Fano’s propensity rule in angle-resolved attosecond pump–probe photoionization

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Interferometric measurements based on combining extreme ultraviolet (XUV) attosecond pulse trains with a weak infrared field, have been successfully used to study time-resolved and recently angle-resolved photoemission in various systems. These measurements rely on quantum interferences between electron wavepackets created by absorption of XUV radiation, with further absorption or emission of laser photons. In this work, we extend Fano’s propensity rule to these above-threshold ionization processes and show that the asymmetry between absorption and emission results in incomplete quantum interference. This leads to periodic emission of photoelectrons perpendicular to the laser polarization at specific delays between the attosecond pulse train and the laser field.

In quantum mechanics, the possible transitions between different states are dictated by selection rules which are based on symmetry arguments. For example, the famous parity and angular momentum selection rules are at the core of our understanding of light-matter interactions. In contrast to these stringent selection rules, the concept of propensity rules, which was introduced by Berry [1], is based on the fact that all allowed transitions are not equally probable. Selection and propensity rules play a fundamental role in physics and chemistry to understand the possible outcomes and reaction rates. One of the most fundamental reactions is the photoionization of an atom following the absorption of a high energy photon, \(A + h\nu \rightarrow A^+ + e^-\), where an electron is promoted to a manifold of degenerate continuum states.

The well-known electric dipole selection rules greatly simplify the problem, restricting the possible transitions to those where the electron angular momentum changes by one unit, \(\Delta l = \pm 1\). Fano’s propensity rule states that out of the two possible transitions, the one increasing the electron angular momentum is favored, due to increase of the centrifugal potential with angular momentum [2]. Conversely, for the time-reversed process, decreasing angular momentum is favored as a photon is emitted.

This Letter deals with laser-assisted photoionization, \(A + h\nu_{\text{XUV}} \pm h\nu_{\text{IR}} \rightarrow A^+ + e^-\), where absorption of an XUV photon brings an electron to the continuum, followed by absorption or stimulated emission of laser radiation, often in the infrared (IR), between unbound states. A natural question is whether Fano’s propensity rule, originally stated between a bound and a continuum state, can be extended to this regime and how a possible asymmetry between absorption and emission affects the photoelectron distribution, both in probability and emission angle.

Laser-assisted photoionization is a cornerstone of attosecond science, used in the temporal characterization of XUV radiation, such as high-order harmonics [3–5], the measurement of attosecond pulses [6, 7] and in many applications, especially the investigation of photoionization dynamics in atoms [8–12], molecules [13–16] and solids [17–19]. Attosecond precision is obtained by laser-assisted photoionization with consecutive, phase-locked harmonics in the so-called RABBIT technique (Reconstruction of attosecond beating by interference of two-photon transitions) [7]. Initially used for the characterization of attosecond pulses, the RABBIT technique has been widely applied to measurements of photoionization time delays in a variety of systems. Recently, angle-resolved RABBIT studies have shown that atomic ionization delays depend on the direction of emission of the photoelectrons [20, 21] and that the photoelectron angular distributions (PAD) vary with the delay between the attosecond pulse train and the probe field [21]. Time- and angle-resolved electron wavepackets have also been studied using a variation of the RABBIT technique, which is based on both even and odd harmonics and leads to an up-down asymmetry in the PADs due to parity mixing of the final states [22, 23].

From a theoretical perspective, angle-resolved laser-assisted photoionization is a long-studied subject [24–27], and angle-resolved RABBIT has been investigated using lowest-order perturbation theory [28–30] as well as by solving numerically the time-dependent Schrödinger equation (TDSE) [20, 31, 32]. While a modification of PADs over delay has been predicted at moderate IR intensities \((10^{11} – 10^{13} \text{ W/cm}^2)\) [33], and the emission of photoelectrons perpendicular to the laser polarization has been discussed in the strong field regime \((10^{13} – 10^{14} \text{ W/cm}^2)\) [34], the underlying reason for delay-dependence of PADs in RABBIT experiments is still not understood.

Here, we show that the RABBIT scheme leads to incomplete quantum interference resulting in the periodic emission of photoelectrons perpendicular to the laser polarization. This result is a consequence of Fano’s propensity rule and it is observed both experimentally and in theoretical calculations based on many-body perturbation theory. Our findings are verified to be universal and Fano’s propensity rule is found to be satisfied in above-threshold transitions for all studied noble gases.
In our experiments, we focus an XUV attosecond pulse train, together with a fraction of the fundamental IR field ($\hbar \omega = 1.58 \text{ eV}$) used for the generation of the pulse train, with a variable delay, into a gas jet of argon atoms. Because the XUV pulse train is composed of coherent odd harmonics with frequency ($n\hbar \omega$), the combined XUV+IR interaction results in above-threshold ionization with increasing sideband order. In our experimental conditions, the values of $\beta_2$ for SB 16 (shown by a dashed line) are affected by the presence of nearby autoionizing resonances and will not be considered in the following.

The interference of the two paths leading to a sideband results in an oscillation of the angle-integrated sideband signal as a function of the relative delay, $\tau$, between the XUV and IR pulses, as can be seen in Fig. 1(a). The sideband signal oscillates according to [7]:

$$P_{SB}(\tau) = A + B \cos(2\omega\tau + \Delta\phi), \quad (1)$$

where $A$, $B$ and $\Delta\phi$ are specific constants for each sideband.

The differential ionization cross-section $d\sigma^{(N)}/d\Omega$ after interaction (absorption or emission) with $N$ photons can be decomposed as a sum of Legendre polynomials $P_n$ weighted by the asymmetry parameters $\beta_n$ [36]

$$\frac{d\sigma^{(N)}}{d\Omega} = \frac{\sigma_0}{4\pi} \left[ 1 + \sum_{n=1}^{N} \beta_n P_n(\cos \theta) \right], \quad (2)$$

where $\sigma_0$ is the total ionization cross section and $\theta$ is the angle of emission with respect to the laser polarization. In the case of a two-photon transition, all the final states have the same parity such that the odd-order asymmetry parameters, $\beta_1$ and $\beta_3$, vanish, giving rise to an up-down symmetric PAD described by $\beta_2$ and $\beta_4$ [25].

Fig. 1(b) presents the delay-dependent PAD of SB 14. Since the signal oscillates as a function of the delay, we normalize the PAD by the angle-integrated signal in order to facilitate the comparison of the PADs at different delays. Around the maxima of the SB oscillations, the PADs are strongly deformed. The two insets in Fig. 1(a) present the PADs at the maximum and minimum of the oscillations, showing clearly the emission of electrons perpendicularly to the polarization axis, close to the minimum of the SB signal. The experimental setup and [35] for details on the VMIS. The interference of the two paths leading to a sideband results in an oscillation of the angle-integrated sideband signal as a function of the relative delay, $\tau$, between the XUV and IR pulses, as can be seen in Fig. 1(a). The sideband signal oscillates according to [7]:

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Figure 2 shows (a) the delay-dependent PAD for SB 14, calculated with the method presented below and (b) the extracted $\beta_2$ parameters for sidebands 14 to 22. The

FIG. 1: Delay-dependent PADs. (a) Angle-integrated signal measured in SB 14 (squares) and fitted oscillation (gray line). (b) Interpolated delay-dependent PAD measured in SB 14 normalized to the angle-integrated signal at each delay. (c) Delay-dependent $\beta_2$ extracted from the experimental angle-resolved RABBIT for SB 14 to SB 22. The insets in (a) show the PAD of SB 14 reconstructed from the measured $\beta$ parameters for delays indicated by the colored squares. The solid arrows indicate the polarization axis $\hat{\epsilon}$ and the dashed arrows correspond to the photoelectron momentum $\hat{k}$.

FIG. 2: Theoretical delay-dependent photoelectron angular distribution. (a) Delay-dependent PAD for SB 14 normalized to the angle-integrated signal at each delay. (b) Delay-dependent asymmetry parameter $\beta_2$ extracted from the calculated PADs for SBs 14 to 22.
delay dependence of the PADs and the sub-cycle variation of the \( \beta_2 \) parameter observed in the experiment is well reproduced by our theoretical results. The weaker oscillation amplitude observed in the experiment can be attributed to several factors such as delay sampling and jitter as well as limited angular resolution of the VMIS or too low signal close to the minima of the SB oscillations. Relative phase shifts between the \( \beta_2 \) oscillations of different sidebands can be observed in both experiment and theory. In the theory, this is solely attributed to the phase of the matrix elements, while in the experiment, part of these phase shifts can also be attributed to the positive chirp of the attosecond pulses [37], which was not included in our calculations for simplicity.

The angular distributions shown in Fig. 2(a) are calculated according to (we now use atomic units \( \hbar = m = e = 4\pi\varepsilon_0 = 1 \) unless stated otherwise)

\[
P_{\text{SIG}}(\theta, \tau) = \int d\phi \sum_m \sum_{L,\lambda} Y_{lm}(\theta, \phi) \exp[-iL\pi/2 + i\eta L]
\times (M^{(+)}_{\lambda L m} e^{i\omega \tau} + M^{(-)}_{\lambda L m} e^{-i\omega \tau})^2,
\]

where

\[
M^{(\pm)}_{\lambda L m} = \sum_p \frac{\langle q | z | p \rangle \langle p | z + \delta \eta \text{RPAE} | a \rangle}{\varepsilon_a - \varepsilon_p + \omega(2n + 1)},
\]

are two-photon transition matrix elements within the dipole approximation from an initial state \( a \) (the 3p orbital of argon with energy \( \varepsilon_a = -15.76 \text{ eV} \)) to a final electron continuum state \( q \) with asymptotic phase, \( \eta_L \), and angular momentum, \( L = 1, 3, 5 \), via all virtual electron states \( p \) with energy, \( \varepsilon_p \), and angular momentum, \( \lambda = 0, 2 \). The common linear polarization of XUV and IR fields ensures conservation of the magnetic quantum number, \( m \), from the initial state. The amplitudes of XUV and IR fields are set to one for clarity. Our calculations are based on a one-electron Hamiltonian, with a Dirac-Fock potential plus a correction that ensures the correct long-range potential for ionized photoelectrons [28]. Electron correlation effects are included by self-consistent changes in the potential, \( \delta \eta \text{RPAE} \), known as the Random Phase Approximation with Exchange (RPAE), for absorption of the XUV photon [38].

Figure 3 shows the ratio between the transition matrix elements [Eq. (4)] with absorption and emission of an IR photon from the same intermediate energy and angular momentum in various noble gas atoms, reached from the outer shell of helium (1s), neon (2p) and argon (3p), or an inner-shell of krypton (3d). The absorption/emission ratio is larger than one when angular momentum is increased, \( L = \lambda + 1 \) [Fig. 3(a)]. Conversely, the emission/absorption ratio is larger than one when angular momentum is decreased, \( L = \lambda - 1 \) [Fig. 3(b)]. This behaviour agrees with Fano’s propensity rule and extends it to the case of laser-assisted photoionization. As illustrated in Fig. 3(c), the transitions indicated by bold arrows are favored compared to those represented by dashed arrows. In all studied atoms, the ratios show an universal decreasing trend with energy that depends on the final angular momentum of the electron. As in the original case of a one-photon transition between a bound and a continuum state [2], the physical origin of the propensity rule comes from the increase of the centrifugal potential with angular momentum.

It is interesting to compare our results with that of the soft-photon approximation (SPA), which is a commonly used approximation to model laser-assisted photoionization [26]. Within the SPA, all interactions of the emitted photoelectron with the parent ion are neglected, which prevents photoemission perpendicular to the polarization of the fields. According to Eq. (2), this implies that the SPA asymmetry parameters, \( \beta_{2}^{\text{SPA}} \) and \( \beta_{4}^{\text{SPA}} \), are simply linked via the relation [27, 32]

\[
\beta_{4}^{\text{SPA}} = \frac{4}{3} (\beta_{2}^{\text{SPA}} - 2).
\]

In order to test the validity of this approximation we calculate \( \beta_{4}^{\text{SPA}} \) using the value of \( \beta_{2} \) from our simulations. The resulting value of \( \beta_{4}^{\text{SPA}} \) does not correspond to that of real SPA calculation but rather provides an insight on the validity of this approximation when compared to
our more advanced calculations. Fig. 4(a) presents the asymmetry parameters $\beta_2$, $\beta_4$ and $\beta_4^{\text{SPA}}$ as a function of the delay between the IR and XUV pulses. The small insets show the angular distribution at delays corresponding to the dotted lines. (b) Variation of the final-state-resolved sideband probability $A_{Lm}$ different final states. (c) Schematic representation of Fano’s propensity rule for the case of sideband formation from two intermediate continuum states with angular momentum $\lambda$.

Fig. 4(b) presents the variations of $|A_{Lm}(\tau)|^2$ for the different final states as a function of delay. Around the maximum, the contribution of the different final states is similar, with the channel with lowest angular momentum being slightly dominant. The fact that the final states have similar contribution is due to Fano’s propensity rule since, for each final state with angular momentum $L$ reached from intermediate states with angular momentum $\lambda$, there is one path with an enhanced transition amplitude and one path with a reduced transition amplitude, as sketched in Fig. 4(c). However, as the delay approaches half of the sideband oscillation period, the contribution of the states with lowest angular momentum drops significantly and the higher angular momentum states become dominant, resulting in a strong modification of the angular distribution. As we will show, this behavior is also a consequence of Fano’s propensity rule.

To address this point, we now investigate why the contribution of the states with highest angular momentum does not cancel at the minimum of the sideband. In the case of a single intermediate channel, we can express $|A_{Lm}(\tau)|^2$ as:

$$ |A_{Lm}(\tau)|^2 = \left| M^{(+)}_{\lambda Lm} \right|^2 \left| M^{(-)}_{\lambda Lm} \right|^2 + 2 \left| M^{(+)}_{\lambda Lm} \right| \left| M^{(-)}_{\lambda Lm} \right| \cos(2\omega \tau + \Delta \varphi_{Lm}) $$

where $\Delta \varphi_{Lm} = \arg\left(M^{(+)}_{\lambda Lm} M^{(-)\dagger}_{\lambda Lm}\right)$. This can be easily extended to the case where there are multiple intermediate channels by summing them coherently. We now introduce the oscillation contrast,

$$ R_{Lm} = \frac{2 \left| M^{(+)}_{\lambda Lm} \right| \left| M^{(-)}_{\lambda Lm} \right|}{\left| M^{(+)}_{\lambda Lm} \right|^2 + \left| M^{(-)}_{\lambda Lm} \right|^2}, $$

which takes values between 0 and 1 and represents the ratio between the amplitude of the oscillations and their mean value. When $R_{Lm} = 1$, the amplitude of the oscillations is as large as the mean value and thus the minima of $|A_{Lm}(\tau)|^2$ are zero. This condition is fulfilled only if $\left| M^{(+)}_{\lambda Lm} \right|^2 = \left| M^{(-)}_{\lambda Lm} \right|^2$, which is the case in the SPA (see supplemental material). However, if $\left| M^{(+)}_{\lambda Lm} \right|^2 \neq \left| M^{(-)}_{\lambda Lm} \right|^2$, the ratio $R_{Lm}$ becomes smaller than 1, which means that $|A_{Lm}(\tau)|^2$ will never completely cancel. Figs. 3(a) and (b) show that the asymmetry between absorption and emission increases with the angular momentum of the final state. This results in the fact that $R_{Lm}$ is always smaller for states with higher angular momentum, which means that the sideband minima are shallower (deeper) at higher (lower) angular momentum. In this way, the modification of the PAD around the minima of the sideband oscillations is a direct consequence of Fano’s propensity rule. Because the SPA does not include Fano’s propensity rule, it works well at the maximum of the sidebands, where all allowed final angular momentum momentum states have comparable contributions, but not at the minimum of the sidebands, where
the high angular momentum states dominate over the low angular momentum states.

In conclusion, we have studied the angle-resolved quantum interference of photoelectrons created by an XUV attosecond pulse train combined with an IR field. When the relative delay of the fields is such that the interference is destructive, the cancellation of the two paths is incomplete, and leads to a strong deformation of the photoelectron angular distribution. The effect is universal and can be interpreted by extending Fano’s propensity rule to above-threshold ionization processes. Increased understanding of above-threshold ionization processes is of great importance for the development of attosecond physics, but also of general interest in other areas of physics where continuum-continuum transitions are present.

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**I. SUPPLEMENTAL MATERIAL**

In the main text we have shown the break-down of the soft-photon approximation (SPA) relation between the asymmetry parameters $\beta_2$ and $\beta_4$ in argon in Fig. 4. Here, we show in Fig. 5 that we observe a similar break-down of the SPA in our experimental data. In addition, in Fig. 6 we show that our calculations also predict a break-down of the SPA in helium, with an even larger deviation of $\beta_4^{\text{SPA}}$ from the value of $\beta_4$ extracted from our simulated PADs. Furthermore, we observe the apparition of lobes at 90° at the minimum, in agreement the predictions in [33] based on solving the time-dependent Schrödinger equation in the single active electron approximation.

In the following we show that SPA is inconsistent with Fano’s propensity rule, which states that absorption (emission) of a photon favors increasing (decreasing) the angular momentum of the electron.

**FIG. 5**: Delay dependent $\beta$ parameters in argon experimentally measured. Asymmetry parameters $\beta_2$ and $\beta_4$ extracted from the measured angular distributions in sideband 18, and $\beta_4^{\text{SPA}}$ calculated using Eq. 5.

**FIG. 6**: Delay dependent $\beta$ parameters in helium. The small insets show the angular distribution at delays corresponding to the dotted lines.

The complex amplitude for laser-assisted photoioniza-
tion within the SPA is given by [39]

\[ T_{fa}(n) = \frac{1}{2} \mathbf{A}_\gamma \cdot \mathbf{k}_f J_n \left( \frac{A_0}{\omega} \cdot \mathbf{k}_f, A_0^2/8\omega \right) \tilde{\phi}_a(\mathbf{k}_f), \]  

(10)

where \( \mathbf{A}_\gamma \) is the vector potential amplitude of the XUV (photoionizing) field and \( A_0 \) is that of the laser (assisting) field, \( \mathbf{k}_f \) is the final momentum of the photoelectron described by a plane wave,

\[ \phi_{k_f}(r) = \left( \mathbf{r} \mid \mathbf{k}_f \right) = \frac{1}{(2\pi)^{3/2}} \exp(i\mathbf{k}_f \cdot \mathbf{r}) \]  

(11)

with energy, \( \epsilon_f = k_f^2/2 \), and momentum normalization. The final state is reached after exchange of \( n \) laser photons, \( \omega \), with an interaction amplitude given by the generalized Bessel function, \( J_n(u,v) \), and one XUV photon, \( \omega_\gamma \), from the initial state, \( \alpha \) with energy, \( \epsilon_\alpha \), and Fourier transform, \( \tilde{\phi}_a(\mathbf{k}_f) \). In Eq. (10) the scalar product, \( \mathbf{A}_\gamma \cdot \mathbf{k}_f = A_\gamma k_f \cos \theta_f \), shows that no photoelectrons will be emitted perpendicular to the laser field polarization, \( \theta_f = \pi/2 \), regardless of the number \( n \) of laser photons exchanged within the SPA. In the limit of a weak assisting field, the generalized Bessel function with one photon exchange reduces to

\[ J_{n+1}(A_0/\omega \cdot \mathbf{k}_f, 0) \approx \pm \frac{1}{2} \frac{A_0 \cdot \mathbf{k}_f}{\omega} = \pm \frac{A_0}{2\omega} k_f \cos \theta_f, \]  

(12)

which gives the two-photon perturbation amplitude

\[ T_{fa}(\pm 1) = \pm \frac{A_f A_0}{4\omega} k_f^2 \cos^2 \theta_f \tilde{\phi}_a(\mathbf{k}_f), \]  

(13)

where \( + (\pm) \) corresponds to absorption (emission) of a laser photon with energy conservation,

\[ \epsilon_f - \epsilon_\alpha = \omega_\gamma \mp \omega. \]  

(14)

The fact that Eq. (13) only differs by a sign between absorption and emission implies that absorption and emission processes have the same strength within the SPA to a given final momentum, \( \mathbf{k}_f \). As a sidenote, Eq. (13) can be easily obtained by evaluating the two photon (XUV±IR) matrix element with all intermediate and final states replaced by plane waves in Eq. (4) in velocity gauge (\( z \to p_z \)). The final plane wave state in Eq. (11) can be expanded on partial waves,

\[ \phi_{k_f}(r) = \frac{4\pi}{(2\pi)^{3/2}} \sum_{\ell,m} i \ell j_\ell(k_f r) Y_{\ell m}(\hat{r}) Y_{\ell m}(\hat{k}_f), \]  

(15)

where \( j_\ell(k_f r) \) are spherical Bessel functions and \( Y_{\ell m} \) are spherical harmonics, which implies that the contribution of a given angular momentum \( L \) and \( M \) is given by

\[ \langle Y_{LM} \mid \mathbf{k} \rangle = \frac{4\pi}{(2\pi)^{3/2}} i^L j_L(k r) Y^{*}_{LM}(\hat{k}). \]  

(16)

By combining Eqs. (13) and (16), we see that absorption and emission processes to a given partial wave have the same strength from any final momentum, \( \mathbf{k}_f \). Adding up coherently all the contributions from all possible final directions of the photoelectron, \( \mathbf{k}_f \), will also result in equal strength of absorption and emission processes,

\[ \epsilon^{(z)}_{fLM} = \int d\Omega_{k_f} \langle Y_{LM} \mid \mathbf{k}_f \rangle T_{fa}(\pm 1) = \pm \frac{4\pi}{(2\pi)^{3/2}} i^L j_L(k r) A_\gamma A_0 k_f^2 \xi_{LM} \tilde{\phi}_a(\mathbf{k}_f) \]  

\[ \times \frac{4\pi}{3} \int d\Omega_{k_f} Y^*_{LM}(\mathbf{k}_f) Y^*_0(\hat{k}_f), \]  

(17)

where we further assumed the initial state to be an \( s \)-wave, so that it can be pulled out from the angular integration of the final momentum. The remaining angular integral is simply a Clebsch-Gordan coefficient equal to \( -1/\sqrt{3} \) for \( L = 0 \) and \( \sqrt{2}/\sqrt{3} \) for \( L = 2 \), respectively. This shows that Fano’s propensity rule is not recovered within the SPA.

Since the SPA does not predict different strengths between absorption and emission depending on the angular momentum of the final state, \( L \), the oscillation contrast is maximal and the same for all the angular channels, \( R_{Lm}^{SPA} = 1 \). This implies that within SPA there will be no delay-dependent modifications of the PAD. SPA works well around the maxima of the sidebands, where the perpendicular photoionization is relatively small, because the contributions to different \( L \) are similar. In contrast, SPA fails to explain the drastic modification of the PAD close to the sideband minima. However, as the photoelectron energy increases, \( R_{Lm} \) tends towards 1, and, therefore, to the delay-independent value predicted by the SPA. This is particularly clear in the case of helium [Fig. 7(a)]. The energy dependence of \( R_{Lm} \) for Ar [Fig. 7(b)] is complex due to the Cooper minimum but also converges towards 1 at high enough kinetic energy.
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