Range and Annealing Behaviour of Pb^+ Ions Implanted into LiNbO_3 Crystals at Moderate Energies *

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Pb^+ ions have been implanted into LiNbO_3 crystals in the energy range of 100–350 keV at doses of 5×10^{15} , 1×10^{16} and 2×10^{16} ions/cm². The profile of the implanted ions was measured by Rutherford backscattering. The mean projected range and the range straggling obtained from the experiment were compared with the TRIM'98 and SRIM 2000 code. In the present case, the TRIM'98 code predicts experimental values better than those of the SRIM 2000 code. The depth distribution is also found to be independent of dose for 350 keV Pb^+ implanted into LiNbO_3 crystals. After 800°C annealing for 60 min in ambient air, obvious diffusion occurs to the implanted Pb^+ ions at 150 keV with a dose of 5×10^{15} ions/cm². After a low-temperature treatment at 77 K in liquid nitrogen, no obvious diffusion phenomenon occurs for the implanted Pb^+ ions in LiNbO_3 .

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In recent years, ion implantation has become a superb method for improving surface properties of materials since it offers accurate control of dopant composition and structural modification at any selected temperature.^[1,2] Recently, there has been an increasing scientific interest in the implantation of polarizable ions for material modification because the polarization in the materials could be remarkably changed by the implanted ions.^[3] Polarizable ions such as Au, Ag, and Bi have successfully been used to implant into many materials for optical and magnetic applications.^[4–6] Since the Pb ion also has high polarizability, many Pb^+ -implanted materials such as lead-doped silicate glass have been of great interest for potential applications.^[3]

Lithium niobate (LiNbO_3) is an outstanding material due to its high electro-optic and nonlinear optical coefficients and due to the fact that large single crystals are available at reasonable cost.^[7] Optical waveguides have been formed in LiNbO_3 crystals by MeV ion implantation, and ion-implanted waveguide lasers have also been prepared with LiNbO_3 .^[8,9] The implantation of polarizable ions into LiNbO_3 crystals and subsequent thermal annealing may provide a method of achieving desirable optical properties. Metal ions such as Au, Ag, and Cu at energies from 100 keV to 3 MeV have been implanted into LiNbO_3 crystals and many experimental data of the optical properties of the implanted LiNbO_3 samples have been obtained.^[4–6,10] Therefore, an accurate knowledge of depth distributions of implanted ions in solids is important for many modern applications.

To our knowledge, there has been no report on the study of the range distribution of implanted keV Pb^+ in LiNbO_3 . In the present work, data are represented

on depth distributions of the implanted Pb^+ ions with moderate energies (100–350 keV) in a LiNbO_3 crystal. The experimental mean projected range and the range straggling are compared with the TRIM'98 Monte Carlo code (transport of ions in matter) and the SRIM 2000 code (stopping and range of ions in solids). The profiles of the implanted Pb^+ ions in LiNbO_3 at low, room and high temperatures are also obtained.

The x -cut LiNbO_3 samples were optically polished and cleaned with the dimensions of $6 \times 5 \times 1.5$ mm³. The samples were implanted with different doses ranging from 5×10^{15} to 2×10^{16} Pb^+ ions/cm² at room temperature. The energies of the implanted Pb^+ ions were ranged from 100 to 350 keV. During the implantation, all the samples were tilted 7° off the incident beam direction to avoid the possible channelling of Pb^+ ions. In order to study the depth distribution of implanted Pb^+ in LiNbO_3 at high and low temperatures, the samples implanted at 150 keV with a dose of 5×10^{15} Pb^+ /cm² were placed at 800°C in ambient air as well as at 77 K in liquid nitrogen.

The range profiles of implanted ions were measured by Rutherford backscattering (RBS) of MeV He^{2+} ions. The backscattered He particles were detected by a Si surface barrier detector. The detector was kept at an angle of 165° with respect to the incident He^{2+} beam. The measurement was performed using the 1.7 MV tandem accelerator at Shandong University. The He^{2+} beam current was typically 1 nA to avoid a pile up.

The present data indicate that the range distribution is nearly symmetrical for our experiments.^[11] Therefore we can use a simple Gaussian to describe the measured depth distributions with only two parameters: the projected range R_p and the range straggling

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ΔR_p . The projected range is determined by positioning the mean value of the fitted Gaussian distribution at the location in the RBS spectra where the concentration of the implanted ions is at maximum. The Gaussian function is given by

Fig. 1. (a) A typical RBS spectrum of 2.1 MeV He^{2+} ions for 100 keV Pb^+ implanted into LiNbO_3 . (b) Comparison of the experimental depth distribution with the theoretical prediction by the TRIM'98 code for 100 keV Pb^+ implanted into LiNbO_3 at a nominal dose of 5×10^{15} ions/ cm^2 . The solid circles represent the experimental data from RBS, the dashed line represents the Gaussian fit, and the histograms indicate the TRIM'98 prediction.

$$C(x) = \frac{D}{\sqrt{2\pi}\Delta R_p} \exp\left(-\frac{(x - R_p)^2}{2\Delta R_p^2}\right), \quad (1)$$

where $C(x)$ is the atom concentration at position x , and D is the implanted dose. The range straggling could be obtained from a well-known correlation

$$\Delta R_p = \frac{w_{\text{FWHM}}}{2.355N\varepsilon}, \quad (2)$$

where w_{FWHM} represents the full width at half maximum of the Gaussian distribution, N is atomic density, and σ is the stopping cross-section factor (see Ref. [11]). In the present calculation, we assume that

Bragg's rule is valid because many experimental results have shown the insignificance of the deviations from Bragg's rule for compound targets and heavy ion beams.^[12] The depth distribution of as-implanted Pb^+ at 100 – 350 keV is approximately Gaussian in shape. The w_{FWHM} value can be found to be 20 – 40 nm after performing the deconvolution process under the assumption that the estimated energy straggling of the He^{2+} ions in LiNbO_3 and the system resolution are both Gaussian.

Figure 1(a) shows the typical RBS spectrum for 100 keV Pb^+ implanted into LiNbO_3 . In the RBS spectrum, the positions of O, Nb and Pb are marked. Similar spectra were also obtained in the cases of implanted Pb^+ ions at energies from 150 to 350 keV into LiNbO_3 crystals. Figure 1(b) shows the depth distributions of 100 keV Pb^+ ions implanted into the LiNbO_3 crystal. The circles represent the experimental data. In order to compare the experimental data with the numerical simulation, the TRIM'98 code was used to simulate the process of the implanted Pb^+ into LiNbO_3 .^[13] The number of ions in this simulation is more than 10 000. The histograms and the dashed line represent the TRIM'98 code prediction and the Gaussian fit curve, respectively. As indicated in Fig. 1(b), both the experimental peak position of the depth distribution and w_{FWHM} of the Gaussian profile are somewhat different from those determined by the TRIM'98 simulation. In the case of 100 keV Pb^+ implanted into LiNbO_3 , the result shows that the experimental mean projected range of $R_p = 28.1$ nm agrees within 5% with the calculated value of $R_p = 26.7$ nm from the TRIM'98 code. However, the experimental range straggling $\Delta R_p = 8.9$ nm is about 13% larger than the calculated value of $\Delta R_p = 7.86$ nm from the TRIM'98 code.

Figure 2 shows the comparison of the measured and calculated values of the mean projected range R_p and the range straggling ΔR_p for Pb^+ implanted into LiNbO_3 . The dots represent the experimental values, and the dashed and solid lines represent the calculated values determined by the TRIM'98 and SRIM 2000 codes, respectively. It is found that the values from the TRIM'98 code are somewhat closer to the experimental data than the values obtained based on SRIM 2000. In Table 1 the experimental results and the calculated values from the TRIM'98 and SRIM 2000 codes for the Pb^+ ion implantation into LiNbO_3 at energies of 100 – 350 keV are compared.

Figure 3 represents the depth distributions of Pb^+ ions in LiNbO_3 at three doses of 5×10^{15} , 1×10^{16} , 2×10^{16} ions/ cm^2 . In order to make a clear comparison, the backscattered yield for three doses was normalized. The results show that the depth distribution of Pb^+ ions in LiNbO_3 is almost independent of dose. This means that sputtering does not play an important role for Pb^+ -implanted LiNbO_3 at 350 keV for doses ranging from 5×10^{15} to 2×10^{16} ions/ cm^2 .

Table 1. Comparison of experimental and calculated values on the mean projected range R_p and range straggling ΔR_p for Pb^+ implanted into LiNbO_3 . The calculated values are obtained based on the TRIM'98 and SRIM 2000 codes, respectively.

Energy (keV) (keV)	Experimental values		TRIM'98 prediction		SRIM 2000 prediction	
	R_p (nm)	ΔR_p (nm)	R_p (nm)	ΔR_p (nm)	R_p (nm)	ΔR_p (nm)
100	281	89	267	78.6	261	75
150	367	113	350	103	343	95
200	454	139	431	125	421	113
250	527	166	506	147	495	130
300	594	195	579	167	567	147
350	670	212	652	190	638	162

Fig. 2. Comparison of the measured and calculated values of mean projected range R_p and range straggling ΔR_p for Pb^+ implanted into LiNbO_3 . The dots represent the experimental values, and the dashed and solid lines represent the calculated values determined by the TRIM'98 and SRIM 2000 codes, respectively.

Fig. 3. Depth distributions of Pb^+ ions in LiNbO_3 at three doses of 5×10^{15} , 1×10^{16} , 2×10^{16} ions/ cm^2 . The backscattered yield for the three doses was normalized.

In order to investigate the diffusion behaviour of implanted Pb^+ in LiNbO_3 at high and low temperatures, the samples implanted at a dose of 5×10^{15}

Pb^+/cm^2 at 150 keV were annealed at 800°C in ambient air and at 77 K in liquid nitrogen, respectively. Figure 4 shows the comparison of shapes of the implanted ion distribution before and after annealing for 150 keV Pb^+ implanted into the LiNbO_3 crystal. Treatments at 800°C for 60 min and at 77 K for 120 min were carried out. In order to make a comparison, the depth of as-implanted Pb^+ at 150 keV is also given in this figure. One can see that after annealing at 800°C for 60 min, the implanted Pb^+ ions have spread towards two sides, the front side and deeper side. It is also observed that after this annealing, the maximum concentration is decreased, and some of them have moved to the surface. The shape of the implanted Pb^+ distribution becomes flatter. Similar phenomena are observed in many cases of heavy ion implantation into some crystals.^[14] While, for the treatment at 77 K for 120 min, it is found that almost no change occurs on the distribution of the implanted Pb^+ ions before and after the annealing. This result indicates that the implanted Pb^+ ions in LiNbO_3 were almost non-mobile at low temperatures down to 77 K.

Fig. 4. Comparison of the shape of the implanted ion distribution for 150 keV Pb^+ implanted into LiNbO_3 . Treatments at 800°C for 60 min and at 77 K for 120 min were carried out. The position of the Pb surface is marked by an arrow.

Pb^+ ions have been implanted into LiNbO_3 crystals in the energy range of 100 – 350 keV at doses of 5×10^{15} , 1×10^{16} and 2×10^{16} ions/ cm^2 . The depth distribution of the implanted Pb^+ ions in LiNbO_3 is

approximately Gaussian measured by RBS of 2.1 MeV He²⁺ ions. Compared to the TRIM'98 and SRIM 2000 codes, the experimental value of R_p is in good agreement with the numerical simulation while the measured value of ΔR_p is different from the calculated value to a certain extent. It is also found that the TRIM'98 code predicts experimental values of both R_p and ΔR_p which are better than those of the SRIM 2000 code. Moreover, it is found that the distribution of the implanted Pb⁺ ions in LiNbO₃ is almost independent of the doses for 350 keV Pb⁺ implantation in the range mentioned above. An obvious diffusion of the implanted Pb⁺ ions at 150 keV with a dose of 5×10^{15} ions/cm² occurs after 800°C annealing for 60 min in ambient air. After a low-temperature treatment at 77 K for 120 min, no obvious diffusion phenomenon occurs for the implanted Pb⁺ ions in LiNbO₃.

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