Generalized Fano parameterization for the Wigner-Eisenbud-Smith time delay in autoionization resonances

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A generalized expression for the complex photoionization amplitude in the vicinity of a Fano resonance is proposed to evalulate the Wigner-Eisenbud-Smith (WES) time delay in the resonant region. The validity of this expression is tested in comparison with accurate numerical calculations empolying the relativistic random phase approximation and multichannel quantum defect theory. The Ne $2s \rightarrow np$ resonance series is used as a convenient test case.

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I. INTRODUCTION

Recent developments in attosecond science have enabled a close examination of ultrafast electron dynamics in atoms and molecules [1–11]. The present study examines the intrinsic Wigner-Eisenbud-Smith time delay ($\tau_{\rm WES}$) [12, 13] associated with photoelectron ejection in the vicinity of an autoionizing resonance. Photoionization time delay information is embedded in the energy dependence of the complex photoionization amplitude and can be studied experimentally using techniques such as attosecond streaking [7] and RABBITT [6, 11]. Measurements of the WES time delay in photoionization provide a signature of the many-electron correlations in atoms and molecules. They also provide new insights into the real-time dynamics of the ionization process [14–16]. A number of theoretical and experimental works have been reported in the literature in the last five years, see e.g. [17–23]. The photoionization WES time delay is given by $\tau_{\rm WES} = d\delta/dE$, where δ is the phase of the photoionization amplitude. Here $\hbar = e = m = 1$. When the phase changes rapidly with energy, there is a significant time delay. A number of earlier studies focussed on the region of the photoionization Cooper minimum where the phase changes rapidly [24, 25]. Most recent studies encompass the autoionizing resonances [26, 27].

Modulations of the phases and their subsequent effects on photoionization time delay due to the presence of an external cage (such as when the atom is embedded in a fullerene molecule) have also been found to be of interest [28–30]. Another region where the phases change rapidly is the region of autoionization resonances, which is governed strongly by electron-electron correlation. The mixing of a bound photoexcitation channel with a continuum channel in a many-electron system results in Fano-Feshbach autoionization resonances. The resonance profiles are often asymmetric owing to the interaction between resonant and background (non-resonant) interfering quantum processes. The Fano resonance shape [31, 32] is of great interest in many contexts, e.g., [33–35]. The resonance profiles have a width Γ about the resonance energy E_r and the profile index q and the correlation factor ρ that define them [31].

The lifetime of an atomic excited state is typically of the order of a few femtoseconds to nanoseconds and can be determined using time-resolved spectroscopy [36]. Resonance lifetime studies also are an important tool to understand electronic structure. The resonance lifetimes are directly related to the photoionization parameters such as oscillator strengths, transition probabilities, and line intensities. Studies of line profiles and resonance lifetimes have important applications in many areas of physics. Of particular importance, they also enable the determination of atomic/ionic relative abundance in stars and other astrophysical objects [37].

The effect of resonances on atomic photoemission time delay has been studied recently for electron transitions of the outermost valence electrons of argon and neon using a coincidence detection technique [38]. The region of autoionization resonance for Mn $3d$ and $4s$ photoionization channels has also been studied recently in which the photoemission time-delay was found to dramatically increase in the region of the $3p \rightarrow 3d$ giant autoionization resonance [39].

The photoionization time delay across the autoionization resonance region is explicitly associated with the resonance lifetime that can be determined with attosecond time delay experiments. As a simple example, for a pure Breit-Wigner resonance, which results from the interference between a single bound-to-bound and a single bound-to-continuum channel, the scattering amplitude, f, is given by [40]

$$f \sim \frac{\Gamma/2}{E_r - E - i\Gamma/2} \tag{1}$$

which gives the WES time delay as

$$\tau = \frac{\Gamma/2}{(E_r - E)^2 + (\Gamma/2)^2} \tag{2}$$

It is seen from Eq. (2) that, for a Breit-Wigner resonance, the maximum value for the time delay is at $E = E_r$, and is $2/\Gamma = 2\tau'$ where $\tau' = 1/\Gamma$ is the lifetime of the excited state. However, in an atomic system in which the autoionization resonance involves additional excitation or ionization channels, this result will be seen to be significantly modified.

In the present work, photoionization time delay in the region of the 2s→np autoionization resonances in atomic neon have been calculated and analyzed. Photoionization and electron-ion scattering have the same final state, but have different initial states. These two processes are related by time reversal symmetry [41]. Accordingly, ingoing wave boundary conditions are employed in photoionization, whereas outgoing wave boundary conditions are employed to describe collisions. In addition, the photoionization WES time delay in the region of the autoionization resonances have been studied in the present work using the Fano formalism [42] to parametrize the autoionization resonance profiles.

II. THEORETICAL METHODOLOGY

A. Generalized Fano parameterization

The autoionization resonances are analyzed using the Fano parameters [31, 32, 42] in terms of which the photoionization cross section in the vicinity of a resonance is given by

$$\sigma(E) = \sigma_0(E)\big[1 - \rho^2 + \rho^2 \frac{(q+\epsilon)^2}{1+\epsilon^2}\big] \tag{3}$$

where σ_0 is the non-resonant (background) cross section, $\epsilon = (E - E_r)/(\Gamma/2)$ with E_r being the resonance energy, Γ the resonance width, q is the Fano shape parameter, and ρ is the correlation coefficient which is required when the resonance is not a pure Breit-Wigner resonance on account of additional continuum channels which may be degenerate at the resonance energy.

In the spirit of this parameterization, we write the photoionization amplitude in the neighborhood of a resonance as

$$f(\epsilon) = f_0\Big[\sqrt{1-\rho^2} + \rho\frac{q+\epsilon}{i+\epsilon}\Big] \propto f_0(\epsilon)\Big[\eta + \frac{q+\epsilon}{i+\epsilon}\Big] \tag{4}$$

with $\eta = \sqrt{1-\rho^2}/\rho$. This parameterization is sonsitent with the cross-section formula (3) because the squared amplitude (4) differs from (3) only by the term

$$2\rho\sqrt{1-\rho^2}\ \frac{q+\epsilon}{1+\epsilon^2}\ \epsilon\ ,$$

which is vanishing at $\epsilon \rightarrow 0$ near the resonance.

The phase of the amplitude, $\arg f(\epsilon)$ is given by

$$\arg f(\epsilon) = -\tan^{-1}\left[\epsilon + \eta\frac{(1+\epsilon^2)}{(q+\epsilon)}\right]^{-1} \tag{5}$$

which yields the time delay

$$\tau = \frac{2}{\Gamma}\frac{1}{1+\left[\epsilon + \eta\ \frac{1+\epsilon^2}{q+\epsilon}\right]^2}\left[1 + \frac{2\eta\epsilon}{q+\epsilon} - \frac{\eta(1+\epsilon^2)}{(q+\epsilon)^2}\right] \tag{6}$$

We note that for a vanishingly small background cross section, i.e., $\eta = 0$ and$\rho = 1$, the time delay (6) reduces to the Breit-Wigner result (2). In addition, the structure of Eq. (6) allows the time delay to be positive or negative. Furthermore, at the resonance energy, $E = E_r$ ($\epsilon = 0$), Eq. (9) reduces to

$$\tau \underset{\epsilon\rightarrow 0}{=} \frac{2}{\Gamma}\frac{1-\eta/q^2}{1+\eta^2/q^2} \tag{7}$$

which is rather different from the Breit-Wigner result. In general, the time delay at the resonance energy must be less than twice the lifetime (the Breit-Wigner result), and can even be negative. In fact, it follows from Eq. (6) that as long as $\eta \neq 0$, the time delay will be negative in the region of $q + \epsilon = 0$. Thus, if $q < 0$, the negative values of time delay will occur for $\epsilon > 0$, i.e., at energies above the resonance position, and vice-versa for $q > 0$. Note also that $q + \epsilon = 0$ corresponds to a minimum in the cross section in the vicinity of a resonance, as seen from Eq. (3). In any case, the WES time delay, in the neighborhood of a resonance, can be conveniently related to the Fano parameters of the resonance.

B. Relativistic-random-phase approximation

The relativistic-random-phase approximation (RRPA) [43, 44] has been used in this work to determine the in-

put parameters for the relativistic multichannel quantum defect theory (RMQDT) [45].

Anatoli's comment: that needs to be explained in greater detail. How do you derive the Fano prameters from the following equations? None of the earlier RMQDT papers explained it to me. If one of the co-authors is puzzled, what happens to the lay reader?!

The RRPA equations are obtained by linearizing the time dependent Dirac-Hartee-Fock (DHF) coupled integro-differential equations. The RRPA accounts for major electron correlations via time forward and time backward ring diagrams, along with corresponding exchange terms. The autoionization resonances have been subsequently generated using RMQDT [46, 47]. The RRPA dipole transition matrix element for a transition effected by the absorption of a photon (represented by the operator $Q_J^{(\lambda)}$, with $J = 1$ for the photon angular momentum and $\lambda = 1$ for the electric dipole interaction) from a bound state $n\kappa$ to a continuum state $\epsilon\bar{\kappa}$ is given by [43]:

$$D_{n\kappa\rightarrow\epsilon\bar{\kappa}} = i^{1-\bar{l}} e^{\iota\delta_{\bar{\kappa}}} \langle \epsilon, \bar{\kappa} \parallel Q_1^{(1)} \parallel n\kappa \rangle_{\rm RRPA}. \qquad (8)$$

In the above equation, $\kappa = \mp\left(j + \frac{1}{2}\right)$ for $j = \left(l \pm \frac{1}{2}\right)$, and $\langle \epsilon, \bar{\kappa} \parallel Q_1^{(1)} \parallel n\kappa \rangle_{\rm RRPA}$ is the reduced matrix element for the electric dipole transition, with $\bar{\kappa} = -\kappa, \kappa \pm 1$. Since the reduced matrix element is complex, the phase of the dipole matrix element (8) is given by

$$\delta_{n\kappa\rightarrow\epsilon\bar{\kappa}}(E) = \tan^{-1}\left\{\frac{{\rm Im}D_{n\kappa\rightarrow\epsilon\bar{\kappa}}}{{\rm Re}D_{n\kappa\rightarrow\epsilon\bar{\kappa}}}\right\}. \qquad (9)$$

The WES time delay (in atomic units) in the photoionization channels is then given by

$$\tau_{WES} = \frac{d}{dE}\Big(\delta_{n\kappa\rightarrow\epsilon\bar{\kappa}}(E)\Big) \qquad (10)$$

III. RESULTS AND DISCUSSION

In the present study, we report the results of RRPA+RMQDT calculations carried out at two different levels of truncation of the RRPA, described below. The different levels of truncation enable us to conduct an examination of selective interchannel coupling, along with effect(s) of the selective coupling upon the parametrization of the resonances and the time delay. The two truncation levels employed are:

(i) 7 relativistic channels from 2p and 2s: $2p_{3/2} \rightarrow \epsilon d_{5/2}, \epsilon d_{3/2}, \epsilon s_{1/2}; 2p_{1/2} \rightarrow \epsilon d_{3/2}, \epsilon s_{1/2}; 2s \rightarrow \epsilon p_{3/2}, \epsilon p_{1/2}$;

(ii) 3 relativistic channels from 2p and 2s: $2p_{3/2} \rightarrow \epsilon d_{5/2}; 2s \rightarrow \epsilon p_{3/2}, \epsilon p_{1/2}$;

Note that the omission of the channels emanating from the 1s shell is essentially irrelevant for the present calculations since the 1s threshold is so far away, almost a keV, from the energies considered in our calculations.

The photoionization cross section of the 2p ($2p_{3/2} + 2p_{1/2}$) shell (with all 7 channels included) in the region of the $2s_{1/2} \rightarrow np$ (spin-orbit unresolved) resonance region is dominated by the $2p_{3/2} \rightarrow \epsilon d_{5/2}$ channel. This is clearly seen in Fig. 1 which shows the absolute values of the complex matrix elements for the possible five relativistically split dipole channels in the region of $2s \rightarrow 3p$ resonance. The present work, therefore, restricts the time delay studies only to the dominant $2p_{3/2} \rightarrow \epsilon d_{5/2}$ channel, as representative of all the possible channels. The phase and time delay results are shown in Fig. 2 for the $2s \rightarrow np$ resonances with n= 3, 4, 5, 6. Across the region for all the resonances shown, the phase changes by π radians and the time delay is positive (time-delay) at an energy just lower than that at the resonance, and it is negative (time advancement) just above.

The negative time delay (time-advancement) is limited by the Wigner causality condition [40] and it corresponds to a repulsive interaction between the escaping photoelectron and the residual ionic field.

Anatoli's comment: I thought the positively charged residual ion always attracts the photoelectron unless there is some exchange modification. Please explain

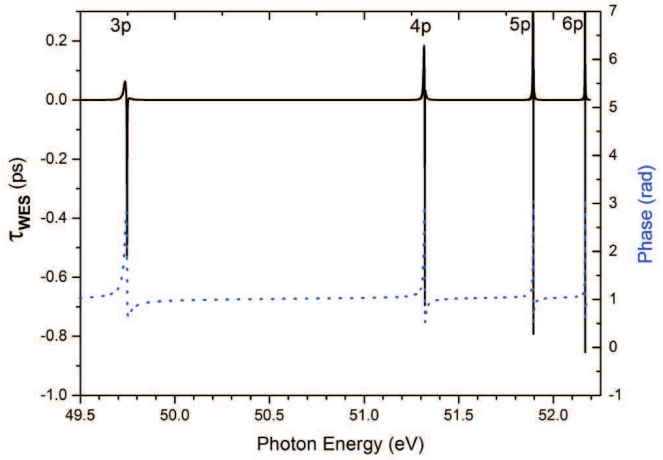

FIG. 1: Absolute amplitudes of the complex matrix elements in the five relativistic dipole channels from the 2p subshells for transitions into the continuum from the 7-channel calculation. The vertical arrow indicates the position of the 3p resonance.

In the 7-channel calculation shown, the $2s_{1/2} \rightarrow np$ resonance lifetime for the is found to be 0.051 ps while the maximum (positive) time delay calculated is 0.061 ps; clearly the time delay is not twice the lifetime as was the case for a Breit-Wigner resonance, discussed above. And, in fact, the same is true for the other resonances shown as well. Furthermore, it is evident from Fig. 2, that the time delay can be positive or negative, unlike the Breit-Wigner case discussed above, where the time delay can only be positive. Thus, the phenomenology exhibited by these resonances is rather different from the Breit-Wigner paradigm.

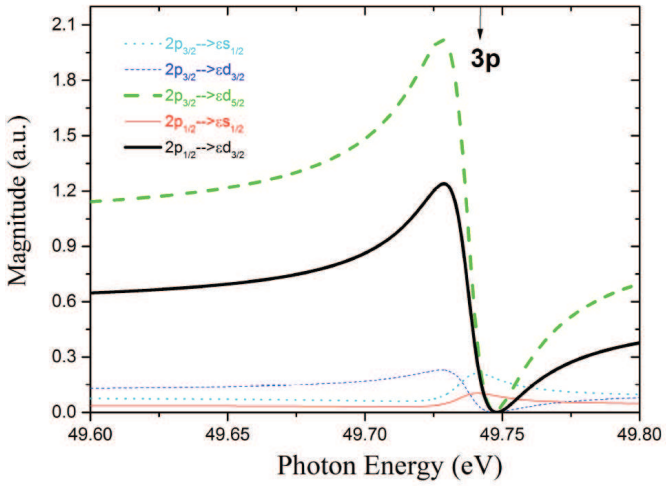

FIG. 2: Photoionization time delay for the $2p_{3/2} \rightarrow d_{5/2}$ channel (black line, left scale) and phase of the matrix element (blue line, right scale) in the region of the $2s_{1/2} \rightarrow np$ $(n = 3 - 6)$ spin-orbit unresolved autoionizing resonances.

TABLE I: Fitted Fano parameters for the $2s \rightarrow np$ resonances. *Anatoli's comment:* Fitted to what? Experimental values in comparison would be very informative

Resonance	$E_r(eV)$	μ	q	ρ^2	$\Gamma(meV)$	σ_0
3p	49.73	0.85	-1.30	0.94	15	8.800
Expt. [49]	45.54	0.83	1.58	0.75	16	
4p	51.3164	0.84	-1.32	0.95	5	8.489
5p	51.8933	0.83	-1.31	0.96	2.24	8.540
6p	52.1674	0.83	-1.35	0.97	1.18	8.440

To demonstrate the utility of Eq. (6), we parametrize the resonances. The Fano parameters for the $2s \rightarrow np$ resonances obtained are given in Table I where it is seen that ρ^2 and q remain fairly constant for the various resonances, as expected in a resonance series. Also, the quantum defects, μ_{nl}, are almost constant, again as expected. Furthermore, the widths, Γ, are expected to fall off as $1/(n^*)^3$, where n^* is the effective quantum number, $n^* = n - \mu_{nl}$, and this is seen to indeed be the case. Using these Fano parameters, the phases and time delays, in the neighborhoods of the resonances, are depicted in Fig. 3 along with the phases and time delays calculated directly.

As seen from Fig. 3, the phases and time delays predicted using Eqs. (5) and (6) and the Fano parameters for the resonances agree reasonably well with these quantities calculated directly. The variation of the phases across the resonances shows excellent qualitative agreement and pretty good quantitative agreement as well. And the same is true for the resonance profile for the time delay, including both the positive maximum below the resonance energy, and the deep negative time delay spike above the resonance energy; the facts that the resonance exhibits negative time delay, and that it occurs above the resonance energy since the Fano q-parameter is negative, are strong indicators that Eq. (6) includes the essential physics of the process.

From these results, seen in close-up in Fig. 3, it is clear that the WES time delay increases with principal quantum number, n, both the positive rise and (particularly) the negative magnitude which reaches the picosecond range for the $2s \rightarrow 6p$ resonance. This might not seem very large, but it is three orders of magnitudes larger than time delays that have been measured in non-resonance regions [6, 7]. In any case, this is no accident and can be easily explained using Eq. (6) which shows that time delay include an overall factor of $1/\Gamma$, where Γ is the resonance width. In a Rydberg series of resonances, as long as there no spectator Auger channels, which is the case for the Ne $2s \rightarrow np$ resonance series, the widths decrease, with increasing n, as $1/(n^*)^3$, n^* being the effective quantum number defined as $n\mu$, where μ is the quantum defect; this is a well-known result of quantum defect theory [46, 47]. Thus, the WES time delay should increase with increasing n as $(n^*)^3$, which is exactly what is seen in Fig. 3. As a consequence, it is evident that the time delay must diverge as $n \rightarrow \infty$. And, this must be true, not only for the particular series studied here, but for any Rydberg series of resonances where are no spectator Augers so that the resonance width tends to zero at the series limit.

Since the resonances scrutinized above are all roughly the same shape, it is not surprising that if Eq. (6) fits one of them, it is likely to fit them all, i.e., going along the series is not as stringent a test of Eq. (6) as we would like. To try a rather different shape, along with attempting to understand the importance of including all relevant channels in the calculation, a truncated RRPA+RMQDT study was performed. Specifically, only a single bound-to-continuum channel $(2p_{3/2} \rightarrow \epsilon d_{5/2})$ was coupled to two bound-to-bound channels $(2s_{1/2} \rightarrow np_{3/2}, np_{1/2})$. Fig. 4 shows the autoionization resonances for such a 3-channel study. Across each resonance, the phase changes by radians and the time delay in the vicinity of each resonance is positive, similar to a pure Breit-Wigner case, but very different from the full 7-channel results for both the time delay and the phase. However, these are not Breit-Wigner resonances; the cross sections (not shown) are rather asymmetric, having values of q of about -2.0. But, since $\eta = 0$, according to Eq. (6), the time delay cannot be negative. Looking at the $2s \rightarrow 3p$ resonance as an example, shown in Fig. 5, this is exactly what is seen. It is also quite clear that the 3-channel time delay and phase results are dramatically different from the 7-channel, thereby demonstrating the importance of including all of the relevant channels in the calculation. Also shown is the magnified time delay across the $2s \rightarrow 3p$ resonance.

In any case, to continue with the 3-channel case as a testing ground for the model based on the Fano parameters, presented in Fig. 5 is the 3-channel WES time delay in the vicinity of the $2s \rightarrow 3p$ resonance obtained from Eq. (9). Both the directly calculated RRPA-RMQDT

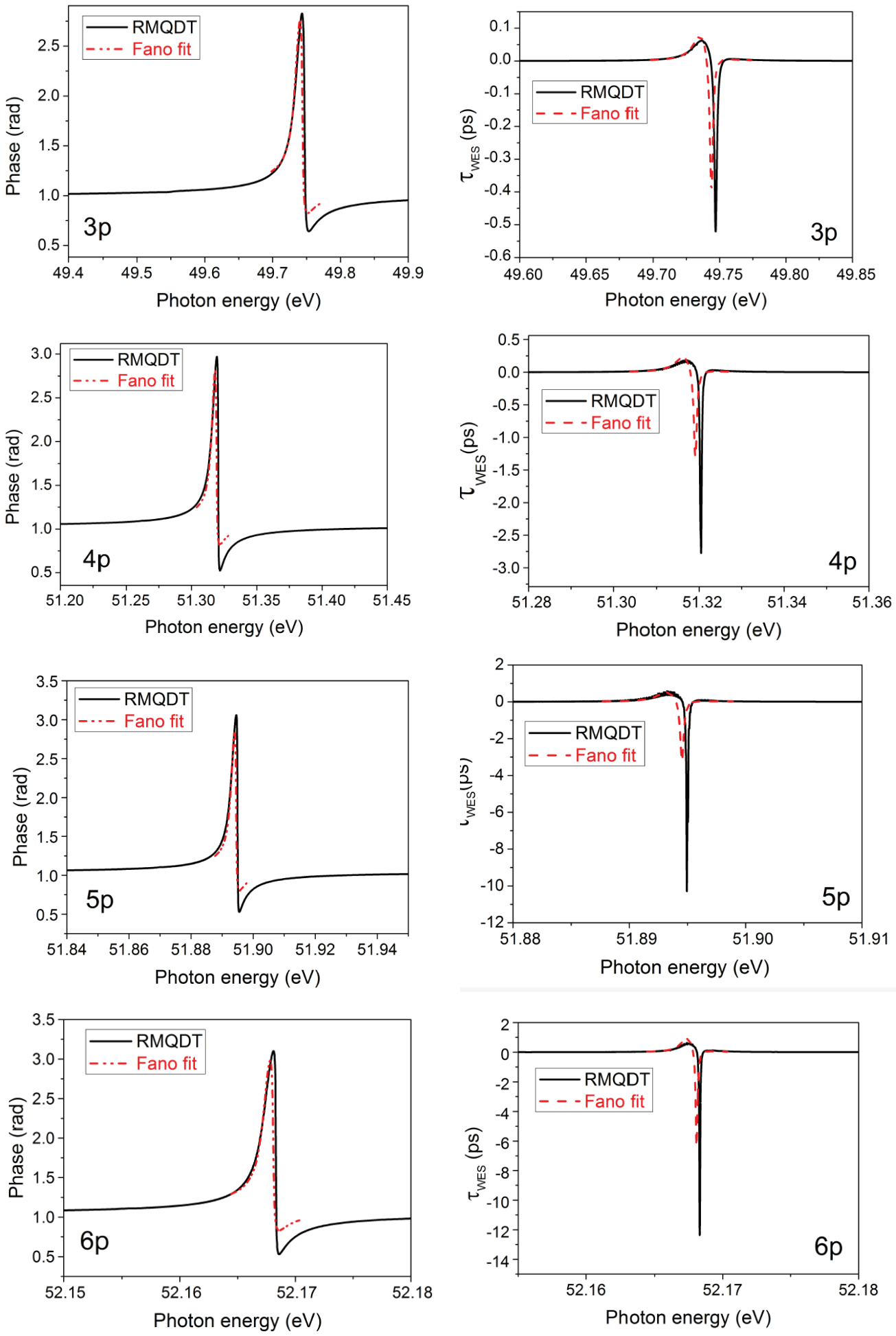

FIG. 3: (color online) Calculated 7-channel $2p$ (spin-unresolved) dipole photoionization phases (left curves) and WES time delays (right curves) in the vicinity of the $2s \rightarrow np$ autoionizing resonances showing the RMQDT results (solid line) and the fits using Fano parameters given in Table I (dashed lines).

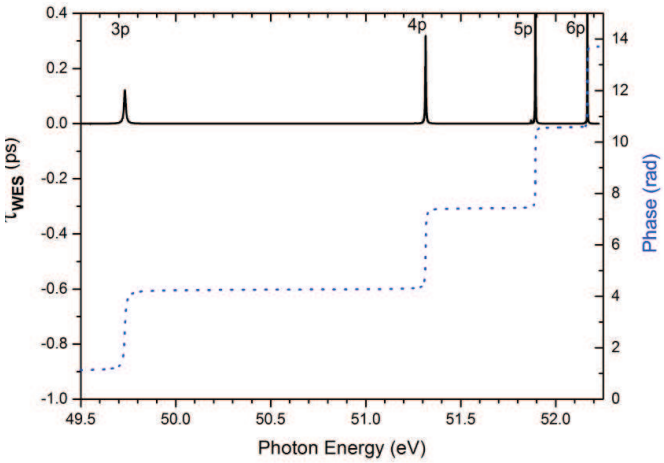

FIG. 4: (color online) WES time delay for the $2p_{3/2} \rightarrow \epsilon d_{5/2}$ photoionization channel (black curve, left scale) and phase (blue curve, right scale) in the $2s \rightarrow np$ autoionization resonance region obtained from the 3-channel RRPA-RMQDT calculation described in the text.

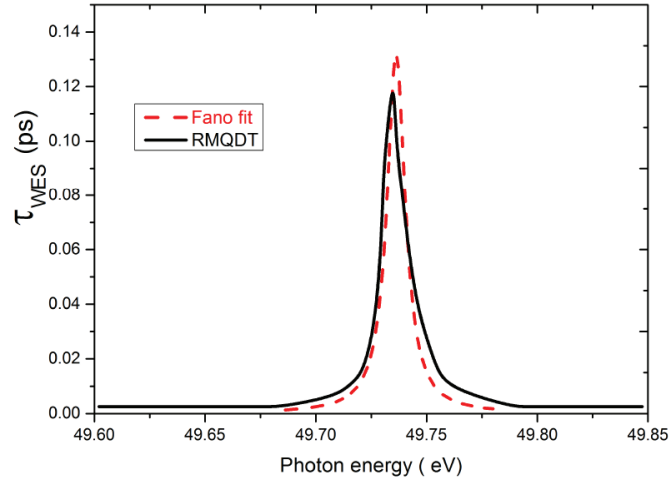

FIG. 5: (color online) WES time delay is the region of the $2s \rightarrow 3p$ resonance resulting from the 3-channel RRPA+RMQDT calculation, as described in the text (black line) along with the result of Eq.(10) using the fitted Fano parameters (red dashes).

results and the results of Eq. (9) using the Fano parameters are shown and it is clear that the agreement between them is excellent. This shows that the physics of the WES time delay is embodied in Eq. (6) in this case as well.

IV. SUMMARY AND CONCLUSIONS

The Wigner-Eisenbud-Smith photoionization time delay has been studied in the region of the $2s \rightarrow np$ autoionization resonances in Ne. The results show a time delay spectrum that has positive and negative components across each resonance and that the magnitudes of both the positive and negative components increase with increasing principle quantum number, n. It was also found that, in the neighborhoods of the resonances, the size of the time delays, both positive and negative, could be many orders of magnitude larger than the times delays measured in nonresonant regions, picoseconds vs. attoseconds.

A model based on the Fano parametrization of the resonance cross section has been developed using the resonance parameters q, Γ, and ρ^2, and the model results are in reasonable agreement with the phases and time delays calculated directly using the combined RMQDT-RRPA methodology. From this model it was determined that for resonances where $\rho^2 = 1$ (a single continuum channel), the time delay can only be positive; the Breit-Wigner case is in this category. But, for $\rho^2 < 1$, the parametric model shows that time delay will have an excursion to negative values and the location of the negative values, at energies higher or lower than the resonance position, will depend upon the sign of the parameter q. It is also noted that, for cases like Ne $2s \rightarrow np$ resonances, where there are no spectator Auger channels, the widths decrease as $1/(n^*)^3$ with increasing n, n^* being the effective quantum number, $n-\mu$, where μ is the quantum defect. Thus, in such a case, the lifetime of the resonances increase as $(n^*)^3$ and, from Eq. (6), the time delay increases as $(n^*)^3$ as well. As a consequence, the time delay, both the positive and native parts, become infinite in the limit of $n \rightarrow \infty$. As a corollary, since quantum defect theory tells us that the time delay is continuous across the ionization threshold, the WES threshold time delay in the $2s$ ionization channels must also diverge at threshold, a fact well-known from other considerations [23]. Finally, we note that a similar model has been proposed for two-photon XUV-IR ionization [50] in which the Fano q parameter is replaced by a complex quantity.

V. ACKNOWLEDGEMENTS

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