On scattering from fractal lattices

HARJINDER SINGH* and P K CHATTARAJ

Theoretical Chemistry Group, Department of Chemistry, Panjab University, Chandigarh
160 014, India

Abstract. Gas-surface scattering is speculated as a meaningful problem for understanding
the physics of fractals. Fractal behaviour can be associated with a self-similar geometry on
a solid surface. The interaction potential for a gas atom or molecule approaching the
lattice depends primarily on local factors but a parametric dependence of the cross-section
data on the fractal dimension can be conceived. Such a dependence on the self-similar
character of a multi-centred target is more explicit when multiple scattering is included.
Application of approximation schemes like the previously developed average wavefunc-
tion method to this problem is suggested.

Keywords. Fractal; scattering; gas-surface.

1. Introduction

Leo Kadanoff, in a recent article (Kadanoff 1986), wrote that '. . . the physics of
fractals is waiting to be born'. Heterogeneous reaction dynamics is an area where
the physics is expected to have a direct correlation with the fractal behaviour
(Mandelbrot 1983; Pfeifer and Avnir 1983) of a surface lattice. Not surprisingly,
among the first problems in chemical physics, where the relevance of fractals is
seen, is that of understanding the catalytic properties of solid surfaces (Meakin
1986). The importance of gas-surface scattering in the understanding of processes
occurring on surfaces is well-known (Singh and Deb 1986). A major problem that
remains unresolved is the physics of scattering from disordered surfaces. The
many-body and abruptly discontinuous nature of an interface presents serious
difficulties. The characterization of a random system usually requires large-scale
computational exercise. The fractal dimension is a possible way out (Mandelbrot
1983). We suggest here a problem based on this possibility.

A completely random fractal, e.g., the Brownian fractal fills up the space
completely. The dimension of such a fractal is known to be that of the euclidean
space (d) that it is embedded in. The degree of disorder in a system, say, a solid
surface, can be characterized in terms of a fractal dimension (d_f) provided a
self-similar behaviour is obtained at a macroscopic level. This is a feature that
should be utilized to explore the physics of surface phenomena. In theoretical work
involving numerical calculations, one has to work with a finite fractal. However,
the dimension d_f can still be well-defined, as long as the self-similarity properties
remain understood. For a gas-surface scattering problem, one can use it as a
description of the relevant geometry and the interaction potential on the solid

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*To whom all correspondence should be addressed.

surface. Thus it is sensible to explore the utility of fractal dimension as a parameter describing the dynamical information inherent in, say, the angular distribution of scattering cross-section. This structure and the dynamics utility of fractal character is well-studied for the case of proteins (Colvin and Stapleton 1985; Helman *et al* 1984; Liu 1985; Wagner *et al* 1985). A detailed stochastic description of a surface that can be extrapolated to all scales has been seen to be related to the fractal dimension for ocean topography (Ausloos and Berman 1985) Montroll and Schlesinger (1983) showed the formal connection between the fractal description of dynamics and other alternatives, namely, the maximum entropy formalism and various distribution functions giving statistical descriptions.

Following a general discussion in §2, we present in §3 the application of the average wavefunction method (AWM) to the scattering from fractal lattices. A number of problems arise in the formulation even at the simplest level for a diagnostic exercise. These are discussed in §4 with brief remarks on the direction of this work.

2. The problem and the methodology

Consider a structureless particle interacting with a solid surface. The target is, by its very nature, a multi-centred one and the interaction potential between the particle and the target is represented by $V^{d_f}(r)$, which has the information about the geometry of the surface. In other words, it has in it the locations of all the atomic sites on the surface. Hence it is meaningful to label it with the parameter d_f. The dimension d_f can be defined in terms of an appropriate scaling property. For example, one may define d_f from $R \propto N^{1/d_f}$, where N is the number of atomic sites in a circle of radius R. Such a potential can also describe approximately, an atom interacting with adsorbate atoms on the surface. The co-ordinate r has all the degrees of freedom in it. Some of the degrees of freedom can be removed depending on the nature of the problem. Realistic systems like fume silica, graphite, crushed glass and colloidal systems are known to exhibit fractal behaviour (Pfeifer and Avnir 1983) and an interaction potential like the one discussed here can be used for these cases.

In any case, the scattering process is described by the following Schrödinger equation,

$$[-(\hbar^2/2m)\nabla^2 + V^{d_f}(r)]\psi(r) = E\psi(r), \tag{1}$$

where m is the mass of the incident particle and E is its energy. The parametric dependence of the interaction potential on the fractal dimension is meaningful even if the surface lattice is not a fractal, except that then $d = d_f = d_T$ (topological dimension). The last equality may not be true for some specific models, e.g., if all the atoms are assumed to be located on a curve coiling like a wire on a surface.

Equation (1) can be written in the integral (Lippmann–Schwinger) form,

$$\psi_k^{+d_f}(r) = \phi_k(r) + \int G_+(r,r')V^{d_f}(r')\psi k'(r')\,dr', \tag{2}$$

where $\phi_k(r)$ is the free-space (or appropriately distorted) plane wave with the incoming wavevector k (given by $\hbar^2 k^2/2m = E$) and $\psi_k^{+d_f}(r')$ is the outgoing scattered wavefunction with the wave-vector k'. The Green's function $G_+(r,r')$

corresponds to the potential corresponding to $\phi_k(r)$, [zero for a plane wave]. The relevant quantity of interest is the T-matrix element,

$$T^{d_f}(\mathbf{k}', \mathbf{k}) = (\phi_{\mathbf{k}'} | V | \psi_{\mathbf{k}}^{+d_f}),$$

(3)

and observable quantities like the differential or total cross-sections follow from the T-matrix. The central question we wish to address is whether the labelling by d_f is appropriate, i.e., whether any memory of the fractal behaviour of the lattice geometry is retained after the dynamics is over. Equation (3) shows that this should be the case though the nature of the dependence of the final results on d_f is not clear.

Thus, analysis of cross-section data from different fractal lattices can be conceived as a feasible way to explore fractal physics. A simple observation follows from (3) immediately. For a potential $V(r)$ approximated by a sum of pairwise interactions,

$$V(\mathbf{r}) = \sum_i v_i(\mathbf{r} - \mathbf{r}_i),$$

(4)

to retain effects arising from the geometry, one has to include multiple scattering. Thus, any approximation scheme designed to evaluate the cross-sections must take this into consideration. For instance, the first order Born approximation cannot be used for this purpose, since then the interaction in (4) gives a sum of single site scattering effects. This is not necessarily true for cases where the pairwise interaction approximation is not used. However, the analysis then becomes more involved, both conceptually and numerically.

3. Average wavefunction method

In the (AWM) approximation scheme, the atom-surface scattering is represented by a potential, which can be written as a sum of two parts. One part is a strong surface potential $V_0(r)$, depending only on the normal distance to surface, the second component $V_1(r)$ is a weaker, short-range corrugation potential that reflects the atomic structure of the topmost layer of the surface. This is taken to be of the form

$$V_1(\mathbf{r}) = \sum_i v_i(\mathbf{r} - \mathbf{r}_i),$$

(5)

Assuming that the dynamics due only to $V_0(r)$ can be solved, we have the Lippmann–Schwinger equation,

$$\psi_{\mathbf{k}}^{+}(\mathbf{r}) = \chi_{\mathbf{k}}^{+}(\mathbf{r}) + \sum_i \int G_+(\mathbf{r}, \mathbf{r}') v_i(\mathbf{r}' - \mathbf{r}_i) \psi_{\mathbf{k}}^{+}(\mathbf{r}') \, d\mathbf{r}',$$

(6)

where $\chi_{\mathbf{k}}^{+}(\mathbf{r})$ are the scattering states of the potential $V_0(\mathbf{r})$; $G_+(\mathbf{r}, \mathbf{r}')$ is the outgoing Green's function of the Schrödinger equation with potential $V_0(\mathbf{r})$. In the AWM scheme, one approximates $\psi_{\mathbf{k}}^{+}(\mathbf{r})$ as,

$$\overline{\psi}_{\mathbf{k}}(\mathbf{r}) = \chi_{\mathbf{k}}^{+}(\mathbf{r}) + \sum_i \lambda_i \gamma_i \int d\mathbf{r}' \, G_+(\mathbf{r}, \mathbf{r}') \, v_i(\mathbf{r}' - \mathbf{r}_i) \omega_i(\mathbf{r}'),$$

(7)

where $\gamma_i = \int d\mathbf{r} \omega_i^*(\mathbf{r}) v_i(\mathbf{r} - \mathbf{r}_i) \overline{\psi}_{\mathbf{k}}(\mathbf{r})$.

(8)

and $\lambda_i^{-1} = \int dr\,|\omega_i(r)|^2\,v_i(r-r_i)$,   (9)

and $\omega_i(r)$ is a weighting factor. In the zeroth order of the approximation, it is taken to be $\chi_k^+(r)$. The reason for this choice is explained in our earlier work (Singh *et al* 1986a, b)

The T-matrix is obtained from

$$T(k',k) = T_0(k',\,k) + T_1(k',\,k),\tag{10}$$

where $T_0(k',k) = (\phi_{k'}|V_0|\chi_k^+)$,   (11)

and $T_1(k',\,k) = (\chi_{k'}|\bar{V}|\bar{\psi}_k)$,

the potential, $\bar{V}$ represents the AWM approximated non-local, separable interaction.

4. Further developments

The application of AWM to model problems (Singh *et al* 1986b, c) has shown that it is a useful time saving approximation, at least for a better qualitative understanding of scattering from a solid surface. For this work, we have used one-dimensional fractal lattices ($d_f = \log 2/\log 3$; $\log 3/\log 5$) with no background potential ($V = 0$; non-zero hard-wall like or more realistic ones being the next step). The preliminary set of calculations are being performed on a set of 16 and 18 atoms respectively with the above mentioned fractal dimensions. The T-matrix shows the following behaviour for spherically symmetric Gaussian repulsive potential at each atomic site.

$$T(k',k) \propto \exp(-\Delta k^2/2)\,\mathrm{erf}(i\Delta k/\sqrt{2}) \cdot \sum_i \gamma_i \exp(i\Delta k \cdot r_i),\tag{14}$$

where $\Delta k = k' - k$, $\mathrm{erf}(z)$ is the error function in z, γ_i is defined in (8) and r_i in (5). The arguments of the first exponential and the error function in (14) are scaled appropriately with respect to the variance of the Gaussian, which is taken to be identical at each site. Even at this simple level, a number of technical as well as conceptual problems arise. First, a sizable computational facility is necessary (a lattice of N atoms requiring an inversion of a matrix of size $N \times N$ for every set of differential cross-section data). Though simplistic, these lattices are expected to show trends that can be investigated (for fractal behaviour) for larger lattices. The calculations done so far have shown distinct features in the cross-section for the two different lattices. More work needs to be done, however, to reach a conclusive stage (Singh and Chattaraj 1987).

5. Conclusion

We presented in this paper a possible way to explore the relevance of fractal behaviour in scattering from solid surfaces. The average wavefunction method is chosen to ensure that multiple scattering effects are taken care of while preserving the unitarity of the Hamiltonian as well as to have analytical expressions that can be evaluated without much labour. However, even at a simple level, the calculations

are reasonably large. Still, we hope to gain some directions towards further research in understanding the physics of fractals. Increasingly more work is being done in chemical physics to explore the significance of fractal behaviour in determining the physics and chemistry of a system rather than merely measuring the dimension.

Though no rigorous analysis of how the fractal behaviour is retained in a dynamical output has been presented here, we wish to explore this in our future work. In particular, an interaction potential with an analytical fractal behaviour can be conceived (Mandelbrot-Weierstrass interaction?) and questions can be asked on what happens to this function after a non-linear transformation of the Lippmann–Schwinger kind.

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