

Near-field measurements of the even-order harmonics undetectable in far-field measurements

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Even-order harmonics (EOHs) are generated for oriented molecules and for atoms under specific conditions. Here, we focus on the most common situations where EOHs are not observed in far-field measurements. We propose an experiment to show that the EOHs are produced close to the nucleus but are not detected in the far-field measurements due to destructive interferences of the propagated EOHs of the emitted radiation. However, Rydberg gas atoms (e.g., rubidium), which are out of the focus of the laser beam, are expected to be ionized due to their weak interaction with a specific EOH of helium, which is not observed by the far-field detector. The ionization energy of the Rydberg gas atoms should be in resonance with the single-photon energy of a specific EOH.

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I. INTRODUCTION

Experimental measurements of the harmonic generation spectra (HGS) of atoms in strong linear polarized light consist of odd-order harmonics only: see, for example, Refs. [1–6] and references therein. There are situations where even-order harmonics are also produced, even when rare-gas atoms interact with continuous-wave (CW) linear polarized light. Another possibility to produce even-order harmonics is when rare-gas atoms interact with bichromatic lasers. The even and odd harmonics are produced when the fundamental laser frequency couples field-free states of different symmetries; see, for example, Refs. [7,8]. We might note in passing that even-order harmonics are generated for oriented (but not for aligned) molecules (see, for example, a theoretical explanation in Ref. [9] and experimental results presented in Ref. [10]). However, this situation is not considered here. Here, we focus on situations where even-order harmonics are *not* observed and only odd-order harmonics are detected.

The reason for the fact that only odd-order harmonics are observed when atoms interact with linearly polarized laser fields when the duration of the laser pulse is sufficiently long is well understood [11]. In these cases, besides the generation of high-frequency radiation, the atoms in the strong laser fields are ionized. Consequently, the single Floquet quasienergy (QE) solution that dictates the photoinduced dynamics is a metastable state. This metastable Floquet state is a complex pole of the scattering matrix, which can be computed when outgoing boundary conditions are imposed on the eigenfunctions of the Floquet operator [12]. It has already been shown that the HGS as observed in experiments can be obtained from the calculations of a single-resonance metastable QE-Floquet solution [13]. In Ref. [13], the numerically exact single-resonance QE-Floquet state for a helium atom in a strong linear polarized laser field was calculated by using a uniform complex scaling transformation. The uniform complex scaling transformation of the electronic Hamiltonian enables the calculation of QE-Floquet states, using computational methods that were originally developed for the calculation of bound QE-Floquet states.

II. CALCULATIONS OF THE LOCAL AMPLITUDE OF THE EMITTED RADIATION

To calculate the amplitude of the emitted radiation with the frequency $\Omega > \omega$ (either by far- or near-field measurement), we need to define the time-dependent local operator $A(x, \Omega)$, which is given by

$$A_x(x, y, z, \Omega, t) \equiv e^{i\Omega t} a_x(x, y, z), \quad (1)$$

where the electron acceleration along the polarization direction x is defined as usual as

$$a_x(x, y, z) = -\frac{1}{m_e} \frac{dV}{dx}, \quad (2)$$

where m_e is the mass of the electron and $V(x, y, z)$ stands for the three-dimensional (3D) effective field-free electronic potential. The complex amplitude of the radiation measured by far-field or near-field detectors is given by

$$\begin{aligned} \mathcal{E}(\Omega, F(\sigma)) &= \int_{-\infty}^{+\infty} dx dy dz \frac{1}{T} \\ &\times \int_0^T dt A_x(x, y, z, \Omega, t) \rho(x, y, z, t; F), \end{aligned} \quad (3)$$

where

$$T = \frac{2\pi}{\omega} \quad (4)$$

and the parameter $1 \leq F \leq 0$ is the far-to-near measurement parameter, and gets a continuous value from 1 (measurement of the far-field radiation, which consists of odd-order harmonics) to 0 (near-field measurements of even- and odd-order harmonics). More specifically,

$$F(\sigma) \equiv 1 - \langle \Psi(t) | \hat{O}(\sigma) | \Psi(t) \rangle, \quad (5)$$

where σ is the controlled parameter, $0 < |\sigma| \leq \infty$, and \hat{O} is defined as

$$\hat{O}(F(\sigma)) \equiv \mathcal{W}(x, \sigma). \quad (6)$$

The function $\mathcal{W}(x, \sigma)$ gets values close to unity when $F(\sigma) \simeq 1$ and only far-field measurements of the HGS are taken. The near-field measurements of the HGS are taken when $F(\sigma) \ll 1$ and $\mathcal{W}(x, \sigma)$ is not a uniform function of x . $x = x_0$ stands for the distance of the Rydberg atom from the rare-gas atoms that are embedded in the focus of the laser beam. x_0 must get a

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sufficiently large value to ensure the negligible effect of the Rydberg atom (e.g., rubidium) on the HGS.

The time-dependent density functions in Eq. (3) are given by

$$\rho(x, y, z, t; F(\sigma)) = [\hat{O}(\sigma)\Psi(x, y, z, -t)]\Psi(x, y, z, t). \quad (7)$$

Then, for bound quasienergy (Floquet) solutions of the time-dependent Schrödinger equation (TDSE), $\Psi(x, y, z, -t) = \Psi^*(x, y, z, t)$, and in far-field measurements, $\rho(x, y, z, t; F(\sigma) = 1) = |\Psi(x, y, z, t)|^2$. When the quasienergy solutions of the TDSE are metastable states, then $\Psi(x, y, z, -t) \neq \Psi^*(x, y, z, t)$ (see the c product as defined in the textbook on non-Hermitian quantum mechanics [12]). The intensity of the emitted radiation with frequency Ω is given by

$$I_0(\Omega, F(\sigma)) = |\mathcal{E}(\Omega, F(\sigma))|^2. \quad (8)$$

The far-field detector shows that $I_0(\Omega, F(\sigma) \rightarrow 1)$ gets large values when $\Omega/\omega = 3, 5, 7, \dots, 2n + 1, \dots$. The near-field detector ($F \simeq 0$) is located out of the focus of the laser that interacts with the noble gas atom that generates the high harmonics. The Rydberg state is a diffuse function which features a tail that reaches the nucleus of the rare-gas atom that emits the high-frequency radiation. The effect of this diffuse function on the symmetric properties of the Floquet Hamiltonian that describes the dressed rare-gas atom is negligible. Therefore, in the far-field measurements, only odd-order harmonics would be observed as before, even when the near-field measurements are taken. It is important to emphasize that the Rydberg atom will only be ionized on resonance conditions, that is, when $I \cdot E_{\text{Rydberg}}/\hbar = \Omega = \omega n_0$, where $I \cdot E_{\text{Rydberg}}$ is the ionization energy of the Rydberg atom and $\Omega = \omega n_0$ is a specific emitted high-frequency radiation that satisfies the resonance condition which is stated above. The experiment should be designed such that n_0 gets a specific even value. The fact that the Rydberg atom is ionized is a proof that even-order harmonics, which are not observed by the far-field detector, are generated with a quite large local intensity.

By using Eq. (7), the following is obtained from Eq. (3):

$$\mathcal{E}(\Omega, F(\sigma)) = \frac{1}{T} \int_0^T dt \langle \Psi(t) | \mathcal{W}(x, \sigma) A_x(x, y, z, \Omega, t) | \Psi(t) \rangle. \quad (9)$$

By changing the order of the integration over time and space, one gets that

$$\mathcal{E}(\Omega, F(\sigma)) = \int_{-\infty}^{+\infty} dz \mathcal{W}(x, \sigma) A_{\text{local}}(x, \Omega), \quad (10)$$

where the local amplitude of the radiation generated by the atom that interacts with the linear polarized laser field, as a function of the polarization direction, is given by

$$A_{\text{local}}(x, \Omega) = \frac{1}{T} \int_0^T dt \int_{-\infty}^{+\infty} dy dz A_x(x, y, z, \Omega, t) \times \Psi(x, y, z, -t) \Psi(x, y, z, t). \quad (11)$$

For bound systems, $\Psi(x, y, z, -t) \Psi(x, y, z, t) = |\Psi(x, y, z, t)|^2$. Here, we take the photoinduced ionization

phenomenon into consideration and therefore prefer to use a more general definition for the inner product (see Ref. [12]). When the duration of the laser pulse is sufficiently long and supports more than 5–10 optical cycles, the dynamics of the photoinduced system can be described by the Floquet theory [11]. Namely, the field-free bound ground state becomes a quasienergy Floquet state, which is the eigenfunction of the Floquet operator, which has the largest overlap with the field-free bound ground state. Consequently,

$$\Psi(\mathbf{r}, t) \propto \Phi(\mathbf{r}, t), \quad \Phi(\mathbf{r}, t) = \sum_{n=0, \pm 1, \pm 2, \dots} \varphi_n(\mathbf{r}) e^{i\omega n t}, \quad (12)$$

where the overlap of $\varphi_0(x)$ with the field-free ground state is large (usually > 0.9). For atoms in a linear polarized laser field, $\Phi(x, y, z, t) = \pm \Phi(-x, y, z, t + T/2)$, and therefore, $\varphi_n(x, y, z) = \pm (-1)^n \varphi_n(-x, y, z)$ [14]. Consequently,

$$\begin{aligned} A_{\text{local}}(x, \Omega) &= \frac{1}{T} \int_0^T dt \langle \Phi(\mathbf{r}, t) | \hat{A}_{x, \Omega}(t) | \Phi(\mathbf{r}, t) \rangle_{y, z} \\ &= \left\langle \left(-\frac{1}{m_e} \frac{dV}{dx} \right) \left| \frac{1}{T} \sum_{n, n'} \int_0^T dt e^{i((n-n')\omega + \Omega t)} \varphi_n \varphi_{n'} \right\rangle_{y, z} \right. \\ &= \left\langle \left(-\frac{1}{m_e} \frac{dV}{dx} \right) \left| \delta_{\Omega, N\omega} \sum_n \varphi_n(\mathbf{r}) \varphi_{n'+N}(\mathbf{r}) \right\rangle_{y, z} \right. \\ &\equiv \left\langle \left(-\frac{1}{m_e} \frac{dV}{dx} \right) \left| \delta_{\Omega, N\omega} \chi(x, y, z; N) \right\rangle_{y, z} \right. . \end{aligned} \quad (13)$$

The near-field emitted radiation (so-called local field) consists of N -order harmonics, where N is an integer number (even or odd). Therefore, the emitted local (near-field) radiation consists of odd- and even-order harmonics, $N = 2, 3, 4, 5, \dots$. Due to the symmetric properties of the quasienergy (Floquet) solutions mentioned above, when N is an even number, then $\varphi_n(x)$ and $\varphi_{n+N}(x)$ have the same symmetric properties (both are either even or odd functions). Therefore, $\chi(x, y, z; N) = \sum_n \varphi_n(x, y, z) \varphi_{n'+N}(x, y, z)$ is an even function of x when $N = 2, 4, 6, \dots$. When $N = 3, 5, 7, \dots$ (odd numbers), then $\chi(x, y, z; N)$ is an odd function and $\chi(-x, y, z; N) = -\chi(x, y, z; N)$. Since the atomic field-free potential $V(x)$ is an even function, and dV/dx is an odd function, it is clear that the integral of $A_{\text{local}}(x, \Omega = N\omega) \propto \langle (dV/dx) \chi(x, y, z; N) \rangle_{y, z}$ over x is zero when N gets even values. This is the reason that only odd-order harmonics are emitted in far-field measurements while the local amplitude of the even-order harmonics is not equal to zero. When the pointer of the detector is a delta function, as first proposed by Berry [15], then even-order harmonics are obtained in weak measurements whenever a local measurement of the emitted radiation is taken. Figures 1–3 show the local amplitude of even- and odd-order harmonics that were obtained for a simple model of Xe in a linear polarized laser field. The results presented in Figs. 1–3 show that $A_{\text{local}}(x, \Omega = N\omega) \propto \langle (dV/dx) \chi(x, y, z; N) \rangle_{y, z}$ is an odd function of x when N gets even values, and it is an even function when N gets odd values. Consequently, in the far-field measurements where the local (near-field) amplitudes are integrated over the entire space (i.e.,

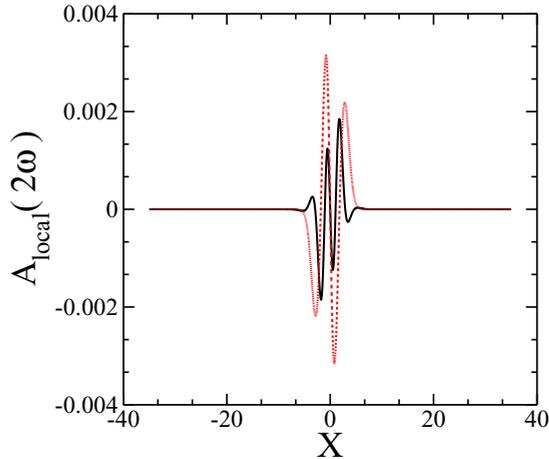


FIG. 1. The complex amplitude of the emitted radiation (black color for real values and red color for the imaginary values) with a photon energy $\hbar\Omega$, where $\Omega = 2\omega$ as a function of the polarization coordinate (a.u.). The laser frequency is $\omega = 0.07$ au (1.9 eV) and the maximum field amplitude $\epsilon_0 = 0.0735$ a.u. ($I_0 = |\epsilon_0|^2 = 1.9 \times 10^{14}$ W/cm²). As one can see from this plot, the local amplitude of the second-order harmonic is an odd function of x (the polarization axis). Therefore, in a far-field measurement where the detector measures the integral over all emitted local radiation, *no* second-order harmonics are emitted.

the polarization direction x), only odd-order harmonics will be detected (see Ref. [14] for the derivation of dynamical selection rules for HHG). The numerical results provide an illustrative example of the analytical properties of the local amplitude of the emitted radiation, as explained above. The model potential of Ar in the linear polarized light and the numerical methods used for calculations of the Floquet solution are given in Ref. [11]. Outgoing boundary conditions were introduced by applying the complex scaling transformation, which enables

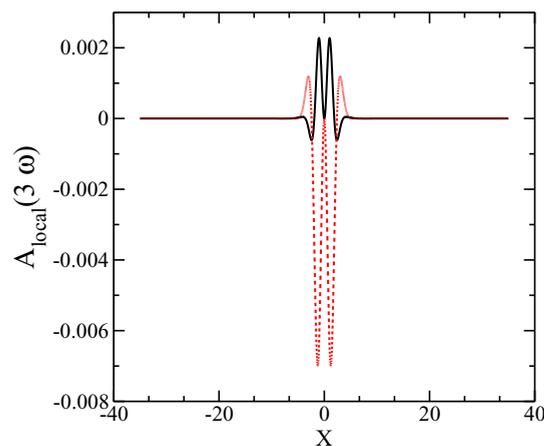


FIG. 2. The amplitude of the emitted radiation with a photon energy $\hbar\Omega$, where $\Omega = 3\omega$, as a function of the polarization coordinate (a.u.). The laser frequency and intensity are as given in the caption of Fig. 1. Since the local amplitude of the third-order harmonic is an even function and therefore in a far-field measurement where the detector measures the integral over all emitted local radiation, the third-order harmonics are emitted.

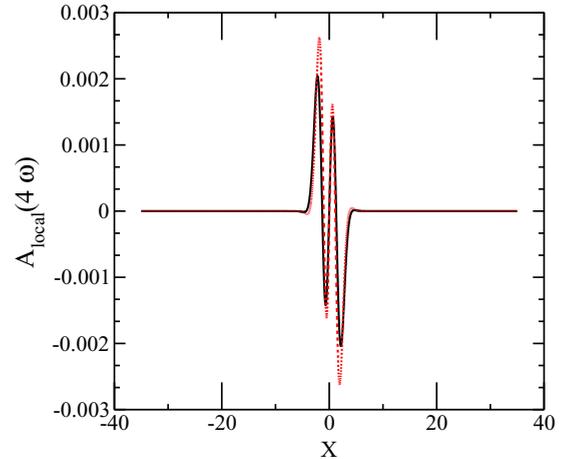


FIG. 3. The amplitude of the emitted radiation with a photon energy $\hbar\Omega$, where $\Omega = 4\omega$, as a function of the polarization coordinate (a.u.). The laser frequency and intensity are as given in the caption of Fig. 1.

the calculations of resonance metastable quasienergy (Floquet) states by using algorithm methods which were originally developed for the calculations of square integrable bound states (see Ref. [12]).

III. MEASUREMENTS OF THE RADIATION EMITTED FROM A SUBATOMIC SPATIAL REGION

The problem is that the detector should be able to measure the radiation emitted from a subatomic spatial region. For illustration reasons only, the detector which measures the near-field-emitted radiation is a super-Gaussian which enables us to control the detection of the high-harmonic-generation spectra from far-field detection to near-field detection by varying the parameter σ ,

$$\mathcal{W}(x, \sigma) = \exp \left[- \left(\frac{x - x_0}{\sigma} \right)^8 \right]. \quad (14)$$

The transition from far-field measurement to near-field measurements is given by the parameter P_F , which is defined by

$$P_F(\sigma) = 1 - \lim_{L \rightarrow \infty} \frac{1}{L} \int_{-L/2}^{+L/2} \mathcal{W}(x, \sigma) dx, \quad (15)$$

such that

$$0 \leq P_F \leq 1, \quad (16)$$

as σ is varied from 0 to ∞ . L stands for the size of the box where the entire system (i.e., rare-gas atom and Rydberg atom) is localized. Note that the parameter $F = 1$ implies that no even-order harmonics are measured and, therefore, $P_{F=1} = 0$.

In our illustrative numerical calculations for the high-frequency radiation which is emitted from Ar that interacts with a linear polarized laser field, we chose x_0 to be “outside” of the atom in the sense that $A_{\text{local}}(x_0, N) = 0$ for any value of N . Note that $\Omega = N\omega$ is the frequency of the emitted high-frequency radiation, where $N = 2, 3, 4, \dots$. Since in our illustrative numerical example $A_{\text{local}}(x, N) \neq 0$ when $|x| \leq L/2 = 6$ is the active atomic region where the even- and

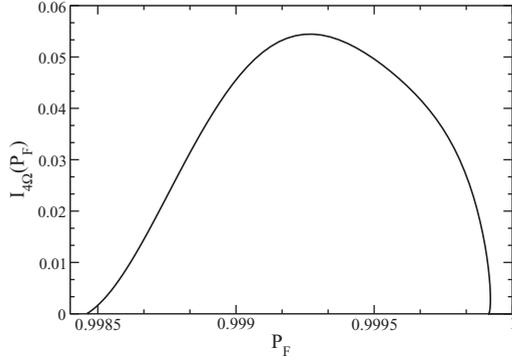


FIG. 4. The intensity, as defined in Eq. (17), of the emitted fourth-order harmonics from Xe, as a function of the far-to-near transition strength parameter. By definition, F is a transition from a far-field measurement ($P_F = 0$) to a near-field measurement ($P_F \cong 1$). The laser frequency and intensity are as given in the caption of Fig. 1.

odd-order harmonics are emitted, we define $P_F(\sigma)$ in our numerical calculations as in Eq. (15), when $L = 12$ a.u. and $x_0 = 10$, where $0 < \sigma \leq 20$.

Thus, $P_F \rightarrow 1$ as $\sigma \rightarrow 0$ and $P_F \rightarrow 0$ as $\sigma \rightarrow 20$, including a continuous change from far-field measurements of the HHG spectra ($\lambda = 1$ for $\sigma \rightarrow 0$) to near-field measurements ($\lambda = 0$ for $\sigma \rightarrow 20$) in our simulations.

The amplitude of the measured N -order harmonics as a function of P_F is obtained from Eq. (10) by substituting $\Omega = N\omega$,

$$A_N(P_F(\sigma)) = \mathcal{E}(\Omega = N\omega, F(\sigma)). \quad (17)$$

Note that due to the fact that the pointer of the detector is located at the region where $A_{\text{local}}(x = x_0, N) = 0$, for any value of N , we will not get any emitted high-frequency radiation for the weakest possible perturbation which breaks the dynamical symmetry of the Floquet operator which is associated with $P_F = 1$. Therefore, we expect to observe a nonmonotonic behavior of the intensity of the emitted even-order harmonics as P_F varies from far- to near-field measurement. The results presented in Fig. 4 confirm our analysis. That is, the even-order harmonics, which are not allowed by the dynamical symmetry rule [14], are detected in a measurement where the postselected state is almost orthogonal to the preselected state. The preselected state is, in our case, a photoionizing resonance eigenstate of the Floquet operator. The postselected state is solely dependent on the location of the detector. It should be emphasized here that x_0 is measured from the origin, where the nucleus of the atom, which serves as a detector (see below), is located.

The calculations we perform will be chronologically presented to show that the pointer does not significantly break the dynamical selection rules which result in far-field measurements of odd-order harmonics only. The pointer couples two resonance Floquet solutions which have different dynamical symmetry. It implies that the solution of the TDSE is now given by

$$\begin{aligned} \Psi_{WP}(\mathbf{r}, t) = & e^{-iE^{\text{res1}}t/\hbar} \Phi_{\text{res1}}(\mathbf{r}, t) \\ & + C_{\text{pointer}} e^{-iE^{\text{res2}}t/\hbar} \Phi_{\text{res2}}(\mathbf{r}, t), \end{aligned} \quad (18)$$

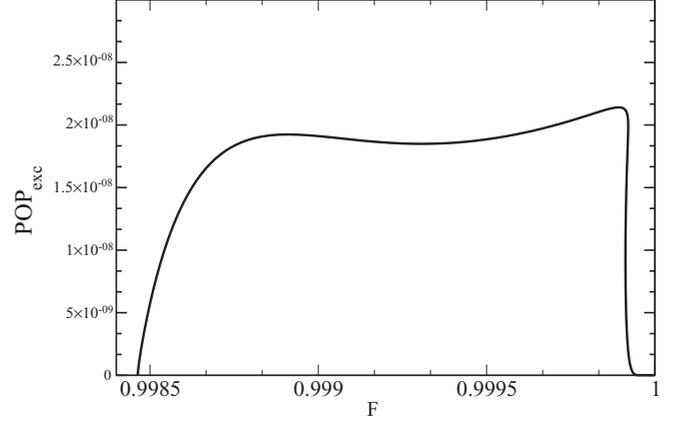


FIG. 5. A low probability to populate the excited Floquet state is required to guarantee the suppression of even-order harmonics in the far-field measurements while the Rydberg atom is ionized due to the local generation of even-order harmonics. By definition, F is a transition from a far-field measurement ($P_F = 0$) to a near-field measurement ($P_F \cong 1$). The laser frequency and intensity are as given in the caption of Fig. 1.

where E^{res1} and $\Phi_{\text{res1}}(\mathbf{r}, t)$ are, correspondingly, the resonance eigenvalue and eigenfunction of the Floquet operator that are associated with the preselected state as defined in Eq. (12), and the other Floquet solution is the solution that is most strongly coupled to the preselected state by the “pointer.” To guarantee that the pointer does not break down the dynamical symmetry of the preselected state, we must confirm that $|C_{\text{pointer}}|^2 \approx 0$. On the basis of standard first-order perturbation theory, C_{pointer} is given by

$$C_{\text{pointer}} = \frac{\langle \langle \Phi_{\text{res1}}(\mathbf{r}, t) | \mathcal{W}(x, \sigma) | \Phi_{\text{res2}}(\mathbf{r}, t) \rangle_{x,y,z} \rangle_t}{E^{\text{res1}} - E^{\text{res2}}}. \quad (19)$$

In Fig. 5, we show, for our model system, the value of $POP_{\text{exc}} = |C_{\text{pointer}}|^2$, which indicates the probability to populate an excited Floquet state due to the near-field measurement. As one can see from the results presented in this figure, the population of another resonance Floquet solution is negligible when the near-field measurement is taken. This is an important result since the near-field measurement is taken *only* when a single Floquet resonance solution controls the photoinduced dynamics and only odd-order harmonics are observed by the standard far-field measurement. Note that in our calculations, we selected the largest possible coupling parameter C_{pointer} . A low value of the population of another Floquet solution due to the near-field measurement is expected since the nominator in the expression of C_{pointer} gets small values and the denominator gets large values, yielding an extremely small ratio, with a negligible effect on the far-field measurements.

IV. VISUALIZATION OF THE PERFORMANCE OF A NEAR-FIELD MEASUREMENT WHERE THE EVEN-ORDER HARMONICS ARE DETECTED

Let us visualize the performance of a near-field measurement where the even-order harmonics are detected. In the proposed measurement, a comparison between two different types of experiments will be taken. In one type of experiment,

using an absorption or fluorescence imaging technique, the fraction of ionized rubidium will be measured in the absence of Ar atoms. In this type of experiments, Rb atoms are ionized because of multiphoton absorption. However, multiphoton ionization in this case will be small because of the low penetration of the rubidium orbitals (even at $n = 10$ Rydberg state) to the focus of the laser. In the second type of experiments, the number of Rb ions will be measured in the presence of Ar atoms. Therefore, in the second type of experiments, the rubidium atoms in the $n = 10$ Rydberg state will be ionized not only by a sequential absorption of two 6400 nm photons but also by a single (second harmonic) 3200 nm photon. In order to reach conditions in which the multiphoton ionization of rubidium is negligible in comparison to the single-photon absorption, the single-photon energy $\hbar\Omega$, where $\Omega = 2\omega$, must be as close as possible to the ionization energy of the highly excited rubidium atoms ($n = 10$ Rydberg p -type orbital). The main contribution to the signal-to-noise ratio is the fluctuation in the power of the harmonic generation laser.

V. CONCLUDING REMARKS

Breakage of the spherical symmetry ground-state resonance Floquet eigenfunction of a noble gas atom due to

the penetration of the tail rubidium is negligible and, in far-field measurements, only the odd-order harmonics emitted by the rare-gas atoms will be observed. However, due to the penetration of the tail of the Rydberg orbital of rubidium to the trap where the noble gas atoms interact with the strong laser field, we expect to ionize significant parts of the rubidium atoms due to the near field of the emitted second-order harmonics. A detailed description of such an experiment is beyond the scope of this work. We leave to the experimentalists, who might be motivated by this paper, to find other means of carrying out the near-field measurements, while only odd-order harmonics are observed in the far-field measurements.

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