Quantum Mechanical Simulation of Nanocomposite Magnets on CRAY XT3

Yang Wang, *Pittsburgh Supercomputing Center*, **G.M. Stocks**, **Aurelian Rusanu**, **D.M.C. Nicholson**, and **Markus Eisenbach** *Oak Ridge National Laboratory*

ABSTRACT: *One of the grand challenges in the development of nanotechnology is how to synthesize and process "designed nano-structured" materials. An important example of these materials is the nanocomposite permanent magnets made of hard and soft phased magnetic nanoparticles located on a superlattice. Binary phase magnets, if they can be devised, could double the energy product of current single-phase magnets. However, such magnetic nanostructures present substantial theoretical challenges due to the need to treat the electronic interactions quantum mechanically whilst dealing with a large number of atoms. In this presentation, we demonstrate our capability of performing the quantum mechanical simulation of nanocomposite magnets using a CRAY XT3 supercomputing system and the Locally Self-consistent Multiple Scattering (LSMS) method, a linear scaling* ab initio *method capable of treating tens of thousands of atoms. The simulation is intended to study the physical properties of magnetic nanocomposites made of FePt nanoparticle and FePt random alloy. We will demonstrate the scalability of the LSMS method on Cray XT3, and will discuss our results on the electronic and magnetic structure of the nanoparticle and the effects of the surrounding metallic random alloy.*

KEYWORDS: teraflop, order-*N*, LSMS, *ab initio*, nanoparticle, CRAY XT3

1. Introduction

Magnetic nanoparticles and their composites are of great interests to us because of their potential applications in data storage industry. Over the past few years, we have witnessed tremendous increase in the data storage density of magnetic disk drives, an increase at a rate of greater than 100% annually achieved by reducing the critical physical dimensions. However experiments and theory indicate that there is a so-called superparamagnetic limit to the size reduction that will soon be reached within just few years. In conventional magnetic recording devices, the recording medium, in the form of granular film, consists of weakly coupled magnetic grains, with grain sizes in the range of 10-15 nm. The signal to noise ratio needed for high density recording is achieved by statistically averaging over about 100 grains per bit. The thermal stability of the grain magnetization has to be maintained in order to store information reliably for 10 years, a typical design criterion used by industry. Increasing the areal density requires a reduction of the grain volume and a tighter grain size distribution. However, decreasing the grain volume, to obtain higher storage densities, results in the moment orientation of individual grains becoming increasingly unstable due to thermal fluctuation. And once the areal density passes the superparamagnetic limit, the thermal instability will give rise to noise, false sensor response, and long-term data loss.

Today's magnetic storage media has reached 100 Gbits/inch^2 level, and the next milestone will be 1 Tbits/inch^2. It is clear that reaching and going beyond this milestone requires major scientific and technological breakthroughs. One possibility for such breakthroughs relies on a radical new design, in which magnetic nanoparticle composite forms a patterned media and each bit (0 or 1) is stored in a single magnetic nanoparticle. Of critical importance for such magnetic storage design is the length of time for which magnetic orientation of individual nanoparticles, once written, is stable at operating (room) temperature. Thus understanding the relationship between materials properties and magnetic stability of magnetic nanocomposite is of crucial importance.

Clearly, the ability to design materials at the nanoscale holds significant future scientific and technological opportunities. However, realizing the ultimate potential of nano-structured materials requires an understanding of the quantum mechanical behavior of the materials at the atomic scale which in fact underpins the new structures and phenomena unique to nanostructures.

Over the last 2-3 decades there has been significant progress in our ability to calculate the properties of materials at the quantum level. These advances have largely been based on the local density approximation (Hedin and Lundqvist, 1971; von Barth and Hedin, 1972) or generalized gradient approximation (Perdew and Wang, 1986; Perdew *et al*. 1996) to density functional theory (Hohenberg and Kohn, 1964; Kohn and Sham, 1965). However, the nano-structured materials place new demands on these *ab-initio* methods because of the large numbers of atoms that are present in even the simplest of nanostructures, an example of which is a 5nm cube of iron that contains the order of 12,000 atoms. Fortunately, recent advances in the locally self-consistent multiple scattering (LSMS) method (Wang *et al*, 1995), an *ab-initio* order-N scaling technique specifically implemented to exploit massively parallel computing, are making the direct quantum simulation of nano-structured materials possible. Previously (Wang *et al*, 2005), we have reported our preliminary results on the calculation of the electronic and magnetic structure of an iron nanoparticle embedded in iron-aluminide crystal matrix, which involves 16,000 iron and aluminum atoms in total. This simulation was our first step toward the direct quantum mechanical simulation of the physical properties of a realistic model of nano-structured materials.

The main objective of this paper is to show our recent study of the electronic and magnetic properties of FePt nanocomposites on CRAY XT3. These are the magnetic nano-structures that have been considered as a potential candidate for the next generation high density magnetic data storage media. This paper is organized as follows. The next section is a brief description of the LSMS method. We show the parallel performance and linear scaling properties of the method on CRAY XT3. The application of LSMS method to the electronic and magnetic structure of FePt nanocomposites is demonstrated in section 3, followed by the conclusion section.

2. The LSMS method and Its Performance on CRAY XT3

The LSMS method is a multiple scattering theory (Korringa, 1947; Kohn and Rostoker, 1954) based approach to the *ab-initio* electronic structure calculation. In the framework of multiple scattering theory, the Green function for the Kohn-Sham one-electron Schrödinger equation is readily given in such a convenient algebraic expression (Faulkner and Stocks, 1980) that its numerical calculation is clearly feasible; particularly when advantage is taken of the analytic properties of the Green's function. An advantage for using the Green function is that the calculation of the crystal wave functions becomes unnecessary, and, as a result, there is no need for the time-consuming orthogonalization and normalization procedure, which is needed in other *ab-initio* methods. Another advantage is that the only global operation required for obtaining the Green function is the calculation of a multiple scattering matrix for each atom which accounts for the major portion of the floating point operations of the entire electronic structure calculation.

In general, given the Green function, the electron density and the magnetic moment density in the vicinity of the i^{th} atom can be conveniently obtained as follows:

$$\begin{aligned} \rho_i(\boldsymbol{r}) &= -\frac{1}{\pi}\,\mathrm{Im}\,Tr\int_{-\infty}^{\varepsilon_F} d\varepsilon\,\underline{G}_i(\boldsymbol{r},\boldsymbol{r};\varepsilon), \\ \boldsymbol{m}_i(\boldsymbol{r}) &= -\frac{1}{\pi}\,\mathrm{Im}\,Tr\int_{-\infty}^{\varepsilon_F} d\varepsilon\left[\underline{G}_i(\boldsymbol{r},\boldsymbol{r};\varepsilon)\cdot\underline{\boldsymbol{\sigma}}\right], \end{aligned} \tag{1}$$

where the Green function $\underline{G}_i(\boldsymbol{r},\boldsymbol{r};\varepsilon)$ is a 2 × 2 matrix in the spinor space and is calculated in the vicinity of atom *i*. The *x*, *y*, and *z* components of vector $\underline{\boldsymbol{\sigma}}$ are the corresponding Pauli matrix, and ε_F is the Fermi energy. The energy integration usually takes place along an energy contour in the upper half complex plane to avoid the singularities of the Green function on the real energy axis. In non-magnetic cases, the Green function matrix is reduced to a scalar function, and the magnetic moment density is simply zero. For ferromagnetic states, the Green function matrix is diagonal in the frame of references that the *z*-axis is along the magnetization direction, and the computational procedure can be much simplified by carrying out a spin-polarized calculation, for which the Green function for spin up and spin down states is decoupled and is calculated separately. For non-collinear magnetic states, the Green function matrix is non-diagonal and its calculation, also known as spin-canted calculation, usually takes four times longer than the spin-polarized calculation and eight times longer than a non-spin-polarized one.

The calculation of the multiple scattering matrix itself scales as N^3, where N is the number of atoms in the unit cell. The crux to making multiple scattering theory order-*N* scaling is an approximation that neglects multiple scattering processes around an atom if they involve atoms from a distance greater than some cut-off radius (R_{LIZ}). The space within R_{LIZ} is called local interaction zone (LIZ). The idea behind this approximation is based on the observation that the scattering processes involving far away atoms influence the local electronic states less and less as the distance form the scatter under study is increased – an example of nearsightedness (Kohn, 1996). Technically, this approximation is implemented as follows: we draw a sphere (or LIZ), with a predefined radius, around atom *i*, we calculate the multiple scattering matrix only for the multiple scattering processes involving the atoms enclosed by the sphere, and we calculate the Green function and use equation (1) to compute the electron and magnetic moment density in the vicinity of the atom. This procedure is repeated for each atom in the unit cell. Obviously, the LSMS method is implicitly parallel over the number of the atoms which makes it an ideal method for the electronic structure calculation for nano-structured materials.

The LSMS method was originally implemented based on one (atom) to one (processor) mapping scheme, which was necessary in the early days when the computing power and memory size was much limited. The code has essentially been rewritten entirely to take advantage of the modularization feature of FORTRAN 90/95 and one-sided communication feature of MPI-2 and, most importantly, to allow flexible association between atoms and processors such that multiple atoms can be mapped onto one processor. The latest measurement of its order-*N* property on Cray-XT3 is shown in figure 1, in which the number of float operations is plotted against the problem size, i.e., the number of atoms involved in the simulation. Pushing the LIZ size to an extreme (including 136 neighboring atoms), where the entire calculation is dominated by the BLAS routine ZGEMM because of large size of the large multiple scattering matrix (3472×3472 double complex matrix), we are able to run LSMS at 80% of the peak performance of the system. The results of strong scaling measurement of LSMS code on a Cray XT3 are shown in figure 2. In this plot, the elapsed time for a fixed size problem, the electronic structure calculation for a magnetic Fe nanoparticle embedded in FeAl matrix with 16,000 atoms altogether, is measured against the number of nodes employed. The ideal strong scaling curve (solid line) representing 100% parallel efficiency is also shown. Departure from the ideal strong scaling at the large number of CPUs usually indicates the existence of the serial components, i.e., tasks that can not be parallelized, and such other factors as the latency for communication, the time spent in performing I/O, etc.

3. The Electronic Structure Calculation for Magnetic Nanocomposites

Here we report our recent studies of electronic and magnetic properties of FePt nanocomposites using LSMS method. The nanocomposite is made of a FePt nanoparticle embedded in $Fe_{0.5}Pt_{0.5}$ random alloy in a face-centered tetragonal (fct) lattice structure. The FePt nanoparticle itself is an ordered compound in $L1_0$ lattice structure, and has the same lattice constants, $a_0 = b_0 = 3.8525$ Å and $c_0 = 3.7133$ Å, as the $Fe_{0.5}Pt_{0.5}$ random alloy. We considered spherical FePt nanoparticles of three different dimensional sizes, 2.5 nm, 3.86 nm, and 5.0 nm in diameter. Each of these nanoparticles contains 711, 2,195, and 4,777 Fe and Pt atoms, respectively. The entire unit cell sample (the nanoparticle plus the random alloy) for each case contains 14,400 Fe and Pt atoms in total.

We performed spin-polarized LSMS calculation for the unit cell samples described above using 1,200 nodes on CRAY XT3, with 12 atoms per node. We simplified the calculation by making the muffin-tin approximation to the potential in which the one-electron potential is assumed to be spherically symmetric within the muffin-tin sphere around each atom and is assumed to be constant within the interstitial region between the muffin-tin spheres. This is a reasonable approximation for most simple and transition metals and intermetallic alloys due to the nature of metallic bonding. In our calculations, we choose the LIZ for each atom to include 54 neighboring atoms which has be proved to be sufficient for close-packed structures.

The calculated results for the electronic and magnetic structure turn out to be very much similar for the three different nanoparticle sizes. We therefore only show the calculated results for the case that the nanoparticle is 3.86 nm in diameter. Figure 3 is a sliced view of the magnetic moment distribution within the nanoparticle and the random alloy. The amount of magnetic moment on each atom is coded in the following color scheme: The balls in silver color are the Pt atoms and the balls in red color are the Fe atoms. The darker color means less magnetic moment value. Obviously, in the interior region, the magnetic moment on Fe and Pt atoms has a rather uniform distribution pattern, that is all the Fe atoms have the same moment and all the Pt atoms also have the same moment. The inhomogeniety effect from the external random alloy has essentially been screened out by the surface layers, the shield region, of the nanoparticle.

More detailed pictures of the electronic and magnetic structure of the nanocomposite are shown in figures 4 and 5, where 14,400 data points, each of which corresponds to the number of excess electrons (figure 4) or the magnetic moment (figure 5) on each atom, are plotted against the atom distance measured from the center of the nanoparticle. Not surprisingly, we find that atoms in the interior region of the nanoparticle have essentially the same amount of net charge and magnetic moment as in the bulk of $L1_0$-FePt crystal. Note that Fe atoms are gaining electrons from the Pt atoms and therefore are negatively charged, while Pt atoms losing electrons are positively charged. For those atoms inside the shield region, the shaded area in the figures in figures 4 and 5, the net charge and the magnetic moment show rather dramatic fluctuations, induced by the influence from the random alloy. This shield region extends as deep as 4 Å inward from the nanoparticle surface, and this depth seems independent of the nanoparticle size. In the random alloy region, as expected, the distribution of the net charge and the magnetic moment is very much random. However, most Fe atoms are negatively charged and carry a magnetic moment larger than 2.6μ_B, while most Pt atoms are positively charged and carry a small magnetic moment, less than 0.4μ_B.

Knowing the charge and moment distribution within a nanoparticle helps to understand the size effect as well as the effect of the surrounding environment. It also helps to determine the electrostatic interaction and the exchange coupling between nanoparticles. A multi-scale model for complex nanocomposites or nano-structured devices in which each nanoparticle is treated as a point particle with proper charge and multipole moment, determined from the quantum mechanical calculation, can thus be built.

4. Conclusion

In summary, we have performed direct quantum mechanical simulation of magnetic FePt nanoparticle composites on CRAY XT3. The results reveal that there exists a shield region, about 4 Å in depth, below the surface of the nanoparticle. This region, whose size is found independent of the nanoparticle dimension, plays an essential role as an electromagnetic shield that screens the effect of the random alloys from influencing the interior region of the FePt nanoparticle. As a result, the electronic and magnetic structure of the interior region shows no difference from the FePt crystal.

The *ab initio* electronic and magnetic structure calculations employing LSMS method and teraflop high performance supercomputers are clearly possible for the large unit cells necessary for modeling interfaces, surfaces, defects, and especially, the nano-structured materials while allowing a rigorous treatment of their electronic and magnetic properties. Despite the successes with our LSMS code, it is clear that nanotechnology, especially new discoveries in nano-structured spintronic devices, pose challenges that go much beyond the capability we just demonstrated. To meet the challenges, we are required to develop computational techniques capable of characterizing the electronic and magnetic structures on sub-hundred nanometers length scales and over a wide range of time scales. Evidently, going from several nanometers to sub-hundred nanometers, an order of magnitude increase in length scale, increases the problem size from tens of thousands to millions of atoms, and consequently increases the computational cost by a factor of hundreds, even assuming the continued order-*N* scaling of our methods. In addition to the challenges in terms of the length scale requirement, we are also facing the challenges in terms of the time scale requirement which can range from picoseconds, for the simulation of the magnetic moment rotation, to tens of nanoseconds or more, for the simulation of nanoparticle movement. Clearly, the success of direct quantum mechanical simulation of nanostructured electronic devices relies on the birth of petaflop computing technology.

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About the Authors

Yang Wang is a senior scientific specialist at Pittsburgh Supercomputing Center. His research interests are mainly focused on computational materials science. He is one of the leading figures in the development of the LSMS method. He can be reached at 300 South Craig Street, Pittsburgh, PA 15213, USA, E-Mail: ywg@psc.edu. G.M. Stocks is Corporate Fellow and research scientist at Oak Ridge National Laboratory. Dr. Stocks can be reached at Metals and Ceramics Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA, E-mail: stocksgm@ornl.gov. A. Rusanu is a postdoctoral researcher at Oak Ridge National Laboratory, and he can be reached at Metals and Ceramics Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA, E-mail: rusanua@ornl.gov. D.M.C. Nicholson is Research Scientist at Oak Ridge National Laboratory. Dr. Nicholson can be reached at Metals and Ceramics Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA, E-mail: nicholsondm@ornl.gov. M. Eisenbach is a postdoctoral researcher at Oak Ridge National Laboratory, and he can be reached at Metals and Ceramics Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA, E-mail: eisenbachm@ornl.gov.

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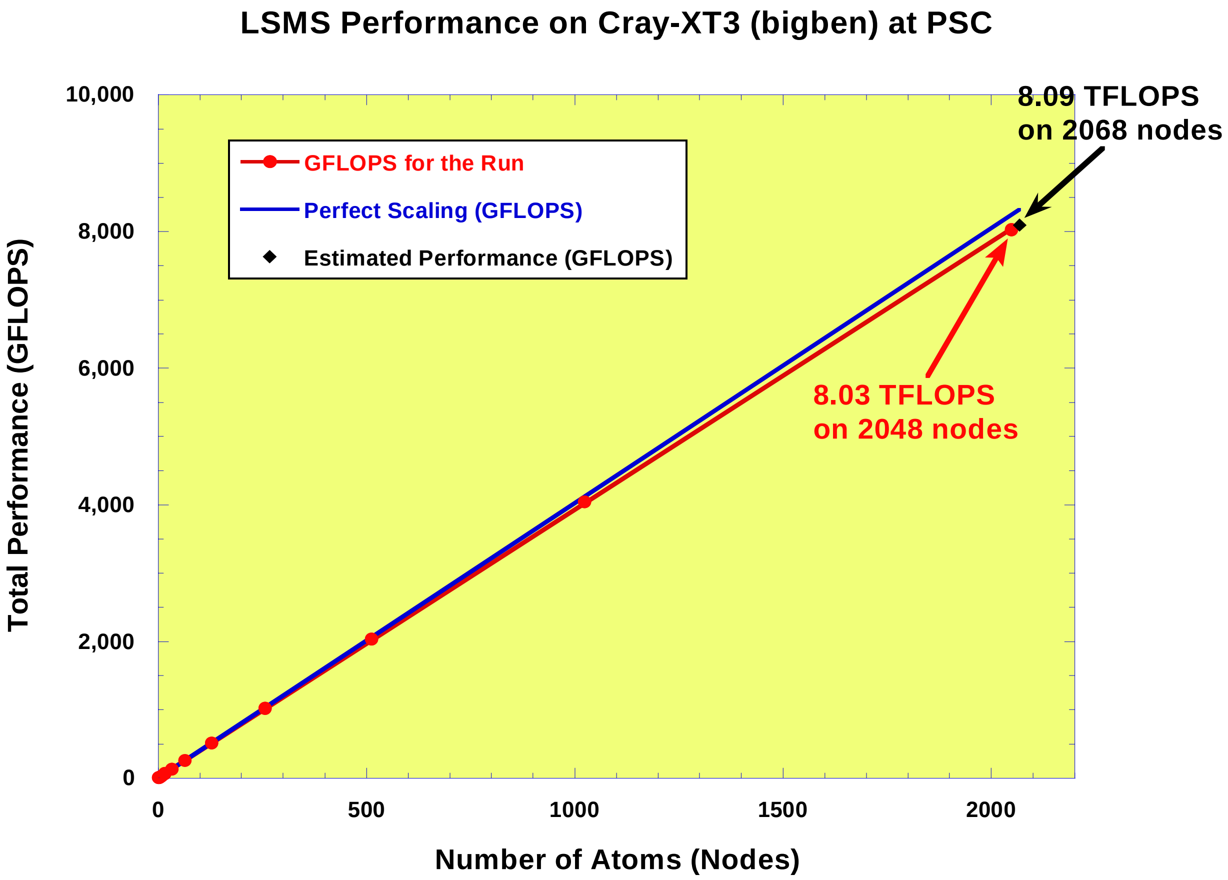

Figure 1. The order-*N* scaling of LSMS code on CRAY XT3. The performance test is a set of non-collinear spin-canted calculations for bcc Fe with 1, 2, 4, 8, 16, 32, 64, 128, 256, 512, 1024, and 2048 atoms, respectively. Each atom is mapped onto a node. The measurement is performed at Pittsburgh Supercomputing Center. With LIZ = 137 atoms, the code achieved 8.03 teraflops on 2048 nodes.

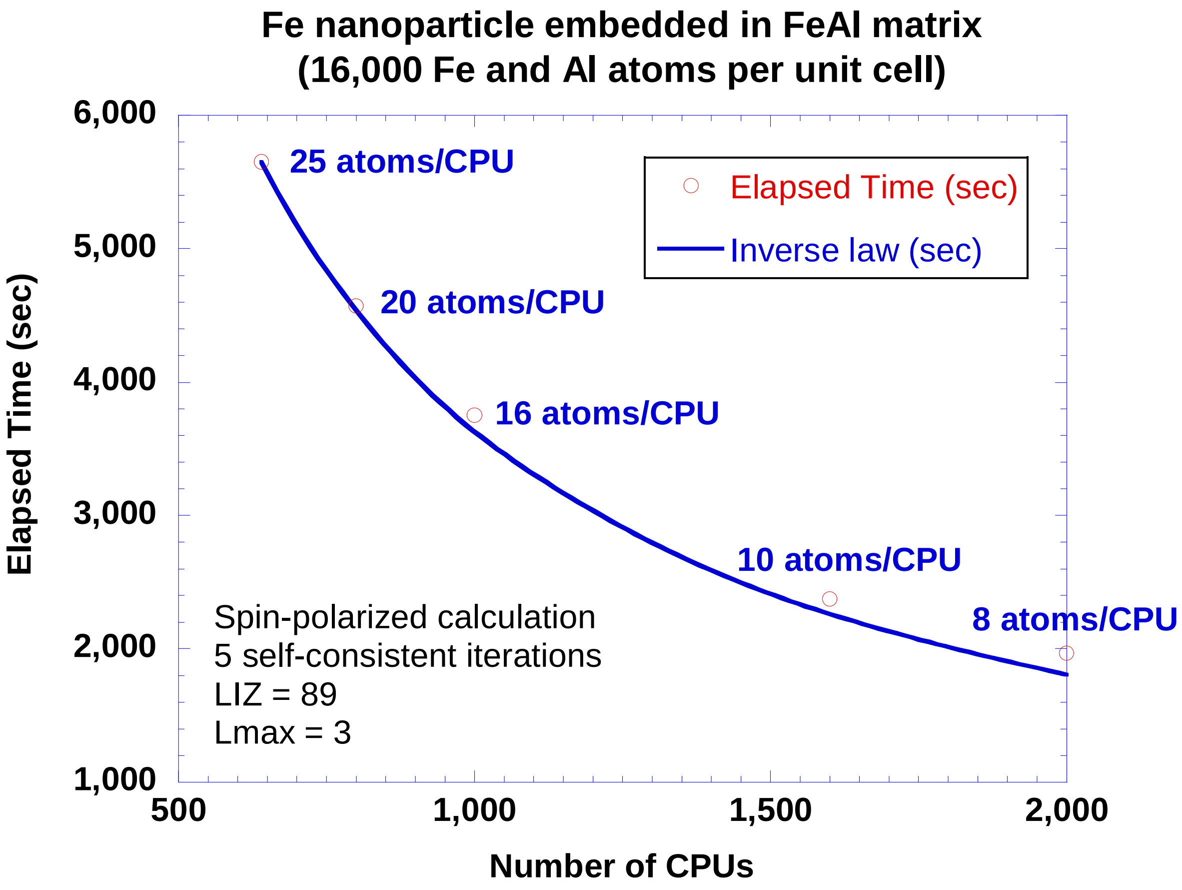

Figure 2. The strong scaling (or Amdahl scaling) of LSMS code measured on CRAY XT3. The same kind of electronic structure calculation (an Fe nanoparticle embedded in FeAl matrix with 16,000 Fe and Al atoms in the unit cell) is performed on 640, 800, 1000, 1600, and 2000 nodes, respectively. The solid curve is the ideal strong scaling curve, and the circles represent the actual elapsed time.

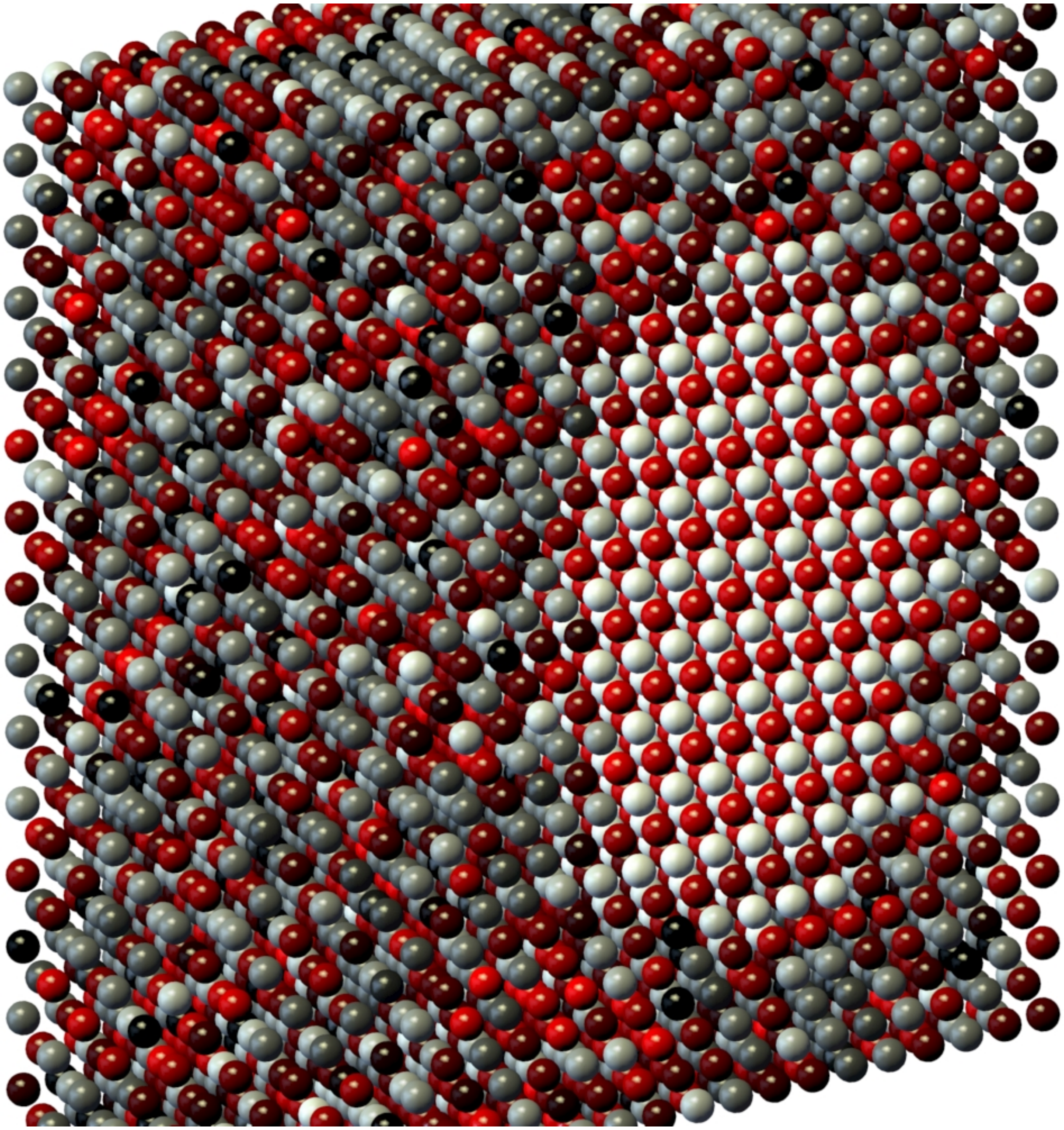

Figure 3. A sliced view of the magnetic FePt nanoparticle together with the surrounding FePt random alloy matrix. The magnetic moment distribution within the nanoparticle and its surrounding atoms is indicated by the color change from the center to the edge. The silver colored balls in the interior region of the nanoparticle represent the Pt atoms and the red balls the Fe atoms. The darker color is used for the atoms with less magnetic moment value.

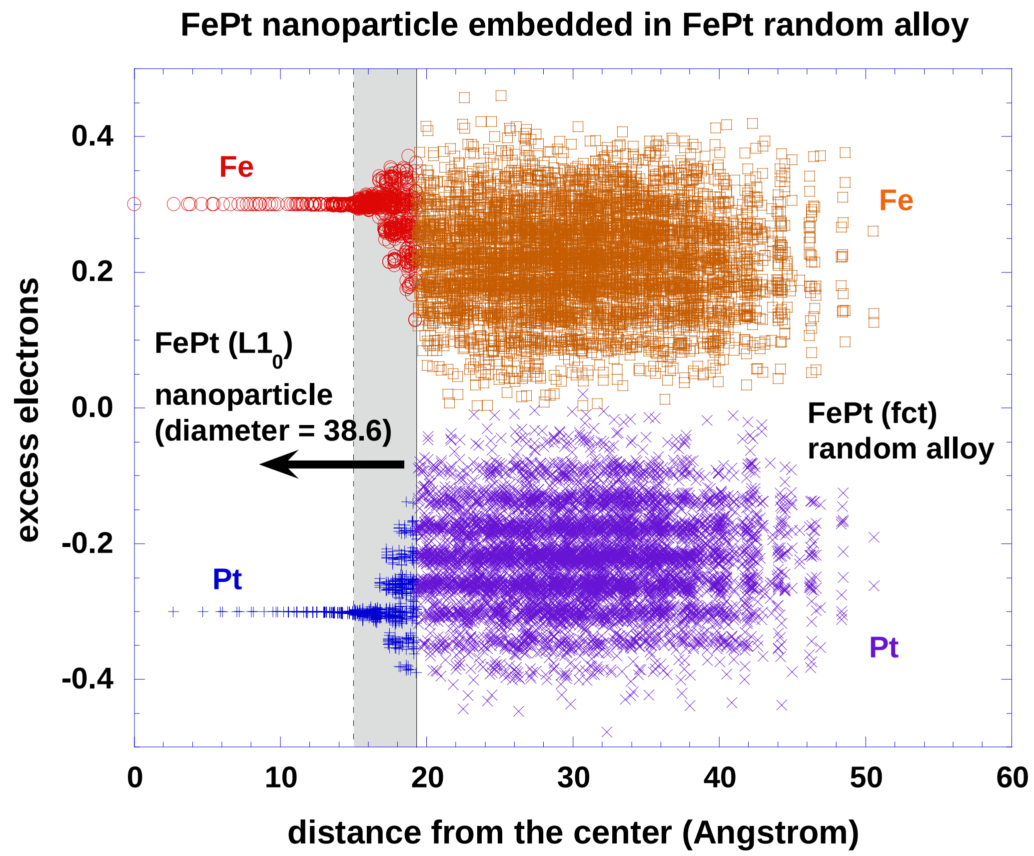

Figure 4. The number of excess electrons on each of the 14,400 atoms is plotted against the atom distance from the center of the nanoparticle. The red circles are the data point for Fe atoms in the FePt nanoparticle, the brown squares are for Fe atoms in the FePt random alloy, the blue crosses are for Pt atoms in the nanoparticle, and the purple crosses are for Pt atoms in the random alloy. The shaded area is the shield region of the nanoparticle which screens out the external random alloy effect from influencing the interior region of the nanoparticle.

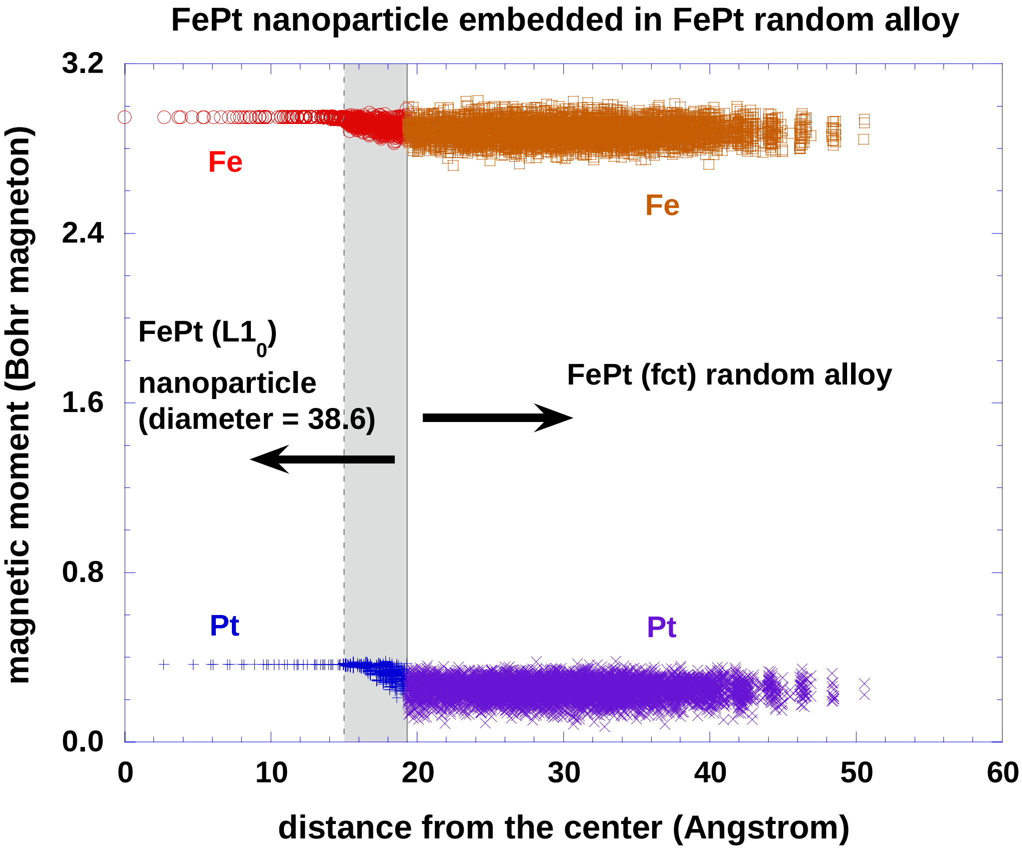

Figure 5. The magnetic moment on each of the 14,400 atoms is plotted against the atom distance from the center of the nanoparticle. The red circles are the data point for Fe atoms in the FePt nanoparticle, the brown squares are for Fe atoms in the FePt random alloy, the blue crosses are for Pt atoms in the nanoparticle, and the purple crosses are for Pt atoms in the random alloy. The shaded area is the shield region of the nanoparticle which screens out the external random alloy effect from influencing the interior region of the nanoparticle.