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Gauge transformations to combine multi-component many-body interatomic potentials

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Many-body interatomic potentials play an important role in atomistic modelling of materials. For pure elements it is known that there exist gauge transformations that can change the form of the potential functions without modifying its properties. These same transformations, however, fail when applied to alloys. Even though different research groups may use the same potentials to describe pure elements, the gauges employed for fitting alloys will generally be different. In this scenario, it is *a priori* impossible to merge them into one potential describing the combined system, and thus no advantage is taken from state-of-the-art developments in the literature. Here, we generalise the gauge transformations applied to pure species in order to leave the properties of alloys invariant. Based on these transformations, a strategy to merge potentials developed within different gauges is presented, aiming at the description of the combined system. Advantage of existing state-of-the-art potentials is so taken, thus focusing the efforts on fitting only the missing interactions. Such a procedure constitutes a helpful tool for the development of potentials targeted to alloys of increased complexity, while maintaining the description quality of their constituents.

Keywords: gauge transformations; many-body potentials; multi-component alloys

By the ever increasing availability of computing power steered in part, the atomistic simulation of materials has become a widespread tool; concurrently, the complexity of the systems aimed for has also increased. At the core of many of these simulations are the many-body interatomic potentials, required to provide energies and forces for arbitrary configurations as reliably as possible. As a consequence, any result obtained with such tools is biased by the quality of the used potential.

Depending on the range of applications desired and the level of detail, the development of many-body potentials can be a straightforward or a formidable task.

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Not very far in the past, only a few material specific properties were considered in the fitting procedure and a typical publication described potentials for a wide range of elements and their alloys, for example [1–5]. Nowadays, however, the reliability demands for specific materials are stressing the quantitative aspects more and more, with a typical publication being devoted to a single element or alloy, for example [6–10].

Recently, advantage has been taken of advanced, state-of-the-art potentials, for the construction of new potentials for binary and ternary alloys, aimed at the modelling of steels, [11–14]. The further combination of such binaries (or ternaries) to tackle more complex multi-component alloys seems, however, hindered by a technical detail stemming from the potentials themselves. Since the introduction of many-body potentials in the early 1980s, it was soon realised that the potential functions for a pure element have no unique form. There exist mathematical transformations [3,15,16] that can change the functional forms without modifying the energetic description, commonly known as gauge transformations. These gauge transformations are no longer valid when the pure elements are part of an alloy, due to the presence of the cross-interactions. On the other hand, if two binary potentials with a common element are to be merged into a ternary potential, the common element must be described by the same potential in the same gauge. The latter poses a major constraint since different authors may use the same potentials to describe the pure elements, but often use different gauges to build their alloys. It is thus unclear if and how advantage of separate efforts can be taken.

In this article, we eliminate this constraint by generalising the gauge transformations valid for pure species potentials so that they also hold for potentials describing multi-component alloys. Based on these transformations, a strategy to combine multi-component potentials is presented and a practical example is given.

The total energy of an atomic assembly, E_{tot} , of most widely used many-body potentials of today may be cast in the general form,

$$E_{\text{tot}} = \frac{1}{2} \sum_{i,j} 'V_{i,j}(r_{ij}) + \sum_i F_i(\rho_i), \quad (1)$$

$$\rho_i = \sum_j ' \varphi_{i,j}(r_{ij}), \quad (2)$$

where i and j run over all the atoms in the system with $i \neq j$. Here, besides a standard distance dependent (r_{ij}) pair term, V , a second term, F , is included to account for the many-body interaction of an atom with its surrounding neighbours. The latter term depends on the so-called local density, ρ , that in turn is built up by the superposition of pairwise contributions from so-called density functions, φ . Moreover, all the interactions generally depend on the constituent species, t_i .

For pure species, the above formalism is known after the names of three independently developed models, namely, ‘glue model’ (GM) [17,18]. ‘embedded atom method’ (EAM) [19], and ‘Finnis–Sinclair’ (FS) formalism [15]. Though, within all of them the pair term is assumed to stand for the repulsive interaction between nuclei, the physical interpretation given to the many-body term is nonetheless different. Irrespective of the model, however, due to the large number of

approximations involved, the connection with their more fundamental theoretical basis remains heuristic. Moreover, the existence of gauge transformations blurs, to some extent, the interpretation of the individual terms themselves. For pure species, separated into two components for clarity, this transformation is given by [3,15,16],

$$\hat{G}_1 \begin{cases} \tilde{\varphi}(r) = S\varphi(r) \\ \tilde{F}(\tilde{\rho}) = F(\tilde{\rho}/S) \end{cases}, \quad (3)$$

$$\hat{G}_2 \begin{cases} \tilde{V}(r) = V(r) + 2C\varphi(r) \\ \tilde{F}(\rho) = F(\rho) - C\rho \end{cases}, \quad (4)$$

with S and C as arbitrary constants. Transformation \hat{G}_1 is nothing but the freedom to choose the density units; transformation \hat{G}_2 shows that the pair term can also contribute to binding. From the fitting point of view, the above is not a problem but a useful tool. In fact, the transformation to the so-called ‘effective gauge’ [15] is a particularly suitable one to obtain simpler equations. There, the constant C is chosen as $C = F'(\rho_{\text{eq}})$, where ρ_{eq} is the equilibrium density of a perfect crystal structure selected as reference. The \tilde{V} so obtained is, in a sense, the closest pair interaction to the full one, in the neighbourhood of the reference. It is also common to choose S such that $\rho_{\text{eq}} = 1$.

The easiest way to see how Equations (3) and (4) can be generalised to alloys is to consider a binary system composed of species A and B . In place of \hat{G}_1 we now set,

$$\hat{G}_1 \begin{cases} \tilde{\varphi}_{AX} = S_A\varphi_{AX}, & \tilde{\varphi}_{BX} = S_B\varphi_{BX} \\ \tilde{F}_A(\tilde{\rho}) = F_A(\tilde{\rho}/S_A), & \tilde{F}_B(\tilde{\rho}) = F_B(\tilde{\rho}/S_B) \end{cases}, \quad (5)$$

where X takes the values A and B . The key point here is that the scaling factors, S , are associated with the target species (first subscript of the pair), thus allowing for a uniform scaling of the total density at the corresponding site, leading to the second line in Equation (5). On the other hand, \hat{G}_2 is now generalised as,

$$\hat{G}_2 \begin{cases} \tilde{V}_{XY} = V_{XY} + C_X\varphi_{XY} + C_Y\varphi_{YX} \\ \tilde{F}_A(\rho) = F_A(\rho) - C_A\rho, & \tilde{F}_B(\rho) = F_B(\rho) - C_B\rho \end{cases}, \quad (6)$$

where X, Y take the values A, B . As before, the central point here is that the constants, C , are associated with the target species. Thus, any linear contribution in the density subtracted from the many-body terms (second line) can be added back symmetrically distributed, into the pair terms (first line). Clearly, the equations for a higher order alloy are the same, namely each species carries its own constants S and C , while allowing the dummy subscripts to vary upon a larger set. Interestingly, from a different viewpoint and for binary alloys within EAM (where $\varphi_{XY} = \varphi_Y$), Equation (6) was also obtained in [3]; clearly, however, their validity extends to a wider context than the one given there.

These arguments can be exploited for the construction of potentials for complex multi-component alloys, starting from simpler ones possessing some species (i.e. potentials) in common. Let us consider the alloys $A_1 = \{X_1, \dots, X_K,$

$Y_1, \dots, Y_M\}$ and $A_2 = \{X_1, \dots, X_K, Z_1, \dots, Z_N\}$ consisting of $(K+M)$ and $(K+N)$ elements, respectively, the goal being to obtain a potential for the combined alloy $A_1 \cup A_2$. First, one disposes of K pairs of constants, (S, C) , within each of the two that transform the overlapping system into a common gauge, such as the effective one mentioned earlier. Then, all the potentials are merged into a single set and the effort is concentrated on determining the missing $(2 \times M \times N)$ cross-density functions, φ_{YZ} , φ_{ZY} , and the $(M \times N)$ cross-pair interactions, V_{YZ} .

As an example of the above procedure, we consider the Fe–Cu–C ternary. We select two potentials found in the literature; namely, the Fe–Cu EAM-type potential by Pasianot and Malerba [11] (henceforth P_{FeCu}), and the Fe–C GM-type potential by Hepburn and Ackland [13] (henceforth P_{FeC}). Both potentials have the Fe part developed by Mendelev et al. [8] in common, but used in different gauges. Strictly, the Fe in [13] is not Mendelev's, but currently there is an alternative based on the latter with equally good performance [20]. For P_{FeCu} , the Fe potential was transformed into the effective gauge prior to fitting the cross-interaction, V_{FeCu} , while for P_{FeC} the Fe potential was used in its original form. The potentials, P_{FeCu} and P_{FeC} , are thus defined by the functions,

$$P_{\text{FeCu}} = \begin{cases} V_{\text{FeFe}}^{\text{eff}}, V_{\text{CuCu}}, V_{\text{FeCu}} \\ \varphi_{\text{Fe}}^{\text{eff}}, \varphi_{\text{Cu}} \\ F_{\text{Fe}}^{\text{eff}}, F_{\text{Cu}} \end{cases}, \quad (7)$$

$$P_{\text{FeC}} = \begin{cases} V_{\text{FeFe}}, V_{\text{CC}}, V_{\text{FeC}} \\ \varphi_{\text{FeFe}}, \varphi_{\text{CC}}, \varphi_{\text{FeC}}, \varphi_{\text{CFe}} \\ F_{\text{Fe}}, F_{\text{C}} \end{cases}. \quad (8)$$

We may only transform P_{FeC} , while keeping P_{FeCu} unchanged. Specifically, by choosing the gauge constants $C_{\text{Fe}} = 0.116$, $C_{\text{C}} = 0$, $S_{\text{Fe}} = 0.038$ and $S_{\text{C}} = 1$, one finds the combined potential,

$$P_{\text{FeCuC}} = \begin{cases} V_{\text{FeFe}}^{\text{eff}}, V_{\text{CC}}, \tilde{V}_{\text{FeC}}, V_{\text{FeCu}} \\ \varphi_{\text{FeFe}}^{\text{eff}}, \varphi_{\text{CuCu}}, \varphi_{\text{CC}}, \tilde{\varphi}_{\text{FeC}}, \varphi_{\text{CFe}}, \varphi_{\text{FeCu}}, \varphi_{\text{CuFe}} \\ F_{\text{Fe}}^{\text{eff}}, F_{\text{Cu}}, F_{\text{C}} \end{cases}. \quad (9)$$

The newly introduced functions, φ_{XFe} and φ_{XCu} , are given by φ_{Fe} and φ_{Cu} , respectively. To complete P_{FeCuC} , the cross-functions $\{V_{\text{CuC}}, \varphi_{\text{CuC}}, \varphi_{\text{CCu}}\}$ need to be fitted to the desired material properties of the Fe–Cu–C alloy.

To conclude, it is worth remarking that both the Fe–Cu and Fe–C potentials cover four different publications; namely, the Fe potential developed by Mendelev et al. [8], the Cu potential developed by Mishin et al. [7], the Fe–Cu cross-interactions developed by Pasianot and Malerba [11] and the Fe–C and C–C interactions developed by Hepburn and Ackland [13]. Thus, much effort went into a detailed description of the interactions of the subsystems. Clearly, this level of performance is unattainable if the potentials of all the subsystems needed to be fitted from scratch. The procedure proposed here is therefore a helpful tool for the development of

potentials targeted to alloys of increased complexity, while maintaining the descriptive quality of their constituents.

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