Electrical Steel: An investigation into the brittleness of the Fe-Si alloy

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Abstract

In this paper, we'll hunt for ordered crystal structures that may be stable, and that would explain the brittleness of electrical steel. We'll do this by exploiting the advantage that ab initio simulations (done using Quantum ESPRESSO (opEn-Source Package for Research in Electronic Structure, Simulation, and Optimization) [1] [2] [3]) give full control over defining the crystal: We'll tell exactly where we want to have every atom, and quantum chemistry will tell us what is the internal energy that corresponds to such crystal. By modifying the positions of the atoms, and by monitoring the corresponding internal energy, we may find ordered crystals that areo thermodynamically stable. (This is, of course, only a draft)

I. Introduction

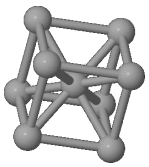
Electric applications such as motors, transformers or generators all have a magnetic material in the core of their electromagnetic coil. Almost always, this is a so-called electrical steel: a Fe-Si alloy with about 3 wt.% Si. It is known since decades that using a steel with 6.5 wt.% Si would be very advantageous[citation needed] over the steel we're now using. Such a steel would reduce energy losses in the application, which would make it possible to build electric machines lighter and more energy efficient. Estimates come up with a saving of several billions of euro worth of electricity every year [citation needed]. So why aren't we using that ideal electrical steel? In contrast to 3 wt.% Si, the 6.5 wt.% Si steel is brittle: you can't press or roll or otherwise form it into the size and shape needed to build the electric apparatus. It would just break apart when trying to do so. Hence finding an electrical steel with 6.5 wt.% Si that is not brittle is quite the holy grail in electrical steel research. There is a hypothesis about why the brittleness appears. Crystals with long range order are usually more brittle than crystals in which the atoms are more disordered [citation needed]. It is assumed that when increasing the silicon content, there is a stronger tendency for the atoms to develop short-range order.

II. Convergence testing

We'll be using the SPSS pseudopotentials [4] [5] for all of our calculations.

i. bcc-Fe

For bcc-Fe iron we use Patrick M. Woodward et.al's cif file. We're also using a pseudopotential generated using "atomic" code by A. Dal Corso v.5.0.99 svn rev. 10869 with the minimum cutoff for wavefunctions being the suggested 64. Ry and the minimum cutoff for charge density the suggested 782. Ry. We use the hydrostatic pressure to test the convergence, for different k-meshes we find the following:



	k mesh	Hydrostatic Pressure (kbar)
1	(1,1,1)	291.45
2	(3,3,3)	76.49
3	(5,5,5)	-99.45
4	(7,7,7)	-71.98
5	(9,9,9)	-109.31
6	(10,10,10)	-98.83
7	(11,11,11)	-80.23
8	(13,13,13)	-92.67
9	(15,15,15)	-84.76

We find it has sufficiently converged at (10,10,10) so that's the k-mesh we'll be using. Now keeping the cutoff charge density ≈ 12 times the cutoff for wavefunctions, we'll vary this wavefunction cutoff:

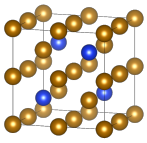
	ecutwfc	Hydrostatic Pressure (kbar)
1	14	-15244
2	24	-1727
3	34	-962
4	44	-457
5	54	-95
6	64	-98

Here we see quite a good convergence at ecutwfc = 54, we'll now take ecutwfc=54 and vary the multiplicity of the charge density:

	factor	ecutrho	Hydrostatic Pressure (kbar)
1	2	108	939
2	4	216	0.81
3	6	324	-69
4	8	432	-96
5	10	540	-92
6	12	648	-95

We already see convergence at a factor 8. Our final values are thus a k-mesh of (10,10,10), ecutwf at 54 and ecutrho at 432.

ii. DO3-Fe3Si



DO3-Fe_3Si is the name of the crystal with a unit cell built from 8 bcc-Fe unit cells stacked in a 2x2x2 way, to make a cube with twice the edge length of a normal bcc-Fe unit cell. Herein every other unit cell's middle iron nucleus is replaced with a Si nucleus, such that these 4 atoms form a tetrahedron. This is a bit hard to recognize in the figure on the left[1] as that one is shifted up and to the left, but this is in fact the same compound. We thus have a unit cell with 16 atoms of which 4 are Si atoms, giving us a concentration of 25 at.% ≈ 14.36 wt.%. Let's now do the same procedure as with bcc-Fe, we wish to have a k-mesh, ecutwf and ecutrho that will work for both so we can later on ignore these parameters. The unit cell has doubled in size so the k-mesh should be halved let's see if we thus get convergence at (5,5,5). Starting with ecutwfc=64 and ecutrho=782 as these, the minimal values for the Fe pseudopotential, are higher than the ones in the Si pseudopotential:

	k mesh	Hydrostatic Pressure (kbar)
1	(3,3,3)	-88
2	(4,4,4)	-43
3	(5,5,5)	-71
4	(6,6,6)	-63
5	(7,7,7)	-73

We "again" see convergence at (5,5,5). Now again keeping the cutoff charge density 12 times the cutoff for the amount of wavefunctions and varying the charge density:

	ecutwfc	Hydrostatic Pressure (kbar)
1	14	-11640
2	24	-1350
3	34	-770
4	44	-355
5	54	-68
6	64	-71

I.e we again see convergence at ecutwfc=54. Let's now search the multiplication factor:

	ecutwfc	Hydrostatic Pressure (kbar)
1	14	-11640
2	24	-1350
3	34	-770
4	44	-355
5	54	-68
6	64	-71

We thus see convergence already at a factor of 5, so we'll take the highest factor of both, i.e 8. Our final values are thus ecutwfc=54, ecutrho at 432 and a k-mesh of (5,5,5).

III. Energies of the end points

We'll do a full geometry optimization by first using the calculation="vc-relax" control parameter and bfgs cell and ion dynamics with 0 pressure (0.5 kBar convergence threshold) for both crystals.
We also did a manual optimization, as to also obtain an equation of state, from which a value for the bulk

[1] cif file by Farquhar M. C. M., Lipson H. and Weill A. R., with COD index 9015110

	factor	hydropressure
1	4	-12
2	5	-61
3	6	-50
4	7	-63
5	8	-68

modulus is derived. To do this, static calculations at 5 slightly different lattice parameters were done, after which a vc-relax calculation with target pressure equal to the weighted average of each of the previously obtained stress tensors was performed. This data was used then used to fit a Birch-Murnaghan equation of state.

i. bcc-Fe

Using the first method, for bcc-Fe we get a final total energy of -329.26 Ry for a unit cell containing 1 atom with a unit cell volume of 76.1642 a.u.^{3} $\implies$ cell length of 4.238871 au, or a length of 0.492942382 alat with a 2.999754 scale.
Using the second method a unit cell volume of 76.52 a.u.^{3}, was found, with lattice parameter 5.35 a.u. = 2.84 Å. This corresponds again with an energy of -329.26 Ry. Using the Birch-Murnaghan fit a value for the bulk modulus of 176.0 GPa was obtained (value on materialsproject.org is 182 GPa).

ii. DO3-Fe3Si

For Fe_3Si we get a final total energy of -999.30084673 Ry for a unit cell containing 4 atoms with a volume of 296.49363 a.u.^{3}

Using the second method a unit cell volume of 301.56 a.u.^{3} was found. This corresponds with an energy of -999.30170 Ry. Using the Birch-Murnaghan fit a value of 190.3 GPa was found for the bulk modulus (value on materialsproject.org is 212 GPa).

IV. Sanity Check

We'll take the DO3-Fe_3Si and change 1 Silicon atom in the unit cell to an Fe atom, for this we'll first have to remove the symmetry and define all the atom's coordinates ourselves. The DO3-Fe_3Si crystal has F m -3 m as symmetry space group (number 225). Looking at the crystallographic database we see that the first Fe atom to consider which has relative atomic

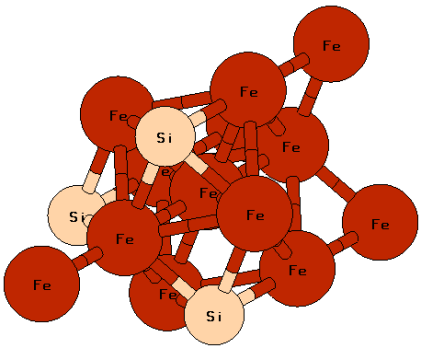


Figure 1: *DO3-Fe_3Si with 1 Silicon changed to Iron*

position (0.5 0.5 0.5) corresponding to Wyckoff Position 4b, the second one has Wyckoff Position 8c and the Si atom has 4a. I.e our unit cell has 4+8+4 = 16 atoms which is correct.
Making a cif file without symmetry can then be done quite straightforwardly. We then check if the energy we get is indeed the same. The energy we get from our Fe_3Si crystal without symmetry is -3997.2 Ry per 16 atom unit cell or -999.3 Ry per 4 which is the same as we found earlier, this is thus a good input file to modify, we'll now replace one Si atom with Iron as shown in figure 1. Doing this gives us a 18.75 at.% $\equiv$ 10.40 wt.% Si crystal. After a 'vc-relax' calculation we get an energy of about -4314.944 Ry, this crystal can be composed of 3 Fe_3Si (containing 4 atoms) and 4 Fe (containing 1 atom) unit cells giving a sum of energies of:

$$3Fe_3Si + 4Fe = -(3 * 999.3 + 4 * 329.26) = 4314.94 \quad (1)$$

So almost exactly the same energy found by doing the simulation. It won't decompose as it's energy is just a little lower than the weighted sum of the two.

References

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