Coherent VUV Emission from Field-Controlled Bound States

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Abstract: We demonstrate a dramatic enhancement of the below-threshold harmonics in the vicinity of atomic resonances. The dependence on the driving laser carrier-envelope phase suggests a nonperturbative mechanism. Phase matching promises scalability to microJoule pulse energies.

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1. Introduction

The generation of high-order harmonics has enabled attosecond pulse generation and ultrafast spectroscopy in the extreme ultraviolet. Recently, ultrafast sources of below-threshold harmonics have been demonstrated, with photon energies below the ionization potential of the generation target. While below-threshold harmonics generation is not compatible with the three-step model of high-order harmonics generation (HHG), recent experiments suggest that the generation process is nonperturbative[1,2]. Here, we uncover a new regime of below-threshold harmonics generation in the vacuum ultraviolet (VUV), which is accompanied by bright, coherent line emission in the vicinity of atomic resonances. Unlike the plasma line emission recently observed in HHG with plasmonic enhancement[3], the resonance-enhanced emission structures are spatially coherent and can be controlled by the sub-cycle field variations of the few-cycle driving laser. Perfect phase matching is enabled by the anomalous dispersion near the atomic resonance, and allows energy scalability to the microJoule level. Since the VUV generation process requires few-cycle driving lasers with intensity of only ~10^{13} W/cm^2, it is readily extendable to multi-MHz repetition rates when driven by commercial Ti:Sapphire oscillators.

2. Experiment and Discussion

In the experiment, near- and below-threshold harmonics were generated using few-cycle (5 *fs*, 730 nm) laser pulses from a Ti:Sapphire amplifier and hollow-core fiber pulse compressor, loosely focused into an argon-filled gas cell with moderate intensities of ~10^{13} to 10^{14} W/cm^2. Within this intensity range, the Keldysh parameter ranges from ~4 to 0.5, spanning the transition from multi-photon excitation to tunnel ionization. The angularly-resolved harmonic spectrum was measured using a flat-field grating spectrometer with resolution of 40 meV at 15 eV.

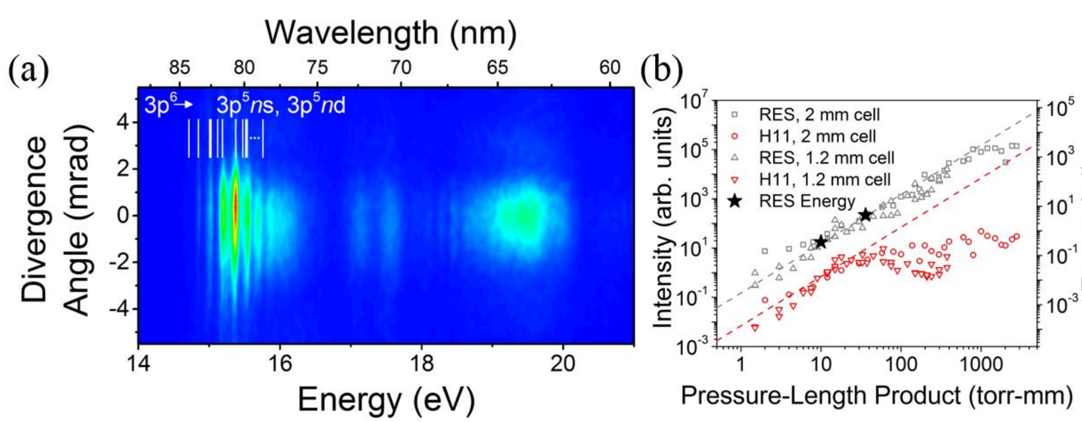

Fig. 1. (a) Angularly resolved RES spectrum. (b) Phase-matched growth of RES emission.

Figure 1(a) shows the angularly-resolved VUV spectrum near the argon ionization threshold (I_p = 15.76 eV) with a driving laser intensity of ~3×10^{13} W/cm^2 and a target pressure-length product of 10 torr-mm. While the 11th harmonic lies completely above the ionization threshold, the 9th harmonic covers energies both above- and below-threshold. Between 14 eV and the ionization threshold, the 9th harmonic exhibits narrow-linewidth resonance-enhanced structures (RESs) which coincide with the excited state resonance energies of argon. The divergence angle of the RESs (~3 mrad) and is in good agreement with that of the above-threshold 11th harmonic and with previous measurements of below-threshold harmonics with multi-cycle lasers[1]. Figure 1(b) shows the scaling of the RESs with the pressure-length product of the argon gas target. The RES energy was measured using an indium foil filter and XUV photodiode. Perfect phase matching is enabled for relatively large pressure-length products as compared with above-threshold HHG, due to the anomalous dispersion of the atomic gas in the vicinity of the resonances as well as the lack of absorption below the ionization threshold, and allows scalability to microJoule VUV pulse energies with nearly 1% conversion efficiency.

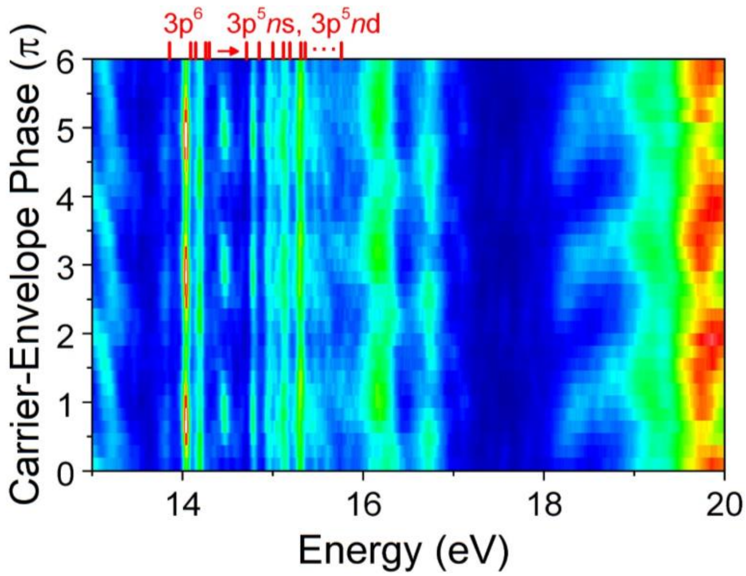

Fig. 2. Carrier-envelope phase dependence of gated RES emission.

Previous studies of phase-matched harmonic generation in the anomalous dispersion regime have focused on the perturbative third harmonic generation[4]. Here, we demonstrate that the RESs observed in below-threshold harmonics generation result from a non-perturbative strong-field interaction. We observe that the RESs depend strongly on the ellipticity of the driving laser pulse, which allows the RES emission to be temporally "gated". Figure 2 shows the carrier-envelope phase (CEP) dependence of the RES generation under the double optical gating[5]. The RESs exhibit a strong CEP dependence, indicating strong-field control on the sub-optical-cycle timescale.

3. Conclusion

We have demonstrated a novel regime of nonperturbative below-threshold harmonics generation accompanied by resonance-enhanced VUV emission structures. The method is scalable to nearly 1% of the driving laser energy and can be controlled by the sub-cycle field of the driving laser. Extending the technique to high-repetition rate few-cycle laser oscillator promises to enable the development of MHz VUV sources for photoemission spectroscopy and coherent imaging without the need for cavity enhancement.

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4. References

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