

Quasicontinuum theory of crystals

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THE paper is concerned with discussing the fundamental concepts of the quasicontinuum theory and its applications to crystalline media. Various relevant methods of interpolation are presented and compared and a definition of the class of QC-functions is given. Some most important properties of the QC-functions are exhibited, and their action in the procedure of passing from a discrete to a continuum model is shown. The notion of equivalence of operators in QC-theory is introduced and a number of examples of mutual equivalences between differential, integral and discrete operators is given. The relation between quasicontinuum and nonlocal continuum as well as between quasicontinuum and discrete lattice is examined. The concept of differential models of discrete lattices is introduced and discussed.

Praca niniejsza dotyczy dyskusji podstawowych pojęć teorii quasi-kontynuualnej i jej zastosowań do ośrodków krystalicznych. Przedyskutowano różne odpowiednie metody interpolacji i podano definicję klasy QC-funkcji. Wskazano na niektóre z najważniejszych własności QC-funkcji i pokazano ich działanie w procedurze przechodzenia z modelu ośrodka dyskretnego do modelu kontynuualnego. Wprowadzono pojęcie równoważności operatorów w QC-teorii i podano szereg przykładów wzajemnej równoważności między operatorami różniczkowymi, całkowymi i dyskretnymi. Zbadano zależność między quasi-kontynuuum i nielokalnym kontynuuum, jak również między quasi-kontynuuum i siatką dyskretną. Wprowadzono i przedyskutowano koncepcję modeli siatek dyskretnych.

Настоящая работа касается обсуждения основных понятий квазиконтинуальной теории и ее применений к кристаллической среде. Обсуждены разные соответствующие методы интерполяции и дается определение класса QC-функций. Указаны некоторые, самые важные, свойства QC-функций и показано их действие в процедуре перехода от модели дискретной среды к континуальной модели. Введено понятие эквивалентности операторов в QC-теории и приведен ряд примеров взаимной эквивалентности между дифференциальными, интегральными и дискретными операторами. Исследована зависимость между квазиконтинуумом и нелокальным континуумом, как тоже между квазиконтинуумом и дискретной сеткой. Введена и обсуждена концепция дискретных сеток.

1. Introduction

FROM the atomistic point of view the set of functions admitted in continuum theory is definitely too large. For example, the displacement field $u(x)$ can be interpreted only at those points at which the atoms are located, i.e. on a discrete set of points

$$x = X_n.$$

Generally, there is a lot of functions of the continuous argument x which take some prescribed values $u(X_n) = u_n$ at a discrete set $x = X_n$. In particular, there are many functions $u(x)$ which are not identically zero but vanish at any $x = X_n$. Such functions cannot be reasonably interpreted in atomistic terms. Moreover, the physical meaning of rapidly oscillating functions of position is always doubtful.

This is a fundamental difficulty of any continuum theory. It suggests the idea of restricting the set of admissible functions from the very beginning and to carry out all the mathematical considerations for this restricted set of functions. This set should contain only the functions which are necessary to describe all possible displacements of the atoms. The corresponding model of material medium differs considerably from the continuous one: contrary to the latter case the number of degrees of freedom per unit volume is finite. The approach based on this idea was developed by KRUMHANSL (1963), ROGULA (1965) and KUNIN (1966), and will be called quasicontinuum theory.

The problem of constructing the required set of functions is essentially an interpolation problem. The set of interpolating functions will be denoted by QC . Without further assumptions this set can not be determined uniquely. We shall discuss three methods of constructing the set QC . Under some additional conditions, they turn out to be equivalent to each other.

For the sake of lucidity, we shall first discuss these methods for scalar fields on a linear chain of atoms. Afterwards we apply this procedure to vector fields on a three-dimensional primitive lattice. The state of a non-primitive structure can be described by multiplets of fields defined on the corresponding Bravais lattice, e.g. one field for each atom in the elementary cell.

2. The sampling function method

Let a denote the distance between neighbouring atoms of a linear chain and let f be a certain quantity taking some values f_n on atoms located at $x = na$ (n - an arbitrary integer).

Let $S(x)$ be a function of the continuous variable x such that

$$(2.1) \quad S(na) = \begin{cases} 1 & \text{for } n = 0, \\ 0 & \text{for } n \neq 0. \end{cases}$$

Any function satisfying (2.1) will be called a sampling function.

Provided that a sampling function has been chosen, one can uniquely associate a function $f(x)$ of the continuous variable x to a function f_n of the discrete variable n :

$$(2.2) \quad f(x) = \sum_n S(x-na)f_n.$$

This formula makes sense whenever the series involved is convergent. Because of (2.1) the function $f(x)$ has the interpolation property

$$(2.3) \quad f(na) = f_n.$$

The function

$$(2.4) \quad S(x) = \frac{a}{\pi x} \sin \frac{\pi x}{a}$$

provides an example of a smooth (analytic) sampling function.

3. The Fourier transformation method

Under some restrictions of its behaviour at infinity, the discrete function f_n can be represented as a Fourier integral

$$(3.1) \quad f_n = \frac{1}{2\pi} \int_{-\pi/a}^{\pi/a} dk e^{ikan} \hat{f}(k)$$

with an appropriate function (or, more generally, distribution) $\hat{f}(k)$. The function $\hat{f}(k)$ is uniquely determined by the function f_n :

$$(3.2) \quad \hat{f}(k) = \frac{1}{a} \sum_{n=-\infty}^{\infty} e^{-ikan} f_n.$$

On the other hand, by writing x instead of an in (3.1), one can define a function $f(x)$ of the continuous variable x :

$$(3.3) \quad f(x) = \frac{1}{2\pi} \int_{-\pi/a}^{\pi/a} dk e^{ikx} \hat{f}(k).$$

The function $f(x)$ has the interpolation property (2.3).

The Fourier transformation method is equivalent to the sampling function method provided that the sampling function (2.4) is chosen. It follows from the fact that to the function $S(x)$ given by the Eq. (2.4) there corresponds the function $\hat{S}(k) = 1$, i.e.,

$$(3.4) \quad S(x) = \frac{1}{2\pi} \int_{-\pi/a}^{\pi/a} dk e^{ikx}.$$

4. The direct interpolation method

According to the generalized Paley-Wiener theorem [4], any function of the form (3.3), with an arbitrary distribution $\hat{f}(k)$, can be continued to the complex x -plane as an entire analytic function. Moreover, it satisfies the inequality

$$(4.1) \quad |f(x)| \leq C(1 + |x|)^N e^{\pi/a \operatorname{Im} x}$$

for some constants C and N . This inequality is also sufficient for an entire analytic function to have a representation of the form (3.3).

In terms of the theory of analytic functions, $f(x)$ is an entire analytic function of order not greater than 1 and type π/a . This class of functions can be identified with QC . Then the problem of fitting a function $f \in QC$ to given values f_n can be stated directly as an interpolation problem. If $f_n \equiv 0$, then

$$(4.2) \quad \int_{-\pi/a}^{\pi/a} dk e^{ikan} \hat{f}(k) = 0$$

and $f(k)$ must be of the form

$$(4.3) \quad \hat{f}(k) = P\left(\frac{d}{dk}\right) [\delta(k - \pi/a) - \delta(k + \pi/a)],$$

where $P(\cdot)$ is a polynomial. Therefore any function $f \in QC$ such that $f_n \equiv 0$ must be of the form

$$(4.4) \quad f(x) = W(x) \sin \frac{\pi x}{a},$$

where $W(x)$ is a polynomial. Thus for the functions $f(x)$ and f_n which tend to zero at infinity the solution of the interpolation problem is unique.

This result is directly related to the distribution of roots of entire analytic functions in the complex plane. Let $f(x)$ be an entire analytic function of order ρ , i.e.,

$$(4.5) \quad \rho = \inf_{\alpha > 0} \alpha: |f(x)| \leq \exp |x|^\alpha$$

for sufficiently large $|x|$ in complex x -plane (see e.g. [5]). Let $\{x_n\}$ be the sequence of non-zero roots of $f(x)$ in complex x -plane, ordered so that $|x_{n+1}| \geq |x_n|$ (multiple roots taken according to their multiplicity). Then the exponent of convergence of these roots, defined as

$$(4.5) \quad \mu = \inf_{\alpha > 0} \alpha: \sum_n |x_n|^{-\alpha} < \infty,$$

satisfies the inequality

$$(4.6) \quad \mu \leq \rho.$$

Hence the condition $\rho \leq 1$ eliminates the possibility for μ to exceed the exponent of convergence of $x_n = na$, which equals 1.

5. The three-dimensional generalization

The above considerations can be generalized directly to multi-dimensional lattices. The atoms of a primitive lattice are labelled by integer multiplerts \mathbf{n} and their positions are given by

$$(5.1) \quad \mathbf{X}(\mathbf{n}) = A\mathbf{n},$$

where A is a certain matrix. The sampling function $S(\mathbf{x})$ should have the property

$$(5.2) \quad S(A\mathbf{n}) = \begin{cases} 1 & \text{for } \mathbf{n} = 0, \\ 0 & \text{for } \mathbf{n} \neq 0. \end{cases}$$

Instead of integrating over the interval $[\pi/a, -\pi/a]$ in (3.1) and (3.3), one should integrate over the first Brillouin zone, e.g.

$$(5.3) \quad f_n = \frac{1}{(2\pi)^3} \int_{BZ} d^3k e^{i\mathbf{k}\mathbf{x}} \hat{f}(\mathbf{k})$$

and

$$(5.4) \quad f(\mathbf{x}) = \frac{1}{(2\pi)^3} \int_{BZ} d^3k e^{i\mathbf{k}\mathbf{x}} \hat{f}(\mathbf{k}).$$

Although it is possible to choose some other equivalent domains of integration, the choice of the *BZ* has the advantage of retaining the point symmetry of the corresponding Bravais lattice.

The sampling function corresponding to the Fourier transformation method is given as

$$(5.5) \quad S(\mathbf{x}) = \frac{1}{(2\pi)^3} \int_{BZ} d^3k e^{i\mathbf{k}\mathbf{x}}.$$

The formulae (5.3), (5.4) and (2.2) can be directly adapted for multi-component quantities, e.g. for displacement fields, force fields etc.

6. Some properties of *QC*-functions

The functions *QC* have been defined as the entire analytic functions satisfying inequality (4.1). In particular, all polynomials are *QC*.

The following simple properties can be checked directly:

- if $u \in QC$, then all the derivatives $\partial^\mu u \in QC$,
- if $u \in QC$ and the convolution u^*v exists then $u^*v \in QC$ (but it can happen that $u \cdot v \notin QC$).

The following equations hold:

$$(6.1) \quad \Omega \sum_{\mathbf{n}} f_{\mathbf{n}} = \int d^3x f(\mathbf{x}),$$

$$(6.2) \quad \Omega \sum_{\mathbf{n}} f_{\mathbf{n}} g_{\mathbf{n}} = \int d^3x f(\mathbf{x}) g(\mathbf{x}),$$

$$(6.3) \quad \Omega^2 \sum_{\mathbf{n}, \mathbf{n}'} u_{\mathbf{n}} \Phi_{\mathbf{n}\mathbf{n}'} w_{\mathbf{n}'} = \int d^3x d^3x' u(\mathbf{x}) \Phi(\mathbf{x}, \mathbf{x}') w(\mathbf{x}'),$$

where Ω denotes the volume of the primitive cell.

In particular, it follows from (6.2) that the kinetic energy equals

$$(6.4) \quad T = \frac{m}{2} \sum_{\mathbf{n}} v_{\mathbf{n}}^2 = \frac{\rho}{2} \int d^3x v^2(\mathbf{x}),$$

where $\rho = m/\Omega$. On the other hand, the potential energy of a crystal can be expressed as

$$(6.5) \quad U = \frac{1}{2} \sum_{\mathbf{n}, \mathbf{n}'} u_{i\mathbf{n}} \Phi_{ij}^{\mathbf{n}\mathbf{n}'} u_{j\mathbf{n}'} = \frac{1}{2\Omega^2} \int d^3x d^3x' u_i(\mathbf{x}) \Phi_{ij}(\mathbf{x}, \mathbf{x}') u_j(\mathbf{x}').$$

The last expression is formally similar to the corresponding expression of non-local (integral) theory. The corresponding expression in (6.4) is also similar to the continuum one. The expressions (6.4) and (6.5) for the kinetic and the potential energies allow us to develop the theory in continuum-like form. The equations obtained, however, describe the crystal exactly.

In the same way as in continuum theory, these equations can be generalized to include dislocation fields. We shall omit here the details of this procedure.

7. Equivalence of operators in QC

Now we briefly consider linear operators L acting in QC . The classification of operators of continuum theory (ROGULA, 1973) does not apply here. According to [6], the singular order of an operator L is determined by the behaviour of $A_{ij}(\mathbf{k})$ at infinity. In QC , however, $\mathbf{k} \in BZ$, so we can not speak of behaviour at \mathbf{k} -infinity.

Consider two linear operators L and L' . If for any $\mathbf{u} \in QC$

$$(7.1) \quad Lu = L'u,$$

these operators will be called equivalent. In fact, from the point of view of QC they are identical, and in any calculation one can put L' instead of L . This circumstance gives the QC -theory great flexibility.

As an example, consider a differential operator

$$(7.2) \quad Lu = P(\partial)u,$$

where $P(\partial)$ is a certain polynomial. Let

$$(7.3) \quad \Psi(\mathbf{x}) = \frac{1}{(2\pi)^3} \int_{BZ} d^3k P(i\mathbf{k}) e^{i\mathbf{k}\mathbf{x}};$$

then we define another operator

$$(7.4) \quad L'u = \Psi^*u,$$

which is an integral operator. Moreover, let

$$(7.5) \quad \tilde{\Psi}(\mathbf{x}, \mathbf{n}) = \Omega \Psi(\mathbf{x} - A\mathbf{n})$$

and

$$(7.6) \quad L''u = \sum_{\mathbf{n}} \tilde{\Psi}(\mathbf{x}, \mathbf{n}) u(A\mathbf{n}).$$

All the operators L, L', L'' are equivalent to each other. This shows that in QC there are equivalencies between differential, integral and discrete operators.

8. Quasicontinuum vs. discrete lattice. Differential models

The quasicontinuum approach, when applied to crystal lattices with given force constants, produces the same physical results as conventional discrete lattice (DL) theory. This, in particular, follows from the formulae (6.4) and (6.5), if we take into account the one-to-one correspondence between force constants and QC -kernels,

$$(8.1) \quad \Phi_{ij}^{\mathbf{n}, \mathbf{n}'} \Leftrightarrow \Phi_{ij}(\mathbf{x}, \mathbf{x}'),$$

which holds precisely under physically plausible conditions of vanishing at infinity. In this sense, there is an isomorphism between QC and DL . The equations of motion of a discrete lattice

$$(8.2) \quad m\ddot{u}_i^{\mathbf{n}} - \sum_{\mathbf{n}'} \Phi_{ij}^{\mathbf{n}\mathbf{n}'} u_j^{\mathbf{n}'} = f_i^{\mathbf{n}}$$

and the corresponding quasicontinuum,

$$(8.3) \quad \rho \ddot{u}_i(\mathbf{x}) - \int d_3 \mathbf{x}' \Phi_{ij}(\mathbf{x}, \mathbf{x}') u_j(\mathbf{x}') = f_i(\mathbf{x})$$

contain the same physical information.

The mathematics of *QC*, however, is much more flexible than that of *DL*. In the framework of *QC* one can use, apart from discrete operators, also other equivalent forms of operators without losing the precise meaning of them. Therefore, the calculational efficiency of *QC* can be much higher than that of *DL*.

Another, and perhaps even more important circumstance consists in the following. The lattice theory does not provide us with any specific set of force constants. These either have to be determined from other physical considerations, or treated as (infinitely many) phenomenological parameters. In any case, establishing a tractable set of force constants involves some approximations.

One of the most popular simplifications in *DL* is a finite (and, as a matter of fact, very short) range of interactions. It is equivalent to approximating the exact dispersion curves by a combination of trigonometric functions.

On the other hand, it is possible to achieve substantial simplification in a different manner, namely by choosing the force constants of the lattice so that the integral operator in (8.3) is equivalent to a differential one. Then, instead of (8.3) we have a differential equation of the form

$$(8.4) \quad \rho \ddot{u}_i(\mathbf{x}) - P_{ij}(\partial) u_j(\mathbf{x}) = f_i(\mathbf{x}).$$

This *QC*-equation describes exactly the dynamics of a certain crystal lattice. The force constants of this lattice do not vanish identically for great distances: instead of that, they tend to zero with some negative power of the distance. In the *k*-space, this corresponds to approximating the dispersion curves by a polynomial instead of trigonometric functions, the polynomial being the simpler but not necessarily the worse approximation.

References

1. J. A. KRUMHANSL, *Generalized continuum field representation for lattice vibrations*, p. 627 in "Lattice dynamics", ed. R. F. WALLIS, Pergamon Press, 1965.
2. D. ROGULA, *Influence of spatial acoustic dispersion on dynamical properties of dislocations*, Bull. Acad. Polon. Sci., Série Sci. Techn., **13**, 337, 1965.
3. I. A. KUNIN, *Prikl. Mat. Mech.* [in Russian] **30**, 542, 1966.
4. L. HÖRMANDER, *Linear Partial Differential Operators*, Springer, 1963.
5. S. SAKS, A. ZYGMUND, *Analytic functions*, Wrocław-Warszawa 1952.
6. D. ROGULA, *On nonlocal continuum theories of elasticity*, Archives of Mechanics, **25**, 233, 1973.

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