We have been using quadrupole mass spectrometers to generate beams of mass-selected ions, which are deposited into growing neon matrices at 5 Kelvins for subsequent analysis by vibrational spectroscopy. Deposition times up to 24 hours are often required to accumulate a sufficient number of ions for detection. This depends on the absorption coefficients of the infrared transitions, which are normally not known. We have been attempting to correlate infrared intensities obtained from ab initio calculations with experimental observations, so that systems can be selected which are most amenable to successful detection of the ions deposited. How should ab initio calculations be performed such that the predicted intensities correlate with experimental observations? To answer this question, calculations employing a variety of basis sets are being evaluated for various odd- and even-electron cationic species. Exemplary computations involving ions containing all heavy atoms, such as the molecular ion of carbon disulfide, as well as a variety of organic cations, will be described.