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Implementation of the Projector Augmented-Wave Method: The Use of Atomic Datasets in the Standard PAW-XML Format

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Abstract. The projector augmented-wave (PAW) method is an important approach for electronic structure calculations based on the Kohn–Sham density functional theory. And the PAW atomic dataset plays an essential role in the implementation and application of the method. The intensive use of proprietary datasets with limited metadata in previous years has led to difficulties in both the cross-validation of PAW codes and the understanding of the accuracy and transferability of PAW atomic data. In this work, we focus on the open-source ABINIT Jollet–Torrent–Holzwarth dataset (JTH) library in the PAW-XML format and investigate the implementation techniques to clarify how the atomic data participate in the computations. We propose an intermediate dataset that extends the original PAW-XML one by atomic quantities in derived forms, which facilitate the PAW implementation using the JTH library and cover the differences between PAW datasets. Our implementation is validated by comparing the structural property results of representative bulk materials and molecules with those calculated by ABINIT using the same datasets. Moreover, we discuss the feasibility of using the intermediate dataset for a quick support of PAW-XML datasets in existing PAW codes.

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Key words: Density functional theory, PAW method, PAW-XML dataset.

1 Introduction

The projector augmented-wave (PAW) method, originally proposed by Blöchl in 1994 [7] and further reformulated by Kresse and Joubert in 1999 [31], plays an essential role nowa-

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days in the Kohn–Sham density functional theory (DFT) calculations [8, 32, 34]. The advantages of the PAW method over traditional pseudopotential methods [23, 46] lie in two aspects. First, the PAW method has an elegant theoretical framework [7, 25, 31]. This framework builds upon a linear transformation between the all-electron (AE) and the pseudo (PS) wave functions, and then taking the PS wave functions as variational quantities, derives the AE densities, energy functionals, the Hamiltonian, and forces in a consistent manner. Second, compared to the ultra-soft pseudopotentials (USPPs) [46] which also offer an elaborate solution for efficient DFT calculations, the construction of the PAW atomic dataset is easier because less parameters are involved [31]. The PAW dataset or the pseudopotential provides necessary atomic data for DFT calculations, and is critical in the sense that the quality of these data determines fundamentally the reliability of a calculation [7, 34, 40].

The implementation of the PAW method, however, is complex [16, 22, 32, 38, 45]. And a lot of efforts have been devoted to the implementation in the last few decades. At the early stage after the development of the PAW method, there were only Blöchl's own implementation, the CP-PAW code [1], and the PWPAW code by Holzwarth [25, 41]. Some detailed formulas used in the PWPAW code have been addressed in [25]. The developers of VASP [29, 30] implemented the PAW method after reformulating Blöchl's original formalism to derive the relationship between the USPP and the PAW methods [31]. From then on, almost all the PAW implementations employed the new Kresse–Joubert formalism. The GPAW code [12] implemented the PAW method using uniform real-space grids, and the grid-based formulas have been described in [37]. In addition to VASP, the ABINIT code [20, 21] supplies another plane-wave implementation of the PAW method. The formulas used in the code, specifically those for the on-site terms, have been given in [44]. Other known implementations of the PAW method include S/PHI/nX [9, 14], PWSCF [19] and ONETEP [24].

Note that these codes use different PAW atomic dataset libraries. The PAW dataset library of VASP is among the most popular ones in DFT calculations [18]. It covers all elements in the periodic table, and usually produces very good results for bulk materials near equilibrium. However, the use of these datasets is limited to VASP users. What is more, the generation code, the full inputs, and the testing data are not distributed with VASP. Therefore, this high quality PAW library offers limited help to the understanding of the accuracy and the transferability of atomic datasets in practical calculations. In contrast, the PWPAW, GPAW, ABINIT, and PWSCF codes employ open-source PAW dataset libraries [10, 12, 26] and the generation codes and input parameters are fully available. Nevertheless, due to the lack of a standard format for the PAW dataset in previous years, the interoperability of these datasets among codes is still not satisfactory.

Recent work on pseudopotentials and PAW datasets laid great emphasis on open-source libraries in a standard file format [10, 16, 18, 38, 42]. From version 0.2, the Jollet–Torrent–Holzwarth (JTH) dataset library [27] provided by ABINIT began to follow the PAW-XML specification as presented on the website of CECAM's Electronic Structure Library [4]. In addition, the accuracy of the recent JTH library has been demonstrated to