Extracting Phase and Amplitude Modifications of Laser-Coupled Fano Resonances

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(Received 10 December 2013; revised manuscript received 14 January 2014; published 10 March 2014)

Fano line shapes observed in absorption spectra encode information on the amplitude and phase of the optical dipole response. A change in the Fano line shape, e.g., by interaction with short-pulsed laser fields, allows us to extract dynamical modifications of the amplitude and phase of the coupled excited quantum states. We introduce and apply this physical mechanism to near-resonantly coupled doubly excited states in helium. This general approach provides a physical understanding of the laser-induced spectral shift of absorption-line maxima on a sub-laser-cycle time scale as they are ubiquitously observed in attosecond transient-absorption measurements.

DOI: 10.1103/PhysRevLett.112.103001 PACS numbers: 32.80.Qk, 07.60.Rd, 32.70.-n, 33.80.Eh

The physics understanding of light-matter interaction is at the heart of any time-resolved experiment that uses electromagnetic fields as a probe of quantum dynamics. Light fields change their spectral amplitude and phase as a result of an interaction, thereby unveiling information about the system under study. The amplitude modification is straightforward to measure by traditional spectroscopy methods by using, for example, dispersive elements and recording the spectral intensity. The spectroscopic fingerprint then usually consists of a superposition of characteristic line shapes, most commonly, the symmetric Lorentzian that is obtained for absorption or emission of light from isolated discrete quantum states.

By contrast, the phase of the light field is more challenging to access and spectral interferometry [1,2] or nonlinear-optical techniques are required for its measurement (e.g., Ref. [3]). Typically, the phase carries the crucial information about the dynamics of the system, e.g., defining the shape of wave packets in a superposition of quantum states. A closer look into the optical response of the system itself, most commonly, its dipole response, identifies the origin of spectral line shapes as the interference of the dipole emission with the transmitted light. Thus, phase information is also encoded in spectral line shapes [4–6].

In atomic physics, asymmetric absorption line shapes have been observed [7,8] and understood to be caused by discrete states coupled to a continuum. Fano's theory introduced the q-asymmetry parameter [9] to describe the line shape. In a recent experiment [6], it has been demonstrated that Fano's spectroscopic line-shape formula $\sigma_{\mathrm{Fano}}(\epsilon) \propto (q+\epsilon)^2/(1+\epsilon^2)$, with the reduced energy $\epsilon = 2(E-E_r)/\Gamma$ with respect to the resonance E_r of width Γ, is directly linked to the phase $\varphi = 2\arg(q-\imath)$ of the system's dipole-response function. Both q and φ encode information about couplings among quantum states, e.g., in Fano's original work, the coupling of a discrete state to a continuum it is embedded in. When quantum states are coupled by short-pulsed laser light of duration shorter than the system's decay time, the phase φ of the dipole response can be externally controlled. In the same way as the Fano q parameter is a sensitive test of atomic structure calculations under field-free conditions [10,11], the measurement of the laser-controlled phase φ may in the future provide a sensitive test of time-dependent quantum-dynamics theory [12]. In Ref. [6], it was shown how a laser pulse at a fixed time delay τ immediately after the excitation of the system is used to control the phase φ of its dipole response. This control mechanism was explained by nonresonant (quasi-classical, ponderomotive) coupling to a nearby continuum of states. By contrast, the resonant coupling of states by pulsed coherent light fields is of substantial interest, e.g., to fully understand the creation and probing of electronic wave packets [13–16].

In this Letter, we present a general scheme to observe and quantify the amplitude and phase changes of excited states coupled by a laser field. It is based on a generalization of the Fano-phase mechanism [6] (i.e., the correspondence between the dipole-response phase φ and the line-shape asymmetry q) to the measurement of dynamical phases induced by resonant laser coupling. We introduce the formalism for the case of doubly excited helium along with an experimental confirmation. By controlling the time delay and intensity of an ultrashort near-visible (VIS) laser field with respect to an exciting attosecond-pulsed extreme-ultraviolet (XUV) field, it is possible to observe, separate, and quantify the effects of different few-photon quantum paths that are involved in the coupling dynamics of auto-ionizing states in a laser field [17,18]. Relative phase and amplitude modifications of the excited states are revealed and represented as a two-dimensional complex-valued function of time delay and intensity of the VIS laser field. While in the past, light-matter interactions using long,

monochromatic laser fields have been intensely studied for such level-coupling situations [19–21], here we focus on the opposite general case of interaction with short pulses and large spectral bandwidth. While in the former case, spectral pictures were important to describe the dynamics (well-defined light-induced states created by laser fields that are δ-functionlike in the spectral domain, i.e., monochromatic), in our case, the pulses are described as δ-like in the time domain, hence calling for a temporal interpretation.

In the experimental setup an XUV light pulse—produced by high-order harmonic generation with a 7 fs 760 nm driver pulse—passes through a helium gas target of ~100 mbar backing pressure. The XUV pulse exhibits a continuous spectrum between 55 and 72 eV. During the interaction with the atoms, it coherently excites the $sp_{2,n+}$ [22] doubly excited state resonance series [see Fig. 1(a)]. The absorption spectra are observed by using a home-built flat-field spectrometer featuring a resolution better than 20 meV (standard deviation), which is crucial to resolve small changes in the observed asymmetric line shapes. In addition, a synchronized VIS laser pulse of controlled intensity and time delay follows the XUV pulse. The VIS laser (photon energy $\hbar\omega_L \approx 1.7$ eV) resonantly couples the lowest-lying dipole-allowed excited states $sp_{2,3+} = |1\rangle$ and $|2s2p\rangle = |3\rangle$ in a two-photon transition via the energetically intermediate $2p^2 = |2\rangle$ state, after their initial XUV excitation from the $1s^2 = |0\rangle$ ground state. The experimental setup is described in more detail in Ref. [6]. In the following, we specifically focus on the intensity- and time-delay-dependent changes of the absorption line shape of the $sp_{2,3+} = |1\rangle$ state excited from the ground state at a photon energy of 63.66 eV. The time-domain interpretation of resonance absorption is the time-dependent dipole response, here originating from the superposition of states $|1\rangle$ and $|0\rangle$, where small phase and amplitude modifications give rise to small changes in the spectroscopic line shape as depicted in Fig. 1(b).

The experimental results of the time-delay-dependent absorption spectrum near the $sp_{2,3+}$ state for a VIS intensity of 0.5×10^{12} W/cm^2 are shown in Fig. 2. The spectrum is modified with a period corresponding to approximately half the optical cycle of the VIS pulse. A small shift of the absorption maximum as a function of τ is observed. In our case, using the fully resolved spectral information, one finds that the shift of the absorption maximum is due to a modification of the line-shape asymmetry on a subcycle time-delay scale. In the following, we explain how such effects generally occur in coherently excited and impulsively laser-coupled few-level

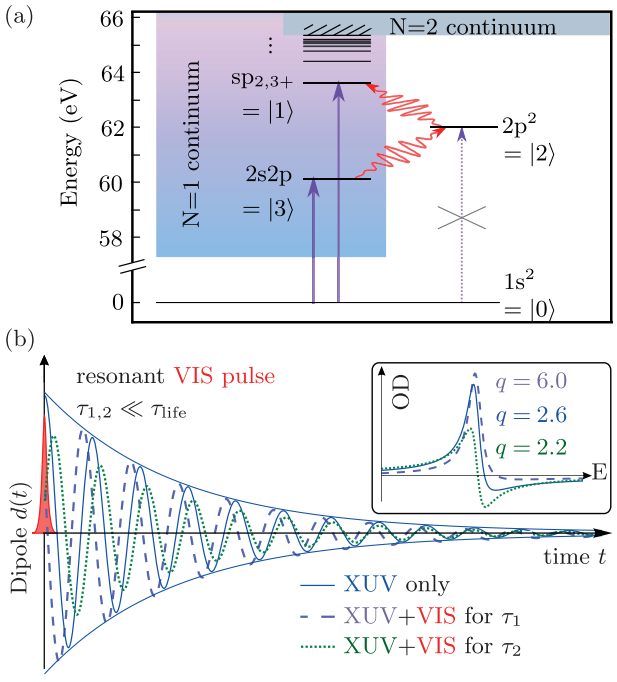

FIG. 1 (color online). (a) Doubly excited states of helium and the embedded three-level laser-coupled system. The XUV pulse coherently excites the dipole-allowed $2s2p$ and $sp_{2,3+}$ states (straight arrows), while the energetically intermediate $2p^2$ state cannot be accessed from the ground state $1s^2$. Both the $2s2p$ and $sp_{2,3+}$ states are resonantly coupled by two photons of a VIS pulse, via the $2p^2$ state. (b) The time-dependent atomic dipole moment $d(t)$, after δ-like XUV excitation at time $t = 0$ followed by natural decay. The observed spectral response [optical density (OD)], see inset, depends on the intensity and time delay of a VIS pulse (red shading), resonantly coupling the autoionizing states.

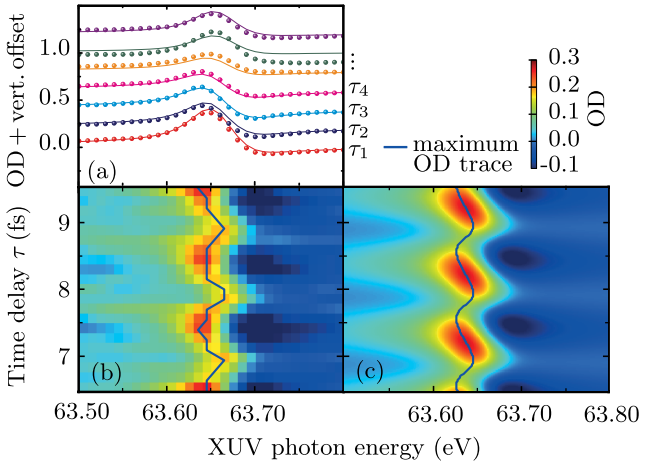

FIG. 2 (color online). (a) Measured (dots) Fano resonance absorption spectrum [optical density (OD)] near the He $1s^2 \rightarrow sp_{2,3+}$ transition. The resonant line shape changes with time delay τ from 8.6 to 9.8 fs of the VIS pulse following the XUV excitation. The VIS intensity was set to 0.5×10^{12} W/cm^2. Analytical formula [Eqs. (6) and (7)] (solid lines) for a fixed resonance-energy position. (b) Measured absorption spectra for varying time delay. (c) Analytically calculated absorption spectra [Eqs. (6) and (7)] after convolution with the spectrometer resolution (20 meV). The maxima of each of the spectra are also highlighted by the solid oscillatory trace. The oscillation of the maxima is due to a line-shape change while the resonance energy remains constant.

systems. We also use this understanding to trace the amplitude and phase changes of the excited state in a laser field of variable intensity.

To understand the few-level coupling case in a short laser pulse, we first describe a general two-level model system consisting of two excited states $|1\rangle$ and $|2\rangle$ with energy spacing $\Delta\omega_{12} \approx \omega_L$ (atomic units $\hbar = e = m = 1$ are used in the following) and opposite parity. Because of dipole selection rules, only $|1\rangle$ is initially populated at $t = 0$ (by the XUV pulse) while $|2\rangle$ remains empty. After excitation, the wave function can be written as $\Psi(t = 0) \propto |0\rangle - \imath a_1|1\rangle$ with complex expansion coefficient $a_1 = A_1 e^{\imath\varphi_1}$. In our case of $|1\rangle = sp_{2,3+}$ in helium, $\varphi_1 = -5.54$ rad corresponds to $q_1 = -2.58$ by using the mapping $q_i = -\cot(\varphi_i/2)$ [6]. We then describe the interaction with the VIS pulse, near-resonantly coupling the states $|1\rangle$ and $|2\rangle$, in second-order perturbation theory. As the VIS pulse is significantly shorter than the lifetime of both states, we approximate it by an infinitesimally short interaction, a δ-like pulse. After the interaction, the coefficient a_1 of $|1\rangle$ is changed to

$$a_1' = a_1 + a_{\text{self}}. \tag{1}$$

The additional contribution

$$a_{\text{self}} = A_{\text{self}} e^{\imath(\varphi_1+\pi)} \propto -a_1 E_{\text{VIS}}^2 |\mu_{12}|^2 \tag{2}$$

of the two-photon laser coupling to state $|2\rangle$ and back to $|1\rangle$ depends on the square of the coupling electric field strength E_{VIS} and the squared modulus of the dipole-matrix element μ_{12} between states $|1\rangle$ and $|2\rangle$. It is π out of phase with the (much larger) original excitation a_1, corresponding to a (small) loss of population from state $|1\rangle$ to $|2\rangle$.

After describing the two-level coupling, we now include the third coherently excited state $|3\rangle$ located energetically below $|2\rangle$ with an energy spacing of $\Delta\omega_{23} \approx \Delta\omega_{12} \approx \omega_{\text{L}}$. The VIS laser pulse couples $|3\rangle$ to $|1\rangle$ by a two-photon transition via $|2\rangle$. As a result, the expansion coefficient of $|1\rangle$ after VIS interaction, within second-order perturbation theory picks up a third term by interstate coupling complementing Eq. (1) to

$$a'' = a_1 + a_{\text{self}} + a_{\text{inter}}(\tau). \tag{3}$$

The third contribution

$$\begin{aligned} a_{\text{inter}}(\tau) &= A_{\text{inter}} e^{\imath(\varphi_3+\pi+\Delta\omega_{13}\tau)} \\ &\propto a_3 E_{\text{VIS}}^2 \mu_{32}\mu_{21} e^{\imath\Delta\omega_{13}\tau} \end{aligned} \tag{4}$$

depends on the time delay τ between the XUV excitation and the VIS coupling pulse. This is due to the fact that $|3\rangle$ was excited coherently by the same XUV pulse as $|1\rangle$ with initial coefficient a_3 but evolving in time with a relative phase $\Delta\omega_{13}\tau$ compared to state $|1\rangle$. It should be noted that all interactions (both XUV and VIS) are treated to take place near $t \approx 0$, while their respective delay τ is cast into the complex coefficient in $a_{\text{inter}}(\tau)$. This is a valid assumption when both τ and the XUV and VIS pulse durations are short compared to the lifetime τ_{life} of the state. The time-dependent XUV dipole response associated to state $|1\rangle$ after VIS interaction is finally given by

$$d_{10}(t) = \langle\Psi(t)|r|\Psi(t)\rangle \propto A_{\text{mod}} e^{\imath\varphi_{\text{mod}}} e^{-(\Gamma/2+\imath\Delta\omega_{01})t} + \text{c.c.}, \tag{5}$$

subject to a natural decay of rate $\Gamma = 1/\tau_{\text{life}}$ for $t > 0$. It involves the complex prefactor $A_{\text{mod}} e^{\imath\varphi_{\text{mod}}} \propto a''$, which is proportional to the expansion coefficient a'' using Eqs. (2)–(4). The Fourier transform of $d_{10}(t)$ given by $\tilde{d}_{10}(\omega)$ is proportional to the background-corrected Fano absorption cross section [6]

$$\sigma_{\text{Fano}}(\omega) - 1 \propto \Im[\tilde{d}_{10}(\omega)] \propto \Im\left[A_{\text{mod}} e^{\imath\varphi_{\text{mod}}} \frac{\imath - \epsilon(\omega)}{1 + \epsilon^2(\omega)}\right], \tag{6}$$

with $\epsilon = 2(\omega - \Delta\omega_{01})/\Gamma$. Because of the linearity of the Fourier transformation, the three contributions to the quantum-state coefficient a'' are now also found in the complex factor

$$\begin{aligned} A_{\text{mod}}(\tau) e^{\imath\varphi_{\text{mod}}(\tau)} = {} & A_1 e^{\imath\varphi_1} + A_{\text{self}} e^{\imath(\varphi_1+\pi)} \\ & + A_{\text{inter}} e^{\imath(\varphi_3+\pi+\Delta\omega_{13}\tau)}, \end{aligned} \tag{7}$$

which governs the line shape of the measured absorption spectrum. As a result, the absorption spectrum $\sigma(\omega)$ also varies as a function of the delay τ. The individual contributions and total sum to the complex dipole response can be represented graphically by phasor addition in the complex plane (see Fig. 3). While the first two components are π out of phase relative to each other and independent of τ, the third component is fixed in amplitude A_{inter} but varies in phase $\varphi_{\text{inter}}(\tau)$. This causes the phasor sum to circle around a fixed center point in the complex plane as the time delay is varied.

We now apply the model to measured absorption spectra across the $sp_{2,3+}$ state in helium for different time delays and intensities of the VIS coupling pulse. We fitted a sequence of absorption line shapes to the state $sp_{2,3+} = |1\rangle$ using the Fano cross section in phase representation [Eq. (6)]. For this fit, the amplitude A_{mod} and phase φ_{mod} were left as free parameters (see Fig. 3), while μ_{12}, μ_{23}, μ_{13}, Γ_i, $\omega_i (i \in [1, 2, 3])$ were fixed to tabulated values [23]. We, thus, assign a pair of amplitude and phase to each pair of time delay and intensity. In Fig. 4, $A_{\text{mod}} e^{\imath\varphi_{\text{mod}}}$ is plotted in the complex plane for half a VIS laser cycle.

Interestingly, we observe an elliptical shape of the curve rather than the circle expected from our model. As a function of the VIS intensity, the center point of the ellipse moves closer to the origin [Fig. 4(b)] as expected from

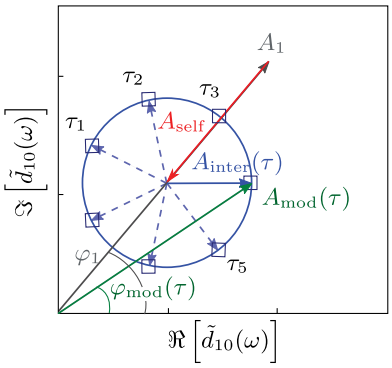

FIG. 3 (color online). Graphical representation of the complex amplitude of an excited state, resonantly coupled by δ-like pulsed laser interaction to two other levels: a (dark) initially non-populated intermediate state and a coherently excited (bright) state. In second-order perturbation theory, the amplitude contribution for transition in and out of the dark state A_{self} is phase shifted by π with respect to the original excitation amplitude A_1, resulting in an amplitude reduction due to population transfer. The contribution of the coherently excited bright state A_{inter} describes a circle in the complex plane as a function of time delay due to the energy difference between the states and their corresponding field-free time-dependent phase evolution.

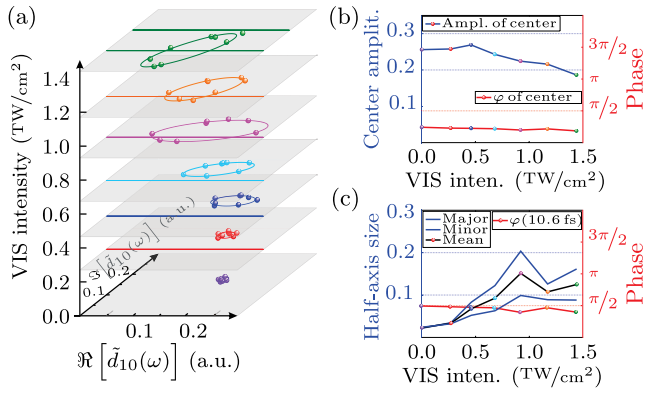

FIG. 4 (color online). (a) Spectral dipole amplitudes and phases as a function of the VIS intensity and time delay. Each layer represents a complex plane in which the time-delay evolution of the dipole moment is shown (clockwise for increasing time delay). The dots show the experimental data, the solid lines are ellipsoidal fits. One half cycle of the VIS pulse corresponds to seven data points. The time delay τ ranges from 9.8 to 11.0 fs. By extracting the circle radius and center as a function of VIS intensity, the different resonant coupling pathways can be separated [as shown in (b) and (c)]. With increasing intensity and correspondingly larger radius approaching the size of the initial amplitude, the circles become more and more elliptical. (b) The absolute value of the complex-valued center point of each ellipse (upper curve and left y axis) and the corresponding phase (lower curve and right y axis) for each measured VIS intensity. The center point of the ellipse changes its position due to population transfer with increasing VIS intensity. The phase of the center point remains approximately constant for all VIS intensities. (c) Length of the major and minor axes of each ellipse, the corresponding mean value, and, relative to the center point, the phase of the point at fixed time delay 10.6 fs for each measured VIS intensity.

population transfer expressed by the contribution a_{self}. The difference of the absolute value of the center point at a given intensity with respect to its zero-intensity value, hence represents the intensity-dependent modulus A_{self}. We can interpret the elliptical shape as the superposition of two counterrotating contributions to the complex coefficient in the observed state $|1\rangle$. A second laser-coupled state energetically above $|1\rangle$, coherently excited by the XUV pulse, would add this counterrotating contribution by the same reasoning as given above for the lower-lying $|3\rangle$ state. In fact, the $N = 2$ continuum in He [see Fig. 1(a)] can contribute by a two-photon transition into state $|1\rangle$, providing such a counterrotating term.

From second-order perturbative two-photon interstate population transfer [as discussed for the circle above, see Fig. 3 and Eq. (4)], an increase of the ellipse half-axis size is expected with increasing VIS intensity. This tendency is confirmed for the measured ellipse (major and minor) axes [Fig. 4(c)]. In future work, the axis sizes of the ellipse, its orientation, and the time-delay dependence can be used to extract more quantitative information on coherent interstate coupling amplitudes A_{inter} and their time-dependent relative phases.

From an entirely different perspective, the experimental results and their interpretation are also of general importance for the budding field of attosecond transient-absorption spectroscopy [24–27]. Figure 2(b) shows the absorption spectrum over a time-delay range of 4 fs. Connecting the absorption maxima of each spectrum reveals an oscillation with the VIS-pulse half-cycle period. The comparison with the fitted line shapes in Fig. 2(a) clearly shows that the time delay induces a phase change in the oscillating dipole as understood by the herein described analytical few-level model of states coupled with δ-like pulses. As a result, the spectral position of the absorption maximum varies as the line shape changes from an asymmetric, Fano-like profile to a more symmetric, close-to-Lorentzian profile, while the resonance energy and width remain constant due to the field-free evolution of the dipole response after the coupling pulse. In the absence of line-shape information at high spectral resolution, such a shift could be confused with a time-dependent energy-level shift, creating an important caveat for transient-absorption spectroscopy.

In conclusion, we developed a generalized model of the Fano-phase approach to the measurement of time-dependent phases and amplitudes induced by short-pulsed resonant laser coupling. This model was confirmed for the example of doubly excited laser-coupled states in the benchmark system of helium. It allows for the separation of different few-photon quantum pathways that are involved in the coupling dynamics of the autoionizing states using laser pulses at controlled time delay and

intensity. Higher-precision measurements and the analyses of the exact variation of the different phase contributions with laser control will help to better understand the laser coupling of autoionizing states to other bound or continuum states. In the future, the same approach also allows for a thorough investigation of the transition from the perturbative to the strong-field regime of resonant and nonresonant laser coupling of electronic quantum states in general.

We thank Jörg Evers and Sebastian Meuren for fruitful discussions. Financial support from the Max-Planck Research Group program and the Deutsche Forschungsgemeinschaft (Grant No. PF 790/1-1) is gratefully acknowledged.

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