Crystal Frameworks, Matrix-valued Functions and Rigidity Operators

S.C. Power

Abstract. An introduction and survey is given of some recent work on the infinitesimal dynamics of crystal frameworks, that is, of translationally periodic discrete bond-node structures in $\mathbb{R}^d$, for $d = 2, 3, \ldots$. We discuss the rigidity matrix, a fundamental object from finite bar-joint framework theory, rigidity operators, matrix-function representations and low energy phonons. These phonons in material crystals, such as quartz and zeolites, are known as rigid unit modes, or RUMs, and are associated with the relative motions of rigid units, such as SiO$_4$ tetrahedra in the tetrahedral polyhedral bond-node model for quartz. We also introduce semi-infinite crystal frameworks, bi-crystal frameworks and associated multi-variable Toeplitz operators.

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1. Introduction

A survey is given of some recent work on the infinitesimal dynamics of crystal frameworks, by which we mean translationally periodic discrete bar-joint frameworks in $\mathbb{R}^d$. This includes a discussion of rigidity operators, matrix symbol function representations and the connections with models for low energy phonon modes in various material crystals. These modes are also known as rigid unit modes, or RUMs, reflecting their origin in the relative motion of rigid units in the crystalline structure. I also introduce briefly the contexts of semi-infinite crystal frameworks and bicrystal frameworks and indicate how their rigidity operators involve multivariable Toeplitz operators whose symbol functions are matrices over multivariable trigonometric polynomials on the $d$-torus.

The topic of infinite bar-joint frameworks, whether periodic or not, can be pursued as a purely mathematical endeavour and many aspects of deformability
and rigidity remain to be understood. The main perspectives below and related issues are developed in Owen and Power [21], [22], [24] and Power [25], [26].

Translationally periodic bond/node bar-joint frameworks or networks are ubiquitous in mathematics (periodic tilings for example), solid state physics (crystal lattices, graphene), solid state chemistry (zeolites) and material science (microporous metal organic frameworks). So there is no lack of interesting examples. I shall illustrate a number of concepts with three examples derived from tilings seen in Seville at the Alcazaar and the Cathedral.

2. Models for material crystals and low energy phonons

We begin by outlining one particular motivation from material science. A crystal framework $\mathcal{C}$ in $\mathbb{R}^3$ can serve as a mathematical model for the essential geometry of the disposition of atoms and bonds in a material crystal $\mathcal{M}$. In the model of interest to us the vertices correspond to certain atoms while the edges correspond in some way to strong bonds. Also the identification of strongly bonded “units” in $\mathcal{M}$ imply a polyhedral net structure and it is this that gives the relevant abstract framework $\mathcal{C}$. A fundamental example of this kind is quartz, SiO$_2$, in which each silicon atom lies at the centre of a strongly bonded SiO$_4$ unit, which in turn may be modeled as a tetrahedron with an oxygen atom at each vertex. In this way the material crystal quartz provides a mathematical crystal framework of pairwise connected tetrahedra with a particular connectedness and geometry.

Material scientists are interested in the manifestation and explanation of various forms of low energy motion and oscillation in materials. Of particular interest are the rigid unit modes in aluminosilicate crystals and zeolites, where quite complicated tetrahedral net models are relevant. These low energy (long wavelength) phonon modes are observed in neutron scattering experiments and have been shown to correlate closely with the modes observed in computational simulations. There is now a considerable body of literature tabulating the (reduced) wave vectors of RUMs of various crystals as subsets of the unit cube (Brillouin zone) and it has become evident that the primary determinant is the geometric structure of the abstract frameworks $\mathcal{C}$. See, for example, Dove et al. [8], Hammond et al. [12], [13], Giddy et al. [10] and Swainson and Dove [31]. Particularly intriguing is the simulation study in Dove et al. [8] which gives a range of pictures of the RUM spectrum and multiplicities for various idealized crystal types.

In the experiments and in the simulations the background mathematical model is classical lattice dynamics and rigid unit modes are observed where the phonon dispersion curves indicate vanishing energy. However, one can also identify such limiting cases through a direct linear approach as we outline below and from this it follows that these sets (at least in simulations) may be viewed as real algebraic varieties. See Theorem 5.4 below, [24], [25] and Wegner [33]. It is convenient to define the RUM dimension to be the dimension of this algebraic variety. (See Section 5.) In 3D it takes the values 0, 1, 2, 3.