

Dipole-phase evolution of laser-dressed autoionizing states in neon

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We present theoretical attosecond transient absorption spectroscopy calculations of neon, focusing on the [He] $2s2p^63p^1P^\circ$ autoionizing state, in an extreme ultraviolet pump, near infrared probe configuration. We find that as the two laser pulses overlap, there is a strong shift in the dipole phase, which we attribute to an effective reduction in the coupling strength between the bound and continuum states as the near infrared pulse makes new pathways accessible. To quantitatively analyze the dipole phase in the presence of perturbed free-induction decay, we demonstrate a method to obtain resonance phase and strength values directly from the dipole spectrum.

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I. INTRODUCTION

Attosecond transient absorption spectroscopy (ATAS) is one of the leading methods for probing electronic dynamics on their natural, attosecond, timescale [1]. Unlike traditional photoelectron spectroscopy that measures ejected electrons, ATAS analyzes light that has been transmitted through a sample, providing time-resolved measurements of both bound-state dynamics and ionization processes [2]. This approach offers unique advantages for studying autoionizing resonances, where interference between discrete bound states and the continuum creates characteristic asymmetric line shapes that can be monitored in real time [3–5].

Attosecond transient absorption spectroscopy has evolved from early measurements of ionization yield [6] to sophisticated phase-resolved spectroscopy [7]. Advances in spectrometer resolution now allow detailed analysis of absorption line shapes, providing access to both amplitude and phase information of the atomic dipole response. This capability is particularly valuable for understanding complex many-body dynamics during strong-field processes, where multiple pathways can contribute to the observed spectral features [8–11]. This was most recently demonstrated in an investigation of the dipole phase during strong-field ionization of xenon [12]. There the observed behavior was attributed to the interplay of strong-field ionization, the time-delayed excitation of multiple excited states, and the polarization of the ground state of the atom. The relative timing of these processes is encoded in the dipole response and made visible through ATAS.

In this work we use theoretical calculations with the *R*-matrix with time-dependence (RMT) code to study the

dynamics of the [He] $2s2p^63p^1P^\circ$ autoionizing state in neon. This resonant state is pumped by an extreme ultraviolet (XUV) pulse and probed by a near infrared (NIR) pulse, causing strong deviation away from typical Fano line shapes. Our results reveal that a strong nonoscillatory phase evolution of the [He] $2s2p^63p^1P^\circ$ resonance stems from field-induced coupling between the autoionizing state and the nearby doubly and triply excited states. We further demonstrate that the observed effects are mediated by electron correlation.

An important challenge in describing these dynamics is the intermediate laser intensity employed. At low intensities, the NIR pulse may be treated as a small perturbation and dynamics can be modeled with traditional ATAS fitting procedures based on Fano line shapes. At high probe intensities, the strong-field dynamics dominate and the detailed atomic structure effects may be ignored, offering a clearer interpretation of the underlying physics [13]. However, at intermediate probe intensities, the regime most commonly encountered in experiments, the situation becomes considerably more complex as multiple processes can contribute on an equal footing.

One mechanism of particular importance in the present study is the optical equivalent of perturbed free-induction decay (PFID), which fundamentally alters the expected Fano line shapes [14–18]. When an autoionizing state is coupled to additional channels by an intense probe field, the assumptions that underlie models of resonance line shapes in absorption spectra break down. This leads to modified line shapes that often cannot be successfully fit even with the best known formalism [13]. Previous studies encountering this phenomenon have avoided quantitative analysis of dipole phase or amplitude, instead focusing on other effects [19]. Here we confirm that conventional fitting procedures fail to capture the dynamics under investigation and we establish the direct dipole calculation approach as a robust alternative for analyzing ATAS data in challenging intermediate-intensity regimes.

II. THEORY

We calculate absorption spectra for neon subject to XUV pump and NIR probe pulses. The XUV pulse has a central photon energy of 45.547 eV, an intensity of

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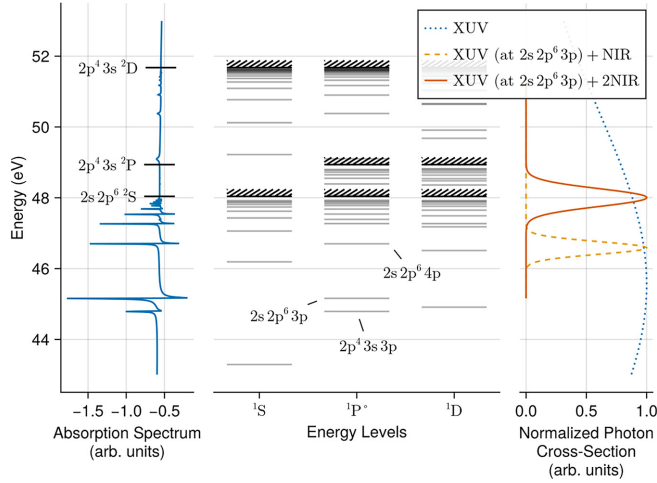


FIG. 1. Atomic structure of neon in the vicinity of the $[\text{He}] 2s2p^6 3p^1 P^o$ state. Energies quoted in this figure, and throughout this paper, are with respect to the neutral neon ground state. These states all lie above the $[\text{He}] 2s^2 2p^5 \ ^2 P^o$ threshold (21.56 eV) and are therefore bound states embedded within the continuum. The locations of the $[\text{He}] 2s^2 2p^4 3s 3p^1 P^o$ (44.79 eV), $[\text{He}] 2s2p^6 3p^1 P^o$ (45.16 eV), and $[\text{He}] 2s2p^6 4p^1 P^o$ (46.70 eV) states and the $[\text{He}] 2s2p^6 \ ^2 S$ (48.04 eV), $[\text{He}] 2s^2 2p^4 3s \ ^2 P$ (48.94 eV), and $[\text{He}] 2s^2 2p^4 3s \ ^2 D$ (51.68 eV) ionization thresholds are indicated. The absorption spectrum for neon subject to a 10^9 W cm^{-2} , 650 as (FWHM) XUV pulse (blue solid line), and normalized cross sections of the 10^9 W cm^{-2} , 650 as (FWHM) XUV (blue dotted line) and $10^{12} \text{ W cm}^{-2}$, 9 fs (FWHM) NIR pulse (orange solid and dashed lines) used in this study are also shown. Each cross section is normalized such that its maximum value is one and the NIR cross sections are plotted with respect to the $[\text{He}] 2s2p^6 3p^1 P^o$ state, to give an idea of what range of states is accessible via two NIR photon absorptions. The XUV (at $2s2p^6 3p$)+2NIR cross section was obtained by convolving the NIR energy spectrum with itself.

10^9 W cm^{-2} , and a Gaussian envelope with a full width at half maximum (FWHM) of seven cycles (650 as). This resonantly excites the autoionizing $[\text{He}] 2s2p^6 3p^1 P^o$ state, but is sufficiently broadband to excite Rydberg series associated with the $[\text{He}] 2s2p^6 \ ^2 S$, $[\text{He}] 2s^2 2p^4 3s \ ^2 P$, and $[\text{He}] 2s^2 2p^4 3s \ ^2 D$ ionization thresholds (see Fig. 1). The NIR pulse has a central photon energy of 1.423 eV (871 nm), an intensity of $10^{12} \text{ W cm}^{-2}$, and a Gaussian envelope with a FWHM of six cycles (9 fs).

Our basis set uses the $[\text{He}] 2s^2 2p^5 \ ^2 P^o$, $[\text{He}] 2s2p^6 \ ^2 S$, $[\text{He}] 2s^2 2p^4 3s \ ^4 P$, $[\text{He}] 2s^2 2p^4 3s \ ^2 P$, $[\text{He}] 2s^2 2p^4 3p \ ^4 P^o$, and $[\text{He}] 2s^2 2p^4 3s \ ^2 D$ residual ion states [20]. We use a close-coupling basis involving these neon residual ion states with a continuum electron, augmented by correlation functions covering 152 configurations where one, two, or three $2s$ and $2p$ electrons have been excited into $3s$, $3p$, $3d$, $4s$, and $4p$ pseudo-orbitals, determined as described in Ref. [21]. This yields energies within 0.08 eV of the literature values for the states under investigation [22–24].

Figure 1 shows the relative atomic structure near the $[\text{He}] 2s2p^6 3p^1 P^o$ state. The energy levels of all states with $1S$, $1P^o$, and $1D$ symmetry are shown, as well as

the $[\text{He}] 2s2p^6 \ ^2 S$, $[\text{He}] 2s^2 2p^4 3s \ ^2 P$, and $[\text{He}] 2s^2 2p^4 3s \ ^2 D$ ionization thresholds. The ground state of neutral neon has $1S$ symmetry, and therefore a single XUV photon absorption can only excite the system into $1P^o$ states. Absorption of an NIR photon can then excite the system into $1S$ or $1D$ states, and absorption of two NIR photons can couple multiple $1P^o$ states via $1S$ or $1D$ states or can excite the system into $1F^o$ states (not shown in Fig. 1).

Figure 1 also shows the absorption spectrum obtained from a calculation using only the XUV pulse, and a measure of the energy cross section of the NIR pulse. The most prominent features in Fig. 1 are the asymmetric absorption line shapes at energies corresponding to the $[\text{He}] 2s2p^6 np^1 P^o$ states.

These asymmetric Fano profiles are due to the autoionizing nature of these states. Since the states lie above the $[\text{He}] 2s^2 2p^5 \ ^2 P^o$ threshold they are embedded within the continuum and therefore competition between the direct, $[\text{He}] 2s^2 2p^5 \ ^2 P^o + \epsilon_{s/d}$, and resonant, $[\text{He}] 2s2p^6 np^1 P^o$, pathways causes interference effects that manifest as asymmetric line profiles [25].

The standard method for reconstructing the atomic dipole proceeds via fitting the resonance line shapes in the absorption spectrum. From Ref. [26], the Fano profile for each resonance takes the form [27]

$$\sigma(\omega) = z_0 \omega \frac{\frac{\Gamma}{2} \cos(\phi) + (\omega - \omega_0) \sin(\phi)}{(\omega - \omega_0)^2 + \frac{\Gamma^2}{4}}, \quad (1)$$

where z_0 , ϕ , ω_0 , and Γ^{-1} are the strength, phase, energy, and lifetime of the resonant state, respectively. In general, z_0 and ϕ are expected to be functions of the time delay τ between the two laser pulses. By convention, τ is the time at which the NIR peak arrives relative to the XUV peak. A positive τ therefore indicates that the NIR peak arrives after the XUV peak.

Clearly, several of the absorption features in Fig. 1 are close enough in energy to overlap, especially in the presence of the strong NIR pulse. The absorption spectrum over some energy range can thus be modeled as

$$\sigma(\omega) = a_0 + a_1 \omega + \sum_i \sigma_i(\omega), \quad (2)$$

where σ_i refers to different resonances in that energy range, each modeled by Eq. (1), and $a_0 + a_1 \omega$ is some empirical background term.

Using RMT we compute $d(t)$, the time-dependent expectation value of the dipole operator. Following Refs. [2,19], the absorption spectrum can be computed from the dipole as

$$\sigma(\omega) = 8\pi\alpha\omega \text{Im} \left(\frac{d(\omega)}{\varepsilon(\omega)} \right), \quad (3)$$

where σ is the optical density, α is the fine-structure constant, ω is the photon energy, and $d(\omega)$ and $\varepsilon(\omega)$ are the Fourier transforms of the time-dependent dipole and electric field, respectively.

Alternatively, because RMT provides $d(\omega)$ directly, we may compute the strength of the resonance z_0 with

$$z_0 = \frac{\Gamma \left| \frac{d(\omega_0)}{\varepsilon(\omega_0)} \right|}{2} \quad (4)$$

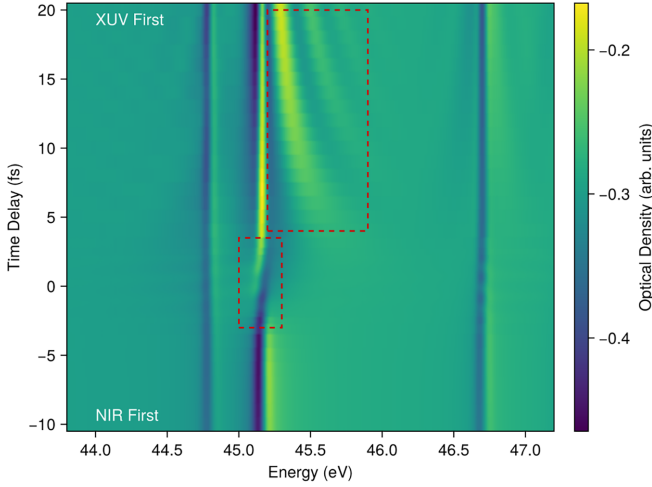


FIG. 2. Delay-dependent absorption of neon subject to a 10^9 W cm^{-2} , 650 as (FWHM) XUV pump and a $10^{12} \text{ W cm}^{-2}$, 9 fs (FWHM) NIR probe pulse. The three absorption lines in the figure correspond to the $[\text{He}] 2s^2 2p^4 3s 3p^1 P^\circ$, $[\text{He}] 2s 2p^6 3p^1 P^\circ$, and $[\text{He}] 2s 2p^6 4p^1 P^\circ$ resonances. At negative time delays the spectrum matches the XUV-only spectrum (Fig. 1), as the NIR has little effect on the ground state. At positive time delays the $[\text{He}] 2s 2p^6 3p^1 P^\circ$ resonance shows fringes associated with the PFID process [13]. In the temporal overlap region (from -4 to 4 fs) the asymmetry of the $[\text{He}] 2s 2p^6 3p^1 P^\circ$ resonance changes, corresponding to a significant change in the dipole phase.

and the phase ϕ with

$$\phi = \arg\left(\frac{d(\omega_0)}{\varepsilon(\omega_0)}\right) - \frac{\pi}{2}, \quad (5)$$

where ω_0 is the energy of the state. To obtain this latter value, we first apply an unwrap algorithm [28] to ensure there are no discontinuities due to the periodic nature of phase, and to obtain both values we use a linear interpolation algorithm [28] to ensure an accurate measurement. These equations are derived in the Appendix.

We perform RMT calculations for pump-probe time delays that span from -10 to 20 fs. For each pump-probe time delay, we calculate the absorption spectrum using Eq. (3) and then fit it with Eq. (2) using three Fano profiles in the range 44 – 47 eV. The three profiles correspond to the $[\text{He}] 2s^2 2p^4 3s 3p^1 P^\circ$, $[\text{He}] 2s 2p^6 3p^1 P^\circ$, and $[\text{He}] 2s 2p^6 4p^1 P^\circ$ excitations. This allows us to obtain the strength z_0 and phase ϕ of the $[\text{He}] 2s 2p^6 3p^1 P^\circ$ resonance. We also extract z_0 and ϕ directly from the energy-dependent dipole response with Eqs. (4) and (5).

III. RESULTS

Figure 2 shows the calculated transient absorption spectrum for neon subject to the XUV pump and NIR probe, as described in Sec. II. For clarity, we restrict the energy range to show only the $[\text{He}] 2s^2 2p^4 3s 3p^1 P^\circ$, $[\text{He}] 2s 2p^6 3p^1 P^\circ$, and $[\text{He}] 2s 2p^6 4p^1 P^\circ$ states and we mark the temporal overlap region and PFID-caused fringes. The optical density is negative for all energies and time delays, indicating that the system absorbs more energy than it emits at all photon frequencies.

There are two key features in Fig. 2, the strong change in phase of the $[\text{He}] 2s 2p^6 3p^1 P^\circ$ resonance (45.16 eV) in the temporal overlap region ($\tau \approx -4$ to 4 fs) and the fringes caused by nonlinear PFID effects at higher time delays. These fringes are only apparent in Fig. 2 at $\tau > 5$ fs, but they begin to affect the shape of the $[\text{He}] 2s 2p^6 3p^1 P^\circ$ resonance at time delays as low as $\tau = 0$ fs, preventing us from using Eq. (1) to study the phase or strength of the dipole in the temporal overlap region. Instead we will use Eqs. (4) and (5) to obtain this information directly from the energy-dependent dipole response, allowing us to quantitatively study the resonance at all time delays. We will begin, however, by briefly justifying the use of this direct dipole method in a more rigorous manner before analyzing the resonance parameters.

Figure 3 shows the result of fitting the absorption spectrum using Eq. (2). At negative time delays, where the NIR pulse has little influence on the spectrum, the equation fits well to the data. However, in the pulse overlap and at positive time delays the fit results often do not match the absorption spectrum well enough to extract meaningful quantitative information. Moreover, the fitting procedure actually fails to converge for time delays between 4 and 7 fs.

The strong modification of the line shape at higher time delays can be explained by the PFID effect on the autoionization process. The dipole response of the autoionizing state can be conceptualized in the time domain as an oscillating exponential decay process [25,26], but if population is driven out of the bound state by a strong external field then this process is truncated, causing a strong modification to the line shape [13,14]. Reference [13] derives an equation to describe these modified line shapes under the assumptions that the autoionizing state interacts only with the continuum and that the population of the state is fully and instantaneously depleted. These conditions can be approximated in our calculations by simplifying the atomic structure and increasing the intensity of the NIR. The line shapes obtained from these simplified calculations (not shown) can then be fit using the equation derived in Ref. [13]. However, at the NIR intensity we employ, these are not reasonable assumptions. The $[\text{He}] 2s 2p^6 3p^1 P^\circ$ state interacts strongly with the surrounding atomic structure and the pulse does not fully or instantaneously deplete the state. Thus, even the modified approach developed in Ref. [13] does not fit the data.

Figure 4 shows the dipole amplitude and phase obtained from the fitting and direct methods [Eqs. (4) and (5)] at the energy of the $[\text{He}] 2s 2p^6 3p^1 P^\circ$ resonance. The phase and amplitude obtained for negative time delays from the two approaches is indeed comparable up to a factor of 2 in the phase. The two methods begin to diverge around $\tau = 0$, as the fits become less robust and the fitting procedure fails completely for time delays between 4 to 7 fs. Figures 3 and 4 together demonstrate that ATAS in this specific regime defies the standard fitting procedure, and from this point on we use the directly calculated dipole data to interpret the underlying physics.

The change in strength in Fig. 4 may be explained through an oscillating dipole model [26]. The excitation of the $[\text{He}] 2s 2p^6 3p^1 P^\circ$ state by the XUV pulse causes an oscillation in the dipole which, if unperturbed, will decay exponentially via autoionization on a timescale of

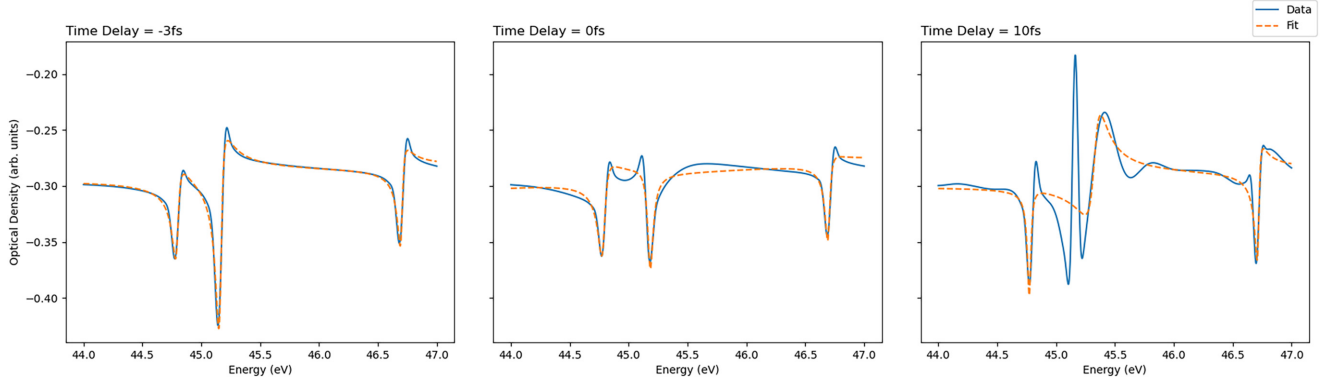


FIG. 3. Absorption spectra (blue solid lines) obtained from calculations with the peak of the NIR pulse arriving at $\tau = -3, 0$, and 10 fs relative to the XUV. Also shown are the fits (orange dashed lines) obtained as explained in Sec. II. At negative time delays the NIR field is predominantly acting on the ground-state system, so the spectrum matches the one in Fig. 1 and fits robustly as the resonances are Fano-like. In the temporal overlap, the NIR acts on the excited system during the excitation, causing the resonances to deviate slightly from the expected Fano profile. At positive time delays the NIR strongly perturbs the excited system, truncating the autoionization process. Here the line shapes, especially that of the $[\text{He}] 2s2p^6 3p^1 P^o$ resonance (around 45.2 eV), no longer resemble a Fano profile.

approximately 50 fs. For negative time delays, this process is essentially unperturbed and the dipole oscillation will decay naturally. This leads to a relatively large dipole amplitude. For time delays around zero, the autoionizing state is depleted by the NIR shortly after excitation, suppressing oscillations and reducing the dipole amplitude. As the time delay increases, the dipole has longer to oscillate before truncation by the NIR, leading to an increase in the dipole amplitude at this energy. If the delay time is large compared to the lifetime of the autoionizing state, we would expect that the unperturbed dipole amplitude would be recovered, but this has not been verified as the autoionization lifetime necessitates prohibitively long calculations.

While we do note that there are 2ω oscillations of the kind described in Ref. [7] present, the most striking feature of the dipole phase shown in Fig. 4 is the large peak in the phase, which follows the intensity profile of the NIR pulse shifted by 0.95 fs. Previous studies of the PFID process [13], as well as our own analytic and numerical investigation

of the oscillating dipole model, suggest that the PFID process does not itself affect the dipole phase. Furthermore, if this phase change were due to the PFID process then the phase would not return to the background phase as early as $\tau = 5$ fs since strong PFID effects are present at much later time delays. This behavior thus indicates some nontrivial dynamics when excitation occurs in the presence of the NIR.

Strong nonoscillatory phase changes have been seen before, most notably in Ref. [26]. In this study krypton was pumped with a strong-field NIR pulse, causing multiphoton ionization, and then holes in the $3d$ orbital were probed with a perturbative XUV laser to stimulate $4p \rightarrow 3d$ transitions. The study identified a nonoscillatory phase change in the resonances associated with these transitions, which is attributed to a coupling effect between the ground and NIR-induced ionized states. However, there are several key distinctions which suggest that different mechanisms are involved. First, the significantly lower NIR intensity, that is, $10^{12} \text{ W cm}^{-2}$ in our study compared to $5 \times 10^{14} \text{ W cm}^{-2}$ in Ref. [26],

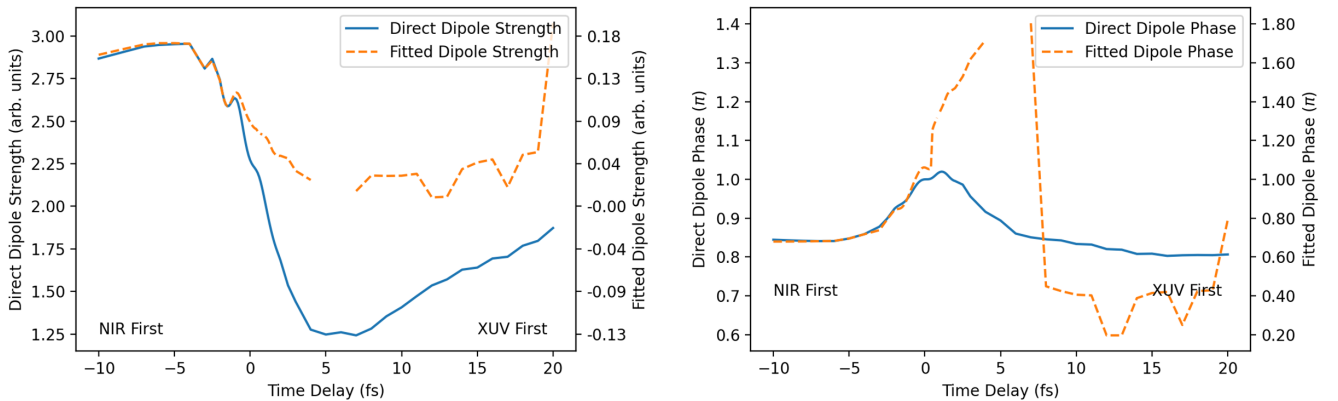


FIG. 4. The blue solid line shows the strength (left) and phase (right) of the $[\text{He}] 2s2p^6 3p^1 P^o$ resonance obtained directly from the dipole spectrum, compared to the phase and strength obtained by fitting to the absorption spectrum (orange dashed line). At negative time delays the quantities from both methods match closely. However, for positive pump-probe time delays, where the peak of the NIR arrives after the XUV, the two methods give wildly different results. In fact, the fitting method fails to converge for time delays between 4 and 7 fs, as shown by the dashed lines.

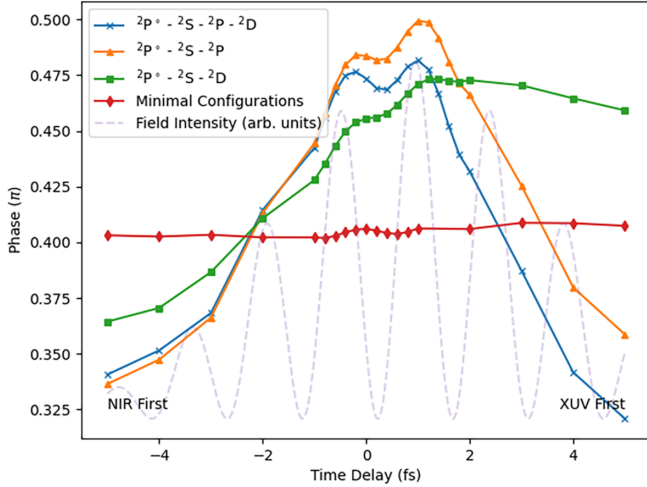


FIG. 5. Phase of the $[\text{He}] 2s^2p^6 3p^1 P^o$ resonance in the temporal overlap region when different atomic structure elements are used in the R -matrix II calculation. The $2P^o - 2S - 2P - 2D$ line (blue crosses) includes the $[\text{He}] 2s^2 2p^5 2P^o$, $[\text{He}] 2s^2 2p^6 2S$, $[\text{He}] 2s^2 2p^4 3s^2 P$, and $[\text{He}] 2s^2 2p^4 3s^2 D$ residual ion states, as well as all configurations in the close-coupling expansion mentioned earlier. The $2P^o - 2S - 2P$ line (orange triangles) omits the $[\text{He}] 2s^2 2p^4 3s^2 D$ residual ion state, and $2P^o - 2S - 2D$ (green squares) omits the $[\text{He}] 2s^2 2p^4 3s^2 P$ residual ion state. Both of these include the same close-coupling configurations as $2P^o - 2S - 2P - 2D$. The “Minimal Configurations” line (red diamonds) includes all the doublet residual ion states above but only includes the $2s^2 2p^5$, $2s^2 2p^6$, and $2s^2 2p^4 3s$ configurations in the close-coupling expansion. No calculations were performed with both the $[\text{He}] 2s^2 2p^4 3s^2 P$ and $[\text{He}] 2s^2 2p^4 3s^2 D$ residual ion states removed, as this caused unacceptably high errors in the convergence tests.

suggests that the so-called highly nonperturbative nature of the cause identified in that study does not translate directly to our scenario. More importantly, the phase shift found in that study is asymmetric; it starts at the background phase value and continuously decreases as the time delay sweeps through the NIR pulse, at variance with our observed phase shift, which is symmetric about the NIR peak and roughly follows the pulse envelope. Finally, the coupling to which the phase shift in that study was attributed was between the ground state and the NIR-induced ionized states whereas no such coupling can occur in our scenario because the ionized states are energetically inaccessible via the NIR pulse.

Therefore, to determine the source of this nonoscillatory phase peak, we perform additional calculations with simplified atomic structure. Figure 5 shows the phase extracted from calculations including either a smaller number of residual ion states or fewer configurations in the close-coupling expansion. The main residual ion states represent the dominant ionization channels, leaving the residual ion in either the $[\text{He}] 2s^2 2p^5 2P^o$ or $[\text{He}] 2s^2 2p^6 2S$ states. The additional residual ion states which are included or excluded are $[\text{He}] 2s^2 2p^4 3s^2 P$ and $[\text{He}] 2s^2 2p^4 3s^2 D$.

When the $[\text{He}] 2s^2 2p^4 3s^2 P$ residual ion state is removed the large increase in phase remains, but the phase does not return to its initial value at high time delays. This residual ion state is close enough to the $[\text{He}] 2s^2 2p^6 3p^1 P^o$ state for the system to ionize into with the absorption of just two NIR

photons, making this a much more likely pathway than into the $[\text{He}] 2s^2 2p^4 3s^2 D$ state. The fact that the initial shift remains suggests that the nonoscillatory phase peak is not related to ionization, at least not into the $[\text{He}] 2s^2 2p^4 3s^2 P$ residual ion state.

The inclusion of a residual ion state allows the system to ionize into that state and also affects the energy and correlations of bound states in the Rydberg series that attach to that residual ion state. The inclusion of configurations in the close-coupling expansion will also affect the energies and correlations of bound states, as well as allowing bound states to be represented that are not part of the Rydberg series converging on one of the included residual ion states.

The most telling detail revealed by Fig. 5, therefore, is the complete elimination of the nonoscillatory phase peak by the removal of configurations in the close-coupling expansion. This shows that it is not the presence or absence of residual ion states that causes these phase dynamics, but rather the electron correlations to nearby bound states.

Up to this point we have been using an oscillating dipole model to explain the underlying mechanics of the system. However, it is instructive to consider an alternative: In the absence of the NIR, the resonance in the absorption spectrum can be modeled as [25]

$$\sigma(\bar{\omega}) = \sigma_0 \frac{(q + \bar{\omega})^2}{1 + \bar{\omega}^2}, \quad (6)$$

where σ_0 is the background optical density far from resonance and $\bar{\omega} = 2(\omega - \omega_0)\Gamma^{-1}$, with ω the photon energy of the laser and ω_0 and Γ the resonance energy and width, respectively. In this model, q is the Fano q parameter defined by [29,30]

$$q = \frac{\langle \psi_b | z | \psi_g \rangle}{\pi V \langle \psi_c | z | \psi_g \rangle}, \quad (7)$$

where $\langle \psi_b | z | \psi_g \rangle$ and $\langle \psi_c | z | \psi_g \rangle$ are the matrix elements coupling the ground state to the bound and continuum states, respectively, and V is the coupling strength between the bound and continuum states [14]. In the absence of a dressing field the coupling strength is equal to the matrix element between the bound and continuum states, $V = \langle \psi_c | \psi_b \rangle$ [14,29].

Although complex dynamics in the presence of a strong field will likely resist any simple model description, it may nonetheless be valuable to develop an intuitive understanding of the mechanisms involved. Thus, we tentatively propose a phenomenological extension to this Fano formalism. An effective coupling strength V^* between the bound and continuum states could in general allow the effect of the NIR to be accounted for in the absorption. In particular, in this case, the NIR dressing field may act to reduce V^* by introducing extra pathways out of the bound state.

To test this, we can quantify how this reduction affects the phase of the dipole by considering the connection between the q parameter and the phase derived in Ref. [25],

$$q = -\cot\left(\frac{\phi}{2}\right). \quad (8)$$

Assuming that $\langle \psi_b | z | \psi_g \rangle$ and $\langle \psi_c | z | \psi_g \rangle$ are not affected by the presence of the NIR—a plausible assumption, considering that these matrix elements represent XUV-caused

transitions—the relationship between ϕ and V^* is therefore

$$\phi = 2 \operatorname{arccot} \left(\frac{-\langle \psi_b | z | \psi_g \rangle}{\pi \langle \psi_c | z | \psi_g \rangle V^*} \right). \quad (9)$$

When $V^* \ll \frac{\langle \psi_b | z | \psi_g \rangle}{\langle \psi_c | z | \psi_g \rangle}$, this will behave with a negative linear relationship

$$\phi \approx -2\pi \frac{\langle \psi_c | z | \psi_g \rangle}{\langle \psi_b | z | \psi_g \rangle} V^*, \quad (10)$$

showing that as V^* decreases (during the temporal overlap region) the phase should increase, as seen in Figs. 4 and 5. Given that the phase shift seen in the [He] $2s2p^63p^1P^\circ$ state has the same profile as the NIR pulse intensity, we hypothesize that in the laser regime used $V^* \approx V - c|\varepsilon(\tau)|^2$ for some constant c .

Further support for such a model is provided by the results shown in Fig. 5. Removing configurations from the close-coupling description should have the effect of closing down the extra laser-enabled pathways out of the bound state. In such a case, we would expect $V^* \approx V$, which should result in a phase that is no longer time-delay dependent, which is the precise behavior observed in Fig. 5. We reemphasize that while this framework does correctly describe the phase shift behavior, further investigation is required to concretely identify exactly what mechanisms cause it.

Any experimental observation of the dipole-phase modification must proceed through fitting Eq. (1) to the absorption spectrum, via the procedure outlined in Sec. II. This poses a significant challenge to experiment, as the PFID process obscures the presence of the phase change in the absorption spectrum at positive time delays. However, the onset of the effect begins at much earlier negative time delays, where Fig. 4 suggests that the fitting procedure is an effective method for obtaining the phase of the resonance. We therefore suggest that collaboration between theory and experiment should allow this phase modification to be observed experimentally and the physics behind the process elucidated.

IV. CONCLUSION

We have performed theoretical ATAS calculations of ground-state neon with a perturbative XUV pump pulse and a midintensity NIR probe pulse, focusing on the [He] $2s2p^63p^1P^\circ$ bound state. In this configuration, there is a strong optical PFID effect that means the absorption spectrum resists standard fitting approaches. We therefore employed the direct dipole approach to perform a quantitative analysis of the phase and strength of the dipole response in this configuration.

This direct dipole method enables the theoretical analysis of laser regimes that previously could not be investigated quantitatively and provides a direct link to the physics, allowing us to begin untangling the processes taking place in complex scenarios. However, while *ab initio* methods like RMT promise a full description of the underlying physics, broader acceptance of such results usually requires some measure of experimental validation, and the direct dipole method requires access to the energy-dependent dipole

response, which is not an experimentally accessible observable. Fortunately, RMT has a rich history of collaborating with experiment and the exceptional agreement between the simulated and experimental observables justifies comparison between the theoretical and experimental insights [5,12,31]. In other words, verifying that there is agreement between the time-delay-dependent absorption spectrum calculated in theory and observed in experiment will give credence to the underlying theoretically computed dipole as a physical reality.

Upon retrieving the dipole phase we found a strong shift away from its background value. This shift was nonoscillatory and therefore not caused by the 2ω coupling that is common in these scenarios. The phase shift did bear similarity to the phase shifts that have been seen in previous strong-field experiments [26]; however, several key distinctions suggest a different cause. While more investigation is needed to determine the precise cause of the shift, we have offered a phenomenological model to describe the phase shift, drawing from the Fano formalism and describing the shift in terms of a field-dependent effective coupling strength between the bound and continuum states involved in the autoionization process.

This study not only cements the direct dipole method as a useful technique for theoretical studies, it also presents an interesting phenomenon that sits outside the usual perturbative and strong-field dichotomy. Interest in these unusual regimes has been increasing recently, and this study showcases the ways in which theoretical techniques are a vital component of their study.

Calculations in this work were performed using the R -matrix II and R -matrix with time-dependence codes, both of which are part of the UK-AMOR suite openly available at [32].

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DATA AVAILABILITY

The data that support the findings of this article are openly available [33].

APPENDIX: DERIVATION OF THE DIRECT DIPOLE EQUATIONS

From the model outlined in Ref. [26], the time-dependent dipole expectation value is

$$d(t) = -2\varepsilon_0 z_0 \sin[\omega_0(t - \tau) + \phi] e^{-(\Gamma/2)(t-\tau)}. \quad (A1)$$

By taking the Fourier transform and applying the rotating-wave approximation, we can show that the energy-dependent dipole response is

$$d(\omega) = \varepsilon_0 e^{-i\omega t} z_0 \frac{(\omega - \omega_0) \cos \phi - \frac{\Gamma}{2} \sin \phi + \left[\frac{\Gamma}{2} \cos \phi + (\omega - \omega_0) \sin \phi \right] i}{(\omega - \omega_0)^2 + \frac{\Gamma^2}{4}}. \quad (\text{A2})$$

Equation (1) can be derived by substituting this into Eq. (3); however, we will proceed by calculating $| \frac{d(\omega_0)}{\varepsilon(\omega_0)} |$ and $\arg(\frac{d(\omega_0)}{\varepsilon(\omega_0)})$.

The relevant real and imaginary parts of the energy-dependent dipole response, after being normalized by the field, are given by

$$\text{Re}\left(\frac{d(\omega_0)}{\varepsilon(\omega_0)}\right) = \frac{-2z_0 \sin(\phi)}{\Gamma} \quad (\text{A3})$$

and

$$\text{Im}\left(\frac{d(\omega_0)}{\varepsilon(\omega_0)}\right) = \frac{2z_0 \cos(\phi)}{\Gamma}. \quad (\text{A4})$$

From this the amplitude of the dipole response is

$$\left| \frac{d(\omega_0)}{\varepsilon(\omega_0)} \right| = \frac{2z_0}{\Gamma}, \quad (\text{A5})$$

which rearranges to give Eq. (4).

To derive Eq. (5) we first generate Table I by considering which quadrant of the complex plane $\frac{d(\omega_0)}{\varepsilon(\omega_0)}$ must be in for any

TABLE I. Quadrant matching table which relates the quadrant of ϕ to the quadrant of $\arg(\frac{d(\omega_0)}{\varepsilon(\omega_0)})$ using the signs of $\text{Re}(\frac{d(\omega_0)}{\varepsilon(\omega_0)})$ and $\text{Im}(\frac{d(\omega_0)}{\varepsilon(\omega_0)})$.

Quadrant of ϕ	$\text{Re}(\frac{d(\omega_0)}{\varepsilon(\omega_0)})$	$\text{Im}(\frac{d(\omega_0)}{\varepsilon(\omega_0)})$	Quadrant of $\varphi = \arg(\frac{d(\omega_0)}{\varepsilon(\omega_0)})$
$-\pi < \phi \leq -\frac{\pi}{2}$	>0	<0	$-\frac{\pi}{2} < \varphi \leq 0$
$-\frac{\pi}{2} < \phi \leq 0$	>0	>0	$0 < \varphi \leq \frac{\pi}{2}$
$0 < \phi \leq \frac{\pi}{2}$	<0	>0	$\frac{\pi}{2} < \varphi \leq \pi$
$\frac{\pi}{2} < \phi \leq \pi$	<0	<0	$-\pi < \varphi \leq -\frac{\pi}{2}$

value of ϕ . We then use the equations

$$\arg(z) = \begin{cases} \arctan\left(\frac{\text{Im}(z)}{\text{Re}(z)}\right), & \text{Re}(z) > 0 \\ \frac{\pi}{2}, & \text{Re}(z) = 0, \text{Im}(z) > 0 \\ \pi + \arctan\left(\frac{\text{Im}(z)}{\text{Re}(z)}\right), & \text{Re}(z) < 0 \\ -\frac{\pi}{2}, & \text{Re}(z) = 0, \text{Im}(z) < 0 \end{cases} \quad (\text{A6})$$

to show that

$$\arg\left(\frac{d(\omega_0)}{\varepsilon(\omega_0)}\right) = \begin{cases} \arctan[-\cot(\phi)], & -\pi < \phi < 0 \\ \frac{\pi}{2}, & \phi = 0 \\ \pi + \arctan[-\cot(\phi)], & 0 < \phi < \pi \\ -\frac{\pi}{2}, & \phi = \pi. \end{cases} \quad (\text{A7})$$

Finally, we apply the identity

$$\tan[\phi + \pi(n + \frac{1}{2})] = -\cot(\phi), \quad n = 0, 1, 2, \dots, \quad (\text{A8})$$

to obtain Eq. (5). When applying this identity we must choose $n = 0$ when $-\pi < \phi < 0$ and $n = 1$ when $0 < \phi < \pi$ to ensure that the quadrants match correctly, as specified in Table I.

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