High-resolution Brillouin spectroscopy with angular dispersion-type Fabry-Perot interferometer and its application to a quartz crystal

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Although the multichannel Brillouin spectroscopy with an angular dispersion-type Fabry-Perot interferometer (ADFPI) becomes a powerful tool for quick measurements, its resolution and contrast are not enough for the study of single crystals. A highly sensitive multichannel detector enables the ADFPI to use a solid etalon with high reflectivity (99.5%); hence, the high resolution and the high contrast of a spectrum are achieved. The finesse, the inverse of the resolution, reaches 100 with a 10 mm diameter of aperture size. The highest finesse of 140 is obtained by using a smaller diameter of 2 mm. The accuracy is examined by the measurement of a quartz crystal. The improvement in the resolution and contrast enables investigations of weak attenuation in a quartz crystal. The elastic anomaly of the α-β transition of a quartz crystal is clearly observed both in sound velocity and attenuation. From the elastic constant c_{11}, the critical parameter $K=0.76$ is determined. © *2007 American Institute of Physics.* [DOI: 10.1063/1.2753593]

Brillouin scattering is a noncontact and nondestructive probe of investigating elastic properties of materials. Brillouin spectra provide the wide variety of information about thermodynamic and acoustic properties in condensed matters.[1] Because of a development of Fabry-Perot interferometer (FPI), Brillouin scattering has been a valuable probe in examining the elastic properties, and significantly improved by the invention of monochromatic laser light sources. The earliest instrument of FPI was an angle dispersion type which combined with an air gapped etalon and a photographic plate detection. Further progress has been accomplished by an advent of a photomultiplier detector with a mechanical scanning FPI. The most sophisticated tandem multipass FPI invented by Sandercock *et al.* has measured an overlap-free Brillouin spectrum in a wide frequency range.[2] However, the conventional scanning FPI requires a long time acquisition and more that the interferometer is mechanically unstable due to its mechanical scanning procedures.

Most spectroscopic instruments, such as Raman spectrometer, have been applied to multichannel detectors and received benefits of short time acquisitions. On the other hand, Brillouin scattering spectroscopy is behind in the application of area detectors due to its complicated optical systems and difficulties of alignment. Therefore, the new technique which investigates the high resolution both in time and frequency has been anticipated. To achieve the shorter acquisition time and stable spectrometer, evolutions of a nonscanning angular dispersion-type Fabry-Perot interferometer (ADFPI) have been the best solution.[3–5] The progress has been achieved by a stable solid etalon combined with a multichannel detector. The advantage of the ADFPI is that all the transmitted light can be collected simultaneously. It enables us to obtain spectra at one time, and many authors focused on this simple spectrometer for several applications.[7–11] Although many applications have been carried out, it is still difficult to investigate the crystals due to the low finesse and contrast of the ADFPI system.

The solid etalon is strongly free from the instability of spectrometer, and the highly sensitive two-dimensional detector leads to the substantial improvement in acquisition time of the light detection.[3] By using a high quantum efficiency line or area detector such as a photodiode array or a charge coupled device (CCD), high reflectivity solid etalon can be applied. By using a high reflectivity solid etalon, it is possible to obtain the high resolution and contrast Brillouin spectra of crystals. In the present study, we have applied the high resolution ADFPI system for a quartz crystal. The α-β phase transition between α and β phases are known, and they have attracted many physicists.[13,14] Since the high quality quartz crystals are commercially available, they are a quite good sample to evaluate the ADFPI system.

The schematic diagram of the ADFPI is shown in Fig. 1. The instrument is a combination of the solid etalon and the CCD detector. A diode-pumped solid-state laser (DPSS532, Coherent) was used as a light source, operating at a single mode of 532 nm with 150 mW. The 5 MHz linewidth, and it is negligibly narrow compared with the general instrumental resolution of Brillouin spectrometers. The focal length of lens L1 is 100 mm, the incident laser beam is focused into a sample, and all lenses applied in ADFPI were achromatic. The scattered light from the sample was collected by lens L2a of 100 mm focal length at the right angle scattering geometry. To eliminate the stray light, a slit was used between the lenses of the same focal length of L2b and L2c ($f=200$ mm). In addition, a bandpass filter was employed to avoid passing the undesired light. The solid etalon of 30 GHz free spectral range (FSR) (reflectivity of 99.5% at 532 nm, SLS optics) was used, and the degree of flatness was better than $\lambda/100$. The solid etalon was made of a single fused silica glass plate, and the mirror was soft-coated dielectric materials. The installation of an area reducer (AR) in front of the etalon could improve the spectral profile and the

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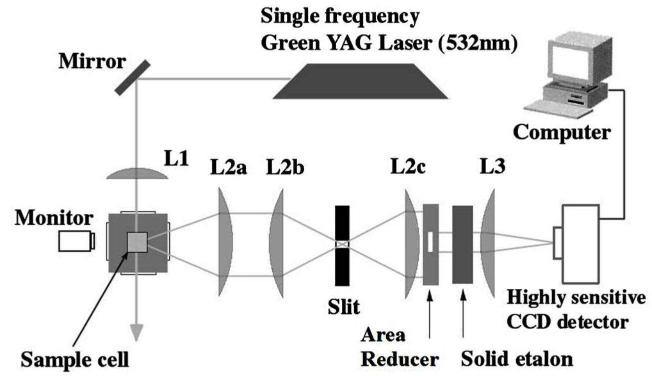

FIG. 1. Schematic diagram of the experimental setup. The mechanically stable solid etalon has been combined with the highly sensitive CCD detector. The AR has been set in front of the etalon for selecting the high flatness area of the etalon and lowering the kinetic broadening effect.

finesse because the AR lowers kinetic broadening and obtains better flatness by choosing the local area of the solid etalon. This is due to the rounded surface of etalon which has better flatness at the center of the etalon. The transmitted light through the etalon was focused onto the CCD detector (AP32E-2, Apogee) by lens L3 ($f=800$ mm). It has a full frame resolution of 2184×1472 with a pixel size of 6.8 $\times6.8\ \mu m^2$. The sample was an optical grade polished quartz crystal (Furuchi). Its size was $6\times7\times8\ mm^3$ along the $X+Y$, $X-Y$, and Z axes, respectively.

The temperature of the sample was controlled by a tubular furnace. The temperature was monitored by a chromel-alumel thermocouple attached onto the sample, and the temperature stability was less than 1 K. The acquisition time of spectra was 120 s in the entire temperature range.

The acquired spectra show nonlinear intervals of FSR because of the angle-dispersive acquisitions. These spectra must be linearized at first, and the linearization procedure is explained in previous work.[4] The transmitted light has a finite spectral width, and generally it is defined as the width at half intensity, called full width at half maximum (FWHM). The sharpness of the fringes increases when the higher reflectivity of the etalon is employed. The ratio of the FSR and obtained spectral width of the peak is defined as finesse, which is the inverse of the resolving power. The inverse of resolution is expressed as $F_R=\pi\sqrt{R}/1-R$, where R is the reflectivity of the etalon. The value of F_R is called reflective finesse. There are three types of finesses: reflective finesse F_R, flatness finesse F_f, and pinhole finesse F_p. The sum of these finesses is called total finesse F_T, which is derived by $1/F_T^2=1/F_R^2+1/F_f^2+1/F_p^2$. The resolving power is limited by these finesses. The finesse of the ADFPI with a 15 mm aperture size of the AR was slightly less than 100, and the finesse increases with decreasing aperture size of the AR, as shown in Fig. 2(a). The finesse reaches 100 with a 10 mm diameter and increases to 140 with decreasing AR size. To take into account the fact that a conventional scanning FPI system does not easily obtain a high finesse above 100, the resolution of the ADFPI is quite notable. To compare with the previous work, the present ADFPI can use a wider aperture size.[12] It has a great advantage in reducing the acquisition time by taking a larger quantity of light from the sample. Another important parameter of contrast C is defined by the ratio of maximum and minimum light intensity. The contrast is expressed as $C=I_{max}/I_{min}=(1+R)^2/(1-R)^2$. It indicates the cability of the interferometer to measure inelastic light scattering. The high contrast is very important in measuring Brillouin peaks when elastic scattering is intense, such as a spectrum of a crystal. Thus, the present high reflectivity etalon drastically raised the contrast of the ADFPI. The calculated values of contrast are 17 512 and 159 201 for reflectivities of 98.5% and 99.5%, respectively. As shown in Fig. 2(b), the contrast of the spectra is improved ten times larger by increasing the reflectivity from 98.5% to 99.5%. Our previous works of Refs. 3–6 and 12 employed a 98.5% reflectivity of the solid etalon, while in the present study, a reflectivity of 99.5% is applied. Therefore, both the resolution and the contrast of the ADFPI are significantly improved.

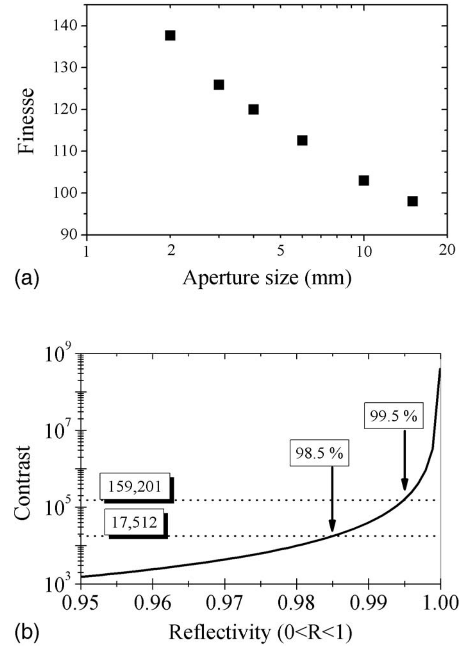

FIG. 2. (a) Aperture size dependence of obtained finesse. (b) A calculated curve of contrast as a function of reflectivity.

Figure 3 shows the Brillouin spectra of the quartz crystal at room temperature with the FSR of 30 GHz in comparison with the spectrum acquired by a Sandercock-type spectrometer. The weak intensity of Brillouin scattering light was effectively accumulated by using the ADFPI with the high re-

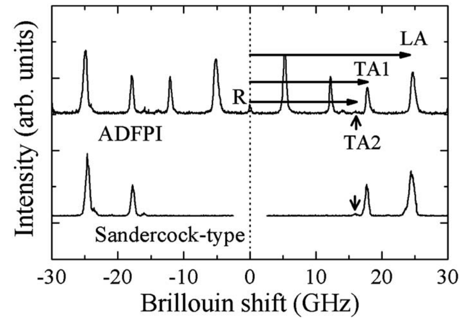

FIG. 3. Brillouin spectra of a quartz crystal at room temperature using the ADFPI with FSR of 30 GHz and using the Sandercock-type spectrometer. R denotes the elastic scattering (Rayleigh) component. The abbreviations of LA, TA1, and TA2 denote the Brillouin components scattered by the longitudinal acoustic phonon and two transverse acoustic phonon, respectively.

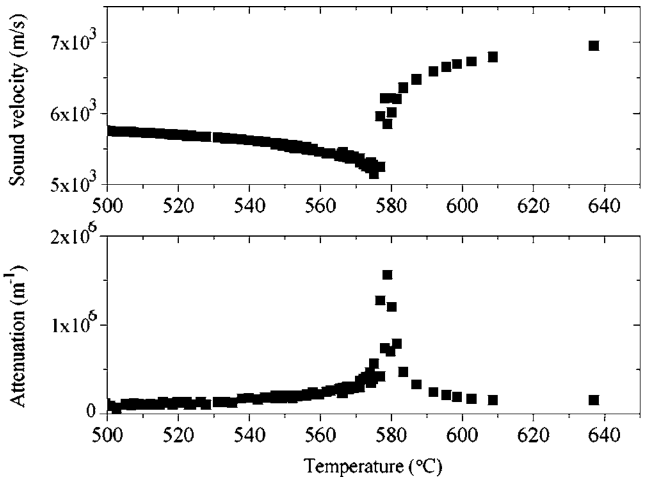

FIG. 4. Temperature dependences of sound velocity and attenuation of LA in a quartz crystal along the [010] axis.

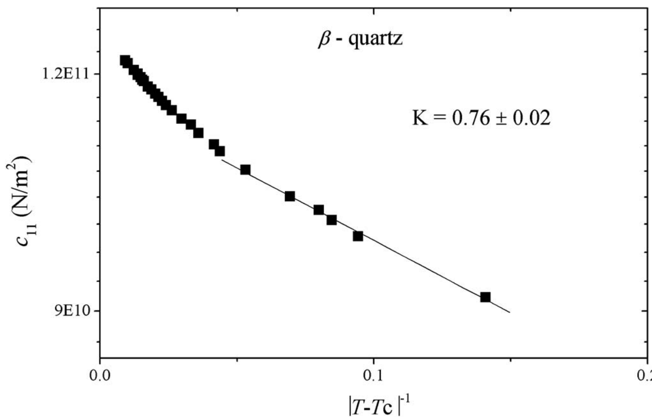

FIG. 5. The plot of c_{11} against $|T\text{-}T_c|^{-1}$. The value of $K=0.76\pm0.02$ is obtained from the slope.

flectivity solid etalon and the highly sensitive CCD. The acquisition time was remarkably reduced by using the ADFPI, and it was one-tenth of the Sandercock-type spectrometer. It shows one longitudinal acoustic (LA) and two transverse acoustic (TA1 and TA2) modes. The Brillouin shift and FWHM have been determined by the Lorentzian fitting. Sound velocity V is obtained from the Brillouin shift ν $(=qV/2\pi)$ and the scattering wave vector q $(=2\pi n \sin(\theta/2)/\lambda)$, where the notations n, λ, and θ are the refractive index of the medium, the wavelength of the laser, and the scattering angle, respectively. The FWHM is related to the attenuation of sound, γ $(=\pi\text{FWHM}/V)$. The temperature dependence of V and γ of the LA component is shown in Fig. 4. The quartz crystal undergoes a phase transition at around $T_c=573$ °C from its low temperature α phase with point symmetry D_3 to its high temperature β phase with point symmetry D_6.[13] In the vicinity of T_c, the anomaly of temperature dependence in sound velocity and γ was clearly observed. The γ becomes quite large around T_c because of the unstable lattice vibrations during the structural transition of the crystal. Due to the improvement of the resolution and the contrast of the ADFPI, it establishes accurate measurements of the sound velocity and the attenuation of a quartz crystal. The anomaly of the longitudinal wave was especially large. In addition, one transverse mode disappeared above T_c. It is attributed to the change of the crystal structure to D_6 at high temperatures. The observed T_c was 574 °C, which promises that the elastic measurements using the ADFPI are sensitive to the structural phase transitions. The critical parameter is calculated as $c_{11}=c_{11}(\infty)+A(T-T_c)^{-K}$, where K denotes the critical parameter. The slope shown in Fig. 5 gives the value of K, and we determined $K=0.76\pm0.02$ by a least squares fit.

In conclusion the high reflectivity of a solid etalon enables the ADFPI to achieve the high spectral resolution and the high contrast. The finesse, inverse of the instrumental resolution, is achieved up to 140. The calculated contrast exceeds 10^5, which is much larger than the previous works of the ADFPI. The ADFPI makes the Brillouin scattering measurements for crystals much simpler and easier. By using the multiplex detection system, the stable and the nonscanning ADFPI gives the rapid investigation of Brillouin scattering compared to the conventional scanning one.

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