

Simulation of x-ray diffraction patterns using a massively parallel SIMD platform

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Abstract

A code for the simulation of X-ray diffraction pattern of a powder has been implemented on a massively parallel SIMD platform developed in the frame of the PQE2000 Project. The code allows the evaluation of the diffraction pattern of atomic-scale models of both perfectly ordered and disordered structures. The code has been used to investigate the structures resulting from the non-equilibrium alloying process of an immiscible metallic couple (Ag-Cu).

1. Introduction

X-ray analysis is the most widely used experimental technique in material science. It is a simple, not expensive, method which allows to have access to the most relevant information on the structure of a given material. A careful investigation of the X-ray diffraction (XRD) pattern can, in fact, reveal the coherence length of the structure (ultimately related to the grain size), the eventual presence of chemical order (presence of "superstructures"), the presence of residual internal stresses etc. All this information is essential for the understanding of the material properties (mechanical, electronic, thermodynamic). The XRD pattern, in a sense, provides a basic "fingerprint" of a material whose structure can be recognized and classified according to it.

The study of the XRD pattern is crucial in the cases

where a clearcut picture of the structure of a given material is not known *a priori* but can be just predicted on the basis of some working hypotheses. This is the case, for instance, when two (or more) elemental materials (type-A and type-B atoms, for instance) are forced to alloy (by some experimental means such as mechanical milling [5]) to form intermetallics or solid solutions (depending on the relative concentrations and on the thermodynamic phase diagram of the couple). In some cases, when the elemental couple has a positive enthalpy of mixing in the solid phase (being, thus, immiscible) the structure of the solid solution resulting from a mechanical alloying of that couple is still the object of a lively debate [3, 6, 2]. In such a case, the availability of a computational tool which can translate the working hypothesis (on the alloying mechanism and the kinetics) into the prediction of an expected XRD pattern, would allow the comparison between the experimental results and the XRD pattern predicted by the theory.

In the case of the alloying of immiscible couples, the occurrence of one of the following structures (each of them resulting from different theoretical predictions) should be assessed :

1. (a)-structure : a solid solution at the atomic scale (i.e. a crystalline structure containing a random distribution of type-A and type-B atoms);
2. (b)-structure : a solid solution of macroscopic units (grains, layers etc.) each of them composed of type-A or type-B atoms

The comparison of the XRD pattern generated by (a) or (b) structures with the available experimental data will be then performed.

The main purpose of this paper is, however, not to participate to the debate concerning the structure of solid solution of immiscible elements; it is just intended to provide a useful tool which might be helpful in discriminating the occurrence of different phases by comparing theoretical prediction to experimental data.

2. Computational layout and tools

The atomic-scale model of the structure is fully defined by specifying the set of atomic coordinates $\{r_i\}$ $i=1,N$ of the lattice sites (in the usual 3-dimensional space) and the set $\{\alpha_i\}$ $i=1,N$ with α =type-A or type-B atoms which defines the atomic species occupancy of the lattice sites. The $\{r_i\}$ and $\{\alpha_i\}$ sets entirely define the entity which we call "grain". The set of coordinates can be either the result of a geometric construction (e.g.the coordinates of a known crystalline lattice) or the outcome of a numerical simulation (Molecular Dynamics and Monte Carlo, MD and MC, respectively, hereafter) which allows to determine the structure which corresponds to the ground state of its hamiltonian. The availability of a technique which does not depend on the presence of particular lattice symmetries is important when one must deal with partially ordered (solid with defects, concentration or stoichiometric modulations) or totally disordered (amorphous, liquid) structures.

Given the structure of the "grain", the XRD intensity $I(2\theta)$ scattered by a "powder" (i.e. a collection of identical "grains" with different orientations in space) can be written as the average of the scattering intensities over all possible grain orientations and all possible angles of incidence of the X-radiation with the grain as follows [4]:

$$I(2\theta) = [1 + \cos^2(2\theta)] \cdot \left[\sum_{i=1}^N f(\theta, \alpha_i)^2 + \sum_{i,j=1}^N f(\theta, \alpha_i) f(\theta, \alpha_j) \frac{\sin [k(\theta) r_{ij}]}{k(\theta) r_{ij}} \right] \quad (1)$$

where $f_i(\theta, \alpha_i)$ are suitable scattering functions which depend on the atomic type present at site i , r_{ij} the distance between atoms i and j , $k(\theta)$ is the scattering vector of the incident radiation

$$k(\theta) = \frac{4\pi \sin \theta}{\lambda} \quad (2)$$

with λ the wavelenght of the incident x-ray radiation.

The mathematical problem in itself, as stated in eq.(1), is intrinsically parallel, as the intensity function at angle θ can be completely determined on the basis of the $\{r_i\}$ and $\{\alpha_i\}$ sets and the knowledge of the values of f_i at all the θ values.

The task can be expressed in a data parallel paradigm and, thus, suitably implemented on a SIMD machine. The parallelization strategy to be adopted is to allott a single θ calculation to a given processor of a MPP platform, whose local memory contains the $\{r_i\}$ and the $\{\alpha_i\}$ sets and the values of the functions $f(\theta, \alpha)$.

The described computational technique for XRD pattern simulation has been implemented on a hybrid machine, called PQE1, containing a MIMD and a SIMD part, as shown in fig.1. The main motivations for the assembly of this platform and the description of its relevant features will be the object of the following section.

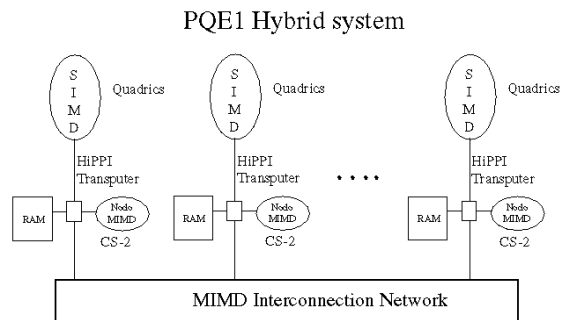


Figure 1. Schematic layout of the HW elements forming the PQE1 platform.

3. PQE1: architecture and computational properties

The PQE1 machine is a first outcome of the Project PQE2000 which groups the three main italian research institutions (CNR, ENEA and INFN) and an industrial partner (QSW, Finmeccanica group). The goals, the organization and other details on the PQE2000 Project can be accessed at the web site <http://www.enea.it/hpcn>. This web site also contains more information on SIMD and MIMD systems (peak performances, interconnections, routing mechanisms, communication bandwidth, MIMD-SIMD interconnection).

The HW structure of the PQE1 platform consists of three parts (see fig.1):

1. a MIMD platform (Meiko CS2) with 8 twin Hyper-Sparc nodes (100 MHz) interconnected via a proprietary fat-tree network;
2. seven SIMD platforms of the series [1] Quadrics/APE100 acting as CPU booster, each of them interconnected to a different MIMD node and having a different number of floating point units (fpu): QH4=512 fpu's, QH1=128 fpu's;
3. the connection between each MIMD node and its own SIMD platform is a two-ways system based on a Transputer and a Hippi channel.

From the SW point of view, the PQE1 platform is characterized by the presence of two different message passing libraries: (1) the usual PVM, MPI libraries for the MIMD nodes interconnection and (2) a message passing library (called QAPI), which allows communications to occur between the MIMD node and the SIMD platform.

The PQE1 architecture has a number of interesting properties:

1. Flexibility: it allows the most suitable architecture to be deployed for a given computational problem (SIMD architecture for data parallelism, MIMD-SMP for task parallelism, possibility of concurrent coarse-grained and fine-grained calculations etc.).
2. Computational power: the whole platform displays a theoretical computational power of ~ 100 GFlops, which is nearly 90 % based on the SIMD part. The HW configuration (i.e. the number of MIMD nodes and/or the size of the SIMD machine to be used) can be suitably selected on the basis of the type of computation to be performed.
3. Code portability: most of the current applications can be easily implemented on the machine. Only the parts of the code which should be ported on the SIMD part must be re-written in the data parallel language of Quadrics/APE100 (called TAO). A Fortran-like language (called QFOR) for the SIMD machine is under development in the frame of the PQE2000 Project.

Within the PQE2000 Project, several other SW products are going to be realized, aimed at a more efficient usability of the machine. They are, essentially:

1. A software-based Distributed Virtual Shared Memory (DVSM), which allows to build up common memory pages among MIMD and SIMD processors, based on a MPI programming of the routing processors of the MIMD machine (Elan, Elite, QSW proprietary).
2. A structured parallel programming environment (called SkIE [8]) based on a skeleton-template approach.

4. Technical layout and computational tasks

In this section, we will focus on the technical features which must be deployed for building up a computational code which allows concurrent calculations between the MIMD and the SIMD parts of the architecture. Although, for this particular application, the computational effort is mostly performed by the SIMD part, in a more general framework the different parts of this architecture could be suitably used for concurrent calculations. The code's sections, which will be reported in the following, are those where calls are performed from the main Fortran code, to the QAPI library to make the two parts of the architecture interacting to each other.

As it has been previously stated, the $\{r_i\}$ point distribution can be either the result of a geometric construction or the outcome of a MD (or MC) simulation. In the latter case, the MD (or MC) code can be made running on the conventional MIMD part. With a given frequency, the MIMD part provides the grain description to the SIMD part, which will be charged to evaluate the XRD pattern. If one adopts this computational scheme, at this point, the code performs the following tasks:

1. The function $f(\theta, \alpha)$ is built up for all the θ values by the SIMD machine.
2. The SIMD system is initialized and the SIMD code (called `name`) is loaded into the Quadrics/APE100 machine as follows:

```
if (AP_Open().eq.QOK) then
  print*, 'Quadrics opened'
else
  print*, 'Quadrics is not opened'
end if
```

```
if (AP_Load(name).eq.QOK) then
  print*, '.zex program loaded'
else
```

```

    print*,'.zex program is not loaded'
end if

```

3. The main code dispatches the $\{r_i\}$ and the $\{\alpha_i\}$ sets to all the SIMD nodes (broadcast). This operation is performed with the QAPI routines as follows: (a) the Quadrics/APE100 memory address and the size of the TAO variable **namef** are defined; (b) the value of the MIMD variable **f** is written into the TAO variable **namef**.

```

(a)add_f = AP_Symaddr(namef,name)
    if (add_f .eq. -1) then
        print*,'Error Variable not found f'
    endif
    size_f = 0
    size_f = AP_Symsize(namf,name)
    if (size_f .eq. -1) then
        print*,'Error Variable not found f0'
    endif

```

```

(b)if(AP_Write(f,QDEV_FPU_SLICE,add_f,
    size_f,0,0,0,iprocx,iprocy,
    iproczz) .eq. QOK) then
    print*,'Writing on Quadrics memories'
end if

```

In this case the Transputer interface has been used for data communication. In fact, due to the small amount of data to be communicated to the SIMD platform (~500 kbytes) the choice of the Transputer interface is more suited as it has faster start-up times with respect to the Hippi channel. In this particular case, however, the MIMD-SIMD communication does not play a crucial role on the whole calculation.

4. This operation is repeated for all the variables which must be given to the SIMD code to allow the evaluation of eq.(1). The order for the execution of the code, previously loaded in task (2), is imparted as follows:

```

    if (AP_Run().eq.QOK) then
        print*,'QUADRICS Program started'
    else
        print*,'QUADRICS Program is not started'
    end if

```

5. At the end of the execution of the code **name**, the main code reads the results on each processor of the SIMD machine (each processor evaluates a single value of θ) and puts together the whole function $I(2\theta)$: (a) the Quadrics/APE100 memory address and the size of the TAO variable **namei** are

defined; (b) the value of the Quadrics/APE100 variable **namei** is downloaded into the MIMD variable **intensity**.

```

(a)add_i = AP_Symaddr(namei,name)
    if (add_i .eq. -1) then
        print*,'Error Variable not found
            intensity'
    endif
    size_i = 0
    size_i = AP_Symsize(namei,name)
    if (size_i .eq. -1) then
        print*,'Error Variable not found
            intensity'
    endif

(b)if(AP_Read(intensity,QDEV_FPU_SLICEMD,
    add_i,size_i,0,0,0,iprocx,iprocy,
    iproczz).eq. QOK ) then
    print*,'Reading from Quadrics memories'
end if

```

The pseudo code for the XRD pattern computation can be summarized as follows:

1. Compute $f(\theta, \alpha)$;
2. Initialize SIMD machine and load the parallel code;
3. Communicate to the SIMD machine all data necessary for eq.(1) calculations;
4. Start the SIMD computation;
5. Upon completion of the SIMD code, communicate results from SIMD to MIMD platform (each SIMD node communicates the proper $I(2\theta)$ value).

5. Code-machine performances

From the algorithmic point of view, the code scales as $O(N^2)$, as the most intense computational part is the double sum i,j of eq.(1).

A crucial parameter of the simulation is the number of θ values which are required to build up the $I(2\theta)$ function. A number of θ values of the order of 512 is usually sufficient, as the θ range which must be spanned is $10 < \theta < 60$ degrees.

A typical XRD calculation with $N=36000$ atoms and 512 angles lasts ~9600 sec. on a QH4 machine (where each fpu performs a single θ calculation of the entire structure) whereas it takes ~30 hours on a Cray J90 vector processor. On a smaller platform (i.e. QH1 with

128 fpu's), 4 different loops are performed (128 θ angles are evaluated at each loop). The execution time, in this case, is nearly four times longer than in the QH4 case, as the SIMD platform does not require to be re-loaded at each loop.

The architecture of the fpu of the SIMD platform [1] is properly exploited by this code, as it performs at a sustained efficiency of $\sim 15\%$.

5.1. Test cases

Two different structures have been thus analyzed to reproduce the experimental XRD pattern of the equiatomic solid solution of Ag-Cu (fig.2) [2].

The (a)-structure is a large fcc lattice (containing $N=36000$ atoms) with the lattice parameter sets at the average value of the lattice parameters of Ag and Cu. An equal number of Ag and Cu atoms has been placed on the lattice sites, with a random distribution. The application of eq.(1) to this structure results in the $I(2\theta)$ pattern reported in fig.3.

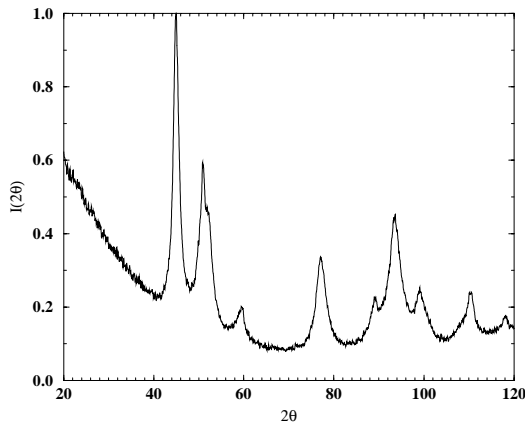


Figure 2. Experimental XRD pattern of equiatomic solid solution of Ag-Cu, mechanically milled for 12 hours and thermally annealed at $T=530$ K.

The (b)-structure has been realized by assembling (along the z axis) a number of alternating layers of pure Ag and pure Cu. The layers have different thickness and are assembled with a random stacking sequence, keeping fixed the overall equiatomic composition. The total number of atoms is still $N=36000$. Each elemental layer exhibits a lattice spacing typical of the pure element. The inter-layer spacing at the boundaries between Ag and Cu regions has been set at the average

lattice spacing of Ag and Cu. The application of eq.(1) to this structure results in the $I(2\theta)$ pattern reported in fig.4.

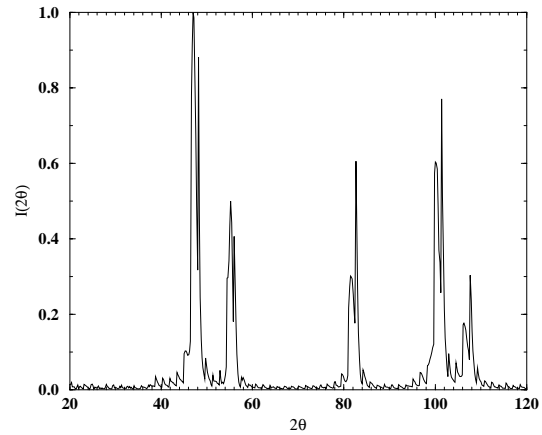


Figure 3. Simulated XRD pattern of the (a)-phase of equiatomic Ag-Cu (see text).

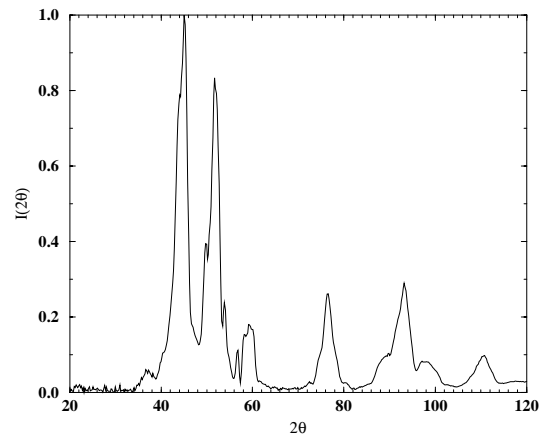


Figure 4. Simulated XRD pattern of the (b)-phase of equiatomic Ag-Cu (see text).

As the geometric positions of a regular lattice do not contain any form of static or thermal disorder (present in a real system) the (b)-phase (which is that which showed the best agreement with experimental data of fig.2), has been equilibrated at room temperature and vanishing external pressure, by Molecular Dynamics (MD) relaxation, using a n -body potential [6], to see if the insertion of realistic static and thermal displacements favours a better agreement with experimental data. The structure resulting after the MD relaxation contains all the static displacements of the atoms (particularly in the interfaces regions, where large static

displacements are introduced by the presence of neighbouring unlike pairs) and the thermal displacements due to thermal motion. The application of eq.(1) to the relaxed structure results in the $I(2\theta)$ pattern reported in fig.5 which shows a better agreement, than fig.4, with the experimental data of fig.2. The agreement to the experimental data has been defined by evaluating the squared distance between the target pattern (fig. 2) and the patterns of figs. 4 and 5, in the whole range $20 < 2\theta < 120$. For $2\theta < 20$ there is a large spike, centered at $2\theta = 0$, corresponding to the not-diffracted term. This term has not been taken into account as, in the experimental pattern, is often affected by spurious effects.

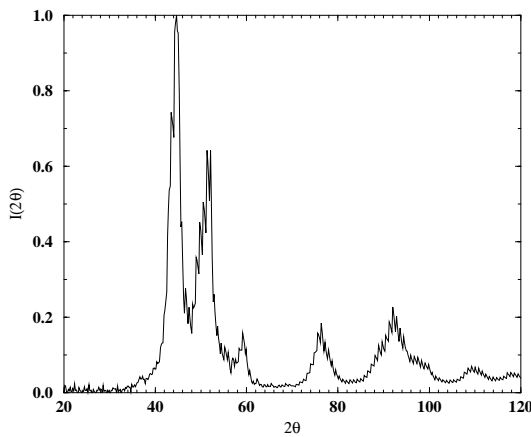


Figure 5. Simulated XRD pattern of (b)-phase of equiatomic Ag-Cu after the MD relaxation (see text).

6. Discussion and conclusions

The availability of a code which allows to perform repeated large $O(N^2)$ calculations with $N \sim 10^5 - 10^6$ is of relevance in the field of materials science. In the present case, it has been used to discriminate among different working hypotheses used for predicting the correct microscopic-scale structure of the solid solution of immiscible elements. The same tool is going [9] to be used within a Reverse Monte Carlo optimization method [7, 10] where the 3-dimensional structure of amorphous Silicon must be inferred from the knowledge of some experimental data (among them, the experimental XRD pattern). The availability of a high performance platform, in this case, is particularly useful as the computation of the XRD pattern, although being performed on a smaller system ($N \sim 10^3$ atoms)

should be repeated at each of the 10^6 MonteCarlo steps required for a typical Reverse MonteCarlo simulation. In this application, the use of the hybrid MIMD-SIMD platform is almost mandatory. In fact, the strategy of the RMC technique is the following. A number of properties (whose experimental values are known) are evaluated on a initial guess structure. A "quality of hit" value is defined as the "generalized" distance between the set of values evaluated on the guess structure and those measured on the experimental sample. The guess structure is then slightly perturbed (e.g. by displacing one or more atoms from the current position) and the set of properties re-evaluated on the perturbed structure. The perturbation is accepted (rejected) according to the decrease (increase) of the quality of hit value. This process is iterated until convergence.

This type of algorithm can highly benefit of a concurrent calculation performed by the different parts of the platform. In this case, in fact, one of the properties which must be evaluated on the current structure is the XRD pattern. This task is totally allotted to the SIMD machine which executes this task. During this time, the MIMD part operates to calculate the values of the other properties. These, differently from the XRD pattern, could not be suited to be evaluated in a data parallel paradigm. In this way the MIMD and the SIMD part are efficiently exploited on computational task where they can better perform. This highly reduces the total computational time.

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