DIRECT OBSERVATION OF RYDBERG-RYDBERG TRANSITIONS IN CALCIUM ATOMS

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Chirped Pulse millimeter-Wave (CPmmW) spectroscopy, an extension of Brooks Pate's Chirped Pulse Fourier Transform Microwave (CP-FTMW) technique, has been adapted to observe electronic transitions between Rydberg states. The feasibility of this method has been demonstrated on calcium atoms of effective principal quantum number n^* of 40 and at frequencies of 70–84 GHz. Whereas Rydberg states are generally detected by ionization, we directly observe the free induction decay that they emit after polarization by the chirped pulse. Spectra with a bandwidth of 10 GHz and resolution of 1 MHz or better may be acquired in several shots, which could enable high-resolution "pure electronic" spectroscopy of molecules in core-nonpenetrating Rydberg states. The available millimeter-wave power is not a limitation in this experiment because the transition dipole moments are large (~5,000 Debye). Furthermore, the ability to quickly and arbitrarily polarize the sample opens avenues for optical plus millimeter-wave coherent experiments. Schemes to extend this technique to molecules are under development.