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Abstract

The nudged-elastic band (NEB) method is modified with concomitant two climbing images (C2-NEB) to find a transition state (TS) in complex energy landscapes, such as those with serpentine minimal energy path (MEP). If a single climbing image (C1-NEB) successfully finds the TS, C2-NEB finds it with higher stability and accuracy. However, C2-NEB is suitable for more complex cases, where C1-NEB misses the TS because the MEP and NEB directions near the saddle point are different. Generally, C2-NEB not only finds the TS but guarantees that the climbing images approach it from the opposite sides along the MEP, and it estimates accuracy from the three images: the highest-energy one and its climbing neighbors. C2-NEB is suitable for fixed-cell NEB and the generalized solid-state NEB (SS-NEB). We validate the C2-NEB method on the solid-solid phase transformations in NiTi, and find agreement with experiment.

Disciplines

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C2-NEB: THE NUDGED ELASTIC BAND METHOD WITH TWO CLIMBING IMAGES, VALIDATED ON THE NiTi SHAPE-MEMORY MARTENSITIC TRANSFORMATION

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For solid-solid phase transformations in inorganic materials, we developed a nudged-elastic band (NEB) method with two concomitant climbing images (C2-NEB) that can find a transition state in complex energy landscapes, including those with a serpentine minimal energy path. We validate it on the solid-solid phase transformations in Ti and NiTi.

New discoveries often require development of new methods. To address a solid-solid phase transformation on the atomic scale at a finite temperature, one should know the involved terminal stable structures and have a method for controlling atomic motion during the transformation. The nudged elastic band (NEB) method coupled with density-functional theory (DFT) is often used to determine the minimum-energy path (MEP) and the transition states (TS). We broaden the NEB applicability and provide an extension to two climbing images (C2-NEB) method [1,2] that is more stable and reliable for complicated potential-energy landscapes.

For validation, we applied the C2-NEB [1] to the solid-solid phase transformations in NiTi [2,3,4]. We found the minimal energy path with a small potential energy barrier for the martensitic transformation between NiTi austenitic and martensitic phases [3]. Considering a 2-dimensional (2D) example with a twisted MEP having a zigzag near the TS (see Fig.5 in [2]), we demonstrated that C2-NEB is more efficient, because it can find the TS with only 3 images, while C1-NEB [5] can require up to 35 images for convergence to the lowest-energy saddle.
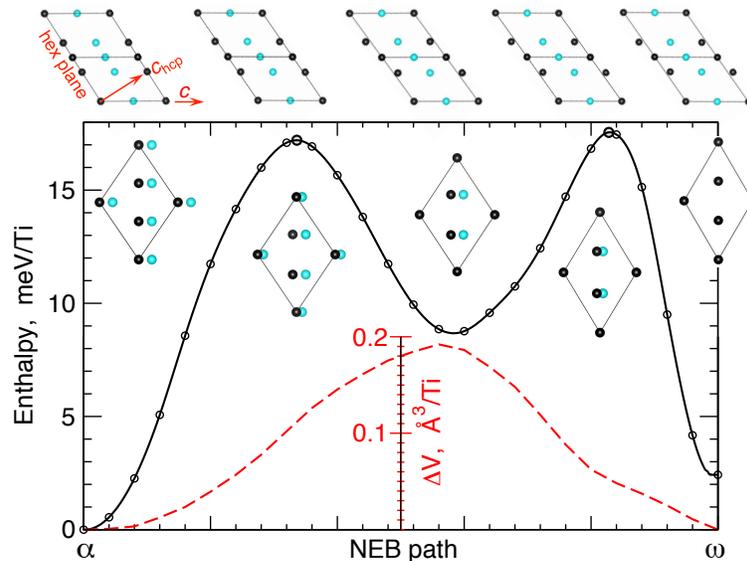

Fig. 1: Ti α–ω transformation under pressure. Enthalpy (solid line) and excess volume (red dashed line, middle scale) from no-climbing NEB (open circles) and C2-NEB (larger circles) methods. Insets are the structural projections of the supercell along [110] and [001] at enthalpy extrema. Ti atoms on the alternating hcp planes are black and blue.

Figs. 1 and 2 in [2] illustrate a 2D potential energy landscape, for which C2-NEB with only 3 images works correctly and finds the TS, while the convergence criteria of C1-NEB with a small number of images will never be satisfied.

Here we apply the C2-NEB code [1] to the α–ω transformation in pure Ti under pressure (Fig. 1). As expected [2], we confirm stability of the C2-NEB code, which is available online [1].

In the C2-NEB method [1,2], at each optimization step, the highest-enthalpy image (HEI) is found. If both neighbors of the HEI are movable (i.e., they are not *the fixed terminal points*), then they climb, approaching the HEI, which is nudged to them. C2-NEB method [2] can be coupled with any DFT or classical molecular dynamics code, capable of computing structural enthalpies and atomic forces. Our code [1] is combined with VASP [6], which provides the atomic forces and the total enthalpy (TOTEN variable) for each image. Implemented to replace the NEB subroutine in VASP, our C2-NEB open-source code is available for download [1].

Martensitic transformation in the NiTi shape memory alloy is addressed in refs. [2,3,4]. Pure Ti is one of the most used structural metals. Here we use C2-NEB to locate two enthalpy maxima for the Ti α–ω (TAO-1) transformation [9], see Fig. 1. To correct the well-known systematic DFT error arising from over-binding of d-electrons, we use DFT+U method [7] with (U–J)=2.2 eV, which provides the α–ω equilibrium coexistence pressure of 2 GPa [8]. For faster convergence, first we obtain the whole trajectory without climbing [5]. Interestingly, Ti α–ω transformation [9] has an intermediate minimum (Fig. 1): this is a lower-density metastable structure, which can be stabilized by negative pressures; it has the maximal excess volume $\Delta V=V(x)–V(0)+x[V(0)–V(1)]$, where the rescaled NEB path coordinate x changes from 0 (α) to 1 (ω). Then, we separately address each local maximum: we use two images on both sides of a maximum as the terminal points, add 3 images between them, and use C2-NEB to converge those 3 images to the saddle point (larger grey circles in Fig. 1); they convergence to the same enthalpy within the DFT error [<0.5 meV/atom], while the central image always remains slightly [~0.1 meV/atom] higher in enthalpy than both climbing images. As expected [2], convergence of the C2-NEB code [1] with only 3 images is stable regardless of any possible zigzags of the MEP.

In summary, we find that the C2-NEB algorithm [1,2] is very useful, especially for systems with a serpentine MEP or unknown behavior of the NEB trajectory near the TS. We validated it on the martensitic transformations in NiTi shape-memory alloy [2,3,4] and in pure titanium under pressure. Our C2-NEB implementation is available as an open-source code [1].

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