A broadband spectroscopy method for ultrasonic wave velocity measurement under high pressure

Zhigang Wang, Yonggang Liu, Wei Song, Yan Bi, and Hongsen Xie

Citation: Review of Scientific Instruments **82**, 014501 (2011);
View online: https://doi.org/10.1063/1.3518953
View Table of Contents: http://aip.scitation.org/toc/rsi/82/1
Published by the American Institute of Physics

A broadband spectroscopy method for ultrasonic wave velocity measurement under high pressure

Zhigang Wang,[1,2,3,a)] Yonggang Liu,[1,a)] Wei Song,[1,3] Yan Bi,[2] and Hongsen Xie[1]
[1]*Institute of Geochemistry of Earth's Deep Interior Materials and Fluid Interaction, Chinese Academy of Science, Guiyang 550002, China*
[2]*Laboratory for Shock Wave and Detonation Physics Research, Institute of Fluid Physics, Chinese Academy of Engineering Physics, Mianyang 621900, China*
[3]*Graduated University of Chinese Academy of Science, Beijing 100048, China*

(Received 17 June 2010; accepted 31 October 2010; published online 20 January 2011)

A broadband spectroscopy method is proposed to measure the ultrasonic wave phase velocity of Z-cut quartz under high pressure up to 4.7 GPa. The sample is in a hydrostatic circumstance under high pressure, and we can get longitudinal wave and shear wave signals simultaneously in our work. By fast Fourier transform of received signals, the spectrum and phase of the received signals could be obtained. After unwrapping the phase of the received signals, the travel time of ultrasonic wave in the sample could be obtained, and the ultrasonic wave phase velocity could also be resolved after data processing. The elastic constant of measurement under high pressure is also compared with previous studies. This broadband spectroscopy method is a valid method to get ultrasonic wave travel parameters, and it could be applied for elasticity study of materials under high pressure. © *2011 American Institute of Physics*. [doi:10.1063/1.3518953]

I. INTRODUCTION

The knowledge of the ultrasonic wave velocity in materials is a question of great interest, for the information that it supplies in a nonintrusive way on these materials. It is widely used in the area of nondestructive testing, measurement of mechanical properties, and elastic constants of materials. Elastic constants like shear modules, bulk modulus, and Poisson's ratio are determined from the measured longitudinal and shear wave velocities and density of materials. As these properties of geological materials at high temperature and high pressure are essential to geophysics to interpret seismic data and study the interior structure of the Earth, in the past several decades, ultrasonic measurement techniques at extreme conditions have been developed extensively.

With recent development, Li and Chen[1] measured the phase velocity of polycrystalline Al_2O_3 sample at high pressure up to 8 GPa using an interference spectrum technique in a multianvil apparatus. The interference spectrum is obtained through varying frequency of triggering transducer, and this method is time-consuming if one wants to get a curve of phase velocity with frequency. In order to simplify the sound velocity measurement, another method is proposed by Li and Chen[2]. In the latter, the transfer function of a piezoelectric transducer, buffer rod, and sample assembly is used to measure the ultrasonic wave velocity of solid materials. From the recorded transfer function, pulse echo patterns at frequencies of the pass band of the input signal are reproduced after convoluting with monochromatic RF input signals. The time delay is obtained by performing pulse echo overlap and phase comparison measurements on reproduced signals. This method has been extensively adopted by other researchers, such as Liu and Liu,[3] Muller *et al.*,[4] and Higo *et al.*[5]

Sachse and Pao[6] first presented the new technique of measuring the ultrasonic wave velocity by using phase spectrum of a broadband pulse. This method is much simpler than the continuous-wave comparison technique with less instruments requirements. One crucial advantage of the broadband pulse method is the ultrasmall duration time of sound pulse which can be utilized to measure specimen with thin thickness, another advantage of this method is that the frequency-dependent phase velocity of specimen can be obtained directly. Based on this method, He[7] proposed a broadband transmission technique to improve the accuracy and precision in determining the acoustic dispersion of specimen under ambient temperature and pressure conditions.

Yoneda and Song[8] carried out a frequency domain analysis of ultrasonic travel time based on the phase difference between the Fourier coefficients of ultrasonic pulses and developed an easier experimental procedure for making accurate ultrasonic velocity measurements at room temperature and room pressure. Yoneda and Ichihara[9] investigated the acoustic shear wave velocity and attenuation of ultrasonic couplers by broadband reflectivity measurements at around room temperature and 15–30 MHz. Gigahertz (GHz) ultrasonic interferometry was used to measure the elastic moduli of olivine by Chen and Spetzler[10] and Chen and Yoneda,[11] and the relation between frequency and travel time could be get by ultrasonic interferometry.

In this paper, this broadband spectroscopy method will be used to measure the ultrasonic wave velocity of Z-cut quartz under high pressure. The principle of our method is based on Sachse and Pao[6] and the originality lies in the automatic signal processing that we use to determine the exact phase shift of each component of the pulse, so that we can get the phase velocity over a large frequency range without too much

a)Authors to whom correspondence should be addressed. Electronic mails: wangzg168@126.com and liuyg@yahoo.com.

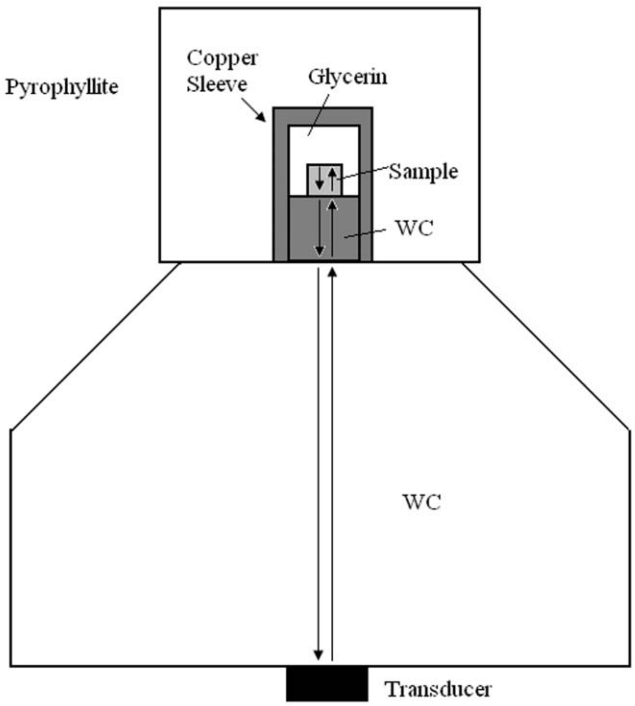

FIG. 1. A cross section of the WC anvil and the pyrophyllite hexahedral cell assembly for the room temperature ultrasonic wave experiments.

adjustment to our previous setup of ultrasonic measurement system.

II. EXPERIMENT SETUP

The experiments are performed in a multianvil pressure apparatus (YJ-3000t) which can generate pressure up to 5 GPa, at the Institute of Geochemistry of Chinese Academy of Sciences of Earth's Deep Interior Materials and Fluid Interaction, Guiyang, China. The apparatus were described in detail by Xie *et al.*[12]

The elastic wave velocities (V_P and V_s) of Z-cut quartz are measured with the pulse-echo method by using the 5077PR ultrasonic square wave pulser/receiver unit (PANAMETRIC-NDT Incorporation, USA), TDS784A digital oscilloscope (Tektronix Corporation, USA) and one immersion piezoelectric transducers (PANAMETRIC-NDT x1013) which can generate 10 MHz longitudinal wave and 5 MHz shear wave in central resonant frequency simultaneously. Detailed descriptions of the ultrasonic measurement system have been given by Liu *et al.*[13–17]

Figure 1 is a cross section of the WC anvil and the pyrophyllite hexahedral cell assembly for the room temperature ultrasonic wave experiments. The cell assembly is something like the solid-liquid hybrid assembly used in the ultrasonic measurements under high pressure by Song and Yoneda.[15,16] In this study, the sample of Z-cut quartz (2.19 mm long, diameter 8 mm) is affixed to the WC buffer rod (12.5 mm long, diameter 10 mm), and the surrounding of the sample is filled with glycerin to applied hydrostatic pressure environment. To prevent the glycerin from extruding into the pyrophyllite while applying pressure, the glycerin, sample and WC buffer

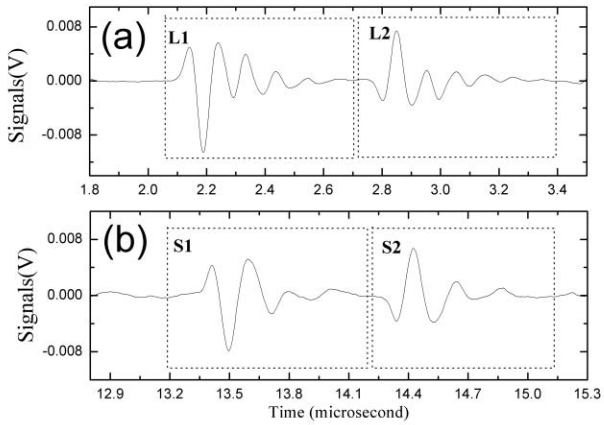

FIG. 2. The received ultrasonic wave signals in an experiment for ultrasonic wave velocity measurement on Z-cut quartz at 0.5 GPa. (a) is longitudinal wave echoes of sample (L1 and L2), and (b) is shear wave echoes of sample (S1 and S2).

rod are inserted into a copper sleeve (20 mm long, outside diameter 12 mm, inner diameter 10 mm) by interference fit sealing. This sealing method of liquid under high pressure is easy to realize because it needs no welding procedures but inserting tightly.

The pulse-echo method is briefly presented here. When traveling along the axis, the ultrasonic wave will be reflected at each interface. The travel time Δt of the sample can be determined by reflected signals at the upper and lower interface of the sample. Panametrics 5077PR is used to generate pulse and receive the successive echo signals. The oscilloscope digitizes the received signal. The received ultrasonic wave signal is shown in Fig. 2. In Fig. 2(a), L1 and L2 are longitudinal wave signals reflected from the upper and lower interface of the sample, and in Fig. 2(b), S1 and S2 are shear wave signals reflected from the upper and lower interface of the sample. The digital signal is downloaded to a computer for signal processing. We will discuss this processing in detail in the next section. What we call ''spectrum'' is the modulus of the signal's fast Fourier transform (FFT).

The pressure dependence of the length change for α-quartz in different cuts has been investigated extensively. In the process of converting the measured travel times to velocities, the length of the specimens at high pressure are corrected by using lattice compression data of α-quartz under hydrostatic pressure.[17]

III. DATA PROCESSING

A. Principle of the method

The principle of the broadband spectroscopy method has been described elaborately by Sachse and Pao,[6] Peters and Petit,[18] and He.[7] If we suppose that the wave is plane, z being the coordinate of the receiver along the anvil axis, we can write the ultrasonic wave pressure as

$$p(z,t) = \int_{-\infty}^{+\infty} p(\nu) e^{-\alpha(\nu)z} e^{2i\pi\nu(t-z/c(\nu))} d\nu, \qquad (1)$$

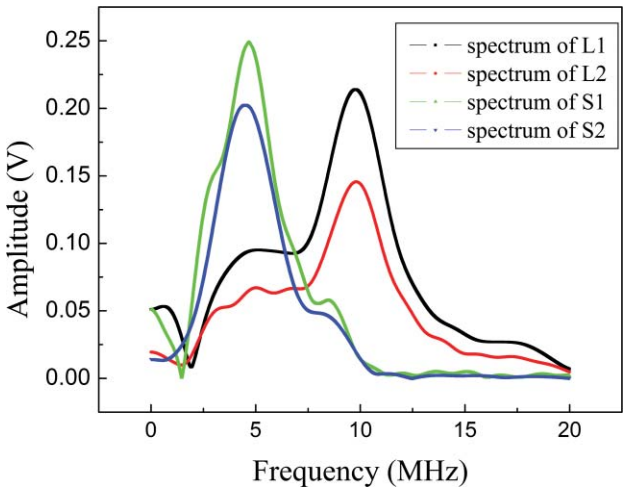

FIG. 3. (Color online) The amplitude spectra of longitudinal wave (L1, L2) of 10 MHz center frequency and shear wave (S1, S2) of 5 MHz center frequency obtained by FFT.

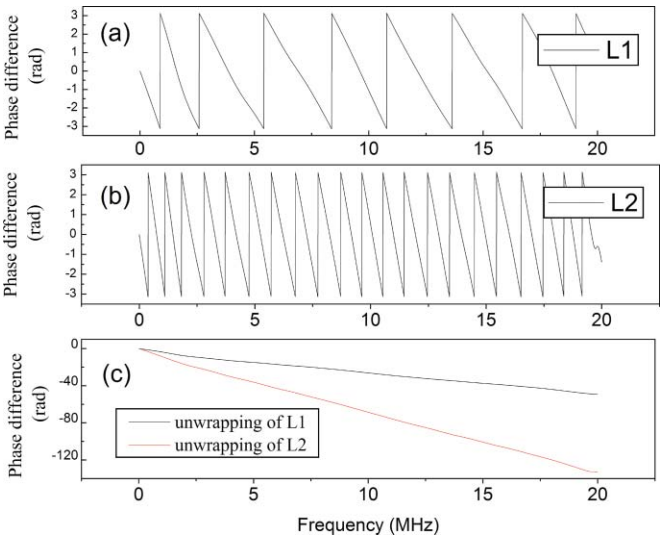

FIG. 4. (Color online) The phase-frequency relation of reflected signals L1 and L2 by FFT and phase unwrapping.

where ν, $c(\nu)$, and $\alpha(\nu)$ are the frequency, phase velocity of the wave, and attenuation coefficient, respectively.

After we get the ultrasonic wave signal for two different positions of the wave, say z_1 and z_2, and then we can calculate the velocity

$$c(\nu) = -\frac{2\pi\nu(z_2 - z_1)}{\mathrm{Arg}\left(\frac{\tilde{p}(z_2,\nu)}{\tilde{p}(z_1,\nu)}\right)} = \frac{2\pi\nu(z_2 - z_1)}{\Delta\varphi}, \quad (2)$$

where $\tilde{p}(z_2, \nu)$ is the time Fourier transformation of $p(z_2, t)$.

B. Experimental data Process

When the ultrasonic wave travels through the WC anvil, buffer rod, and sample, the ultrasonic wave will be reflected at each interface. The reflected signals of the upper and lower interfaces of sample are the exactly signals what we need to deduce the ultrasonic velocities. Before applying FFT, the interval selection of each signal is shown as the dashed window in Fig. 2 which should keep the integrity of each reflected signal. Obviously, suitable interval of each signal is critical to avoid the influence of coda wave while doing signal truncation. In general, there are two methods to increase the interval between successive echoes. One is to adopt highly damping transducers to get shorter duration time of each echoes, another one is to increase the sample thickness appropriately. We found the signal quality as shown in Fig. 2 is good enough to decide each integral signal for applying FFT, because slightly changing the right side of dashed window will scarcely affect the results of FFT, regardless of amplitude or phase spectrum.

Figure 3 shows the amplitude spectra of longitudinal wave (L1, L2) of 10 MHz center frequency and shear wave (S1, S2) of 5 MHz center frequency obtained by FFT. Their center frequencies are 10 MHz for longitude waves and 5 MHz for shear waves, respectively. When applying FFT, the resolving power of frequency should be paid special attention to. In this work, the sampling rate that we used is 250 MS/s, the recorded time of each signal is about 0.6 μs, so the sampling points of each signal is just 150 points. If signals are processed by FFT directly, the resolving power of frequency domain is about 1.7 MHz, and there will be less than ten points in frequency domain that we use, and this result cannot satisfy our need. To increasing the resolving power of frequency, the zero-padding method must be used. In this work, 50 K zeros are padded to the tail of the signals before applying FFT, and the resolving power of frequency is 5 kHz.

In the phase spectra of FFT, we can find that the phase changes from $-\pi$ to π with the change of frequency as shown in Figs. 4(a) and 4(b). Figure 4(a) is the phase-frequency relation of L1, and Fig. 4(b) is the phase-frequency relation of L2. By phase unwrapping method[12], the separated phase of L1 and L2 can be acquired as shown in Fig. 4(c). With frequency increasing, the phase difference of L1 and L2 also increases linearly.

The different phase $\Delta\varphi$ related with frequency of the two signals can be calculated, and the travel time t of the sample can be calculated out as

$$t = \Delta\varphi/(2\pi\nu), \quad (3)$$

where ν is the frequency.

For example, Fig. 5 shows the travel time of longitudinal wave in Z-cut quartz by phase unwrapping method and travel time by reading the corresponding peaks at 0.5 GPa. The curve is the travel time by phase unwrapping method, and it is nearly straight in the valid frequency range from 8 to 12 MHz. It is concluded that the ultrasonic wave attenuation of quartz is small and the dispersion can be neglected. The dot is the travel time by reading the corresponding peaks of reflected signals of sample. There is about 2 ns difference between the dot and the line at the center frequency 10 MHz in Fig. 5. That is owing to the resolving power of the method by reading corresponding peaks. The sampling rate of digital oscilloscope we use is 250 MS/s, and the resolving power is just 4 ns related to the inverse of sampling rate. As the resolving power by phase unwrapping method is extremely high according to the FFT theory, it is possible to

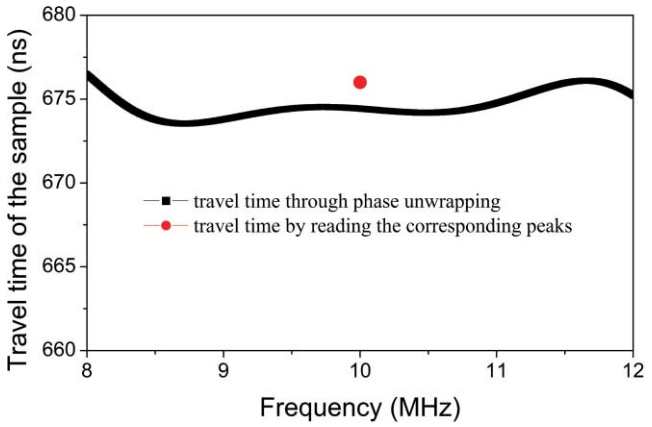

FIG. 5. (Color online) Travel time through phase unwrapping and travel time by reading the corresponding peak of longitudinal wave in Z-cut quartz at 0.5 GPa.

see there is about 2 ns difference between the travel time by reading the corresponding peaks and by the phase unwrapping method. It can be concluded that the method by reading corresponding peaks is hard to distinguish the little change of travel time, but the phase unwrapping method can.

All the signals under different pressures are processed by the broadband spectroscopy method, then we get the travel time of sample under different pressures as shown in Fig. 6. The travel time of sample decreases with the pressure increasing steadily. It can be said that the broadband spectroscopy method is effective.

IV. RESULT AND DISCUSSION

Single crystals of Z-cut quartz are chosen for our feasibility study for following reasons: (1) the ultrasonic wave attenuation of quartz is very small, and it is a benefit for checking the broadband spectroscopy method; (2) high quality single crystals of quartz specimens are commercially available; and (3) elastic constants for single crystals are available from hydrostatic experiments at low pressure and theoretical calculation. The crystal structure of quartz is well established and has been thoroughly investigated as a function of pressure;[19–23] and (4) quartz does not undergo

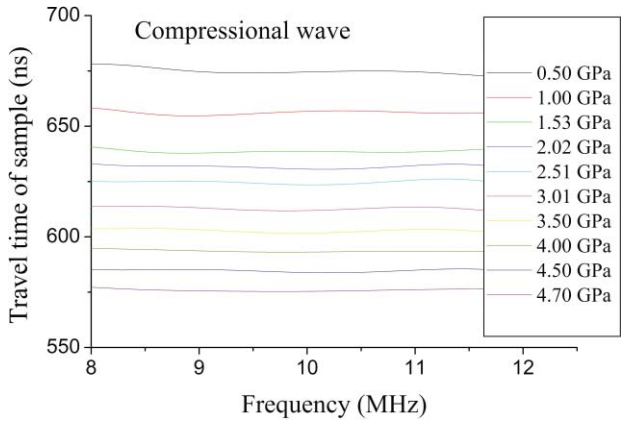

FIG. 6. (Color online) Longitudinal wave travel time of Z-cut quartz under pressure.

TABLE I. Ultrasonic wave velocity of Z-cut quartz under pressure.

Pressure (GPa)	t_p (μs)	t_s (μs)	V_L (km/s)	V_S (km/s)
0	...	...	6.319[a]	4.688[a]
	...	...	6.31[b]	4.68[b]
0.50	0.674	0.944	6.479	4.629
0.78	0.664	0.943	6.577	4.629
1.00	0.657	0.932	6.639	4.680
1.23	0.647	0.928	6.732	4.694
1.53	0.639	0.923	6.810	4.713
1.74	0.631	0.921	6.890	4.720
2.02	0.624	0.920	6.960	4.718
2.25	0.618	0.918	7.020	4.722
2.51	0.612	0.917	7.077	4.719
2.75	0.608	0.916	7.112	4.722
3.01	0.602	0.915	7.180	4.721
3.25	0.597	0.915	7.227	4.717
3.50	0.593	0.915	7.267	4.710
3.75	0.588	0.915	7.319	4.707
4.00	0.584	0.917	7.366	4.691
4.25	0.580	0.916	7.408	4.690
4.50	0.575	0.917	7.458	4.680
4.70	0.572	0.915	7.489	4.683

[a]Reference 24.
[b]The ultrasonic wave velocity of zero pressure is from linear fitting the velocity under high pressure.

any phase transitions in the pressure range in this room temperature study. Quartz remains stable up to approximately 18 GPa at ambient temperature, several experimental results show a gradual amorphization, producing heterogeneous samples of coexisting crystalline and amorphous phases at higher pressure.[20,21] And so, it is a very suitable material for our ultrasonic experiments at high pressure in our multianvil apparatus and also for interlaboratory comparison.

Table I, shows ultrasonic wave velocity of Z-cut quartz under high pressure. The round-trip travel time of longitudinal wave t_p is got from phase unwrapping at the center frequency 10 MHz in Fig. 6, and the round-trip travel time of shear wave t_s is got from phase unwrapping at the center frequency 5 MHz. The length of sample under pressure is corrected as mentioned in Sec. II, and the ultrasonic wave velocity V_L and V_S is calculated after length correction. The ultrasonic wave velocity in first line is at zero pressure experimental data of Kushibiki and Ohtagawa,[24] and the second line data are the extrapolating value to zero pressure of ultrasonic wave velocities under high pressure of this study. The two results are consistent with each other very well.

The experimental data of this study are also compared with other previous research works. Mcskimin and Anderrich[25] measured the ultrasonic wave velocity of single-crystal quartz as a function of temperature and pressure in helium gas pressure apparatus. The modulus of C_{33} and C_{44} is used to compare with this study in Fig. 7. Calderon and Gauthier[19] measured all the elastic compliances of α-quartz, which had been obtained as a function of hydrostatic pressure up to 1 GPa by using the ultrasonic pulse-echo technique and a newly developed method to treat the data. By theoretical calculation method, Kimizuka and Ogata[26] calculated all the independent elastic constants of α-quartz under hydrostatic

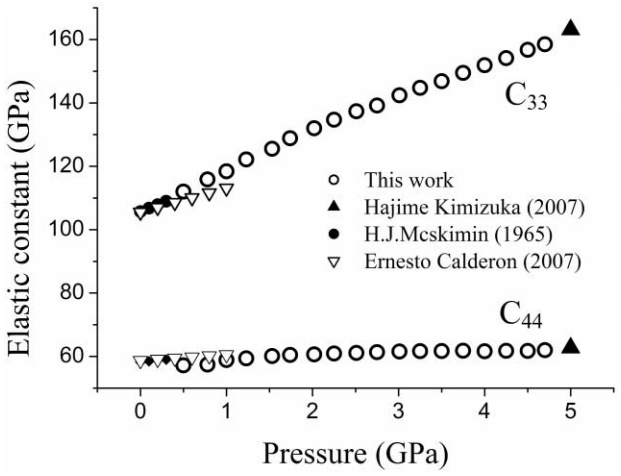

FIG. 7. Comparison of C_{33} and C_{44} of quartz.The open circles are experimental data from this study. The open triangles are experiment data of Calderon and Gauthier (Ref. 19). The solid triangles are theoretical data of Kimizuka and Ogata (Ref. 26). The solid circles are experiment data of Mcskimin and Anderrich (Ref. 25).

pressure up to 20 GPa using density functional theory. Their results are all consistent with this study in Fig. 7.

A broadband spectroscopy method based on the pulse-echo method can estimate the two parameters, phase velocity and ultrasonic wave attenuation, that uses both the amplitude and phase information of received signals in a pulse-echo method. But the accoustic attenuation of quartz is very small, so we just calculate the phase velocity of quartz by using phase information in this study. In some cases, if a material has a large attenuation coefficient, that is to say, the material disperses with the frequency observable, it will be advantageously to use this broadband spectroscopy method for getting the phase velocity and attenuation information under pressure.

V. CONCLUSION

We have applied a broadband spectroscopy method based on the pulse-echo method to measure the phase velocity of Z-cut quartz under the pressure of 0–4.7 GPa. The experimental result is consistent with previous studies. By this method, the spectrum of ultrasonic wave is easy to get, which is helpful for studying ultrasonic velocities and attenuation coefficient of materials.

Comparing the method of reading the corresponding peaks with the method of phase unwrapping, the phase unwrapping method gives higher resolving power, which can distinguish the very little change of travel time, but the method of reading the corresponding peaks is limited by the digital oscilloscope sampling rate.

A broadband spectroscopy method is a valid method to get ultrasonic wave travel parameters, and it is worth being widely applied in elasticity study of materials under high pressure.

ACKNOWLEDGMENTS

This work is supported by the National Natural Science Foundation of China (Grant No. 40974052), the Knowledge Innovation Program of the Chinese Academy of Sciences (Grant No. KZCX2-YW-Q08–3-4), and the Mutual Foundation of Huge Scientific Equipment of National Natural Science Foundation of China and Chinese Academy of Science (Grant No. 10979053). Z.W. also thanks Dr. Wenge Zhou and Dr. Dawei Fan at Institute of Geochemistry in Chinese Academy of Sciences for helpful discussion.

[1] B. Li, G. Chen, G. D. Gwanmesia, R. C. Liebermann, in *Properties of Earth and Planetary Materials at High Pressure and Temperature*, Geophysical Monograph Series 101, edited by M. H. Manghnani and T. Yagi (American Geophysical Union, Washington, DC, 1998), pp. 41–61.
[2] B. Li, K. Chen, J. Kung, R. C. Liebermann, and D. J. Weidner, J. Phys.: Condens. Matter **14**, 11337 (2002).
[3] W. Liu, Q. Liu, M. L. Whitaker, Y. Zhao, and B. Li, J. Appl. Phys. **106**, 043506 (2009).
[4] H. J. Mueller, F. R. Schilling, C. Lathe, and J. Lauterjung, in *Advances in High-Pressure Technology for Geophysical Application* (Elsevier B.V., Amsterdam, 2005), Chap. 4, pp. 67–94.
[5] Y. Higo, T. Inoue, T. Irifune, K. Funakoshi, and B. Li, Phys. Earth Planet. Inter. **166**, 167 (2008).
[6] W. Sachse and Y. H. Pao, J. Appl. Phys. **49**, 4320 (1978).
[7] P. He, Ultrasonic **37**, 67 (1999).
[8] A. Yoneda, M. Song, and E. Ito, J. Appl. Phys. **97**, 024908 (2005).
[9] A. Yoneda and M. Ichihara, J. Appl. Phys. **97**, 054901 (2005).
[10] G. Chen, H. Spetzler, I. Getting, and A. Yoneda, Geophys. Res. Lett. **23**, 5 (1996).
[11] G. Chen, A. Yoneda, I. Getting, and H. Spetzler, J. Geophys. Res. **101**, 25161 (1996).
[12] H. Xie, Y. Zhang, H. Xu, and G. W. Hou, Science in China, Ser. B **36**(10), 1276 (1993).
[13] Y. Liu, H. Xie, and J. Guo, Chin. Phys. Lett. **17**(12), 924 (2000).
[14] Y. Liu, H. Xie, and J. Guo, J. Phys.: Condens. Matter **14**(44), 11381 (2002).
[15] M. Song, A.Yoneda, and E. Ito, Rev. Sci. Instrum. **76**, 033906 (2005).
[16] M. Song, A. Yoneda, and E. Ito, Chin. Sci. Bull. **52**, 1600 (2007).
[17] B. Olinger and P. M. Halleck, J. Geophys. Res. **81**(32), 5711 (1976).
[18] F. Peters and L. Petit, Ultrasonics. **41**(5), 357 (2003).
[19] E. Calderon, M. Gauthier, F. Decremps, G. Hamel, G. Syfosse, and A. Polian, J. Phys.: Condens. Matter **19**, 436228 (2007).
[20] K. Kingma, R. Hemley, H. Mao, and D. Vleben, Phys. Rev. Lett. **70**, 3927 (1993).
[21] K. Kingma, H. K. Mao, and R. Hemley, High Press. Res. **14**, 363 (1996).
[22] R. Hazen, L. Finger, R. Hemley, and H. Mao, Solid State Commun. **72**, 507 (1989).
[23] L. Levien, C. Prewitt, and D. Weidner, Am. Mineral. **65**, 920 (1980).
[24] J. Kushibiki, M. Ohtagawa, and I. Takanaga, J. Appl. Phys. **94**, 295 (2003).
[25] H. J. McSkimin, P. Andreatch Jr, and R. N. Thurston, J. Appl. Phys. **36**, 1624 (1965).
[26] H. Kimizuka, S Ogata, J. Li, and Y. Shibutani, Phys. Rev. B **75**, 054109 (2007).