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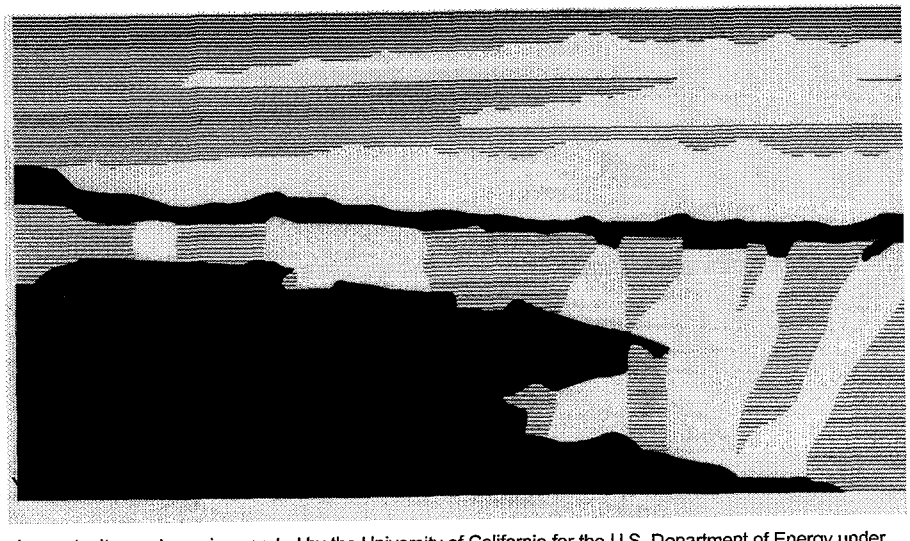

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Femtosecond Coherent Spectroscopy at 800nm: MI-FROG Measures High-Field Ionization Rates in Gases

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Abstract. We report the first quantitative phase-sensitive measurement of ultrafast ionization rates in gases. Ultrafast probe depletion via frequency mixing in the ionization front is observed.

The detailed dynamics of an atom in a strong laser field is rich in both interesting physics and potential applications. The source of both high-harmonic UV-VUV-XUV radiation [1] and ionization-based sources of tunable radiation [2,3], successful "mode-locking" of ultrafast ionization may soon yield the first attosecond pulses [4].

Previous studies [2,3] of ultrafast ionization have relied on spectral power density measurements using 100 fs pulses. As the detailed structure of the ionization-front blue-shifted spectra depends on a complicated interplay of the input pulse intensity/phase structure and the ionization dynamics [5], the conclusions made from such experiments are at best well-thought-out and limited inferences.

To achieve a quantitative measure of the ionization rate independent of probe structure, we used an experiment configuration which, by utilizing three power spectra channels and one optically gated frequency resolved channel per each of four CCD images (pumped or unpumped cases with one or two probe pulses, see figure 1), provides simultaneous and self-consistent dual-channel power spectra and frequency domain interferograms, Multi-pulse Interferometric Frequency Resolved Optical Gating (MI-FROG), [6], and FROG [7] data. Using an experimental setup similar to that previously described [6], data was taken with various gas densities, N_{gas}, of He, Ne, Ar, Kr, Xe, H_2, N_2, O_2, and Air at pump-probe increments of 12 fs. The polarization of the 124 fs,

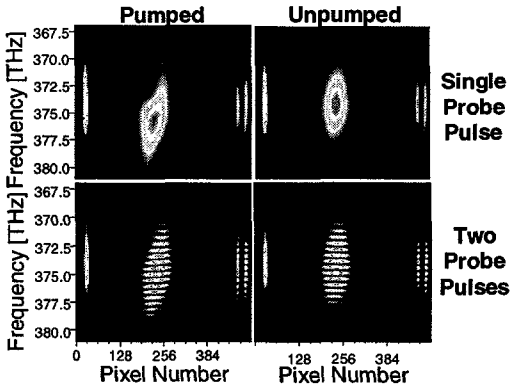

Figure 1: Example data.

400 nm pump was oriented either parallel or orthogonal to that of the weak 800 nm, 175 fs, multi-pulse probe.

The power-spectra channels allow both centroid-based power spectrum and interferogram analyses, which provide pulse-width averaged measures of frequency and phase shift, respectively. They can depend sensitively [5,3] on the detailed temporal and frequency-domain structure of the probe pulse. As MI-FROG recovers, to all orders, the phase difference between the pumped and unpumped probe pulses, we can utilize it to a) verify the intensity and phase structure of the probe pulse and b) recover the sub-pulsewidth time-resolved phase and frequency shifts impressed upon the probe by the ionization front, *independently* of the probe pulse structure.

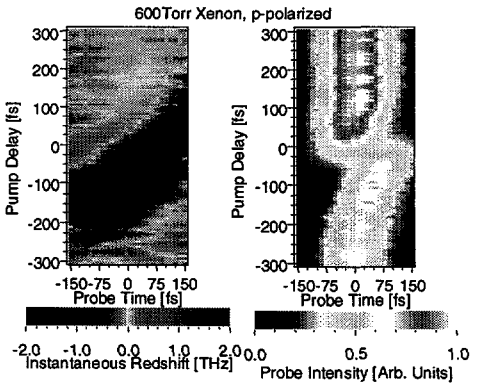

Figure 2: a) Doubly time-resolved frequency shift. b) Probe intensity profile.

Figure 2a illustrates the doubly time-resolved probe pulse frequency shift in the case of above-threshold ionization of Xe. The upper, red, diagonal corresponds to n_2 I cross-phase modulation via the neutral gas and is strongest for early pump delays. This asymmetry suggests defocusing is significant, a hypothesis supported by independent probe spectral data. The lower, blue, diagonal is predominately due to blueshifting in the ultrafast ionization front, though comparison of the p and s-polarized data indicates that neutral gas contributions are not always negligible. Figure 2b similarly illustrates the probe pulse intensity profile versus time. It should be stressed that by using MI-FROG a zero of time is well defined and the apparent motion of the pulse profile is genuine.

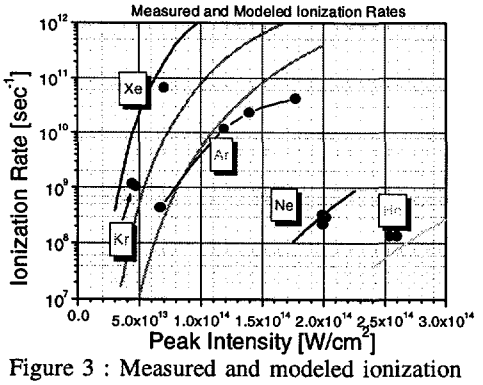

Figure 3 : Measured and modeled ionization rates for the Nobel gases, obtained assuming an interation length of two Rayleigh lengths.

To compare the measured ionization-front blueshifts with existing models of atomic ionization, we used a reduced frequency-shift parameter δ_ν given by $\Delta\nu/r_e\lambda N_{gas}$, where $\Delta\nu$ is the measured shift, $r_e = 2.8 \times 10^{-13}$ cm, and λ the laser wavelength. Thus, δ_ν is determined solely by the product of the single-atom ionization rate and an interaction length. Figure 3 presents the measured δ_ν divided by the confocal parameter. For those cases where the confocal parameter well approximates the plasma length (evidenced by no pump or time-resolved probe absorption and by

n_2I-like features in the frequency-shifts.), we find good agreement between the measured ionization rates with those calculated with a 1D plasma fluid model and Ammosov ionization rates [8]. Using Keldysh rates yielded ten times smaller predicted rates.

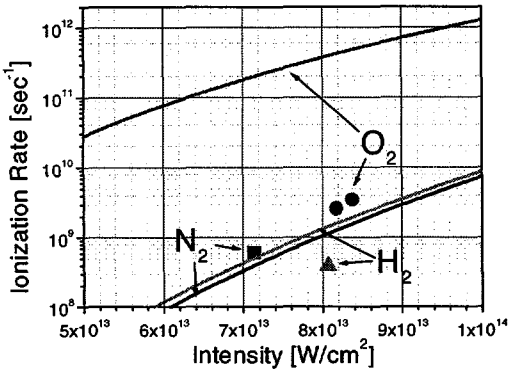

Figure 4: Measured and modeled ionization rates for three homonuclear diatomics.

In the case of the homonuclear diatomic gases H_2 and N_2 (figure 4), we find similar good agreement for the model, using published molecular ionization potentials. However, in the case of O_2, we find a two order of magnitude disagreement, with tunneling theory *overpredicting* the ionization rate. Similar disagreement has been observed by Gibson [9], who has proposed an alternative calculation method for the tunneling rate.

Finally, modeling of the Figure 2 and similar cases reveal frequency mixing via the transverse plasma current [10] on such a large scale that ultrafast depletion of the probe pulse occurs within the ionization front. Though this mixing is clearly accompanied by 2D effects not included in the model, the observed ultrafast depletion, as it occurs only *within* the ionization front and for *both* polarizations, cannot be due to defocusing or neutral gas effects. We conclude, then, that this work represents the first direct, ultrafast-time-resolved observation of this phenomenon.

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