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Development of non-contact and non-destructive method for estimating the thermal properties by using the laser heterodyne photothermal displacement method

光ヘテロダイン光熱変位法を用いた熱物性の非破壊・非接触評価技術の開発

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1. Introduction

The figure of merit is a quantity used to characterize the performance of thermoelectric devices.[1)] The smaller the thermal conductivity (κ) becomes, the higher the performance is. The decrease in κ by introducing the nanostructures has been reported.[2,3)] The 3ω and time domain thermoreflectance (TDTR) methods were used to measure κ of thin films and nanostructures.[4,5)] But, they are destructive evaluation methods that require the formation of a heating electrode or Al reflective film on the sample. Destructive measurement may change the condition of the sample. In addition, it is difficult to measure the anisotropy of κ using those methods.

We have evaluated the thermal properties of Si-nanopillars/SiGe composite films[6)] utilizing the frequency-dependent piezoelectric photothermal method.[7)] The composite film is supposed to have anisotropic κ. We found that the observed signal intensity was affected by the contact condition between the detector and sample, thereby complicating quantitative measurements. Hence a noncontact and nondestructive method that can measure anisotropic thermal properties is necessary.

We developed a laser heterodyne photothermal displacement (LH-PD) method to evaluate κ by noncontact detection of thermal expansion of the sample surface caused by heat generation after light absorption using a heterodyne interferometer.[8)] The LH-PD has the potential to measure the anisotropy of thermal properties by controlling the positions of excitation and probe beams. As a preliminary step to measuring the anisotropy, we measured the isotropic thermal conductivity in this study. It was applied to the Si and GaAs bulk samples to confirm the measurement method, and κ is estimated by fitting with the theoretical calculations.

2. Experimental Procedure

When a modulated excitation beam was irradiated on the sample surface, the light absorbed area was periodically heated owing to the nonradiative recombination of photoexcited carriers. The following surface displacement was measured by the heterodyne interferometric method. The He-Ne laser (632.8 nm) was used as the probe beam and a laser diode (808 nm) was used as an excitation beam. The LH-PD can observe the time variation of displacement with an accuracy of subnanometer order. The thermal diffusion length (L) varies as $L=(2\kappa/\rho c\pi f_{ex})^{-1/2}$ where ρ is the density, c the specific heat, and f_{ex} the frequency of excitation beam. Thus, κ can be estimated from the time and frequency dependence of surface displacement. The f_{ex} was changed from 200 to 7000 Hz.

The theoretical calculations were performed by the finite element method by COMSOL Multiphysics considering the heat diffusion equation and solid mechanics in cylindrical coordinates. We applied the Robin boundary conditions in the light-illuminated surface and the side of the cylinder. The Dirichlet boundary condition was applied for the opposite side of the light-illuminated surface. κ is estimated by reproducing the displacement.

The *n*-type Si and semi-insulating (SI) GaAs substrate samples were prepared. Since the physical properties of Si and GaAs are widely reported, we assumed that they are appropriate materials to confirm the validity of our experiments and model calculations.

3. Results and Discussion

Figure 1 shows experimental and calculated time variations of surface displacement. Solid squares and circles were the experimental results of *n*-Si and SI-GaAs, respectively. Surface displacement was increased as soon as the excitation beam was turned on and decreased when the excitation beam was turned off. They were well reproduced by the theoretical calculations using literature values.[9)] The calculation showed that the smaller κ becomes, the larger the displacement and

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the slower the time variation are. This implies that κ can be easily estimated from the time variation of the surface displacement. Since the decay curve of the μ-TDTR follows a single exponential function,[5] a time variation of the surface displacement was attempted to be fitted by a single exponential function. However, present results could not be reproduced with a single exponential function. As a result, it could be fitted with two exponential functions $\exp(-t/\tau_f)+\exp(-t/\tau_s)$, where t is time and τ_f and τ_s are the time constant for fast and slow components, respectively. Since the τ_f and τ_s become longer when κ is smaller, both components are the κ-related time changes of the surface displacement. However, it is unclear what τ_f and τ_s mean and need to be discussed.

Figure 2 shows f_{ex} dependences of surface displacement of n-Si and SI-GaAs. The maximum values in the time variation were adopted. They were decreased as increasing f_{ex}, whereas that of Si showed a constant value in low f_{ex}. Although there was discrepancy in the high f_{ex} region, the experimental and calculated results generally accorded for both samples. In this figure, f_{ex} dependences of L also plotted. The gradient of L did not change when κ was changed. On the other hand, the experiments and calculations showed that when κ becomes smaller, its gradient becomes steeper. Hence, the frequency dependence of the displacement could not be discussed only in terms of the L. In addition, we could not define the contributions of τ_f and τ_s to the present f_{ex} dependence. To estimate exact κ from the time variation and f_{ex} dependence of the surface displacement, further analysis concerned with the heat generation, diffusion, and following the thermal expansion of the material is needed.

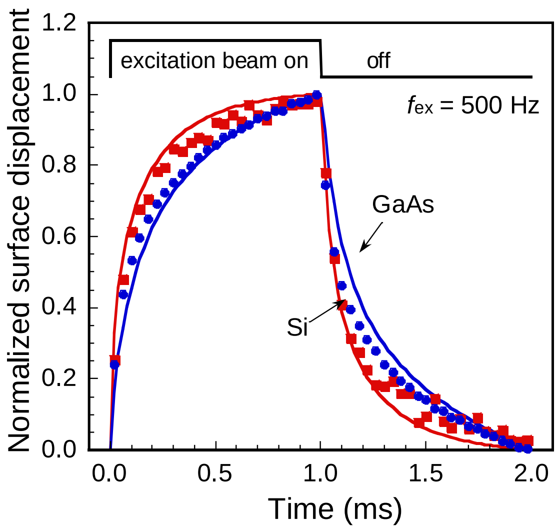

Fig. 1 Time variation of surface displacement. Surface displacement was normalized. Solid curves and symbols are calculated and experimental results, respectively.

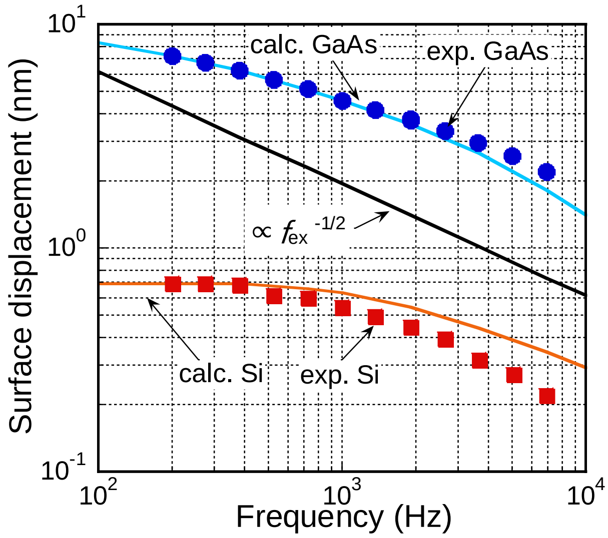

Fig. 2 Frequency dependence of surface displacement. Solid curves and symbols are calculated and experimental results, respectively.

4. Conclusion

We constructed the LH-PD method for detecting surface displacement. The calculation well reproduced the observed time variation of the surface displacement. In addition, the experimental and calculated results for f_{ex} dependence generally accorded. However, the physical meanings of τ_f and τ_s, estimated from the fast and slow components were not clear at present.

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