Quantum systems in time-dependent fields — We have derived new theoretical results for transition probabilities in quantum systems in time-dependent electromagnetic fields. These results challenge Dirac’s expression for the transition probability, which depends on the norm-square of the coefficient $c_k(t)$ for the excited state $|k_0\rangle$ of the original unperturbed Hamiltonian $H_0$, in the time-dependent wave function. By integrating by parts, Landau and Lifshitz separated $c_k(t)$ into an adiabatic term $a_k(t)$ that follows the adiabatic theorem of Born and Fock, and a nonadiabatic term $b_k(t)$ that depends on the nonadiabatic term of the perturbation up to time $t$. The adiabatic term describes the adjustment of the initial state to the perturbation without actual transitions, while the transition probability is given by the norm-square of $b_k(t)$. Our work reinforces this statement and goes beyond Dirac’s expression for the transition frequency to state $k$; while the opposite is true when $\omega < \omega_0$. These results are independent of the phase of the oscillating wave relative to the peak of the Gaussian envelope. The differences are also quite stark for a perturbing pulse that rises to a level plateau, and later falls off. While the perturbation is constant, the nonadiabatic transition probability is constant as required physically, since a static perturbation cannot induce transitions; in contrast Dirac’s form of the transition probability continues to oscillate while the perturbation is constant, as shown in Figure 1.

Collision-induced spectroscopic processes — Spectroscopic processes that are forbidden for single molecules are observed in dense gases and liquids, because the electronic charge distorts during molecular collisions. Our work has focused on collision-induced absorption in the IR by $\text{H}_3^+$ gas. $\text{H}_2/\text{H}_3^+$ and $\text{He}$ mixtures, $\text{N}_2$ gas and $\text{O}_2$ gas, with applications in astrophysics and in atmospheric profiling. Interaction-induced absorption affects the radiative profiles of gases in star-forming nebulae; very old, very cool white dwarf stars; the outer planets and exoplanets that are termed “hot Jupiters” and “warm Neptunes,” and the atmospheres of satellites of the outer planets. My research group calculates the total dipole moments $ab\textit{ initio}$ and then expresses the results in spherical-tensor form for subsequent line-shape calculations. Dipole surfaces for $\text{H}_2/\text{H}$ are shown in Figure 2. This research area involves collaborations with Lothar Frommhold and Martin Abel (University of Texas, Austin), Magnus Gustafsson (Luleå, Sweden), Tijs Karman, Gerrit Groenenboom, and Ad van der Avoird (Nijmegen, the Netherlands), and Richard Dawes (Missouri University of Science and Technology).