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Photoexcitation of gigahertz longitudinal and shear acoustic waves in BiFeO₃ multiferroic single crystal

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Using femtosecond laser pulses, coherent GHz acoustic phonons are efficiently photogenerated and photodetected in BiFeO₃ (BFO) multiferroic single crystal. Due to the crystal lattice symmetry, longitudinal as well as two transverse acoustic modes are generated and detected, and the corresponding sound velocities are determined. This provides the opportunity to experimentally evaluate the elastic coefficients of the multiferroic compound BiFeO₃ that have been estimated so far only through *ab initio* calculations. The knowledge of the elastic properties of BFO is highly desired for BFO integration in nanoelectronic devices. Moreover, our findings highlight also that BFO may be a good candidate for light-controlled coherent acoustic phonons sources. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4719069>]

In recent years, multiferroics which are materials exhibiting simultaneous coexistence of polar and magnetic orders have attracted extensive attention due to their tremendous potential for various applications including memories, spintronic devices, sensors/actuators, or electro-optical systems.¹ Among them, BiFeO₃ (BFO) is the only compound so far displaying room temperature multiferroicity.² As a result, BFO has received special attention that led to a plethora of unexpected and exotic properties. Indeed, the various lattice order parameters^{3,4} are at the origin of several striking features. Remarkable examples are (i) the decrease of the Curie temperature with the magnitude of the strain^{5,6} in (100)-oriented BFO thin films, in contrast to normal ferroelectrics such as BaTiO₃, (ii) the observation of giant electromechanical properties,⁷ or (iii) multiple pressure instabilities.⁸ Besides, because BFO has a small bandgap (in contrast to normal ferroelectrics), properties such as visible-light photovoltaic effect,^{9,10} photostriction properties,¹¹ or THz electromagnetic wave generation by laser illumination have been recently explored.^{12,13} The mechanisms behind these properties are not fully understood yet as some traps arising from defects or Schottky barriers at electrode interface can affect the experimental observations.^{10,13} Nevertheless, it is believed that light excitation allows injection of photocarriers whose motion is driven by the built-in electric field linked to the large polarization of BFO. As a matter of fact and in addition to magnetic and/or electric field, light appears as a powerful tool to probe and tune BFO multiferroic properties as well as an original way for the discovery of unrevealed yet phenomena. More specifically, ultra-fast optical pump-probe spectroscopy can bring insights into the fundamental microscopic dynamics, important in many

applications, that are at the origin of functional properties of various materials such as metals,¹⁴ semiconductors,¹⁵ colossal magnetoresistive manganites,¹⁶ superconductors,¹⁷ or multiferroics.^{12,13,18} Such technique was used to investigate the model multiferroic BFO. As a result using femtosecond laser pulses, it was shown that coherent terahertz electromagnetic waves emission is possible.^{12,13} More interestingly, it was shown that actually the emitted terahertz radiation reflects the ultrafast modulation of the spontaneous polarization of BFO which offers approaches to readout in nonvolatile random access memories and ferroelectric domain imaging systems. In addition, such pump-probe technique allows access to the photo-induced coherent acoustic phonons dynamics¹⁸ which provides access to the determination of the elastic properties and also provides insight on the coupling of phonons and light. In this letter, we show that coherent acoustic waves are efficiently photogenerated and photodetected in BFO by using femtosecond laser pulses. We have determined the sound velocity and some elastic moduli for bulk BFO. These experimental values, which are very desired for BFO integration in micro-nanoelectronic devices, are in very good agreement with recent theoretical findings.¹⁹

At room temperature, the bulk BFO structure is described by a rhombohedral *R3c* space group with a perovskite pseudocubic unit cell elongated along the [111] polarization direction. For the purpose of this study, bulk BFO crystal was grown using flux method.²⁰ Our measurements were performed at room temperature on a [110] pseudocubic-oriented crystal (or [010] direction for rhombohedral symmetry) corresponding to a direction allowing the observation of both longitudinal acoustic (LA) and transverse acoustic (TA) phonons. It is worth to remind that in that direction the TA branches are non-degenerated. The pump probe technique used here consists in the optical excitation of the system by a femtosecond laser pump and the detection of the corresponding transient optical reflectivity

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using a femtosecond laser probe. The pump beam excites the crystal and acoustic phonons are emitted. These acoustic phonons create a strain field that propagates and modulates the refractive index. In turn, the refractive index modulation disturbs the optical reflectivity of the probe beam through photoelastic effect. Thanks to a control of the time arrival of the probe beam relative to the excitation by the pump beam, it is possible to monitor in the time domain the transient optical reflectivity of the BFO single crystal and then detect the propagation of the photogenerated acoustic phonons. The delay of the probe beam is accomplished by controlling its optical path with a moving mirror. The setup used in our study follows the classical scheme of picosecond acoustics.^{14,15,21,22} In the particular case of our study, the BFO sample is excited by a UV pump beam with 3.1 eV photon energy (see inset of Fig. 1). We remind that the direct band gap of BFO is around 2.8 eV.^{23,24} Thus these conditions permit above band-gap optical interband transitions and also lead to efficient deposition of energy, particularly for oxides,²⁵ within a rather thin skin depth that is estimated at about 40 nm.²³ The pump beam diameter of 15 μm is much larger than the pump optical penetration depth. As a result the photogenerated acoustic front is well described as a plane wave front. Due to the symmetry properties of the [110] pseudo cubic direction longitudinal as well as transverse acoustic modes are excited.^{26,27} The photogenerated coherent acoustic phonons are detected by the probe beam of 1.55 eV energy (i.e., below band-gap energy). This below-band-gap detection of phonons allows to probe elasticity deeply beneath the free surface (depth penetration larger than 1 μm). Due to momentum conservation during the interaction between the probe light and the moving acoustic front, only the acoustic phonon component with the Brillouin frequency (f_B) is detected.²¹ In the back-scattered configuration of our experiments, that momentum conservation is achieved for $q = 2k_{probe}$ where q is the momentum of acoustic phonon and k_{probe} the photon momentum of the probe beam with $k_{probe} = 2\pi\sqrt{n^2 - \sin^2(\theta)}/\lambda$ and λ , n , θ , the probe wavelength in vacuum, the refractive index of BFO at the wave-

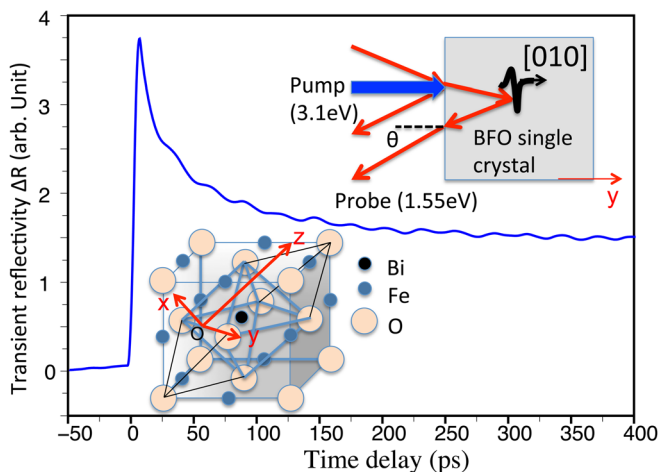


FIG. 1. Time resolved optical reflectivity of a [010] BiFeO₃ single crystal. (inset) Sketch of the two colors pump-probe experiment performed on a [010] BFO single crystal and representation, inside a face centered cubic cell, of the rhombohedral cell with the crystallographic axis (x , y , z).

length λ and the incidence angle of the probe beam respectively. Assuming the phonon dispersion law $2\pi f_B = V_S q$, where V_S is the sound velocity in BFO, the Brillouin frequency f_B component is then given by

$$f_B = \frac{2V_S\sqrt{n^2 - \sin^2\theta}}{\lambda}. \quad (1)$$

For the experimental signal this corresponds to a sinusoidal modulation of the optical reflectivity ΔR in the time domain. The origin of this oscillation can also be understood as the result of optical interferences between the beam reflected by the free surface and the beam scattered by the moving acoustic front (see inset of Fig. 1).²¹ In our case $\theta = 0^\circ$ and thus the Brillouin frequency is reduced to $f_B = 2V_S n/\lambda$. According to the dispersion curve in the [110]-pseudocubic direction,²⁸ the momentum conservation ($q = 2k_{probe}$) is achieved by the three modes (LA, TA1, TA2) that lead to a superposition of three sinusoidal components in the transient reflectivity signal with the frequencies of the corresponding acoustic modes. The transient optical reflectivity has been recorded up to a time delay of 500 ps (Fig. 1). Long-living oscillations are clearly detected. The first strong peak at a time delay of 0 ps is related to the ultrafast electronic dynamics that has been already studied^{12,13} and is out of scope of this paper. After subtracting the baseline of the transient reflectivity signal, the acoustic signal is extracted as shown in Fig. 2(a). Note that Takahashi *et al.*¹² found in their spectrum a broad oscillatory feature whose specific origin has remained obscure. Based on our measurements, we argue that this oscillatory feature corresponds to the acoustic waves and has nothing to do with any nonequilibrium dynamics in ferroelectrics.¹² An appropriate fast Fourier transform reveals three peaks at 12.5, 20, and 36 GHz (see Fig. 2(b)) corresponding to the three acoustic modes we have expected to observe.

Following Eq. (1) and assuming a refractive index ranging from $n = 2.6$ (Ref. 23) to $n = 2.9$,²⁴ the sound velocity of

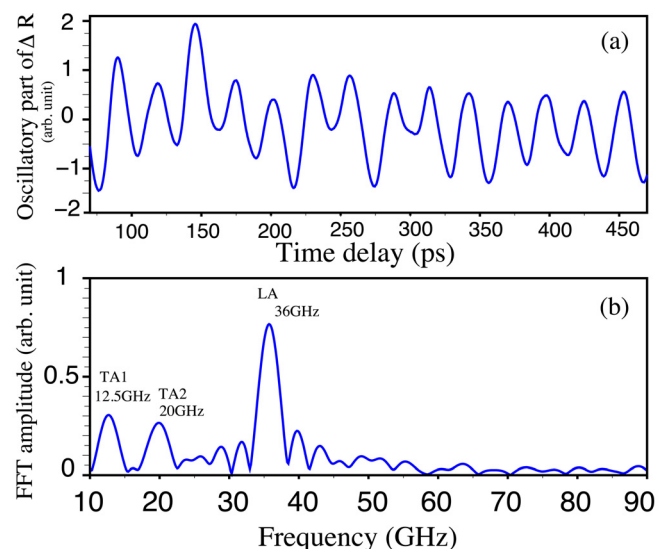


FIG. 2. (a) Brillouin oscillations detected in [110]-pseudocubic direction of BFO single crystal. (b) Fast Fourier transform revealing the three acoustic modes (TA1—12.5 GHz, TA2—20 GHz, and LA—36 GHz).

LA, TA1, and TA2 acoustic modes can be deduced. We obtain: $V_{LA} = 4965 - 5538 \text{ ms}^{-1}$, $V_{TA1} = 1720 - 1920 \text{ ms}^{-1}$, and $V_{TA2} = 2750 - 3070 \text{ ms}^{-1}$. Despite uncertainties of the order of 10% or less, arising from the experimental refractive index evaluation, these results are in good agreement with theoretical predictions based on first-principle calculations.¹⁹ From the relationships between the three differently polarized sound velocities in rhombohedral crystal and the elastic coefficients that follow²⁸

$$\rho V_{LA}^2 = C_{11}, \quad (2)$$

$$\rho V_{TA1}^2 = \frac{1}{2} \left[(C_{66} + C_{44}) - \{(C_{44} - C_{66})^2 + 4C_{14}^2\}^{1/2} \right], \quad (3)$$

$$\rho V_{TA2}^2 = \frac{1}{2} \left[(C_{66} + C_{44}) + \{(C_{44} - C_{66})^2 + 4C_{14}^2\}^{1/2} \right], \quad (4)$$

it is possible to extract some elastic coefficients. Using the BFO density of $\rho = 8340 \text{ kgm}^{-3}$, longitudinal acoustic mode leads to a value of $C_{11} = 205 - 255 \text{ GPa}$ which is in a very good agreement with the theoretical predictions $C_{11} = 220 \text{ GPa}$.¹⁹ From Eqs. (3) and (4), a single experimental measurement of V_{TA1} and V_{TA2} cannot allow the determination of the full set of the additional elastic constants because there are three unknown constants against two experimental inputs. However, we can show in the following that our experimental findings are in agreement with the theoretical estimates. The elastic constants C_{ij} have been calculated for the rhombohedral structure and for various unit cell volumes.¹⁹ At room temperature, x-ray diffraction experiments give a mean volume per atom of 0.0125 nm^3 . Therefore at this unit cell volume, we can extract from Shang *et al.*¹⁹ the elastic constants and deduce the transverse acoustic phonons velocities (TA1, TA2) that we can expect in the [110]-pseudocubic propagation direction (or [010] direction in the rhombohedral axis). Taking the following predicted values:¹⁹ $C_{11} = 220 \text{ GPa}$, $C_{33} = 140 \text{ GPa}$, $C_{12} = 120 \text{ GPa}$, $C_{13} = 60 \text{ GPa}$, $C_{44} = 35 \text{ GPa}$, $C_{14} = 20 \text{ GPa}$, $C_{66} = 1/2(C_{11} - C_{12})$ and using Eqs. (3) and (4), we find $V_{TA1} = 1625 \text{ ms}^{-1}$ and $V_{TA2} = 2825 \text{ ms}^{-1}$. These values are in good agreement with the measurement data, even if the TA1 mode value is slightly out of the range determined experimentally.

In conclusion, we have shown that GHz acoustic waves can be generated and detected in the model BiFeO₃ multiferroic compound using femtosecond light excitation. We have determined the sound velocities of longitudinal and transverse acoustic phonons along the [110]-pseudocubic direction. A good agreement is found with previous *ab initio* calculations. These constants, which were so far unknown, are unavoidable for future integration of BFO in micro/nano-devices. By highlighting the photo-induced acoustic waves, our findings bring an essential brick to the large potential applications of the famous BFO multiferroic compound. As a perspective work, the mechanism of photogeneration of these LA and TA waves remains to be elucidated. It is expected that the inverse piezoelectric process is an efficient mechanism^{14,29} because of the large internal remanent polarization in BFO. However, this is not the only possible mechanism to explain these induced

coherent acoustic phonons as it has been already discussed in piezoelectric materials,²⁹ and further investigations are required in order to establish the physics behind the emitted acoustic waves. Nonetheless BFO system could be a good candidate for the development of optically triggered longitudinal and shear piezotransducers.

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