

Reconstruction of attosecond beating by interference of two-photon transitions with higher-order processes

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The reconstruction of attosecond beating by interference of two-photon transitions (RABBITT) method is a leading technique for time-resolved investigation of photoionization processes. Interpretation of RABBITT spectra is limited to a lowest-order description, while new variants of the technique necessitate higher intensity infrared fields where this description breaks down. Here, we present an extension of RABBITT theory which can account for higher-order processes (RABBITT-HOP), and demonstrate RABBITT-HOP's ability to predict and describe higher-order processes through *ab initio* simulation of a RABBITT experiment targeting the high-lying $1snp$ Rydberg series of helium. We demonstrate that the signature line shape imparted by the resonances in the 2ω phase of the lowest sideband can also be observed in the 2ω phase of the next mainband, as well as in the 4ω phases of the next sideband and mainband—each with additional phase shifts predicted under RABBITT-HOP. Repeating this analysis for a range of infrared intensities, we determine the lower limits of intensities required for these higher-order processes to become observable and upper limits beyond which nonperturbative effects begin to dominate. This new theoretical framework forms a much-needed basis for accurate interpretation of RABBITT spectra in experiments involving more intense fields, and paves the way for the development of new RABBITT-like techniques which take advantage of this new understanding of how atomic resonance effects may be propagated beyond their nearest sideband.

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I. INTRODUCTION

One of the fundamental objectives in attosecond science is the resolution of ultrafast electron dynamics. Thus, several methods have been developed for the time-resolved measurement of these dynamics [1–6]. Among these methods, the reconstruction of attosecond beating by interference of two-photon transitions (RABBITT) technique has become a particularly popular approach [7–12]. RABBITT was conceptualized as a two-color interferometric technique to identify phase differences between consecutive harmonic components (attochirp) generated through high harmonic generation via an intense infrared (IR) laser field [13], allowing the characterization of an attosecond pulse train (APT) [7]. It has since been realized that the phase information extracted via the RABBITT technique can be decomposed to also recover *atomic* phase information and, thereby, to resolve electron dynamics in the time domain [8–12].

Figure 1(a) shows an example energy-level diagram covering the main transitions within the RABBITT scheme. An APT comprising odd harmonics of the driving IR pulse

(frequency, ω_{IR}) ionizes the system and excites a valence electron from the ground state into the continuum. The IR dressing field induces a second transition, which causes the excited electron either to absorb or to emit an IR photon, which changes its energy by $\pm\omega_{\text{IR}}$, and creates a “sideband” in the photoelectron spectrum. Thus, each sideband may be reached by two different pathways: absorption of a $(2n+1)\omega_{\text{IR}}$ XUV photon and an ω_{IR} IR photon, or absorption of a $(2n+3)\omega_{\text{IR}}$ XUV photon and emission of an ω_{IR} IR photon. These two allowed pathways will interfere with each other, and Fig. 1(c) shows that this interference can be observed by varying the delay of the APT relative to the IR field. The sideband signal oscillates sinusoidally with a frequency of $2\omega_{\text{IR}}$ —matching the spectral separation of the two contributing XUV harmonics—and a phase related to their phase difference.

One key aspect of RABBITT is the requirement for a weak IR field. The sidebands show interference between pathways involving the neighboring harmonics, but to interpret these interference patterns higher-order ionization paths are considered negligible. Previous research has investigated how RABBITT may be pushed past the lowest order. Notably Swoboda *et al.* observed signals at higher IR intensities which could not be explained using only 1-XUV-1-IR-photon paths [14]. Bharti *et al.* demonstrated a novel “multisideband” RABBITT scheme in which the XUV-APT was generated by a frequency-doubled IR to produce harmonics separated by $4\omega_{\text{IR}}$. The three sidebands between these harmonics correspond to interference between two- and four-photon paths and between three- and three-photon paths [15,16].

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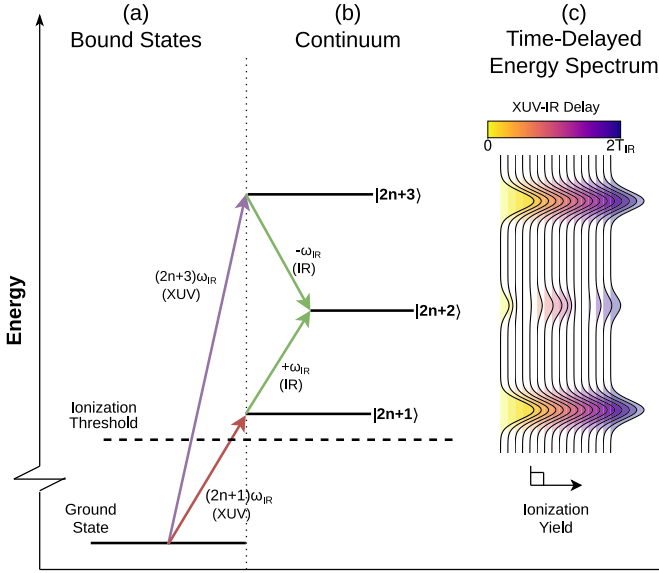


FIG. 1. Diagrammatic explanation of the RABBITT Technique. (a) XUV photons (purple and red arrows) from the attosecond pulse train (APT) ionize the system. (b) Absorption (upwards green arrow) or emission (downwards green arrow) of an IR photon changes the photoelectron's energy by ω_{IR} to form a “sideband” in the photoelectron energy spectrum. (c) Contributions from the two possible pathways interfere, and this is observed as an oscillation of the sideband signal by varying the relative delay of the XUV-APT with respect to the IR field.

More recently, Lucchini presented a Floquet model to account for interfering paths discarded under the lowest order approach [17]. At high intensities ($7 \times 10^{11} \text{ W/cm}^2$) and higher photoelectron energies the traditional interpretation breaks down and sideband oscillations at more than one frequency are observed.

Lucchini's approach, based on the strong-field approximation, is valid only for high photoelectron energies. However, we recently observed higher-order effects within a RABBITT sideband at photoelectron energies just above the ionization threshold [18]. Analysis of the interferences in the photoelectron angular distribution reveal the contribution of transitions involving a minimum of three IR photons. Thus, an extension of the RABBITT technique to account for higher-order effects is needed—not just to explain specifically constructed experimental setups such as multisideband RABBITT [19], but also for the interpretation of traditional RABBITT setups, even at moderate ($\approx 10^{11} \text{ W/cm}^2$) IR intensities.

In this paper, we derive expressions for RABBITT signals which account for all ionization paths involving up to some specific higher order: RABBITT extended to higher-order processes (RABBITT-HOP). These expressions are then verified through a RABBITT investigation of neutral helium using the *R* matrix with time dependence (RMT) method. Our focus is on understanding the contribution of *atomic resonances* (which are ignored in Ref. [17]) to the measured, high-order signals. The presence of resonance contributions from the $1s4p$ and $1s5p$ Rydberg states in the RABBITT spectra provides a clear marker for the progression of higher-order processes. We also analyze how the multimodal oscillation of

sidebands and mainbands varies with IR intensity, providing an estimate of the intensities required to evoke multiphoton transitions with sufficient probability to produce measurable effects.

II. RABBITT FORMALISM FOR HIGHER-ORDER PROCESSES

A. Standard RABBITT

Figure 1 shows the standard scheme for a RABBITT study, with the RABBITT sideband formed from the coherent superposition of two two-photon ionization paths. The evaluation of this coherence relies on the transition matrix elements for each “step” along a path, accounting for both a probability amplitude and an imparted phase shift [1,13,20,21]. We present here only a simplified model in order to track the contributions of atomic resonances to the measured RABBITT phase. A more detailed description will also require consideration of the partial-wave decomposition across each ionization path [10,22], as well as the structure of the electric field in time [20,23–25].

For the general description of the RABBITT process, we express the time-dependent XUV field as a coherent superposition of odd harmonics of the driving IR field with frequency ω_{IR} :

$$D_{\text{XUV}}(t) = \sum_{n, n \text{ odd}} 2E_n \cos(n\omega_{\text{IR}}t - \phi_n), \quad (1)$$

where ϕ_n is a phase delay parameter associated with the n th harmonic component, the derivative of which with respect to frequency is commonly referred to as the “attochirp.” Under experimental conditions, different harmonics have different delays, although in our calculations, the phase delays are chosen to be zero for simplicity. The evaluation of XUV ionization in each pathway then requires just the specific transition dipole matrix element between the ground state and a well-defined continuum state [20,23]. Note that for the IR field, we associate E with the field strength explicitly associated with transitions within the continuum, which requires a factor 2 in the overall driving fields. We simplify our initial analysis of the RABBITT scheme further by considering general matrix elements, in which we ignore the decomposition into partial waves, and assume that all intensities can be treated perturbatively. The former assumption does not substantially impact our current considerations on higher-order processes as we do not deal with photoelectron angular distributions, which need a more stringent treatment of the amplitude and phases of multiple partial waves [18].

Following these assumptions, the probability amplitude for photoionization from the ground state into the continuum, via absorption of a single n th-harmonic XUV photon, is given by integration of the time-dependent Schrödinger equation (TDSE) as

$$a_n^{(1)} = \frac{\mu_{g,n} E_n e^{i\phi_n}}{i}, \quad (2)$$

where $\mu_{g,n}$ is a time-integrated generalized transition dipole matrix element associated with transitions from $|g\rangle \rightarrow |n\rangle$, and E_n is the strength of the n th harmonic component of the XUV field [26]. The superscript (1) indicates the total number

of photons involved in the transition between initial and final state. For simplicity, we assume a chirpless pulse train, i.e. that all phases in the XUV field are zero, yielding

$$a_n^{(1)} = \frac{\mu_{g,n} E_n}{i}. \quad (3)$$

In the RABBITT scheme, photoionization by an XUV photon is followed by emission or absorption of an IR photon transferring the photoelectron to sidebands of order $(n+1)$ or $(n-1)$. We denote probability amplitudes for IR transitions by $b_{n\pm}^{(k)}$ to distinguish them from XUV transitions, with the superscript (k) indicating the number of IR photons absorbed or emitted to reach the final state. The subscript $n\pm$ denotes absorption or emission starting from a continuum state with an energy $n\omega_{\text{IR}}$ above the ground state. Integration of the TDSE then gives the probability amplitude for a specific, one-photon transition as

$$b_{(n)\pm}^{(1)} = \frac{\mu_{n,n\pm 1} E_0 e^{\pm i\phi}}{i}, \quad (4)$$

where E_0 and ϕ indicate the amplitude and phase, respectively, of the IR field. The phase of the IR is determined by the time delay τ relative to the XUV-APT. Thus we have $\phi = \omega_{\text{IR}}\tau$. Note that $\mu_{n,n\pm 1}$ should be treated as a generalized amplitude, as the initial state in the transition will start to receive population during the IR pulse. Using Eqs. (3) and (4), we may write the total probability amplitude of two-photon transitions from the ground state, $|g\rangle$, to the sideband $|n-1\rangle$ in terms of one-photon transitions as

$$\begin{aligned} a_{n-1}^{(2)} &= a_n^{(1)} b_{(n)-}^{(1)} + a_{n-2}^{(1)} b_{(n-2)+}^{(1)} \\ &= \frac{\mu_{g,n} E_n}{i} \frac{\mu_{n,n-1} E_0 e^{-i\omega_{\text{IR}}\tau}}{i} \\ &\quad + \frac{\mu_{g,n-2} E_{n-2}}{i} \frac{\mu_{n-2,n-1} E_0 e^{i\omega_{\text{IR}}\tau}}{i} \\ &= -\mu_{g,n} \mu_{n,n-1} E_n E_0 e^{-i\omega_{\text{IR}}\tau} \\ &\quad - \mu_{g,n-2} \mu_{n-2,n-1} E_{n-2} E_0 e^{i\omega_{\text{IR}}\tau}. \end{aligned} \quad (5)$$

The first term represents XUV-induced excitation from $|g\rangle$ to $|n\rangle$ and emission of an IR photon, while the second represents excitation to $|n-2\rangle$ and absorption of an IR photon.

The total population within $|n-1\rangle$ can then be obtained by multiplying expression (5) by its complex conjugate. This gives an average yield in the sideband given by the sum of the squared magnitudes of each of the pathways plus an interference term given by

$$|a_{n-1}^{(2)}|_{2\omega}^2 = \mu_{g,n} \mu_{n,n-1} \mu_{g,n-2}^* \mu_{n-2,n-1}^* E_n E_{n-2} E_0^2 e^{2i\omega_{\text{IR}}\tau} + \text{c.c.} \quad (6)$$

$$= C_1 \cos(2\omega_{\text{IR}}\tau - \phi_C), \quad (7)$$

where C_1 and ϕ_C are determined by the magnitudes and phases of the transition dipole matrices in the prefactor.

The interference term thus manifests as an oscillation in the sideband yield. For a chirpless pulse, the attophase is zero, and the phase of the oscillation is determined entirely by the atomic phases present in the matrix elements. The frequency of the oscillation, $2\omega_{\text{IR}}$, is determined by the energy gap between the mainbands involved [n and $n-2$ in Eq. (6)].

This idea becomes more important as we consider higher-order (and higher-frequency) terms originating from different mainbands with larger energy separations.

B. RABBITT signal within a mainband

Typically, RABBITT studies focus on the oscillation within a sideband, arising from two-photon transitions as encapsulated in Eq. (7). However, population transfer to a sideband should deplete the population in a mainband. Since the population transfer depends on interference between adjacent harmonics, mainband depletion must also contain signatures of adjacent harmonics. Hence mainband depletion already contains effects of higher-order (multiphoton) processes.

The lowest order of perturbation theory that can generate interference terms with frequency $2\omega_{\text{IR}}$ is second order. The important contributions to the population of mainband n , up to second order, are (a) direct excitation by an XUV harmonic of order n , (b) excitation by an XUV harmonic of order $n-2$ followed by absorption of two IR photons, and (c) excitation by an XUV harmonic of order $n+2$ followed by emission of two IR photons. An additional contribution is present: direct excitation by an XUV harmonic of order n followed by emission and absorption of an IR photon. This contribution can be neglected for interference terms with frequency $2\omega_{\text{IR}}$ in lowest order, as the main contribution arising from harmonic n is the lowest-order direct excitation term. The higher-order contribution will not introduce any new frequency components in RABBITT oscillations, and, in the perturbative regime, will be a minor correction to the interference created by the direct excitation.

We can thus restrict considerations to the population amplitude arising from a single component contributed by each harmonic, giving the probability amplitude for one- or three-photon transitions into mainband n as

$$\begin{aligned} a_n^{(3)} &= a_n^{(1)} + a_{n+2}^{(1)} \frac{b_{(n+2)-}^{(1)} b_{(n+1)-}^{(1)}}{2} \\ &\quad + a_{n-2}^{(1)} \frac{b_{(n-2)+}^{(1)} b_{(n-1)+}^{(1)}}{2}. \end{aligned} \quad (8)$$

As we observed for sideband oscillations in Eq. (6), oscillations at a frequency of $2\omega_{\text{IR}}$ will arise from the interference between pathways originating in mainbands separated by $2\omega_{\text{IR}}$. When considering the magnitude of this probability amplitude, these correspond to one interference term connecting harmonics n and $n+2$ and one connecting harmonics n and $n-2$. The $2\omega_{\text{IR}}$ interference term of the mainband is then given by

$$\begin{aligned} |a_n^{(3)}|_{2\omega}^2 &= -\frac{\mu_{g,n} \mu_{g,n-2}^* \mu_{n-2,n-1}^* \mu_{n-1,n}^* E_n E_{n-2} E_0^2 e^{-2i\omega_{\text{IR}}\tau}}{2} \\ &\quad - \frac{\mu_{g,n} \mu_{g,n+2}^* \mu_{n+2,n+1}^* \mu_{n+1,n}^* E_n E_{n+2} E_0^2 e^{2i\omega_{\text{IR}}\tau}}{2} \\ &\quad + \text{c.c.} \end{aligned} \quad (9)$$

Comparing Eqs. (6) and (9) we can see that oscillations in sidebands are mirrored by corresponding depletion of the associated mainband signals.

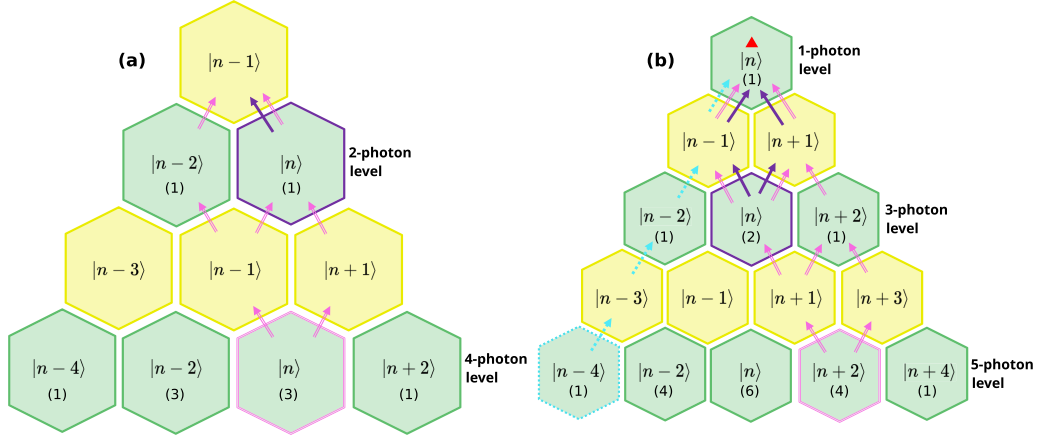


FIG. 2. Schematic of the relationship between binomial coefficients described in Pascal's triangle and the number of possible ionization paths from odd harmonics to (a) a chosen sideband, $|n-1\rangle$, at the two- and four-photon levels, and (b) a chosen mainband, $|n\rangle$, at the one-, three-, and five-photon levels. Green hexagons (every second layer starting with the bottom layer) correspond to mainbands, while yellow hexagons (alternate layers) correspond to sidebands. Arrows represent IR absorptions and emissions from several example cases. In (b), the red arrowhead in the $|n\rangle$ mainband at the one-photon level serves as a reminder that a direct XUV-only path exists, and that there is an XUV transition from the ground state to every other green hexagon omitted in this diagram.

This analysis can now be extended to higher-order frequency components in both sidebands and mainbands.

C. Higher-order signals within a sideband

At higher IR laser intensities, more than one IR photon may be absorbed to reach a sideband. In that case, the total final population amplitude of sideband $|n-1\rangle$ can be expanded as

$$a_{n-1}^{(\text{all})} = \sum_{j=0}^{\infty} a_{n-1}^{(2j+2)},$$

which is the coherent sum of all possible paths to reach sideband $|n-1\rangle$. The superscript on a indicates the total number of photons involved, with a minimum of two photons (one XUV and one IR) needed. Each term in this expression can be decomposed into a sum of $(2j+1)$ products of one-IR-photon paths, following a single XUV transition to mainbands within $(2j+1)\omega_{\text{IR}}$ of the sideband. A single term of order $(2j+2)$ can be written as

$$a_{n-1}^{(2j+2)} = \sum_{k=(n-2j-2),2}^{n+2j} [a_k^{(1)} b_{(k \rightarrow n-1)}^{(2j+1)}], \quad (10)$$

where $b_{(k \rightarrow n-1)}^{(2j+1)}$ is the coherent sum of all $(2j+1)$ -IR-photon paths from $|k\rangle$ to $|n-1\rangle$.

The transition from $|k\rangle$ to sideband $|n-1\rangle$ involving $2j+1$ IR photons, $b_{(k \rightarrow n-1)}^{(2j+1)}$, can be written as a product of $2j+1$ individual IR transitions, comprising n_{b+} IR absorptions (b_+) and n_{b-} IR emissions (b_-):

$$b_{(k \rightarrow n-1)}^{(2j+1)} \simeq \binom{n_{b+} + n_{b-}}{n_{b+}} \frac{b_+^{n_{b+}} b_-^{n_{b-}}}{(2j+1)!}, \quad (11)$$

where n_{b+} and n_{b-} indicate the number of IR photons absorbed and emitted, respectively, and account is taken of the number of possible orderings of absorption and emission processes. The probability amplitude for a process of order

$(2j+2)$ can then be expressed as

$$a_{n-1}^{(2j+2)} = \sum_{k=(n-2j-2),2}^{n+2j} \left[\binom{2j+1}{j+d} a_k^{(1)} \frac{b_+^{j+d} b_-^{j-d+1}}{(2j+1)!} \right], \quad (12)$$

where $d = (n-k)/2$.

Following Eq. (12), the combination of paths to the $|n-1\rangle$ sideband, truncated at p -photon paths, is then given by

$$a_{n-1}^{(p)} = \sum_{j=0}^{\lfloor (p-2)/2 \rfloor} a_{n-1}^{(2j+2)} = \sum_{j=0}^{\lfloor (p-2)/2 \rfloor} \left\{ \sum_{k=(n-2j-2),2}^{(n+2j)} \left[\binom{2j+1}{j+d} a_k^{(1)} \frac{b_+^{j+d} b_-^{j-d+1}}{(2j+1)!} \right] \right\}. \quad (13)$$

The $4\omega_{\text{IR}}$ component in a RABBITT sideband signal originates from the interference of pathways involving the absorption or emission of a single IR photon and emission or absorption of three IR photons. At perturbative intensities, these additional three-IR-photon pathways will not significantly alter the main $2\omega_{\text{IR}}$ RABBITT interference signal, but they are source terms for the $4\omega_{\text{IR}}$ interference. Figure 2 demonstrates the possible pathways that could be followed to reach a specific sideband. To reach sideband $|n-1\rangle$ using three IR photons, the initial XUV excitation can involve harmonics $n+2$, n , $n-2$, and $n-4$. However, the dominant contributions to the $4\omega_{\text{IR}}$ signal will only involve single-photon transfer from harmonics n and $n-2$, and three-photon transfer from harmonics $n-4$ and $n+2$. Thus, the $4\omega_{\text{IR}}$ frequency component within a sideband can be analyzed using just a single term for each contributing harmonic:

$$a_{n-1,\text{min}}^{(4)} = a_n^{(1)} b_{(n)-}^{(1)} + a_{n-2}^{(1)} b_{(n-2)+}^{(1)} + a_{n+2}^{(1)} \frac{b_{(n+2)-}^{(1)} b_{(n+1)-}^{(1)} b_{(n)-}^{(1)}}{6} + a_{n-4}^{(1)} \frac{b_{(n-4)+}^{(1)} b_{(n-3)+}^{(1)} b_{(n-2)+}^{(1)}}{6}. \quad (14)$$

The photoelectron yields associated with a $4\omega_{\text{IR}}$ signal will then be formed by the (incoherent) sum of interference between terms in which XUV harmonics are separated by

$$\begin{aligned} |a_{n-1}^{(4)}|_{4\omega}^2 = & - \frac{\mu_{g,n}\mu_{n,n-1}\mu_{g,n-4}^*\mu_{n-4,n-3}^*\mu_{n-3,n-2}^*\mu_{n-2,n-1}^*E_nE_{n-4}E_0^4e^{-4i\omega_{\text{IR}}\tau}}{6} \\ & - \frac{\mu_{g,n-2}\mu_{n-2,n-1}\mu_{g,n+2}^*\mu_{n+2,n+1}^*\mu_{n+1,n}^*\mu_{n,n-1}^*E_{n-2}E_{n+2}E_0^4e^{4i\omega_{\text{IR}}\tau}}{6} + \text{c.c.} \end{aligned} \quad (15)$$

The $4\omega_{\text{IR}}$ interference can thus be written as the sum of two cosine terms with amplitude and phase given by the various dipole matrix elements.

D. Higher-order signals within a mainband

The description of higher-order effects in mainbands proceeds in a similar manner. In this case, an even number of IR photons must be absorbed in addition to a single XUV photon. A full account of allowed pathways to the mainband $|n\rangle$ is thus given by

$$a_n^{(\text{all})} = \sum_{j=0}^{\infty} a_n^{(2j+1)}. \quad (16)$$

Following the same process as above, we obtain

$$a_n^{(2j+1)} = \sum_{k=(n-2j),2}^{n+2j} \left[\binom{2j}{j+d} a_k^{(1)} \frac{b_+^{j+d} b_-^{j-d}}{(2j)!} \right], \quad (17)$$

$4\omega_{\text{IR}}$: n and $n-4$, and $n-2$ and $n+2$. This gives for the $4\omega_{\text{IR}}$ signal strength

and the full combination of paths to mainband $|n\rangle$, truncated beyond p photons, as

$$a_n^{(\text{1top})} = \sum_{j=0}^{\lfloor p/2 \rfloor} \left\{ \sum_{k=(n-2j),2}^{n+2j} \left[\binom{2j}{j+d} a_k^{(1)} \frac{b_+^{j+d} b_-^{j-d}}{(2j)!} \right] \right\}. \quad (18)$$

For the analysis of the $4\omega_{\text{IR}}$ component, the treatment of the amplitude can be restricted to the lowest-order contributions arising from each of the harmonics that are within $4\omega_{\text{IR}}$ of harmonic n . This gives

$$\begin{aligned} a_{n,\text{min}}^{(1 \text{ to } 5)} = & a_n^{(1)} + a_{n-2}^{(1)} \frac{b_{(n-2)+}^{(1)} b_{(n-1)+}^{(1)}}{2} + a_{n+2}^{(1)} \frac{b_{(n+2)-}^{(1)} b_{(n+1)-}^{(1)}}{2} \\ & + a_{n+4}^{(1)} \frac{b_{(n+4)-}^{(1)} b_{(n+3)-}^{(1)} b_{(n+2)-}^{(1)} b_{(n+1)-}^{(1)}}{24} \\ & + a_{n-4}^{(1)} \frac{b_{(n-4)+}^{(1)} b_{(n-3)+}^{(1)} b_{(n-2)+}^{(1)} b_{(n-1)+}^{(1)}}{24}. \end{aligned}$$

By considering the interference terms arising from pathways involving harmonics that differ in energy by $4\omega_{\text{IR}}$, the $4\omega_{\text{IR}}$ interference signal is obtained as

$$\begin{aligned} |a_n^{(1 \text{ to } 5)}|_{4\omega}^2 = & \frac{\mu_{g,n}\mu_{g,n-4}^*\mu_{n-4,n-3}^*\mu_{n-3,n-2}^*\mu_{n-2,n-1}^*\mu_{n-1,n}^*E_nE_{n-4}E_0^4e^{-4i\omega_{\text{IR}}\tau}}{24} \\ & + \frac{\mu_{g,n}\mu_{g,n+4}^*\mu_{n+4,n+3}^*\mu_{n+3,n+2}^*\mu_{n+2,n+1}^*\mu_{n+1,n}^*E_nE_{n+4}E_0^4e^{4i\omega_{\text{IR}}\tau}}{24} \\ & - \frac{\mu_{g,n+2}\mu_{n+2,n+1}\mu_{n,n+1}\mu_{g,n-2}^*\mu_{n-2,n-1}^*\mu_{n-1,n}^*E_{n-2}E_{n+2}E_0^4e^{4i\omega_{\text{IR}}\tau}}{4} + \text{c.c.} \end{aligned} \quad (19)$$

Interferences in the central path [the interference term involving harmonics $(n+2)$ and $(n-2)$] are thus significantly more prominent compared to the upwards-only or downwards-only paths. This is in contrast to any of the previously derived interference expressions, where all pathways are equally important.

E. Resonance treatment

The treatment developed thus far is valid when the IR field is coupling continuum states only. However, many recent applications of RABBITT have probed the role of atomic structure, and specifically its effect on the measured RABBITT phase. This has been formalized in extensions to the RABBITT technique such as rainbow-RABBITT—

revealing signatures of autoionizing states (see for instance Refs. [27,28])—and underthreshold RABBITT (uRABBITT). In uRABBITT, the XUV field is chosen such that at least one harmonic resonantly excites a near-threshold bound state and absorption of an IR photon ionizes the system. The phase of the resonant bound state is then visible in the phase of the resulting sideband [11,18,29].

In the expressions for the interference terms derived above [Eqs. (6), (9), (15), and (19)] the phase imparted by an excited state $|n\rangle$ is implicitly included through its transition dipole matrix element, $\mu_{g,n}$, and complex conjugate. A full treatment will require a detailed integration over all intermediate states. For the present purpose, it is sufficient to consider the final outcome of such integration for a sideband within the continuum. The $(+/-)$ signs within the different expressions are

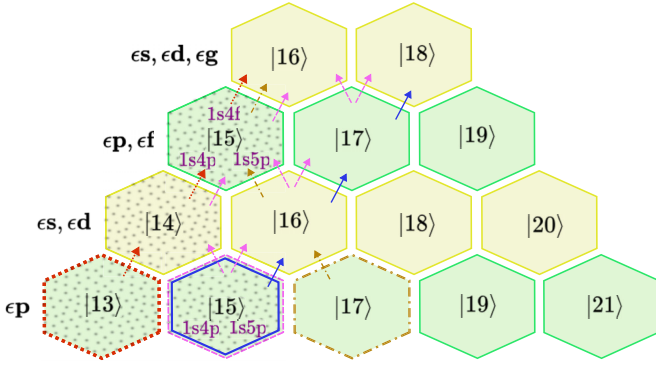


FIG. 3. Schematic in the style of Fig. 2, demonstrating the ionization paths to SB16 and SB18 in the helium uRABBITT investigation. Sidebands and mainbands lying below the ionization threshold are shaded, and the accessible bound states are labeled in purple. The partial waves permitted at every level of photon interaction (assuming four-photon paths) have been labeled to the left of each row. Four-photon paths to SB16 (dotted red, dashed pink, dot-dashed brown) and SB18 (solid blue) which pass through H15 have been marked to highlight that (at the four-photon level) SB18 can contain a resonance imprint from the $1s4p$ and $1s5p$ states, but not the $1s4f$ state.

important to keep track of, as they manifest as phase changes of $\pm\pi$.

Once the signature of a resonance has been established within a sideband above the ionization threshold, the further propagation of resonance signatures is visualized in Fig. 3. In the presented setup, the $1snl$ states cover the energy range spanned by harmonic 13 to 15, including sideband 14. H15 in the APT allows excitation of the $1s4p$ and $1s5p$ states. Subsequent absorption of an IR photon then connects these resonant states to SB16. Once SB16 is reached, the resonance signature can progress within the continuum to H17 and SB18 by further absorption of IR photons. The dominant pathways contributing at a sideband or mainband level will come from the diagonals; these connect the level of interest with an initial harmonic through the minimum number of IR photons. Interference terms for frequency $2\omega_{\text{IR}}$ or $4\omega_{\text{IR}}$ are then obtained via diagonals originating from initial harmonics separated by two or four IR photons in energy.

The figure also demonstrates that H15 pathways through the $1snp$ states can contribute to SB18, but those through $1snf$ states cannot (to lowest order). This is because excitation from the $1s$ ground state to the $1s4f$ state (aligned with H15) requires the absorption of at least three photons. Subsequent excitation to SB18 requires three further photon absorptions. This also means that the $4\omega_{\text{IR}}$ phase in SB16 is complicated by the presence of the $1s4f$ states. The $2\omega_{\text{IR}}$ phase in SB16, by contrast, is dominated by the two photon pathways, and thus the influence of the $1snf$ states will not be visible therein.

III. ATOMIC RESONANCES IN RABBITT-HOP SPECTRA

A. Helium description and simulation setup

To investigate the propagation of resonance imprints, we numerically replicate an energy-resolved RABBITT-HOP experiment for He, set up such that the H15 XUV component

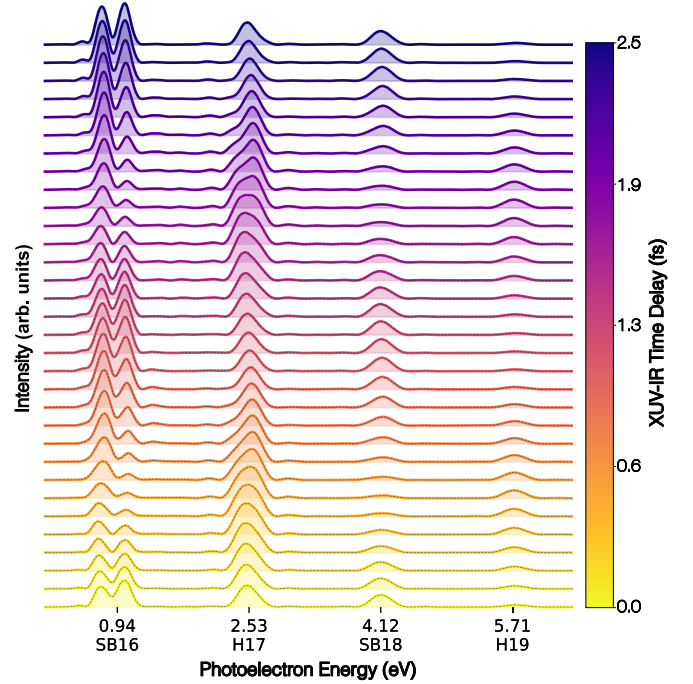


FIG. 4. Emission angle integrated, XUV-IR delay resolved photoelectron energy spectra resulting from the simulated RABBITT-HOP experiment of helium described in Sec. III A. Sidebands and mainbands are marked by both their photoelectron energies and the harmonics of the IR field to which they correspond. The double peak structure in SB16 is indicative of the contribution of pathways via the below-threshold $1s4p$ and $1s5p$ states, as are the changes in the shape of the H17 and SB18 peaks with respect to time delay. All raw data and analysis scripts can be obtained via Ref. [43].

dresses the $1s4p$ and $1s5p$ Rydberg states. By tuning the laser parameters thus, SB16 results from interference between the laser-assisted resonant ionization pathway via H15, and a direct photoionization pathway via H17 [11,18,30–38]. We study how these resonant states modify the photoelectron yield measured in SB16 and also in higher-lying sidebands and mainbands as a function of delay time of the IR field with respect to the XUV field. This behavior with delay time is then analyzed through a Fourier decomposition to determine the RABBITT phase as a function of energy.

Simulations are performed with the RMT code [39,40]. Ionization channels of the form $1s\ell l$ $\{l < 32\}$ are included in the model. No higher-lying thresholds are included in the calculations. The ground-state energy of He was set to its experimental value. The IR field was set to a wavelength of 775.5 nm and an intensity of $1 \times 10^{12} \text{ W cm}^{-2}$. The corresponding $2 \times 10^{10} \text{ W cm}^{-2}$ XUV pulse train is centered on the 15th harmonic of the IR (23.98 eV). These parameters were established previously to approximate experimental conditions targeting excitation of the $1s4p$ and $1s5p$ bound states [18]. From the final state of the simulated system, we calculate the photoelectron energy spectrum, and repeat this for 32 XUV-IR delays spanning one IR period (Fig. 4). The double peak structure, due to the 0.3-eV energy gap between the $1s4p$ and $1s5p$ states, is visible in SB16 in Fig. 4. The figure furthermore shows significant dynamics within the H17

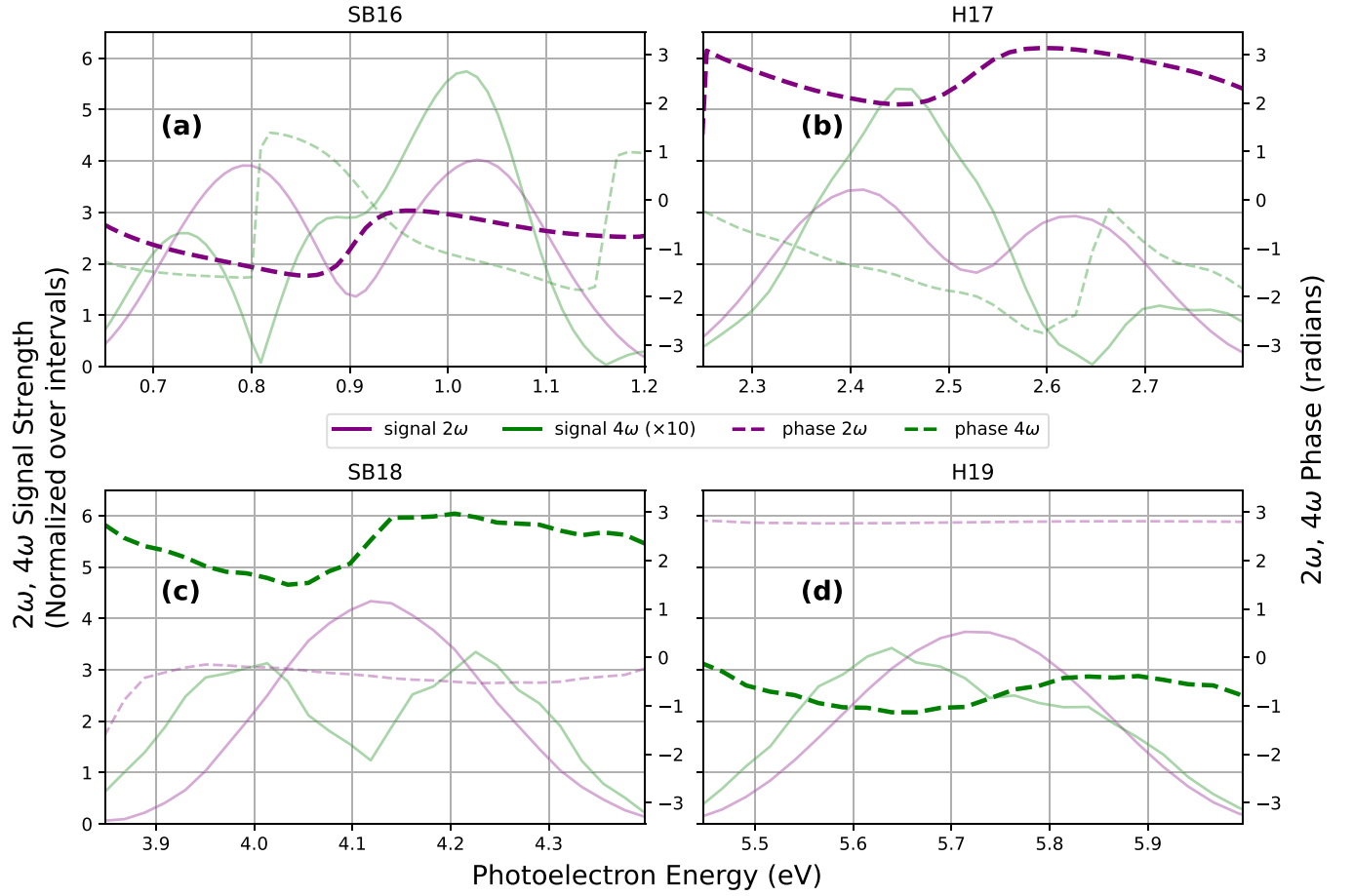


FIG. 5. Sideband and mainband $2\omega_{\text{IR}}$ and $4\omega_{\text{IR}}$ signals (solid lines) and phases (dashed lines) in SB16 (a), H17 (b), SB18 (c), and H19 (d) for two color photoionization of He. The resonance information imprinted from the $1snp$ bound states causes a distinct antiresonance in the SB16 $2\omega_{\text{IR}}$ phase centered on 0.9 eV (a). Pathways involving three photons translate this imprint into the H17 $2\omega_{\text{IR}}$ phase (b), and four- and five-photon processes translate into the $4\omega_{\text{IR}}$ phase of SB18 (c) and H19 (d), respectively.

peak as a function of time delay. The most likely origin of this behavior is the $1s4p$ and $1s5p$ states. This provides motivation to analyze higher-order contributions of the RABBITT phase.

To decompose the time-delay spectrum into its frequency components, we compute the Fourier transform of the photoelectron yield along the time-delay axis resolved in energy across SB16, H17, SB18, and H19. This yields Fourier components $y_{n\omega}(\epsilon)$ for $n = 0, 1, \dots, 15$, from which the energy-resolved amplitude, A , and phase Φ are extracted via

$$A_{n\omega}(\epsilon) = \sqrt{\text{Re}[y_{n\omega}(\epsilon)]^2 + \text{Im}[y_{n\omega}(\epsilon)]^2},$$

$$\Phi_{n\omega}(\epsilon) = \arctan\left(\frac{\text{Im}[y_{n\omega}(\epsilon)]}{\text{Re}[y_{n\omega}(\epsilon)]}\right),$$

for each $n\omega$ signal component.

B. $1snp$ imprints in multiple sidebands and mainbands

Figure 5 shows the $2\omega_{\text{IR}}$ and $4\omega_{\text{IR}}$ signal amplitudes and phases for SB16/18 and H17/19 for an IR intensity of $10^{12} \text{ W cm}^{-2}$. The $2\omega_{\text{IR}}$ phase across SB16, Fig. 5(a), varies strongly as a function of energy, with a characteristic “antiresonance” shape centered on 0.9 eV [30]. This phase behavior

is a signature of resonant $1s4p$ and $1s5p$ states’ contribution to the photoelectron yield at this energy.

The same characteristic phase behavior is observed in the H17 $2\omega_{\text{IR}}$ phase in Fig. 5(b), and in the SB18 and H19 $4\omega_{\text{IR}}$ phase in Figs. 5(c) and 5(d), as predicted in Sec. II E.

The π shift in the phase for H17 and SB18 arises from the sign with which the $\mu_{g,15}^*$ matrix element is included in the interference expressions in Eqs. (6), (9), (15), and (19), as discussed in Sec. II E.

As shown through Eqs. (9) and (6), the $2\omega_{\text{IR}}$ phase across a mainband may be estimated from the $2\omega_{\text{IR}}$ phase across its neighboring sidebands. Figure 5(b) shows the $2\omega_{\text{IR}}$ phase for H17. We can see that this phase mirrors the change in $2\omega_{\text{IR}}$ phase seen for SB16. Figure 5(c) shows that the $2\omega_{\text{IR}}$ phase for SB18 is constant across the sideband, as expected for a structureless continuum, which confirms that the origin of the phase change in SB16 and H17 is the same. However, as the $2\omega_{\text{IR}}$ signal in H17 corresponds to depletion of the SB16 yield, the phase change must be shifted by π compared to SB16. This confirms that the $2\omega_{\text{IR}}$ phase of H17 [Fig. 5(b)] can be attributed to the SB16 signal component, and thereby to the Rydberg states dressed by H15.

The next-order influence of the resonance imprint on the phase is visible in the $4\omega_{\text{IR}}$ phase in SB18, as shown in

Fig. 5(c). As described by Eq. (15), this component is composed of two contributions: interference between pathways through H15 and H19, and between pathways through H17 and H21. The $2\omega_{\text{IR}}$ component in H19 [Fig. 5(d)] is constant across all energies, showing absence of resonance terms associated with harmonics H17 to H21. Thus the phase variation can be ascribed to the $1s4p$ and $1s5p$ resonances reached at H15. Figure 5(c) shows that the phase variation remains shifted by π from the imprint seen for SB16 [Fig. 5(a)].

The resonance remains imprinted on the $4\omega_{\text{IR}}$ yield in H19 as shown in Fig. 5(d). However, here we can see a reduction in the overall variation of the phase compared to the overall variations seen in SB16, H17, and SB18. As shown in Eq. (17), the interference terms leading to a $4\omega_{\text{IR}}$ oscillation are H15-H19, H17-H21, and H19-H23. The relative strength of these components depends (a) on the relative strength of the different harmonic components and (b) on the combinatorics of emission and absorption processes. The combinatorics strongly favors the H17-H21 interference term over the H15-H19 and H19-H23 terms. The consequence of this is a reduction in the overall phase variation seen in the $4\omega_{\text{IR}}$ frequency component of H19. Analysis of Eq. (17) shows that a phase change of π relative to the $4\omega_{\text{IR}}$ frequency component of SB18 should be expected, and this is indeed seen in Fig. 5(d).

Analysis of the amplitude and phase of the $4\omega_{\text{IR}}$ component in SB16 is not as straightforward. As shown in Fig. 5(a), its amplitude crosses zero near 0.8 eV and near 1.15 eV, with a rapid phase change at these crossings. However, a phase change of about π is still observed between these two zero crossings. Many resonant ionization paths can contribute to the SB16 $4\omega_{\text{IR}}$ signal: H15 can excite the $1s4p$ and $1s5p$ resonances directly, whereas two-photon absorption from H13 can excite not just these resonances, but also $1s4f$ and $1s5f$. Detailed analysis of the H13 contribution is required, so it is difficult at present to ascribe this phenomenon to a particular process. Nonetheless these results provide compelling evidence that further bound-state dynamics can be explored using the RABBITT technique, involving more deeply bound atomic structure.

The complexity of the $4\omega_{\text{IR}}$ frequency component in SB16 is mirrored in the $4\omega_{\text{IR}}$ frequency component of H17 [Fig. 5(b)]. A zero crossing in the amplitude is seen at an energy of 2.65 eV. This does not match the positions of either of the zero crossings seen in SB16. For these higher-order processes, a more detailed treatment of all possible pathways below threshold is thus required. However, the influence of resonant processes remains visible, as the phase of the $4\omega_{\text{IR}}$ frequency component varies steadily with energy.

C. Intensity analysis of $2\omega_{\text{IR}}$ signals

Application of the RABBITT-HOP formalism requires limiting the description to processes involving at most p photons. The standard RABBITT approach involves two photons (one XUV and one IR photon), but the validity of this assumption breaks down here at a moderate intensity of 10^{12} W/cm². Using the present formalism, we may use H17 to calibrate the range of intensities for which this standard approach is valid.

We mentioned earlier that the H17 $2\omega_{\text{IR}}$ phase is a function of the SB16 and SB18 $2\omega_{\text{IR}}$ phase (see the Appendix):

$$\phi_{(\text{H17}, 2\omega)} \simeq \arctan \left(\frac{|E_{15}|\sin(\phi_{(\text{SB16}, 2\omega)} + \pi) + |E_{19}|\sin(\phi_{(\text{SB18}, 2\omega)} + \pi)}{|E_{15}|\cos(\phi_{(\text{SB16}, 2\omega)} + \pi) + |E_{19}|\cos(\phi_{(\text{SB18}, 2\omega)} + \pi)} \right). \quad (20)$$

This reconstruction will be accurate only if the contribution of four-photon pathways is negligible.

Figure 6 shows the $2\omega_{\text{IR}}$ phase of H17 as extracted directly from the RMT calculation results, alongside its reconstruction as a combination of the SB16 and SB18 $2\omega_{\text{IR}}$ phases, for six IR intensities spanning 5×10^{10} to 2×10^{12} W/cm². At 5×10^{10} W/cm², the H17 $2\omega_{\text{IR}}$ signal is weak, and may be influenced by spectral “spillover” from the numerical description of the XUV pulse. Nevertheless, even at this low intensity, the general behavior still replicates the antiresonance behavior. For even lower intensities, the oscillations obscure the behavior, and we thus regard 5×10^{10} W/cm² as a lower limit on the intensity required to extract useful phase information from a mainband in our calculations.

The antiresonance is observed in the reconstruction for all intensities shown in Fig. 6. Quantitative agreement between the extracted and reconstructed phases breaks down at 2×10^{12} W/cm². However, the lower intensity of 10^{12} W/cm² is already showing a greater deviation between the two phases compared to 5×10^{11} W/cm². This suggests that even at a relatively modest intensity of 10^{12} W/cm², four-photon processes are playing a role. This corroborates the evidence of Fig. 5 which showed clear signatures of four- and five-photon processes at this intensity.

D. Intensity analysis of $4\omega_{\text{IR}}$ signals

Figure 7 demonstrates the range of IR intensities for which the $1snp$ antiresonance can be observed in the $4\omega_{\text{IR}}$ phase of SB18 and H19. In SB18, a minimum intensity of 1×10^{11} W/cm² is needed for antiresonance to be clearly observed. In H19, a higher minimum intensity of 5×10^{11} W/cm² is required. As in Fig. 6, at lower intensities, the extracted phases are “noisy” due to the low signal-to-noise ratio. In both SB18 and H19, the antiresonance is no longer unambiguously observed beyond 2×10^{12} W/cm². Thus the intensity regime between 5×10^{11} and 2×10^{12} W/cm² can be considered as a transition regime, below which one can fully ignore higher-order processes in the RABBITT scheme, and above which one has to apply a fully nonperturbative method.

IV. CONCLUSIONS

We present RABBITT-HOP, which allows interpretation of RABBITT schemes at higher intensities such that pathways involving more than two photons contribute non-negligibly to the observed photoelectron spectra. In particular, by tracking the contribution of a specific transition dipole matrix element—which involves a resonant state—to the measured oscillations, we predict the appearance of

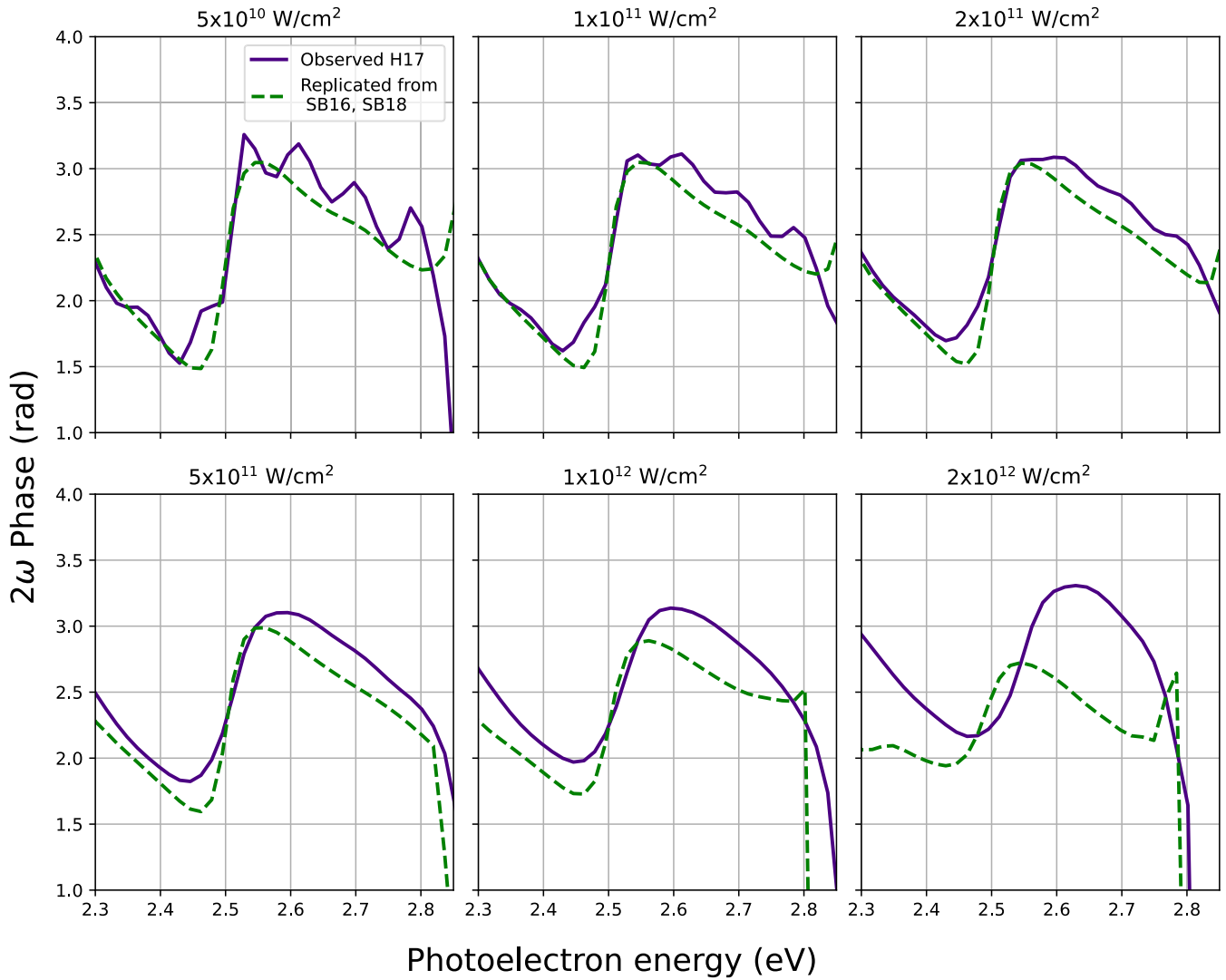


FIG. 6. Extracted (solid) and reconstructed (dashed) H17 $2\omega_{\text{IR}}$ phases for a range of IR intensities. The reconstructed phases use the neighboring SB16 and SB18 signals [Eq. (20)] and should be accurate if four-photon processes are negligible. For all intensities, the characteristic “antiresonance” shape is visible in both the directly extracted and reconstructed phases. At the lowest intensities, additional oscillations are visible in the directly calculated phase, which we attribute to background contributions from the numerical description of the XUV pulse. At an intensity of $2 \times 10^{12} \text{ W/cm}^2$, the quantitative agreement between extracted and reconstructed phases breaks down, indicating the non-negligible contribution of four-photon processes.

signatures of that resonance in the higher-lying sidebands and mainbands.

To verify these predictions we performed a simulated RABBITT investigation of the $1s4p$ and $1s5p$ bound states of neutral helium. We identified the characteristic antiresonance signature of the energy-resolved resonance in the phase of the $2\omega_{\text{IR}}$ signal in SB16, the sideband immediately above the dressed bound states, and showed how this resonance imprint propagates to the $2\omega_{\text{IR}}$ oscillation of the H17 mainband, and further to the $4\omega_{\text{IR}}$ oscillations in SB18 and H19. Finally we analyzed the ranges of IR intensities where we expect to see these behaviors, i.e., the window of intensities such that the multiphoton pathways contribute non-negligibly, and the resonance effects are not washed out by other, nonresonant pathways. We find that, for a wavelength around 800 nm, the intensity range between 5×10^{11} and $2 \times 10^{12} \text{ W cm}^{-2}$ holds the most promise.

Further structure is observed in $4\omega_{\text{IR}}$ oscillations in SB16 and H17. These imprints cannot be readily interpreted in terms of the $1s4p$ and $1s5p$ resonances, and may contain effects arising from other bound states, either lower in the Rydberg series, or with higher angular momentum. However, to date no uRABBITT investigation of a more “deeply” bound state lying more than around 1.6 eV (approximately the energy of one IR photon) has been conducted. With the RABBITT-HOP interpretation, we speculate that one could instead analyze a higher-energy, above-threshold, mainband or sideband to investigate the photoionization delays relating to the deeply bound state. However, the number of possible pathways in this specific instance meant the analysis was beyond the scope of this paper.

The *phase* of the different $n\omega_{\text{IR}}$ oscillations provides a more sensitive measure of the contributing processes than could be achieved by considering only the amplitude of the

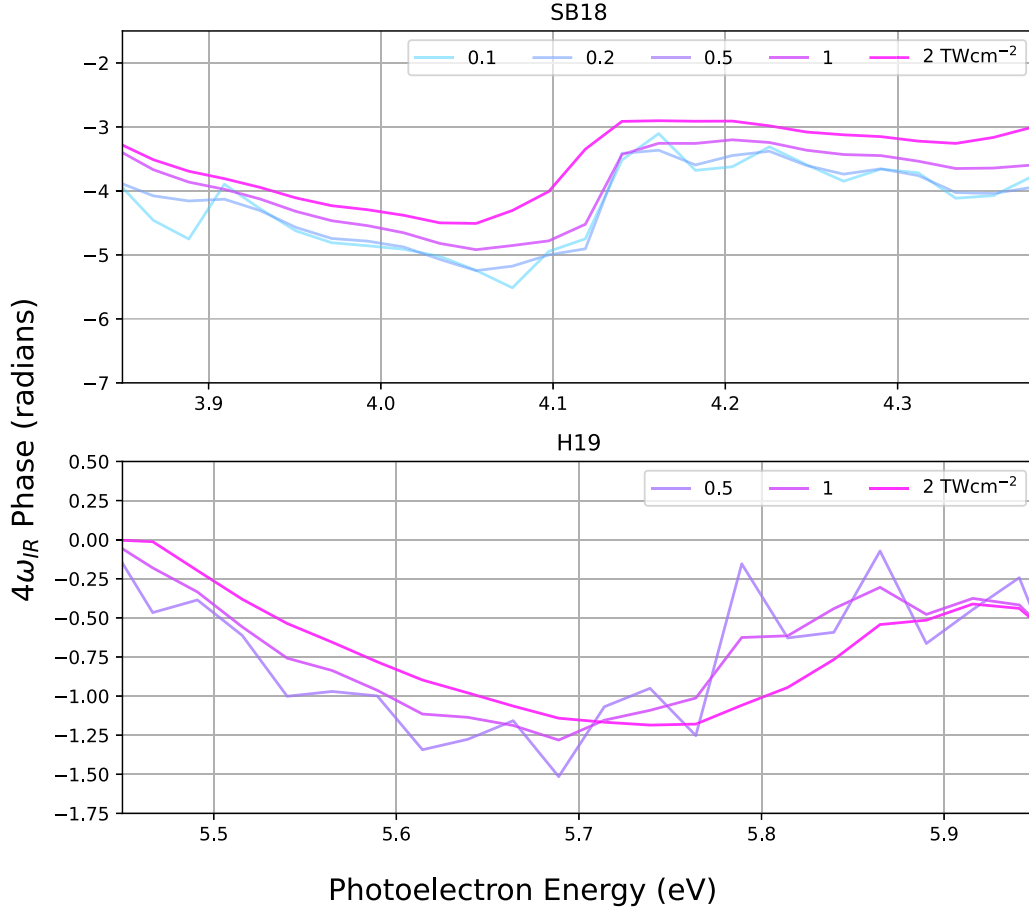


FIG. 7. Calculated $4\omega_{\text{IR}}$ phase across SB18 (top panel) and H19 (bottom panel) for the range of IR intensities where the characteristic antiresonance is visible. The $4\omega_{\text{IR}}$ signal was too weak to meaningfully extract the phase for IR intensities below 10^{11} W/cm² in SB18 and below 5×10^{11} W/cm² in H19. Beyond 2×10^{12} W/cm² in both cases the antiresonance is not visible.

measured spectra. These phases show signatures of the resonant states in the amplitude of the $2\omega_{\text{IR}}$ and $4\omega_{\text{IR}}$ oscillations (Fig. 5), even in more distant sidebands. We speculate, therefore, that extraction of the $2\omega_{\text{IR}}$ and $4\omega_{\text{IR}}$ phase may provide a sensitive measure of these resonant effects even in challenging experimental conditions.

Previous studies have addressed these higher-order effects, but our approach differs in two important respects. First, we predict and describe the role of high-order effects in *standard* RABBITT schemes, as opposed to those designed specifically to suppress the lowest-order contributions (such as Ref. [15]). More importantly, while the earliest work in this area treated atomic structure effects as secondary corrections that complicate the analysis [14], our RABBITT-HOP approach demonstrates how atomic resonances propagate systematically through higher-order processes, transforming higher-order effects from a limitation of standard RABBITT into a new spectroscopic tool for probing atomic structure and electron dynamics.

The present study has focused on the energy-resolved phase and amplitude of the frequency components of the sidebands. In principle, this study can be extended further to resolve the angular distributions as well. This may provide additional information on the interplay between the different partial waves.

ACKNOWLEDGMENTS

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DATA AVAILABILITY

The data that support the findings of this article are openly available [43].

APPENDIX: PHASE OF COMBINED SIGNALS

Let S be the sum of n $2\omega_{\text{IR}}$ -frequency signals, $S_1, \dots, S_n$, each defined as

$$S_i = A_i \cos(2\omega_{\text{IR}}t + \phi_i).$$

Then we have that

$$\begin{aligned}
 S &= \sum_i^n A_i \cos(2\omega_{\text{IR}}t + \phi_i) \\
 &= \sum_i^n A_i \cos(\phi_i) \cos(2\omega_{\text{IR}}t) - A_i \sin(\phi_i) \sin(2\omega_{\text{IR}}t) \\
 &= \left[\sum_i^n A_i \cos(\phi_i) \right] \cos(2\omega_{\text{IR}}t) - \left[\sum_i^n A_i \sin(\phi_i) \right] \sin(2\omega_{\text{IR}}t).
 \end{aligned}$$

As the sum of $2\omega_{\text{IR}}$ -frequency signals will also be a $2\omega_{\text{IR}}$ -frequency signal, we may write S as

$$\begin{aligned}
 S &= A \cos(2\omega_{\text{IR}}t + \Phi) \\
 &= A \cos(\Phi) \cos(2\omega_{\text{IR}}t) - A \sin(\Phi) \sin(2\omega_{\text{IR}}t),
 \end{aligned}$$

and equate the $\cos(2\omega_{\text{IR}}t)$ and $\sin(2\omega_{\text{IR}}t)$ components directly:

$$A \cos(\Phi) = \sum_i^n A_i \cos(\phi_i), \quad A \sin(\Phi) = \sum_i^n A_i \sin(\phi_i).$$

Dividing the second component by the first, we may obtain the phase of the overall signal:

$$\begin{aligned}
 \tan(\Phi) &= \frac{\sum_i^n A_i \sin(\phi_i)}{\sum_i^n A_i \cos(\phi_i)} \\
 \Rightarrow \Phi &= \arctan\left(\frac{\sum_i^n A_i \sin(\phi_i)}{\sum_i^n A_i \cos(\phi_i)}\right).
 \end{aligned}$$

In the main text, this is applied to interpret the $2\omega_{\text{IR}}$ phase across H17 in terms of the contributions from the neighboring SB16 and SB18. Using the expressions for the sideband signal given by Eq. (6) with $n = 17$ and 19 (for SB16 and SB18), respectively, we can calculate that the phase of the H17 $2\omega_{\text{IR}}$ oscillation is

$$\begin{aligned}
 \phi_{(\text{H17}, 2\omega)} &\simeq \text{atan} \\
 &\times \left(\frac{|E_{15}| \sin(\phi_{(\text{SB16}, 2\omega)} + \pi) + |E_{19}| \sin(\phi_{(\text{SB18}, 2\omega)} + \pi)}{|E_{15}| \cos(\phi_{(\text{SB16}, 2\omega)} + \pi) + |E_{19}| \cos(\phi_{(\text{SB18}, 2\omega)} + \pi)} \right). \quad (\text{A1})
 \end{aligned}$$

This is the formula used to reconstruct the phase in Fig. 6.

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