Zn Vacancy Ordering in $Ba_3(Zn_{1/3}Ta_{2/3})O_3$ Ceramics

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There have been a lot of investigations on the dielectric properties and microstructure of $Ba(Zn_{1/3}Ta_{2/3})O_3$ (BZT)[1,2] because of its application to satellite communication. According to the previous studies, Zn:Ta (1:2) ordering exists in BZT and influences the dielectric properties of the system[3,4]. In this work, a new type of ordering different from the 1:2 ordering was found in BZT using transmission electron microscopy (TEM). Moreover, unit cell of ordered structure was suggested and confirmed by the computer simulation of diffraction pattern.

$Ba(Zn_{1/3}Ta_{2/3})O_3$ pellets were prepared by the mixed oxide method and the relevant homogeneous mixtures were pre-fired at 1100°C for 2 hours, ground, pressed into pellets and finally sintered at 1400°C for 2 - 90 hours. TEM specimens were prepared by the conventional method. Hitachi H-9000 NAR and Philips CM20T/STEM microscopes were used to observe the specimens.

Figure 1 is the [010] zone axis diffraction pattern of specimen sintered at 1400°C for 90 hours. As shown in this figure, 1:2 (Zn:Ta) ordering is developed along the c-direction with the wavelength of 0.7 nm. Most of the grains in the specimen have the diffraction pattern shown in Fig. 1. However, diffraction pattern shown in inset of Fig. 2

was also observed in the limited area. The analysis revealed that it is the [001] zone axis diffraction pattern with extra reflection at 1/3(110) position. The interplanar distance calculated from the extra reflection was about 0.9 nm which is three times larger than the interplanar distance of (110) plane.

In order to develop a new ordered structure along the [110] direction observed in this study, one of the defects in cation sites should be ordered. Since ZnO compound is volatile and ZnO loss was observed in the specimens sintered at high temperature (>1400°C) or for long time (> 60 hours) at 1400°C[4], a new type ordering is due to the Zn vacancy ordering. A superlattice containing three BZT unit cells is suggested as the unit cell of ordered structure. In this unit cell, Zn vacant sites were located at (00½). Figure 3(a) shows the ordered Zn vacancy structure projected along [001] direction. The computer simulation of diffraction pattern was carried out using the structure shown in Fig. 3(a) and the simulation result is shown in Fig. 3(b). The simulated diffraction pattern matches exactly the experimental result shown in inset of Fig. 2.

References

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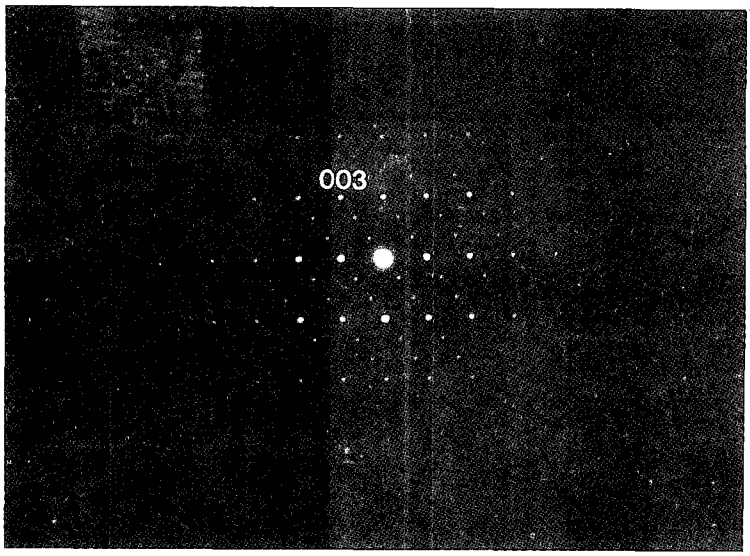

Fig. 1. The [010] zone axis diffraction pattern of specimen sintered at 1400°C for 90 hours.

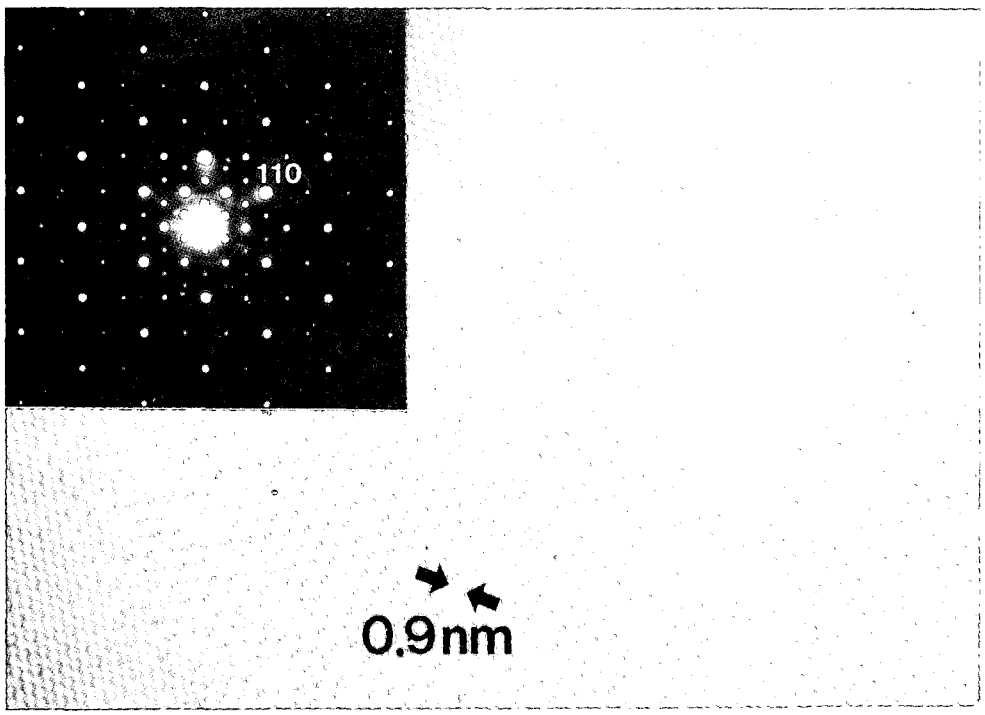

Fig. 2. High resolution lattice image with [001] beam direction showing the modulation along the [110] direction taken from BZT sintered at 1400°C 90 hours.

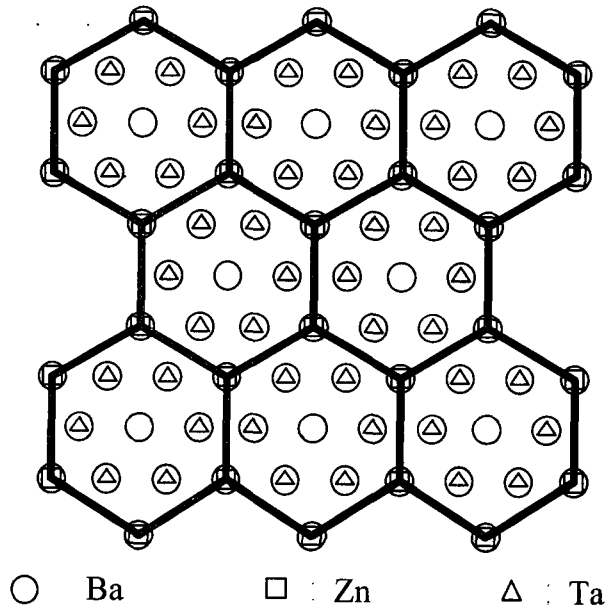

Fig. 3. (a) Ordered Zn vacancy structure projected along the [001] direction.

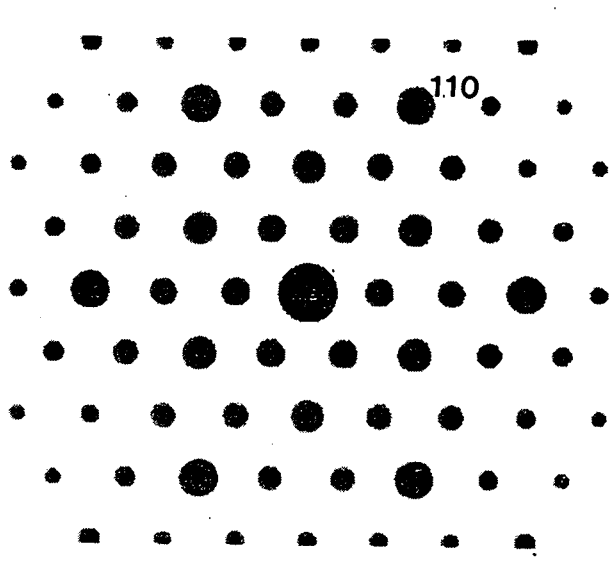

Fig. 3. (b) Computer simulated diffraction pattern with [001] beam direction using the structure shown in Fig. 3(a).