High-throughput DFT study on MM'X alloys for magnetic refrigeration

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In this study, we developed a high-throughput DFT method to identify promising MM'X alloys for magnetocaloric applications. We first screened for stable structures using formation energy and our newly defined distortion parameter. Next, we estimated the Curie and structural transition temperatures to ensure magnetostructural transitions occur near room temperature. Finally, further filtering with Σ_M and λ_2 narrowed our candidates to 7 alloys with high magnetocaloric potential. This framework not only expanded the chemical space for screening but also reduced computational costs for designing complex alloys.

Keywords: magnetic refrigeration; MM'X alloys; high-throughput DFT; magnetostructural transitions

1. Introduction

Achieving carbon neutrality is a global priority focused on energy conservation and emissions reduction. Room-temperature magnetic refrigeration utilizing the magnetocaloric effect (MCE), offers a promising alternative and solution [1]. MM'X alloys (where M and M' are transition metals and X is a p-block element), with compositionally tunable magnetostructural transitions (low-temperature orthorhombic (ort) to high-temperature hexagonal (hex) structures), exhibit high MCE potential. To accelerate material discovery, this study introduces a novel and efficient high-throughput computing and density functional theory (HTP-DFT) screening framework (Fig.1) to facilitate the sought of MM'X compositions based on properties of interest. This framework efficiently evaluates MM'X alloys, enabling rapid identification of optimal magnetocaloric materials within established computational constraints. By streamlining the selection process, this approach aims to advance magnetic refrigeration technology for sustainable cooling solutions.

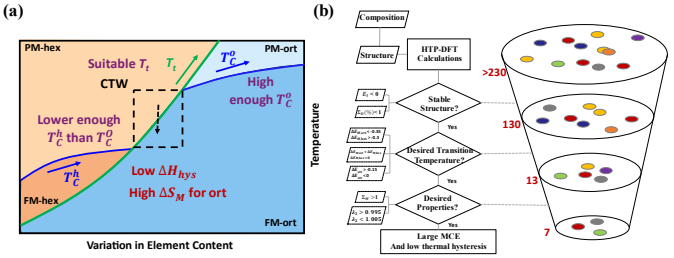

Figure 1: (a) A schematic representation of a MM'X compound exhibiting the desired properties and a magnetostructural transition, specifically when the structural phase transition temperature (T_t) falls within the Curie Temperature Window (CTW). (b) HTP-DFT workflow designed to screen novel magnetic refrigeration materials for MM'X compounds.

2. Results and discussion

The thermodynamic stability was evaluated using the formation energy (E_f), and we defined a new relative structural distortion parameter $\Sigma_D = \Sigma\left(\frac{|\theta_{cal}-\theta_{ex}|}{\theta_{ex}}\right)$ to assess structural viability as it measures changes in the unit cell angles after full relaxation. Of 230 compounds, 181 show stable (negative E_f) low-temperature ort. structures. Applying our Σ_D criterion, we narrow this down to 130, including known experimentally validated MM'X alloys (Fig. 2(a)).

Using the mean-field approach based on the DFT-calculated energy differences, we estimated the Curie temperature (T_C) and structural transition temperature (T_t) and established the CTW [2] —T_C from the FM-AFM energy difference (ΔE_M) and T_t from the ort.-hex. energy difference (ΔE_{str}). Screening for room-temperature magnetostructural transitions yields 13 candidates (Fig. 2(b)). For further evaluation of MCE and thermal hysteresis (ΔT_{hys}), we set the screening criteria as $\Sigma_M > 1.5$ [3] and $0.995 < \lambda_2 < 1.005$ [4], where 7 promising compositions are identified (Fig. 2(c)). This HTP framework efficiently highlighted 7 potential high-performance magnetocaloric materials, demonstrating its utility in complex alloy designs for magnetic refrigeration [5].

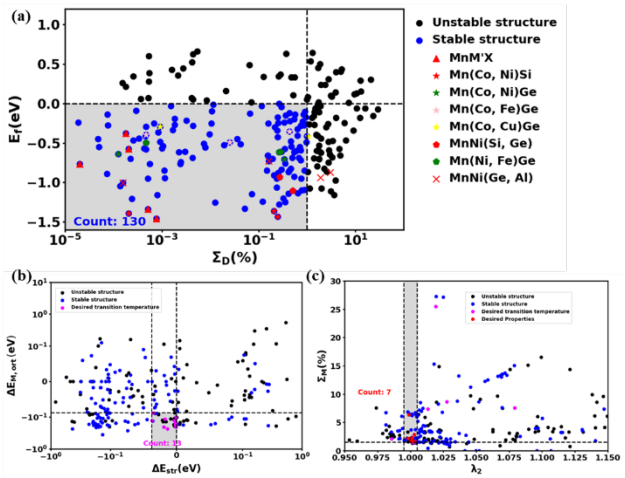

Figure 2: Screening process: (a) DFT-calculated formation energy and structural deformation; the grey regions are screened to identify (b) the desired transition temperature and (c) additional desired properties. Well-known experimental MM'X alloys are included for validation in (a).

References

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