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Non-Hermitian quantum mechanics versus the conventional quantum mechanics: Effect of the relative phasing of bichromatic fields on high-order harmonic generation

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We perform a dynamical symmetry analysis (DSA) of the high-order harmonic generation (HHG) spectrum of an atom interacting with a bichromatic laser field. Within the framework of the conventional Hermitian quantum mechanics (QM), the HHG spectrum calculated using a single Floquet state, or *any finite number of Floquet states*, is invariant under the inversion of the relative phase of the two-frequency components, $\phi \rightarrow -\phi$. The asymmetry with respect to the phase inversion seen in the simulated HHG spectra is obtained in the conventional QM *only* when the Floquet spectrum is continuous and ionization is taken into consideration. However, when the Hamiltonian is complex scaled the description is different. Even a single eigenstate of the complex scaled Floquet operator is enough to describe the breaking of the $\phi \rightarrow -\phi$ symmetry in the HHG spectra. We find that there is a direct correlation between the strength of the asymmetry with respect to the relative phase inversion and the magnitude of the ionization rate. For illustration purposes, the DSA is accompanied by the results obtained for a one-dimensional effective single-electron model Hamiltonian mimicking xenon atom interacting with strong laser field.

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I. INTRODUCTION: THE “PUZZLE”

Since the beginning of the 1990s, many experimental and theoretical investigations have been carried out, in which the photoinduced dynamics was controlled by means of varying the relative phase of two laser beams. The laser beams employed in these studies were typically first and second or third harmonics having comparable intensities. The two particular phenomena that attracted the attention of the researchers have been the high-order harmonic generation (HHG) and above threshold ionization (ATI). Let us mention here only those publications which are most relevant to the subject of the present work, that is, to the phase-asymmetry effect in the HHG spectra in bichromatic fields.

In 1992 Schafer and Kulander showed in their calculations that the change in sign of the relative phase in 1ω-2ω fields can lead to completely different directional ionization rates, angular distributions, and ATI spectra [1]. The asymmetry in the directional ionization rates in the tunneling regime was explained by the authors by comparing the electric fields resulting from the opposite values of the relative phase. In 1995, Alon and Moiseyev used the dynamical symmetry properties of the Floquet Hamiltonian to show that the phase asymmetry in the directional ionization rates is a general effect holding for an arbitrary ionization regime [2]. The predictions of Schafer and Kulander were confirmed in 1994 by the experiment of Schumacher, Weihe, Muller, and Bucksbaum summarized in Ref. [3]. The observed asymmetry in the ATI rates regime has been attributed to the recollision of the ionized electron with the core (see Ref. [4]). Since in the low-frequency regime HHG is a consequence of the recollision of the ionized electron with its parent ion (tunneling mechanism) [4,5], one might expect to get the similar phase-asymmetry effect for the HHG spectra too. Indeed, Zuo and Bandrauk together with Ivanov and Corkum showed in 1995 that the phase asymmetry is characteristic also of the HHG spectra [6]. Analytical calculations for a model potential which consists of two δ functions performed by Long, Becker, and McIver have also shown that there is no symmetry in HHG spectra with respect to inversion of the sign of the relative phase, i.e., $\phi \rightarrow -\phi$, although the $\phi \rightarrow \phi + \pi$ symmetry is obeyed [7]. However, in 1999 Bivona, Burlon, and Leone argued that since the time-periodic Hamiltonian does not change under $\phi \rightarrow -\phi$, $t \rightarrow -t$, and $z \rightarrow -z$, the power spectrum of the radiation is unaffected by the phase inversion, $\phi \rightarrow -\phi$ [8]. This general statement was illustrated by the results of the numerical calculations for two-level model.

The experimental results obtained by Andiel, Tsakiris, Cormiris, and Witte in 1999 [9] do not show a dramatic asymmetry with respect to the phase inversion, although the scatter of the measured harmonic intensities could mask some degree of phase asymmetry. Consequently, the experiment leaves the question “is the HHG spectrum invariant under the transformation $\phi \rightarrow -\phi$ or not?” unanswered.

More recently, in 2000, Faria, Milošević, and Paulus carried out numerical studies of the phase-dependence effects in bichromatic HHG [10]. In the experimental conditions their solution of the one-dimensional (1D) time-dependent-Schrödinger equation (TDSE) clearly shows that the HHG spectrum is not invariant under the $\phi \rightarrow -\phi$ transformation (see Fig. 7 in Ref. [10]).

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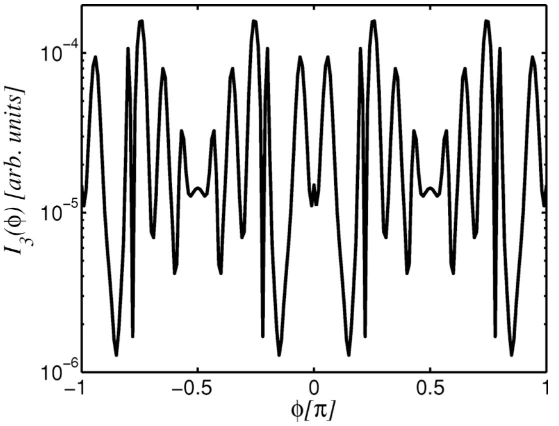

FIG. 1. The third harmonic as a function of the relative phase ϕ, $I_3(\phi)$, as obtained from the propagation of a single Floquet state within the framework of the conventional (i.e., Hermitian) QM. The spectrum is *invariant* with respect to the translational $\phi \to \phi + \pi$ and inversional $\phi \to -\phi$ transformations.

The "puzzle" is, thus, in the apparent contradiction between the results obtained from propagation calculations solving the TDSE (e.g., Ref. [10]) and the results obtained by using Floquet theory [8]. Does it imply that Floquet states do not form a complete basis set? This might be a very serious problem due to the fact that Floquet theory enables us to carry out analytical investigation of the problem (see, for example, the analysis of the HHG based on dynamical symmetry properties of the Hamiltonian [11]), and also enables one to use time-independent scattering theory for time-dependent Hamiltonian [12,13].

Let us illustrate the puzzle by representing the results obtained for a 1D Gaussian potential well, $V(x) = -0.63\exp[-(x/2.65)]^2$. This potential supports three bound states: $E_0 = -0.4451$ a.u., $E_1 = -0.1400$ a.u., and $E_2 = -0.00014$ a.u. The first two eigenstates mimic the lowest electronic states of xenon atom. The electric field generated by the bichromatic laser beam is given by $E(t,\phi) = \epsilon_0 f(t)[\cos(\omega t) + \cos(2\omega t + \phi)]$, where $f(t)$ represents the shape of the pulse envelope. In the case of cw lasers $f(t) = 1$. In Fig. 1 we represent the third-harmonic intensity as a function of the relative phase ϕ as obtained for a single Floquet state. The laser frequency is $\omega = 0.0924$ a.u., ($\hbar\omega \sim 2.5$ eV, $\lambda = 493$ nm), and the field amplitude, strength parameter is $\epsilon_0 = 0.035$ a.u. In two-color laser experiment the field intensity depends on the relative phase, $I = c/(8\pi)\max|E(t,\phi)|^2$. For $\phi = 0$, $I \sim 1.3 \times 10^{14}$ W/cm^2 and for $\phi = \pi/2$, the laser intensity is equal to $I \sim 1.76 \times 10^{14}$ W/cm^2. The Floquet state $\Phi(x,t,\phi) = \Phi(x,t+T,\phi)$, where $T = 2\pi/\omega$ was calculated numerically by the diagonalization of the time evolution operator according to the (t,t') algorithm [14] using 500 particle-in-a-box basis functions with the box size $L = 150$ a.u.

For time-periodic systems (i.e., when the shape of the laser pulse is not taken into consideration and the laser is described as a cw laser) the intensity of the nth harmonic, $I_n(\phi)$, is given by $I_n(\phi) \propto |(NT)^{-1}\int_0^{NT}\ddot{d}(t,\phi)\exp(\pm i\omega nt)dt|^2$, where $T = 2\pi/\omega$ is the time period, and the time-dependent dipole is given by $d(t,\phi) = \langle\Phi(x,t,\phi)|x|\Phi(x,t,\phi)\rangle$. N is the number of optical cycles which are taken into consideration in our calculations of the HHG spectra. In propagation calculations, $\Phi(x,t,\phi)$ is the solution of the time-dependent Schrödinger equation which can be described as a linear combination of Floquet states. In such a case one should take the limit of $N \to \infty$. However, for systems being described by a single Floquet state, $N = 1$ and the time-periodic part of the Floquet wave function, $\Phi(x,t,\phi)$, is the eigenfunction of the Floquet Hamiltonian: $\mathcal{H}_f\Phi(x,t,\phi) = E^{QE}(\phi)\Phi(x,t,\phi)$, $\mathcal{H}_f = -i\partial/\partial t + H(x,t,\phi)$. The time-dependent Hamiltonian of our model is $H(x,t,\phi) = -0.5d^2/dx^2 + V(x,t,\phi)$. The time-dependent potential is defined as $V(x,t,\phi) = V(x) - exE(t,\phi)$, where e is the charge of the electron.

As one can see, the third-harmonic generation intensity $I_3(\phi)$ is invariant under the transformation $\phi \to -\phi$ and also under the transformation $\phi \to \phi + \pi$. Note that in our calculations we followed the same Floquet state as ϕ was varied. This behavior is obtained for *any* high-order harmonic and for *any* Floquet state $\Phi_\alpha(x,t,\phi)$, where $\alpha = 1,2,\ldots$ denotes the αth quasienergy (QE) Floquet state. It is possible to prove that the same two types of symmetry behavior, i.e., $I_n(\phi) = I_n(-\phi)$ and $I_n(\phi) = I_n(\phi + \pi)$, are obtained for *any* linear combination of discrete Floquet states. The Floquet spectrum is discrete only due to the use of finite number of basis functions and a finite box size (or finite number of grid points). However, this proof holds for any finite number of basis functions and for any value of L which can be taken as large as one wishes. The proof, based on dynamical symmetry analysis (see Ref. [15] for the treatment of dynamical symmetries of time-periodic Hamiltonians) is as following.

Let P_2 be the dynamical symmetry operator of order 2 which is defined as $P_2 = (t \to -t; \phi \to -\phi; i \to -i)$. Since the commutator $[\mathcal{H}_f, P_2] = 0$, the Floquet states, eigenfunctions of $\mathcal{H}_f$, are also eigenfunctions of P_2. Therefore, $\Phi_\alpha(x,t,\phi) = \pm[\Phi_\alpha(x,-t,-\phi)]^*$. The Floquet eigenstates of P_2 which are associated with the +1 eigenvalue are denoted by the index j, whereas the Floquet states associated with the eigenvalue −1 are denoted by the index k. Since the diagonal dipole matrix elements $d_\alpha(t,\phi) = [d_\alpha(-t,-\phi)]^*$, regardless if $\alpha = j$ or $\alpha = k$, it is clear that $I_n^{(\alpha)}(\phi) = I_n^{(\alpha)}(-\phi)$. Note that the same HHG spectra are obtained when propagation is carried out forward or backward in time; namely, the calculation of the time-dependent dipole for $-\phi$ rather than $+\phi$ requires us to carry out backward time propagation rather than forward time propagation as in the case where $d_\alpha(t,\phi)$ is calculated. In the case of the conventional quantum mechanics (QM) this is not a problem at all since time is symmetric. This is an important point in our proof given above for the relative-phase symmetry properties of the nth HHG spectra calculated from a *single Hermitian Floquet state*. Note by passing that in non-Hermitian QM (NH-QM) time is asymmetric. We will return to this point later in the following section. In the case that the initial state is a square integrable function $\Psi(x,t=0)$ (for example can be taken as the field-free ground state) we can use the Floquet states as a basis set and the solution of the TDSE $\Psi(x,t)$ is given by

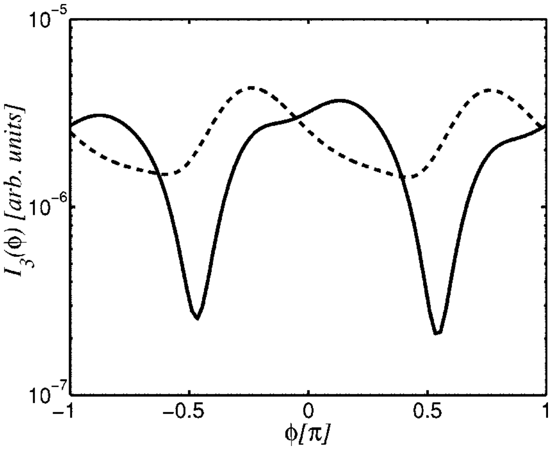

FIG. 2. $I_3(\phi)$, as obtained from the propagation of the ground field-free state within the framework of the conventional QM. The full solid line stands for the HHG spectrum obtained when the laser is adiabatically turned on [the envelope of the pulse is given by $f(t)=1-\exp(-0.000\,02t^2)$]. The dashed line stands for the HHG spectra obtained for the case where the laser pulse envelope is $f(t)=\sin^2[\omega/(2N)]$, where $N\geqslant 20$. These results show that the functional behavior of the HHG spectra with respect to the variation of the relative phase ϕ is sensitive to the shape of the laser pulse although the phase-asymmetry property is alike.

$$\Psi(x,t) = \sum\nolimits_\alpha C_\alpha \exp[-iE_\alpha^{QE}(\phi)t]\Phi_\alpha(x,t,\phi),$$

where $C_\alpha=\langle\Phi_\alpha(t=0,\phi)|\Psi(t=0)\rangle$. It is easy to see that the contributions to the exact harmonics in the Fourier transform of the dipole moment $\langle\Psi(t)|x|\Psi(t)\rangle$, and/or its second-order time derivative, are *only* from the diagonal terms $d_\alpha(t,\phi)$. Therefore, the HHG intensity is invariant under the transformation $\phi\rightarrow-\phi$ and $I_n(\phi)=I_n(-\phi)$. *This proof supports the statement of Bivona, Burlon, and Leone under the restriction that the initial state has to be defined as a linear combination of finite number of discrete Floquet states [8]*. As we will show later the key solution to the puzzle is in that restriction.

To complete the representation of the puzzle we carried out numerical calculations where $\Psi(x,t=0)$ was the field-free ground state. The TDSE has been solved when the box size L is taken to be large enough to avoid the artificial reflections from the edge of the box during the propagation. Two cases have been studied. In the first one the field was adiabatically turned on whereas in the second case the envelope of the laser pulse supported $N\geqslant 20$ optical cycles. The propagation was carried out as long as the acceleration $\ddot{d}(t)=\langle\Psi(t)|-\partial V(x,t,\phi)/\partial x|\Psi(t)\rangle$ had not been converged to that of the free particle in the field. The results for the third harmonic are presented in Fig. 2. As one can see the HHG spectra are invariant against $\phi\rightarrow\phi+\pi$ but $I_n(\phi)\neq I_n(-\phi)$. *This result is in a complete agreement with the results of Long-Becker-McIver and of Faria-Milošević-Paulus which were mentioned above*. Note by passing that the functional behavior of the HHG spectra with respect to the variation of the relative phase ϕ is sensitive to the shape of the laser pulse although the symmetry properties [i.e., $I_n(\phi)$ vs $I_n(-\phi)$ and $I_n(\phi)$ vs $I_n(\phi+\pi)$] are alike.

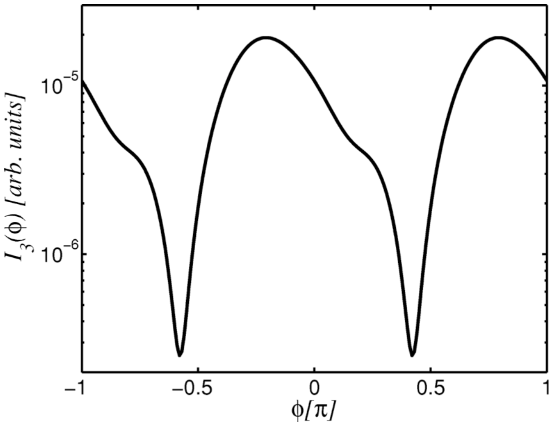

FIG. 3. $I_3(\phi)$, as obtained from the propagation of a single-resonance (longest-living) Floquet state within the framework of the non-Hermitian (NH) QM. The spectrum is invariant with respect to the translational $\phi\rightarrow\phi+\pi$. However, it is *not invariant* under the inversion of $\phi\rightarrow-\phi$.

Before closing the introductory part of this paper, let us present in Fig. 3 the results we have obtained for the third harmonic when the time-dependent dipole was calculated using a *single complex scaled resonance Floquet state*. We selected the longest-living resonance Floquet state assuming that this state controls the photoinduced dynamics. The resonance Floquet state is an eigenfunction of the complex scaled (non-Hermitian) Floquet operator, $\mathcal{H}_f^\theta=-i\partial/\partial t+H(x\exp(i\theta),t)$. For the calculations of resonances by the complex scaling procedure (complex coordinate method) see Ref. [16]. As one can see from the results presented in Fig. 3, $I_3(\phi)=I_3(\phi+\pi)$ but definitely $I_3(\phi)\neq I_3(-\phi)$. The results obtained from a single resonance Floquet state using (NH-QM) are very different from the results obtained from a single Floquet state calculated within the framework of the conventional QM. Moreover, the results obtained from a single resonance Floquet state are in good agreement with the results obtained from simulation calculations when the TDSE has been solved within the framework of the conventional QM. Due to the sensitivity of the HHG spectra to the shape of the laser pulse as stated above and shown in Fig. 2, we do not expect to have a quantitative agreement between the HHG spectra obtained from the time-dependent simulations. See the results for the HHG spectra presented in Fig. 4 obtained for the three different cases mentioned above. That is, in two out of the three cases the initial wave packet was propagated within the framework of the conventional QM (in one case the field was adiabatically turned on and in the second case the laser envelope supported 20 optical oscillations), whereas in the third case the HHG spectra were obtained from a single resonance Floquet state calculated within the framework of the NH-QM. As one can see on a logarithm scale all spectra look alike. However as we have shown in Figs. 2 and 3 the functional behavior of $I_n(\phi)$ phase dependency is quite different although in the three different cases the phase asymmetry is similar.

The strategy of solving the puzzle is as following. First we will show that the dynamical symmetry proof given above for the Hermitian time-dependent periodic Hamil-

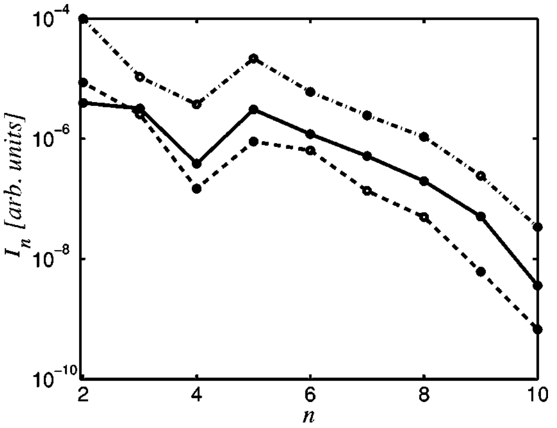

FIG. 4. The high-order harmonic generation spectra as obtained when $\phi=0$. The solid line stands for the results obtained by solving the time-dependent Schrödinger equation when the ac field is adiabatically turned on. The dashed line stands for the results obtained when the sine-square envelope of the laser pulse supports 20 optical oscillations. Both results are obtained within the framework of the conventional QM. The solid-dotted line denotes the results obtained by calculating the long-living resonance Floquet state using the complex scaling approach (non-Hermitian QM calculations).

tonian does not hold for the non-Hermitian complex scaled Hamiltonian. We base our arguments on physical grounds associating the discrepancy between the NH-QM formalism to the conventional one with the resonance lifetime. As the resonance lifetime is longer the discrepancy between the results should be smaller. Indeed our numerical calculations presented in Sec. II support this assumption. In Sec. III we show that when a *discrete* Floquet spectrum is used for the representation of the time evolution operator, the interference among Floquet states which are nondegenerate states of the dynamical symmetry operator are canceled. This interference effect results in the breaking of the symmetry of the HHG spectra to the inversion of the relative phase. This interference effect is taken into consideration when the TDSE is solved by carrying out propagation calculations as described above in this section. Moreover, we show that in the propagation calculations using the conventional (Hermitian) QM, the phase breaking symmetry effect results, as in NH-QM, from the photoinduced ionization phenomena.

II. EFFECT OF THE RELATIVE PHASE ON THE HHG SPECTRA IN NON-HERMITIAN QM CALCULATIONS

In Fig. 3 we demonstrate that the intensity of the third harmonic calculated from a single-resonance, complex scaled, Floquet state changes as $\phi\rightarrow-\phi$. This result is very different from the result obtained from the calculations of $I_3(\phi)$ with a single Hermitian Floquet state as shown in Fig. 1. Moreover, in Sec. I we proved that within the framework of the Hermitian QM $I_n(\phi)=I_n(-\phi)$ when the initial state is constructed from a linear combination of discrete Hermitian Floquet states. Why the results for the HHG spectra are different when NH-QM is used?

In order to answer this question we will briefly describe how the HHG spectrum is calculated in NH-QM. For the sake of simplicity and without loss of generality we denote the internal coordinate by x. The derivations we represent here (and also in the preceding section) hold also for many-electron systems where the electron correlations are taken into consideration. The resonance Floquet states are the eigenfunctions of the complex scaled Floquet Hamiltonian,

$$\mathcal{H}_f(x,t,\phi,\theta)\Phi_\alpha(x,t,\phi,\theta)=E_\alpha^{QE}(\phi)\Phi_\alpha(x,t,\phi,\theta), \tag{1}$$

where

$$\mathcal{H}_f(x,t,\phi,\theta)=-i\frac{\partial}{\partial t}+\hat{H}(x\exp(i\theta),t)+\epsilon_0 ex\exp(i\theta)\times[\cos(\omega t)+\cos(2\omega t+\phi)]. \tag{2}$$

The resonance complex quasienergies,

$$E_\alpha^{QE}(\phi)=\mathcal{E}_\alpha(\phi)-\frac{i}{2}\Gamma_\alpha(\phi), \tag{3}$$

are θ independent provided $\theta\geqslant\theta_c$ where the critical angle to explore the resonances is given by[16]

$$\theta_c=\tfrac{1}{2}\arctan[\Gamma_\alpha/(2\mathcal{E}_\alpha)]. \tag{4}$$

$\Gamma_\alpha(\phi)$ is the total ionization rate of decay of the αth resonance Floquet state. The time-periodic Floquet states can be expanded in a Fourier basis set,

$$\Phi_\alpha(x,t,\phi,\theta)=\sum_{n=-\infty}^{+\infty}e^{i\omega nt}\varphi_{n,\alpha}(x,\phi,\theta). \tag{5}$$

The Fourier components are the eigenvectors of the Floquet matrix **F** whose (n',n)th matrix element is defined as

$$F_{n',n}(x,\phi,\theta)=\frac{1}{T}\int_0^T e^{-i\omega n't}\mathcal{H}_f(x,t,\phi,\theta)e^{+i\omega nt}\,dt. \tag{6}$$

It is important to realize that the Fourier components are complex functions and are not analytical continuations to the complex plane of the solutions obtained in the conventional (Hermitian) QM.

For the calculations of the time-dependent dipole moments we should define the "bra" states in NH-QM. The bra states are associated with the left eigenvectors of the matrix **F**. Therefore, the bra states are the right eigenvectors of the transpose of **F**. Let us denote the bra Fourier components as $\varphi_{n,\alpha}^t(x,\phi,\theta)$. Note that at $\phi=0$, **F** is a complex and symmetric matrix and consequently, $\varphi_{n,\alpha}^t(x,\phi=0,\theta)=\varphi_{n,\alpha}(x,\phi=0,\theta)$. If $\phi\neq0$, then $\varphi_{n,\alpha}^t(x,\phi,\theta)\neq\varphi_{n,\alpha}(x,\phi,\theta)$. The bra resonance Floquet states (denoted by the superscript "t" which stands for "transposed") are given by

$$\Phi_\alpha^t(x,t,\phi,\theta)=\sum_{n=-\infty}^{+\infty}e^{-i\omega nt}\varphi_{n,\alpha}^t(x,\phi,\theta). \tag{7}$$

In NH-QM the time-dependent dipole moment amplitude calculated from a single-resonance Floquet state is given by

$$d^{NH\text{-}QM}(t,\pm\phi,\theta)=\int_{-\infty}^{+\infty}\Phi^{t}_{\alpha}(x,t,\pm\phi,\theta)x\Phi_{\alpha}(x,t,\pm\phi,\theta)dx. \tag{8}$$

The time-dependent dipole moment amplitudes $d^{NH\text{-}QM}(t,\pm\phi,\theta)$ as the resonance complex quasienergies $E^{QE}_{\alpha}(\phi)$ and as any other physical quantity are θ independent provided $\theta \geqslant \theta_c$ where the critical rotational angle is defined in Eq. (4). From Eq. (4) one can see that for sufficiently narrow resonances, $\theta_c \propto \Gamma_{\alpha}(\phi)$. Therefore, we will change the notation of the NH-QM value for the time-dependent dipole moment amplitude from $d^{NH\text{-}QM}(t,\pm\phi,\theta_c)$ to $d(t,\pm\phi,\Gamma_{\alpha}(\phi))$. The linear dependence of θ_c on Γ_{α} implies that θ_c gets smaller values as the resonance width becomes smaller. Since the resonance width is defined as the inverse of the resonance lifetime it implies that θ_c gets smaller values as the resonance lifetimes are larger. In Hermitian QM the quasienergies are real and therefore we may say that as the resonance width $\Gamma_{\alpha}(\phi)$ gets smaller values, the differences between $d(t,\phi,\Gamma_{\alpha}(\phi))$ and the values obtained for the time-dependent dipole moment amplitudes in the conventional QM become smaller as well. This is another property that will be clarified later and will help to explain the different results for the HHG spectra and its dependence of ϕ, which were obtained from a single Floquet state in the NH-QM and in the conventional one. Moreover, it will help us to understand why the strength of the phase-asymmetry effect on the HHG, quantified by

$$\Delta_n(\phi)=|I_n(\phi)-I_n(-\phi)|, \tag{9}$$

gets large values for specific values of ϕ and small for others and why it varies with the field intensity. Let us return to the calculation of the HHG spectra from a single resonance Floquet state. The HHG spectrum is given by the Fourier transform of the second-order time derivative of the dipole moment amplitude calculated from the αth resonance Floquet state,

$$I_n(\phi,\Gamma_{\alpha}(\phi)) \propto \left|\frac{1}{T}\int_0^T e^{i\omega nt}\ddot{d}(t,\phi,\Gamma_{\alpha}(\phi))dt\right|^2. \tag{10}$$

The variation of $I_n(\phi,\Gamma_{\alpha}(\phi))$ under the transformation $\phi\rightarrow-\phi$ can be understood by carrying out the dynamical symmetry (DS) analysis of the problem. The dynamical symmetry operator $P_2(t\rightarrow -t,\phi\rightarrow-\phi,\theta\rightarrow-\theta,i\rightarrow -i)$ commutes with the complex scaled Floquet operator. Therefore,

$$\mathcal{H}_f(x,t,\phi,\theta)=[\mathcal{H}_f(x,-t,-\phi,-\theta)]^{*} \tag{11}$$

and consequently the "ket" and the "bra" Floquet states satisfy the following equalities:

$$\Phi_{\alpha}(x,t,\phi,\theta)=\pm[\Phi_{\alpha}(x,-t,-\phi,-\theta)]^{*}, \tag{12}$$

$$\Phi^{t}_{\alpha}(x,t,\phi,\theta)=\pm[\Phi^{t}_{\alpha}(x,-t,-\phi,-\theta)]^{*}, \tag{13}$$

and

$$E^{QE}_{\alpha}(\phi)=E^{QE}_{\alpha}(-\phi). \tag{14}$$

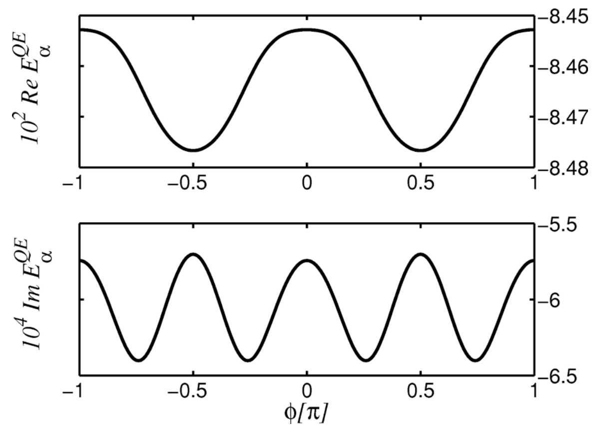

FIG. 5. The complex quasienergies of the longest-living resonance Floquet state, as obtained from complex scaled NH-QM calculations, as a function of the relative phase ϕ. The resonance position Re E^{QE}_{α}, and width (inverse lifetime) Im E^{QE}_{α}, are invariant with respect to the translation $\phi\rightarrow\phi+\pi$ and to the inversion of $\phi\rightarrow-\phi$ transformations.

The last equality is obtained due to the fact that when the coordinates are rotated backward (i.e., $\theta\rightarrow-\theta$) while the propagation is backward in time (i.e., $t\rightarrow -t$) the complex QE is embedded in the upper-half complex energy plane $[E^{QE}_{\alpha}(-\phi)]^{*}$ rather than in the lower one where the physical resonances are located. However, when the complex conjugate is taken following the requirement of the dynamical symmetry operator, the resonance energy $E^{QE}_{\alpha}(-\phi)$ is obtained. Indeed the phase inversion symmetry of the complex QE as given in Eq. (14) has been obtained in all our numerical calculations. For illustration purposes we present in Fig. 5 the ϕ trajectory of the resonance Floquet state which is associated with the narrowest (i.e., longest-living) resonance state. Now we return to the equality given in Eq. (12). If we were able to carry out backward propagation in time, then the right-hand side of the equality for the ket Floquet state would be associated with the bra Floquet state which is defined below:

$$\Phi_{\alpha}(x,-t,-\phi,-\theta)=\Phi^{t}_{\alpha}(x,t,-\phi,-\theta). \tag{15}$$

However, unlike $\Phi_{\alpha}(x,-t,-\phi,-\theta)$ and $\Phi_{\alpha}(x,+t,-\phi,+\theta)$ which are normalizable functions, $\Phi_{\alpha}(x,+t,-\phi,-\theta)$ and $\Phi_{\alpha}(x,-t,-\phi,+\theta)$ are not. The last two functions diverge exponentially in time. One should keep in mind that the motivation to the rotation of the internal coordinates in the Hamiltonian to the complex plane was to bring back the resonance wave functions (associated with outgoing Siegert/Gamow boundary conditions) which diverge exponentially in the coordinate space to the Hilbert space [16]. This is the source of the time-asymmetry problem in NH-QM [17]. If we would not have the time-asymmetry problem in NH-QM we would get the following equality:

$$d(t,\phi,\Gamma_\alpha(\phi)) = [d(t,-\phi,-\Gamma_\alpha(\phi))]^*, \tag{16}$$

which reminds us of the equality $d(t,\phi)=[d(-t,-\phi)]^*$ as obtained in the introduction part of the paper for the Hermitian Floquet state. Similarly to our derivation in the Introduction one might expect that if Eq. (16) holds then $I_n(\phi,\Gamma(\phi)) = I_n(-\phi,\Gamma(\phi))$. However, as we will explain below the assumptions that lead to Eq. (16) are not valid. In our derivation we assumed that the dipole moment on the right-hand side of Eq. (16) has been calculated with the nonphysical poles of the scattering matrix which are embedded in the upper-half complex energy plane rather than the lower one (for the association of the resonances with the poles embedded in the lower-half complex energy plane see, for example, Ref. [18]). These nonphysical poles diverge exponentially in time as $t\rightarrow\infty$. Due to the time-asymmetry problem in NH-QM (i.e., $\theta>0$ in our case) we can carry out only forward time propagation calculations [note that $\exp(-iE_\alpha^{QE}t)$ diverges when $t\rightarrow-\infty$ since $\mathrm{Im}\,E_\alpha^{QE}<0$]. Similarly when $\theta<0$ we have to carry out backward time propagation calculations. However, in the right-hand side of Eq. (16) we assume that it is possible to carry out forward time propagation while $\theta<0$. Since it is impossible to carry out these calculations Eq. (16) does not hold. We show here that the time asymmetry in NH-QM is the reason for the breaking of the symmetry of the HHG spectra to the inversion of the relative phase, even when it is calculated from a single resonance Floquet state.

The conclusion is that the breaking of the symmetry of the HHG under the transformation $\phi\rightarrow-\phi$ results from the time asymmetry in NH-QM which avoids the equality we obtained in Hermitian QM for a single Floquet state, $d(t,\phi) = [d(t,-\phi)]^*$. From Eq. (16) it is clear that the phase-symmetry HHG would be obtained as $\Gamma_\alpha(\phi)\rightarrow 0$. Consequently we argue that a quantitative measure for the strength of the breaking symmetry parameter is the value of $\Gamma_\alpha(\phi)$. As the resonance width becomes smaller, $\Delta_n(\phi)$ (see the definition given above) is reduced. Our numerical calculations support this conclusion. See, for example, Fig. 6 where we show the correspondence between $\Delta_3(\phi)$ and $\Gamma_\alpha(\phi)$ as calculated from the same Floquet state (denoted here by α).

III. EFFECT OF THE RELATIVE PHASE ON THE HHG SPECTRA IN CONVENTIONAL QM CALCULATIONS

In this section we address the following question. How come that when we solve the TDSE the HHG spectrum is phase asymmetric, while it is phase symmetric when the propagated wave packet is described as a linear combination of finite number of Floquet states?

To this end, we shall first recall a few notions of the time-independent scattering theory for time-dependent Hamiltonians. This can be conveniently done using the (t,t') formalism [14]. Within this method, the solution of the TDSE is given by

$$\Psi(x,t) = \psi(x,t'=t,t), \tag{17}$$

where the function $\psi(x,t'=t,t)$ is obtained by the action of the time evolution operator on the initial condition:

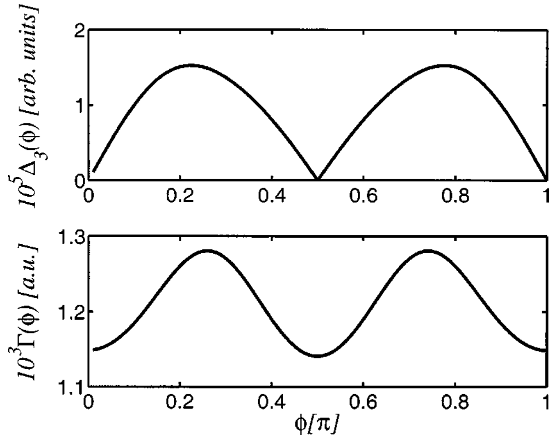

FIG. 6. The correspondence between the HHG asymmetry strength parameter $\Delta_n(\phi)=|I_n(\phi)-I_n(-\phi)|$ and the longest-living resonance Floquet width $\Gamma(\phi)$ as obtained from complex scaled NH-QM calculations.

$$\psi(x,t',t) = e^{-i\mathcal{H}_f(x,t')t}\Psi(x,0). \tag{18}$$

The fact that (t,t') formalism provides an analytical expression for the time evolution operator when the Hamiltonian is time dependent enabled us to develop the time-independent scattering theory for time-dependent Hamiltonians [14,16]. The Fourier components of $\psi(x,t',t)$ are given by

$$\psi_E(x,t') = iG_E(x,t')\Psi(x,0), \tag{19}$$

where the Green operator in the extended (x,t') space is defined as

$$G_E(x,t') = \lim_{\epsilon\rightarrow 0^+}\{E-[\mathcal{H}_f(x,t')-i\epsilon]\}^{-1}. \tag{20}$$

In the spectral representation of the Green operator,

$$G_E(x,t') = \lim_{\epsilon\rightarrow 0^+}\int dE'\frac{|\Phi_{E'}\rangle\langle\Phi_{E'}|}{E-E'+i\epsilon}, \tag{21}$$

where $\Phi_{E'}(x,t')$ are the eigenfunctions of the Floquet operator. The time-dependent solution of the TDSE, expanded in the Floquet basis set, is given by

$$\Psi(x,t) = i\int_{-\infty}^{+\infty} d\,Ee^{-iEt}\lim_{\epsilon\rightarrow 0^+}\int_{-\infty}^{+\infty} dE' \times\frac{\langle\Phi_{E'}|\Psi(t=0)\rangle}{E-E'+i\epsilon}\Phi_{E'}(x,t). \tag{22}$$

Due to the use of finite number of basis functions or finite number of grid points in the numerical calculations the QE spectrum of $\mathcal{H}_f$ is discrete. This approach which yields a discrete spectrum is known as the "box-quantization" approach. Therefore, when the box quantization is applied, the integrals over E and E' in Eq. (22) are replaced by summations over E_α and $E_{\alpha'}$. The corresponding discrete time-periodic Floquet states are given by,

$$\Phi_\alpha(x,t) = \sum_n e^{i\omega n t}\varphi_{\alpha,n}(x). \tag{23}$$

The expression for the HHG spectra, where Ω is the frequency of the emitted radiation, is obtained by taking the Fourier transform of the time-dependent expectation value of the acceleration [i.e., time second-order derivative of the dipole moment, $d(t)=\langle\Psi(t)|x|\Psi(t)\rangle$],

$$I(\Omega) \propto \left| \lim_{\epsilon\to 0^+} \sum_{\alpha,\alpha'} C^*_{\alpha'} C_\alpha \sum_{n,n'} A_{\alpha',n',\alpha,n}(\Omega,\omega)\langle\varphi_{\alpha',n'}|x|\varphi_{\alpha,n}\rangle \right|^2, \tag{24}$$

where

$$C_\alpha = \sum_{n=-\infty}^{+\infty} \langle\varphi_{\alpha,n}|\Psi(t=0)\rangle \tag{25}$$

and

$$A_{\alpha',n',\alpha,n}(\Omega,\omega) = \frac{[(E_\alpha - E_{\alpha'}) + \omega(n-n')]^2}{\Omega - [(E_\alpha - E_{\alpha'}) + \omega(n-n')] + i\epsilon}. \tag{26}$$

From Eq. (26) it is clear that the peaks in the HHG spectra should be obtained when $\Omega=(E_\alpha - E_{\alpha'})+\omega(n-n')$. Therefore, as mentioned in Sec. I, the only terms that contribute to the HHG (i.e., $\Omega=m\omega$ where m gets integer values only) spectra are obtained when $\alpha'=\alpha$ and the ratio Ω/ω gets integer values only. Moreover, as explained in the introduction part of this paper, due to the fact that only the diagonal terms survived the Fourier transform of $\ddot{d}(t)$, the HHG spectrum is invariant under the $\phi\to-\phi$ transformation. However, this is not the case when the spectrum is continuous. What wrong have we done by using the box-quantization condition in the conventional QM? The answer is that by carrying out the integration over time from 0 to ∞ when the expression for the HHG has been derived, we explored the artificial revivals which result from the use of the box-quantization condition. To avoid the effect of the artificial revivals, ϵ in Eq. (26) should be larger than a given nonzero value (we will discuss this point in detail later) and therefore the nondiagonal terms in Eq. (24) contribute to the calculations of the HHG spectra. These nondiagonal terms are associated with quasienergy Floquet states that are nondegenerate eigenfunctions of the dynamical symmetry operator. This explains the breaking of the symmetry properties of the HHG spectra obtained when only the diagonal terms were involved in the calculations. The phase asymmetry of the HHG spectra results from the interference between nondegenerate eigenfunctions of the dynamical symmetry operator.

Let us explain why when the wave packet is expanded in terms of finite number of Floquet states we cannot take the limit of ϵ to zero. First we wish to consider a simple case where the initial state is a linear combination of two Floquet states, $\Psi(x,t=0,\phi)=[\Phi_\alpha(x,0,\phi)+\Phi_{\alpha'}(x,0,\phi)]/\sqrt{2}$. The solution of the TDSE at $t=nT$ is given by $\Psi(x,t=nT,\phi)=\exp[-iE_\alpha(\phi)nT](\Phi_\alpha(x,0,\phi)+(\exp[-i\Delta E(\phi)T]]^n\Phi(x,0,\phi))/\sqrt{2}$ where $\Delta E(\phi)=E_{\alpha'}(\phi)-E_\alpha(\phi)$. It is clear that up to a phase factor, the solution oscillates in time with the period of $\Delta E/(2\pi)$. In our case the initial field-free state populates finite number of Floquet states only due to the use of box-quantization condition. These oscillations (and the related wave-packet revivals) are artificial and cancel the interference among Floquet states which have different dynamical symmetry properties in Eq. (24) when $\epsilon=0$. There are several ways to avoid these artificial revivals. One way is to carry out the Fourier transform from $t=0$ to $t=t_f<2\pi\Delta E$ rather than to $t=\infty$. Another approach is not to take the limit of $\epsilon\to0$ in Eq. (24) but substitute in that equation $\epsilon=\Delta E$ where ΔE is the smallest energy gap between two states that are populated by the initial field-free state. Here ΔE serves as the width of the discrete quasienergies. In this way we apply the NH-QM approach in the most simple way in order to avoid the artificial revivals which are introduced by the numerical calculations where box-quantization condition is imposed. In Ref. [19] a simple two-level model to illustrate the transition from beat to relaxation has been introduced. It has been shown that when the width (i.e., ϵ in our case) is equal to or greater than the energy gap between the two states, rather than having a damped oscillatory behavior of the survival probability it decays exponentially in time with a rate constant which is linearly proportional to $(\Delta E)^2/\epsilon$ when $\epsilon\geqslant\Delta E$. Here, ϵ has the effect of preventing the artificial revivals and in the two-level model prevents the relaxation of the initial state. This approach of substituting $\epsilon=\Delta E$ into the cross-section expressions has been taken, for example, in the calculations of the crosssection for interatomic Coulombic decay in van der Waals clusters [20]. Another approach which is very close in its spirit to the last one is the approach where a complex absorbing potential is added to the Hamiltonian. Again by doing it the Hamiltonian becomes non-Hermitian. This approach has been taken extensively in different fields of physics and chemistry for many decades. However, we want in this section to calculate HHG spectra, while avoiding the artificial revivals, within the framework of the conventional QM. Therefore, in our time propagation calculations the number of grid points are increased (and thereby the energy splitting due to the use of box-quantization condition is reduced) as time passes to avoid the situation where $\Psi(x,t,\phi)$ gets nonzero value at the edge of the grid. Obviously, in our numerical calculations the propagation of the initial state has been carried out not until $t=\infty$ but until $t=t_f$. The final time of the propagation was taken as the time where the dipole converges to one of the free particle in the field [21].

IV. CONCLUSIONS

Recent experiment and theoretical studies have shown that the HHG spectra of atoms in bichromatic field are affected by the relative phase between the two-color laser beams. The question we addressed here was whether the effect of the sign of the relative phase can be obtained when the lasers are adiabatically switched on and the photoinduced dynamics is solely controlled by a single Floquet state. Dynamical symmetry analysis of the problem shows that the HHG spectrum is invariant under the transformation of phase, $\phi \to \phi + \pi$, but however it *changes under the transformation* $\phi \to -\phi$. We prove that within the framework of the conventional Hermitian QM, the effect of the transformation $\phi \to -\phi$ on the HHG spectra is not obtained when the initial wave packet populates a finite number of Floquet states (also when a single Floquet state is populated). The puzzle we solved is associated with the two quasicontradictive results: (a) the HHG spectrum calculated by a single Floquet state (solving the time-dependent Schrodinger equation with Hermitian Hamiltonian) is invariant under the phase inversion transformation (unlike the result obtained from wave-packet propagation calculations); (b) upon complex scaling, when the Hamiltonian becomes non-Hermitian, the HHG spectrum *changes* under the phase inversion transformation as seen even from the calculations of a single complex scaled Floquet state.

We show that the strength of the effect of the $\phi \to -\phi$ transformation on the HHG spectrum is increased as the photoinduced ionization rate of decay is increased (for example, by varying the relative phase ϕ). The photoinduced ionization rates are directly obtained from the non-Hermitian QM calculations of the complex quasienergies of the complex scaled Floquet operator, and indirectly from motion of wave-packet calculations within the framework of the conventional (Hermitian) QM calculations.

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