Title: Radiation Damage Effects in Ferroelectric LiTaO3 Single Crystals

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RADIATION DAMAGE EFFECTS IN FERROELECTRIC LiTaO$_3$ SINGLE CRYSTALS

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ABSTRACT

Z-cut lithium tantalate (LiTaO$_3$) ferroelectric single crystals were irradiated with 200 keV Ar^{++} ions. LiTaO$_3$ possesses a structure that is a derivative of the corundum (Al$_2$O$_3$) crystal structure. A systematic study of the radiation damage accumulation rate as a function of ion dose was performed using ion-beam channeling experiments. An ion fluence of $2.5 \cdot 10^{18}$ Ar^{2+} ions/m^2 was sufficient to amorphize the irradiated volume of a LiTaO$_3$ crystal at an irradiation temperature of ~120K. This represents a rather exceptional susceptibility to ion-induced amorphization, which may be related to a highly disparate rate of knock-on of constituent lattice ions, due to the large mass difference between the Li and Ta cations. We also observed that the c$^-$ end of the ferroelectric polarization exhibits slightly higher ion dechanneling along with an apparent greater susceptibility to radiation damage, as compared to the c$^+$ end of the polarization.

INTRODUCTION

Lithium tantalate (LiTaO$_3$) is a ferroelectric oxide possessing large nonlinear optical coefficients [1] as well as large electro-optic coefficients [2]. As such, LiTaO$_3$ crystals are of interest for nonlinear optical applications as well as for surface acoustic wave and piezoelectric devices [3]. LiTaO$_3$ is a derivative structure of corundum (α-Al$_2$O$_3$) with space group R3c. Recently, there has been considerable interest in the radiation damage tolerance of corundum structural derivative rhombohedral oxides [4-7]. The purpose of the study presented here is to assess the ion beam radiation damage resistance of lithium tantalate, especially in the context of ion beam irradiations under cryogenic conditions.

EXPERIMENT

Synthetic Z-cut LiTaO$_3$ crystals were obtained from Yamaju Ceramics Co., Ltd., (Crystal Technology Division, No. 1123 Sango-Cho, Owariasashi-shi Aichi-ken, Japan). Crystal wafers were 51 mm diameter and 0.5 mm thick, polished to a mirror finish on both sides. Samples were irradiated with 200 keV Ar^{++} ions at an inclination of a few (~10) degrees to the c <0001> crystallographic axis, in order to minimize channeling effects. The ferroelectric polarization of these crystals, **P**$_s$, is oriented along the c axis, i.e. normal to the front and back surfaces of each LiTaO$_3$ wafer. The c$^+$ axis is defined as the surface normal parallel to **P**$_s$, while the c$^-$ axis is antiparallel to **P**$_s$. Samples were irradiated at cryogenic temperature using a sample stage cooled to ~120K by liquid nitrogen conduction cooling.

TRIM simulations [8] were performed to determine the projected range of 200 keV Ar ions in LiTaO$_3$ and to determine the instantaneous displacement damage induced by this irradiating

species. Simulations indicate that the longitudinal range of Ar ions in $LiTaO_3$ is ~120 nm, while the longitudinal straggling is ~51 nm. The peak Ar concentration is about 0.8 at.% per 10^{19} Ar/m^2, while the peak number of displacements per atom (dpa) is approximately 0.8 dpa / 10^{19} Ar/m^2.

Unirradiated and irradiated lithium tantalate crystals were analyzed by Rutherford backscattering spectroscopy and ion channeling (RBS/C), using 2 MeV He^+ ions for the analyses. Minimum backscattering yield, χ_{min}, as defined by the ratio between the RBS/C ion yield in an aligned spectrum versus a random spectrum, was measured in both unirradiated and irradiated samples to determine radiation damage accumulation rates in $LiTaO_3$.

RESULTS

Figure 1 shows ion channeling spectra from both surfaces of an $LiTaO_3$ unirradiated single crystal wafer. A difference in He^+-ion backscattered yield is apparent in Fig. 1, wherein more dechanneling is observed along c^- compared to c^+. This difference may be due either to: (1) dechanneling induced by the polarization P_s; or (2) to the existence of an internal field, E_{int}, whose presence is attributed to lattice defects such as OH- ions and nonstoichiometric, charge-compensating defects [3, 9].
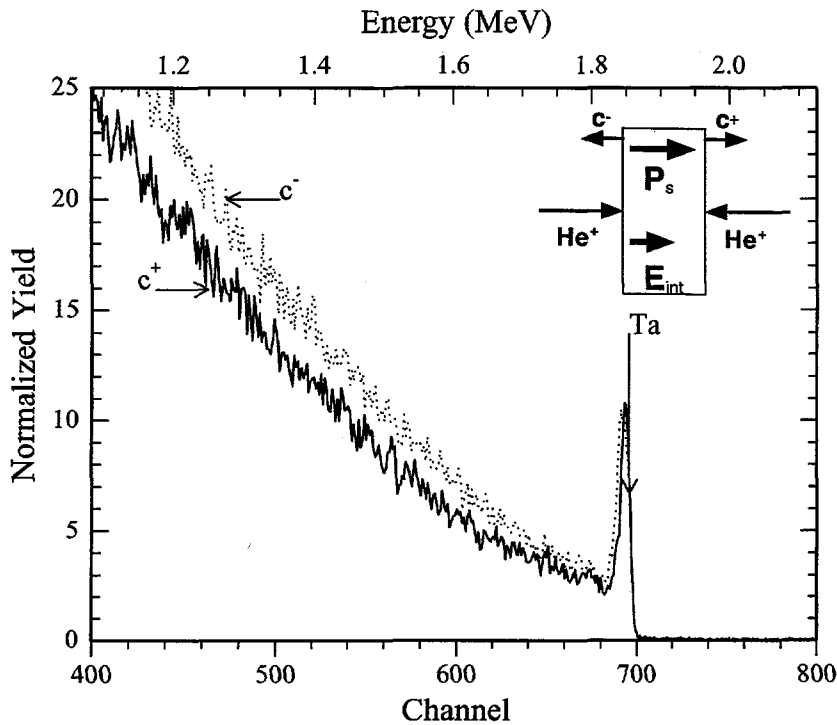

Figure 1. He^+ ion channeling from an unirradiated $LiTaO_3$ single crystal, with the He^+ beam aligned along the c-axis and with the ferroelectric polarization, P_s, aligned either parallel (c^+) or antiparallel (c^-) to the c-axis.

Figure 2 shows He+ ion RBS/C spectra obtained from the same crystal as in Fig. 1. In this plot, more dechanneling is apparent along c^+ compared to c^-. The difference compared to Fig. 1 is that the ferroelectric polarization of the crystal has been reversed at room temperature using an electric field of ~21 kV/mm. At room-temperature, this applied field is able to reverse the polarity, P_s, but is not of sufficient strength to reverse the sense of the internal field, E_{int} [3].

Figure 2. He$^+$ ion channeling spectra from an unirradiated LiTaO$_3$ single crystal, with the He$^+$ beam aligned along c$^+$ and c$^-$, respectively.

Referring to the schematic insets in Figs. 1 and 2 (representing edge-on views of a thin, single crystal LiTaO$_3$ wafer), the maximum dechanneling is observed on the right-hand surface, irrespective of the orientation of $\mathbf{P}_s$.
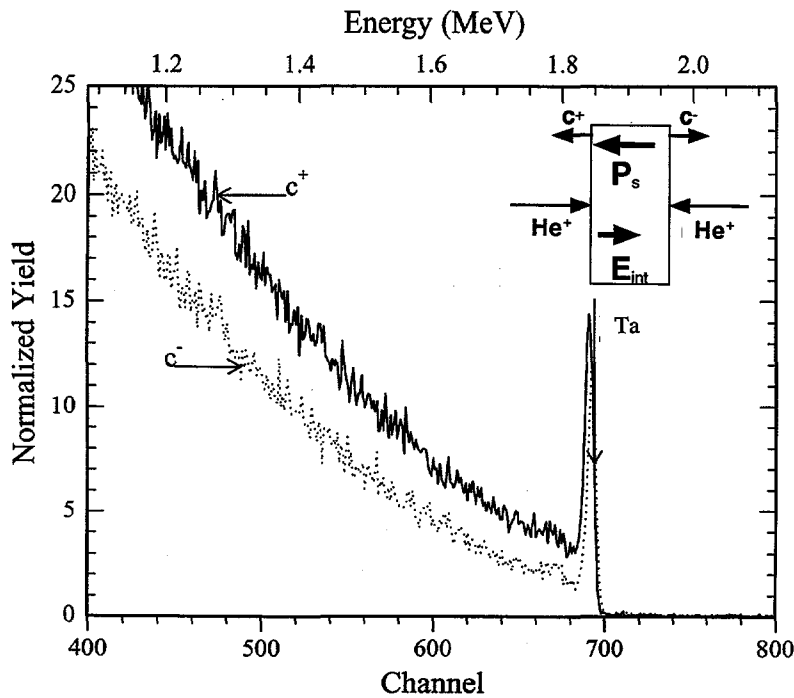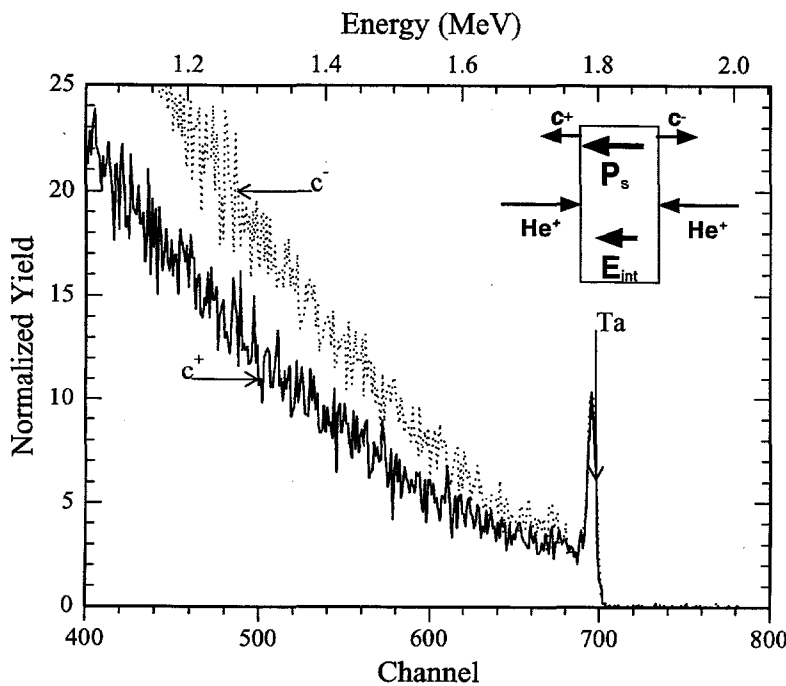

Figure 3. He$^+$ ion channeling spectra from an unirradiated LiTaO$_3$ single crystal, with the He$^+$ beam aligned along c$^+$ and c$^-$, respectively.

Figure 3 show RBS/C spectra from the same LiTaO$_3$ crystal as in Figs. 1 & 2. More dechanneling is apparent along c⁻ compared to c⁺. The difference here compared to Fig. 2 is that the crystal was annealed at 520K for one hour in air. This treatment is sufficient to reverse the sense of the internal field, $\mathbf{E}_{int}$, without affecting the polarization, $\mathbf{P}_s$ [3, 9]. Clearly, the results in Figs. 1-3 indicate that c-axis dechanneling differences are dependent on the orientation of $\mathbf{E}_{int}$, not $\mathbf{P}_s$.
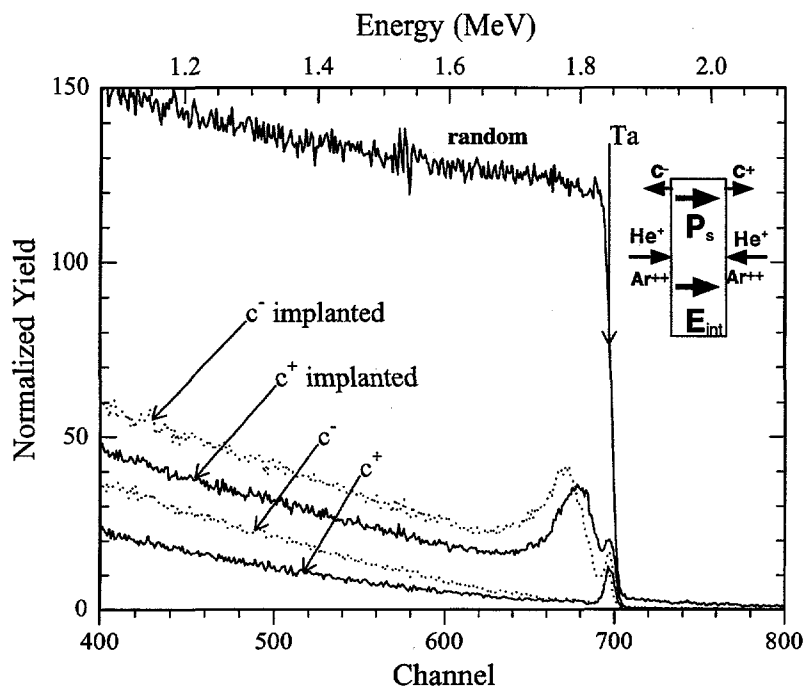

Figure 4. Ion channeling spectra from: (1) an unirradiated LiTaO$_3$ single crystal (spectra labeled "c⁺" and "c⁻"); and (2) an LiTaO$_3$ crystal irradiated with 200 keV Ar⁺⁺ ions to a fluence of 1•10¹⁸ Ar⁺⁺/m² at a substrate temperature of ~120K (spectra labeled "c⁺ implanted" and "c⁻ implanted"). The He⁺ beam is aligned along either the c⁺ or c⁻ direction in each spectrum, except for the spectrum with highest ion yield (labeled "random").
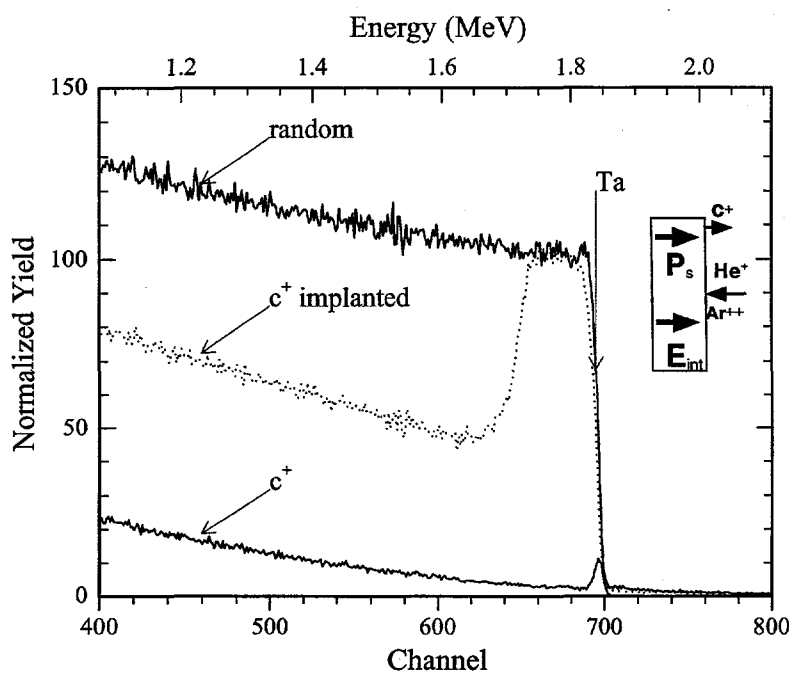

Figure 5. 2 MeV He⁺ RBS/C spectra from: (1) an unirradiated LiTaO$_3$ single crystal (channeling spectrum labeled "c⁺" and random spectrum labeled "random"); and (2) an LiTaO$_3$ crystal irradiated with 200 keV Ar⁺⁺ ions to a fluence of 2.5•10¹⁸ Ar⁺⁺/m² at a substrate temperature of ~120K (spectrum labeled "c⁺ implanted").

Figure 4 shows 2 MeV He+ RBS/C spectra from both an unirradiated LiTaO$_3$ wafer (both c- and c+ surface orientations, and a LiTaO$_3$ crystal irradiated to a fluence of $1 \cdot 10^{18}$ Ar^{++}/m^2 at a substrate temperature of ~120K. In this plot, more dechanneling is observed following Ar^{++} ion implantation along both c$^+$ and c$^-$ orientations and dechanneling along c$^-$ is greater than c$^+$, in either the unirradiated or irradiated condition. As indicated in the schematic inset, the enhanced dechanneling along c$^-$ is due to the antiparallel sense of the internal field, $\mathbf{E}_{int}$, with respect to the c$^-$ axis used in these experiments.

Figure 5 shows RBS/C spectra from an unirradiated LiTaO$_3$ single crystal an LiTaO$_3$ crystal irradiated with 200 keV Ar^{++} ions to a fluence of $2.5 \cdot 10^{18}$ Ar^{++}/m^2 at a substrate temperature of ~120K. In this plot, the dechanneling yield in the near surface region (i.e., for energy losses in the vicinity of the energy for surface tantalum scattering (labeled "Ta")) reaches the random level. This is indicative of an amorphization transformation in the implanted volume. Amorphization of LiTaO$_3$ due to ion irradiation has been confirmed in a related study [4].

Figure 6 summarizes the radiation damage accumulation results of several Ar^{++} ion irradiations of c$^+$ oriented LiTaO$_3$ crystals (with $\mathbf{E}_{int}$ parallel to c$^+$). These results indicate that LiTaO$_3$ is rather easily amorphized, upon a peak damage level of about 0.2 dpa. This represents a rather exceptional susceptibility to ion-induced amorphization, which may be related to a highly disparate rate of knock-on of constituent lattice ions, due to the large mass difference between the Li and Ta cations.
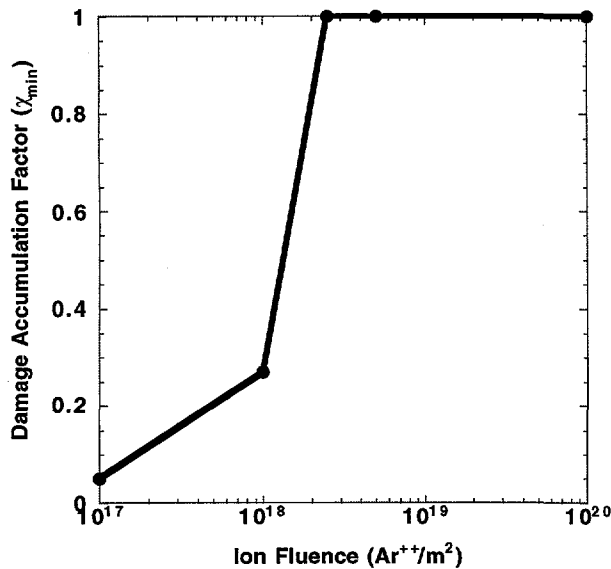

Figure 6. Summary of ion channeling measurements of Ar^{++} ion irradiated LiTaO$_3$ single crystals. This plot shows the damage accumulation fraction, χ_{min}, as a function of Ar^{++} ion fluence. χ_{min} is the ratio of He$^+$ ion backscattered yield in a c-axis channel orientation to the He$^+$ ion yield in a 'random' orientation, for ion backscattering over an energy range equivalent to 100 RBS/C multi-channel analyzer channels.

CONCLUSIONS

- c-axis ion channeling in $LiTaO_3$ single crystals is dependent on the orientation of the internal field, E_{int}, a field associated with lattice point defects.

- At cryogenic temperatures, ion irradiation damage accumulation in $LiTaO_3$ is evident at low doses and amorphization is observed at a peak damage level of about 0.2 dpa.

ACKNOWLEDGMENTS

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