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Precise Measurement of Elastic Constant of Isotopically Controlled Diamond Thin Films using Brillouin Oscillation Method

ブリルアン振動法による同位体比制御ダイヤモンド薄膜の弾性定数の精密計測

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1. Introduction

Elastic constants are strongly relevant to atomic-bond strength. Thus their information is crucial for not only practical purposes but also condensed matter physics. Especially, diamond shows the highest stiffness in every natural material as well as other remarkable mechanical properties. Recent advances in synthesis technique allow the synthesis of highly pure diamond thin films, so that diamond comes to be used for more diverse applications, and allow the synthesis of isotope-controlled diamond, so that the isotope effects of diamond come to be studied experimentally. We expect the significant isotope effects of diamond because of light atomic mass and large mass ratio between the isotopes for carbon. In fact, 50% increase in the thermal conductivity was indicated by controlling the isotopes in diamond [1]. However, the mechanism of the isotope effects has not been clarified, so reliable experimental results are definitely needed.

Concerning the isotope effect on the elastic constants, the previous paper reported that there was no significant effect in ^{13}C enriched single crystal bulk diamond specimens within the experimental accuracy [2]. However, it is difficult to synthesize highly pure ^{13}C enriched single crystal bulk diamond largely enough to measure the elastic constants using existing methods. Thus, the reported result presumably doesn't show the whole isotope effect on the elastic constants. As contrasted to the bulk diamond, highly pure and isotopically controlled diamond can be synthesized in terms of thin films. However, due to the difficulties in measuring the elastic constants of thin films, the measurement has not been conducted. In this report, we measure the out-of-plane elastic constant of highly pure and isotopically controlled diamond thin films using Brillouin oscillation method.

2. Brillouin oscillation method

Brillouin oscillation can be detected by the picosecond ultrasound technique, and it provides us with the out-of-plane elastic constant of thin films through interaction between probe light and a propagating ultrasound pulse along the thickness direction. Differing from Brillouin scattering method, it can precisely determine the elastic constant of a thin film formed on a substrate, because the probe light pulse interacts with ultrahigh-frequency (~200 GHz) coherent phonons. The probe light is backward diffracted by the acoustic wave, and the diffracted light causes the interference with the surface reflected probe light, causing the oscillation in the total reflectivity of the probe light, so-called Brillouin oscillation. The oscillation frequency is identical to the acoustic-wave frequency causing the diffraction of the probe light, from which the out-of-plane stiffness is obtained. A typical Brillouin oscillation signal is shown in **Fig. 1(a)**.

3. Experiment

Three specimens were prepared. They are isotopically controlled homoepitaxial diamond thin films on a (100) substrate. Their ^{13}C concentrations are 0%, 25% and 50%. The substrate was synthesized by high pressure high temperature method and it has natural isotopic ratio, and the films were deposited on the substrate by the microwave plasma-assisted chemical vapor deposition method. The film thickness is about 3 μm and a 20 nm Al thin film is deposited on the specimen surface as the acoustic-wave generator.

We use Ti-Sapphire pulse laser with duration of 140 fs and repetition rate of 80MHz. The wavelength of pump light is 800nm, and that of the probe light is 400nm. In **Fig. 1(b)**, we calculated the change in the Brillouin-oscillation frequency using FFT with a short FFT gate to estimate the interface between the film and the substrate. The result of conducting FFT for the signal of the film and the substrate separately is shown in **Fig. 1(c)**.

4. Result & Discussion

With the optics we originally developed, we succeeded in determining the elastic constant of the

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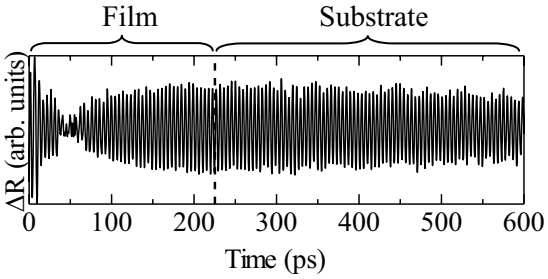
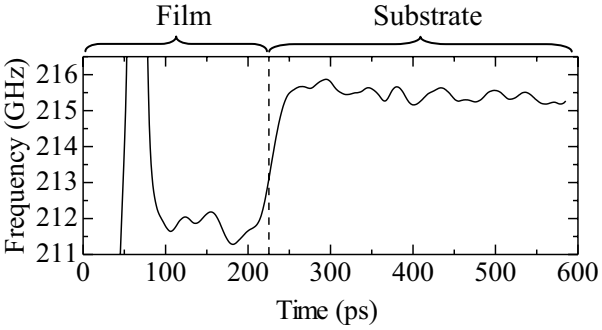
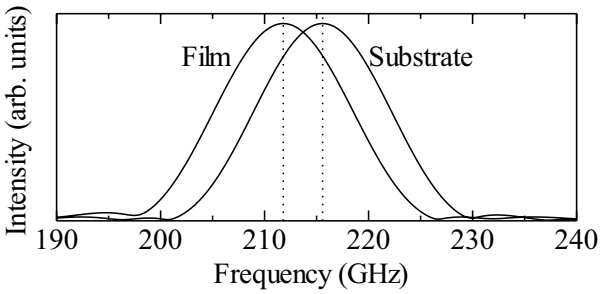


(a) Observed Brillouin oscillation.



(b) Brillouin-oscillation frequency shift versus propagating time of the ultrasound from the surface of the film.



(c) Fourier spectra of Brillouin oscillations in the film and the substrate derived from Fig. 1(a). Dotted lines represent center axes calculated from the Lorentzian-fitting procedure.

Fig. 1 The results of measurement and analysis in a specimen, whose film is composed of ^{12}C and ^{13}C in equal parts. In (a) and (b), a dashed line represents the interface between the film and the substrate.

thin-film diamond with accuracy of 0.2%. As the result of the measurement, the elastic constant of the diamond film increases by 0.6% as the ^{13}C concentration increases from the natural abundance to 50%. The enhancement is more noticeable than that of Ref. [2] with the result relating to the natural diamond being in good agreement with the reported one in Ref. [2]. So we also succeeded in observing the isotope effect on the elastic constant experimentally. Theoretically, we speculate that the mechanism of the isotope effects on the stiffness is similar to that of temperature reduction effects. Both would be attributed to the reduction of lattice vibrational amplitudes. With this hypothesis, we predict the change rate of the elastic constant. Firstly, from Ref. [3] we estimate the change rate of the lattice constant caused by the change of the ^{13}C concentration from natural abundance to be 0.25%. Secondly, from Ref. [4] we estimate temperature reduction of room temperature causing the same rate change of the lattice constant as described above. Finally, from Ref. [5] we calculate the corresponding change rate of the elastic constant to be 0.05%, caused by the estimated temperature reduction. In our measurement, the elastic constant increases by 0.4% as the ^{13}C concentration increases from the natural abundance to 25%. As the result, an indefinable difference appears between the change rates of the elastic constant, and it suggests that there are some differences between the mechanism of the isotope effects and that of the temperature reduction effects. We will able to observe the difference experimentally by conducting the same measurement at low temperature and the experimental result will be useful to clarify the isotope effects.

5. Conclusion

We succeeded in measuring the out-of-plane elastic constant of isotopically controlled diamond thin films formed on diamond substrate precisely and observing the systematic isotope effect on the elastic constant experimentally. In the range of ^{13}C concentration of 0% to 50%, the elastic constant increases with the concentration rising. The enhancement of the elastic constant in the specimens is much larger than both the reported one [2] and the estimated one by taking account of temperature reduction. We can expect to get information which is useful to clarify isotope effects by conducting the same experiment at low temperature.

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