ACCURATE X-RAY PHASE MEASUREMENTS WITH GENETIC ALGORITHM: APPLICATIONS IN DOPED NLO CRYSTALS

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Since it was reported decades ago that information on structure factor phases – or more precisely, on invariant triplet phases - could be accessed via X-ray multiple diffraction (XRMD) experiments, many efforts have been carried out to use this phenomenon as a physical solution of the phase problem in X-ray crystallography or, at least, as a general tool for studying crystalline and non-periodic materials. In this latter context, XRMD can be seen as a specific method to provide singular pieces of information that could not be retrieved by any other method and thus make it possible to distinguish one atomic structure from another among several possibilities. For instance, this was demonstrated in elucidating the mechanism of resonant scattering in $LaMnO_3$ [1]. However, the potential field of applications of XRMD in phase measurements is not widely exploited. It has potential for new opportunities in the dynamical theory of X-ray diffraction. Phase sensitivity in XRMD experiments is essentially a dynamical diffraction process well described by the N-beam dynamical theory in perfect crystal slabs. There is, however, a difficult point to be settled, which is how to account for crystalline imperfections on the dynamical interference process of simultaneously diffracted waves.

In this work, we experimentally exploited the potential of XRMD phase measurements for investigating structural changes in doped NLO crystals. In particular, we studied ADP ($NH_4H_2PO_4$) and KDP (KH_2PO_4) doped with Ni and Mn, respectively. In the former, the evidence of structural changes caused by doping are remarkable, and beyond any doubt that could be raised due to the presence of lattice imperfections. An example of similar phase shifts as that cause by doping of ADP were obtained in GaAs for the sake of comparison. In the case of Mn-doped KDP, synchrotron X-ray energy and polarization were adjusted to enhance the phase sensitivity for quantitative data analysis by using a genetic algorithm [2,3]. Samples with different contents of Mn in the crystal lattice were investigated. The influence of lattice imperfections on measured phase values was minimized by using a criterion to identify XRMD cases suitable for accurate phasing. Structural changes in the doped samples accounting for the observed phase shifts are discussed. This work has been supported by FAPESP, CNPq, and LNLS.

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