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Infrared laser frequency combs: generation and spectroscopic applications

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The laser spectroscopy revolution that began in the 1960's largely bypassed the infrared and terahertz regions of the electromagnetic spectrum. This includes the so-called molecular fingerprint region (500 - 1500 cm^{-1}, or 6.7 - 20 μm), which is appropriately named because of the rich spectral information it holds for identifying many molecular species. And despite the steady progress of more than 50 years of laser-based research, it is remarkable that one of the most widely used spectroscopic tool in this spectral region continues to be the Fourier transform infrared spectrometer, consisting of a thermal light source and mechanically-scanned Michelson interferometer. In this talk, we present recent laser and nonlinear optics advances that bring the power and precision of laser frequency combs to the long-wave infrared. We have developed a simple and robust method for generating super-octave (4-12 μm), optical frequency combs in the fingerprint region through intra-pulse difference frequency generation in an orientation-patterned gallium phosphide crystal. This frequency comb is orders of magnitude brighter than thermal light sources and comparable to infrared beam lines at synchrotron user facilities. We demonstrate the utility of this unique, coherent light source for high-precision, dual-comb spectroscopy in methanol and ethanol vapor. These results highlight the potential of laser frequency combs for a wide range of infrared molecular sensing applications, from basic molecular spectroscopy to nanoscopic imaging.