Anomalous Fano Profiles in Strong Fields

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(Dated: May 19, 2014)

In presence of an infrared control field, a Fano resonance line acquires a complex $q$-parameter, reflecting the non-stationary nature of the control process. We derive a simple expression for $q$ and compare to numerical solutions of the TDSE of Helium in three dimensions. Control during excitation is dominated by a modification of the continuum states. For a control pulse after excitation, bound state distortions by infrared two-photon transitions dominate and lead to characteristic interference fringes.

The celebrated Fano formula

\[
\sigma(\Delta E) = \sigma_0 \left( \frac{q\Gamma + 2\Delta E}{\Gamma^2 + 4\Delta E^2} \right)
\]

characterizes the cross section of any process involving a continuum that is structured by interaction with a single embedded bound state relative to the cross-section $\sigma_0$ in absence of the embedded state. Apart from the width $\Gamma$ and the detuning from the resonance energy $\Delta E$, there is the $q$ parameter, which produces a characteristic asymmetry and — if it is real — an exact zero of the cross section. The Fano profile is one of the prominent manifestations of quantum mechanical interference in scattering. The mechanism is ubiquitous and independent of the particular nature of the transitions involved. In recent years it was proposed to control the line shape by external parameters and schemes in diverse fields of physics were experimentally realized (see review in [1]). In particular, for a quantum dot system controlled by a magnetic field [2] it was observed that complex $q$ was needed to fit the control-dependence of the line shape and the observation was associated with breaking of time-reversal symmetry. In contrast, in standard Fano theory [3], a sudden excitation of an otherwise stationary, time-reversal symmetric system results in real-valued $q$ (see, e.g., [4]). Complex $q$ has also been discussed as a signature of decoherence in atoms [5] as well as in quantum dots [6]. From a more general point of view, complex $q$ can be expected whenever an additional decay channel turns the embedded state into a decaying state that cannot be associated with a real-valued eigenfunction.

In the present work we show that also an electrical control field leads to a complex Fano $q$-parameter. We demonstrate this for a pump-probe scenario as in Ref. [7]: a short extreme ultraviolet (XUV) pulse combined with a weak, time-delayed near-infrared (IR) pulse ionizes a He atom, producing Fano lines in the photo-electron spectrum at the positions of the 2snp series of doubly excited states. The two primary mechanisms of controlling the line shapes by the IR field are to modify energies (by Stark shifts) and to modify the involved bound and continuum states. When additional couplings are induced by the IR, the elementary Fano scenario breaks down and the universal line shape Eq. (1) is lost. This happens for multiple resonant states coupled by the IR pulse, as first considered in [8]. Near-resonant coupling, including possible Autler-Townes splitting were discussed in [9]. In the theoretical interpretation of [7], focus was on Stark shifts, but without direct reference to an microscopic model of the Fano resonance. A conjecture on the structural impact of delayed (partial) depletion of the embedded state was made in [10]. To our knowledge, the influence of non-resonant IR multi-photon processes has not been considered so far. Another quite obvious process whose impact on the line shapes was not discussed before is the “streaking” of electrons, i.e. the electron momentum boost from XUV electron emission time $t_0$ until the end of the IR pulse at time $t_1$

\[
\vec{k} \rightarrow \vec{k} + \vec{A}(t_0), \quad \vec{A}(t_0) := -\int_{t_0}^{t_1} \vec{E}(\tau) d\tau,
\]

which in particular redistributes amplitudes between the partial waves. (Unless indicated otherwise, we use atomic units, where electron mass, proton charge, and $\hbar$ are all set equal to 1.)

Using analytical as well as complete numerical solutions we will show that it is mainly the last two processes which determine the line-shapes. We find that for overlapping XUV and IR pulses, the line-shape distortions are caused by a field-induced boost of photo-electron momenta: the profile remains Fano-like, albeit with complex $q$. In contrast, if the XUV precedes the IR pulse, modification is predominantly through the distortions of the resonant state: interference fringes from weak IR two-photon coupling appear already well below the IR intensities considered in [7]. As a result, the line no longer has a Fano-like profile and shows additional delay-time dependent modulations.

Fig. 1 gives an overview of the photo-emission spectra in the vicinity of the He(2s2p) line as a function of XUV excitation time $t_0$ with an IR pulse centered at $t = 0$. The results were obtained by numerically solving the time-dependent Schrödinger of the He atom in full 3+3 spatial dimensions. Spectra were computed using...
FIG. 1: Photo-electron spectra of He in the vicinity of the 2s2p resonance as a function of XUV excitation time $t_0$ ($l = 1$ partial wave). IR peak intensity $2 \times 10^{12} \text{W/cm}^2$. Solid line: IR field, dotted lines: maxima due to IR two-photon interferences. Dashed lines indicate the lineout times of Fig. 2.

the time-dependent surface flux method (tSURFF, [11]). The XUV wavelength of 21 nm was chosen to match the excitation to the 2s2p state, but the spectral width of $\sim 10 \text{ eV}$ at the pulse duration of 0.15 fs evenly covers the whole 2snp series of doubly excited He states. Calculations were performed for IR wavelength of 800 nm with pulse duration of 2 optical cycles and peak intensity $2 \times 10^{12} \text{W/cm}^2$.

We first concentrate on the temporal overlap region around $t_0 = 0$. Fig. 2 shows lineouts of the spectra at $t_0 = -3/4$ and $+1/2$ in units of IR optical cycles. At $t_0 = -3/4$, where the peak of the XUV intensity coincides with a node of the IR electric field, one sees a near Fano profile. In contrast, at $t = 0.5$, peak of the IR field, the profile is Lorentz-like. At suitable delays, the Fano minimum appears at the energetically lower side of the resonance (see Fig. 5 below). Neither width nor position of the resonance are affected by the weak IR. The pattern qualitatively repeats itself for all $t_0$ during the IR pulse.

Fig. 2 also contains fits to the lines by Eq. (1), where $\Gamma$ was taken from the IR-free case and only the overall intensity $\sigma_0$ and $q$ and were adjusted, restricting $q$ to real values and admitting complex $q$, respectively. Where the XUV falls onto node of the IR (right panel of Fig. 2), only the fit with complex $q$ is satisfactory: phenomenologically, there is no exact zero in the spectrum when IR and XUV pulse overlap, which trivially rules out an exact fit by Eq. (1) with real $q$.

We now show how complex $q$ arises in the framework of a generalized Fano theory. A standard Fano Hamiltonian has the form

$$H_f = |\varphi\rangle E_\varphi \langle \varphi| + \int d^3k \left[ \langle \vec{k}\rangle^{2} \frac{k^2}{2} + \langle \vec{k}\rangle V_k \langle \varphi| + h.c. \right],$$

where the embedded bound state $|\varphi\rangle$ interacts with the continuum states $|\vec{k}\rangle$ through $V_k = \langle \vec{k}|V|\varphi\rangle$. The exact scattering solutions $|\xi_k\rangle$, the resonance width $\Gamma$, and the shift of the resonance position from the non-interacting $E_\varphi$ are all given in closed form. The Fano transition amplitude $\langle \xi_k|T|\phi_0\rangle$ for an arbitrary transition operator $T$ from some initial state $|\phi_0\rangle$ leads to the Fano cross section (1). Introducing the wave packet after transition

$$T|\phi_0\rangle =: |\psi_0\rangle = |\varphi\rangle X_\varphi + \int d^3k |\vec{k}\rangle X_{k}$$

and assuming for notational simplicity that $|\varphi\rangle$ decays into a well-defined angular momentum state, the $q$-parameter in the corresponding partial wave cross section (in case of the 2s2p doubly excited state $l = 1$) is

$$q_0 = \frac{\langle \varphi|\psi_0\rangle + P \int \frac{k^2dk'}{2\pi^2} V_k^{\ast} (k'|\psi_0\rangle}{\langle \vec{k}|\psi_0\rangle}$$

where $|k\rangle$ denotes partial wave continuum state with $k = \sqrt{k^2}$. When the $\langle k'|\psi_0\rangle$ and $|\varphi\rangle|\psi_0\rangle$ all share the same phase, $q$ is real. This is in particular the case, when the initial state $\phi_0$ is a true bound state and the transition $T$ is “sudden”, i.e. without energy-dependent phase.

For the overlap region, we restrict the effect of the laser on the embedded state $|\varphi\rangle$ to a Stark shift of its energy $E_\varphi(t)$. We further assume that the IR pulse duration is short compared to the decay time of the embedded state. The net effect of the IR pulse is to replace $|\psi_0\rangle$ by a modified initial wave packet

$$|\psi(t)\rangle = |\varphi\rangle X_\varphi + \int_{t_0}^{t_1} d^3k \int_{t_0}^{t} e^{-i\varphi_{t_0}(t')} |\vec{k} - \vec{A}(t) + \vec{A}(t_0)| X_{k} dt_{t_0},$$

where $|\vec{k} - \vec{A}(t) + \vec{A}(t_0)|$ are the continuum states including the momentum boost $\vec{A}(t) - \vec{A}(t_0)$ imparted by the IR between between $t_0$ until $t$. The phase offset $\Phi_k(t)$ between embedded and continuum states accumulated from excitation at $t_0$ until $t$ is

$$\Phi_k(t) = \int_{t_0}^{t} \frac{|\vec{k} - \vec{A}(t) + \vec{A}(t_0)|^2}{2} - E_\varphi(t) + E_\varphi(t_0)dt. \quad (7)$$
Clearly, even if $X_\varphi$ and $X_\xi$ are all real, the interaction with the IR imprints a phase-modulation on $|\psi_1\rangle$ and the Fano parameter becomes complex. The redistribution of partial waves by the momentum boosts can be easily computed in the so-called “strong field approximation” for the continuum states: when the IR field prevails over the atomic potential, the continuum states at time $t$ are can be approximated as plane waves with wavevector $\vec{k} - \vec{A}(t)$. A short calculation using the standard spherical expansion of plane waves leads to the IR modification of the Fano parameter

$$q_1 = q_0 + a \left( e^{-i\chi} / J - 1 \right),$$

where $a = \langle \varphi | \psi_0 \rangle / (\pi V^2 k^2 |k| |\psi_0\rangle)$ denotes the relative strength of initial embedded to continuum amplitudes, and

$$\chi = \int_{t_0}^{t_1} dt [\vec{A}^2(t)/2 - \vec{A}^2(t_0)/2 - E_\varphi(t) + E_\varphi(t_0)]$$

is a laser-induced phase shift between embedded and continuum states. Although the phase-shift does give a numerically discernable contribution, the $t_0$-dependence of $q_1$ is dominated by

$$J = j_0(|\vec{a}|k) - 2j_2(|\vec{a}|k) - ij_1(|\vec{a}|k) \frac{\vec{a} \cdot \vec{A}(t_0)}{3|\vec{a}|},$$

where the spatial offset of a free electron by the IR pulse

$$\vec{a} = \int_{t_0}^{t_1} dt \vec{A}(t)$$

appears in the argument of the spherical Bessel functions. The $J$-term accounts for the “streaking effect”, i.e. the energy modulations due to $\int_{t_0}^{t_1} \vec{k} \cdot \vec{A}(\tau) d\tau$, which re-distributes the original purely $l = 1$ continuous functions into $l = 0$ and $l = 2$. Due to dipole selection rules, only these terms contribute to the $l = 1$ partial wave. As decay of the embedded state is exclusively into $l = 1$, the Fano interference phenomenon is affected by depletion of the direct contribution to the $l = 1$ continuum.

In Fig. 3 we compare Eq. (8) with fits to the numerical simulation data for the 2s2p line. Sign-changes of the real part corresponding to transition from Fano to “anti-Fano”, and peaks in the imaginary parts are all well reproduced. Quantitative deviations must be expected, for example, due to the simple approximation of the scattering solutions by plane waves. In addition, there is a non-negligible background of IR two-photon coupling, see discussion below.

Within the model of Eq. (6), the factor $J = 1$ at excitation times $t_0 = t_n$ where the spatial offset vanishes $\vec{a} = 0$ and the imaginary part of $q_1$ is exclusively due to the phase-shifts $\chi$. Up to small offsets due to the short IR pulse duration, the $t_n$ coincide with zeros of the field. For these excitation times, the profile is Fano-like (Fig. 2), except that the characteristic minimum remains slightly above zero. The minima for subsequent $t_n$’s grow monotonically from the IR-free value of 0 (Fig. 3, right panel), reflecting the slow accumulation of the phase shift $\chi$, Eq. (9).

When the XUV precedes the IR pulse without overlap, the spatial offset goes to zero ($\vec{a} \approx 0$) and therefore $J \approx 1$. Here, line-shapes are the combined effect of the phase-shift $\chi$ and IR two-photon coupling between embedded and continuum states, which is beyond the basic Fano model (3). Two-photon coupling clearly manifests itself in a side-band like interference structure at energies around $E_\varphi - 2\omega$ due to stimulated 2-photon emission. By $\omega$ we denote the IR photon energy. Two-photon (absorption-emission) transitions that couple the embedded state to the continuum in its immediate surrounding are of the same magnitude. We fit both spectral features for time-delay $t_0$ and IR peak field strength $E_0$ by

$$\sigma_c(E) = |f(E) + e^{-iE_0 t_0} c_0(E)|^2,$$

where $f(E)$ is the IR-free Fano transition amplitude and $c(E)$ is the unknown two-photon transition amplitude.

In Fig. 4 the cross-section Eq. (12) at delay $t_0 = 12$ IR opt.cyc. is compared to the TDSE result. For $c(E)$, we use Gaussians centered at $E_\varphi$ and $E_\varphi - 2\omega$, respectively. Both structures are fitted with the same width of $c(E)$ and only the magnitudes were adjusted independently, admitting for the different nature of the transitions into a structured and unstructured continuum, respectively. As one can see, interference fringe separation is $2\pi/t_0$, which proves the nature of the spectral modulation as the interference of photo-electrons emitted at relative delay $t_0$. Keeping $c(E)$ fixed, the fringes are equally well reproduced for intensities up to $I \lesssim 10^{12} W/cm^2$ and for all $t_0$. The quadratic dependence on IR field strength $E_0$ shows that this is a true two-photon process without resonant coupling to neighboring states. We further confirmed this by calculations at IR wave length up to $\lambda = 2\mu m$, where
the fringes consistently appear. At short time-delays $t_0$, fringe separation increases and is hardly discernable as an independent feature, affecting only larger detunings from the resonance. Note that the effect is “heterodyning”, i.e. a re-distribution of probability generated by the original Fano resonance, little extra electron probability added by the coupling.

The main characteristics of the line-shape modulations are independent of the exact structure of the resonance. Fig. 5 shows the delay-dependence of the $(2s)(np)$ resonances up to $n = 7$: although polarizability of the embedded states vastly differs, transition from Fano to anti-Fano all occur at the same laser parameters. This further corroborates that the modulations is dominated by the field-dressing of the continuum states.

Complex $q$ necessarily appears whenever a non-trivial phase is imprinted on the Fano system. This phase may be due to inherent dynamics of the embedded state, i.e. when $\ket{\phi}$ is not strictly an eigenstate of a stationary Hamiltonian, as for decaying states and decoherence. Here we have considered a different route to complex $q$, namely, manipulation of the phase of embedded and continuum states after both were populated. Note that also in the case of full temporal overlap between the (very short) excitation process and the control pulse, one can consider control to start only after excitation and that the convolution of both processes is of secondary importance.

Combining existing laser beam lines, e.g. as in Ref. [7], with precision photo-electron spectrograms should allow to experimentally detect complex $q$: the profile in Fig. 2 shows an unambiguous signature in the departure from an exact zero of the cross section as described by Eq. (8). One needs to be aware that the presence of the IR introduces additional partial waves beyond the Fano decay channel $l = 1$. These can exceed the Fano background $\sigma_0$ Eq. (1) and need to be carefully subtracted by an angle-resolved measurement.

Our analytical approach to the time-dependent control of complex $q$ can be generalized to any system that allows a sensible description of the impact of the control on bound and embedded states. Other than short laser pulse controls, this may include time-dependent electric or magnetic fields for quantum dot systems.

We acknowledge support by the excellence cluster “Munich Center for Advanced Photonics (MAP)” and by the Austrian Science Foundation project ViCoM (F41).

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