Tbit/inch² Ferroelectric Data Storage Based on Scanning Nonlinear Dielectric Microscopy

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Scanning Nonlinear Dielectric Microscopy (SNDM) is the method for observing ferroelectric polarization distribution, and now, its resolution has become to the sub-nanometer order, which is much higher than other scanning probe microscopy (SPM) methods for the same purpose. Up to now, we have studied high-density ferroelectric data storage using this microscopy. In this study, we have conducted fundamental experiments of nano-sized inverted domain formation in LiTaO₃ single, and successfully formed inverted dot array with the density of 1.5 Tbit/inch².

Key words: Ferroelectric data storage, Scanning nonlinear dielectric microscopy, Tbit/inch²

1. INTRODUCTION

With the advance of information processing technology, the importance of high-density data storage is increasing. Studies on thermal fluctuation predicts that the magnetic storage, which plays a main role in this field, will reach a theoretical limit in the near future, and a novel high-density storage method is required. So, we have studied the application of ferroelectric materials to high-density storage media. Ferroelectrics can hold bit information in the form of a polarization direction. Moreover, the domain wall of typical ferroelectric material is as thin as the order of a few lattices [1]. This feature is favorable for high-density data storage.

Recently, there are many reports on inverted domain formation in ferroelectric thin film using scanning probe microscopy (SPM). In this technique, inverted domain dots are formed by applying a relatively large voltage to the probe. These dots can be detected by piezoelectric imaging technique. Up to now, 6 Gbit/cm² (39Gbit/inch²) artificial domain dot array formed in lead zirconate titanate (PZT) thin film has been reported [2]. However, in order to achieve the recording density of Tbit/inch² order, improving the resolution of the domain detection device and the physical properties of the ferroelectric medium are required.

The present authors have recently developed and reported scanning nonlinear dielectric microscopy (SNDM) for observing ferroelectric polarization distribution [3-5]. The resolution of this microscopy is sub-nanometer order [4], which is much higher than other scanning probe microscopy (SPM) methods for observing polarization distribution [6]. So SNDM is considered to be a powerful tool for ferroelectric domain engineering. Moreover, SNDM is promising for a pickup head of a ferroelectric high-density storage system, because this method is purely electrical, and therefore high-speed data transfer can be realized using this method.

On the other hand, in regard to recording medium, we consider that single crystal materials are more suitable than thin film materials for studying nano-domain engineering quantitatively with good reproducibility, because atomic scale non-uniformity in current thin film becomes serious obstacle to nano-sized inverted domain formation. To date, barium titanate (BaTiO₃) is the only material that has been studied as a single-crystal material for nano-domain switching [7]. Although BaTiO₃ is a typical ferroelectric material, it is not suitable for a recording medium with a number of reasons. Most importantly, BaTiO₃ single crystal belongs to the tetragonal system, with the result that it has two possible domains in which bits are stored, a 90° a-c domain and a 180° c-c domain. Such a structure introduces a level of complexity not desired at present. Furthermore, the phase transition from the tetragonal phase to the orthorhombic phase occurs at 5 °C, which is dangerously close to room temperature for a storage medium, possibly resulting in data loss as a result of ambient temperature drift. Lastly, it is difficult to fabricate large, high quality, and practical usable BaTiO₃ single-crystal at low cost.

A ferroelectric nano-domain engineering material suitable for practical application as a storage medium should have a single 180° c-c domain, an adequately high Curie point without phase transitions below the Curie point, and be producible as large single-crystals with good homogeneity at low cost. Lithium tantalate (LiTaO₃) single-crystal satisfies all these conditions. In this study, we used stoichiometric LiTaO₃ (SLT) [8] besides congruent LiTaO₃ (CLT), which is used widely in optical and piezoelectric devices. A number of the pinning sites of SLT, which is derived from nonstoichiometric defects, is much less than that of CLT. The pinning sites have strong effects on domain inversion characteristics as follows: (1) The coercive field of SLT is much smaller than that of CLT [9]. (2) The domain switching time of SLT is much less than that of CLT. (3) The sidewise-growth velocity of SLT is much faster than that of CLT [10,11]. So we expected that high-speed recording could be realized using SLT, whereas high-density data storage could be realized using CLT.

2. EXPERIMENTAL DETAILS

SNDM observes the distribution of ferroelectric domains by detecting the lowest order of nonlinear dielectric constant ε_{333} , because the sign of ε_{333} changes in accordance with the inversion of the spontaneous polarization. Fig. 1 shows the schematic diagram of SNDM domain engineering system. The probe composed of a LC resonant circuit and a metal needle detects the capacitance of the specimen immediately under the tip (denoted by C_s). When alternating voltage $E_p \cos \omega_p t$ is applied on the specimen, C_s alters on account of nonlinear dielectric response. The ratio of the alternating variation ΔC_s to the static value of capacitance C_{s0} without time dependence is given as

$$\frac{\Delta C_{\rm s}(t)}{C_{\rm s0}} = \frac{\varepsilon_{333}}{\varepsilon_{33}} E_{\rm p} \cos \omega_{\rm p} t + \frac{\varepsilon_{3333}}{4\varepsilon_{33}} E_{\rm p}^2 \cos 2\omega_{\rm p} t \qquad (1)$$

where ε_{33} is a linear dielectric constant and ε_{333} and ε_{3333} are nonlinear dielectric constants. Therefore, ε_{333} is



Fig. 1. Schematic diagram of SNDM domain engineering system.

obtained by means of detecting the component of the probe signal with frequency of ω_p using lock-in technique. In this study, we used a conductive cantilever with a radius of 25nm as a needle of a probe, and contact load of the cantilever is kept constant using AFM mechanism.

Inverted domains are formed by applying a voltage pulse between the probe needle and the metal stage of SNDM. In this case, the electric field just under the tip is highly concentrated, which is much different from the case of using parallel plate electrodes. The necessary condition that an inverted domain remains stably after finishing voltage application is that the inverted region penetrates to the backside of specimen. This means that, in order to form an inverted domain using a needle with a sharp point, it is necessary to reduce the thickness of the specimen. In this study, we made SLT and CLT plates with a thickness of about 100 nm using dry etching technique. At first, we studied domain inversion characteristics in small area using SLT thin plate. Fig. 2 shows the SNDM images of an inverted domain formed in SLT by means of applying a voltage pulse with amplitude of 15 V and duration time of (a) 500 nsec (b) 100 nsec (c) 60 nsec. From these figures, we found that the area of the domain decreases with decreasing voltage application time. More detailed experimental result with regard to the relationship between the radius of the inverted domain and voltage application time is shown in Fig. 3.







Fig. 3. Relationship between the radius of the inverted domain and voltage application time.

From this result, we found the power law dependence of the radius of the inverted domain r on voltage application time t. We also found that domain wall velocity v = dr/dt declined with expansion of the



Fig.4. The smallest inverted domain at the present time, which was formed in SLT by applying 15V, 60nsec pulse. inverted domain.

Fig. 4 shows the smallest inverted domain at the present time. The radius of this domain is 6 nm, which corresponds to storage density of 4 Tbit/inch² if more than one dots can be formed closely without interference.

The result of studying the retention of small inverted domains is depicted in Fig.5. These images are observed (a) 50 minutes after pulse application (b) 8 hours after pulse application (c) 24 hours after pulse application. From this result, we found that small inverted domains remained stably for a long time.

Subsequently we have also studied inverted domain formation in CLT. Figure 6 shows the images of the inverted domain pattern with density of (a) 0.62 Tbit/inch² (b) 1.10 Tbit/inch² (c) 1.50 Tbit/inch². These domain patterns were formed by applying voltage pulses with amplitude of (a) 11 V (b), (c) 12 V, and duration time of (a) 10 µsec (b) 500 nsec (c) 80nsec.

Fig. 7 (a) shows the SNDM image of 5×5 inverted domain dot array formed in CLT by means of applying 12V, 1ms voltage pulses, and inverted domain dots were sequentially erased by means of applying -12V, 10ms voltage pulses as shown in Fig. 7 (b)-(e).



Fig.5. Images of inverted domain dot array in SLT. These images are observed (a) 50 minutes after pulse application (b) 8 hours after pulse application (c) 24 hours after pulse application.



Fig.6. Images of the inverted domain pattern formed in CLT with density of (a) 0.62 Tbit/inch² (b) 1.10 Tbit/inch² (c) 1.50 Tbit/inch².



Fig. 7. (a) Image of 5×5 inverted domain dot array formed in CLT by means of applying 12V, 1ms voltage pulses. (b)-(e) Inverted domain dots were sequentially erased by means of applying -12V, 10ms voltage pulses.

3. CONCLUSIONS

We have conducted fundamental experiments of inverted domain formation in LiTaO₃ single crystal using SNDM. First, we studied the relationship between the size of inverted domain and voltage application time in SLT. As a result, we successfully formed inverted domain with a radius of 6 nm. We also confirmed that small inverted domain remained stably at least for 24 hours. Subsequently we studied inverted domain formation in CLT, and we achieved 1.5 Tbit/inch² data storage. Therefore we consider that ferroelectrics are promising materials for high-density data storage in a next generation.

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