One- and two-photon phase-sensitive coherent control of total ionization yields in the presence of static electric fields

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We experimentally demonstrate one- and two-photon phase sensitive coherent control over the excitation and ionization of Rydberg states of Sr atom in the presence of a static homogeneous electric field. Without the field this excitation scheme can only be employed for the manipulation of angular distributions of photoelectrons and molecular photofragmentation products. Total atomic excitation-ionization yields cannot be modulated because the final states excited by each pathway are orthogonal to each other. When, however, a static electric field is applied, *s*, *p* and *d* character is admixed into the final Rydberg state, which has no definite parity anymore. Hence, the excitation of the target Stark state from the ground state is possible with either two (fundamental laser frequency) photons or one (second harmonic frequency) photon and its population and further ionization can be controlled by varying the relative phase between the two radiation fields. The concept is successfully tested below as well as above the classical saddle point and with either mutually crossed or parallel linear laser beam polarizations (while the second harmonic beam polarization vector is always parallel to the static field direction). We examine the behavior of the obtained photoionization signal modulation depth V as a function of the static field strength F for otherwise identical experimental conditions. The $V(F)$ curve exhibits a maximum (typically $\sim 65\%-85\%$) at a field strength value that is dictated by the interplay between the employed laser power densities, one- and two-photon transition dipole moments, and relative amounts of field-dependent *s*, *p* and *d* character. It is therefore shown that the static field strength may serve as an additional, experimentally adjustable, control parameter in a fashion complementary to both the intensities and relative phase of the two light beams.

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I. INTRODUCTION

Since the advent of coherent light sources, numerous methodologies designed so as to allow efficient control on atomic and molecular processes were intensively investigated. Two major approaches were continuously developed during the last fifteen years or so towards this goal. The first relies on the manipulation of electronic or vibronic wave packets produced and controlled by short laser pulses [1] and the second exploits the interference between alternative transition amplitudes generated by a bichromatic laser field [2]. Restricting the discussion to the latter approach, the most familiar scheme involves a fundamental frequency laser beam and one of its mutually coherent harmonics. Each beam is associated to a single transition amplitude (excitation pathway) and couples the same initial and final states with a different number of photons. Phase sensitive coherent control is then achieved by externally adjusting the relative phase between the two fields [2]. Due to the parity and other electric dipole selection rules, *total* excitation-ionization yields of isolated atomic systems may be controlled by involving only odd [for example, $1\times(3\omega)+3\times\omega$] [3,4] or only even [e.g., $2\times(2\omega)+4\times\omega$] [5] number of photons for each excitation pathway. Evidently, the even number of photons case is by far the most experimentally convenient one since the second harmonic field is easily produced by appropriate frequency doubling crystals, thus avoiding the complications accompanying the generation of odd harmonics, such as the third harmonic, for example. Nevertheless, due to the aforementioned selection rules for isolated atoms, the simplest phase control scheme, utilizing two photons from the fundamental beam and one photon from its second harmonic [i.e., $1\times(2\omega)+2\times\omega$], cannot be applied for controlling total yields. Still, it can be and has been productively applied for manipulating angular distributions of photoelectrons in atomic [6,7] or molecular [8] ionization and photocurrents in semiconductors [9] where, strictly speaking, the two transitions do not lead to the same final state but to a coherent combination of different continua. Ponderomotive potential and other strong laser field effects also allowed the application of the $1\times(2\omega)+2\times\omega$ scheme in above threshold ionization [10]. Finally, it has been particularly fruitful to the study of phase sensitive molecular processes such as photodissociation [11], photofragment angular distributions [12], and forward-backward asymmetries [13].

For extending the application of this simplest and very convenient scheme to the manipulation of total ionization yields (while remaining well within the low intensity, perturbative laser-atom interaction domain) the implication of external static electric fields which mix states of opposite parity was theoretically investigated [14]. Experimentally however, the concept was not implemented until very recently [15]. In that work the Cs $6s\rightarrow 8s$ transition was used and a dc electric field of fairly high strength (~ 4 kV/cm) mixed a small amount of *p* character into the low-excited $8s$ level. As a result, a weak one-photon dipole transition amplitude was induced, simulating feeble electric quadrupole, magnetic dipole, and parity nonconserving interactions. The purpose of that experiment was to propose a methodology for measuring

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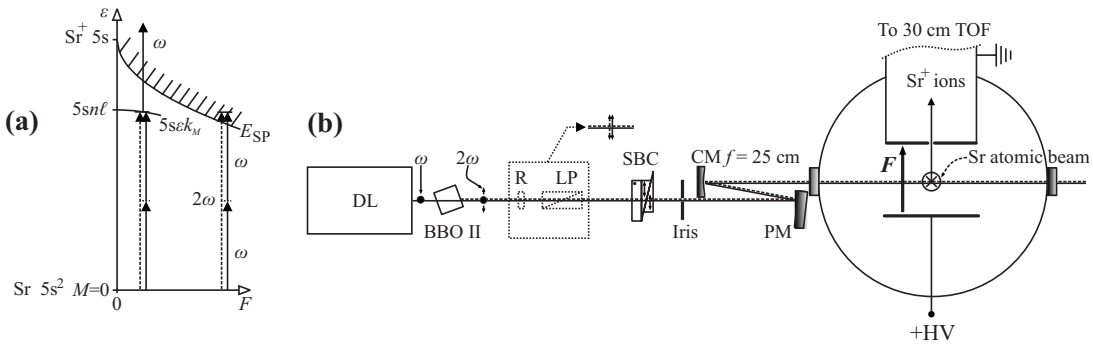

FIG. 1. (a) Excitation schemes employed for phase-sensitive coherent control over the population of Sr Stark $5s\varepsilon k_M$ states for excitation energies ε below as well as above the saddle point $E_{SP}=-2\sqrt{F}$ (in atomic units). For $\varepsilon<E_{SP}$ the 2ω uv beam cannot photoionize the Stark level. Sr^+ ions are produced by the absorption of an extra ω photon. (b) Schematic drawing of the experimental setup. DL: Nd:yttrium aluminum garnet (YAG) pumped dye laser. BBO II: frequency doubling crystal. SBC: Soleil-Babinet compensator for varying the relative phase $\varphi_{2\omega}-2\varphi_{\omega}$ between the doubled and fundamental frequency beams. PM: plane mirror. CM: $f=+25$ cm focal distance concave mirror (the angle of incidence to both mirrors is ~ 3 deg). TOF: Time of flight. The polarization of the 2ω beam is parallel to the direction of the applied static electric field $\vec{\mathbf{F}}$ while the polarization of the fundamental beam is originally perpendicular to it. For making both polarizations parallel to $\vec{\mathbf{F}}$ a quartz polarization rotator (R) and an alfa-BBO Rochon prism linear polarizer (LP) are placed immediately after the doubling crystal.

these interactions with improved signal-to-noise ratio after employing heterodynelike detection techniques. In the present work, we explore further the concept experimentally from another perspective and by using an excitation scheme which differs from that proposed in the theoretical analysis of [14]. Our intention is to introduce the static electric field strength as an additional adjustable parameter that may dictate the efficiency of phase sensitive coherent control over total excitation-ionization yields in a manner complementary to the relative phase and power densities of the radiation fields. That is successfully demonstrated using Stark final states corresponding to $5snp\ ^1P_1$, $5sns\ ^1S_0$, and $5snd\ ^1D_2$ zero-field Rydberg levels of Sr with $n\sim 20$ as well as quasidiscrete Stark states in the vicinity and above the classical saddle-point energy. It is thus shown that the concept can also be applied to a two-valence electron atom and it is operational from the ℓ-mixing region up to the above-the-barrier one. Moreover, the results of this study may be used as a guideline for applications to systems which, in general, do not possess inversion symmetry.

II. PHASE SENSITIVE EXCITATION SCHEME

Before going into the experimental details it is useful to present the phase-sensitive excitation scheme. In the presence of a static electric field $\vec{F}$ of strength F the parity is no longer a good quantum number. In fact, for a multielectron atom the only reliably good quantum number is M, the projection of total angular momentum on the dc field direction, which we define as the quantization axis. Let us use the notation $5s\varepsilon k_M$ for characterizing a particular Sr Stark state of given M. The Rydberg electron is assumed to lie at energy ε, within the ℓ-mixing or n-mixing region, or even above the classical saddle-point energy $E_{SP}=-2\sqrt{F}$ (in atomic units) [16] [see Fig. 1(a)]. The Rydberg electron's wave function $|\varepsilon k_M\rangle$ may, in a simplistic manner, be represented as a sum of different ℓ-character terms whose amplitudes are field dependent, i.e.,

$$|\varepsilon k_M\rangle = c_{s,M}(F)|\varepsilon s\rangle + c_{p,M}(F)|\varepsilon p\rangle + c_{d,M}(F)|\varepsilon d\rangle + \cdots . \tag{1}$$

In Eq. (1) $|\varepsilon s\rangle$, $|\varepsilon p\rangle$, and $|\varepsilon d\rangle$ denote collectively all the zero-field s, p, and d states contributing to $|\varepsilon k_M\rangle$, but for small enough field strengths only the immediate neighbor states around the level of interest possess significant amplitudes $c_{\ell,M}$. The dipole one-photon $5s^2(M=0)\rightarrow 5s\varepsilon k_M$ transitions out of Sr ground state populate the p component of $|\varepsilon k_M\rangle$ while the dipole two-photon transitions excite the s and d components. For achieving phase sensitive coherent control over the population transferred to the $5s\varepsilon k_M$ state, the one-photon and two-photon transitions should necessarily excite the same M value. This is accomplished by adjusting the linear polarization of each light beam with respect to direction of the static electric field. A light beam whose linear polarization is parallel to the dc field (π polarization) induces $\Delta M=0$ dipole transitions so only $M=0$ final states can be reached from the $M=0$ ground state and irrespectively of the number of absorbed photons. If, on the other hand, the beam is linearly polarized perpendicularly to the dc field (σ polarization) the corresponding dipole selection rule reads $\Delta M=\pm 1$/per photon [17]. Consequently, a two-photon transition leads to $|M|=0,2$ final states in our case. Therefore, as long as the laser field inducing the one-photon transition is π polarized, $M=0$ final states are commonly excited by the two light beams with either mutually parallel or perpendicular polarizations. Using Eq. (1) the rate of $5s^2$ $(M=0)$ $\rightarrow 5s\varepsilon k_{M=0}$ excitation induced by a bichromatic laser field $E_{tot}=E_{\omega}\exp(\mathbf{i}\varphi_{\omega})+E_{2\omega}\exp(\mathbf{i}\varphi_{2\omega})$ is written as

$$W_{\mathrm{exc},M=0} \propto |c_p\mu_{sp}^{(1)}|^2 I_{2\omega} + |c_s\mu_{ss}^{(2)} + c_d\mu_{sd}^{(2)}|^2 I_\omega^2 + 2|c_p\mu_{sp}^{(1)}(c_s\mu_{ss}^{(2)} + c_d\mu_{sd}^{(2)})|\sqrt{I_\omega^2 I_{2\omega}}\cos(\varphi_{2\omega} - 2\varphi_\omega), \quad (2)$$

where $I_{2\omega}$ and I_ω are the intensities of the 2ω and ω beams, $\varphi_{2\omega}$ and φ_ω their respective phases, and $\mu_{sp}^{(1)}$ and $\mu_{ss,d}^{(2)}$ one- and two-photon single-electron transition dipole moments, respectively. The efficiency of a phase sensitive control scheme is customarily evaluated on the basis of the modulation depth

$$V \equiv \frac{S_{\max} - S_{\min}}{\frac{1}{2}(S_{\max} + S_{\min})} \quad (3)$$

with $S_{\max}$ $[\cos(\varphi_{2\omega}-2\varphi_\omega)=1]$ and $S_{\min}$ $[\cos(\varphi_{2\omega}-2\varphi_\omega)=-1]$ the maximum and minimum signal, respectively. From Eq. (2) it is obvious that the efficiency achievable via the $M=0$ pathway strongly depends on the field strength through $c_{\ell,M}(F)$, but this dependence is in general difficult to predict as it relies on the specificities of the zero-dc field spectrum of a particular atom.

One may argue that with crossed polarizations a parasitic $|M|=2$ background would reduce the observed modulation depth. Note, however, that a π-polarized one-photon field may also induce electric quadrupole $\Delta M = \pm 2$ transitions to the d component of $|\varepsilon k_M\rangle$ [18]. The importance of this second $W_{\mathrm{exc},|M|=2}$ term, representing an additional phase control contribution, depends, of course, on the magnitude of the relevant quadrupole excitation rate as well as on the amount of d character admixed to the $|\varepsilon k_M\rangle$ wave function.

III. EXPERIMENTAL PROCEDURE

The above-described phase-sensitive control scheme is realized by the experimental setup schematically shown in Fig. 1(b). A Nd:yttrium aluminum garnet (YAG) pumped dye laser (Lambda Physik Scanmate 2EC-400) operates within the 441–437 nm wavelength range at a repetition rate of 10 Hz and delivers linearly polarized pulses of ~5 ns duration and ~0.1 cm^{-1} linewidth. The dye laser output is frequency doubled in a BBO type-II crystal whose phase matching angle is automatically adjusted for optimized second harmonic output as the wavelength is scanned. After the crystal, the fundamental (ω) and frequency doubled (2ω) beams have mutually orthogonal linear polarizations and propagate collinearly through a Soleil-Babinet compensator for manipulating their relative phase $\varphi_{2\omega}-2\varphi_\omega$. Subsequently, they are focused to a Sr atomic beam which they cross perpendicularly. The achromatic focusing system consists of a plane and a concave mirror, the latter having a ≈25 cm focal length. The interaction chamber is evacuated by a 250 l/min turbo molecular-pump–rotary-pump system and it is also equipped with a liquid nitrogen trap. The achieved background pressure is $\sim 5\times 10^{-7}$ mbar and the Sr atomic beam density $\sim 10^8$ atoms/cm^3. The laser atom interaction takes place within two circular stainless steel plates separated by ≈1 cm. One of the plates is grounded while a positive voltage is applied to the other for creating a static electric field of strength F between 0.2 and 2 kV/cm. Sr^+ ions produced by photoionization are pushed by the field into a direction perpendicular to both the atomic and laser beams and pass through the grounded plate via a <1 mm hole drilled on its center and toward a 30-cm-long grounded time-of-flight (TOF) tube where they are discriminated against parasitic ions. At the end of the tube the ions are detected by a pair of microchannel plates (MCPs). The MCP signal is monitored by a digital oscilloscope, processed by a boxcar integrator and fed to a chart recorder and a personal computer where the data are finally stored.

The polarization of the uv 2ω beam is horizontal and parallel to the static electric field, while the polarization of the fundamental ω beam is originally perpendicular to it. When the $|M|=2$ two-photon excitation is unwanted a system consisting of a 2.44-mm-thick quartz polarization rotator followed by an alfa-BBO (beta barium borate) Rochon prism linear polarizer is placed immediately after the doubling crystal. The system matches with minimal power losses the polarization of the fundamental beam to that of the uv one, leaving the latter unaffected.

The radius of the ω beam is larger than the radius of the 2ω one. Only their common central parts are allowed to enter the interaction chamber by placing an iris of ~1 mm diameter in front of the mirror system. The iris serves the task of minimizing the creation of parasitic Sr^+ ions in laser-atom interaction regions where the two light beams do not overlap and thus phase control cannot be attained. It is indeed found that there is an optimum iris diameter leading to the highest modulation depth achievable for a given transition. This is attributed to a better matching between the two beam diameters near the focus since it is observed that the waists depend on the iris diameter. However, it was not possible to measure them with reasonable precision and this effect was not explored further. It is, nevertheless, expected that equal beam diameters in front of the focusing mirror would generally result in unequal waists at the interaction point so parasitic Sr^+ production cannot be completely eliminated with this setup. More appropriate would be to use a set of dichroic mirrors for splitting and then recombining the two beams in a Mach-Zehnder interferometer-type arrangement [15], in order to independently manipulate each beam's power and waist. Nevertheless, our intention here is to explore the capabilities of a simple and most general experimental setup where the needs for laborious overlap procedures and wavelength-specific dichroic mirrors are removed.

The use of a Soleil-Babinet compensator as a relative phase manipulator deserves a final comment. As is well known, the compensator is composed of a block and two identical prisms [Fig. 2(b)], all made of the same birefringent material (MgF_2 in the present case). For simplicity we denote as fast axis (FA) of the device as a whole the direction of the fast axes of the two prisms which are parallel to each other but perpendicular to the block's fast axis. All axes are perpendicular to the laser beams propagation direction. In our experiment the light beam polarizations are set either parallel or perpendicular to FA (with an uncertainty of ~1 deg) and the variation of $\varphi_{2\omega}-2\varphi_\omega$ is accomplished by sliding one prism relative to the other over a given maximum range. For that same range however, the number of observed 2π cycles

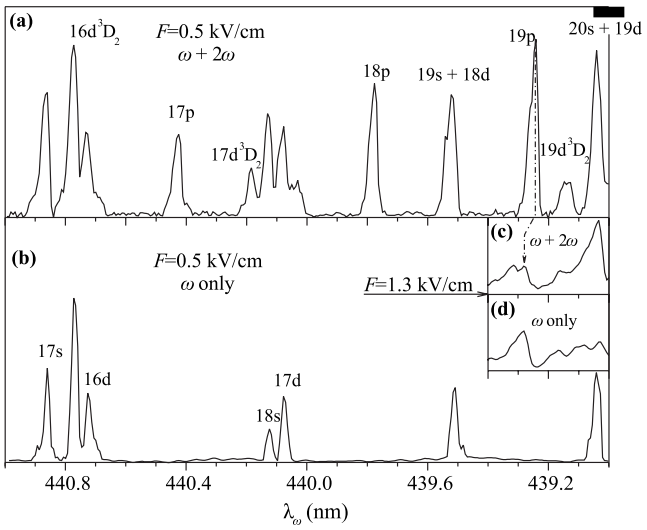

FIG. 2. (a) Sr Stark spectra recorded for a field strength $F=0.5$ kV/cm with both laser beams present. The observed resonances are marked by their zero-electric-field labels. Only the Rydberg $n\ell$ electron label of each (singlet) $5sn\ell$ level is given except for triplet states where their symmetry is also specified. In this range some $5sns$ and $5sn'd$ lines lie very close to each other and are not resolved. (b) Same as in (a) but with only the ω beam present. The uv beam is suppressed by means of a filter placed after the doubling crystal. Note the absence of $5snp$ resonances. (c) Same as in (a) but for a field strength of $F=1.3$ kV/cm. The shifted location of the $5s\epsilon k_M$ state corresponding to the zero-field $5s19p\ ^1P_1$ level is denoted by the dashed-dotted line. At that field strength many additional resonances appear one of which is positioned very close to the "$5s19p\ ^1P_1$" one. (d) Same as in (c) but with only the ω beam present. The "$5s19p\ ^1P_1$" resonance is now observed in the two-photon spectrum because the state has acquired substantial s and d character. On the opposite, the $5s(21s+19d)$ level has acquired substantial p character and it is less efficiently excited as compared to (c).

of $\varphi_{2\omega}-2\varphi_\omega$ depends on both the relative polarization directions (parallel or crossed) as well as on their orientation with respect to FA (four combinations in total). As an example, consider the crossed polarizations case where FA may be parallel or perpendicular to the ω beam. Then, it may be easily predicted that for MgF_2 every cycle of the former case corresponds to ~2.4 cycles of the latter. This, experimentally confirmed, fact provides an efficient means for unambiguously verifying the observation of phase sensitive coherent control and distinguishing it from parasitic, artifact signal variations.

IV. RESULTS AND DISCUSSION

Typical recordings of bound level $\varepsilon<E_{SP}$ spectra are given in Fig. 2. It should be emphasized that the studied energy range presents a rather complicated zero-dc-field energy level structure due to the occurrence of doubly excited states perturbing the $5snd\ ^1D_2$ and $^3D_2$ bound Rydberg series and causing their singlet-triplet mixing [19] (a similar situation holds for the $5snp$ series but without mixing between the $^1P_1$ and $^3P_1$ states [20]). Therefore, the simple picture implied by Eqs. (1) and (2) is not very precise in this case. Nevertheless, that specific part of the spectrum is deliberately chosen for demonstrating the generality of the method. A comparison between Figs. 2(a) and 2(b) reveals that for $\varepsilon<E_{SP}$ the resonances originating from one-photon absorption appear only when both laser fields are present but not when only the uv (2ω) beam enters the interaction chamber. This is in accordance with earlier experiments where one-photon excited $|M|=1$ states manifested themselves by means of field ionization, but not photoionization [21], and it is due to the fact that these highly excited states present a very low photoionization cross section under uv illumination. On the contrary, the corresponding cross section for visible light is apparently substantial. Due to the above observation, at this energy range both beams need to be present while verifying that one-photon excitations are not saturated. For a given $5s^2\rightarrow 5snp$ transition and iris diameter the dye laser output is reduced before passing through the doubling crystal until the Sr^+ signal varies linearly with uv pulse energy. If necessary, the procedure is repeated after setting the phase matching angle of the doubling crystal at a slightly nonoptimized value. Measurements are carried out preferably at the lowest dc field strength allowed by our TOF collection efficiency (~0.2 kV/cm) where we expect the Stark mixing effect to be small. Under such conditions the lines corresponding to $5s^2\rightarrow 5snd$ transitions are found unsaturated as well (ω-beam pulse energy) and there is no observable nonresonant background. At $F=0.5$ kV/cm [Figs. 2(a) and 2(b)] which is also relatively low (considering the excitation energy) ℓ mixing does occur, although this is not very evident at first glance. To realize this, let us note first that at this field strength the line intensities of predominantly p resonances are weaker than those recorded at the aforementioned lowest operational strength of ~0.2 kV/cm while they are further decreasing as F increases. Second, for the iris diameter employed for recording those spectra the corresponding intensities of predominantly s and d resonances are comparable to those of p resonances at $F=0.5$ kV/cm, but much lower than them at ~0.2 kV/cm. In addition, at $F=0.5$ kV/cm s and d lines are stronger when both laser fields are present than with the ω beam alone. Third and more important, the observed signal's modulation depth is small but nonzero for the majority of lines, as will be shown shortly. At higher electric field strengths (while still being within the $\varepsilon<E_{SP}$ range) many new resonances appear and most of them are common to both the $(2\omega+\omega)$ and ω only spectra. Moreover, they are found shifted from their zero-field positions (the effect is less pronounced for the s states). As an example, consider the zero-field $5s19p\ ^1P_1$ level. From Figs. 2(c) and 2(d) it is evident that for a field strength of $F=1.3$ kV/cm the corresponding Stark state has acquired comparable amounts of p and $(s+d)$ character. Figure 3(a) shows the signal modulation curve $S(\varphi_{2\omega}-2\varphi_\omega)$ recorded at that field strength with the laser wavelength tuned to the line peak. By fitting the curve to a form $S=A[1+(V/2)\cos(Bx+C)]$ (with x being the Soleil-Babinet compensator setting in arbitrary distance units) we find a modulation depth of $V\approx 65\%$. This modulation depth value is typical for all reso-

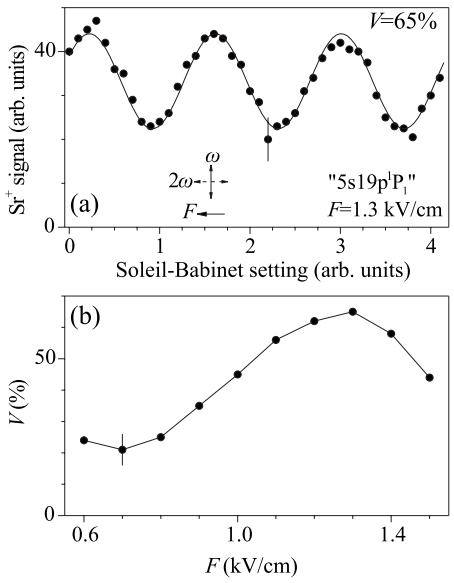

FIG. 3. (a) Sr^+ signal modulation as a function of the Soleil-Babinet compensator setting x (in arbitrary distance units) which is proportional to the relative phase $\varphi_{2\omega}-2\varphi_{\omega}$ between the two laser beams. The static field strength is $F=1.3$ kV/cm and the laser wavelength is tuned to the maximum of the "$5s19p\ ^1P_1$" line. Also shown are the laser polarization directions with respect to $\vec{F}$. The iris diameter is ~1 mm. The pulse energies for the ω and 2ω beam just before entering the interaction chamber are ~100 and ~30 μJ resulting in roughly estimated intensities of ~30 MW/cm^2 and ~10 MW/cm^2, respectively. Typical error bars are shown for a single point. The solid line is the result of a least squares fit to the form $S=A[1+(V/2)\cos(Bx+C)]$ giving $V=(65\pm3)\%$. (b) Fitted modulation depth values as a function of F for the "$5s19p\ ^1P_1$" line. All parameters are identical to those of curve (a) except that for each field strength the laser wavelength is tuned on the corresponding line maximum. Typical error bars are shown for a single point. The line connecting the experimental data is drawn for guiding the eye.

nances of Fig. 2 that correspond to zero-field $5sns\ ^1S_0$, $5snp\ ^1P_1$, and $5snd\ ^1D_2$ states (measurements were not performed for $5snd\ ^3D_2$, $n=17-19$ states due to their weak ion signal level). More importantly, that fairly high value of V is obtained with no particular effort concerning the iris diameter (~1 mm). For a particular transition and field strength, a more careful adjustment of this diameter [ensuring approximately a doubled ($\omega+2\omega$) signal with respect to the ω-only one] leads to modulation depths reaching or exceeding ~85%.

In Fig. 3(b) the fitted modulation depth values for the "$5s19p\ ^1P_1$" state are plotted as a function of the static field strength F. All measurements are recorded under identical conditions, apart from the laser wavelength which is always set at the field-dependent line maximum. The observed behavior of the $V(F)$ curve is a consequence of the variation of the admixture coefficients $c_{s,p,d}(F)$ of Eq. (1). Due to the above mentioned spectral complexity, the experimental spectroscopic data concerning the presently studied range are incomplete and not analyzed in detail [22], while calculations on the Stark effect of Sr have been limited to lower excitation [23]. Therefore, the coefficients $c_{s,p,d}$ are unknown and we restrict ourselves to a qualitative discussion. For low F the state exhibits a dominant p character, its two-photon excitation is weak, and consequently V is low. As F increases the c_p coefficient decreases in favor of the $c_{s,d}$ ones, leading to a growing modulation depth. At some appropriate field strength their relative values are such so as to maximize it. However, as F increases further it is very likely for c_p to become practically zero (or at least very small) as was observed in Stark effect computations on high-ℓ states of Ba [24]. In that high-ℓ case the relevant coefficient was found negligible at ~10 V/cm but since here we are dealing with a low-ℓ state it would not be surprising for the same effect to occur at ~1.5 kV/cm. Then, the dipole one-photon transition becomes progressively forbidden and the modulation depth decreases. The behavior of "$5sns$" and "$5snd$" states is similar but the maximum modulation depth occurs at state-specific field strengths for the same pulse energies and iris diameter. It has to be noted further that the data of Fig. 3 are acquired with crossed laser polarizations. Under otherwise identical conditions the parallel-polarizations recorded modulation depth is found systematically lower by ~20%. We tentatively attribute this behavior to the absence of the above mentioned $W_{\mathrm{exc},|M|=2}$ phase control contribution caused by the combination of one-photon-quadrupole two-photon-dipole excitations. This contribution could also partly explain the fact that, apart from the $5s16d$ resonances where the singlet and triplet components are comparable [19], the lines corresponding to (nominally) $5snd\ ^3D_2$ states are observed only when both beams are present. Although the singlet component of $5snd\ ^3D_2$, $n=17-19$ states is nonzero, it is apparently not sufficiently high to allow their detection via the ω beam alone, even if its intensity is increased to such a level where the onset of three-photon nonresonant ionization is evident. Given that the zero-field $5snp\ ^1P_1$ and $^3P_1$ series of Sr are not mixed [20] and that the electric field does not couple states of different total spin, the one-photon dipole transition amplitude is also expected to be very weak. As a consequence, constructive interference solely between dipole excitation amplitudes appears insufficient to explain their emergence in Fig. 2(a) and $W_{\mathrm{exc},|M|=2}$ might be of particular importance, especially at low Stark field strengths. Certainly, this point has to be clarified by further experiments with $F=0$.

Let us now discuss the $\varepsilon \geqslant E_{\mathrm{SP}}$ range where level mixing is by far more intense and complicated due to the high density of quasidiscrete Stark states. The spectra corresponding to $F=2$ kV/cm are plotted in Figs. 4(a) and 4(b). Here the uv beam directly photoionizes Sr atoms and the Sr^+ signal appears slightly below E_{SP}. In fact, uv photoionization turns out to be much more efficient than two-photon ionization. For approximately equalizing the two signals it is found necessary to decrease the pulse energy of the 2ω beam by about a factor of 10 with respect to the $\varepsilon < E_{\mathrm{SP}}$ spectral range. Furthermore, these experiments are conducted using parallel polarizations because, otherwise, the two-photon excited $|M|=0$ and 2 spectra merge together, resulting in a seemingly structureless signal background with no apparent resemblance to the $M=0$ single-photon ionization spectrum. On the contrary, the parallel polarization one- and two-photon

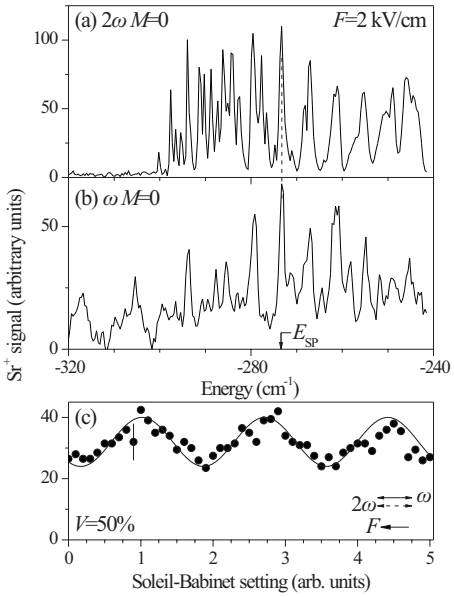

FIG. 4. (a) One-photon $M=0$ Stark spectrum in the vicinity of the saddle point for a field strength $F=2$ kV/cm. The energy scale refers to the field-free first ionization threshold of the Sr atom. Note that below -300 cm^{-1} the uv laser beam does not produce any photoionization signal. (b) Two-photon $M=0$ Stark spectrum for the same energy range and field strength. The vertical dashed line marks the resonance at $\approx$437.9 nm whose location practically coincides with the saddle point $E_{SP}=-273.8$ cm^{-1} (noted by an arrow). (c) Sr^+ signal modulation as a function of the Soleil-Babinet compensator setting. Also shown are the laser polarization directions with respect to $\vec{F}$. The iris diameter is $\sim$1 mm. The pulse energies for the ω and 2ω beam just before entering the interaction chamber are $\sim$70 and ~ 2 μJ (roughly estimated intensities $\sim$20 MW/cm^2 and $\sim$1 MW/cm^2), respectively. The solid line is the result of a least squares fit of the same form as in Fig. 3 yielding $V=(50\pm3)\%$.

spectra, although not identical, exhibit many similarities, in particular a number of strong, sharp, and more or less equidistant resonances. Figure 4(c) shows the $S(\varphi_{2\omega}-2\varphi_{\omega})$ curve recorded on the maximum of the $\approx$437.9 nm resonance marked by the vertical dashed line in Figs. 4(a) and 4(b). That resonance is located practically at the classical saddle-point energy [see Fig. 4(b)]. Without taking any particular maximization measures concerning the iris diameter a modulation depth of $\sim$50% is obtained. Similar results are recorded for all the sharp and intense lines which are common to both the 2ω and ω spectra.

V. CONCLUSION

We have experimentally demonstrated that by applying a dc electric field $\vec{F}$ which breaks the radiative transitions selection rules it is possible to use the phase sensitive coherent control technique for manipulating the total atomic excitation and ionization yields using the simplest $(2\omega)/\omega$ excitation scheme, i.e., by one- and two-photon dipole transitions. The method has been tested to the high-lying Stark states of Sr atom and it is found applicable irrespectively of the complexity of either the zero-field Rydberg spectrum or the Stark spectrum below as well as above the classical saddle-point energy. For a given set of ω- and (2ω)-beam power densities, chosen within a certain reasonable range, there is a static field strength value that maximizes the depth V of signal modulation as a function of the relative phase between the two laser fields. The discussion on that observation remained here qualitative. Nevertheless, in the future this kind of $V(F)$ measurements could be complementary to traditional experimental means for quantitatively testing theoretical treatments of the Stark effect. They could also be proved quite useful in situations where several constrains on the choice of laser intensities apply, thus offering great flexibility to the design of phase sensitive interactions.

Our simple experimental setup can be improved in a number of ways. The aforementioned splitting and recombination of the fundamental and second harmonic beam in a Mach-Zehnder interferometer-type arrangement, combined with an unfocused laser beam geometry is one of them. Furthermore, at present a single dc field plays the double role of a Stark field and a TOF ion extracting field. By using a separate, pulsed TOF (or ionizing) field, measurements down to $F\approx 0$ will be possible. Apart from the increased detection efficiency and functionality implied by that modification, there are also other advantages, such as, for example, the use of $5snd$ Rydberg target states and low field strengths for independently evaluating the importance of the $W_{\mathrm{exc},|M|=2}$ phase control term caused by the combination of one-photon-quadrupole and two-photon-dipole excitations.

The extension of the present methodology to autoionizing Rydberg states, where experimental investigations are scarce [4,7], appears as the natural continuation of the present study. Particularly motivating is the possibility of (dc field plus phase) control of total autoionization rates and spectral line shapes. Moreover, the dc field can be employed even beyond the phase control of quantities related to total yields. Note, for example, that the presence of low field strengths (a few tenths of V/cm or even lower) during excitation and detection is at the heart of electron imaging techniques via which phase control experiments on angular distributions and branching ratios of photoelectrons were previously conducted in alkaline earth atoms [7]. Such fields, on the other hand are sufficient for inducing a high degree of Stark mixing to Rydberg states located sufficiently close to their respective ionization threshold. It would be highly interesting to study the influence of the field strength on those phase-manipulated observables (especially the branching ratios) for these high-lying autoionizing Stark states.

Finally, while in our study an atomic system was employed, the concept may have implications for and applications to systems which, in general, do not possess inversion symmetry. Admixture ions in crystalline host materials are an example of such systems where state mixing and splitting is determined by the symmetry and strength of the crystal field, thus allowing parity nonconserving transitions. The results of studies such as the one presented herein may provide useful clues for devising excitation schemes that may selectively enhance or suppress coherent excitation of certain states.

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