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Mollow sidebands in macroscopic high-harmonic generation¹ TENNESSE JOYCE, AGNIESZKA JARON-BECKER, University of Colorado, Boulder — In the high-harmonic spectra of many molecular ions, it was recently predicted that the usual odd harmonics can be accompanied by sidebands at noninteger multiples of the fundamental frequency, analogous to the appearance of Mollow triplets in quantum optics. These sidebands result from competition between two nonperturbative processes, Rabi oscillation and high-harmonic generation, and therefore offer additional insight into the dynamics of the molecule in a strong laser field. However, it is not immediately clear whether the effect is robust enough to appear in a real experiment. To answer this question, we estimate the macroscopic harmonic spectrum by combining many single-molecule calculations at different intensities, obtained with either time-dependent density functional theory or a 1D model potential. Our results suggest that not only do the Mollow sidebands remain around the same order of magnitude as the main harmonics in intensity, but they are also radiated at wider angles, and can therefore be distinguished from the main harmonics, and in principle also isolated as an alternative radiation source.

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