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STRUCTURAL EFFECTS OF Li AND Nb CODOPING IN A $KTaO_3$ MATRIX: RAMAN INVESTIGATIONS

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New attention has recently been devoted to $K_{1-x}Li_xTa_{1-y}Nb_yO_3$ compounds (KLTN), owing also to perspective applications (e.g., photorefractivity). Attempts are in progress, in particular, to identify the structural effects arising from the interplay of Li and Nb in the $KTaO_3$ matrix. Actually, there is evidence that Li^+, with its strong dipolar displacement and slow relaxation, is able to condition the symmetry of the low temperature phase and to increase transition temperatures, when added to $KTa_{1-y}Nb_yO_3$ (KTN) (1). Conversely, small additions of Nb have been shown to modify the usual dipole-glass picture of KLT, introducing new elements of critical behaviour (2).
To gain new insight on these phenomena, we have undertaken Raman investigations on a series of KLTN compounds of varying stoichiometry. The present study concerns two samples with relatively high proportions of Nb, namely: x = 0.0028 and y = 0.013 (sample 1) and x = 0.0014 and y = 0.024 (sample 2). These samples contained minor additions of Cu, which improves crystallization. Dielectric data on the same samples are also presented in a separate paper (3), in which it is suggested that a reentrant dipole glass phase is realized in sample 2 at low temperatures. Systematic observations of TO1 and TO4 spectra were performed in the range from 12 K to 100 K, following preparation in different poling conditions. As a general feature, a sudden increase in the intensity of these spectra was observed below a critical temperature Tc, higher than that characterizing the onset of the soft modes in KTN of comparable Nb concentrations. In sample 2 (larger Nb/Li ratio) a new remarkable feature was the appearance of two distinct stages. In the first, below the softening point in the range 40 - 45 K a single component of the TO1 spectrum is seen to develop with increasing frequencies. Successively, a second low energy component begins to emerge from the central background at about 30 K, hardening therefrom in parallel with the high-energy component. At temperatures around 15 K the two frequencies stabilize, with a final splitting which depends on the state of poling (25 cm^{-1} and 12 cm^{-1}, respectively, in the poled or the unpoled sample). These results are remarkable, inasmuch as they suggest the presence of two different transitions, of which there is no mention in previous literature on KTN of this concentration. It is straightforward to think of this as a proof of the dramatic effects that a small Li addition can have in these compounds. Hints of the same phenomena were found, with less clearcut manifestations, also in sample 1. Here, however, the features described above were observed only in the poled sample, whereas the unpoled sample showed no evidence of critical behaviour.

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[2] P. Camagni, P. Galinetto, E. Giulotto, G. Samoggia, V. A. Trepakov, P. P. Syrnikov, Ferroelectrics (in press)
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