Chapter 7
Conclusions

In this thesis I have attempted to relate the structural, thermodynamic and dynamic behaviour of simple atomic clusters to the underlying PES and the form of the potential. This has led to the following accomplishments:

- I have created a comprehensive structural database cataloguing the effects of the range of the potential. It should provide candidate structures for comparison with the indirect structural information available from experimental studies of clusters in molecular beams, and for theoretical studies using computationally expensive methods for which comprehensive searches of the PES are not feasible.[*] It is also an ideal system to benchmark global optimization methods.

- I have developed a statistical method which can accurately reproduce the thermodynamic properties of clusters from a sample of minima, and which provides insight into the respective roles of different regions of configuration space.

- I have provided a microscopic understanding for the destabilization of the liquid phase that occurs for short-ranged potentials.

- I have shown that the structure of the liquid-like state of small clusters has many of the characteristics of the bulk liquid, and that this structure evolves through a rapid increase in the disclination density with size.

- I have suggested an explanation for the transition from electronic to geometric magic numbers observed in mass spectroscopic studies of sodium clusters.

[*] For this purpose, the points files of all the global minima will soon be made available on the world wide web at URL <http://brian.ch.cam.ac.uk/ jon/jon.html>

- I have characterized some of the features of a model PES which play key roles in aiding or hindering relaxation to the global minimum.

The work presented here also suggests a considerable number of applications, extensions and fresh challenges. As is often the case, the more we know the more we realize our ignorance.

- The predictions for the structure of neutral clusters of C_{60} molecules presents both a challenge to experiment to probe the neutrals through techniques, such as electron diffraction, and a challenge to theory to quantitatively understand the differences between neutral and charged clusters.

- The range of applicability of the superposition method could be extended by incorporating a 'multi-histogram' approach to combine the information from samples of minima produced by systematic quenching at different energies.

- There are many possible extensions to the work relating the dynamics to the PES, since this field is, in many ways, still in its infancy. There is the application of the master equation method to the M_{13} data that we have obtained, and the development of statistical methods to accurately model the dynamics when only an incomplete sample of stationary points is available, as is inevitable for all but the smallest sizes. In this thesis, the aspect of the dynamics upon which I have focussed most attention is relaxation to the global minimum, but there are many other possible research avenues. One area of particular promise is the dynamical properties of supercooled liquids[287,288], including such topics as self-diffusion and the structural origins of the α and β mechanisms of relaxation[272,289].

- The unusual structural (Chapter 2), thermodynamic (§2.4) and dynamic (§6.4) properties of LJ_{38} and LJ_{75} make them particularly attractive cases for more detailed study. Such an investigation could provide insights into why global optimization methods can fail, and possibly even suggest improvements to these methods to help circumvent such difficulties.

Although this thesis has concentrated on the properties of clusters, the ubiquity of PES's means that some of the ideas contained here are relevant to a much wider domain. For example, in Chapter 5, I attempted to make detailed connections between the behaviour of clusters, proteins and glasses. Furthermore, I hope the allusions and asides to seemingly unrelated subjects, such as polytopes in 4D space, the optimal shapes of the bubbles in an idealized head of beer and the heptagonal barrel-shaped chaperones that assist the folding of proteins, have illustrated the rich and interdisciplinary nature of research into clusters. Moreover, I hope this thesis has shown the utility of simple models that capture aspects of the essential physics in providing a framework for understanding a diverse range of behaviour. Indeed, the complexity that emerges from these simple models is quite remarkable. Do complex systems, such as these, herald, as some have suggested, the dawn of a post-reductionist science? Certainly, a cluster is much more than just a collection of atoms.