

STRONG-FIELD IONIZATION OF DIATOMIC MOLECULES AND MOLECULAR ANIONS: INTERFERENCES AND CLASSICAL MODEL

D. Habibović¹, A. Gazibegović-Busuladžić¹, M. Busuladžić^{2,1}, D. B. Milošević^{1,3}

¹ Faculty of Science - University of Sarajevo, ² Faculty of Medicine - University of Sarajevo, ³ Academy of Sciences and Arts of Bosnia and Herzegovina

dhfizika1@gmail.com

Introduction

When molecules are exposed to a strong laser field many novel phenomena can be observed due to the interference effects. The required intensities of the applied field are of the order of 10^{14} W/cm². The archetypal description of the underlying physics relies on an electron, liberated by the multiphoton or tunnel ionization, which propagates in the continuum and can directly reach the detector (ATI) or first recollide with the parent ion (HATI). Already in early studies it was shown that the molecular symmetry is imprinted to the low-energy photoelectron spectra [1], i.e. the ATI spectra depend on the symmetry of the molecular orbital.

Theory

Differential ionization rate can be calculated using the theory based on the strong-field approximation (SFA):

$$w_{fi}(n) = 2\pi \mathbf{k}_f |T_{fi}(n)|^2, \quad (1)$$

where \mathbf{k}_f is the final electron momentum, and the T -matrix element consists of six contributions, two of which correspond to the direct electrons, while other four correspond to the rescattered electrons. Because the matrix elements $\mathcal{F}_{fi}^{(0)}(t)$ and $\mathcal{F}_{fi}^{(1)}(t)$, which correspond to the direct and rescattered electrons, respectively satisfy $\mathcal{F}_{fi}^{(0)}(t) \propto \sum_{q=\pm 1} e^{iq\mathbf{k}_f \cdot \mathbf{R}/2}$ and $\mathcal{F}_{fi}^{(1)}(t) \propto \sum_{q=\pm 1} e^{iq[\mathbf{k}_{st} + \mathbf{A}(t_0)] \cdot \mathbf{R}/2}$, the conditions for the interference minima which correspond to the atomic orbitals of a given parity can be obtained [2]. Here \mathbf{R} is the internuclear distance and \mathbf{k}_{st} is the stationary momentum. For the direct electrons and electrons which are born and rescattered at the same atomic center the contribution to the T -matrix element is proportional to $\cos \alpha$ ($\sin \alpha$) for even (odd) atomic orbitals where $\alpha = \mathbf{k}_f \cdot \mathbf{R}/2$, while the rescattering contribution is proportional to the product $\cos \delta \cos \gamma$ ($\sin \delta \cos \gamma$), $\gamma = (\mathbf{k}_f - \mathbf{k}_{st}) \cdot \mathbf{R}/2$, $\delta = \mathbf{k}_{st} \cdot \mathbf{R}/2$ for even (odd) atomic orbitals.

The other way to calculate the (H)ATI spectra is by using the improved classical model where the electron's final momentum is calculated classically as a solution of the Newton's equation of motion, while the ionization rate is obtained using the imaginary-time method [3]. Here we analyze the orthogonally polarized two-color (OTC) laser field which consists of two linearly polarized fields with mutually orthogonal polarizations and commensurable frequencies.

Numerical results

First, we analyze the interference effects using the ω - 2ω OTC laser field. The intensity of the field components is $I_1 = I_2 = 1.2 \times 10^{14}$ W/cm², the fundamental wavelength is 800 nm, while the relative phase is $\varphi = 0^\circ$. In Fig. 1 we present various contributions to the T -matrix element.

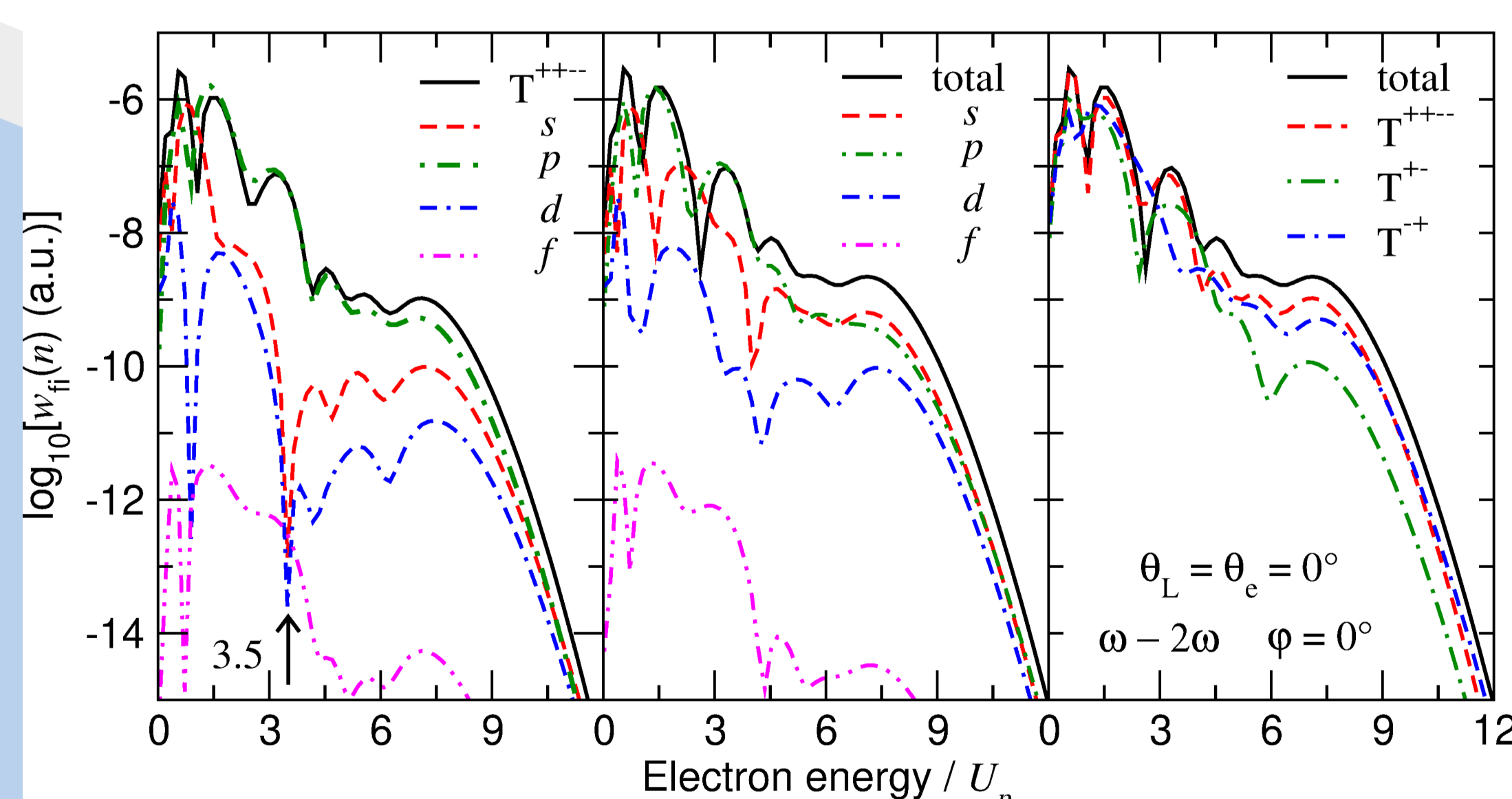


Figure 1: Logarithm of the differential ionization rate of the N_2 molecule exposed to the ω - 2ω OTC field as a function of the photoelectron energy together with various partial contributions.

In Fig. 2 we present the modulus of the phase factor which appear in the T^{++--} contribution to the T -matrix element as a function of the electron's energy for s and d (black solid line) and p and f (red dashed line) atomic orbitals. Clearly, for s and d atomic orbitals this phase factor is equal to zero for the electron energy around $3.5 U_p$, so that the corresponding spectra also exhibit minimum for this electron's energy (see red and blue lines in the left panel of Fig. 1).

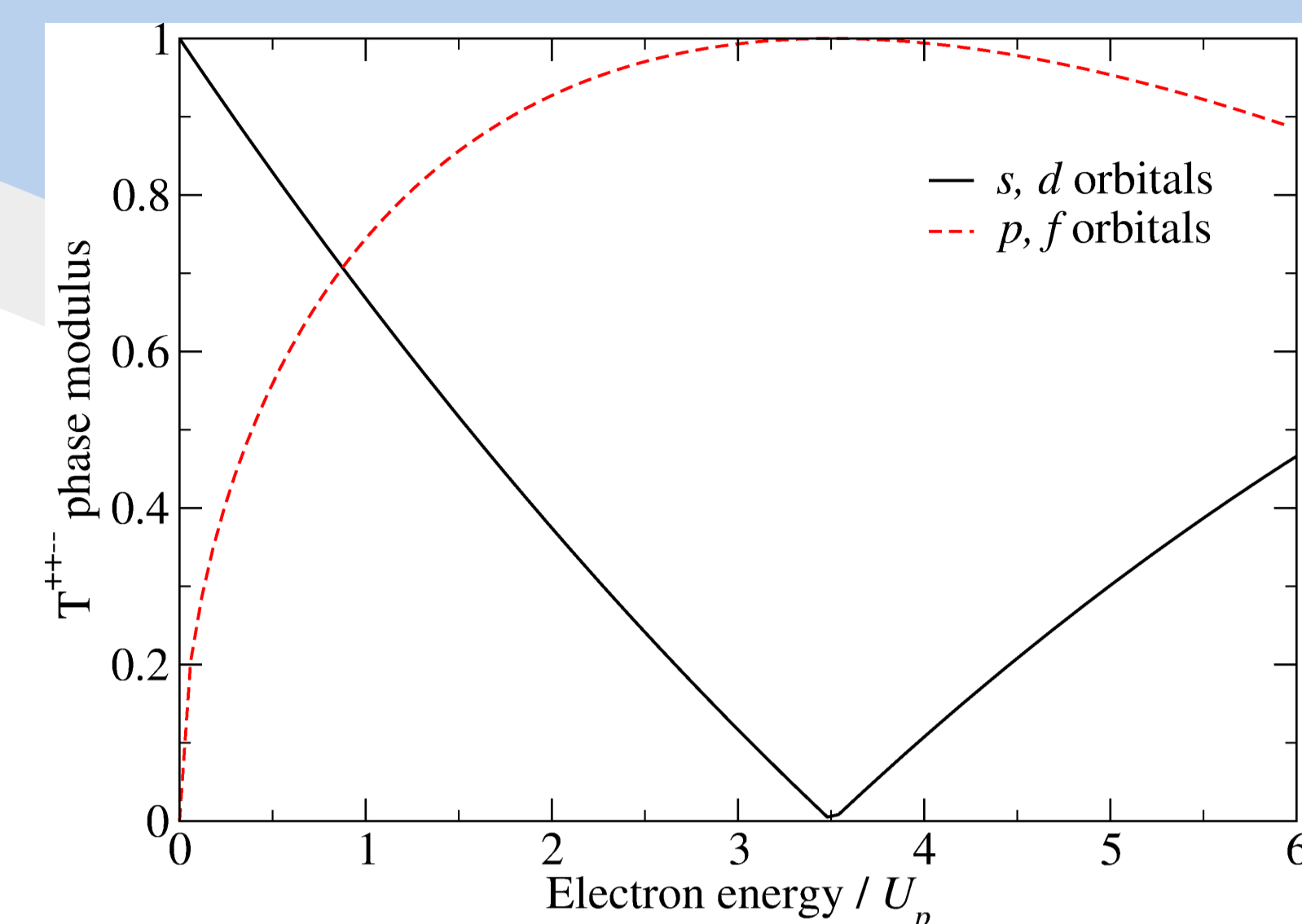


Figure 2: Modulus of the phase factor which appears in the T^{++--} contribution to the T -matrix element as a function of the electron's energy for s and d (black solid line) and p and f (red dashed line) atomic orbitals.

In order to explain how the particular atomic orbitals contribute to the total spectra in the left (right) panel of Fig. 3 we present the modulus of the product $\cos \delta \cos \gamma$ ($\sin \delta \cos \gamma$) which appear in the rescattering contribution as a function of the emission angle and photoelectron energy.

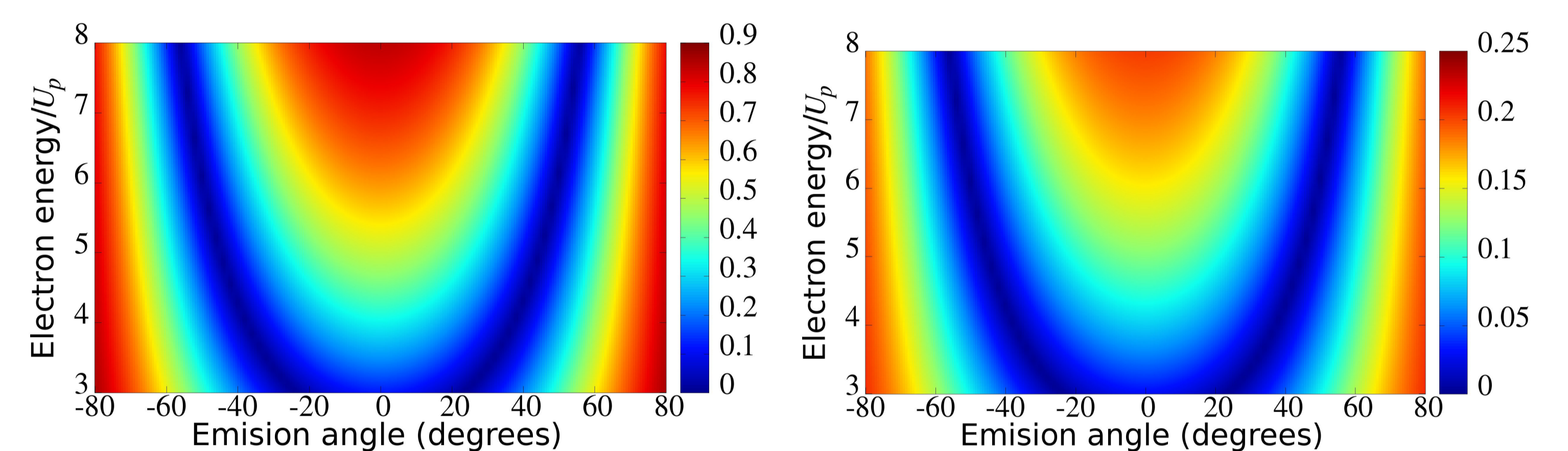


Figure 3: Left (right) panel: Modulus of the product $\cos \delta \cos \gamma$ ($\sin \delta \cos \gamma$) which appears in the rescattering contribution as a function of the emission angle and photoelectron energy for s (p) atomic orbitals.

Finally, in Fig. 4 we present the comparison between the SFA results and results obtained using the improved classical model.

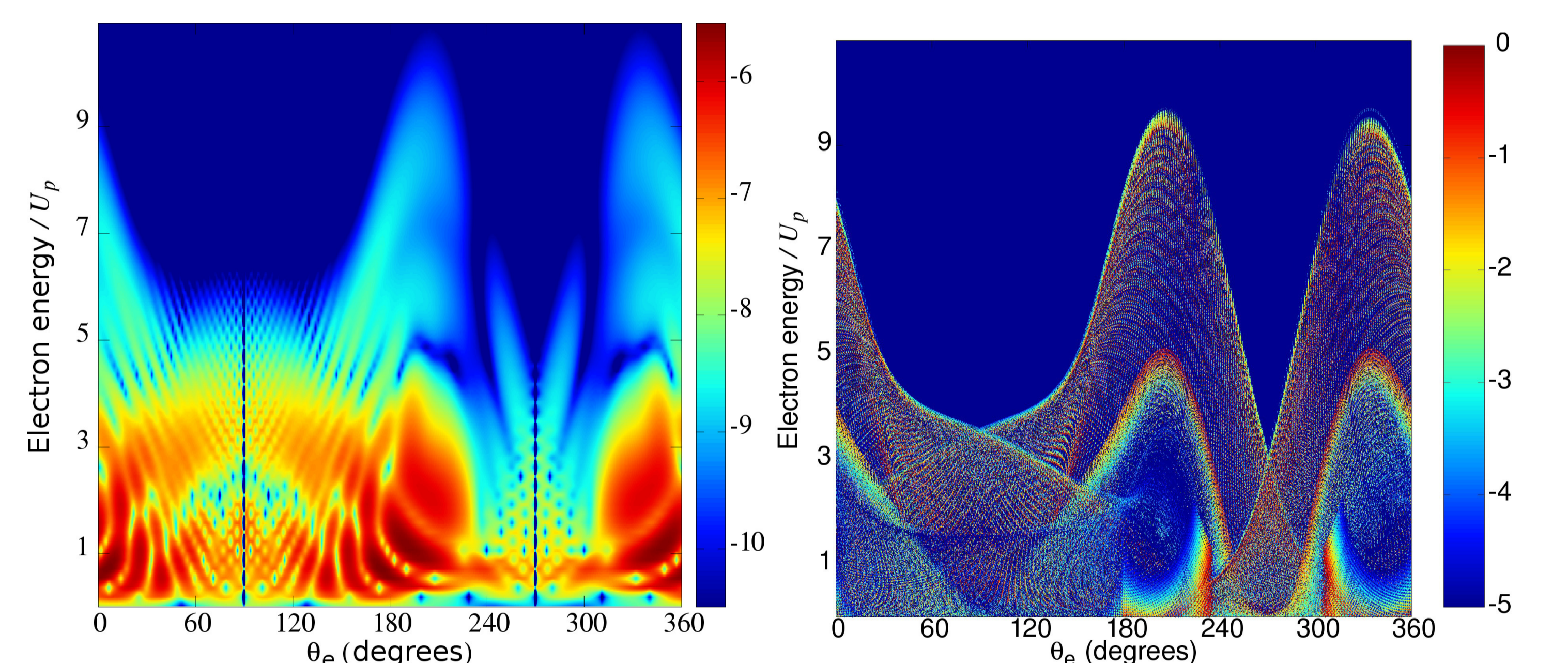


Figure 4: Left panel: SFA results. Right panel: Improved classical results.

Conclusion

The photoelectron spectra obtained using only the contribution which corresponds to the situation where the electron is liberated and rescattered at the same atomic center exhibit minima for the particular values of the photoelectron energy in accordance with the analytically derived formula.

The conclusions about which atomic orbital is dominant can be made by inspecting the value of the phase factor which appears in the direct and rescattering contributions to the differential ionization rate.

The HATI molecular spectra can be well reproduced using the improved classical model.

References

- [1] F. Grasbon, G. G. Paulus, S. L. Chin, H. Walther, J. Muth-Böhm, A. Becker, and F. H. M. Faisal, Phys. Rev. A **63**, 041402 (2001).
- [2] M. Busuladžić, A. Gazibegović-Busuladžić, D. B. Milošević, and W. Becker Phys. Rev. A **78**, 033412 (2008).
- [3] V. S. Popov, Phys.-Usp. **47**, 855 (2004).