



A highly-efficient technique for evaluating bond-orientational order parameters



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ABSTRACT

We propose a novel, highly-efficient approach for the evaluation of bond-orientational order parameters (BOPs). Our approach exploits the properties of spherical harmonics and Wigner 3j-symbols to reduce the number of terms in the expressions for BOPs, and employs simultaneous interpolation of normalised associated Legendre polynomials and trigonometric functions to dramatically reduce the total number of arithmetic operations. Using realistic test cases, we show how the above, combined with a CPU-cache-friendly data structure, leads to a 10 to 50-fold performance increase over approaches currently in use, depending on the size of the interpolation grids and the machine used. As the proposed approach is an approximation, we demonstrate that the errors it introduces are well-behaved, controllable and essentially negligible for practical grid sizes. We benchmark our approach against other structure identification methods (centro-symmetry parameter (CSP), common neighbour analysis (CNA), common neighbourhood parameter (CNP) and Voronoi analysis), generally regarded as much faster than BOPs, and demonstrate that our formulation is able to outperform them for all studied systems.

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1. Introduction

Bond-orientational order parameters (BOPs) were proposed by Steinhardt et al. in 1981 [1,2] as a generalisation of the two-dimensional hexatic order parameter [3]. Initially BOPs were applied to the study of the orientational order in liquids and glasses, later to become a standard tool in all of solid state physics. Nowadays BOPs are chiefly used to differentiate between crystalline phases, such as sc, bcc, fcc or hcp [4–11].

BOPs have been used to study nucleation and crystal growth [6,10–17], helping to elucidate the structure of critical nuclei [5] and nucleation kinetics [18]. They also constitute a standard tool for the study of melting processes [19–21], where global BOPs are used as a direct indicator of a phase transition, while local BOPs serve as measures for the determination of solid and liquid fractions. Studies of undercooling and glassification [10,17,22,23] also employ BOPs, as do investigations of local icosahedral order in liquid metals [21,24] and in other systems [1,2,25,26].

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Many model systems have been studied with the aid of BOPs: hard- [6,16,27–29] and soft-spheres [4], Lennard-Jones systems [5,9,13,14,18,20,30,31] (including binary [32] and polydisperse [22]), quantum Lennard-Jones solids [19,33] and Gaussian-core systems [9,17]. Systems described with more complex potentials, such as Morse [34], modified Buckingham [15] and many-body potentials (pair functionals) [7,8,35,36] have also been studied using BOPs.

The BOP technique is commonly used in the investigations of nanoscale systems, such as atomic clusters [35–37], and gold nanowires [7,8]. BOPs have also been used in studies of shear-induced phenomena (e.g. shear-induced ordering [38], shear-induced crystallisation [39], and shear-induced overaging [40]), anomalies in liquids [41,42], the freezing of argon in porous carbon [43], quasicrystals [26], and even plasma [44].

The widespread use of the BOP technique spurred a number of extensions or generalisations over the last decade. A modification, where an additional averaging over nearest neighbours is performed during the calculation of local BOPs was proposed by Lechner et al. [9] in order to improve identification of a variety of crystalline structures. A new formulation, where BOPs are combined with Voronoi tessellation, was proposed by Mickel et al. [45]. This formulation removes the ambiguities introduced by the

arbitrariness in the choice of a cutoff radius, allowing a better characterisation of the orientational order of disordered systems.

The fact that calculating BOPs involves repeated evaluation of spherical harmonics (SHs) [46] means that it is a computationally intensive approach. In a benchmark of methods for structural analysis Stukowski [47] assigns it a computational cost factor of 100, compared with 50 for Voronoi analysis [48,49], 3 for common neighbour analysis (CNA) [50] and 1 for the centro-symmetry parameter technique (CSP) [51]. The high computational effort associated with BOPs narrows the spectrum of their potential applications, and we are not aware of any examples in the literature where they would be used for the analysis of large-scale simulations—these typically employ computationally cheaper methods, such as energy filtering (e.g. Refs. [52,53]), or CSP (e.g. Ref. [54]). Being able to use a more involved method that BOP constitutes for large-scale systems is an enticing prospect. A discussion of accuracy and limitations inherent in a number of approaches to structural analysis is given in Ref. [47].

Moreover, the availability of the BOP approach to researchers is limited. To our knowledge, its implementation is not bundled with any of the well-known molecular dynamics codes or visualisation tools, while CSP, CNA or Voronoi analysis are offered by e.g. LAMMPS [55] or OVITO [56]. The authors are aware of only two implementations available to the scientific community: one due to Lechner et al. [9], and another one due to Wang et al. [36]. Both of these implementations are less computationally efficient compared to competing, simpler approaches.

With the above considerations in mind, we feel a highly-efficient approach for the evaluation of bond-orientational order parameters has the potential to widen their spectrum of application. In this paper we propose a novel, approximate method for efficient calculation of BOPs, which can reduce the computational effort by a factor of up to 50, allowing it to outperform even the four approaches generally regarded as faster, i.e. CSP, CNA, CNP (common neighbourhood parameter) [57] and Voronoi analysis.

The paper is organised as follows. In Section 2 we describe the BOP approach, highlighting the steps in the calculation that can be optimised. Section 3 outlines the proposed technique for evaluating BOPs. Section 4 is devoted to benchmarking the efficiency and accuracy of the proposed approach. Section 5 contains conclusions.

2. The bond-orientational order parameter (BOP) method

Bond-orientational order parameters [1,2] (BOPs) are used to characterise short-range order by classifying each atom as belonging to one of a number of close-packed structures. For every reference atom i , the classification is performed in four stages, outlined below.

Stage 1.

The set $B(i)$ of nearest neighbours j of atom i is determined. Nearest neighbours are defined as atoms that are no further away from atom i than a prescribed cutoff radius r_c . The bond vectors $\mathbf{r}_{ij} = \mathbf{r}_j - \mathbf{r}_i$ joining atom i with the neighbours j are calculated as Cartesian components. The number of neighbours of atom i (cardinality of $B(i)$) will be denoted by $N_b(i)$.

Stage 2.

Each bond vector \mathbf{r}_{ij} is projected to the unit sphere, and its spherical coordinates $\theta(\mathbf{r}_{ij})$ and $\phi(\mathbf{r}_{ij})$ are calculated.

Stage 3.

A vector of complex spherical harmonics (SHs) [46] $Y_l^m(\theta(\mathbf{r}_{ij}), \phi(\mathbf{r}_{ij}))$ is evaluated for every bond vector \mathbf{r}_{ij} , for a chosen value of l and $m \in \{-l, \dots, l\}$.

Table 1
BOP values for typical ideal structures.

Structure	Parameter			
	Q_4	Q_6	\hat{W}_4	\hat{W}_6
sc	0.76376	0.35355	0.15932	0.01316
bcc	0.08202	0.50083	0.15932	0.01316
fcc	0.19094	0.57452	-0.15932	-0.01315
hcp	0.09722	0.48476	0.13410	-0.01244
icos	0	0.66332	0.	-0.16975

Stage 4.

A vector of complex quantities $Q_{l,m}(i)$, defined as

$$Q_{l,m}(i) = \frac{1}{N_b(i)} \sum_{j \in B(i)} Y_l^m(\theta(\mathbf{r}_{ij}), \phi(\mathbf{r}_{ij})) \quad (1)$$

is constructed. Subsequently so-called second-order (Steinhardt) invariants are constructed, according to

$$Q_l(i) = \left(\frac{4\pi}{2l+1} \sum_{m=-l}^l |Q_{l,m}(i)|^2 \right)^{1/2}. \quad (2)$$

Third-order invariants [58] can also be constructed:

$$\hat{W}_l(i) = W_l(i) \times \left(\sum_{m=-l}^l |Q_{l,m}(i)|^2 \right)^{-3/2}, \quad (3)$$

where

$$W_l(i) = \sum_{\substack{m_1, m_2, m_3 \\ m_1 + m_2 + m_3 = 0}} \begin{pmatrix} l & l & l \\ m_1 & m_2 & m_3 \end{pmatrix} Q_{l,m_1}(i) Q_{l,m_2}(i) Q_{l,m_3}(i). \quad (4)$$

The quantities

$$\begin{pmatrix} l_1 & l_2 & l_3 \\ m_1 & m_2 & m_3 \end{pmatrix}$$

are Wigner 3j-symbols [59].

Invariants for $l \leq 3$ vanish for lattices with cubic symmetry, and in practice bcc, fcc and hcp are differentiated using $l = 4$ and $l = 6$. Each atom i is classified as belonging to a particular structure by direct comparison of calculated invariants $Q_4(i)$, $Q_6(i)$, $\hat{W}_4(i)$ and $\hat{W}_6(i)$ with reference values. The values for a number of ideal structures are given in Table 1.

At non-zero temperatures thermal motions lead to a smearing of the BOP values, and in practice classification is performed on a two-dimensional plane of parameter values (e.g. $Q_4(i) - Q_6(i)$ or $Q_6(i) - \hat{W}_4(i)$), on which regions corresponding to particular packings are defined. For examples see e.g. Refs. [7,15]. However, no single, consistent approach to the classification itself has been proposed to date.

3. A highly-efficient approach for evaluating BOPs

The four stages of the calculation of BOPs outlined in Section 2 differ in computational effort. For the identification of neighbours (stage 1) efficient, linear-scaling algorithms, such as the linked-cell approach [60,61] are typically used, and calculating bond vectors \mathbf{r}_{ij} and their spherical coordinates (stage 2) are very simple operations. The computational effort of stages 1 and 2 is thus very small.

3.1. Evaluation of spherical harmonics by fast simultaneous interpolation

The most computationally demanding (about 60% of total effort for a standard implementation) is stage 3, where a large number of

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