Controlling the 2p Hole Alignment in Neon via the 2s–3p Fano Resonance

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We study the state-resolved production of neon ion after resonant photoionization of Ne via the 2s–3p Fano resonance. We find that by tuning the photon energy across the Fano resonance a surprisingly high control over the alignment of the final 2p hole along the polarization direction can be achieved. In this way hole alignments can be created that are otherwise very hard to achieve. The mechanism responsible for this hole alignment is the destructive interference of the direct and indirect (via the autoionizing 2s−13p state) ionization pathways of 2p. By changing the photon energy the strength of the interference varies and 2p-hole alignments with ratios up to 19:1 between 2p0 and 2p±1 holes can be created—an effect normally only encountered in tunnel ionization using strong-field IR pulses. Including spin-orbit interaction does not change the qualitative feature and leads only to a reduction in the alignment by 2/3. Our study is based on a time-dependent configuration-interaction singles (TDCIS) approach which solves the multichannel time-dependent Schrödinger equation.

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I. INTRODUCTION

Fano resonances [1] appear in almost any field of physics ranging from atomic physics to solid-state physics and to optics [2]. Their most characteristic feature is the asymmetric line profile [3] which results from the coherent interference of a direct continuum channel and an indirect channel which involves a discrete quasi-bound state [4]. These asymmetric line shapes have been first discussed in atomic physics in the context of photoabsorption [5] and electron scattering [6].

In the last decades, there has been an increasing interest in Fano resonances in the presence of strong-field [7, 8] and ultrashort [9, 10] pulses. Strong-field pulses modify the ionization continuum and alter [11] or even destroy [8] the characteristic Fano profile. With attosecond and femtosecond pulses the electron motion of an autoionization process can be studied [12, 13]. Also the interplay of Fano resonances with free-electron laser pulses has been investigated [14].

With the rapid advances in laser technology, it is nowadays possible to study the influence of the details of the ionization process on the parent ion and not only on the ionized photoelectron [15, 16]. Here, transient absorption spectroscopy has been used to probe population and coherences within the parent ion [17]. The high control of the delay between the pump and probe pulses makes it possible to measure even the sub-cycle ionization dynamics and the hole population build-up [18–20]. Also the magnitude of the magnetic quantum number of the hole can be resolved providing information about the hole alignment within an nl-subshell [16, 17, 19].

In this paper, we show how in photoionization the interference of the direct and indirect ionization pathways results in an unusual ionic state with a highly aligned ionic hole. Specifically, we consider photoionization of neon with a photon energy that is resonant with the autoionizing 2s−3p state. We investigate what influence this resonance has on the ionic hole that is eventually formed in the 2p shell. Our calculations are performed using the time-dependent configuration-interaction singles (TDCIS) approach [21]. The correlation-driven autoionization process (2s−13p0 → 2p−1mε l,m) produces the same final states as the direct 2p photoionization (2s22p6 + γ → 2p−1ε l,m). The constructive and destructive interferences of these two pathways lead to the characteristic Fano profile in the photoionization cross section [12, 22–24]. Also the photoelectron angular distribution (characterized by the asymmetry parameter β) varies strongly across the Fano resonance [24]. In addition, as we demonstrate here, there is also a profound effect on the 2p hole, which cannot be deduced from the angular photoelectron distribution. When tuning the photon energy across the resonance, the hole alignment (the ratio between 2p0 and 2p±1 hole populations) varies dramatically from ratios around 1:6 in the non-resonant case to ratios as large as 19 in the resonant case.

These high ratios, signaling that the hole is dominantly located in the 2p0 orbital, are unusual in the XUV regime and are normally only encountered after tunnel ionization with strong-field IR pulses [18, 25, 26]. The maximum hole alignment occurs when the direct 2p photoionization pathway interferes most destructively with the indirect 2p ionization pathway (2s22p6 + γ → 2s−13p0 → 2p−1mε l,m). The photon energy where this destructive interference is the strongest coincides with the minimum position of the Fano profile. By tuning the photon energy above or below the 2s–3p resonance, one controls how these two pathways interfere and, consequently, one controls the 2p hole alignment.

The rest of the manuscript is structured as follows:
II. THEORY

Our implementation of the TDCIS approach to solve the multichannel Schrödinger equation has been described in previous publications [21, 27]. We have applied our TDCIS approach to a wide spectrum of processes [26], ranging from attosecond photoionization [28] to nonlinear x-ray ionization [29] and strong-field tunnel ionization [19, 20, 30].

The N-body TDCIS wave function reads

$$|\Psi(t)\rangle = \alpha_0(t) |\Phi_0\rangle + \sum_{a,i} \alpha_i^n(t) |\Phi_i^n\rangle,$$

where $|\Phi_0\rangle$ is the Hartree-Fock ground state and $|\Phi_i^n\rangle = \hat{c}_i^\dagger \hat{c}_i |\Phi_0\rangle$ are singly excited configurations with an electron removed from the initially occupied orbital $i$ and placed in the initially unoccupied orbital $a$. Since Eq. (1) describes all $N$ electrons in the atom, an electron can be removed from any orbital. This multichannel approach is very helpful in describing ionization processes with XUV and x-ray light where more than one orbital is accessible. By limiting the sum over $i$, specific occupied orbitals can be picked to be involved in the dynamics, thereby testing the multichannel character of the overall dynamics. Inserting Eq. (1) into the full time-dependent Schrödinger equation, one finds the following equations of motion for the CIS coefficients:

$$i \dot{\alpha}_0(t) = -E(t) \sum_{a,i} (\Phi_0|\hat{\mathcal{Z}}|\Phi_i^n) \alpha_i^n(t),$$

$$i \dot{\alpha}_i^n(t) = (\Phi_i^n|\hat{\mathcal{H}}_0|\Phi_i^n) \alpha_i^n(t) + \sum_{b,j} (\Phi_i^n|\hat{\mathcal{H}}_1|\Phi_j^b) \alpha_j^b(t)$$

$$-E(t) (\Phi_i^n|\hat{\mathcal{Z}}|\Phi_0) \alpha_0(t) + \sum_{b,j} (\Phi_i^n|\hat{\mathcal{Z}}|\Phi_j^b) \alpha_j^b(t),$$

where $\hat{\mathcal{H}}_0 = \sum_n \left[ \frac{\hat{p}_n^2}{2} - \frac{Z}{\hat{r}_n} + \hat{V}_{MF}(\hat{r}_n) - \imath \eta \hat{W}(\hat{r}_n) \right]$.

The second term in Eq. (2b), which describes the electron-electron interaction, is the only term within the TDCIS theory that leads to many-body effects. Electronic correlation effects, which within TDCIS can only occur between the ionic state (index $i$) and the photoelectron (index $a$), are captured in the interchannel coupling terms ($i \neq j$) where both indices ($a$ and $i$) are changed simultaneously. It means that the ionic state changes due to the interaction with the excited electron. Intrachannel interactions do not change the ionic state ($i = j$) and describe the long-range $-1/r$ Coulomb potential for the excited electron. Intrachannel interaction can be viewed in terms of a one-particle potential and cannot lead to electron-electron correlations. The importance of many-body correlation effects [28, 34] can be easily tested by either allowing (full TDCIS model) or prohibiting (intra-channel TDCIS model) interchannel interactions which are captured in the $\hat{\mathcal{H}}_1$.

III. RESULTS

We begin in Sec. III A with a discussion of the spectral and temporal properties of the 2s–3p Fano resonance in neon, which we exploit in Sec. III B to control the hole alignment by tuning the XUV pulse across the Fano resonance.

A. 2s–3p Fano Resonance

1. Spectroscopic Features

The photoabsorption cross section, $\sigma(\omega)$, of neon around the 2s–3p resonance obtained within TDCIS is shown in Fig. 1, both with and without interchannel coupling between the 2s and 2p shells. They are both calculated via an autocorrelation function (see Refs. [3, 26]). Strictly speaking, the 2s–3p resonance has in principle no line width in the intrachannel model since the state 2s–3p cannot autoionize and, therefore, lives forever. In Fig. 1, this resonance has a finite width that is artificial and has been introduced by hand for better visualization [42].

With the addition of interchannel coupling of the electrons in the full model, the excited 2s–1p state autoion-
will be in reality photon energies at the minimum p interferes and the overall 2 TDCIS model (blue-dashed line) and the full TDCIS model in an asymmetric Fano line shape [crepancy has no effect on the results and the conclusions. Figure 1: (color online) Photoabsorption cross section of neon in the vicinity of the 2s → 3p resonance for the intrachannel TDCIS model (blue-dashed line) and the full TDCIS model (red-solid line). Fano profile fits [24] give the resonance frequency for the intrachannel TDCIS model ω_intra, and the resonance frequency for the full TDCIS model ω_res. The curve for the intrachannel model is shifted up by +6 Mb for better visualization.

izes to a singly charged ionic state 2p^−1\epsilon l. This indirect ionization of a 2p electron (2s^22p^0 + γ → 2s^−13p → 2p^−1\epsilon l) and the direct one-photon ionization of a 2p electron (2s^22p^0 + γ → 2p^−1\epsilon l) can now interfere, resulting in an asymmetric Fano line shape [1] (see Fig. 1). We fit both curves (with and without interchannel interactions) to the characteristic Fano profile [1, 24] given by

$$\sigma(\omega) = \sigma_0 \frac{(q + \epsilon)^2}{1 + \epsilon^2} + \sigma_b, \quad \text{with} \quad \epsilon = \frac{\omega - \omega_{\text{res}}}{\Gamma/2}.$$  

where q is the Fano parameter describing the asymmetry of the line shape, ω_res is the resonance frequency of the transition, and Γ is the width of the resonance structure. These fits give the transition frequencies for both models as well as the transition width and Fano parameter for the full model: ω_intra = 45.511 eV, ω_res = 45.538 eV, Γ = 31.8 meV, and q = −1.32. The experimentally obtained value for the resonance position is 45.546 eV, for the line width is 13 meV, and the Fano parameter is q = −1.58 [22, 24]. Since the experimental line width is much than twice as small as our theoretical one, the spectral features presented in Figs. 3-4 will be in reality not as broad. Qualitatively, however, this line width discrepancy has no effect on the results and the conclusions.

At frequencies below ω_res, the two ionization pathways constructively interfere and the overall 2p ionization is increased. Above ω_res, the two pathways destructively interfere and the overall 2p ionization is suppressed. The photon energies at the minimum ω_min = 45.559 eV and the maximum ω_max = 45.525 eV are determined visually.

2. Temporal Features

In order to investigate the temporal character of the autoionization process, we resonantly excite neon with a relatively short 2.4 fs pulse of frequency ω_res and a peak intensity of 5.6 × 10^{13} W/cm^2. The duration of this pulse is purposely chosen to be much shorter than the lifetime of the 2s hole given by T_{2s-1} = 1/Γ = 20.7 fs, in our calculations.

Figure 2: (color online) (a) Hole population of the 2s (red-solid line), 2p_0 (green-dashed line), and 2p_{±1} (blue-dashed line) orbitals as well as the ground state depopulation (pink-dashed line) for the full CIS model. The pulse has a Gaussian shape and is 2.4 fs long (FWHM of the intensity) centered around t = 0, and has the carrier frequency ω_res. Also shown are scaled close-ups of the ground state depopulation (b), and of the hole populations of the different orbitals (c-e).

The hole population for the 2s, 2p_0, and 2p_{±1} orbitals as well as the depopulation of the neon ground state are presented in Fig. 2. Note that for linearly polarized light the sign of the magnetic quantum number m is important and the +m and −m electrons behave exactly in the same way when the initial state is an M = 0 state as it is the case for closed-shell atoms. At the end of the pulse, all 2p_m depopulations increase while that of the 2s decreases. The total depopulation, which is the sum of the 2s and all 2p_m orbitals, remains constant, indicating that the 2p and 2s hole populations vary equally but oppositely. Note that in Fig. 2(b-e) the time scale is changed to visually emphasize these temporal trends. This is also consistent within TDCIS, where the depopulation of the ground state can no longer change when the pulse is over [see Eq. (2a)]. Only the hole rearranges with time from the 2s orbital to the 2p orbitals.

This hole rearrangement is the resonant Auger decay (or the autoionization process). The energy released by the hole movement, 26.9 eV, is sufficient to knock the excited electron residing in the 3p shell, which has a binding energy of 2.9 eV, into the continuum [43].
**B. Hole Alignment**

As we have seen in Sec. III A 1, the indirect ionization pathways via the autoionizing $2s^{-1}3p$ state interferes constructively or destructively with the direct ionization pathway depending on the detuning of the photon energy. The spectral information (in Fig. 1) does, however, not contain channel-resolved cross sections. Particularly, it cannot answer the question as to which extent the interference affects all $2p_m$ ionization channels equivalently or whether there is a preferred $m$ ionization channel. A non-uniform behavior would result in different effective ionization rates for $2p_0$ and $2p_{\pm 1}$ and, consequently, in a modified ratio between $2p_0$ and $2p_{\pm 1}$ hole populations compared to the ratio expected for non-resonant one-photon ionization at similar photon energies.

By studying theoretically and experimentally the angular distribution of the photoelectron [24], a large variation of the asymmetry parameter $\beta$ has been found. Therefore, we also expect an variation in the hole ionization rates for $2p_0$ and $2p_{\pm 1}$ ionization channels equivalently or whether there is a preferred $m$ ionization channel.

The relative enhancement of the $2p_{\pm 1}$ ionization channel leading to an $m$-resolved $2p_{\pm 1}$ hole alignment, since an $s$-wave photoelectron can only originate from the $2p_0$ orbital. Such a connection to the ionization states has, however, not been made in earlier studies.

As we can see from Fig. 3(a) the $2p_m$ populations do vary across the resonance. Especially the ionization for $2p_{\pm 1}$ is much more suppressed at $\omega_{\text{min}}$ than for $2p_0$. In the following, we investigate in more details why the ionization of the $2p_m$ orbitals behave so differently by having a closer look at the partial-wave contributions leading to $s$-wave and $d$-wave photoelectrons.

1. **$d$-wave photoelectron**

The $2p_{\pm 1}$ ionization is much more suppressed than $2p_0$ around $\omega_{\text{min}}$ [see Fig. 3(a)]. For $2p_{\pm 1}$, the destructive interference is so strong that it leads to a suppression of almost 2 orders of magnitude compared to non-resonant photon energies. All $2p_m^{-1}\varepsilon d_m$ partial-wave channels show the same degree of suppression. To be more precise, the ratio between $2p_0^{-1}\varepsilon d_0$ and $2p_{\pm 1}^{-1}\varepsilon d_{\pm 1}$ is exactly $4/3$. A detailed analysis shows that this ratio between the $m = 0$ and $|m| = 1$ appears in both, the direct and the indirect, ionization pathways and can be explained by the Wigner-Eckart theorem [35]. Consequently, the behavior of constructive and destructive interference is exactly the same for all $d$-wave channels, $2p_m^{-1}\varepsilon d_m$.

2. **$s$-wave photoelectron**

To generate $2p_0$ holes there exists another ionization channel leading to an $s$-wave photoelectron, i.e., $2p_0^{-1}\varepsilon s_0$. The behavior of this partial-wave channel is different than the behavior of the $2p_m^{-1}\varepsilon d_m$ partial-wave channels [see Fig. 3(a)]. For $2p_0^{-1}\varepsilon s_0$, the destructive interference happens at $\omega_{\text{max}}$ and constructive interference occurs at $\omega_{\text{min}}$.

The overall trend is dominated by $2p_0^{-1}\varepsilon d_m$, since the probability of ejecting an electron from a $p$-orbital into an $s$-continuum is generally much smaller than ejecting the electron into a $d$-continuum [36]. Only around $\omega_{\text{min}}$, where the ionization into a $d$-continuum is strongly suppressed, the situation changes and ionization into the $s$-continuum becomes the dominant ionization channel (corresponding to an asymmetry parameter of $\beta = 0$). The relative enhancement of the $2p_0^{-1}\varepsilon s_0$ partial-wave channel results in a ten times smaller overall suppression for $2p_0$ ionization than for $2p_{\pm 1}$ ionization [see Fig. 3].

3. **The ratio of $2p_m$ hole populations**

In Fig. 3(b), the hole population ratio $2p_0/2p_{\pm 1}$ is shown as a function of the photon energy for the full TDCIS model (orange-solid line) and the intrachannel TDCIS model (green-dashed line). This ratio is a direct measure of hole alignment, where 1 stands for an isotropic hole distribution, $\infty$ for perfect hole alignment along the polarization direction, and 0 for perfect hole
antialignment in the plane perpendicular to the polarization direction.

Strong variations of the hole alignment across the Fano resonance are found resulting in ratios that vary by more than one order of magnitude (between 1.6 and 18). A ratio of 18 means the 2p hole is primarily located in the 2p₀ orbital, and only a 10% chance exists to find the hole in either the 2p₁₋₁ or 2p₁₊₁ orbital. Such strong hole alignment is normally only encountered in the strong field regime where tunnel ionization almost exclusively ionizes the outermost p₀ orbital (when using linearly polarized light) [25, 27].

In the off-resonance limit, the intrachannel TDCIS model and the full TDCIS model approach the same value for the 2p₀/2p±₁ ratio (1.6). Such values are very common in the XUV and x-ray regimes where an almost isotropic distribution of the hole is found with a slight preference for the polarization direction (i.e., m = 0).

The maximum hole alignment is reached when the photon energy is εdmin, located at the minimum of the Fano resonance, which is exactly the energy where the suppression of the dominant ionization channels (leading to 2p−₁ εdmax) is most pronounced, and only s-wave photoelectrons are formed which leave a 2p₀ hole behind.

C. Spin-orbit coupling

Up to now, we have ignored that the 2p shell is actually split due to spin-orbit coupling into two subshells 2pj with j = 1/2 and j = 3/2. As a result, the hole alignment has to be defined with respect to mj and not m0. In particular, the 2pj/2 holes for mj = ±1/2 and m j = ±3/2 have to be compared. Here, mj refers to the projection of the total angular momentum j along the XUV polarization axis. In our TDCIS approach, we consider only the spin-orbit interaction within the ion, where it is the strongest, and we neglect it for the photoelectron (see Ref. [20] for details).

Figure 4 (a) shows the hole populations of 2p±₀/₂, 2p±₁/₂, and 2p±₁/₅, and (b) shows the ratio between 2p±₁/₅ and 2p±₁/₅ defining the hole alignment. Figure 4 shows the same trends as Fig. 3. The mixing of 2p₀ and 2p±₁ orbitals in the spin-orbit case reduces the maximum hole alignment within the 2p₁/₂-shell by ~2/3 in comparison to the non-spin-orbit case [44], which results in a maximum alignment ratio of ~13 instead of 18.

Figure 4: (color online) (a) Hole population is shown as a function of the photon energy for the 2p±₁/₁₀ orbital (solid-red line), the 2p±₁/₅ orbital (dashed-blue line), and the 2p±₁/₅ orbital (dotted-light-blue line). (b) The ratio 2p±₁/₁₀/2p±₁/₅ is shown for the full TDCIS model (solid-brown line) and the intrachannel TDCIS model (dashed-green line). The same pulse parameters as in Fig. 3 has been used.

The maximum hole alignment is reached when the photon energy is εdmin, located at the minimum of the Fano resonance, which is exactly the energy where the suppression of the dominant ionization channels (leading to 2p−₁ εdmax) is most pronounced, and only s-wave photoelectrons are formed which leave a 2p₀ hole behind.

\[
\begin{align*}
|2p_{0,5}^{±0.5}\rangle &= \pm \sqrt{\frac{2}{3}} |2p_{±1,±\frac{1}{2}}\rangle + \frac{1}{3} |2p_{0,±}\rangle \\
|2p_{1,5}^{±0.5}\rangle &= ± \sqrt{\frac{1}{3}} |2p_{±1,±\frac{1}{2}}\rangle + \sqrt{\frac{2}{3}} |2p_{0,±}\rangle \\
|2p_{1,5}^{±1.5}\rangle &= |2p_{±1,±\frac{1}{2}}\rangle
\end{align*}
\]

where |2p₀,σ⟩ refers to the spatial 2p₀ orbital with the spin projection σ. Note that in Sec. III B we focused only the spatial part of the orbitals because the spin-up and spin-down components behave exactly the same [45]. The spin-orbit interaction is treated here in degenerate perturbation theory (see Ref. [20, 37]) where only the impact on the angular momentum is considered. The radial part is unaffected by the spin-orbit interaction which leads to errors of few per cent [38].

By using Eqs. (4a–4c), all populations shown in Fig. 4(a) can be written in terms of the non-spin-orbit-split populations shown in Fig. 3(a), and, consequently, also the alignment ratio in the case of spin-orbit splitting can be expressed in terms of the ratio without spin-orbit splitting as done earlier.

IV. CONCLUSION

We have shown that resonant excitation of the autoionizing 2s−13p state leads to a second ionization pathway that can interfere with the direct 2p photoionization pathway and strongly influences the state of the parent ion. This interference is well known as the origin of the characteristic Fano profile. Also the asymmetry parameter β measuring the angular distribution of the photoelectron varies strongly across the Fano resonance but a direct relation to the hole alignment cannot be made.

We showed that this interference has destructive character at εdmin and creates a dark-state in the photoelectron continuum. As a result, the 2p−₁ εd ionization chan-
nel is strongly suppressed, and the photoelectron is emitted as a pure s-wave. Consequently, the only orbital that is ionized is the $2p_0$ orbital. The imbalance of ionizing $2p_0$ and $2p_{\pm 1}$ orbitals leads to a large hole alignment along the XUV polarization direction.

The ratio between the populations of $2p_0$ and $2p_{\pm 1}$ goes as high as 19—localizing the hole in the $2p_0$ orbital—and is significantly different than the off-resonant value (1.6), which possesses only a slight hole alignment. Strong hole alignments are usually only encountered after tunnel ionization with strong-field IR pulses, where the Keldysh parameter is well below 1 [26]. Here, we used XUV pulses and we are in the perturbative one-photon regime, where the Keldysh parameter is well above 1 and large anisotropies in the hole states are not expected.

When disabling interchannel coupling effects, i.e., disabling the correlation-driven autoionization mechanism of the excited $2s^{-1}3p$ state, no interference of the ionization pathways occurs and no hole alignment modulation appears when tuning across the $2s-3p$ resonance. Including spin-orbit interaction within the ion does not change the picture. Only the strong hole alignment within the $2p_{3/2}$-shell is reduced by a factor 2/3, which still results in a strong hole alignment with ratios up to 13:1 between $2p_{3/2}$ and $2p_{5/2}$ hole populations.

Controlling the hole alignment via the $2s-3p$ Fano resonance serves as an example of how correlation effects can be explicitly targeted and exploited to create new and exotic electronic states in atoms and molecules. Similarly other Fano resonances can be used where the strength of the resonance determines how strongly the hole alignment can be tuned. Furthermore, with a second pulse the Fano resonance could be modified within attoseconds [11] to gain an even larger control of the electronic motion. Also the extension to high-intensity pulses is interesting, which can be realized with currently available seeded free-electron lasers like FERMI [39] or sFLASH [40]. First preliminary results we have obtained suggest that completely different ionization behavior occurs when a Fano resonance is driven by a high-intensity pulse.

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[42] The autocorrelation function is exponentially damped by hand. The damping factor is directly related to the width of the Lorentzian.

[43] These energies were calculated using values found in the National Institute of Standards and Technology (NIST) database [41].

[44] The exact transformation from the hole alignment ratio without spin-orbit coupling, $r_{nols}$, to the ratio when spin-orbit coupling is included, $r_{ls}$, reads: $r_{ls} = 1/3 + 2/3 r_{nols}$. For large hole alignments, the constant can be ignored.

[45] To be more precise, the non-spin-orbit-split hole populations $2p_m$ are the sums of the corresponding spin-up and spin-down hole populations.