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Fine splits of photon emission spectrum of hydrogen atom caused by transitions between different dressed states in intense high frequency laser field*

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The photon emission spectrum of the hydrogen atoms in an intense high-frequency laser pulse is simulated by using one-dimensional soft Coulomb potential. Regular fine structures appear on the two sides of both the odd and even multiples of photon energy of the laser field besides the ordinary odd harmonic peaks. It is proved that the splits of the fine structures are responsible for hyper-Raman lines and the energy spacing between the odd harmonic lines is equal to the difference in energy between the eigenstates with the same parity of the time averaged Krameters–Henneberger (KH) potential. By analysing the features of the fine structures, we also verify that the so-called even order harmonics under the stabilization condition are indeed hyper-Raman lines caused by the transitions between the dressed atomic states with different values of parity.

Keywords: high-order harmonic generation, hyper-Raman line
PACC: 4265K, 3280K, 3115

1. Introduction

In recent years, the rapid development of the laser technology has speeded up the research on the nonlinear and the non-perturbation phenomena in the interaction between intense laser field and atoms. One of these effects is stabilization,[1−3] i.e. when one photon energy of the laser is high enough, i.e. it equals or even exceeds the ionization energy of the atoms, the ionization probability of the atom will decrease with the laser intensity increasing. This effect is often understood by means of the so-called Kramers–Henneberger (KH) transformation,[4] in which the rapid oscillations of the external field are transformed into the atomic potential. The average potential with high laser frequency $\omega_{\rm L}$ is given by[4−6]

$$V_{\rm eff} = \frac{\omega_{\rm L}}{2\pi}\int_0^{2\pi/\omega_{\rm L}} {\rm d}t V\left[\boldsymbol{r}+\alpha(t)\right]. \qquad (1)$$

Here, $\alpha(t)$ is the classical electron displacement in the incident electric field, $\alpha_0 = |\boldsymbol{E}_{\rm L}|/\omega_{\rm L}^2$ denotes the maximum classical displacement of the electron in the incident field related to the strength $E_{\rm L}$ and frequency $\omega_{\rm L}$ of the incident laser field. Then $V_{\rm eff}$ has two attraction centres at $\pm\boldsymbol{E}_{\rm L}/\omega_{\rm L}^2$ separately, for the linearly polarized laser field.[7] $V(\boldsymbol{r})$ is the field-free atomic potential without applying any external field. In order to avoid the numerical problems caused by the singularity of the potential at $r = 0$, the soft Coulomb potential model $V(\boldsymbol{r}) = -1/\sqrt{r^2+a}$ is often used.[8−10] In our one-dimensional calculation, we take $a = 2$ so that the energy eigenvalue of ground state is -0.5 a.u., which is equal to the value of a real hydrogen atom.[8,9]

High-order harmonic generation (HHG), as a source of coherent x rays and attosecond extreme ultraviolet (XUV) light, is also an attractive subject. It is generated by the radiative recombination of the ionized electron and the parent ion as often interpreted in the three-step model[8,11,12] and it is located at odd multiples of the photon energy required by the parity conservation. Besides the odd order harmonic lines, it is predicted that there are hyper-Raman lines[13−15] as well. Their positions are given by $\omega_{\rm hR} = \omega_{\rm i} \pm n\omega_{\rm L}$, where $\omega_{\rm i}$ is the difference in energy between the dressed atomic bound states and $\omega_{\rm L}$ is the frequency of the laser pulse. Most investigations for the HHG were performed for the laser pulse of inten-

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sity and frequency far below those at the stabilization. In the present study, we will show the fine structures of the harmonic and hyper-Raman spectra of atoms in an intense and high-frequency laser field when the stabilization occurs such that the ionization is restrained and the requirements for the hyper-Raman line generation are satisfied.[13,15]

2. Brief description of the theory

Calculations in the present study are performed by using the symmetrized split operator fast Fourier transform method (SOFFT).[16−18] A smooth cosine mask function confined to a moderately narrow band near the boundaries is adopted in order to prevent reflection of the wave function from the edges of the grid and force the electron density to be zero on these edges. Therefore, the norm of the wave function is not conserved. We treat the electron density removed by the mask function as the density of ionized electrons and thus the decrease of the norm is considered to be the probability of ionization.[3]

The interaction between one-dimensional hydrogen atom and the laser pulse is expressed as the interaction Hamiltonian in the dipole approximation, i.e.

$$\hat{H}_{\rm I} = E_{\rm L} f(t) x \sin(\omega_{\rm L} t), \tag{2}$$

$f(t)$ is the envelop of the laser pulse chosen to have $\sin^2$ turn-on and $\cos^2$ turn-off of three optical cycles and to have a flat top between them unless otherwise stated. The whole duration of the laser pulse is one hundred optical cycles for the full evolution of the wave packet. The frequency of the laser $\omega_{\rm L}$ is chosen to be 1 a.u. throughout this paper, for half-photon ionization and the conditions necessary for stabilization which can be easily satisfied. For the well studied case of linear polarization, the stabilization condition is expressed as $E_{\rm L}^2/2\omega_{\rm L}^2 \geq \omega_{\rm L} \geq I_0$,[19] where I_0 is the ionization potential of the atom and it is 0.5a.u. for the atomic hydrogen.

3. Results and discussion

3.1. Both harmonic and hyper-Raman lines generated

Here we mainly consider the harmonic generation of atom in this high-frequency and ultra-intense laser field. It was first reported by Kulander *et al* [3] that the even harmonics, which are forbidden by parity conservation, appeared in the harmonic spectrum. It was explained as the fact that the localized wave function contained components of odd symmetry and the nonsymmetric medium is capable of containing even harmonics. Reed *et al* [20] calculated the harmonic spectrum under this condition by using the 1D soft Coulomb potential and found that the even harmonic strength was reduced for a longer turn-on time of the field. The explanation given by the authors is that the fast turn-on means that the evolution process is no longer symmetrical so that the parity is no longer an appropriate concept. However, we give a different explanation and prove that the even harmonics do not really exist for the soft Coulomb potential.

The harmonic spectrum is illustrated in Fig.1 with a solid line for $E_{\rm L}$=5.0 a.u. and a dashed line for $E_{\rm L}$=10.0 a.u. One can see the sharp peaks around even and odd multiples of the photon energy. There are clear and regular splits among the fine peaks. To see the fine structures clearly, the magnified details around the fourth and fifth harmonics are shown in the insets of Fig.1. For the fifth harmonic there are peaks symmetrical with respect to the main fifth harmonic line, while for the fourth harmonic the central peak does not actually exist, only lines symmetrically appear on both sides of the central position of the fourth order harmonic. We will demonstrate that these symmetric lines are hyper-Raman lines caused by the transitions between the dressed bound states.

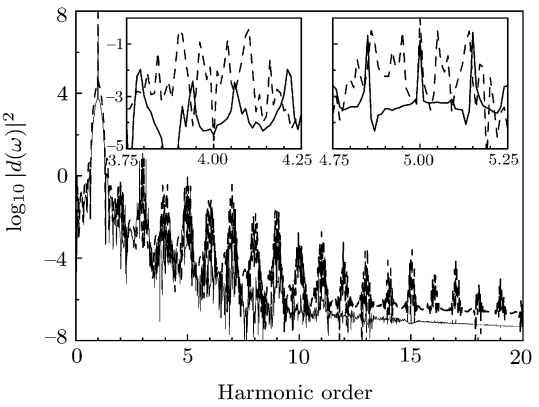


Fig.1. Photon emission spectra of the one-dimensional soft Coulomb potential $-1.0/\sqrt{x^2+2}$ in a laser field of frequency 1 a.u. with the values of amplitude being 5 (solid line) and 10 a.u. (dashed line). The magnified spectra around the fourth and fifth harmonic orders can be seen in the insets.

In Fig.1, the distribution and the separation of the lines around even and odd harmonic orders are different in the same laser field. This can be interpreted by the corresponding transitions among different eigenstates. For the Raman process, the atom first absorbs photons to a virtual energy level from the initial state and immediately transits to another state to emit a photon. If the atom first absorbs an even number of photons, the corresponding hyper-Raman lines should be located at $\omega_{\rm hR} = 2n\omega_{\rm L} \pm \omega_{\rm i}$, where $\omega_{\rm i}$ is the difference in energy between the initial and final states of different values of parity. If an odd number of photons are absorbed, the hyper-Raman lines will be located at $\omega_{\rm hR} = (2n+1)\omega_{\rm L} \pm \omega_{\rm i}$, with $\omega_{\rm i}$ being the difference in energy between the initial and the final states of the same parity. The dashed line in Fig.1 represents the harmonic spectrum with $E_{\rm L}$=10.0 a.u. Besides the difference in cut-off position, the solid and the dashed lines have a quantitative difference in fine structure. The detailed hyper-Raman lines with different laser intensities are not located at the same position because of their different eigenstates of the time averaged KH potential. We give the first six eigenenergies of the KH potential with $E_{\rm L}$=5.0 and 10.0 a.u. in Table 1. We examine the two features presented above and confirm the existence of hyper-Raman lines as compared with the fine structures illustrated in Fig.1. For example, the nearest peaks to the fifth harmonic at 5.1509 and 4.8491 when $E_{\rm L}$=5.0 a.u. correspond to the energy difference of 0.1509 a.u. between the ground state and the second excited state of KH potential with the same parity and the nearest peaks to the fourth harmonic at 4.064 and 3.936 to the energy difference of 0.064 a.u. between the ground and the first excited state with different values of parity. The transitions between the dressed bound states are mainly from the ground state. The energy difference will decrease with the intensity of the laser field increasing, and the separation of the fine structure is smaller for $E_{\rm L}$=10.0 a.u. What we have to clarify here is that the lines located on the two sides of both the odd and the even multiples of $\omega_{\rm L}$ are from the true Raman processes but not the three-step processes. In order to answer this question, the state populations of three situations with $E_{\rm L}$=5.0 a.u. including the present subsection will be discussed in the following in order to see clearly the features of the two kinds of lines.

Table 1. Energies (in a.u.) of the first six low-lying eigenstates of the 1D time averaged KH potential as a function of $E_{\rm L}$.

n	0	1	2	3	4	5
$E_{\rm L}=5$	–0.31473	–0.25066	–0.16383	–0.097512	–0.066571	–0.047193
$E_{\rm L}=10$	–0.21515	–0.20103	–0.16402	–0.11951	–0.07876	–0.053519

We illustrate the time-dependent populations of the first three bound states of the KH potential in Figs.2(b) and 2(c) with the corresponding harmonic spectrum in Fig.2(a). Figure 2(c) shows the changes in population in one optical cycle. In the high frequency and intense laser field the wave packet oscillates along the polarization direction with the same frequency as that of the laser pulse train. During each optical half cycle, the ionized flux leaves the vicinity of the atom along the direction of the instantaneous electron field. Then the time-dependent populations of the bound states reach theirs twice maximum in one optical cycle, resulting in the production of the normal harmonic lines. One can also see the Raman transitions from the first and second excited states to the ground state and then from the ground state to the first and second excited states within a very short time corresponding to the electron recombination time. To see the general change tendency, we choose the maximum in half the optical cycle and connect the 200 maximum points of the population of the first three bound states as shown in Fig.2(b). The electron is mainly located in the ground and the first excited states by turns. There are also clear oscillations with a period of about six optical cycles. This is the Rabi oscillation, and it is proportional to the intensity of the laser field. This oscillation is slow and the maximum populations of the bound states do not change much from one optical cycle to another. From the Rabi oscillation we can also see the transitions between the bound states with the same and different parities separately.

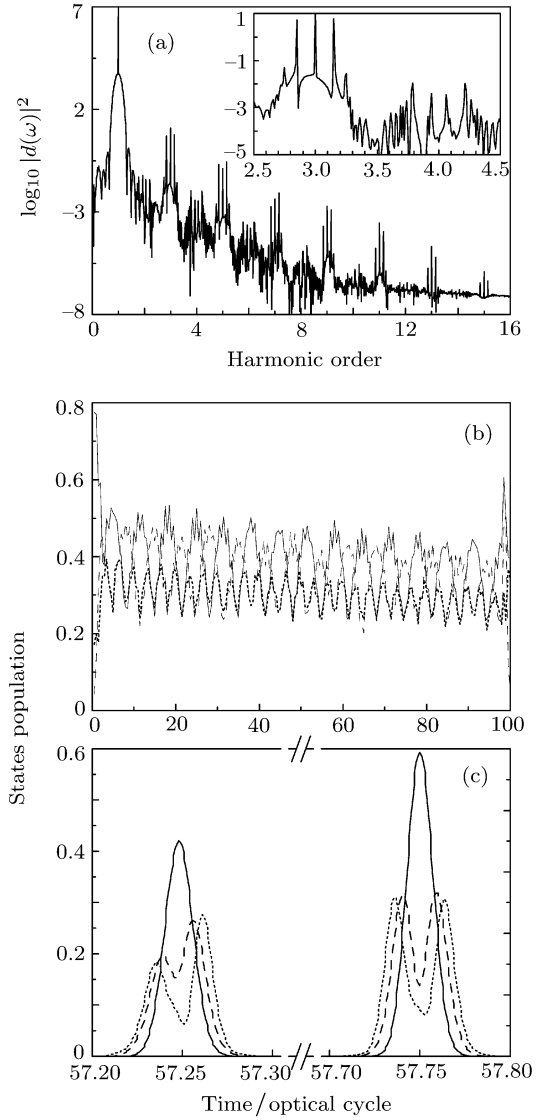


Fig.2. The photon emission spectra shown in Fig.1 by solid line (a); connection of the two maximum points of the time-dependent populations of the first three states of the KH potential in one optical cycle (b); time-dependent populations of the first three bound states of the KH potential in one optical cycle (c). In (b) and (c), solid, dashed and dotted lines refer to the data of the ground, the first excited, and the second excited states, respectively.

3.2. Only hyper-Raman lines generated

It is shown in Refs.[21, 22] that by choosing an appropriate coherent superposition state as the initial state one can generate only the hyper-Raman lines especially for the diabatic case $T_{\rm on} = 0$. However, an appropriate turn-on time is essential to making the population spread across many of the bound eigenstates of KH potential. With too short a turn-on time, the electron cannot be trapped effectively and stabilization will not take place[3,4] then the electron is ionized rapidly and both hyper-Raman and harmonic lines do not appear. Therefore, we do not change the parameters of the laser pulse. Although the conditions of the atom model and the laser pulse are different from those in the previous work, we obtain a similar result when the initial state is chosen to be the combination of the ground state and the first excited state of the KH potential. The emission spectrum is shown in Fig.3(a). We can see the strengthening of the hyper-Raman lines as compared with that in Fig.2(a). The increase of the population of the first excited state enhances the probability of the Raman transition from it. The lines around the fourth order in Fig.2(a) indicate the transitions between the ground state and the first excited state and between the ground state and the third excited state. Hyper-Raman lines caused by the transition between the first excited state and the second excited state are also obvious in Fig.3(a). The third order normal harmonic line is effectively restrained. This also indicates that the peaks near the normal harmonic lines are mainly from the Raman transitions between the dressed states too. The strengthening of the hyper-Raman lines corresponds to the strong transitions between the bound states as shown in Fig.3(b) where the oscillation of the population is too rapid to be seen clearly and the electron also has the possibility to stay mainly in the second excited states. Besides the regular Rabi oscillation, the population changes much in one optical cycle particularly for the ground state, which can be seen clearly from Fig.3(c). Each time the electron comes back, the population distribution among the bound states changes a lot, i.e. strong Raman process occurs.

3.3. Only harmonic lines generated

As mentioned above, turn-on time is necessary for the transitions between the dressed atomic states. The increase of the turn-on time can reduce the spread of the population among the bound states and locate most of the population in the ground state, and the transitions between the bound states are also reduced. The harmonic intensity changes slightly with the turn-on time increasing, while the hyper-Raman lines do

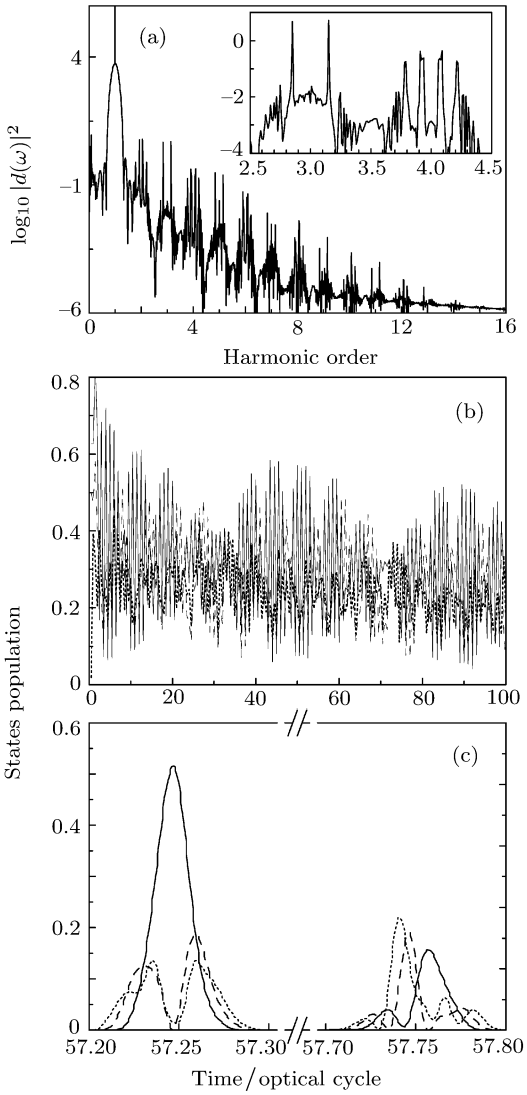


Fig.3. As Fig.2 but for the initial state being the superposition of the ground and the first excited states of the KH potential.

not exist any more as shown in Fig.4(a), where the turn-on and turn-off times of the laser pulse are increased up to forty optical cycles with the same total laser duration and the field-free ground state as the initial state. Obviously there are only odd harmonics in this figure. The harmonic lines are considerably broadened compared with those in Fig.2(a). The broadening arises from the gradual variation in ground state energy from –0.5 to –0.31473 a.u. During the slow turn-on process of the laser pulse, when the electron ionized from the ground state comes back and recombines the ion, the energy of the ground dressed state is enhanced by the laser field, resulting in a slight reduction in energy of the emitted photon and the odd harmonic lines deviating to the left of the normal position. One can see the overlapping small peaks on the broad profile, which are corresponding to the process enhancing the ground state for each optical circle. The turn-off process causes an opposite shift of the emitted photon. So the harmonic peaks are broadened up to a half width of about 0.18527, the difference in energy of the ground state between with and without the laser field.

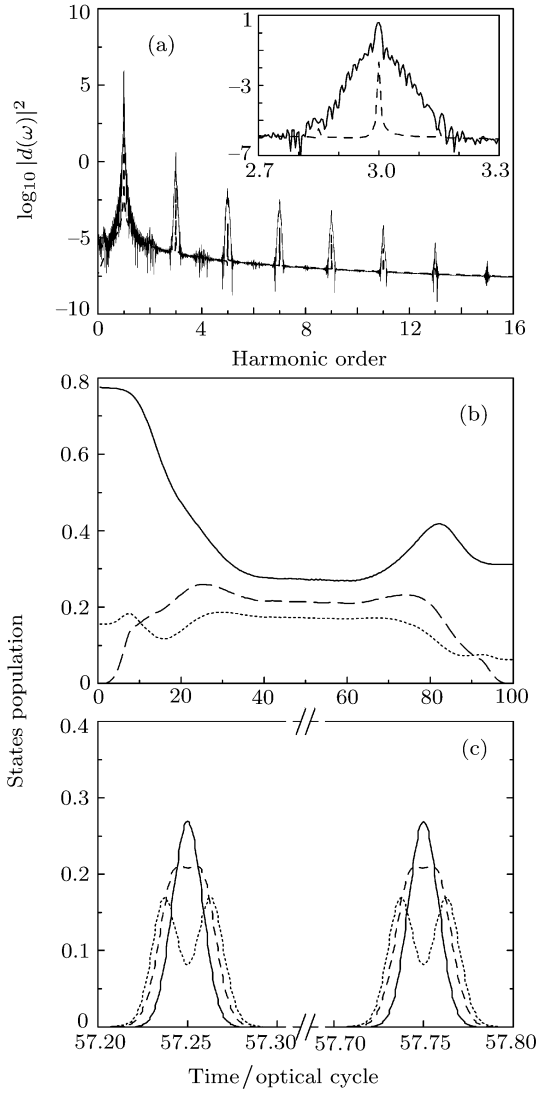


Fig.4. As Fig.2 but for the laser pulse with forty optical cycle turn-on and turn-off times, with the whole duration being unchanged.

To remove the broadening caused by the turn-on and turn-off processes, narrow harmonic lines can be obtained by taking only the spectrum after the turn-on and before the turn-off of the laser pulse, i.e. the Fourier transform is performed only from forty to sixty optical cycles, i.e. in the flat part of the laser pulse. The result is shown in Fig.4(a) with the dashed line. For the case of only harmonic lines, the populations of the bound states nearly are the same during the flat part of the laser pulse and most of them are lo-

cated in the ground states as shown in Fig.4(b). The transitions between bound states can be hardly seen in Figs.4(b) and 4(c). The reduction in hyper-Raman line with a longer turn-on time accords with the result presented in Ref.[20], while our explanation as the transitions between bound states seems to be more reasonable.

4. Conclusion

The photon emission spectra of the atoms interacting with a high-frequency and intense laser field are calculated by using one-dimensional soft Coulomb potential and the obtained results, including the time-dependent population of bound states and the harmonic spectrum, are interpreted in terms of the time averaged KH potential. When the stabilization occurs, a considerable part of electrons oscillate between the bound states of the time averaged KH potential rather than being ionized. The photon emission spectrum under this condition is more complex because of these oscillations and the population spreading across many eigenstates in the KH potential during the turn-on time. The hyper-Raman processes are strengthened and shown as clear splits around both even and odd harmonics. We have calculated the spectrum and deduced the splits of the hyper-Raman lines around the even harmonics, which are determined by the difference in energy between eigenstates with different values of parity and those around the odd harmonics calculated by these of the same parity because of the parity conservation. We construct three cases (only harmonic lines, only hyper-Raman lines, and both lines exist), by using the simple one-dimensional model, to investigate the correlations and differences between the two lines with the help of the time-dependent population of the first three bound states of the time-dependent KH potential.

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References

[1] Pont M and Gavrila M 1990 *Phys. Rev. Lett.* **65** 2362
[2] Liu J, Chen S G, Li B W and Hu B 2000 *Chin. Phys.* **9** 24
[3] Kulander K C, Schafer K J and Krause J L 1991 *Phys. Rev. Lett.* **66** 2601
[4] Gajda M, Grochmalicki J, Lewenstein M and Rzażewski K 1992 *Phys. Rev.* A **46** 1638
[5] Potvliege R M 2000 *Phys. Rev.* A **62** 013403
[6] Grochmalicki J, Lewenstein M and Rzażewski K 1991 *Phys. Rev. Lett.* **66** 1038
[7] Patel A, Protopapas M, Lappas D G and Knight P L 1988 *Phys. Rev.* A **58** R2652
[8] Zhou Z Y and Yuan J M 2007 *Chin. Phys.* **16** 675
[9] Javanainen J, Eberly J H and Su Qichang 1998 *Phys. Rev.* A **38** 3430
[10] Liu W C and Clark C W 1992 *J. Phys.* B **25** L517
[11] Yang Y J, Chen G, Chen J G and Zhu Q R 2004 *Chin. Phys. Lett.* **21** 652
[12] Corkum P B 1993 *Phys. Rev. Lett.* **71** 1994
[13] Millack T and Maquet A 1993 *J. Mod. Opt.* **40** 2161
[14] Pons M L, Taïeb R and Maquet A 1996 *Phys. Rev.* A **54** 3634
[15] Zhou Z Y and Yuan J M 2007 *Chin. Phys. Lett.* **24** 683
[16] Feit M D, Fleck J A, Jr and Steiger A 1982 *J. Comp. Phys.* **47** 412
[17] Fleck J A, Jr, Morris J R and Feit M D 1976 *Appl. Phys.* **10** 129
[18] Li P C, Zhou X X, Dong C Z and Zhao S F 2004 *Acta Phys. Sin.* **53** 750 (in Chinese)
[19] Chism W, Choi Dae-Il and Reichl L E 2000 *Phys. Rev.* A **61** 054702
[20] Reed V C, Burnett K and Knight P L 1993 *Phys. Rev.* A **47** R34
[21] Liu C P, Gong S Q, Li R X and Xu Z Z 2004 *Phys. Rev.* A **69** 023406
[22] Piazza A D and Fiordilino E 2001 *Phys. Rev.* A **64** 013802