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Report on the Doctoral Dissertation of Ms Sangami Ganesan-Santhi

The doctoral dissertation “Interactions and cold collisions involving AlF molecules” presents a substantial and coherent body of work that addresses an important and timely topic in the field of ultracold molecules. AlF has recently emerged as one of the most promising candidates for direct laser cooling and the creation of ultracold dipolar gases, and the theoretical foundations explored in this thesis are directly relevant to ongoing experimental efforts. The thesis combines rigorous ab initio electronic-structure calculations with scattering theory to investigate the interaction properties of AlF with He, with another AlF molecule, and with Rb. These systems are central to understanding buffer-gas cooling, evaporative cooling, and atom–molecule collision processes, and the new results contained in the dissertation will be of considerable value for future work in the field.

The thesis is generally well written, clearly structured, and easy to follow. Only minor linguistic and formatting issues remain, none of which distract from the clarity or scientific content. The figures are for the most part of high quality, with well-prepared captions that reach the standard of journal publications, with only isolated formatting inconsistencies (such as in Fig. 6.18). The inclusion of short summaries at the end of each section is helpful for guiding the reader through the work, although some of these summaries could have been expanded to place the results in the broader context of collisional cooling and ultracold-molecule research. In particular, the implications of the deep global minimum found in the AlF–AlF potential for evaporative cooling and the likely importance of inelastic channels would have benefitted from a brief discussion, even if full scattering calculations lie beyond the scope of the thesis.

The introduction provides a broad overview of ultracold physics, production methods for molecules, and the applications of ultracold systems, followed by a longer discussion of electronic-structure methods. Ms Ganesan-Santhi demonstrates a strong command of the relevant literature and has clearly studied an extensive body of work. The introduction is comprehensive and informative, but at times reads more like a series of individual descriptions than a fully integrated narrative. Nevertheless, it remains well organised and accessible, and the presentation of the research aims is concise and clear. The concluding chapter could have referred back to these aims more explicitly, but the overall structure of the thesis is sensible and coherent.

The chapter on methods summarises the essential ideas of ab initio electronic-structure theory and scattering theory in a clear and accessible manner. The discussion is well judged and provides sufficient detail for a reader who is not a specialist in quantum chemistry. As an experimental physicist myself, I found this chapter particularly useful as a compact and accurate refresher of the theoretical tools used throughout the thesis. It also demonstrates that the candidate has gained a solid understanding of both the approximations and the practical considerations that determine the reliability and convergence of high-level

computational methods.

The main scientific results are presented in four chapters. The first concerns the electronic structure of AlF itself, including ground- and excited-state potential energy curves and molecular properties. The results are clearly presented and the improvement of the potentials with increasing methodological sophistication is well documented. It would have been even more valuable to include a direct comparison between the ab initio potentials and those inferred from high-resolution spectroscopy, beyond the brief reference to the experimentally determined separation of the A-X potentials, but this is minor.

The chapter on AlF–He interactions is very strong. Ms Sangami provides accurate potential energy surfaces for the ground and excited electronic states and presents scattering calculations that directly address the feasibility of buffer-gas cooling. The analysis of elastic and inelastic collision cross sections is physically well grounded. This work has already been published in a peer-reviewed journal, indicating that both the methodology and the results meet high scientific standards.

The chapter on AlF–AlF interactions presents the first four-dimensional potential energy surface for this system. This is an impressive and technically demanding calculation, and the resulting surface will be highly relevant to future studies of evaporative cooling and the dynamics of dense AlF samples. The identification and characterisation of a deep global minimum is an important finding. Although the thesis does not attempt a full analysis of the collisional implications, even a qualitative estimate of the likely impact on elastic and inelastic collision rates would have strengthened the discussion. Nonetheless, the work is thorough, carefully executed, and represents a significant contribution.

The final results chapter provides two-dimensional potential energy surfaces for the interaction between AlF and Rb in several electronic states. These results are original and timely, given current interest in atom–molecule mixtures and magnetically tunable Feshbach resonances in such systems. The short-range anisotropy observed in the potentials is likely to play an important role in the resonance structure, and it is notable that these results are already being used in ongoing collaborative work.

Taken together, the scientific results in this dissertation constitute a coherent and valuable contribution to the field of ultracold molecules. The work shows that Ms Ganesan-Santhi can work independently, execute advanced computational tasks with care and precision, and interpret the resulting physics appropriately. Her calculations provide theoretical foundations that will be immediately useful for forthcoming experiments on AlF cooling and trapping, and it is clear that she has acquired a strong technical skillset that will allow her to contribute effectively to future research.

In conclusion, I evaluate the dissertation positively. It presents new and important results, is written to a high standard, and meets the criteria for a doctoral thesis. Its scientific quality corresponds to the good–average level expected at my institution. I recommend that Ms Sangami Ganesan-Santhi be admitted to the subsequent stages of the doctoral proceedings.

Sincerely,