Experimental study of acoustical memory in lithium niobate

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In this paper, the acoustical memory effect is studied from 2.5 to 10 MHz. This acoustical memory effect has been observed in both columnar and cubic samples. The observed phenomenon of amplitude hysteresis of the acoustical memory signal suggests nonclassical nonlinear properties, caused by the structural inhomogeneity and inevitable flaws in $LiNbO_3$. Experimental results also show that the amplitude of the acoustical memory signal is enhanced by increasing the temperature at these frequencies. The shape of the memory signal changes greatly if the driving voltage is sufficiently high or each burst contains enough cycles; second and even third parts of the acoustical memory signals may appear. Amplitude hysteresis of the fundamental and second harmonic of the direct wave can also be found. Some experimental results are qualitatively explained in this paper.

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I. INTRODUCTION

Lithium niobate ($LiNbO_3$) is a type of ferroelectric material whose properties are still of interest to fundamental and applied science due to its physical and chemical properties. It is a strongly piezoelectric material with nonlinear optical and electro-optical properties and is widely used in sensors, ultrasonic motors, actuators, ultrasonic transducers, and many other electromechanical devices [1–3]. The acoustical memory in $LiNbO_3$ has different frequency and amplitude characteristics from the known ferroelectric memory (which is related to polarization hysteresis [4,5]), and is also different from the electro-optical memory [5,6]. Observations of the acoustical memory may aid in interpretation of acoustical losses in the medium and are helpful in understanding of the acoustical nonlinear response of lithium niobate. A study of the acoustical energy redistribution that occurs in $LiNbO_3$ single crystals has been reported [5,7], in which the acoustical tone burst stores energy within the crystal that is reemitted later. This effect can be characterized as an "acoustical memory" and the later part of the signal is the so-called acoustical memory signal.

Previous studies [6,7] have been carried out for the frequency range between 16 and 30 MHz. The acoustical memory in $LiNbO_3$ has been investigated at 4.6 MHz [8]. In this paper, we have extended the frequency range to cover 2.5–10 MHz. The mechanism of the effect of ultrasound on the energy state of $LiNbO_3$ is related to the frequency of the mechanical stress. Studies at a different frequency range have not only theoretical value but also practical interest. The acoustical memory in $LiNbO_3$ at the low-frequency range from 2.5 to 10 MHz has not been adequately investigated. In this study, we have discovered amplitude hysteresis phenomena in the memory signal. The strength of this memory signal has also been studied as a function of temperature. Furthermore, we have discovered second and even third parts of the acoustical memory signal behind the first one. Amplitude hysteresis of the fundamental and second harmonic of the direct wave has been found, which reflects the nonclassical nonlinear characteristics of $LiNbO_3$. The nonlinear frequency shift also shows this special feature of $LiNbO_3$.

II. EXPERIMENTAL TECHNIQUE

The experimental system was as follows. A tone-burst signal from an arbitrary wave form generator (Agilent 33250, Loveland, CO) is fed into a power amplifier (ENI A150, Rochester, NY). The output of the amplifier is applied to the transmitting transducer and the acoustic wave arriving at the other end of the sample is detected by a receiver. Both of these are lead zirconate titanate (PZT) transducers (Shantou Ultrasonic Co., Ltd., diameter 20 mm). The acoustic pressure is recorded by a digital oscilloscope (Agilent 54810, Colorado Springs, CO).

Two samples of single-crystal $LiNbO_3$ were used for measurements of the acoustical memory effect. One sample was a cylinder 3.5 cm long and 2.0 cm in diameter; the other was a cube $3.3\times3.4\times3.4$ cm^3. These two samples are strictly regular and the parallelism of the faces or the end planes of the $LiNbO_3$ samples was assured first before the experiments; otherwise the acoustical memory signal may be affected. The active surfaces of the emitter and receiver were 20 mm in diameter. It was crucial to keep the acoustic axes of both transducers strictly in one line and both surfaces parallel in order to avoid wave form conversion. In this situation, the amplitude of the receiving signal was largest. It was also necessary to put two transducers at the central location of the samples in order to avoid wall reflection. In our experiment, a trestle table was designed to have two spatially symmetrical jacks to keep the axes of the two transducers and cylindrical sample along one line. We used vaseline as the coupling agent and then fixed the two transducers and a lithium niobate sample on the trestle table in order to maintain the position of the sample and transducers.

Acoustical memory signals were obtained in both the cylindrical and cubic samples at 2.5 MHz. The driving voltage

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was 200 mV in amplitude, 8 cycles in width, and with a 100 Hz pulse repetition frequency; the temperature of the experiment was 20 °C. We first tested the experimental system using aluminum. The exponential echo train in aluminum proved that the acoustical memory signals caused by lithium niobate samples in the following experiments were not caused by the experimental system. In order to demonstrate the relationship between the memory signal and the piezoelectrically active direction in the cubic sample, under the same conditions, we compared the memory signals along the piezoelectrically active direction [001] and nonpiezoelectric direction [010]. In the experiment to observe the amplitude hysteresis of the memory signal, the amplitude of the acoustical memory signal was measured at room temperature while the driving voltage was increased from 100 mV to 1 V at 100 mV intervals, and then decreased gradually. We also analyzed the hysteresis effects in the cylindrical sample at room temperature, and showed that the results were similar to those in the cubic sample. Other experimental conditions were kept the same in order to make the results more reliable.

In the experiment where the temperature dependence (5–55 °C) of the memory signals of the cylindrical sample was measured, we used an oven to heat the sample, and then cooled the sample in air after turning off the oven. In order to reduce temperature variations across the sample, the two processes (heating and cooling) must be very slow. To increase the temperature by 5 °C took about 15 min while to decrease by 5 °C took about 20 min. The temperature in the $LiNbO_3$ sample was the average value measured by two temperature probes, which were stuck on to the two opposite sides of the sample. We chose a high-temperature coupling agent instead of ordinary vaseline, which would melt above 50 °C. The performance of the PZT transducers was nearly identical in the range of 15–45 °C, but their performance decreases by 4% at 5 °C and increases by 11% at 55 °C; therefore all results must be corrected appropriately.

For measurement of the memory signal dependence on the number of cycles, the number of cycles was changed from 4 to 64 under the same driving voltage (200 mV). In order to avoid overlapping between the direct wave and the first reflected wave, the limit of the number of cycles was set to 67. Once the number of cycles was reached, the sample was left for 10 min to achieve equilibrium.

A PZT transducer whose central frequency was 2.5 MHz was adopted as transmitting transducer. One PZT transducer whose central frequency was 2.5 MHz was used as receiver to measure the change of amplitude of the first harmonic with the driving force. Another transducer whose central frequency was 5 MHz was chosen to measure the amplitude of the second harmonic. In addition, two analog bandpass filters, with frequency ranges 2–3 MHz and 4–6 MHz, respectively, were used to measure the amplitude of the fundamental and second harmonic of the direct wave.

III. EXPERIMENTAL RESULTS

Figure 1 shows the acoustical memory signal in the cubic sample at 2.5 MHz. The signal from 0 to 50 μs comprises

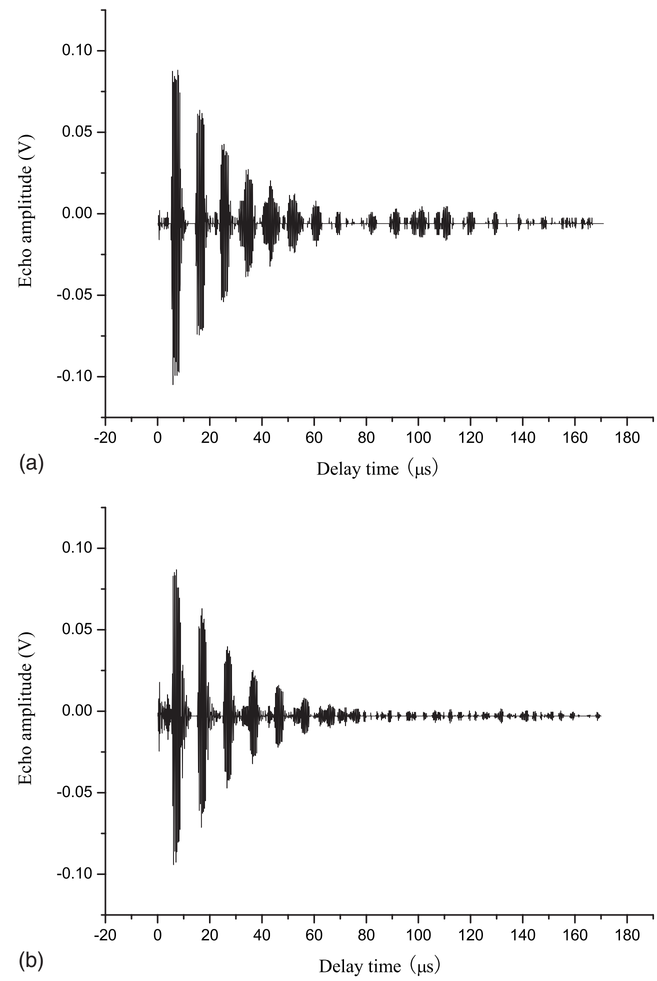

FIG. 1. Oscillogram of the echo taken from the cubic sample $LiNbO_3$ along the piezoelectrically active directions at 2.5 MHz: (a) [001] and (b) [010].

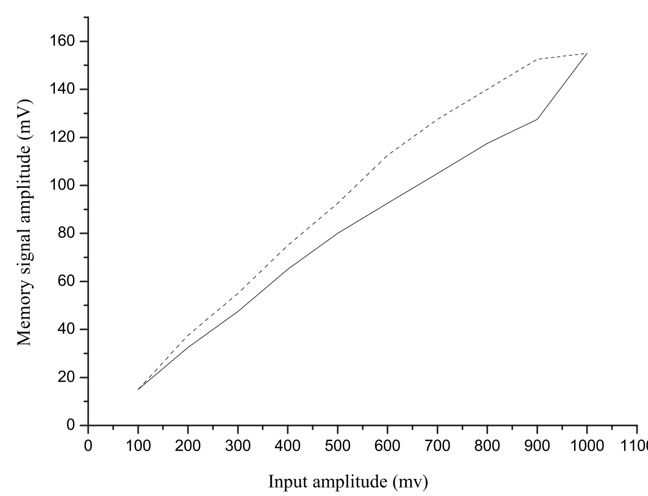

FIG. 2. Change of the amplitude of the acoustical memory signal with the input amplitude. The solid line represents the amplitude of the acoustical memory signal in the process of increasing input amplitude; the dashed line shows the amplitude of the acoustical memory signal in the process of decreasing input amplitude.

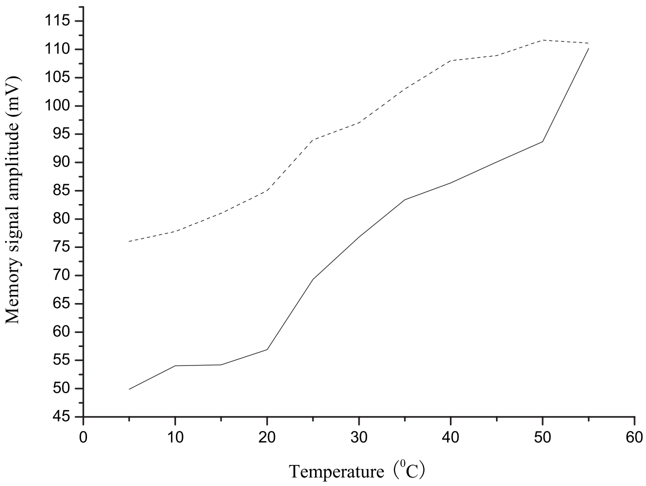

FIG. 3. Temperature characteristics of the acoustical memory signal. Change of the memory signal with increase (solid line) and decrease of the temperature (dashed line).

the first eight echoes. The echo train dies down to the noise level after a few round trips through the sample. The eighth echo is already very weak. A little time (about 10–20 μs) later, another set of signals can be detected; these appear from a release of energy that has been stored within the crystal. This part of the signal is the memory signal. Figures 1(a) and 1(b) demonstrate the memory signal obtained using acoustic longitudinal waves propagating along the piezoelectrically active [001] and nonpiezoelectric directions [010], respectively. The amplitude of the memory signal is obviously larger along the piezoelectrically active direction. Moreover, the central frequency of the memory signal is 2.44 MHz, which means a 60 kHz frequency shift. Nonexponential echo patterns were also observed, which substantiate the previous observations [7].

Figure 2 shows the amplitude characteristic of the acoustical memory signal as a function of the driving voltage. Since the emitting or receiving efficiency of the transducers varies with the driving force, corrections were necessary. The memory signal increases if the driving signal increases, and vice versa. At the same input voltage, the amplitude of the memory signal for decreasing driving voltage is higher than that for increasing driving voltage. From a loop consisting of two different curves, we can easily obtain the amplitude hysteresis loop of the crystal, which is a notable characteristic of nonclassical nonlinear acoustics.

Figure 3 shows the results of measurements of the memory signal dependence on temperature ranging from 5 to 55 °C. The sample was heated up from the lowest temperature (5 °C) to a peak temperature (55 °C) and then cooled down. The results shown in Fig. 3 indicate that the amplitude of the memory signal is higher in the cooling (compared with the warming) process at the same temperature. In addition, the lower the temperature, the bigger is the difference in the amplitude of the acoustical memory signal between the warming and cooling processes.

We also find second and third parts of the acoustical memory signal if the driving force is large enough or if each burst includes enough cycles. Figure 4 shows the change of the memory signal with increasing numbers of cycles at 10 MHz. We used two PZT transducers whose central frequency was 10 MHz as emitter and receiver, and kept the input voltage at 200 mV and cycle period 10 ms. The numbers of cycles in Figs. 4(a)–4(f) are 4, 16, and 64, respectively. When each burst contains 4 cycles, we can observe a clean memory signal envelope, just as shown in Figs. 4(a) and 4(b). But the second and third parts of the memory signals are so weak that we consider that only the first part of the memory signal is present. When each burst contains 16 cycles, the second part of the memory signal can be observed clearly, as shown in Figs. 4(c) and 4(d). Although the gap between the first and the second signals is not very large, the response of the first and second parts to changing the number of cycles is so different (Fig. 5) that we believe they are two distinct parts of the memory signals. Finally, as shown in Figs. 4(e) and 4(f), when we increase the number of cycles to 64, the gap between the first and second parts is obvious enough to differentiate them, and the signal of the third part is clear. In fact, the amplitude of the first part also increases when the number of cycles changes from 4 to 64. Although the first part partly overlaps with the second part, the gap becomes larger and differentiation becomes clearer with the increase in the number of cycles.

From measurements of the amplitude of the direct wave, we find that it becomes weaker as the number of cycles increases (Fig. 6) and a part of its energy shifts into the memory signals, so that the acoustic memory signal in each part increases. However, the rates of increase for different parts are quite different (Fig. 5). We have measured the change of amplitude of the direct wave with the number of cycles while an ultrasonic wave passes through a piece of aluminum (Table I), and we find that the emission performance of the transducer is not sensitive to the number of cycles.

We have also analyzed how the acoustical memory signals change with the driving amplitude. The results were similar, and the memory signals in the second and third parts can be clearly observed. We also believe that fourth and fifth parts of the memory signals could be obtained if the number of cycles or driving amplitude is large enough.

The change of the fundamental signal with the amplitude of the driving voltage can be seen in Fig. 7(a). The amplitude hysteresis of the second harmonic can also be found [Fig. 7(b)]. The amplitudes of the first and second harmonics in the process of decreasing the driving voltage are higher than those in the process of increasing driving voltage, as for the acoustical memory signal. The power spectrum of the PZT transmitting transducer shown in Fig. 8 demonstrates that the influence of the second harmonics induced by the transmitter on the receiving signal is very small and can be ignored.

IV. DISCUSSION

When ultrasonic waves pass through $LiNbO_3$ samples, a portion of the sound energy will shift to substructures of superlattices and anomalous regions because of the defects of inner substructures, which form subsources of ultrasonic vibrations. After a nonlinear interaction between the primary

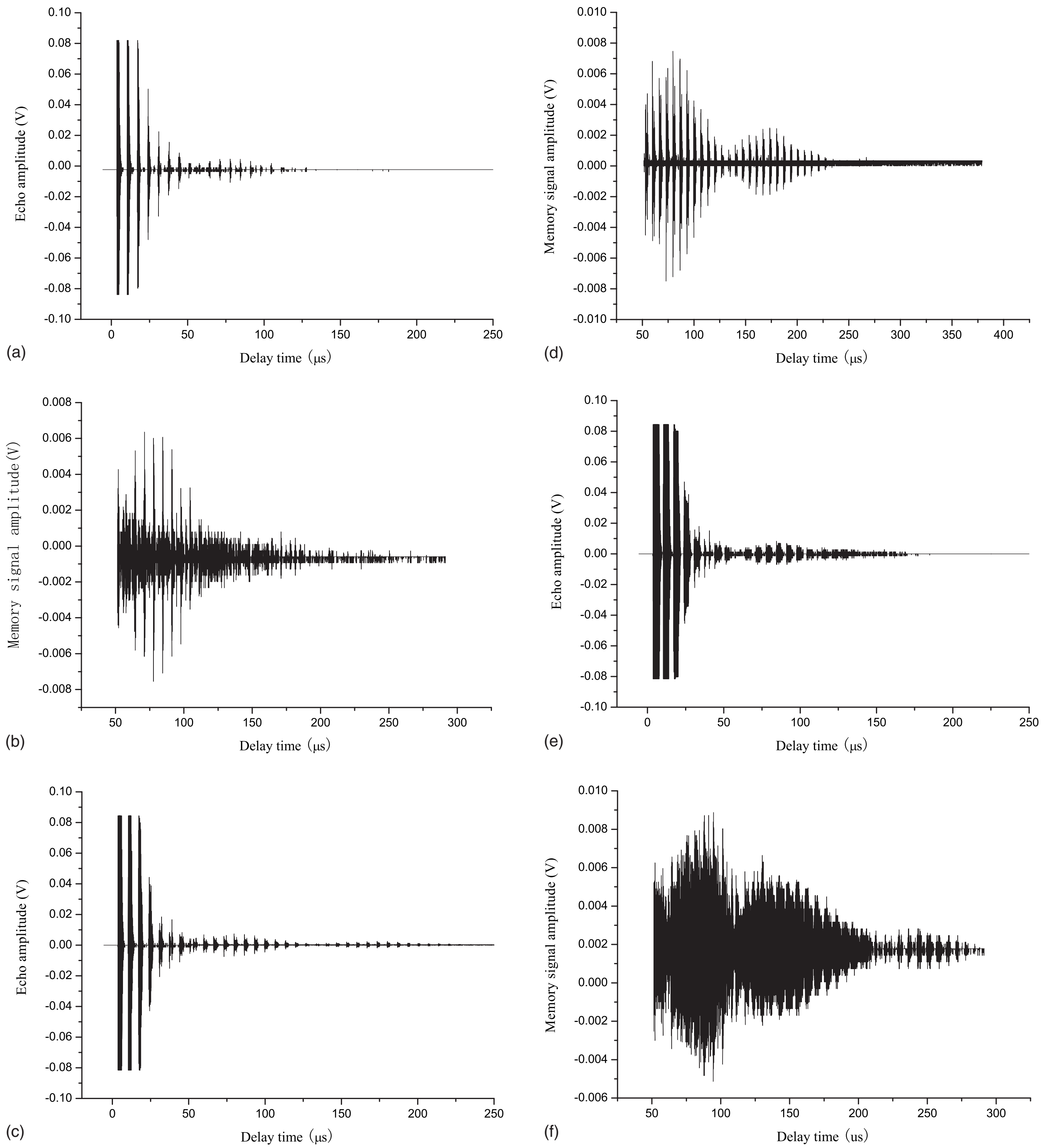

FIG. 4. Oscillogram of the echo when the number of cycles in every burst is 4 (a),(b); 16 (c),(d); 64 (e),(f). (a),(c),(e) are the figures that give the echo and (b),(d),(f) show the acoustical memory signals.

sound source and subsource, the sound energy is retransmitted and a memory signal appears. On the other hand, acoustical stress leads to a redistribution of ferroelectric domains. The domain walls can be placed in motion by the piezoelectrically active acoustic wave. The physical mechanism of interaction between ultrasonic waves and ferroelectric domains originates from the piezoelectric field. The vibration of ferroelectric domains and defects, which originates from the piezoelectric effect, still remains after ultrasonic waves have passed through the sample. This kind of phenomenon occurs only in crystals because other media do not have domain structures. The asymmetrical distribution of crystal lattices results in an interaction between domain walls and ultrasonic waves, which induces the acoustical memory signal.

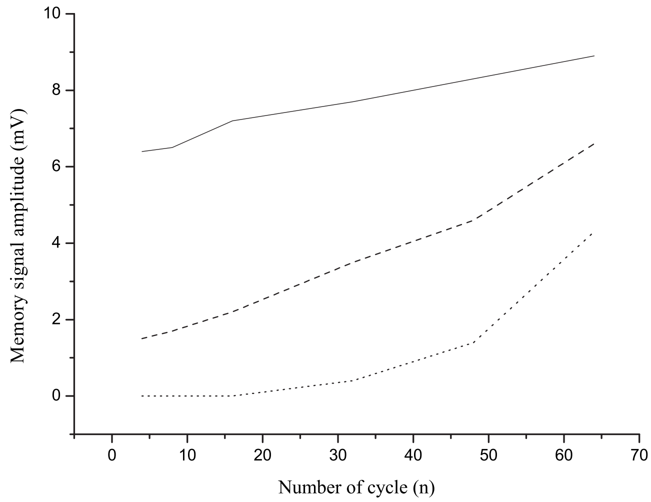

FIG. 5. Change of the amplitude of the memory signal with the number of cycles. The solid line represents the amplitude of the first part of the memory signal, the dashed line that of the second part, and the dotted line that of the third part.

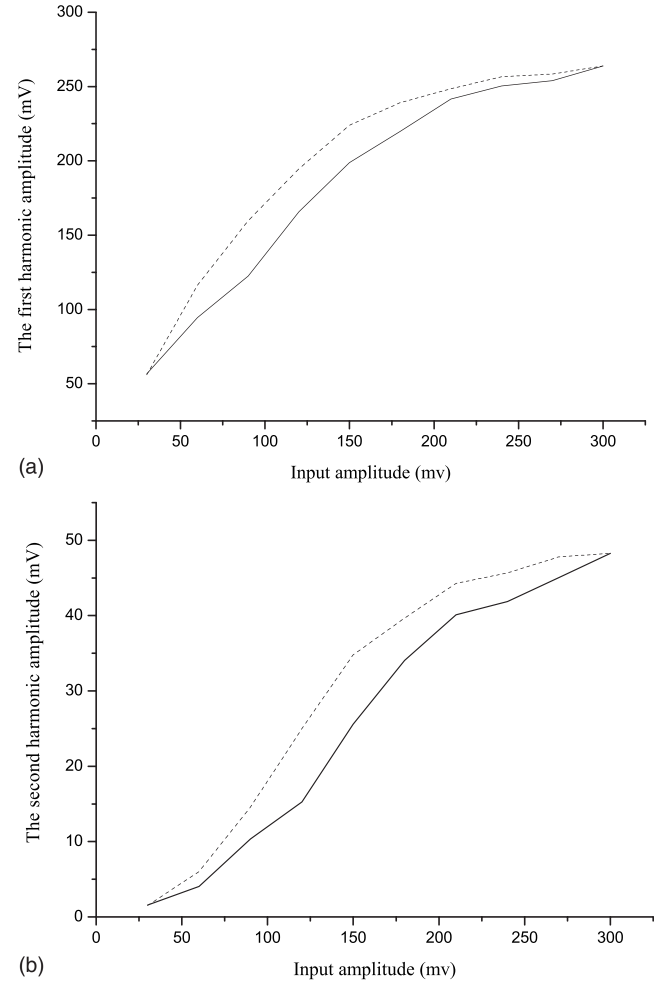

FIG. 7. Change of the amplitude of the fundamental (a) and the second harmonic (b) with the input amplitude. The solid line represents the amplitude in the process of increasing input amplitude; the dashed line shows the amplitude in the process of decreasing input amplitude.

The memory signal is closely related to the crystal's microstructure because the memory signal is also quite different in different directions. Therefore, the memory signal can indicate the microcharacteristics of crystals.

The domain structure of ferroelectrics can be influenced by an ultrasonic wave passing through the crystal. The increase of ultrasound in the energy state of the domain structure reduces the dielectric viscosity and increases the mobility of the domain walls. If we increase the number of cycles or amplitude of the ultrasound, more ferroelectric domain walls can be driven into motion by external stress, which can be used to explain the occurrence of the second and third acoustical memory signals.

In our paper, the amplitude of the acoustical memory signal at 2.5 MHz increases with increasing temperature, but in the papers by Breazeale and co-workers [7], the amplitude decreases with increasing temperature at 25.9 MHz. The acoustical memory is related to the movement of the ferro-

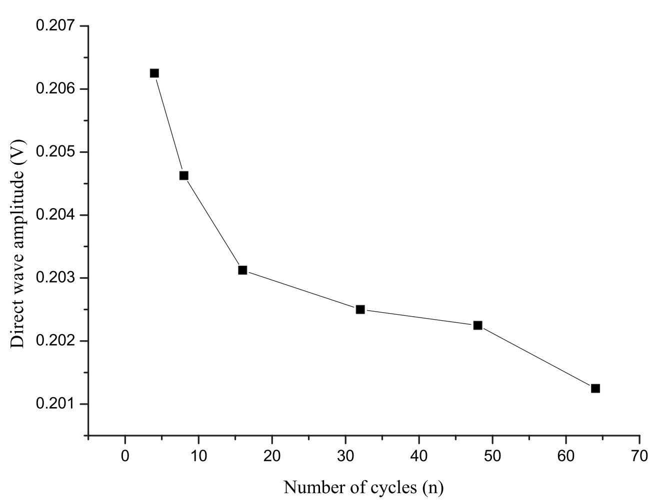

FIG. 6. Change of the amplitude of the direct wave with the number of cycles in each burst.

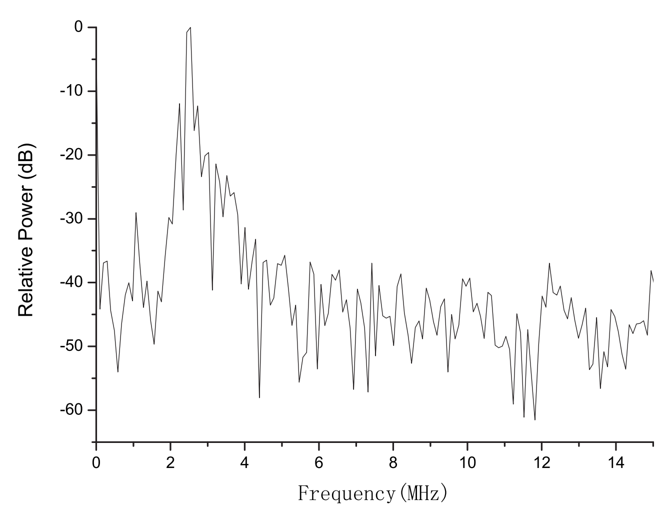

FIG. 8. Relative power spectrum of the transmitting transducer.

TABLE I. Amplitude of the direct wave versus the number of cycles.

Number of cycles	4	8	16	32	48	64
Amplitude of the direct wave (mV)	759.4	759.8	760.1	762.3	763.9	761.1

electric domains, which is mainly determined by external stress and viscous forces [9]. The viscous constant that determines the viscous force is a function of the temperature and the frequency. Therefore the acoustical memory signals may be different at different frequencies and temperatures.

The hysteresis of the amplitude of the ultrasonic signals reflects the existence of defects in the crystal. No matter what kind of crystal, there are some defects in it. Because of the existence of these defects, the relationship between the stress and strain is no longer linear. Both Breazeale and co-workers' and our experiments testify that the acoustical memory is caused by the nonclassical nonlinearity of $LiNbO_3$; and the observed phenomenon of amplitude hysteresis of the acoustical memory is one of the features of nonclassical nonlinear properties. The central frequency of the memory signal is 2.44 MHz, so the frequency shift is about 60 kHz, which is another characteristic of nonclassical nonlinear acoustics.

The clear hysteresis of the acoustical memory signal and the first two harmonics of the direct wave is quite similar to that in magnetics. In magnetics, a hysteresis loop reflects a kind of memory phenomenon and this effect has been widely used. So the acoustical memory signal's hysteresis phenomenon also reflects some features of acoustical memory. The mechanism and application of this technique will need further study.

V. CONCLUSION

In this paper, the acoustical memory phenomenon is studied from 2.5 to 10 MHz. We have found that there is a hysteresis relationship between the memory signal and the driving force, which reflects the characteristics of nonclassical nonlinearity. The amplitude hysteresis of the first and second harmonics of the direct wave further reflects this kind of characteristic. The relationship between the amplitude of the memory signal and temperature is different from that found by Breazeale and co-workers. Second and third parts of the acoustical memory signal appear if a tone burst includes enough cycles or the driving force is high enough. We have made a qualitative analysis of these observations. Further research to be carried out as part of this project will help to explain the physical mechanism of the acoustical memory phenomenon. It may also provide further understanding of the crystal's inner structure and possible applications of the crystal using the acoustical memory phenomenon.

ACKNOWLEDGMENTS

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