Superspace description of two-dimensional distributions of defects. Application to the analysis of vacancy ordering. , Luis Elcoro[a]* and J. Manuel Perez-Mato[a],[a]*Dpto. de Física de la Materia Condensada. Facultad de Ciencia y Tecnología. Universidad del País Vasco/Euskal Herriko Unibertsitatea. Apdo. 644, Bilbao 48080, Spain*. E-mail: wmpelcel@lg.ehu.es

The superspace framework has probed to be a useful tool for the quantitative description of compositionally flexible layered systems. Starting from an ideal structure (for a specific composition) with a small number of layers in the unit cell, the layer stacking sequence for other compositions is obtained by the ordered removal of complete layers (or substitution of some layers with different ones) from the ideal structure, according to the composition. The vacant or substituting layers can be interpreted as *defects* in the ideal stacking sequence. It is an experimental fact that these defects are distributed as uniformly as possible in the resulting stacking sequence and the distribution is unique for a given composition. In the superspace analysis of these systems, a single model is sufficient to describe the structure for all the compositions. The atomic layers are represented by occupational step-like (crenel) functions (named atomic domains, AD) along the 1-dimensional internal space and the number of ADs in the superspace unit cell is the same for all the compositions. The parameters which depend on composition are just the modulation wavevector and the length of the crenel function which represents each layer. The experimental uniform distribution of defects is realised when the projections onto the internal space of the ADs associated to consecutive layers are connected (the so-called *closeness condition*). This condition fixes the relation between the modulation wavevector and the composition.

In this work, we analyze the possibility of having uniform distributions of point defects in two dimensions, giving rise to alternative compositionally flexible systems. In those families, the number of layers is the same for all the compositions but the periodicity inside the layers is composition-dependent. Starting from an ideal compound with small unit cell vectors parallel to the layers, the structure for each composition is obtained by an ordered removal (or substitution) of atoms, according to the composition, inside the layer. The superspace analysis of these systems requires a 2-dimensional internal space, and the atoms are represented by 2-dimensional ADs. The assumption of the closeness condition for these 2-dimensional ADs gives also rise in this case to *uniform* defect distributions in two dimensions, which can be interpreted as long-period or aperiodic configurations resulting from the tendency to distribute the defects as uniformly as possible, within the restrictions imposed by the discrete character of the basic 2-dimensional array on which the defects are introduced. For these uniform distributions a relation between the modulation wavevector and the composition is predicted. A successful comparison of the proposed (3+2)-dimensional superspace construction with the structures reported for three real compounds having some vacancy ordering (with tetragonal, hexagonal and pseudo-hexagonal symmetries) proves the general soundness and efficiency of this approach when dealing with this type of systems.