Any chemical process can be, in principle, understood and manipulated through electron dynamics. Such dynamics occur on what is known as the “ultrashort” time scale, taking place in $10^{-15}$ of a second (a femtosecond). Controlling or even observing these processes requires electromagnetic forces that can be arbitrarily shaped in space and controlled on the sub-femtosecond time scale, i.e. ultrashort laser pulses. Unlocking this capability has wide-ranging applications to a variety of chemical reactions (like photosynthesis) and electronic processes (leading, in principle, to PHz electronics).

While lasers of sufficiently short pulse duration are not currently available, we propose and develop a technique which will lead to the needed duration using coherently generated Raman sidebands. Our technique simultaneously produces and characterizes the pulses by using dichroic mirrors and the cross-correlation frequency resolved optical gating (XFROG) technique [1, 2] without the need for costly acousto-optic pulse shaping, as shown in Fig. 1(a). Our preliminary results are consistent with theory, and indicate that we have generated a 4 fs FWHM isolated pulse, the shortest isolated pulse synthesized with a Raman-based technique.

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