Large molecules, molecules with about 15 or more heavy atoms, have high angular momentum quantum states populated even at the low temperatures produced by seeded pulsed jet expansion. The rotational spectroscopy of these molecules is often more easily described using semiclassical energy expressions and selection rules. One prominent feature of the rotational spectra of molecules in the semiclassical limit is a set of strong, equally spaced transitions that correlate to the collapse of the well-known quartets in the rotational spectra of asymmetric top molecules. These quartets occur for classically stable rotation about the principal axis with both the smallest and largest moments-of-inertia. Over several measurements of the broadband, chirped-pulse Fourier transform microwave spectra of large molecules with semiclassical limit spectra, we have noticed that the quartets for motion around the a-principal axis (prolate quartets) frequently show very fast decay of the free-induction decay (FID) relative to the oblate quartet transitions and the “normal” asymmetric top rotational transitions. To further explore this effect, we have performed Hahn echo experiments that show the rapid decay is caused by dephasing instead of population relaxation. It has also been observed that these transitions do not show significantly faster FID dynamics in a coaxial nozzle cavity FTMW instrument. A summary of results over a series of molecules and possible physical origins of the short dephasing times of the prolate quartets will be presented.