, a regeneratively amplified titanium sapphire laser laser (CPA-1000; Clark-MXR Inc.) was used as a light source. The pulse train wavelength was 800 nm with a repetition rate of 1 kHz and pulse width of 200 fs in full width at half maximum. Only the pump pulses were frequency doubled to a wavelength of 400 nm. The probe pulses remain 800 nm.

Results and Discussion

TRG responses measured with the sub-nanosecond time resolution is shown in Fig.2. The signal had a small peak just after photo-excitation at the almost same time as incident pulse. The following signal shows an oscillating decay consisting of an exponential decay and an oscillating decay. The signal was separated to each component, which is shown in the right side of Fig.2. Considering the time range observed, the first peak corresponds to a refractive index change due to photo-excited carriers, and the exponential decay and the oscillating decay mean a thermal decay and a surface acoustic wave (SAW), respectively. It is very difficult to discuss the photo-excited carrier dynamic because the dynamics is considered to be faster processes than the used pulse width. About the thermal component, the decay occurs due to a thermal diffusion parallel to the interface, which disappears the temperature distribution like a grating pattern. Thus the decay time corresponds to the time during which heat diffuses for the length of the grating spacing. The grating spacing, $\Lambda$is expressed as $\Lambda = \lambda/2\sin(\theta/2)$, where λ is the wavelength of the pump pulses, and θ is the intersection angle of the two pump pulses. From the thermal decay time and the value of Λ, a thermal diffusion coefficient can be calculated, and this value is a property value in the depth region of Λ, which can be controlled by changing θ and has a typical length of 1 – 10 μm. Then, this signal provides a depth profiling of the thermal diffusion coefficient. The acoustic oscillation of the SAW originates in thermal expansion due to heat generation with a grating pattern. The wavelength of SAW must agree with the grating spacing, so that the SAW with a controllable wavelength can be generated and detected. Since a SAW have an elastic information in a surface region of the wavelength, this acoustic signal also offers a depth profiling of elastic properties. Actually the depth profiling of thermal and acoustic properties were applied to 10 keV nitrogen ion-implanted DLC films (40 nm) on multiplayer substrates, with Λ from 0.86 to 3.96 μm [9], and for 300 keV Ar ion-implanted Si wafers (ion-projected range, 0.31 ∓ 0.09 mm) with Λ from 1.65 to 3.66 μm [11]. Effective surface thermal conductivities and elastic constants were obtained. Effective thermal conductivities approached the real values for DLC films as the observed effective thickness is decreased, and the obtained value measured with the least depth resolution is good agreement with the real one. For the ion-implanted Si wafers, measurements of effective elastic constants revealed that the ion-implantation hardened the surface layer, though the surface-hardening mechanism is not clear yet.

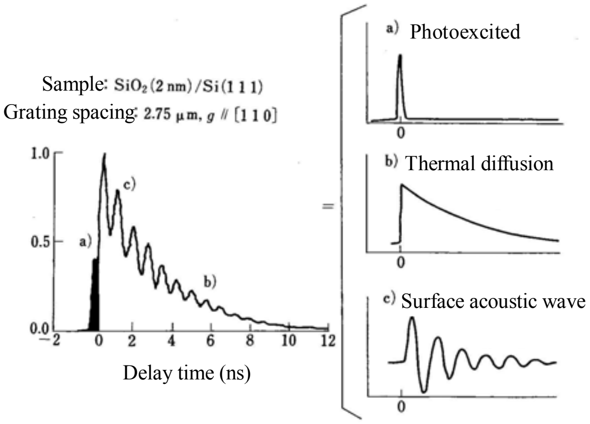

Fig. 2 A response of transient reflecting grating for a silicon (111) surface with a time resolution of 100 picoseconds. Three components comprising the response are also shown on the right side.

Furthermore, it is expected that this TRG method is used to investigate in-plane local properties for surface analysis in the depth range of nanometers because thermal and elastic properties at local places are very important for inhomogeneous surfaces including a partially ion-implanted region, structural defects, impurities and so on. The TRG method gives such properties at the focused spot of a probe light with a diameter of several micrometers. Imaging of the TRG signal can be obtained by measuring it while scanning a sample surface. This new type of imaging method features in providing a set of time-resolved images by changing delay times. By using the images, images for thermal relaxation rate or surface acoustic wave frequency can be deduced. We call this new microscope *Sazanami* imaging. *Sazanami* is a Japanese word that refers to the small rippling waves caused by a light wind blowing over a water surface. TRG looks like the *sazanami* ripples on a material surface, but are generated by light.

Time-resoled sazanami images are shown in Fig. 3. The sample was a Si wafer, partly 300 keV He-ion-implanted (dose, $10^{15}/cm^2$). The initial parts of the TRG responses for both non-implanted and implanted regions are shown at the top. The two arrows at t=0.0 and 0.5 ns indicate the photoexcited carriers' peak and the first acoustic peak, respectively. By using the two delay times, two different sazanami images were obtained. Although no sturucture apart from dust appears in the dark field image, the ion-implanted region is distinguishable in the sazanami images. The dark area in the 0.0 ns image indicates a small number of carriers. The bright area in the 0.5 ns image means that a large amount of heat is generated. The results are simple that, ion-implantation makes the carrier density small and heat generation large as a result of acceleration of carrier recombination. With time-resolved imaging we can easily grasp such dynamics.

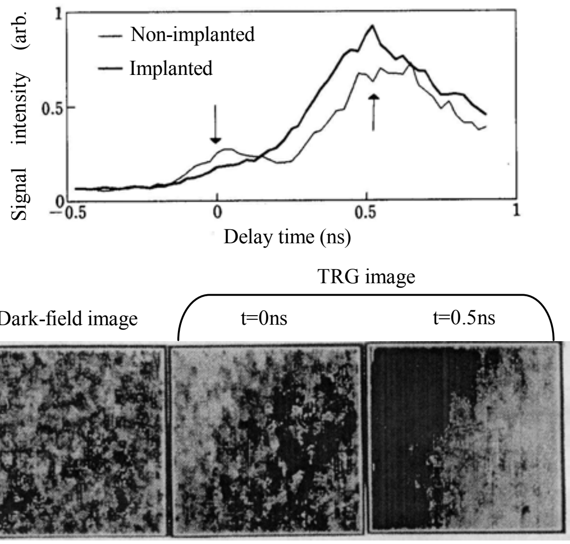

Fig.3 Top: TRG responses at He-implantated and non-implanted place. Bottom: A dark-field image and time-resolved TRG images of a partially He ion-implanted silicon.

Next, the photoexcited carrier dynamic was focused on. The dynamics was observed in the first peak of the sub-nanosecond time-resolved TRG signal. To clarify the temporal response, the TRG measurement with the time resolution of 200 fs was applied to the same sample. The TRG response is shown in Fig.4. The signal rose just after photo-excitation and decayed in 2ps. The photo-excited carriers lose their energy due to scattering by phonons, that is, an energy transfer from photo-excited electrons to phonons. The process is the origin of a temperature rise. In the literature [12], it was reported that the scattering due to phonons occurs about a few picoseconds. Thus it was concluded that the TRG signal in the picoseconds time range includes information on the origin of heat generation, that is, an energy transfer from photo-excited electrons to phonons.

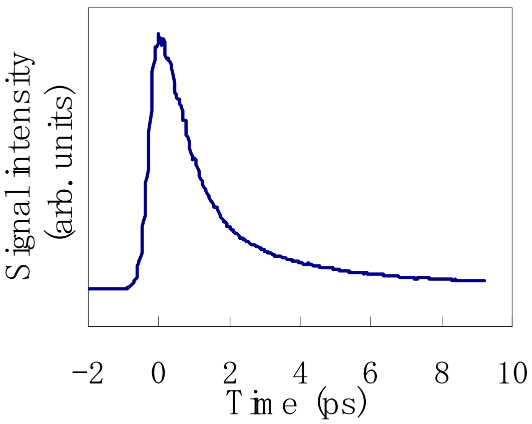

Fig. 4 A response of transient reflecting grating for a silicon surface with a time resolution of 200 femtoseconds.

Conclusion

The TRG method was applied to various solid surfaces in the time range from femtoseconds to nanoseconds. In a measurement with a time resolution of sub-nanoseconds, the signal gave information on thermal diffusion and SAW. This method provides a depth profiling of thermal and acoustic properties by changing the experimentally controllable observed depth. Also, we propose a new microscope using the TRG technique and it provide a thermal and elastic characterization at local places with a spatial resolution of several micrometers. Recently developed ultrafast time-resolved measurement with 200 femtosecond time resolution offers an investigation of an initial heat generation, that is, an energy transfer from electrons to phonon.

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