Supporting information

Broad negative thermal expansion operation-temperature window achieved by adjusting Fe-Fe magnetic exchange coupling in La(Fe,Si)$_{13}$ compounds

Shaopeng Li,†, §   Rongjin Huang,*,† Yuqiang Zhao,†, §   Wen Li,†, §   Wei Wang,†
Chuanjun Huang,† Pifu Gong,‡, §   Zheshuai Lin,‡ and Laifeng Li*,†

† State Key Laboratory of Technologies in Space Cryogenic Propellants, Technical Institute of Physics and Chemistry, Chinese Academy of Sciences, Beijing, P.R. China

‡ Beijing Center for Crystal R&D, Key Lab of Functional Crystals and Laser Technology, Technical Institute of Physics and Chemistry (TIPC), Chinese Academy of Sciences (CAS), Beijing, P.R. China

§ University of Chinese Academy of Sciences, Beijing, P.R. China

* To whom correspondence should be addressed. E-mail: huangrongjin@mail.ipc.ac.cn; laifengli@mail.ipc.ac.cn

The calculated method details of Fe^I-Fe^{II} interatomic distance for $LaFe_{13-x}Si_x$ (x=2.8, 2.9, 3.0, 3.1)

For $LaFe_{13−x}Si_x$ in tetragonal phase exhibiting no negative thermal expansion (NTE) effects, only the cubic phase in the two-phase mixture was considered. In order to investigate the relationship between Si content x and the Fe-Fe interatomic distance, the first-principles geometry optimization calculations for $LaFe_{13−x}Si_x$ (2.8≤x≤3.1) at 0 K are performed using the plane-wave pseudopotential method[1] implemented in CASTEP package.[2] Virtual Crystal Approximation (VCA)[3,4] is also implemented in the calculations for the statistical distribution in the periodic structure. The initial structures for different Si contents were extrapolated from $LaFe_{11}Si_2$ determined by experiment. The normal-conserving pseudopotential[5] and the generalized gradient approximation (GGA) with Perdew, Burke and Emzerhof (PBE) functionals[6] are chosen to describe the exchange and correlation (XC) potentials in the calculation of the optical properties. The plane-wave cutoff energy was set at 550eV and the Monkhorst-Pack k-point meshes[7] were set with a density of 4×4×4 points in the Brillouin zone.

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