Atomic Floquet Physics Revealed by Free Electrons

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Abstract: We theoretically investigate the ability of free electrons to probe the nonlinear Floquet dynamics of atoms under intense illumination. We observe multiple features in the electron energy-loss spectra associated with electron-photon exchanges and Floquet resonances. © 2022 The Author(s)

Free electron beams allow us to probe optical excitations sustained by electronic and phononic dynamics with an unparalleled combined spatial and energy resolution through electron energy loss spectroscopy (EELS) [1]. Novel techniques such as photon-induced near-field electron microscopy (PINEM) [2] permit shaping of the electron wave function and exploring samples typically holding bosonic excitations. In a separate but related context, the interaction of electrons with atoms under strong illumination, in which the probed excitations have fermionic character, has attracted increasing attention in the past due to the observation of multiphoton processes [3]. More recently, the use of periodic optical driving on several types of physical systems [4,5] has been a very active field of research due to the possibility of exploiting Floquet physics to induce topological states of matter. In this work, we combine these elements to explore the physics of an optically driven atomic system in the steady state using EELS.

We consider a two-level system with transition frequency ω₀ and an inelastic decay rate κ₀ = 10⁷ω₀, under monochromatic external illumination of frequency ω_L. The Hamiltonian of the atom-light interaction, which includes a coupling strength quantified by the Rabi frequency Ω₀, is time-dependent but periodic with period 2π/ω_L. Due to the external drive and the inherent decay, the atom + light system reaches a specific steady state, independent of the initial conditions. The steady-state dynamics is probed using free electrons, which can gain or lose energy during the interaction.

![Evolution of the EELS probability as a function of energy loss and light-atom coupling strength Ω₀ for a two-level system under different illumination frequencies ω_L. (F-J) Evolution of the EELS probability with ω_L or different light-atom coupling strengths. We normalize Ω₀, ω₀, and ω_L to Ω₀ and the loss probability to the non-illuminated atom at ω = ω₀.](image-url)
A dramatic departure from the signal found in the interaction of the electron and a non-illuminated atom, which only showed a loss peak at \( \omega = \omega_0 \), is now clearly observed in the illuminated system. Firstly, we note the appearance of PINEM-like sidebands [2] corresponding to the exchange of an odd number of photons with the laser field, emerging as straight lines with slope \( \pm \hbar \omega_1 \) in the EELS spectrum. A second feature is the spectral shift of the peak positions as the intensity of the field is increased (i.e. a dynamical Stark effect [6]). Finally, avoided crossings of the PINEM-like peaks and those corresponding to transitions between Floquet states [7] arise in the \( \omega_2 \)-dependent spectra, with growing separation between peaks as the intensity \( \Omega_0 \) is increased. We conclude that free electrons offer a suitable platform to probe the steady-state dynamics of strongly non-linear multi-level fermionic systems such as atoms and excitons.

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