The Efficiency of Evolutionary Optimization of Magnetic Properties of Ternary Alloy

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Abstract. The area of magnetism offers a wealth of possibilities to study different algorithmic approaches. The reason is the diversity of different forms which can be used to describe magnetic samples. One can start from the most simple Ising-like system which, being of physical origin, became an inspiration for many models in a various areas of science and the formulation of which fits ideally the earliest representations of genetic algorithms. The recent problems concern elaborate molecular structures, like eg. spin glasses where the character of interaction between the spins of the same type may be different depending on some random variable or mixed alloys which can have different stoichiometry or structure and therefore present visibly distinct magnetic characteristics. In this paper we will deal with the mixed magnetic ternary alloy - an analog of prussian blue. Some results concerning the physical conclusions coming from applying the evolutionary computation to analyse this material have already been published and here the attention will be paid to the influence of parameter change on the efficiency of procedure.

1 Introduction

In a previous paper I described the possibilities of the evolutionary optimization approach to the problems of the physics of magnetism [4]. They also arise from the good match between the classical description of genetic representation and the formulation of some basic physical problems connected to magnetism at the atomic level (such as the Ising model) and from the calculational complexity of large magnetic systems. This paper is devoted to the study of the so called "ground state" of mixed (ternary) magnetic alloy with antiferromagnetic coupling between nearest neighbours. From the physical point of view configuration-based studies of magnetic phenomena using the evolutionary approach may be divided into two groups. The phrase "ground state", used earlier in this paragraph means the search of minimum energy state of the system neglecting thermal effects. For such calculations the main computational effort is spent on overcoming difficulties resulting from the complicated structure of the material. One has to notice that even the most famous and most simple system (above mentioned Ising model) was only solved in 1991 [3], almost 30 years after EA was discovered. These calculations were made for the ± 1 system with the hamiltonian

$$H = -J \sum_{ij} S_i S_j \tag{1}$$

and the ferromagnetic exchange constant $J > 0$ which lead to purely parallel ordering of all spins.

The second group of magnetism related evolutionary calculations concerns the problem of thermodynamic minimization. The questions mainly concerning the way of entropy calculation are addressed. In order to deal with such problems in their analytical form one needs to consider set nonlinear equations. which are, especially for higher order of entropy approximation, impossible to solve. From the point of view of EA the inclusion of temperature leads to increase of complexity due to necessity of performing the local spin configurations calculations.

In this paper we deal with the search for the ground state of a ternary alloy being an analog of prussian blue. Materials belonging to this group are built in characteristic way with two distinctly distinguishable sublattices interacting antiferromagnetically one with another and consisting of spins of different kinds. The complications with optimization of such a system has been mentioned earlier [4] and here I want to present some more detailed analysis of the dependence of the results on the different evolutionary parameters. Finally I am going to present remarks concerning the optimal set of parameters for the studied problem.

2 Model

The structure of ternary magnetic prussian blue analog is presented in Fig.1.

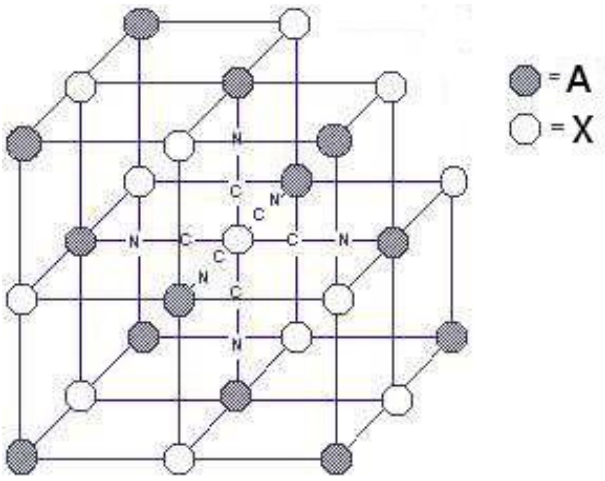

Figure 1. The structure of typical Prussian blue analog. Two interpenetrating fcc lattices A nad X are presented as grey and white circles respectively. The middle atom is shown with surrounding cyanide bonds and the upper right corner is intentionally empty due to possible lack of A type atom.

The grey and white circles correspond to the positions occupied by the spins belonging to the two sublattices, denoted A and X respectively. Both of them separately form the fcc lattice so their superposition together with relative translation along the $[a/2, 0, 0]$ vector (a is the lattice parameter) leads to the structure where all the simple cubic posi-

tions are filled. Both A and X structures are not homogenous but must be described by factors defining their stoichiometry. In the A sublattice there exist vacancies, an example of which can be seen in the upper right corner of Fig.1. The number of occupied positions is defined by the relative factor N_A/N_X and for these calculations is assumed to be equal 2/3. In the X structure all positions are occupied but they can be filled with two types of spins, say B and C. The ratio of their appearance is described by the number p which refers to the relative number of B spins. Thus generally we can present the studied structure as AB_pC_{1-p}, corresponding to the real prussian blue formula $K[Fe^{3+}Fe^{2+}(CN)_6]$. The A sublattice is formed by the alkali metal ions, and the X one consists of iron atoms, however the physics of phenomenon requires that they interact in a different way with potassium. Certainly a similar mechanism may be observed for other structures such as high temperature stable $K[V^{2+}Cr^{3+}(CN)_6]$. The assumption $p = 0$ is, as well as that about the value N_A, related exactly to the prussian blue.

The value which undergoes minimization is the total magnetic energy of system.

$$U = -\sum_{ij} J_{ij} S_i S_j - D \sum_i S_i^2, \tag{2}$$

where sumation is taken only over nearest neighbours and the second term, called single-ion anisotropy prefers either larger or smaller spin modules dependending on the D constant sign. The exchange constants J_{ij} determines the character of interaction between nearest neighbours. The need to exact enumeration follows the existence of two possible spins in X sublattice. Usually $J_{AB} > 0$ so this is the ferromagnetic interaction and $J_{AC} < 0$ so is the antiferromagnetic one. The phase diagrams are presented in the relative coordinates:

$$R = \frac{|J_{AC}|}{J_{AB}}, \qquad\qquad d = \frac{D}{J_{AB}}. \tag{3}$$

The values of modules are equal $S^A = \frac{3}{2}$,$S^B = 1$,$S^C = \frac{5}{2}$, and the possible z-components which are used in energy calculations belong to the sets , $S^A = [-\frac{3}{2}, -\frac{1}{2}, \frac{1}{2}, \frac{3}{2}]$, $S^B = [-1, 0, 1]$, $S^C = [-\frac{5}{2}, -\frac{3}{2}, -\frac{1}{2}, \frac{1}{2}, \frac{3}{2}, \frac{5}{2},]$. The size of sample used in optimization is the result of some system requirements, ie. in order to apply periodic boundary conditions it must be even and, due to N_A/N_X ratio, must be divisible by three. We selected the smallest acceptable number and in calculations the cubic sample with edge length 6 was used. This value together with listed above possible spin projections allows us to estimate the complexity of the problem: with $N_X = 108$,$N_A = 72$, taking into account possible spin values and the number of combinations obtained from distributing of A spins over possible lattice sites, the size of configurational space is about 10^{150}. Even considering that much of this configurations may be obtained from others with rotations or translations the number still remains large.

In the calculations we used the "spin-oriented" approach to sample representation the partially mixed crossover and random mutation. This operators combination was described earlier [4] as the most effective.

The fitness function was selected in the same form as in cluster structure minimization:

$$f(U) = \exp\left(-\alpha \frac{U - U_{min}}{U_{max} - U_{min}}\right) \tag{4}$$

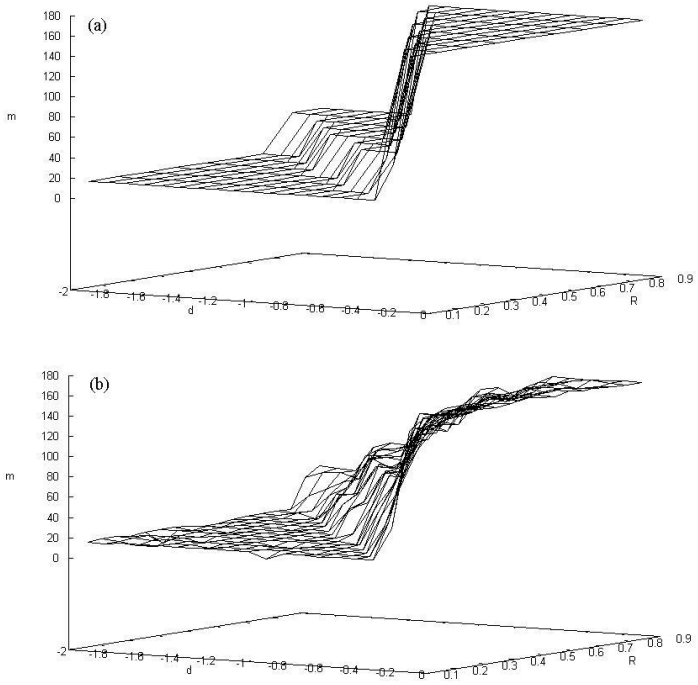

Figure 2. Phase diagram for ground state of magnetic ternary alloy: (a) analytical [1], (b) from evolutionary optimization.

which gives a good compromise between elitism and diversity of population [2]

3 Results and conclusions

The previous paper presented results of energy minimization for the algorithm parameters listed in table 1.

parameter	value
population size	50
crossing probability	0.5
mutation probability	0.01
number of generations	dependent on size of sample {2000,10000}

Table 1. Parameters of evolutionary optimization for multispin problem.

The results though promising were characterized by some inaccuracies. The reference data for our calculations were the results of analytical computations made by Bobak et al. [1]. The main difference is that analytical data were obtained with assumption about completely pure phase structure, which means that all spins of the same type have the same z-component. For magnetic samples of such complicated construction internal substructures or periodic forms may exist which may lead to lower energy states. The comparison of analytical and evolutionary minimized result is shown in Fig.2.

On the z axis there is presented magnetization of whole sample, calculated simply as the sum over all spins.

$$m = \sum_i S_i \tag{5}$$

Some questions arise when looking at the plot presented. The main physical effect is that there are some intermediate states between 'pure' phases. They may be observed as small plateau areas. On the other hand the roughness of obtained surface causes doubt about the correctness of data presented. While it is certain that the majority of energy values are smaller than obtained by Bobak some of them are higher, so one has to say that not all optimization runs lead to the best result. This was why we tried to perform some statistical analysis on the influence of different evolutionary operators on the obtained results.

For the tests the well defined point $R = 0.3, d = -1.0$ was selected for which the $[1/2, -1/2]$ phase dominates and this result is confirmed in our calculations. This point is however relatively close to the phase boundary (appr. $R = 0.41, d = -1.0$) where the phase $[3/2, -3/2]$ starts to dominate and we expected that our procedure could be decieved by some low energy structures. For this point the total energy in relative units (with $J_{AB} = 1$) is equal to 12.6 and the magnetization to 18. Because we are only interested in the best point we are going to determine the efficiency of procedure by calculating the percentage of runs leading to this best result.

The effects are presented in Fig.3 where there are shown the data averaged over all runs with the same parameters and grouped according to the optimization time measured in the number of generations. The number of runs used in averaging was different for every point, but is always between 600 for the longer runs and 2000 for shorter runs. Table 2 contains the values of correlation coefficients calculated for these points according to the well known formula:

$$\rho = \frac{\langle xy \rangle - \langle x \rangle \langle y \rangle}{\sigma_x \sigma_y} \tag{6}$$

It may be observed that mutation no influence on the optimization effects. The deviations from the horizontal line, parallel to the x-axis is here only statistical. More interesting is the dependence on the crossing probability For the lower value (0.25) the efficiency is also significantly lower and increases for greater values but the analysis of data for $p_{cross} = 0.5$ shows that it isn't the decisive factor as the points are spread over a large range. However a small positive correlation is observed. The population size turns out to be the most important factor. There were taken into account different numbers of personals from μEA (popsize=5) up to 50 and the tendency is clearly visible. It may be interesting however that correlation weakens with increasing number of generations.

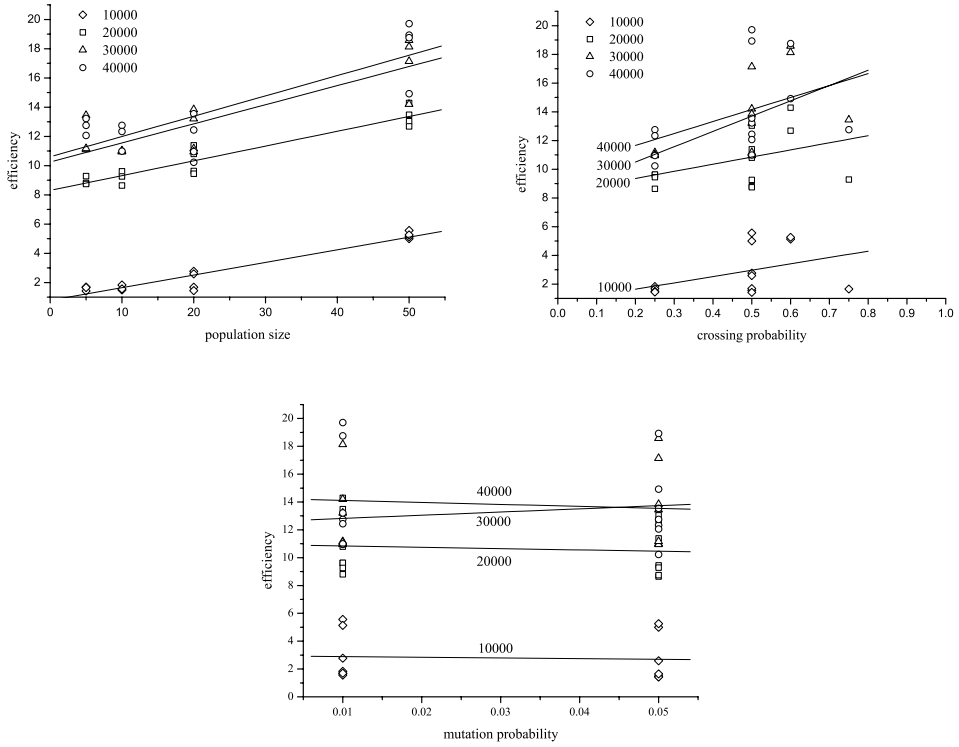

Figure 3. The trend and correlation between the efficiency and evolutionary process parameters for different number of generations.

The question may arise whether the selection of parameters was sufficient to draw such general conclusions. In this calculations we used 4 different values for population size, 4 for crossing probability and 2 for mutation probability. The analysis of all possible combinations gives the total number of 32, 14 of which were selected. The choice was performed in a way to cover approximately uniformly the parameter space, ie. calculations were made also for $p_{cross} = 0.5, p_{mute} = 0.05, popsize = 50$ as $p_{cross} = 0.5, p_{mute} = 0.01, popsize = 5$ or $p_{cross} = 0.25, p_{mute} = 0.5, popsize = 20$.

In the Fig.4 there are shown averaged (with standard deviations), minimum and maximum efficiencies in the function of number of generations. This dependence starts to saturate for number exceeding about 30000. The comparison of deviation bars and the positions of squares showing min and max value leads to the conclusion that there are grouped points below the averaged value and single points lying above it. This curve shows also that the values used earlier (see tab.1) were incorrect from the point of view of efficiency.

There is also a question concerning the equality of efficiency over the whole phase diagram. Although the (R,d) point was selected close to the phase boundary, as mentioned,

number of generations	10000	20000	30000	40000
population size	0.962	0.951	0.858	0.826
crossing probability	0.412	0.391	0.586	0.415
mutation probability	-0.061	-0.100	0.169	-0.097

Table 2. The correlation coefficient values between the efficiency and the parameter pointed in the left column.

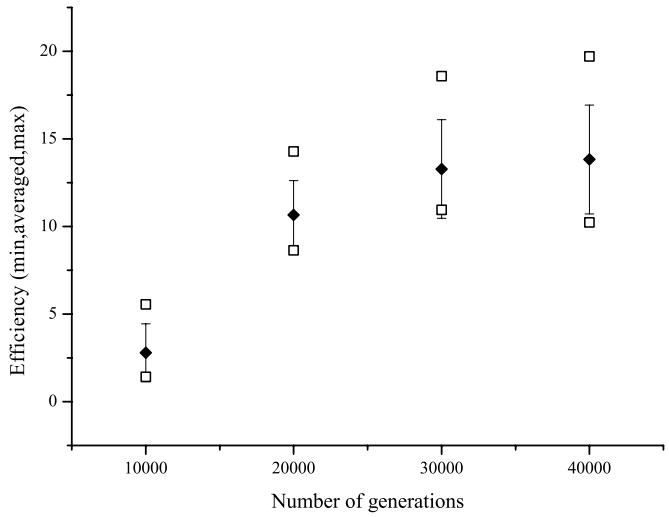

Figure 4. Efficiency averaged over all parameters set for the same generations number.

the suspicion still exist that for the other regions, especially those near the $[3/2, -3/2]$ to $[3/2, -5/2]$ phase transition it may be significantly lower.

In this paper I presented some efficiency estimations for the computationally hard problem of energy minimization for mixed molecular magnetic material. The results show that for this type of process the crucial factor is the size of population which allows a greater number of configurations to survive and increases the diversity of population.

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