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ABSTRACT

By a roller quenching technique amorphous ribbons of $LiNbO_3$ and $PbTiO_3$ were obtained. These ribbons were examined by electron diffraction as well as by X-ray diffraction experiments, resulting in obtaining them in amorphous state.

INTRODUCTION

By rapid cooling from the liquid state, ferromagnetic amorphous materials[1] had been previously obtained in iron group alloys with Si, B, C and P as glass form atoms. We have reported many unusual magnetic properties, in particular, magnetoelastic coupling phenomena[2] including giant magnetomechanical coupling and giant ΔE effect which, in turn, realize a dramatic change in the propergation time of the magnetoelastic wave in a highly magnetostrictive amorphous transmission ribbon by applying a magnetic field. It is of great interest if we can obtain dielectric amorphous ribbons by a similar rapid quenching technique from their melts. When we can obtain such a ribbon, it is possible to expect similar unusual dielectric properties, as mentioned above, by prohibiting the crystal formation to kill the crystalline anisotropy.

An attempt has been made of the preparation of such highly dielectric amorphous ribbons by a modified roller quenching method employed in the above study. In this paper, we report the evidence of amorphous state in $LiNbO_3$ and $PbTiO_3$ ribbons thus obtained.

EXPERIMENTAL PROCEDURES AND RESULTS

A ferroelectric oxide, $LiNbO_3$ was melted in a platinum crucible at 1350°C for a few hours, and an ingot was obtained. In order to get the stoichometric composition, a special care was taken about the composition before melting. A platinum-rhodium nozzle with a small hole 0.3 mm in diameter was used, in which a rod cut out from the above ingot was melted again at the temperature just above its melting point. By employing the roller rapid quenching technique, thin ribbons were obtained. They were about 5~20 μm in thickness and about 0.4 mm in width. Ribbons of $PbTiO_3$ could also be prepared by a similar technique. It should be noted that an addition of a certain amount of suitable glass formers enables us to obtain very long, wider and even ribbons.

Before the structure analysis, ribbons were examined by a polarizing microscope with cross Nicol's prisms. In one series of this observation the temperature of the specimen was changed from room temperature to 1000°C in a special micro-furnes. The ribbons were found to be transparent, very clear and optically isotropic at room temperature. During heating, slight gradual changes of color and transparency, which are corresponding to crystalization, were observed between 400~500°C and 450~500°C for $LiNbO_3$ and $PbTiO_3$ respectively.

For the direct observation of the structures of these ribbons, transmission electron diffractometry was applied. Each ribbon was cut into short piece about 1 mm in length and placed between two copper meshes. In most observations using JEM-1000 electron microscope, an accelerating voltage was 1 million volts and $L\lambda$ = 20 mmA. In one series of experiments, the amorphous ribbons were beam-heated in the microscope, and the same thin heated area was re-examined. Typical electron diffraction pattern is shown in Fig. 1 for an amorphous $LiNbO_3$ as-rolled ribbon.

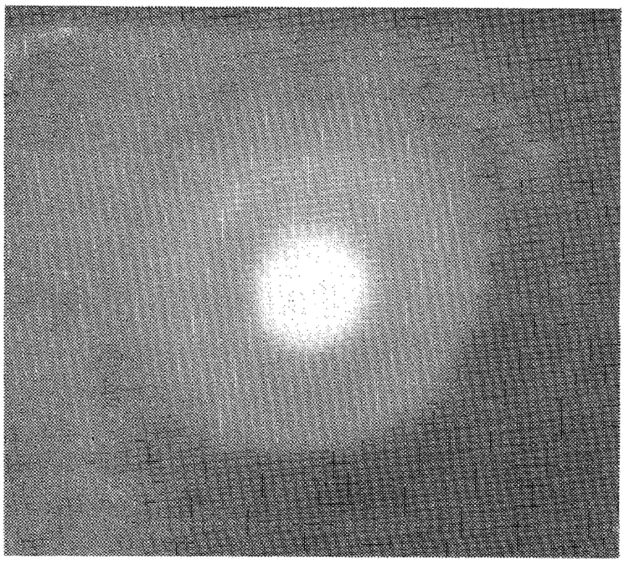

Fig. 1 Electron diffraction pattern of an amorphous $LiNbO_3$ as-rolled ribbon.

The line profile of this halo pattern is qualitatively similar to that of typical amorphous materials. It is quite clear that the electron diffraction pattern does not have sharp Bragg-type diffraction maxima.

The structures of their ribbons were also examined by X-ray diffraction. The X-ray measurements were made by D-9C diffractmeter using Cu-K_α radiation. Incident beam was monochromatized by a quartz single crystal. Special pracautions were taken to set the goniometer and the monochrometer so as to reduce the difference in the intensities of halo reflection as both sides of the incident beam to a few percent. An example of X-ray diffraction pattern is shown in Fig. 2 for an

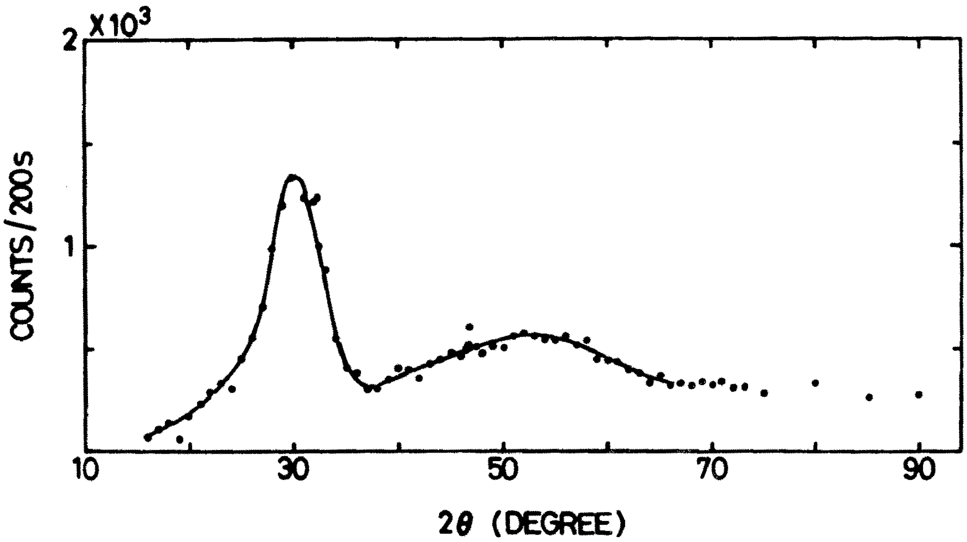

Fig. 2 X-ray diffraction pattern of an amorphous $PbTiO_3$ ribbon. Where the vertical axis shows stop counts of diffraction X-ray per 200 seconds, and 2θ is the angle between incident and diffracted X-ray.

amorphous $PbTiO_3$ ribbon. This pattern is also a typical one of the amorphous state.

It is interesting to mention the dielectric properties of these amorphous ribbons which exhibit no local symmetry but a certain symmetry as a whole. At room temperature, the value of dielectric constant in amorphous $LiNbO_3$ was approximately twice that in a single crystalline platelet. When heating, Glass, Lines, Nassan and Shiever found remarkable maxima of the dielectric constant[3], about $4x10^5$, at about 350 and 500°C in a roller quenched $LiNbO_3$. We believe that their ribbon is in amorphous state, although its structure analysis has not been published.

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