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# Quantitative determination of antiphase boundary profiles in long period superstructures. I: real space analysis

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Résumé. — Nous présentons une méthode d'analyse numérique de clichés de microscopie électronique à haute résolution pour déterminer, au moins semi-quantitativement, la fonction de modulation de toute structure à parois d'antiphase périodiques qui présente des parois larges induisant un désordre chimique en son voisinage. On montre que ces structures peuvent être décrites de façon quantitative par une fonction de modulation générique en forme de créneau arrondi dépendant de trois paramètres: le paramètre de maille, a<sub>0</sub>, l'espacement moyen des antiphases, M, et l'extension de l'arrondissement du créneau, z<sub>0</sub>. Les propriétés topologiques et thermodynamiques des différentes structures peuvent être exprimées à l'aide des rapports  $M/a_0$ , qui détermine la période, commensurable ou non, de la structure,  $z_0/M$ , qui définit le profil des parois et  $z_0/a_0$  qui mesure l'épinglage des parois sur le réseau. La méthode a été utilisée pour comparer les structures observées dans les systèmes Cu-Pd et Ti-Al. On montre que, dans les deux cas, les structures présentent des profils de paroi semblables. Dans le système Cu-Pd,  $M/a_0$  et  $z_0/a_0$  sont grands si bien que les structures sont incommensurables : elles relèvent d'un régime de haute température. Par ailleurs, avec de faibles rapports  $M/a_0$  et  $z_0/a_0$  les structures dans le système Ti-Al sont commensurables avec des parois épinglées, ce qui est typique d'un régime de température intermédiaire.

Abstract. — We present a method combining chemical lattice imaging, using high resolution electron microscopy, and statistical analysis of numerically processed micrographs for determining, at least semi-quantitatively, the modulation function in any long period antiphase boundary structure (LPS) with wide APB generating a chemical disorder in their neighborhood. It is shown that these structures can be quantitatively represented by a generic modulation function in the form of a smoothed square wave involving three characteristic lengths: the lattice spacing,  $a_0$ , the mean APB spacing, M, and the width of the smoothing,  $a_0$ . The topologic and thermodynamic properties of the different structures can be expressed using the ratios  $M/a_0$ , which determines the period, commensurate or not, of the structure,  $a_0/M$ , which defines the APB profile and  $a_0/a_0$  which measures the pinning of the APB to the lattice. The method has been applied to compare the LPS observed in Cu-Pd and in Ti-Al. It is shown that in both cases the LPS have a similar APB profile. In Cu-Pd, both  $a_0/a_0$  and  $a_0/a_0$  are large so that the structures are incommensurate and belong to a high temperature regime. On the other hand, with small  $a_0/a_0$  and  $a_0/a_0$  the LPS in Ti-Al are commensurate with pinned APB, which is typical of an intermediate temperature regime.

#### 1. Introduction.

Modulated (commensurate and incommensurate) structures form an important class of structures, observed among all categories of materials: metallurgical or magnetic compounds, minerals, ionic or molecular insulators, inorganic metals of low dimensionality, organic charge transfer metallic salts, etc. The modulation, which is a periodic perturbation of a basic structure, has frequently a one-dimensional character. In numerous cases it involves a continuous variable, for example the atomic position in the case of displacive modulations. The modulations are generally incommensurate with, in some cases, the occurrence of lock-in transitions when lowering the temperature. In the other class of modulation, the modulation involves a discrete variable which takes a *finite* number of values: this variable can be the nature of the atoms, of an intercalation layer, etc.

The so-called long period antiphase boundary structures observed in various binary alloys belong to the latter class of modulations. In that case, the atomic sites are those of a FCC lattice and the basic structure is the L1<sub>2</sub> cubic structure with stoichiometry A<sub>3</sub>B (Fig. 1a). The modulation has a one-dimensional character and modulates the site occupancies by a given chemical species in the atomic planes perpendicular to the modulation axis, i.e. to a cube axis. These planes are alternatively pure A planes and mixed AB planes. The mixed planes contain two different sites occupied differently and an exchange in their occupation defines a domain wall called an antiphase boundary (APB) (Fig. 1b), hence the name given to these structures. This kind of ordering has been discovered in Cu-Pd by Jones and Sykes [1] and in various noble metal alloys like Cu-Au, Ag-Mg, Au-Zn etc. (for a review see [2]). In the last decade, long period structures have been observed in new systems, Ti-Al [3], Pt-V [4, 5], Cu-Al [6] with very interesting thermal behaviours.

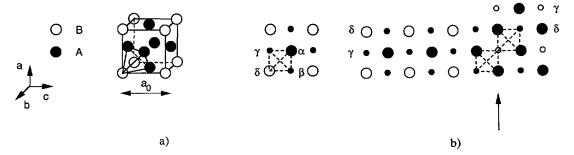


Fig. 1. — a) Sketch of the L1<sub>2</sub> structure (A<sub>3</sub>B). It contains four atomic sites defining four cubic sublattices,  $\alpha\beta\gamma\delta$ , one of them being occupied by the minority atoms B. Along a cube axis it consists of a stacking of alternating pure A planes (sites  $\alpha$  and  $\beta$ ) and of mixed AB planes (sites  $\gamma$  and  $\delta$ ). b) Sketch of a conservative APB lying in a (001) plane indicated by the arrow. The site occupied by the B atoms is changed from  $\delta$  to  $\gamma$ . This APB conserves the environment of first neighbours as shown by the tetrahedron of first neighbours drawn in dashed lines.

Long period structures have been thus extensively studied since about forty years with two main preoccupations; i) to find a general microscopic description of the structures, ii) to explain the origin of their stability and of their structural properties. Pioneering studies used X-ray diffraction and electron diffraction. On this basis, two kinds of structure models have been proposed. In the first one, Fujiwara [7] describes the structure assuming sharp domain walls strictly contained in cube planes whereas the second model proposed by Jéhanno and Pério [8] takes into account wavy antiphase boundaries. This problem has been solved in the

seventies-eighties using the high resolution electron microscopy (HREM) which allows a direct observation on an atomic scale of the shape and of the distribution of the antiphase boundaries. It has been shown that the boundaries can be sharp or spread over a few atomic planes, which become more or less chemically disordered, depending on the temperature and/or on the chemical composition [3, 9, 10]. High resolution observations of different systems have lead to a general qualitative description accounting for the different experimental situations, using a generating smoothed square wave function (for a review, see [11]).

Much effort has been spent, especially in the last decade, to characterize and to understand the thermodynamic behaviour of long period structures. The modulation is not directly associated with a physical property like electrical resistivity, magnetization, as in most modulated structures. As a consequence, it is not easy to study the thermodynamic properties by measuring a simple physical macroscopic property. The only possible investigations are thus diffraction methods and electron microscopy to determine their whole structure. In situ characterizations are difficult because X-ray and neutron measurements require large single crystals. Although it is possible to heat samples in the microscope, there is to our knowledge no reliable in situ observation of equilibrium states. Thus observing equilibrium states is achieved in annealed and then subsequently quenched samples provided the quench is efficient. It is known since the sixties that the period of the modulation can vary continuously or discontinuously with the chemical composition in noble metal alloys: this is the case in Cu-Au [12], in Ag-Mg [13, 14], and in Cu-Pd [9, 15]. Recently, variations with temperature of the modulation period and of the shape of the domain walls have been shown in Au-Zn [16], Ti-Al [3], Pt-V [5], Cu-Pd [10]. The most spectacular behaviour has been found in Ti-Al which shows between 700 °C and 1 300 °C a devil's staircase with more than twenty different structures [3, 17], characterized by sharp APB at low temperature and by jogging APB at high temperature.

Considering their properties, long period structures can be studied within the general framework of one-dimensional modulated structures. The electronic model proposed by Sato and Toth [8] relating the period to the size and the shape of the Fermi surface explains satisfactorily the observed variations with the chemical composition in noble metal alloys. However, this model only includes internal energy contributions independent of the temperature and cannot explain why most long period structures are observed at high or at intermediate temperatures. A thermodynamic theory is necessary. General Landau arguments of the theory of the phase transitions can be applied. They show that the structures are incommensurate at high temperatures and consist of commensurate regimes separated by discommensurations or solitons [19]. These solitons exactly correspond to the wide APB observed in Cu-Pb for instance [10]. At lower temperatures, the solitons become narrower and pinned to the lattice, so that commensurate structures appear through lock-in transition. At intermediate temperatures, complicated structures can appear, arising from the competition between two natural periodicities, that of the underlying lattice and that related to the interactions. Much progress has been made in the understanding of such competitive effects within the so-called ANNNI model (for a review see [20, 21]). It has been first developed for the modulated structures in magnetic systems. It is now well-known to account for the general features encountered in many long period structures [22, 23]. It has also been generalized to the FCC lattice [24] and successfully applied to the Cu-Pd system [25].

The ANNNI model is a microscopic model with competing short range interactions in one direction. As a result, the ground state is strongly degenerate and many different structures are found at finite temperature. They appear through branching processes experimentally found in Ti-Al, with the possible occurrence of devil's staircase at intermediate temperatures. The structures are commensurate at low temperatures and become mainly incommensurate at high temperatures. They can all be described using more or less smoothed square wave

functions, the smoothing being a function of temperature as shown by Monte Carlo simulations [26]. One of the main interest of this microscopic model is then to show the crucial role played by temperature in the stabilization of long period structures and in the form of the generating function describing them.

It therefore appears very important to determine quantitatively the shape of this generating function. The aim of this paper is to show how to obtain at least a semi-quantitative information from high resolution images. A method using a numerical treatment and a statistical analysis of the pictures has been developed and its feasability tested on various examples. The paper is organized as follows: the qualitative description of the LP structures and the definition of the generating function are given in section 2. Section 3 presents the experimental systems which have been chosen to develop and to test the method. This method is described in section 4 and applications are given in section 5. Finally the efficiency and the limits of the method are discussed in section 6.

# 2. Qualitative approach.

The basic structure L1<sub>2</sub> is drawn in figure 1a. As shown in section 1, long period structures are obtained by introducing periodic antiphase boundaries in (001) cubic planes. APB can be described by an in-plane displacement vector  $\mathbf{R} = (\mathbf{a} + \mathbf{b})/2$ , which exchanges atom occupancies of the  $\gamma$  and  $\delta$  sites in (001) mixed planes (Fig. 1b). The APB distribution is uniform in the sense defined by Fujiwara [7] and can be described using a one-dimensional square wave function  $\theta(z)$  of period 2 M defined as follows:

$$\theta(z) = +1$$
 if  $2kM \le z < (2k+1)M$   
 $\theta(z) = -1$  if  $(2k+1)M \le z < 2(k+1)M$ 

where k is an integer number. This function determines the occupation numbers  $\theta_n$  related to the occupation probabilities  $p_n$  of the sites  $\delta$  by minority atoms, in each mixed plane n defined by its position along the modulation axis,  $z = n - \alpha$ , where  $\alpha$  is an arbitrary phase, by the relation:

$$\theta_n = 2 p_n - 1$$
  $p_n = 0, 1$ ;  $\theta_n = -1, 1$ .

With this definition the occupation function for the other sublattice  $\gamma$  is just  $-\theta(z)$ . The function is uniquely determined by M which represents the mean distance between two consecutive APB. The discontinuities of the function therefore define the ideal positions of the APB. The real positions are imposed by the crystal lattice and can only be at integer values n of z. These appropriate values minimize the distance to a discontinuity. The structure given in figure 2 is the straightforward result of this simple rule, for M = 4/3. Due to the periodicity and the symmetry between the sublattices, all the information lies in an interval  $[\alpha + (k-1/2)M, \alpha + (k+1/2)M[. \theta(z)]$  as defined above provides us with univocal positions for the APB, which are sharp and strictly planar. Such a function describes well some alloys  $(Ag_{3\pm x}Mg [13, 14], Pt_3V [4, 5], Cu_{3\pm x}Al [6])$  where the antiphase boundaries are perfectly straight and where no apparent disorder occurs in equilibrium states (Fig. 3). This is not the case for other alloys such as  $Cu_{3\pm x}Pd$  [9],  $TiAl_{3-x}$  (Fig. 4) [3] and  $Au_{3\pm x}Zn$  [16, 27] where the APB extend over a few atomic planes giving rise to strong contrast variations in their neighbourhood. This phenomenon can be interpreted by considering that the order in these planes is lower than in the other planes — the natural order parameter being the concentration difference between the two sites  $\delta$  and  $\gamma$  in a given plane n (Fig. 1) i.e. precisely  $\theta_n$  [28]. Disordered planes thus have an average occupation parameter  $0 \le |\theta_n| < 1$ , the complete disorder corresponding to  $\theta_n = 0$ . The function  $\theta(z)$  becomes a square wave

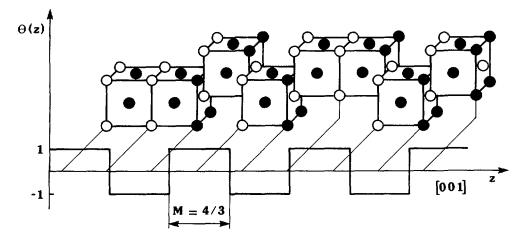


Fig. 2. — The modulation function  $\theta(z)$  for the LPS M=4/3 generating the uniform sequence  $\langle 211 \rangle$ . In the mixed planes, white dots represent the minority atoms which occupy either the site  $\gamma$  or the site  $\delta$ . The nature of the site is changed when crossing a discontinuity of the function. The function has an arbitrary origin with respect to the lattice.

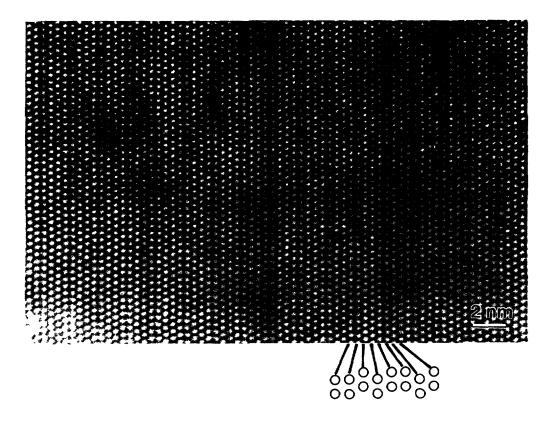


Fig. 3. — High resolution image of the LPS M = 4/3 observed in Pt<sub>3</sub>V, projected along the a axis. The white dots represent the projected columns of minority atoms.

function with rounded corners over a width related to the number of planes affected by the disorder. The smoothing of the function is therefore a representation of the APB profile along the modulation axis.

The periodicity hypothesis for the modulation function (uniformity) implies long-range correlations between disordered planes: if n and m differ by a multiple of M (half period of the modulation) then  $|\theta_n| = |\theta_m|$ . Another consequence is that there is not always a simple relationship between  $\theta_{n_1}$  and  $\theta_{n_2}$  when  $n_1$  and  $n_2$  are neighbouring disordered planes. This effect is particularly striking for structures corresponding to complex rational numbers M = P/Q i.e. with large values of P and Q such that there are many levels in the continuous fraction decomposition of  $M(^1)$ . When M is between 1 and 2, two disordered planes cannot be consecutive — such a situation would mean that the modulation function is an almost sine curve — so that the different disordered planes are spread over the period of the structure (see Fig. 4a). These planes become consecutive in a representation of  $\theta(z)$  modulo M as shown in figure 5; they are classified in order of increasing disorder and reconstruct the APB profile. This reconstruction is a signature of the uniformity criterium. In fact, if P and Q have no common divisor, the values n modulo M are separated by 1/Q and the numbers n modulo M, (n+1) modulo M, ..., (n+P-1) modulo M are all distinct and assume all P possible values of n modulo M. This means that for a complex M, the APB profile  $\theta(z)$  is sampled with a high precision 1/Q and the distance between equivalent planes is equal to P.

Finally the reciprocal space of LPS is recalled in figure 6. There are three different contributions: the fundamental reflections of the FCC host lattice (**K**), the superstructure reflections that arise from ordering in the (001) planes ( $\mathbf{k}_3 + \mathbf{K}$ ) and the satellite reflections of the antiphase modulation along the (001) axis ( $\mathbf{k}_{1(2)} + \mathbf{q}_0 + \mathbf{K}$ ) where  $\mathbf{q}_0 = \frac{2m+1}{2M}\mathbf{k}_3$ , m integer and where  $\mathbf{k}_1 = (100)$ ,  $\mathbf{k}_2 = (010)$  and  $\mathbf{k}_3 = (001)$ . The latter reflections can be viewed as a splitting of the L1<sub>2</sub> superstructure reflections ( $\mathbf{k}_{1(2)}$ ).

Let us mention at last that two secondary modulations are also frequently present. The first one is a displacement modulation affecting the atomic positions in the neighborhood of APB [8]. However, the amplitudes of these displacements are too small to be detected on high resolution images. In the following, we thus consider a rigid lattice with a pure occupation modulation. The third modulation affects the chemical composition of (001) planes, inducing satellites at positions  $(\mathbf{q}_1 + \mathbf{K})$  and  $(\mathbf{k}_3 + \mathbf{q}_1 + \mathbf{K})$  where  $\mathbf{q}_1 = \frac{m}{M} \mathbf{k}_3$  (m integer). It can also be observed in real space using atom probe investigations [32] but not using HREM.

#### 3. Samples.

The quantitative analysis presented below for determining the function  $\theta(z)$  has been applied to two complementary systems which are representative of the diversity of LPS in binary alloys. We were first interested in Cu-Pd, one archetypal system, which is well known to exhibit such structures with an half period M varying from 3 to 15 as the Pd atomic concentration decreases from 30 % to 17 %. The phase diagram, shown in figure 7, has been extensively studied [9, 15, 33, 34]. The structures are mainly incommensurate and characterized by broad APB. An example is given in figure 4b. In that case, there are one APB per period of the structure and 3-4 consecutive disordered planes, so that the qualitative shape of the APB profile is well imaged and can be measured by eye. We investigated several compositions taken out from single-crystalline samples. Because of a concentration gradient

<sup>(1)</sup> The crystallographic period of the structure is P if Q is even and 2P if Q is odd.

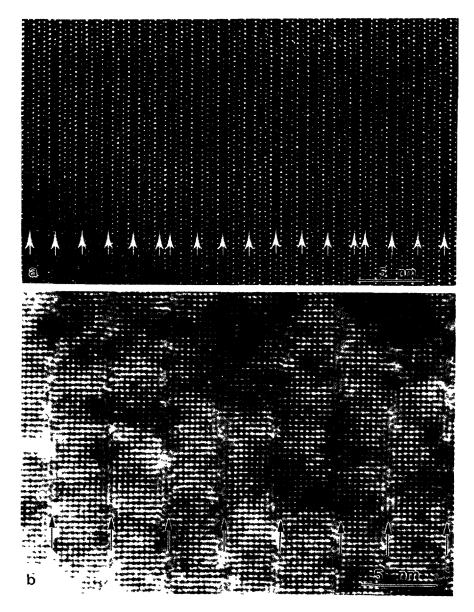


Fig. 4. — a) High resolution image of the LPS M=27/16 observed in  $\mathrm{Ti}_{28}\mathrm{AI}_{72}$  annealed at 1 150 °C. The objective aperture includes the fundamental reflections due to the underlying FCC lattice. White arrows indicate partially disordered planes: in these planes, all columns are mixed  $\mathrm{A}_{1-x}\mathrm{B}_x$  columns imaged as fine grey dots. In other planes columns of minority atoms are imaged by white dots. b) High resolution image of the LPS M=10.35 in Cu-18.5 % Pd annealed at 485 °C. The arrows indicate the mean position of the APB, which extend over 3-4 partially disordered planes. These planes are imaged with a diffuse contrast, the objective aperture used excluding the fundamental reflections, the spacing between two neighbouring mixed columns is not resolved.

in the single crystals (cylindrical ingots) along the growth axis, the composition of a given sample is not accurately known but varies monotonically with its position in the ingot. Slices are cut perpendicularly to the growth axis, then subsequently annealed under vacuum, first in

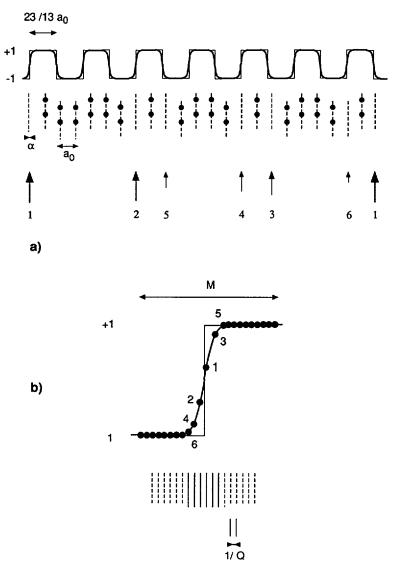


Fig. 5. — a) Schematic representation of a smoothed modulation function for the LPS M = 23/13. The arrows indicate the partially disordered planes classified in order of increasing disorder (decreasing values of  $|\theta_n|$ ) with the numbers 1 to 6. b) Reduced representation of the function *modulo M*: ordered planes are drawn in dashed lines and disordered ones in solid lines.

the disordered state (520 °C) for a few hours, then ordered at 485 °C for a week, with the purpose of developing large single variants. The temperature is then gradually lowered at the required level and the samples are finally oil quenched (2).

The second system of interest is Ti-Al around the 72 % at. Al concentration, which has proved to be an experimental realization of a devil's staircase [3]. Some simple commensurate

<sup>(2)</sup> X-ray experiments performed at the annealing temperature and on quenched samples give identical results showing that the equilibrium state is preserved by the quench [41].

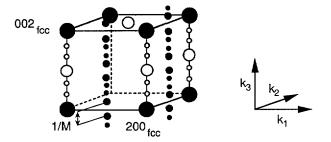


Fig. 6. — a) Sketch of the reciprocal space as experimentally observed: (•) fundamental reflections; (O) reflections due to the chemical ordering of the (001) planes; (••) reflections due to the antiphase boundary modulation; (O) reflections due to a secondary composition modulation.

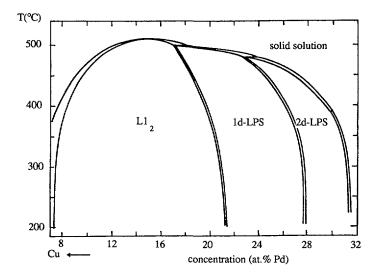


Fig. 7. — Phase diagram of the Cu-Pd system in the Cu-rich part (after [33]).

LPS are observed over a fairly wide temperature range (700-900 °C): M = 4/3, M = 3/2. At higher temperatures, very complex commensurate LPS arise in narrow stability regions. The M value always lies between 1 and 2, so that the real period as well as the hierarchical arrangement of antiphase boundaries are not easily seen on micrographs. Moreover, at high temperatures, the APB are not perfectly thin and planar but can jog over one atomic plane which is more or less disordered, indicating a slightly smoothed APB profile. An example of a complex structure (M = 27/16) is given in figure 4a. This case corresponds to the situation shown in figure 5: the APB profile cannot be easily seen on the micrograph since the different disordered planes, which are characterized by different values of the occupation number  $\theta_n$  defining the APB profile, are not consecutive as in Cu-Pd, but spread over the period of the structure. It is not easy to estimate the relative degree of disorder of the atomic planes and a quantitative analysis of the micrographs is necessary to determine the APB profile.

#### 4. Quantitative analysis of micrographs.

All high resolution images analyzed in the following are projections of the structure along a  $\langle 100 \rangle$  direction, that is a cube axis perpendicular to the modulation axis, so that the boundaries are seen edge on. They are taken with the incident beam along (100) by interfering a large numer of reflections of the (100) zone axis. Along the projection direction, only atoms of the same chemical species overlap. It has been shown by numerical simulations using the method developed by Van Dyck and Coene [35] that for foil thicknesses ranging from 7 to 25 nm, a square pattern of high contrast bright dots is obtained in a rather large defocus interval around the Scherzer defocus, and that the bright dots correspond to the atomic columns of the minority atoms [3, 36]. These conditions realize chemical contrast images and provide therefore a direct analysis of the distribution of the APB on an atomic scale as sketched in figures 2, 3. In the APB neighborhood, the mixed planes can be partially disordered, that is the atomic columns are mixed  $A_xB_{1-x}$  columns. These columns are imaged as fine grey dots (see Fig. 4a, the planes indicated by the arrows), if the resolution of the microscope allows to separate distances shorter than 0.2 nm. If not, it has been shown from numerical simulations [3, 36] that these columns are imaged with a diffuse grey contrast (see Fig. 4b). We have verified that the results of the analysis presented now do not depend on the choice of resolution conditions.

These imaging conditions provide a convenient tool to analyse the dot intensities in terms of chemical composition of the atomic columns. The most direct way to reach the quantitative APB profile is therefore to extract the modulation function from the intensities measured on the micrographs. We first developed a « by hand » method [37, 38]. It consisted in a 3-value reading of the image. White dots with maximum contrast are assumed to be pure minority atomic columns. When the dot is too faint or in phase with none of its neighbours, the atomic column is assumed to be a mixed AB column. Working with these simple rules, the occupation numbers  $\theta_n$  are obtained by summing the minority atomic composition of the atomic columns belonging to a given sublattice in each mixed plane n. Considering an arbitrary origin on the  $\gamma$  sublattice for instance, the maximal value for the occupation function  $(\theta_n = 1)$  is assumed for each plane whose all white points are equivalently bright and which are in-phase (Fig. 8,  $\gamma$  marks);  $\theta_n = -1$  being assumed for out-of-phase planes (Fig. 8,  $\delta$  marks). The occupation function of other (diffuse) planes is proportional to the difference in the number of in-phase and out-of-phase dots.

This analysis is naturally very sensitive to eye appreciation. Nevertheless, it confirmed the feasability of such a method in that sense that uniformity and periodicity can be quantitatively attested [37, 38]: the  $\theta_n$  values are well arranged on a single smooth hull function as shown in figure 9. In particular, the representation  $\theta(z)$  modulo M emphasizes the hierarchy between the different disordered planes which is not visible to eye on the micrograph. In this representation, the M value giving rise to the minimal dispersion of the data is determined with an accuracy better than 5 ‰ and is consistent with the analysis of the diffraction pattern. The structure is thus definitely commensurate with a period  $p = P = 27 a_0$ . Within a period, seven planes are more or less diffuse and the hierarchy in the disorder is well defined by the quasi exact superposition  $\theta_n$ ,  $\theta_{n+P}$ ,  $\theta_{n+2P}$ , etc. (Fig. 9c) with a dispersion less than 5 ‰.

In order to improve the statistical quality (as well as the comfort!) of the analysis we found it necessary to implement some automation through digital image processing. The negatives were digitized via a CCD camera by the Synoptics Synergy board, mounted in an IBM-Pcat. Processing and the analysis of  $512 \times 512$  pixels images were subsequently performed by Synoptics Semper 6 software, following an algorithm which is now described.

The first condition to be fulfilled is the ability of the system to define, throughout the image, reference values that correspond to extreme values  $\pm 1$ . The image must therefore be

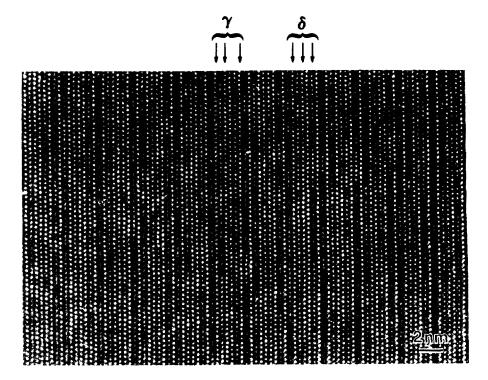


Fig. 8. — High resolution image of the LPS M=23/13 observed in  $Ti_{28}Al_{72}$  at 1 150 °C. The objective aperture used excluding the fundamental reflections, disordered columns appear diffuse. The marks  $\gamma(\delta)$  indicate planes in which columns of minority atoms are in-phase (out-of-phase).

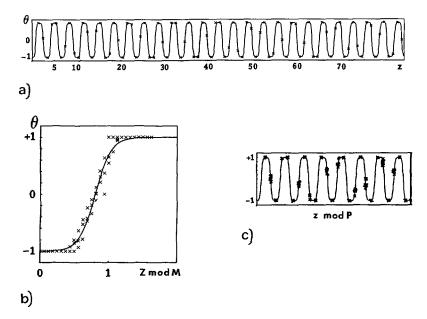


Fig. 9. — Statistical analysis of the LPS M = 27/16. a) Extended representation of the modulation function resulting from the analysis of the image reproduced in figure 4a; b) Representation *modulo* M; c) Representation *modulo* P.

as uniform as possible, on length scales greater than atomic distances. The inhomogeneities are essentially due to slight variations of the sample thickness and to local defects such as surface oxidation or contamination. Because of the clear-cut difference in characteristic wavelengths between defects and lattice information, they are best discriminated in Fourier space. More precisely, the information we are interested in is located in fundamental and satellite Bragg spots whereas the irrelevant data produces a weak diffuse scattering. We thus perform Fourier filtering, which consists in an inverse Fourier transform, after setting to zero the amplitude and the phase for all  $|\mathbf{q}| \langle q_{\min} \text{ and } |\mathbf{q}| \rangle q_{\max}$  in the direct Fourier transform (Fig. 10). This is a two-dimensional band-pass filter, whose real space equivalence is a convolution with Bessel functions. High-pass filtering eliminates low frequencies in the image as desired. A cleaned image is presented in figure 11a.

The next step consists in collecting in each mixed plane the intensities at the nodes of the square lattice, which is the projection of the cubic sublattice containing the minority atoms. It is therefore necessary to determine its parameters (origin and basis vectors) with sufficient accuracy. Because of the discretization process this step is crucial. The parameters one can obtain in Fourier space or by lattice fitting in real space are not accurate enough and must be refined by an trial-and-error method. No better quality criterion could be found than eye control, except of course that the final sampling actually leads to a square-like wave function after summing the intensities collected in each plane. This obvious statement is in fact very constraining as can be seen numerically in figures 11b-e. Figures 11b and c present two good samplings of the original image 11a in that sense that the projection perpendicular to the

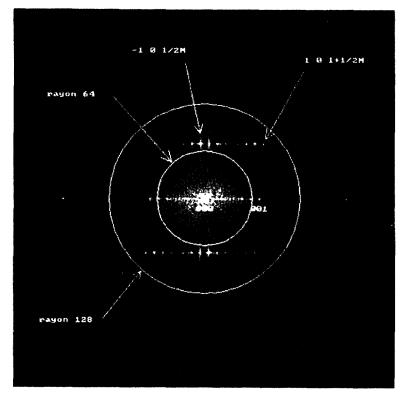


Fig. 10. — Numerical Fourier transform of a digitized image and the band pass filtering used: only the frequences between the two circles are kept.

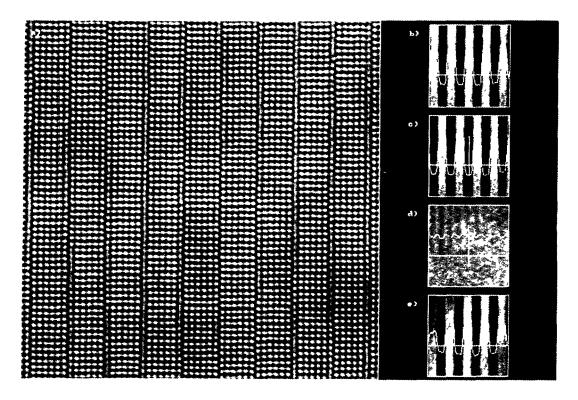


Fig. 11. — Image analysis of the LPS M = 6.21 in Cu-20.5 % Pd at 485 °C. a) Digitized and filtered image; b) and c) Sampling of the image after projection perpendicularly to the modulation axis; in b) origin on the  $\gamma$  sublattice; in c) origin on the  $\delta$  sublattice. d) Summation of the samplings b) and c), showing that the total dot intensity is dispatched over the two sublattices and that the representations b) and c) are complementary, e) Sampling showing the effect of a small mismatch of lattice parameters.

modulation axis (hereafter designed by z, y being the other direction in the image), gives a hull function with well defined minimum and maximum plateaus. The images are determined by I(n, j), the intensity of the pixel located at site (n, j):

$$I(n, j) = I_0(w_z + nu_z + jv_z, w_y + nu_y + jv_y)$$

where  $(w_z, w_y)$ ,  $(u_z, u_y)$  and  $(v_z, v_y)$  are couples of real numbers in pixel units that represent the origin and basis vectors mentioned earlier.  $I_0(z, y)$  is the intensity of the (z, y) pixel which is interpolated if necessary. Figure 11b is obtained with:

$$w_z = 4.3$$
  $w_y = 0$   $u_z = 8.66$   $u_y = -0.05$   $v_z = 0$   $v_y = 8.55$ .

The complementary sublattice (Fig. 11c) with identical vectors  $\mathbf{u}$  and  $\mathbf{v}$  but its origin is shifted by  $\mathbf{v}/2$ :

$$w'_z = w_z - v_z/2 = 4.3$$
  $w'_y = w_y - v_y/2 = -4.27$ .

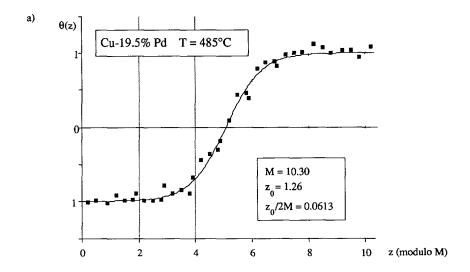
Both images and projected profiles are shown with the same intensity scale, showing the good quality of the choice of parameters. Another test is achieved in figure 11d where the average of the two previous images is computed and displayed at the same intensity scale. It proves

that the resulting intensity is constant and that there is no particular loss at the antiphase boundary; the assumption that this total intensity is dispatched over the two sublattices is not invalidated. Figure 11e shows the effect of a small mismatch of lattice parameters. It results from the following choice:

$$w_z = 4$$
  $w_y = 0$   $u_z = 8.7$   $u_y = 0$   $v_z = 0$   $v_y = 8.5$ ,

which is just an approximate version of the first one. It can be seen that corners in the images are blurred, meaning that sampling points fall in between lattice sites. As a consequence it becomes impossible to determine extrema reference values.

Once the occupation values are obtained, it remains to determine the best hull function fitting these values, its period and its smoothing. We describe the APB profile f(z) by a



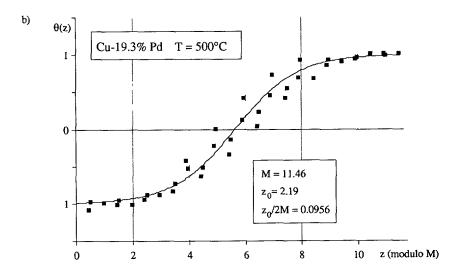
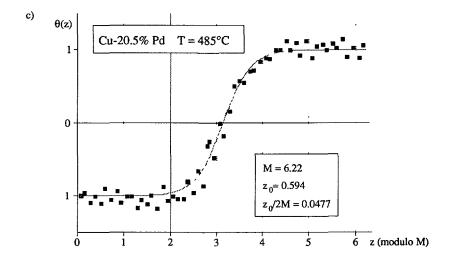


Fig. 12. — Some examples of modulation function (represented modulo M) determined in Cu-Pd alloys.



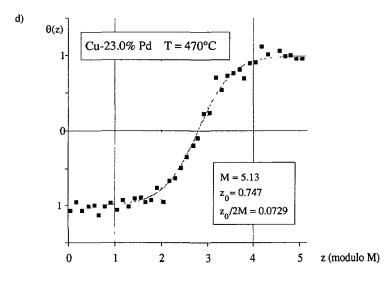


Fig. 12 (continued).

simple hyperbolic tangent law, which accounts for the width through a parameter  $z_0$  and gives an exponential decay towards the domains of in-phase planes. We write:

$$f(z) = \tanh \frac{(z \, modulo \, M) - \alpha}{z_0}$$

and  $\theta(z) = f(z) * \sum_{k} (-1)^{k} \delta(z - kM)$ 

where  $\alpha$  is the phase. This parameter is meaningless for true incommensurate structures but appears in this analysis because only a small area of the total function is fitted. For commensurate structures (M = P/Q) however, it yields indication on the position of the APB with respect to the atomic planes. If the boundary lies on a plane,  $\alpha$  vanishes, whereas the

maximum value  $\alpha = 1/2 Q$  indicates that it remains as far as possible from any atomic plane. Clearly, this only makes sense in the case of simple commensurate structures where 1/2 Q remains significantly different from 0.

From a computational point of view,  $\alpha$  and  $z_0$  can be directly obtained through a bilinear regression procedure, but M cannot because the modulo operation is too singular. The method thus consists in scanning over a reasonable range of M values and selecting the one that minimizes the variance of the regression data.

#### 5. Results.

The method has been applied to the alloys described in section 3. All the measurements are grouped in table I. Some profiles of LPS in Cu-Pd are presented in figure 12. In all cases, the  $\theta_n$  values are well fitted by the hyperbolic tangent law with a relative small dispersion that insures a precise determination of M and of  $z_0$ . An example of a LPS observed in Ti-Al is shown in figure 13. In that case again, the fit is very satisfying and the dispersion of the data very small. The M value (23/13  $a_0$ ) resulting from the fit exactly corresponds to that found from the diffraction pattern (Fig. 14) [11]. Over a crystallographic half-period ( $p/2 = P = 23 a_0$ ), there are seven, i.e. about  $z_0 \times P$ , non perfectly ordered planes and the hierarchy in this disorder is well defined by the quasi exact superposition of  $|\theta_n|$ ,  $|\theta_{n+P}|$ ,  $|\theta_{n+2P}|$ , etc. (Fig. 13b).

One can easily check the validity of the numerical value of  $z_0 \cdot 2 z_0$  is indeed the width where the hyperbolic tangent is significantly different from  $\pm 1$ ; the number of consecutive diffuse planes on the image should thus be about  $2 z_0$  (see Fig. 8). This is well the case when comparing micrographs and numerical APB profiles. In particular, in Ti-Al  $2 z_0$  is below 1 which is consistent with the fact that two diffuse planes are never consecutive. The other interesting value is the dimensionless ratio  $z_0/2 M$  which should permit to compare the nature of the profiles for different samples in different thermodynamic conditions. From the

Table I. — Results of the statistical analysis of LPS images in Cu-Pd and in Ti-Al.

Sample	Annealing temperature	M/a <sub>0</sub> diffraction	M/a <sub>0</sub> HR	$z_0/a_0$	z₀/2 M
Cu-18.5 % Pd	485 °C	13.10	12.90	1.71	0.0663
Cu-19.5 % Pd	485 °C	10.75	10.30	1.26	0.0610
Cu-19.3 % Pd	~ 500 °C	10.35	11.46	2.19	0.0956
Cu-20.5 % Pd	485 °C	6.26	6.21	0.595	0.0479
—	485 °C (2)	6.34	6.32	0.624	0.0494
—	491 °C	6.43	6.43	0.566	0.0440
Cu-23 % Pd	430 °C	5	5	0.720	0.0720
—	470 °C	5.22	5.13	0.747	0.0729
—	485 °C	5.35	5.28	0.751	0.0712
Ti <sub>28</sub> Al <sub>72</sub>	1 150 °C	23/13	23/13	0.280	0.0793

collected results it seems impossible to draw any conclusion concerning the evolution with temperature and concentration. For Cu-Pd alloys, the relative widths vary from 0.044 to 0.073 without visible correlation with thermodynamic parameters. Only one sample presents a wider profile  $(z_0/2\ M=0.0956)$ ; the thin foil comes from a polycrystalline specimen, annealed at a temperature very close to the disorder transition (Fig. 4b). For the Ti-Al alloy (Fig. 13),  $z_0$  is obviously small  $(z_0=0.28)$  but it is striking that the relative width  $z_0/2\ M=0.08$  is quite comparable to that of Cu-Pd alloys.

We have tested the sensitivity of the method to the parameters introduced in the algorithm [28]. The stability with respect to sublattice exchange was mentioned in section 4, with the following results: M = 6.21,  $z_0/2$  M = 0.0477 and M = 6.22,  $z_0/2$  M = 0.0481. Secondly the choice of the frequencies during band-pass filtering especially influences the point dispersion but does not affect systematically the M and  $z_0$  values. At last, we studied the validity of the sampling by changing the number of representative pixels per white dot. It implies of course that image capture is done at a higher magnification and thus reduces the number of analyzed planes. The test was performed with a 81 pixel matrix for sample  $n^{\circ}$  4 and gave M = 6.24,  $z_0/2$  M = 0.0495 whereas, at the same magnification, the standard analysis (1 pixel per dot) results in M = 6.24,  $z_0/2$  M = 0.0487.

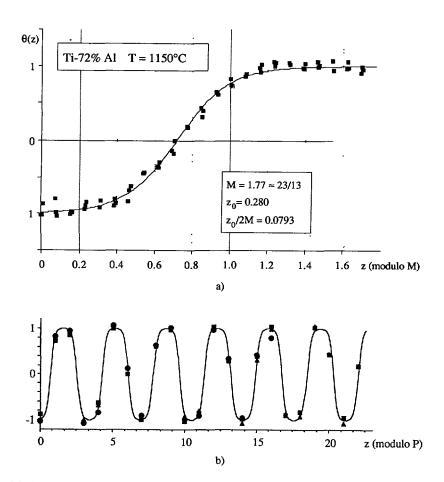


Fig. 13. — Modulation function determined for the LPS M = 23/13 resulting from the analysis of the image reproduced in figure 8. a) Representation modulo M; b) Representation modulo P.

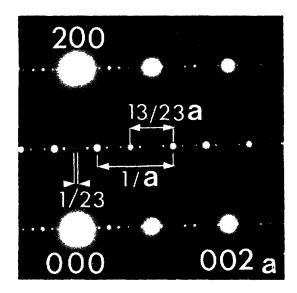


Fig. 14. — Diffraction pattern of the (010) zone axis of the LPS M = 23/13. The satellites due to the modulation are located on the line [10  $\zeta$ ] line with  $\zeta = (2 p + 1)/2 M$  (modulo 1) [11]. The five first harmonics have a significant intensity. Their relative positions are consistent with M = 23/13: the first order harmonics are separated by 13/23 i.e. 1/M and the shortest distance between two consecutive harmonics is 1/23 i.e. 1/P.

#### 6. Discussion.

From the examples presented in section 5, it clearly appears that the power of the method is to show the validity of a quantitative description of any LPS by a generic regular periodic function. One of the most striking results is the numerical proof of the uniformity of the APB distribution following the uniformity criterion defined by Fujiwara [7] even for the complex structures observed in Ti-Al. Here is confirmed the existence of very long range correlations between the positions of the APB.

A single function accounts for any LPS in spite of their apparent diversity when considering their imaging in high resolution electron microscopy. The description involves three lengths: the period of the underlying lattice i.e. the spacing of the mixed planes,  $a_0$ , which represents the natural length scale, the periodicity related to the interactions between APB, M, and the width of the APB,  $z_0$ . Topological differences between the LPS come from the relative values of these three characteristic lengths. The periodicity of the function is expressed through the ratio  $M/a_0$  whereas the smoothing of the APB profile is characterized by the ratio  $z_0/2 M$ ; finally the ratio  $z_0/a_0$  measures the pinning of the APB to the lattice.

In all cases, the larger the number of disordered planes, the more precisely defined is the APB profile. This is realized either when both  $M/a_0$  and  $z_0/a_0$  are large or when Q and  $z_0/2$  M are large. The first condition is realized in Cu-Pd and the second one in Ti-Al. In both systems, the ratio  $z_0/2$  M is about 0.1, meaning that the profiles are of the same nature. However, in Cu-Pd,  $M/a_0$  is large i.e. larger than 2 and incommensurate. As a consequence, there is only one APB per period and all the different disordered planes are located in the neighborhood of the APB, which allows to image directly the shape of its profile. Ti-Al corresponds to an opposite situation.  $M/a_0$  takes complex rational P/Q values between 1 and 2. The APB profile produces a certain number of different disordered planes which are at least separated by one perfect mixed plane since 2  $z_0$  is smaller than 1. The larger Q, the larger

this number. The disordered planes are spread over the whole crystallographic period of the structure, each being close to a different APB. As a consequence, the Q APB are imaged in different ways. APB appear either perfectly thin and planar or jogging over one atomic plane. This hierarchy in the disorder and the selective imaging of the APB are the signature of the property of uniformity.

The theoretical framework recalled in section 1 can be considered and rendered more precise in terms of the characteristic lengths involved in the present description. A high temperature regime is characterized by large values of the ratios  $z_0/a_0$  (i.e. larger than 1) and of  $z_0/2$  M (i.e. at least 0.1). In particular, a large  $z_0/a_0$  ratio (i.e. larger than 1) means that the APB are weakly pinned by the lattice, which is typical of solitons in incommensurate structures. These conditions are realized in Cu-Pd. In Ti-Al, the second ratio only is large so that the structures remain commensurate with pinned APB: this situation is typical of an intermediate temperature regime. Not here that temperature is not the only relevant parameter; for instance long-ranged oscillatory interactions favour incommensurate structures at fixed temperature. This has been recently discussed in detail by Ceder et al. [24-25].

The determination of the modulation function and of its parameters by the method presented here is only semi-quantitative, although the results are fairly consistent with the appreciation by the eye of the micrograph. Considering the tests presented in section 5, we can reasonably assume that the results are not dramatically dependent of the numerical processing: Fourier filtering, sample size of each dot, and that the micrographs are correctly and unambiguously analyzed. One of the factors restricting the quantitative character of the method is the fact that a high resolution image is a two-dimensional projection of the structure and that we have no direct information along the direction of projection. This is in particular crucial for the disordered planes. The analysis of two-dimensional projections is valid since the wavelengths of the composition fluctuations along a column (about 2-3 nm, see e.g. Fig. 4b) are much shorter than the foil thickness (10-20 nm).

A more crucial problem concerns the imaging conditions in the microscope. These are chosen in order to realize chemical images in the sense that a pure column of minority atoms is seen as a white dot, the columns of majority atoms being dark. In the present analysis, the dot intensity has thus been related to the chemical composition of a column. This relation has been used in the same way by Ourmazd et al. [39] in the method of image analysis they have developed for analysing the roughness of interfaces in semiconductors. We want to point out the difficulty of contrast interpretation when the conjugate effects of dynamic interaction and chemical disorder add together. To our knowledge, no complete treatment of this problem has been achieved up to now. When a chemical disorder is introduced in a column, some partial simulations performed by Coene et al. [40] have shown that the linear relation assumed here does not apply to any thickness and that as soon as the foil thickness is not « kinematic », the intensity of the dot does not follow any single monotonic law relating it with the amount and/or the nature of disorder.

The present analysis needs therefore to be improved by more quantitative experiments. This can only be done in reciprocal space using X-ray or neutron scattering techniques which are not affected by dynamical effects. The measurement of satellite intensities allows us to recover the hull function  $\theta(z)$  through its Fourier transform. For a pure sine wave, a single satellite is present, whereas the amplitude of higher order satellites is reinforced as the modulation tends to a square wave. Preliminary X-ray diffraction experiments have been performed on the same Cu-Pd single crystals used in this study. They confirm the validity of a description by a smoothed regular function. However, they indicate that the intensity of high order harmonics is too weak with respect to the APB profile determined from micrographs [28]. This would mean that the APB should be wider than their H.R. image or, in other

words, that some weakly disordered columns are seen with the same dot contrast as pure monoatomic columns. It should be said, however, that the intensities of the satellites are disturbed by the existence of a displacive modulation that affects atomic positions in the neighborhood of APB, with amplitudes of opposite sign for both species [2, 8, 29-31]. This disturbance, increasing with |q|, can be all the more important as measurements are made at large |q|. To overcome this difficulty, neutron diffraction experiments, allowing measurements near the origin, are currently performed on the same samples. These comparative studies, which will be published in a forthcoming paper, should help to discuss in more detail the relation between the dot intensity and the chemical composition of a column in high resolution imaging.

# Acknowledgments.

We are indebted to G. Van Tendeloo and D. Broddin for numerous fruitful discussions.

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# Revue de livres

# Atomic and Molecular Beam Methods. Volume I

Edited by G. Scoles

(Publ. Oxford University Press, 1989) 720 p., £ 75.00.

Until the publication of this compendium the standard work for any scientist be they physicist, chemist or molecular biologist was the classic text by Norman Ramsey in 1956. The development of modern ultra-high vacuum systems, supersonic gas sources and the pioneering work on clusters has superceded Ramsey's work. A single volume describing the various facets of the production, detection and monitoring of molecular beams is therefore long overdue. The editor has collected contributions of an outstanding quality from the foremost scientists in the field, yet by careful editing ensured few repetitions and achieved a continuity that is to be praised.

In the opening chapters of this work the basic formulative mechanisms of molecular beams are discussed. The editor himself providing a brief, informative overview of the whole work. David Miller (San Diego, USA) neatly summarises the physics of supersonic expansion, the role of skimmers and free jet design while H. Pauly (Gottingen, Germany) summarises effusive sources and the production of beams of free radicals and metastable species. Ronald Gentry (Minnesota, USA) concentrates upon those designs necessary to produce pulsed gas beams. In three chapters D. Bassi (Trento, Italy) discusses the different methods for the detection and monitoring of molecular beams, a topic extended by U. Hefter (Munich) and K. Bergmann (Kaiserslautern) in discussing spectroscopic detection techniques. M. Zen (Trento, Italy) extends the discussion to the detection of excited species (e.g. metastable species). J. Reuss (Nijmegen, Holland) and K. Bergmann discuss those state selective detection techniques that isolate vibrational and/or rotational states of molecules and the separation of different magnetic sublevels of a degenerate atomic state.

In two excellent chapters, C. J. Van der Meijdenberg (Leiden, Holland) and Daniel J. Auerbach (John Hopkins University, USA) describe techniques to velocity select the individual atoms/molecules within the beam. Van der Meijdenberg describes mechanical methods and Auerbach time of flight techniques, the latter contribution is guaranteed to become a basic reference for any worker in this field.

M. Kappes and S. Leutwyler (both Bern, Switzerland) attempt the complex task of discussing the formation of beams of atomic and molecular clusters. In just thirty six pages they can but provide a modest overview of a complex field but the lucidity of their article is to be commended.

In the final chapters of part one of this volume, K. Ploog (Max Planck Institute, Stuttgart, Germany) reviews molecular beam epitaxy and U. Valbusa (Genoa, Italy) the construction of molecular hydrogen beams within the ring of an accelerator (e.g. the CERN SPS), vacuum and molecular beam physics on the largest possible scale.

The second part of the volume is devoted to molecular scattering and crossed beam experiments. Introductory chapters by U. Buck (Gottingen, Germany) introduces the reader to the general principles and methods of molecular scattering, the measurement of elastic scattering differential cross sections and energy loss methods. Elastic integral cross sections are summarised by J. J. H. Van der Biessen (Philips Laboratories, Eindhoven). Reactive scattering is discussed in two well written chapters by Y. Lee (Berkeley, USA) and P. J. Dagdigan (John Hopkins University, USA). Dagdigan also introduces the concepts of laser induced fluorescence and bolometric detection. The final three chapters bring this volume up to date with recent work at the forefront of this ever evolving field of study. Excellent contributions by S. Stotte (Nijmegen, Holland) and R. Durren (Kaiserslautern, Germany) on scattering experiments with state selected species describe the role of lasers to select atomic states both before and after collision events. S. Iannotta (Trento, Italy) concludes the work with a discussion of new experiments using spin polarised beams. The reader can but be convinced that the study of molecular beams remains an important and challenging field of research, as when Ramsey wrote his classic work.

In conclusion, this single volume should be essential reading for any experimentalist using (or planning to use) molecular beams. It is well written and edited but the price probably confines it to libraries. The publishers should be encouraged to produce a paperback version as soon as possible.

N. J. MASON.

# Solids far from equilibrium

Edited by Claude GODRÈCHE

Collection Aléa Saclay (Cambridge University Press, 1992) 588 p., £ 60.00, \$ 120.00.

The authors of this book are among the most distinguished specialists of non-equilibrium statistical mechanics. Langer's monography of dendritic growth in Reviews of Modern Physics is still a basic reference. Sander is the co-inventor of Diffusion Limited Aggregation (DLA). The group of the Ecole Normale Supérieure (Y. Pomeau, B. and C. Caroli, M. Ben Amar, B. Roulet...) is among the most productive in the field of dendritic growth. Nozières is better known for his contribution to the Kondo problem for instance, but his work with Gallet on the dynamic renormalisation of the roughening transition is also a classic. Finally, Krug and Spohn have done a very interesting work on stochastic growth models.

The book contains the lectures delivered in 1989 at the summer school directed by Claude Godrèche, and which takes place every year in Britanny. Godrèche's main recommendation is to be didactic (use of transparencies is prohibited in order to slow down the speakers). Indeed, the book is perfectly accessible to students and newcomers; even those who are not familiar with the Gibbs-Thomson relation, though the latter is qualified « familiar » (p. 17).

There are 6 parts written by 6 different authors or groups of authors. They will be reviewed according to their increasing degree of specialisation.

Nozières' part, full of many (68) helpful drawings, is a didactic introduction to crystal growth theory. Yet it is rich in original views and things which will be discoveries even for many specialists. Let me test your knowledge, dear reader:

Why is the elastically mediated interaction between two parallel steps of identical sign on a surface repulsive while an analogous mechanism in the bulk leads to the attraction between electrons which gives rise to superconductivity? (Answer p. 44).

Does a crystal with several Frank-Read sources grow faster than with a single one? (Answer: NO, p. 96).

If you press a crystal perpendicularly to its surface, will the latter remain flat? (Answer: NO, p. 24). The first theories of the roughening transition gave correct results, but the calculation was wrong. Who made the first correct derivation? (Answer: Knops and Den Ouden in 1980. See p. 119).

Can the roughening transition be first order? (Answer: maybe, in the strong coupling case, p. 146). Nozières' bibliography contains only 41 references, but it is rich in russian and french citations, and will be useful to those who read only Physical Review Letters.

Langer's contribution, remarkably didactic too, is an introduction to the classics of nucleation and spinodal decomposition: Becker and Döring, Kolmogorov and Avrami, Cahn and Hilliard, Lifshitz and Slyozov. Binder and Stauffer... all names which are always quoted, but no much read. In which library can the works of Becker and Döring (1935) or Kolmogorov (1937) be found? The student will also learn from Langer's text what a Langevin or Fokker-Planck equation is, what it is good for, what a Zel'dovich factor is. One may just be surprised not to find references to Kramer's and Eyring's works.

The chapters written by Sander and by Krug and Spohn are related to the effect of stochastic fluctuations in growth problems. Sander's short part contains much: an introduction to fractals, a review of numerical, analytical and experimental results on fractal growth (DLA) and a simple but interesting presentation of the Eden model (fixed growth probability per unit area) and related models. This point is developed much more in detail by Krug and Spohn, but Sander reviews among other things an exactly solvable 1+1 dimensional model he invented with Meakin et al. The results are the same as can be obtained from renormalisation group in the continuum model of Kardar, Parisi and Zhang... but

how simpler is the derivation! Sander's main merit is to be easy and pleasant to read. Obviously he did not try to make a complete review. In such a review, Pietronero's theory of DLA should be mentioned and the author's contribution to electrolytic growth should not be mentioned without giving some references on the relevant basic mechanisms of electrochemistry.

The last two contributions to be reviewed are devoted to deterministic growth in the strongly nonlinear (dendritic) or moderately nonlinear (Mullins-Sekerka) regime. They are still accessible to beginners, but they are rather specialized and detailed. B. and C. Caroli and Roulet try to make reading easier by putting the most complicated algebra into appendices. On the contrary, nonlinear algebra is the heart of the contribution of Pomeau and Martine Ben Amar, just as mystery is the heart of a detective story. In order to solve the problem of dendritic growth, the detectives invent a « geometrical model » which can be solved by methods which, after many episodes, allow them to prove that there is no solution, but can be applied to the real case too, and in that case there is a solution. The conclusion is an impressive agreement (except at weak undercooling) with experiments on dendritic growth of nickel.

The publisher is perfectly right when he writes that « statitical physicists, condensed matter physicists, metallurgists and applied mathematicians will find a stimulating and valuable introduction » to an « important area of research ». However, this presentation is too limitative. For instance, semiconductor physics is also concerned by crystal growth. And, before all, this book is not *only* an introduction. The monograph of Caroli *et al.* on the « Instabilities of planar solidification fronts » is, with its 140 pages and 134 references, much more than an introduction. The book edited by Godrèche will be a basic reference for the beginner *and* for the specialist.

Jacques VILLAIN.

# The structure of the proton — Deep inelastic scattering

R. G. ROBERTS

(Cambridge University Press, 1990) 182 p., £ 30.00, \$ 49.50.

Although they are many processes where perturbative quantum chromodynamics has been tested our detailed knowledge of the quark distributions of the nucleon still relies only on data from deep inelastic scattering experiments. This book discusses the structure of the proton as revealed by such experiments.

It starts with a brief introductory overview and a chapter on the definitions of the various structure functions encountered in both the scattering of charged leptons and neutrinos from nucleons. These are followed by a chapter on the predictions of the parton model and their comparison with experimental data. Deviations from the parton model are predicted by QCD and are observed in experimental data. This is the subject of the next three chapters. Firstly, there is a short formal introduction to perturbative QCD; then a chapter on the application of QCD to the analysis of the moments of structure functions; and finally a chapter discussing the special problems encountered at very small and very large x values. The former region will soon become of great interest when data appear from the new generation of colliders recently completed or currently under construction. A summary of the parton distributions of various types obtained from analyses of experimental data is given in chapter 7. In the case of the gluon distribution, which only enters indirectly in deep inelastic scattering, information from other sources is also briefly reviewed. The final chapter is devoted to how the structure of the proton manifests itself in the case of bound nucleons. This is of practical importance because many deep inelastic experiments have been done using heavy nuclei as targets. There is also a very short appendix on radiative corrections.

This relatively short monograph is easily the most complete, accessible and up-to-date review of deep inelastic scattering. Although it is primarily theoretical in its approach, experimental data are always clearly in view, and the role of phenomenology is emphasised. It would make excellent reading for anyone wanting to know the state of this field, which looks set to enter a new exciting phase when data from HERA, and later the SSC, become available.

# **Experimental Physics, Modern Methods**

#### R. A. DUNLAP

(Oxford University Press, 1988) 373 p., £ 40.00.

Ainsi qu'il est annoncé dans la préface de ce livre, il s'agit d'un texte qui résulte d'un enseignement d'une année, destiné à familiariser les étudiants avec les méthodes du laboratoire. Il serait plus correct de dire « avec un certain nombre » de méthodes du laboratoire. L'accent est mis en effet sur trois domaines de la physique moderne :

- la physique du solide
- l'optique
- la physique nucléaire.

L'auteur insiste aussi beaucoup sur la physique de l'électronique. On trouve donc un chapitre d'introdution, « The electrical properties of solids », qui résume le modèle de l'électron libre et la théorie des bandes. Suivent deux chapitres qui traitent de certains systèmes semiconducteurs :

- diodes
- transistors à effet de champ
- systèmes analogiques
- amplificateur opérationnel
- systèmes digitaux.

Le chapitre 4 présente une revue assez complète des techniques utilisées en traitement du signal et accumulation de données.

Viennent ensuite quelques chapitres spécialisés consacrés à des technologies d'expérimentation générale (techniques du vide, mesures des pressions, thermométrie, cryotechniques).

Les six derniers chapitres ont, deux par deux, l'intention de décrire successivement l'instrumentation optique, l'instrumentation nucléaire et l'instrumentation propre à la physique du solide (diffraction X et mesure magnétique essentiellement).

Ce livre est certainement utile. Il recouvre une bonne part de ce que les étudiants sont souvent supposés apprendre par eux-mêmes lorsqu'ils débutent leur travail en laboratoire de recherches.

Christian JANOT.

# Random Processes in Physical Systems.

# An introduction to Probability-based Computer Simulations

Charles A. WHITNEY

(Wiley Interscience, 1990) 320 p., £ 36.00.

Il existe beaucoup de bons livres de probabilité. Un des plus connus est le livre de Feller, écrit dans les années 50. Il est à l'origine d'une lignée de manuels d'introduction aux probabilités (Papoulis, Ross, ...) qui — tout en évitant son côté foisonnant — lui empruntent l'essentiel de leurs contenus. Ils suivent tous la trame classique de présentation des concepts : espaces discrets, problèmes combinatoires, variables aléatoires, addition des variables aléatoires et convergence, introduction aux processus stochastiques.

Le livre de Whitney se démarque de ce schéma classique par deux aspects. D'une part, il renonce à un exposé linéaire où chaque notion est définie, chaque théorème est démontré etc. A la présentation traditionnelle et déductive par les concepts, il préfère une présentation par les problèmes. Par exemple il introduit dès les premières sections les marches au hasard qui sont généralement reléguées dans les derniers chapitres des livres classiques de probabilité. D'autre part, l'auteur utilise des simulations numériques pour repésenter des exemples concrets de phénomènes stochastiques ou des notions probabilistes. Le livre fourmille ainsi de programmes écrits en Pascal ou en Basic ou encore faisant appel à l'utilisation d'un tableur. Il faut reconnaître que ce créneau de présentation des probabilités et des phénomènes stochastiques par des simulations est peu occupé. C'est là que réside à mon avis le principal intérêt du livre, en montrant la voie à suivre.

Malgré les intentions affichées par l'auteur dans son avant-propos : « My intent is to discuss physical properties of gases and plasmas from a quantitative, but physical, rather than a mathematical point of view. I feel that most introductions to statistical physics and thermodynamics are so formalized and so steeped in the classical assumptions of equilibrium theory that they have little bearing on practical problems », ce livre est en vérité au moins autant — sinon plus — consacré aux probabilités qu'à la physique. Les deux premières parties traitent en effet majoritairement des espaces de probabilité discrets (première partie) et continus (deuxième partie), avec pour finir deux sections d'introduction à la mécanique statistique. Les exemples d'application à la physique se trouvent regroupés dans la troisième partie : deux sections sur les gaz d'atomes et de photons, et une section d'exemples empruntés à l'astrophysique — l'auteur étant astrophysicien. La dernière partie concerne les méthodes d'estimation et une rapide incursion dans le domaine de l'optimisation avec une présentation succincte du problème du voyageur de commerce et de la méthode Monte Carlo.

Quelques réserves pour finir. Ce qui fait la force de ce livre fait aussi sa faiblesse. En effet le piège qui guette les livres novateurs — par opposition aux exposés linéaires et déductifs où la théorie est bien assise — est qu'ils demandent une agilité d'esprit bien supérieure. Bien que le livre se veuille « self-contained », il faut aller chercher les axiomes des probabilités, les définitions importantes (probabilités conditionnelles, théorème des probabilités totales, etc.) dans un court appendice. « A formalist will notice that in many instances I have used a concept or term before defining it. This is intentional, as I believe that this is the natural way to acquire new vocabulary ».

Enfin ce livre est plus un livre d'initiation qu'une véritable introduction. Il s'adresse donc plutôt aux étudiants de 1<sup>er</sup> cycle. Le chercheur ressentira une certaine frustration de ne pas disposer d'une présentation plus fouillée des problèmes introduits. Ce livre pourra néanmoins stimuler son imagination, surtout s'il est amené à enseigner le sujet.

Claude GODRÈCHE.

# Electrons in metals and alloys

#### J. A. ALONSO and N. H. MARCH

(Academic Press, 1989) 595 p., \$ 49.50.

Ce livre se veut une étude plutôt exhaustive du comportement des électrons dans les métaux (cristallins, désordonnés et en alliage). Il ne s'adresse pas au débutant et requiert au préalable la lecture de plusieurs « bons ouvrages » de physique du solide et du liquide, de mécanique quantique, de physique statistique. L'accent est mis sur les aspects théoriques et sur le rôle des électrons de conduction dans le comportement thermodynamique des métaux et alliages, au sens le plus large.

Cela donne un ensemble compact, bien documenté mais de lecture plutôt difficile, où sont traités avec un certain luxe de détails :

- la structure électronique des métaux
- les défauts ponctuels
- les alliages cristallins
- les systèmes désordonnés
- les propriétés optiques et de transport
- le magnétisme
- les surfaces et interfaces.

Le livre se termine sur 17 « appendices » de quelques pages chacun et se référant aux différents chapitres précédents. On y trouve les compléments les plus variés, comme la règle de somme de Friedel, la méthode des moments ou la méthode du groupe de renormalisation.

Cet ouvrage a les caractéristiques de la « bible » qu'on tentera certainement de consulter lorsqu'il sera nécessaire de se rassurer sur tel ou tel aspect du comportement des électrons dans les métaux et alliages. Cependant, il existe sans doute dans la littérature des présentations du sujet sensiblement plus pédagogiques.

# Initiation à la physique du solide.

### Exercices commentés

J. CAZAUX

(Masson, 1988) 405 p., FF 168.

Ce livre est constitué de quatre grandes têtes de chapitre de la physique du solide :

- édifice cristallin et diffraction des rayonnements
- liaison cristalline et constantes physiques
- vibrations et chaleur spécifique du réseau
- électrons libres
- théorie des bandes et semiconducteurs.

L'originalité de l'ouvrage vient de la présentation. Chaque grande partie commence par un « résumé de cours » qui contient les bases nécessaires et la terminologie courante. Ensuite viennent des exercices dont les questions permettent une découverte active et une construction progressive de la connaissance des sujets. Le lecteur paresseux, ou pressé par le temps, peut consulter immédiatement les solutions des exercices et procéder ainsi comme pour un livre classique. Il est clair qu'il est préférable de procéder autrement, en privilégiant l'aspect recherche personnelle, comme l'a voulu l'auteur, de toute évidence. La fin du volume propose des associations des exercices précédants qui permettent soit de constituer des examens complets d'un objet particulier à travers ses différentes propriétés, soit de traiter des problèmes centrés sur la similitude des raisonnements physiques. Des questions sont également posées aux pages 393 à 399; elles renvoient aussi à des associations d'exercices et incitent à comprendre les aspects physiques, en se dégageant de l'aspect parfois étouffant des calculs.

La publicité de l'ouvrage n'est peut-être plus à faire puisqu'il s'agit d'une seconde édition, la première ayant été épuisée en moins de six ans. Il reste incontestablement le seul livre en langue française de ce type et sur ce sujet. Il a même, à vrai dire, peu de concurrents en langue anglaise. Cette seconde édition améliore encore la première publication, dans le sens d'une évidente modernisation. Parmi les 130 exercices présentés, 50 sont entièrement nouveaux et traitent des sujets les plus actuels (super-réseaux quantiques, EXAFS, spectroscopie Auger, supraconducteurs, effet Hall quantique, microscopie Tunnel, etc...).

S'agissant de façon avouée d'un ouvrage d'initiation, le public directement concerné reste celui des étudiants de second cycle des Universités (Physique, Science des Matériaux, Chimie-Physique). D'autres, à des niveaux différents (DEA, début de thèse, etc...) peuvent certainement y trouver de l'intérêt.

Christian JANOT.

#### Photons and Atoms: Introduction to Quantum Electrodynamics

C. COHEN-TANNOUDJI, J. DUPONT-ROC and G. GRYNBERG

(J. Wiley and Sons, New York, 1989), 468 p., £ 47.50.

Although there are a number of textbooks on quantum electrodynamics now available, this book is unique in the choice and presentation of the material covering basic concepts of quantum electrodynamics. The book is aimed at filling the gap between ordinary quantum mechanics courses and general quantum field theory, and it is addressed to an increasing number of researchers working in the field of low-energy interaction between matter and radiation, i.e., in widely understood light-matter interactions. The book explains how basic concepts (such as photons, vacuum fluctuations, wave-particle duality, etc.) arise, and how interactions between particles and photons must be described.

In chapter I authors begin with the classical electrodynamics description of the evolution of an ensemble of charged particles coupled to the electromagnetic field using the Maxwell-Lorentz equations in both real and reciprocal spaces. The normal variables for the field are introduced and their evolution studied. Chapter I concludes with short discussion of various possible quantization schemes.

Chapter II is devoted to the Lagrangian and Hamiltonian approaches to electrodynamics. The standard Lagrangian of classical electrodynamics is introduced and discussed in both real and reciprocal spaces. The Coulomb gauge is used to eliminate redundant variables, and the Coulomb gauge electrodynamics is formulated. The canonical quantization scheme is applied to quantize the field.

In chapter III a general presentation of quantum electrodynamics in the Coulomb gauge is given. The observables and states of the quantized free field are discussed, and the notions of photons, vacuum fluctuations, and coherents states are introduced.

Chapter IV is devoted to a presentation of other equivalent formulations of electrodynamics. The authors show how, applying a unitary transformation to the standard representation in the Coulomb gauge, one can obtain a different, although equivalent, formulation of electrodynamics. The Göppert-Mayer and Power-Zienau-Wooley transformations are thoroughly discussed, and the problem of the transition from  $\mathbf{A} \cdot \mathbf{p}$  to  $\mathbf{E} \cdot \mathbf{r}$  interaction Hamiltonian is clarified.

Finally, chapter V is an introduction to the covariant formulation of quantum electrodynamics. The difficulties that arise with quantization of the field with the Lorentz gauge and the ways to go out of them are shortly discussed.

Each chapter is supplemented by a number of Complements that are, in fact, independent sections of the book presenting additional and essential material. The last Complement to each chapter contains Exercises, a set of problems with the detailed solutions. The Complements form a rather important part of the book filling about one half of the volume. In the Complements one can find such things as the analysis of interference phenomena in the quantum theory of radiation, squeezed states of the radiation field, an elementary introduction to the electric dipole Hamiltonian, and the justification of the nonrelativistic theory in the Coulomb gauge starting from relativistic quantum electrodynamics, to mention only a few topics. *Photons and Atoms* covers a number of subjects that could be found only in the original articles before.

In my view, the choice of the material and its thorough presentation make *Photons and Atoms* an excellent textbook for those who start studying quantum optics or laser physics and a nice reference for those who already work in this field. Certainly, this is the book I would like to have at hand in my work.

R. TANAŚ.

# Foundations in Statistical Mechanics

Vol. 1: Equilibrium Theory

Vol. 2: Non-Equilibrium Phenomena

W. T. Jr. GRANDY

(D. Reidel Publishing Co., 1987 for Vol. 1 and 1988 for Vol. 2), Dfl. 165.00, \$ 79.00, £ 62.00.

These books are intended to provide an introductory course at graduate level. The first four chapters are essentially a revision of undergraduate material: probability theory, equilibrium thermodynamics and quantum mechanics. The «principle of maximum entropy» is introduced in the chapter on probability and it is used throughout both volumes. Although Grandy mentions, both in his introduction and later on in the book, that it is not universally well accepted in all the situations that he uses it, he does not explain why this is the case. This leaves the reader with a very subjective view of statistical mechanics, which is not desirable for a foundation course.

The first volume then progresses naturally from non-interacting particles to particles in an external field to interacting particles. It ends with a discussion of the phases of matter. Although the material presented will be of interest to the graduate physicist, the level at which it is written is somewhat uneven. The author introduces bosons and fermions as if they were new concepts, which they certainly shouldn't be for a graduate physicist. Similarly the introduction to chapter nine, « phases of matter », would seem more suitable for first year undergraduates.

The text as a whole appears to be a little cluttered with standard mathematical formulae, which could have been placed in an appendix, or just referenced to Abramowitz and Stegun. A case in point is the half-page on recursion relations for Bessel functions in the chapter on non-interacting particles.

The second volume begins with a chapter on classical hydrodynamics, necessary because it is the main worked example in the first part of this volume. The general theory of non-equilibrium phenomena is then set up in terms of the « principle of maximum entropy » ensemble. The first non-equilibrium calculations dealt with are those of a system driven from equilibrium by well defined perturbations. These are studied both theoretically and through some real examples. The steady state and the linear approximations are developed in order to make the non-equilibrium calculations tractable. Covariance functions, which are used extensively later on, are then introduced in the context of hydrodynamics. The remainder of the book considers specific examples of non-equilibrium phenomena in thermally driven systems, transport processes and sound propagation, all in terms of the maximum entropy description.

Throughout the books Grandy provides copious references, a practice that should be encouraged for books at this level. It is however unfortunate that he feels the need to abbreviate so many of his own phrases. In so doing he creates his own jargon and makes the use of the text as a reference more difficult.

In summary, this book is unlikely to become a standard work, although it may find a place as additional reading for the student interested in statistical mechanics.

Nicola K. WILKIN.

# A random walk through fractal dimensions

B. H. KAYE

(VCH, 1989) 421 p., DM 138.00.

Comme son nom l'indique, ce livre se propose d'aborder à travers une marche quelque peu aléatoire des aspects aussi divers que possible des caractéristiques, des exemples et des applications des géométries fractales.

Les deux premiers chapitres sont consacrés à la description de quelques exemples de courbes et surfaces fractales (naturelles ou mathématiques) et à l'introduction de définitions et techniques de détermination de dimensions fractales. On appréciera tout particulièrement la discussion détaillée dans le deuxième chapitre des problèmes qui peuvent apparaître quand on applique sans précautions ces techniques de détermination. Comme le rappelle l'auteur, on peut en effet, en s'y prenant avec suffisamment de foi, trouver une dimension fractale à tout objet (y compris un ensemble d'ellipses euclidiennes). L'auteur présente, entre autres, des déterminations de dimensions fractales à partir de techniques utilisant des outils de mesure de morphologie mathématique de type dilatation ou érosion qui sont intégrés dans un nombre croissant de systèmes d'analyse d'images.

Le chapitre 3 présente (avec, comme fil conducteur, l'idée de démontrer l'utilité pratique et l'applicabilité des approches fractales) des résultats expérimentaux de mesures effectuées sur des milieux granulaires, des poussières et des agrégats. Ces problèmes correspondent d'ailleurs aux intérêts de chercheur de l'auteur. Si certains exemples sont abordés trop brièvement à travers une photo et un diagramme log-log, d'autres donnent lieu à une discussion détaillée et analysent la liaison entre paramètres physiques et géométrie fractale : attaque d'un grain métallique par un acide, croissance d'un dépôt électrolytique sous différents voltages, relation entre dimension fractale et danger des particules inhalées. Des exemples intéressants pris sur des fragments de processus de broyage ou d'éclatement sont également indiqués.

L'auteur aborde ensuite dans les deux chapitres suivants l'analyse de structures fractales engendrées par des processus de marche ou de tirage aléatoire. Les exemples utilisés comprennent aussi bien la marche aléatoire à une dimension (évolution du déplacement global en fonction du temps), les profils de rugosité d'une roche fracturée, la limite d'un panache turbulent que des processus à deux dimensions : structure de dendrites, croissance de dépôts électrolytiques, formation d'amas de particules. Une part importante est donnée dans le chapitre 5 à la description mathématique détaillée des amas fractals ainsi qu'à la structure des figures de diffraction d'images de structures fractales à deux dimensions. Ce chapitre se termine par l'analyse de deux modèles statistiques importants qui donnent naissance à des géométries fractales : la percolation et l'agrégation limitée par diffusion.

Les derniers chapitres sont consacrés respectivement à la recherche de systèmes physiques de propriétés similaires à celles du tamis de Sierpinski, à la discussion de l'existence et de la caractérisation de volumes poreux de géométrie fractale à la structure fractale des fronts d'invasion instables de poreux par des fluides peu visqueux et, enfin, de la structure fractale des fronts de fractures et de systèmes fragmentés. Les discussions sur ces derniers sujets sont quelque peu rapides, particulièrement en ce qui concerne l'invasion diphasique. Il s'agit en effet d'un des problèmes pour lesquels l'approche fractale a été la plus élaborée et la mieux reliée aux phénomènes physiques : une discussion plus complète aurait été souhaitable (signalons par exemple les résultats d'auteurs tels que Stanley, Daccord et Jossang sur les caractéristiques multifractales du processus de DLA). La marche au hasard de l'auteur à travers les fractales se conclut enfin par un bref inventaire « à la Prévert » incluant les estampes japonaises, les îles de la mer Egée, la côte de Terre-Neuve, un mélange de cailloux et de graviers...

Globalement, ce livre se distingue très nettement de la plupart des ouvrages publiés sur le même sujet car ce n'est ni un simple recueil d'images, ni un livre insistant sur les propriétés mathématiques des structures fractales. (Une bonne part de ces dernières ne sont guère utiles au physicien dont les systèmes ne peuvent avoir de caractéristiques proches des courbes ou surfaces fractales que sur une gamme finie d'échelles de longueur : ce ne sont donc jamais des fractales au sens strict mathématique du terme).

On appréciera tout particulièrement la présentation concrète par l'auteur des techniques de détermination pratique des dimensions fractales de systèmes réels et des problèmes qu'elles posent. Comme l'indique l'auteur dans sa préface, c'est un livre de « nuts and bolts » (écrous et boulons) sur la géométrie fractale appliquée : c'est ce qui fait son originalité et son intérêt. Il intéressera tout particulièrement les scientifiques qui cherchent comment caractériser en pratique telle ou telle structure rugueuse ou tel ou tel agrégat de particules par une approche de type géométrie fractale.

On pourra seulement regretter (mais c'est encore à l'heure actuelle une question des plus ouvertes) que le livre ne donne que peu de résultats quantitatifs sur la relation entre la dimension fractale et les autres paramètres physiques caractérisant la structure du matériau ou importants d'un point de vue pratique. Le chapitre 3 intitulé « à quoi servent les fractales » discute en effet surtout de manière quantitative divers problèmes (surtout de sécurité) liés à la présence de fines particules dans l'atmosphère. On pourra également regretter que le livre n'aborde que peu les approches (multifractales par exemple) qui cherchent à aller plus loin que la simple détermination d'une valeur de géométrie fractale. Alors que la partie analyse des matériaux granulaires est complète et bien détaillée, les aspects « structure des écoulements dans les poreux » qui représentent un des exemples qui ont été analysés le plus complètement ne sont que très brièvement analysés.

Remarquons par ailleurs qu une abondante bibliographie est fournie à la fin de chaque chapitre aussi bien en ce qui concerne les fondements théoriques des résultats présentés qu'en ce qui concerne les détails de ceux-ci. L'auteur cite également des types et des marques d'équipements.

En conclusion, le livre de B. H. Kaye est original, très bien documenté et donne des fractales une vision d'expérimentateur critique et réfléchie. Il nous paraît indispensable à tous ceux qui s'intéressent à l'application pratique et expérimentale de ces modèles; il donne de ces problèmes une vision juste et concrète qu'on ne trouvera pas dans d'autres ouvrages plus inconditionnels et/ou plus superficiels.

J. P. HULIN.

#### Quantum Mechanics II; a second course in quantum mechanics

R. H. LANDAU

(Wiley, 1990) 482 p., £ 39.30.

This book represents a general one year graduate course in quantum mechanics for students who will have already studied a standard text such as Merzbacher. The style is informal and often, when working through it, I could imagine the author giving the lectures upon which it is based.

Though designed to be suitable for a wide range of physics students, its flavour tends to be that of the particle physicist, with most space being devoted to two-particle scattering theory.

The book divides naturally into three parts. The first of these treats non-relativistic scattering theory, mainly from the standpoint of integral equations, up to and including the interactions of spin-half

particles with a fixed potential. It is well known that most books on scattering theory and the operator formulation of the Lippman-Schwinger equation are (with the possible exception of Joachim) rather daunting to the students. However, in setting up and solving the equations, the author stresses the underlying Physics. This, together with the fact that the work is only an introduction to the subject rather than a work of reference, make it far more palatable to the reader. At the end of this section there are two short chapters on many body problems with discussions of the Hartree-Fock and Thomas-Fermi equations.

The second part is concerned with relativistic potential scattering. Again, this is no substitute for a specialist tome, e.g. Bjorken and Drell, but rather an introduction for the student who first needs to acquire a broader perspective. Of especial interest here is the chapter on how to solve the Dirac, as well as non-relativistic equations, numerically in momentum space.

The last section introduces quantum field theory, using mainly old-fashioned perturbation theory and, where possible, avoiding lengthy proofs. In this way the author could get as far as discussing the Bethe-Salpeter equation for two-particle scattering.

Given the large number of good volumes on quantum mechanics, is there a market for this new one? There might be when the book is recognised as giving a user-friendly overview of a wide area of physics before the student launches into more refined but narrower texts.

Colin WILKIN.

# Acoustic and Electromagnetic Waves

D. S. JONES

(Clarendon Press, Oxford, 1989) 745 p., £ 25.00.

This is an unchanged reprint, in paperback form, of the well-known treatise which first appeared in 1986. The basic equations for the propagation of sound and light waves are very similar, leading to analogous phenomena of reflection, refraction and diffraction in the two cases. The major difference between the two is that the former deals with the propagation of scalar fields, whereas in the latter case they are of vector nature. In addition though, Maxwell's equations are exact in vacuum whereas in a medium, to which acoustic disturbances are always confined, both become effective theories.

In this book, Jones develops the two theories in parallel, thus showing the strong similarities and allowing the formalism and approximation techniques learned in one area to be used in the other. As an example, Babinet's acoustic principle is first derived to be followed immediately by the equivalent theorem in electromagnetism, with its associated complications due to the polarisation of the light.

Though the equations of acoustic and electromagnetic waves are derived in the first chapter, it is assumed that the reader has familiarity with the formalism of fluid dynamics and Maxwell's equations respectively. The book is thus aimed at final year undergraduate or research students in physics or engineering. The whole approach is heavily theoretical with 50 pages of mathematical appendices.

Relativity is of course the crucial difference between the two theories, so that this is treated extensively in a special chapter. In the case of the propagation of light in vacuum there is no preferred reference frame and this leads to the covariance of the theory under Lorentz transformations. On the other hand, sound needs a medium in which to propagate, which gives rise to an obvious definition of an inertial system.

The author only discusses classical macroscopic physics, though the similarity between the theories persists in quantum mechanics with photons and phonons.

The approach presented here is innovative although the number of students who will be capable of benefitting from both facets of the material might be relatively small. Nevertheless it should remain a valuable standard reference for many years.

Colin WILKIN.