Direct theoretical method for the determination of peak laser intensities from Freeman resonances in above-threshold ionization

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(Received 8 June 2009; published 17 November 2009)

Freeman resonance is one of the most important phenomena in strong-field laser physics. The previous calculations to determine peak laser intensities were mainly based on solving the entire time-dependent Schrödinger equation, which requires tedious theoretical and numerical work. The newly obtained exact solutions to a driven two-level atom made accurate calculations of quasienergies and the Bloch-Siegert shifts with an arbitrary laser-beam intensity possible and practical. With the recent progress in the numerical calculations of driven two-level atom, we find a direct theoretical method, without solving the entire time-dependent Schrödinger equation, to calculate the peak laser intensities which can excite a ground-state electron to a resonant Rydberg state with shifted energy level followed by an above-threshold ionization. Due to the tranquility of the nonresonant Rydberg states, Freeman resonance fits the driven two-level atom theoretical model well. With accurate calculation of Bloch-Siegert shift as a function of laser-beam intensity, we determine the peak laser intensities from Freeman resonances of different Rydberg states. Some important features of Freeman resonances are also discussed with this method.

DOI: 10.1103/PhysRevA.80.053417 PACS number(s): 32.80.Ee, 32.70.−n

I. INTRODUCTION

In 1987, Freeman *et al.* found experimentally [1] that the above-threshold ionization (ATI) peaks [2] broke up into many small peaks when the laser pulses were short. The appearance of the small peaks was interpreted as multiphoton resonances between the ground state, say $5P_{3/2}$ for the outermost shell electrons of xenon atoms, and Rydberg states with a shifted energy level. In the literature these resonances are called Freeman resonances. From the nineteenth century, the main work force to explore the microscopic world is the study of light spectroscopy, while the study of electron spectroscopy is far behind. The study of photoelectron spectra by Einstein is one of the most glorious examples of using electron spectra to study the microscopic world. Einstein's relation for photoelectric effect is $E_k+E_b=h\nu$, where E_k+E_b is the sum of the photoelectron kinetic energy and the binding energy of the initial bound electron. The sum is equal to the energy of the absorbed photon. For a known photon frequency, the photoelectron kinetic energy signal in a photoelectron energy spectrum therefore directly gives the electron bound-state energy level $-E_b$. In ATI, the kinetic energies of photoelectrons forming Freeman resonances satisfy a relation $E_k+E_r=nh\nu$ [1], where n is an integer, similar to the Einstein's relation. With this relation and a known laser frequency Freeman-resonance peaks on a photoelectron energy spectrum can be recognized when these resonances occur at various laser intensities. This allows a direct visualization of all Rydberg-state energy levels.

After Freeman *et al.*'s finding, a number of experimental [3–10] and theoretical studies [11–22] were devoted to Freeman resonances. Van Woerkom's group observed various participating Rydberg states in Freeman resonance [6]. Recently, Liu's group investigated experimentally the condition of the laser polarization and wavelength to produce Freeman resonances [9]. Now Freeman resonance, ATI peaks, and high harmonic generation [23,24] are the three most important phenomena in strong laser physics in nonrelativistic regime.

In the past, peak laser intensities were obtained by measuring the pulse duration, the size of the focus spot, and the total energy of the pulse. Since the time duration and the size of the spot are all geometric quantities, we may call this kind of methods geometric methods. Unfortunately, these methods do not provide accurate measurements of peak laser intensities. The best solution to improve the measurements is using the observation of some known physical effects which occur at certain peak laser intensities. We may call this kind of methods physical methods. A physical method will provide more subjectively accurate measurement than those from geometric methods. Freeman resonance is one of the best candidates among these physical effects. One of the main purposes of this paper is to facilitate the measurement of the peak laser intensities from geometric methods to physical methods. There are already several experiment groups which use Freeman resonance to calibrate the laser-beam intensities [3,9,25].

To utilize Freeman-resonance peaks in photoelectron spectra as marking events in the measurement of peak laser intensities, it is necessary to make the physical mechanism of Freeman resonance more clear. Currently, there are two interpretations for the energy shifts leading to Freeman resonances. One interprets it as the ponderomotive shift [1,3], the other as the Bloch-Siegert (BS) shift [26,27]. In this paper, we treat the energy shift leading to Freeman resonance as the

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BS shift. The BS shift is a relative energy shift of the energy spacing between two atomic energy levels of a two-level atom in a driving radiation field. Our treatment is based on the following two reasons: (1) Since a Freeman resonance is a true resonance [27], when it appears at a certain laser intensity, the transitions between the ground state and other excited states which are not in resonance can be ignored. Thus, the ground state and the excited state involved in a Freeman resonance should fit the driven two-level atom model very well. (2) It looks as, apparently, there are three states, the ground state, the resonant Rydberg state, and a continuum state, involved in the transition leading to a Freeman resonance. In our treatment the ground state and the Rydberg state form a two-level atom, while the continuum state for a photoelectron to reach is regarded as a derived state of the resonant Rydberg state according to the Floquet mechanism, which is automatically included in our theory.

The previous calculations to determine the triggering peak laser intensities of Freeman resonances were mainly based on solving the entire time-dependent Schrödinger equation, which required tedious theoretical and numerical work. Not long ago, the first analytically exact solution to the Bloch equation which described a two-level system interacting with a driven field was obtained [27]. The exact solution showed that the resonances occur only when the shifted energy spacing equals to an integer (measured by laser photon energy). This conclusion coincided with the resonances found experimentally by Freeman *et al.* [1]. The new solution made accurate numerical calculations of two-level atom interacting with a laser beam possible. A numerical solution in a special case [28] was obtained soon from the analytical solution and readily to be generalized to any arbitrary case. With further developed algebraic method, we calculate the quasienergies for all two-level atoms, which have an integer energy spacing originally. The results in all integer cases allow us to calculate shifted quasienergies for a generic two-level atom with an original energy spacing of a noninteger [29].

These calculated quasienergies are of a function of laser intensity. Our calculation shows that, at a certain laser intensity, the energy spacing of two-level atom may reach to an integer. Thus, a resonance occurs. These resonances correspond to Freeman resonances found experimentally. With this understanding, we apply the driven two-level-atom model to calculate the laser intensities which trigger the Freeman resonances. Theoretically, a calculated laser intensity is a constant. Experimentally, the laser-beam intensity of a pulse varies with time. In a laser pulse, when the beam intensity ramps to its lower bound which triggers the Freeman resonance, the Freeman resonance occurs. The laser beam with an intensity higher than the lower bound still triggers the Freeman resonance in the ramping process when the beam intensity passes through the lower bound. Thus, the laser intensities involved in the calculations correspond to the lower bound of peak laser intensities which trigger Freeman resonances in experiments.

This method brings three advantages. (1) With this method, one can evaluate the required peak laser intensities which trigger any Freeman resonance. (2) The energy levels of Rydberg states for an atom can be measured or remeasured through the measurement of Freeman resonances. This method can help to identify uniquely the resonant Rydberg states on the photoelectron energy spectrum. (3) One can apply this method to calibrate laser intensities using Freeman resonances as marking events.

In the current paper, we start with a brief description of the further developed algebraic method [27–29], which provides the basis for our calculation, then apply the method to calculate the Freeman resonances for xenon atoms exposed to an intense laser field.

II. ALGEBRAIC BASIS FOR THE NUMERICAL CALCULATION

The Bloch equation which describes a two-level atom interacting with single-mode light is ($\hbar = c = 1$) [27]

$$\left(\frac{d}{d\tau} - iD\cos\tau\sigma_x + i\Delta\sigma_z + iE\hat{I}\right)Y = 0, \tag{1}$$

where 2Δ stands for the original energy spacing (without light) in the unit of the laser photon energy ω ($\omega=1$), $2\Delta \neq 0$ in general; σ_x and σ_z are Pauli matrices; $\hat{I}$ is the 2×2 identity matrix. The quantity D relates to the ponderomotive parameter u_p ($u_p \equiv U_p/\hbar\omega$) and the field intensity I by $D = 2\sqrt{u_p m_e/\omega} = e\sqrt{8\pi I}/\omega^2$. The shifted energy E and wave function Y are quantities to obtain. The wave functions read

$$Y^{\pm} = \frac{1}{\sqrt{2}} e^{iD\sin\tau} \sum_{s=-\infty}^{\infty} A_s^{\pm} e^{-is\tau} \left(\frac{\Delta}{s \pm E} + 1, \frac{\Delta}{s \pm E} - 1\right)^t. \tag{2}$$

The accurate wave functions $Y^{\pm}$ can only be obtained with accurate E. The expression of E is given in terms of a set of infinite determinants [27]. When the original energy spacing is an integer, i.e., $2\Delta = n$ ($n=1,2,3,\ldots$), the shifted energy E_n can be evaluated with a single infinite determinant r_n as

$$E_n = \frac{1}{2\pi}\arccos[(-1)^n + (-1)^n 2\pi^2 r_n]. \tag{3}$$

For the noninteger case, the shifted energy E can be evaluated with all of the infinite determinants r_n as

$$E = \frac{1}{\pi}\arccos\sqrt{\cos^2(\pi\Delta) + \pi R_+ \sin(2\pi\Delta)}, \tag{4}$$

where

$$R_+ \equiv \sum_{n=1}^{\infty} \frac{4\Delta r_n}{4\Delta^2 - n^2},$$

and r_n are the infinite determinants described in Ref. [27], as a power expansion of D^2 determined by the laser intensity and frequency,

$$r_n = a_0 D^2 + a_1 D^4 + a_2 D^6 + a_3 D^8 + \ \ldots\ . \tag{5}$$

With the above formulas, we calculate accurately the shifted energies and corresponding wave functions directly. The detailed calculations of these infinite determinants are shown in another paper to be published in *Atomic Data and Nuclear Data Tables* [29].

TABLE I. Six-photon Freeman resonances from the ground state $5P_{3/2}$ to the Rydberg states $7p$, $4f$, $8p$, $5f$, respectively, with the corresponding resonance peak laser intensities and the kinetic energies of photoelectrons emitted from xenon.

Energy level	I_{fr} (W/cm^2)	Photoelectron kinetic energy (eV)
$7p$	1.79×10^{15}	0.88
$4f$	1.33×10^{15}	1.14
$8p$	9.66×10^{14}	1.33
$5f$	7.60×10^{14}	1.46

III. DETERMINATION OF PEAK LASER INTENSITIES FROM FREEMAN RESONANCES

Now we focus on how to use Freeman resonances as marking events to predict the resonant intermediate states, their locations in the photoelectron kinetic-energy spectra, and corresponding peak laser intensities. We choose xenon atom, which is irradiated by laser light of wavelength 616 nm, as the sample atom. The laser photon energy is 2.01 eV. The original energy spacing between any of the selected unoccupied excited states, i.e., Rydberg states, and the ground state $5P_{3/2}$ is less than six-photon energy. As the laser intensity increases, the energy levels of these unoccupied excited states all shift up. At some laser intensity, one of the shifted energy spacings reaches to an integer (measured by the laser photon energy 2.01 eV). Then a resonance, the Freeman resonance, occurs and the ionization rate is enhanced. In this laser intensity, the initially bound ground-state electron goes through the energy-shifted resonant Rydberg state, as a real intermediate state, and then automatically ionized due to the Floquet mechanism. Different unoccupied excited states resonate at different laser intensities because their original energy spacings with the ground state are different. In general, to drive a Rydberg state with a deeper binding energy into a Freeman resonance, a higher laser intensity is needed. Theoretically, the laser intensity used in the calculation is a constant parameter. Experimentally, once the peak laser intensity reaches the constant theoretical value, the corresponding Freeman resonance occurs. The following formula shows the photoelectron kinetic energy which appears on a photoelectron spectrum as a Freeman-resonance peak:

$$\frac{\mathbf{P}^2}{2m_e} = (j-m)\omega - E_r, \tag{6}$$

where $-E_r$ is the original energy level of the unoccupied excited state involved in the Freeman resonance, j is the total absorbed photon number for the photoelectron to ionize, and m is the absorbed photon number for the initially bounded electron to reach the shifted Rydberg state. This equation is the modified Einstein relation for photoelectric effect and completely equivalent to the one given by Freeman *et al.* [1]. Table I shows the calculated peak laser intensities to trigger a Freeman resonance and the kinetic energies of photoelectrons ionized through the intermediate states $7p$, $4f$, $5f$, and $8p$, respectively. These resonant Rydberg states, the corresponding triggering peak laser intensities, and locations in the photoelectron kinetic-energy spectrum agree with the original measurements made by Freeman *et al.* Figure 1 shows the energy spacing between $5f$ state and ground state varying with the peak laser intensity. Before the xenon atom exposes to laser light, the energy spacing between $5f$ and the ground state is 5.75 (measured by the laser photon energy 2.01 eV). The energy spacing increases with the increase of the laser-beam intensity. As the peak intensity reaches 7.60×10^{14} W/cm^2, the shifted energy spacing hits integer 6, the Freeman resonance occurs.

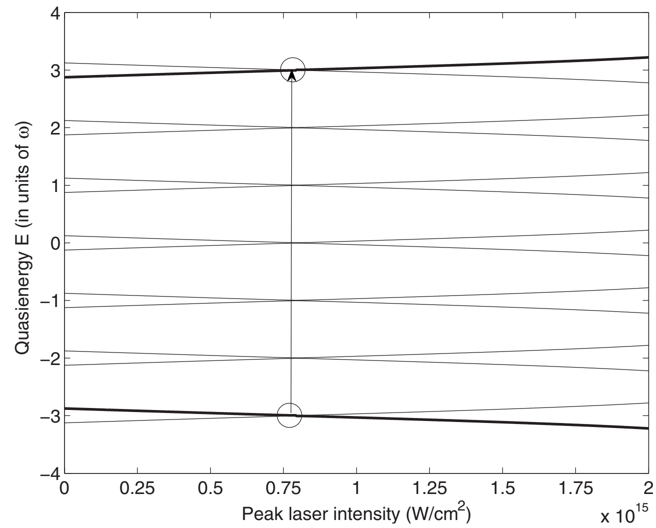

FIG. 1. The energy spacing (measured by the laser photon energy) between $5f$ and the ground state $5P_{3/2}$ as a function of the peak laser intensity. The main energy spacing is presented by the two thick lines while the derived quasienergy levels from the Floquet periodicity are presented by the thin lines in each respective group. The energy zero is chosen at the midpoint of the two main levels. When two lines intersect, the energy spacing becomes an integer, then a Freeman resonance occurs.

Our calculation method manifests the following features of Freeman resonances which were observed in experiments: (1) Different unoccupied Rydberg states resonate at different laser-beam intensities. (2) For a particular unoccupied Rydberg state, once its Freeman resonance appears in the photoelectron spectrum, this Freeman resonance has a fixed position as the laser-beam intensity increases. It becomes prominent as the peak laser intensity further increases. (3) To trigger the Freeman resonance of a Rydberg state with a deeper binding energy, a higher peak laser intensity is needed. When the peak laser intensity increases further, the Freeman resonance of another Rydberg state with a further deeper binding energy may occur.

IV. DISCUSSION

A. Some complications involved in identifying resonant Rydberg states

The theoretical method developed here may help experimentalists to identify different Freeman resonances in photoelectron kinetic-energy spectrum in different peak laser intensities. Following are two cases which involve some complications. (1) Two Rydberg states with an integer energy spacing (without light). One may think that two Rydberg states with an integer energy spacing may resonate simultaneously. According to the exact solution to a driven two-level atom [27], a true resonance occurs only when a laser-shifted energy spacing equals to an integer. These two Rydberg states make two different two-level atoms of different original energy spacings with the same ground state; thus the shifted energy spacings under the same peak laser intensity are different. So, the two Rydberg states cannot have simultaneous Freeman resonances. (2) Two different Rydberg states resonant at the same peak laser intensity. Under the same peak laser intensity, when two Freeman-resonance peaks appear simultaneously on the photoelectron energy spectrum at different locations, they belong to two different two-level systems with the same ground state. Thus they must have undergone different energy shifts. The original energy spacing between these two Rydberg states, from the point 1 discussed above, cannot be an integer.

B. Limitation of this method

For better applications and further developments of this method, the following aspects are to be considered.

(1) *Limitation of this method in the quality of laser pulses.* The driven two-level-atom model derived from quantum-field theory has no dipole approximation [27]. The derived exact wave functions and quasienergies automatically include the contributions from both rotating frequency and counter-rotating frequency. Thus, this method is expected to have a wide range of applications. In the current stage, this method may have the limitation in the periodicity of laser light in the case of extremely short pulses. In the few-cycle case, the laser pulses can be considered as a superposition of three neighboring modes in different frequencies. While in the subcycle case, the laser pulse can be considered as a superposition of three far separate modes. To break the limitation in the periodicity, the generalization of the driven two-level-atom model from the single-mode case to the multimode case is to be considered. In Freeman *et al.*'s original experiment, the laser pulses are of picoseconds far beyond femtoseconds and attoseconds; thus, our method applies.

(2) *Limitation of this method in the energy regions of photoelectron spectra.* Based on exact solutions, this method automatically incorporates with Floquet mechanism. All Freeman-resonance peaks on the high energy region can be reproduced by adding integer multiple photon energies with theoretical calculations. Freeman-resonance peak was discovered in low energy region on photoelectron energy spectra. As a very preliminary work, we only compare our result with Freeman *et al.*'s original measurement. In the near future, we will apply our method to Freeman resonances in higher energy regions.

(3) *Limitation of this method in nonisolated resonances.* Since this method is derived from the exact solution of a driven two-level atom, it may not apply to the cases when the atomic transitions are involved with multiple energy levels. For example, to treat the case when two Freeman resonances are not well isolated on an electron energy spectrum, one may have to develop a driven three-level atom theory.

C. Discussion of the calculated peak laser intensities

The magnitude of calculated peak laser intensities ($\sim 8\times 10^{14}$ W/cm^2) which trigger Freeman resonances is only a half-order higher than that in the original experiment by Freeman *et al.* ($\sim 1.5\times 10^{14}$ W/cm^2). This discrepancy is from the different methods in estimating the laser-beam intensities. The laser-beam intensities in Freeman *et al.*'s experiment were, most likely, estimated by a geometric measurement, and should stand for the average values. For a laser pulse, the peak laser intensity that differs from the average intensity by a half-order may be considered as reasonable. Moreover, one of the main purposes of this paper is to facilitate the measurements of laser-beam intensity from rough geometric methods to more accurate physical methods. The initial discrepancy between theoretical calculations and experiment measurements can certainly be achieved with future calculations and measurements.

V. CONCLUSION

In the end of this paper, we conclude the following:

(i) Isolated Freeman resonances fit the driven two-level atom model very well because all nonresonant Rydberg states do not play any role in the resonant transition in both experiments and the theoretical model.

(ii) This calculation method developed for a generic driven two-level atom applies to Freeman resonances to predict the resonant intermediate states, their corresponding locations in the photoelectron energy spectra, and corresponding peak laser intensities.

(iii) The energy shifts leading to Freeman resonances are the Bloch-Siegert shifts in nature.

ACKNOWLEDGMENTS

This work was supported by the Shanghai Rising-Star Program, the China National NSF under Grant No. 10774153, and the 973 program of China. Y.S.W. was supported in part by the U.S. NSF under Grant No. PHY-0756958.

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