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A STUDY ON THE RAMAN SPECTRUM OF THE LiNbO_3 -DOPED Mg CRYSTAL AT LOW TEMPERATURE

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ABSTRACT

The Raman Spectra of $\text{LiNbO}_3\text{:MgO}$ (6.7 mol%) at both low temperature and room temperature were studied. The results showed that the crystal structure has changed little after doping Mg^{++} . At room temperature the lattice distorted a little, which caused the appearance of coupling phenomenon of some individual scattering peaks. As the temperature decreased, the coupling reduced gradually.

Key Words: $\text{LiNbO}_3\text{:MgO}$ Crystal; Raman spectrum; Low temperature; Laser

INTRODUCTION

The LiNbO_3 crystal is now widely used in laser techniques for its superior electro-optic and nonlinear characteristics. Nevertheless, the applications are somewhat limited due to its poor ability of resisting laser damage. Recently, some researchers attempt to enhance this ability by doping Mg^{++} in LiNbO_3 crystals. In 1980, Zhong Giguang et al. found that

doping 4.6 mol% of MgO in LiNbO₃ made an improvement of laser damage resistance by two orders of magnitude. This was a significant advancement in the study of LiNbO₃ applications. Hence, a further study of the characteristics of the LiNbO₃:MgO crystal is necessary. In this paper, we studied the Raman spectra of the LiNbO₃:MgO crystal (doping with 6.7 mol% Mg⁺⁺) at both low temperature and room temperature. We also compared these spectra with that of the pure LiNbO₃ crystal, and no clear distinction had been found. No obvious frequency shift of the scattering peak appeared. There was coupling between the two low frequency modes at room temperature. This phenomenon disappeared at low temperature, and meanwhile, the peak width became narrower. This meant that the symmetry of the crystal was not changed when the Li⁺ was substituted equivalently by Mg⁺⁺ and the strong Raman spectra of the A₁-mode and E-mode were determined mainly by the characteristic vibration of the oxygen octahedron (NbO₃).

Crystal Structure and Vibration Modes

At room temperature, LiNbO₃ is an ferroelectric crystal with space group R_{3c}(C_{3v}⁶), and point group (C_{3v}) in which each crystal cell contains two molecules. In other words, it contains ten total atoms. From the analysis of the lattice vibration on the basis of the symmetry group, it is well known that the vibration modes are as follow :

$$4A_1 + 9E + 5A_2 \quad (1)$$

where A₁ and E are Raman and infrared active modes respectively, while A₂ are not Raman active modes.

The Characteristics of the Raman Spectrum at Room Temperature

The experimental apparatus used for this work is a Raman spectrometer, Model SPEX 1403, configured with a microcomputer system. Specimens of size of 10 × 10 × 2 mm³ were prepared from a LiNbO₃:MgO (6.7 mol%) crystal provided by the crystal laboratory of the Harbin Institute of Technology.

Figure 1 shows the Raman spectra of the A₁ mode (a) and E mode (b) at room temperature, respectively. From Figure 1(a) we can see two scattering peaks of low frequency A₁ modes overlap each other, so that they are indistinguishable. From the profile of the peak, we can assume that it has resulted from two peaks in superimposition. The non-harmonic interaction

between the modes was enhanced by lattice distortion due to the large amount of Mg⁺⁺ doping in the crystal lattice of LiNbO₃, therefore the coupling of the two low frequency A₁ modes was enhanced, which resulted in superimposition of the scattering peaks. The other two scattering peaks of the A₁ mode remained unchanged.

From Figures 1(a) and 1(b) we can see eight scattering peaks of the E mode. As compared with the result of the group theory analysis, the peak of the $\Delta\gamma \sim 630\text{ cm}^{-1}$ did not exist¹. Table 1 gives the Raman frequency shifts of the modes.

Because the 277 cm^{-1} vibration of A₁ (TO) model cannot be discriminated when doping 6.7 mol% Mg⁺⁺, it is difficult to determine whether it is due to a frequency close to 258 cm^{-1} or due to other affecting factors rather than the change of frequency. This problem cannot be solved by

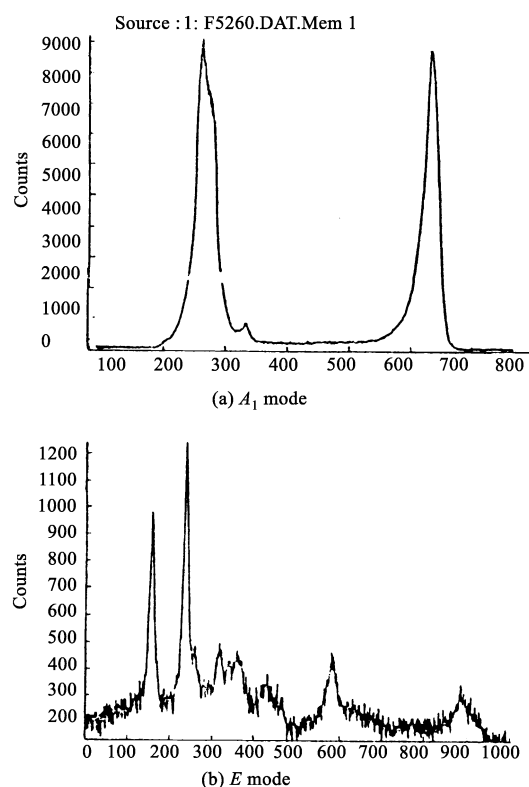


Figure 1. The Raman spectra of the A₁ mode and E mode at room temperature.

Table 1. Raman Frequencies of Vibration Modes of the $\text{LiNbO}_3\text{:MgO}$ (6.7 mol%) at Room Temperature (in cm^{-1})

Mode		Band Location (in cm^{-1})						
A_1			251		334		614	
E	155	236	265	325	371	431	582	890

measuring a Raman spectrum at room temperature, so we should study the Raman spectrum at low temperature.

The Raman Spectrum of the $\text{LiNbO}_3\text{:MgO}$ (6.7 mol%) Crystal at Low Temperature

We first obtained the Raman spectra of the $\text{LiNbO}_3\text{:MgO}$ crystal at low temperature. Figures 2 and 3 show the measurement results of $A_1(\text{TO})$ mode under coordinate $X(\text{ZZ})Y$ and $E(\text{TO})$ mode under $X(\text{ZY})Z$ respectively during the process of the temperature drop. The degree of superimposition of the scattering peaks of two low frequency $A_1(\text{TO})$ reduces with the decrease of temperature, until 260 K; the A_2 peak reappears sharply, as shown in Figure 2. Further decrease of temperature separates the two peaks far apart from each other, this means that the nonharmonic interaction between the two modes are weakened. Actually there is no frequency shift. In Figure 3, it is obvious that the intensity of the scattering peak of the E mode is increased during the process of the temperature drop.

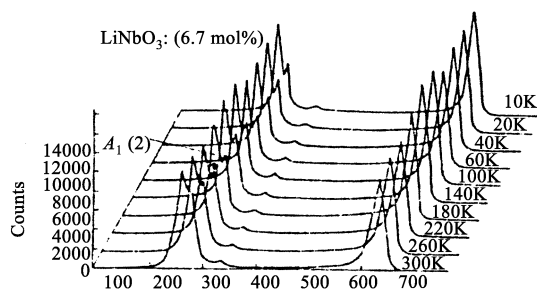


Figure 2. The measurement results of $A_1(\text{TO})$ mode under co-ordinate $X(\text{ZZ})Y$ during the process of temperature drop.

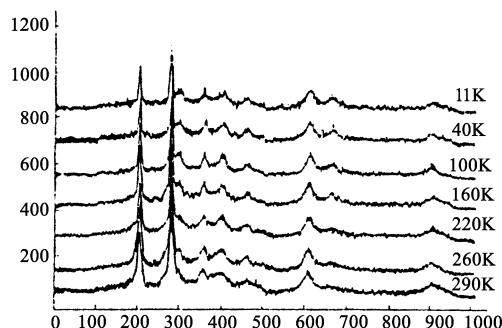


Figure 3. The measurement results of E(TO) mode under co-ordinate X(ZY)Z during the process of temperature drop.

Table 2. Raman Frequencies of Vibration Modes of the LiNbO₃:MgO (6.7 mol%) at 11 K (cm⁻¹)

Mode		Band Location (in cm ⁻¹)								
A ₁				277	285	339			620	
E	157	240	269		328	374	435	589	638	897

SUMMARY

We have revealed that there is no distinct effect of doping on the vibrational modes in Raman spectra at both low temperature and room temperature. Table 2 gives the Raman frequency shifts of the modes at 11 K. Comparing Tables 1 and 2, it is known that at room temperature and low temperature, the peak of Raman spectra of the LiNbO₃:MgO crystal was not shifted and distorted, it shows that the crystal structure of LiNbO₃:MgO was not changed after substituting Li⁺ by Mg⁺⁺. The stronger Raman spectra of the A₁(TO) and E(TO) vibrational modes were contributed to mainly by the characteristic radicals of the oxygen octahedron. The coupling of individual peaks of vibration was observed due to the crystal lattice distortion at room temperature. This coupling was also weakened at low temperature.

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