Peer Review Report

1. General Overview and Recommendation

The manuscript presents a thorough, pedagogically sound, and visually enriched examination of the quantum mechanical principles governing molecular rotations, vibrations, and their coupling. It is **well-structured**, **mathematically rigorous**, and **computationally substantiated**, offering value to both didactic and research-oriented readers.

Recommendation: *Minor Revisions*

While highly commendable in scope and clarity, a few minor aspects warrant revision before acceptance in an average-tier peer-reviewed journal.

---

2. Title, Abstract, and Keywords

- **Title:** The title accurately reflects the scope and content, although it is slightly long. Consider shortening it for indexing purposes in average journals (e.g., removing "A Comprehensive Analysis...").
- **Abstract:** Exceptionally well-written. It outlines the main models (rigid rotor, harmonic oscillator), highlights centrifugal distortion and Born–Oppenheimer breakdown, and ends with relevance to quantum technologies. No revision needed.
- **Keywords:** Highly appropriate for searchability and thematic indexing.

✅ *No action required.*

---

3. Introduction

The introduction is excellent. It:

- Builds historical continuity (Dennison, Wilson, Pekeris),
- Clarifies foundational models and approximations (Born-Oppenheimer, rigid rotor, harmonic oscillator),
- Establishes practical relevance (quantum control, spectroscopy),
- Clearly states the paper's aim.

✅ *No revision needed.*

---

4. Methodology

This section is **exemplary for a pedagogical manuscript**:

- Begins with classical-to-quantum derivations (rotational Hamiltonian, vibrational potential),
- Includes **detailed mathematical transitions**, e.g., harmonic oscillator reformulated in dimensionless variables,
- Introduces realistic corrections (Morse potential, centrifugal distortion, B_v dependence),
- Includes **selection rules**, a critical aspect often omitted in teaching-level reviews,
- Ends with a competent **computational approach**, suitable for DFT-level simulations of rovibrational systems.

✅ Strengths:

- Clear, progressive derivations.
- Proper use of classical foundations leading into quantum descriptions.
- Good referencing of canonical works.

🛠️ Minor suggestion:

- The Morse potential could benefit from a comparison of *experimental versus theoretical* dissociation energies for a common molecule (e.g., HCl) to emphasise its empirical usefulness.

---

5. Results and Analysis

This section is **very strong and visually enriched**:

- **Figure 1–5** (pp. 12–19): Present *rotational levels*, *centrifugal distortion*, *harmonic vs. anharmonic*, *rovibrational couplings*, *spectral transitions*, and a *3D potential energy surface*. These are **rare in average-level submissions** and highly valuable for readers.

🛠️ Suggestions:

- A few figures could benefit from **axis unit clarifications** (e.g., eV vs cm^{-1} in some vibrational graphs).
- Consider explaining more explicitly that **Figure 3's right panel** is based on experimental B_v data fitted to α_e, not ab initio calculations.

✅ Computational simulations are solid and based on well-defined molecular parameters (CO, HCl), showing transitions, energy level spacings, and selection rule consequences.

---

6. Discussion

This section is extensive and divided into **seven meaningful subsections**, covering:

- Theoretical and practical advantages of quantum mechanical treatment,
- Limitations of harmonic and rigid rotor approximations,
- Applications in quantum control and spectroscopy,
- Educational value,
- Technological applications (quantum computing, sensing),
- Future directions.

🛠️ Suggestions:

- Some sections (especially 4.3 and 4.7) could benefit from clearer **segmentation between theoretical and computational perspectives**. For instance, the discussion on ultracold molecules is largely experimental but tied to theoretical treatments—clarify which predictions are computational vs. experimental observations.
- 4.6 (Pedagogical) is strong, but adding a reference to an actual curriculum or course structure (e.g., undergraduate molecular spectroscopy syllabus) would elevate its utility for teaching staff.

---

7. Conclusion

A **lucid and cohesive summary**:

- Reiterates all critical elements of the paper,
- Highlights the robustness of basic models (Bv dependence, anharmonic corrections),
- Acknowledges limitations (Born-Oppenheimer approximation, centrifugal distortion at high J),
- Reconnects theory to computational predictions and visual outputs.

✅ No changes necessary. It convincingly synthesises the manuscript's purpose.

---

8. Attachments and Python Code

The **Python code** (pp. 30–36) is clean, readable, and annotated, using:

- `matplotlib`, `numpy`, `seaborn`,
- Diagrams of rotational levels, Morse potentials, rovibrational couplings, and 3D surfaces,
- Spectral simulations with Boltzmann factors and selection rule compliance.

✅ Strengths:

- Reproducible, compact, and educational.
- Visuals produced are of publication quality.

🛠️ Minor technical suggestion:

- Consider including a `.notebook` version (`.ipynb`) in addition to the script, especially if the journal accepts supplementary materials.

---

9. References and Acknowledgements

The references are **well-selected**, classic (Dennison, Schrödinger, Wilson) and modern (Jacob & Reiher, Koch et al., Schrader). APA style is followed with DOI links present.

✅ Suggestions:

- Reference saturation is high; that's good for didactic purposes but might be trimmed if required by length constraints.

---

10. Suitability for Average-Level Periodical

This manuscript significantly **exceeds the average standard** for many peer-reviewed teaching-oriented periodicals and is close to upper-tier review level due to:

- Depth of theory,
- Breadth of coverage,
- Strong computational reproducibility,
- High-quality visual materials.

However, its **pedagogical clarity** and **modular structure** still make it well-suited to average readership, especially in journals focusing on molecular quantum chemistry, spectroscopy, or chemical education.

Final Remarks

Overall Evaluation: ★★★★☆ (4.5/5)
Recommendation: *Minor revisions – Ready for publication after minimal improvements*

This manuscript is a **model of clarity, comprehensiveness, and computational support** for a topic central to molecular quantum chemistry. It will serve both as a reference and a teaching tool. With a few small refinements in figure annotation and theoretical-to-computational transitions, it will be an excellent addition to the literature.
