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FAST TRACK COMMUNICATION

Three-photon double ionization of Ar studied by photoelectron spectroscopy using an extreme ultraviolet free-electron laser: manifestation of resonance states of an intermediate Ar\textsuperscript{+} ion

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Abstract

Three-photon double ionization of an Ar atom has been investigated by photoelectron spectroscopy using an extreme ultraviolet free-electron laser. When the photon energy of the laser has been tuned to the resonances of the intermediate Ar\textsuperscript{+} ion, photoelectron peaks due to the resonant two-photon single ionization of the Ar\textsuperscript{+} ion have been observed. And it has been revealed that the appearance of the peaks is controlled by the mixing ratios of 3p\textsuperscript{4} core-configurations of the multi-configuration states. Furthermore, it has been shown that the laser intensity dependence of the Ar\textsuperscript{2+} ion production rate due to the resonant two-photon single ionization strongly deviates from that expected by perturbation theory.

(Some figures in this article are in colour only in the electronic version)
Multi-photon multiple-ionization experiments of free atoms with the new intense sources of extreme ultraviolet (EUV) radiation, the free-electron lasers and the high harmonics of optical lasers, have been reported in [1–5] and references therein. In such experiments, the charge states of product ions have been measured by the ion time-of-flight (TOF) technique. Although the charge states provide the primary information on multiple ionization, from them one cannot directly distinguish non-sequential (direct) multiple ionization, in which electrons are emitted simultaneously and thus share the total energy provided by photons, and sequential multiple ionization, in which electrons have discrete energies corresponding to the relevant sequential processes. In due course, photoelectron spectroscopic studies on the two-photon double ionization of Ne [6, 7] and Ar [8] atoms have appeared, in which the sequential processes, two discrete photolines corresponding to the first and second steps, have been obviously demonstrated. And also a general expression for the angular correlation function of the two photoelectrons emitted in the sequential ionization has been derived [9]. On one hand, in the previous studies [6–8], photon energies higher than the first ionization potential of the singly charged ion were selected. On the other hand, if one selects the photon energies of resonance excitation of the singly charged ions, one can expect three-photon double ionization via the resonance states of the intermediate singly charged ions. This approach enables one to study the resonance effect of photoionization in intense light. It should be noted that this kind of research is limited to the very recent publications [10, 11] and theoretical works [12–16] up to now as far as we know. In the previous works [10, 11], however, the mechanism of the resonant two-photon ionization process and the laser intensity dependence of the ionization rate have not been investigated.

In this context, we have performed photoelectron spectroscopic studies for a sequential three-photon double ionization of the Ar atom by an extreme ultraviolet free-electron laser (EUV-FEL). Namely, first the Ar atom is singly ionized by the first photon in an EUV-FEL pulse:

\[ h\omega_1 + A(J_0) \rightarrow A^+(J_i) + e_1(\epsilon_1 \ell_1 j_1) \]  \hspace{1cm} (1)

with the emission of the first electron \( e_1 \). The energy of this electron is determined by the energy conservation, \( \epsilon_1 = h\omega_1 - E_1 \), where \( E_1 \) denotes the ionization energy of the neutral atom. Then, in the subsequent second step, the singly charged ion is resonantly excited by the second photon in the same pulse:

\[ h\omega_2 + A^+(J_i) \rightarrow A^{**}(J_f) \]  \hspace{1cm} (2)

Last, in the subsequent third step, the resonantly excited ion is ionized by the third photon in the same pulse:

\[ h\omega_3 + A^{**}(J_f) \rightarrow A^{3+}(J_f) + e_2(\epsilon_2 \ell_2 j_2) \]  \hspace{1cm} (3)

The energy of this second electron is \( \epsilon_2 = h\omega_3 - (E_2 - E_r) \), where \( E_2 \) and \( E_r \) are the ionization and resonance energies of the singly charged ion, respectively. The atomic and ionic states in (1)–(3) are characterized by the total angular momenta of \( J_0, J_1, J_f \) and \( J_f \) for the initial, lower ionic, excited ionic and final states, respectively. Here the photon energy \( h\omega \) of EUV-FEL is denoted by \( h\omega_1 \) in (1), \( h\omega_2 \) in (2) and \( h\omega_3 \) in (3), i.e. the suffixes are merely used to distinguish the sequential processes. In this communication, we report the experimental results on photoelectron spectroscopic studies for a sequential three-photon double ionization of the Ar atom when the photon energy \( h\omega \) of EUV-FEL was tuned to the resonance energies \( E_r \) of the ion in (2).

The present experiment has been performed at the focused beam line of the Spring-8 Compact SASE Source (SCSS) [17, 18]. A linearly polarized EUV-FEL light pulse with the repetition rate of 20 or 30 Hz and the pulse duration \( \Delta t \approx 100 \text{fs} \) was focused onto a 25 × 25 \( \mu \text{m} \) or 13 × 14 \( \mu \text{m} \) spot within the collimated supersonic gas jet in the centre of the vacuum chamber. The maximum light pulse energy was 30 \( \mu \text{J} \) with the root-mean-squared fluctuation of \( \sim 10\% \) at (21.65 ± 0.2) eV, so that the maximum light intensity was estimated to \( 3.5 \times 10^{13} \text{ W cm}^{-2} \) from the pulse duration, spot size and reflectivity of focusing mirrors. Here 0.2 eV refers to the standard deviation times 2 for fluctuations of the photon energy, see figure 3. Photoelectrons emitted along the electric vector of the incident light were detected by the TOF spectrometer with 1/3000 of the total 4\( \pi \) solid angle under the field-free condition, which was built on the idea of Hemmers et al [19]. Electron pulses from a Chevon arrangement of micro-channel plates equipped with the TOF were registered directly by a 7.25 GHz digital oscilloscope, which was triggered by pulses generated by a delay unit connected to the master oscillator of SCSS.

Photoelectron spectra of Ar measured at four different photon energies of (21.4 ± 0.2), (21.65 ± 0.2), (23.8 ± 0.2) and (24.6 ± 0.2) eV are shown in figure 1. The fine structure of the total angular momentum, \( J_i, J_f \), and \( J_f \), was not resolved. The sequential two-photon double ionization of Ar is energetically forbidden at the present photon energies, because the threshold energy of the second step for the sequential process is 27.6 eV (see figure 2). The non-sequential two-photon double ionization of Ar, which has the threshold energy of 43.4 eV, is energetically forbidden at the photon energy of 21.4 and 21.65 eV, but allowed at the photon energies of 23.8 and 24.6 eV. The excess energy of 4.2 and 5.8 eV is shared by two electrons at 23.8 and 24.6 eV, respectively. Therefore, two electron continuum contributions might be expected in the lower energy side of the Ar 3p photoline, although they were not observed in the present light intensity regime, \( I_{\text{ph}}(h\omega) < 3.5 \times 10^{13} \text{ W cm}^{-2} \). The direct two-photon single ionization of \( \text{Ar}^+ \) is energetically allowed. However, such an ionization process may be ruled out, because it was reported, in the case of Ne\( ^+ \) [3], that the cross section of the direct two-photon process is approximately one order smaller than that of the resonant two-photon single ionization. In fact, at the off-resonant photon energy below 21.4 eV we could not detect photoelectrons due to the direct two-photon single ionization of \( \text{Ar}^+ \).

Then, photoelectron peaks except for the Ar 3p photoline in figure 1 are assigned to a sequential three-photon double ionization via the resonance states of the intermediate \( \text{Ar}^+ \) ion: \( (1) \rightarrow (2) \rightarrow (3) \) (see figure 2). Candidates of the resonances at 23.8 and 24.6 eV are summarized in table 1, taking into account the band pass of the EUV-FEL light. Considering the
Figure 1. Photoelectron spectra of Ar irradiated by FEL for four mean photon energies: (a) 21.4, (b) 21.65, (c) 23.8 and (d) 24.6 eV. Each spectrum was obtained by $10^5$ FEL shots. Shaded peaks are due to the two-photon single ionization via the Ar$^+$ resonance states (see figure 2).

In contrast to the cases at 23.8 and 24.6 eV, at the photon energy of 21.4 eV the photoelectron peak corresponding to the third step (3) splits into three lines having the kinetic energies of 10.9, 13.6 and 15.3 eV (see figure 1(a)), which are assigned to the ionic states of Ar$^{2+}$ 3p$^4$ (1S), 3p$^4$ (1D) and 3p$^4$ (3P), respectively. At the photon energy of 21.65 eV the photoelectron peak corresponding to the third step (3) splits into three lines having the kinetic energies of 11.4, 14.2 and 15.9 eV (see figure 1(b)), which are assigned to the ionic states of Ar$^{2+}$ 3p$^4$ (1S), 3p$^4$ (1D) and 3p$^4$ (3P), respectively.

This significant difference on the appearance of the photolines due to the third step (3) comes from the nature of the resonance states of the many-electron systems with open sub-shells. In general, their electronic states are linear combinations of different open sub-shell terms; they are mainly 3p$^4$ 1S, 1D and 3P in the case of Ar$^+$ 3p$^4$ 3d $^2$D states. When the interaction between the sub-shells is significant, the mixing between the sub-shell terms becomes substantial and the states are required to be represented in terms of multi-configurations. However, the spectroscopic terms themselves are, normally, identified in terms of their single leading configurations in convention, which may sometimes prevent us from understanding the
Table 1. Excitation energy of the Ar⁺ resonance states from the Ar⁺ 2P½, 1/2 states. The optical transitions of ΔJ = 0 and ±1 are listed. The spectral term and level energy are taken from NIST [20].

<table>
<thead>
<tr>
<th>Configuration</th>
<th>Term</th>
<th>J</th>
<th>Energy from ²P½ (eV)</th>
<th>Energy from ²P½ (eV)</th>
<th>Photon energy (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3p⁴ (⁴P) 5p</td>
<td>²D</td>
<td>5/2</td>
<td>23.620</td>
<td>23.620</td>
<td>23.620</td>
</tr>
<tr>
<td>3p⁴ (⁴P) 5p</td>
<td>²S</td>
<td>1/2</td>
<td>23.682</td>
<td>23.505</td>
<td>23.682</td>
</tr>
<tr>
<td>3p⁴ (⁴P) 5p</td>
<td>⁴S</td>
<td>3/2</td>
<td>23.701</td>
<td>23.496</td>
<td>23.701</td>
</tr>
<tr>
<td>3p⁴ (⁴S) 4p</td>
<td>²P</td>
<td>3/2</td>
<td>23.801</td>
<td>23.624</td>
<td>23.801</td>
</tr>
<tr>
<td>3p⁴ (⁴P) 4d</td>
<td>²D</td>
<td>5/2</td>
<td>23.873</td>
<td>23.669</td>
<td>23.873</td>
</tr>
<tr>
<td>3p⁴ (⁴P) 6s</td>
<td>⁴P</td>
<td>5/2</td>
<td>24.649</td>
<td>24.512</td>
<td>24.649</td>
</tr>
<tr>
<td>3p⁴ (⁴D) 4d</td>
<td>²P</td>
<td>1/2</td>
<td>24.727</td>
<td>24.550</td>
<td>24.727</td>
</tr>
<tr>
<td>3p⁴ (⁴D) 4d</td>
<td>²D</td>
<td>5/2</td>
<td>24.756</td>
<td>24.617</td>
<td>24.756</td>
</tr>
<tr>
<td>3p⁴ (⁴P) 5d</td>
<td>⁴D</td>
<td>5/2</td>
<td>24.791</td>
<td>24.668</td>
<td>24.791</td>
</tr>
<tr>
<td>3p⁴ (⁴P) 6s</td>
<td>²P</td>
<td>3/2</td>
<td>24.800</td>
<td>24.623</td>
<td>24.800</td>
</tr>
<tr>
<td>3p⁴ (⁴P) 6s</td>
<td>²F</td>
<td>5/2</td>
<td>24.825</td>
<td>24.696</td>
<td>24.825</td>
</tr>
</tbody>
</table>

Figure 3. Oscillator strength distribution for the 3p→3d excitations of the Ar⁺ ion. Solid and dashed bars: oscillator strength by the Coulomb gauge, i.e. velocity form calculation. Squares and circles: oscillator strength by the Babushkin gauge, i.e. length form calculation. Positions of solid bars and squares correspond to the excitation energies from the Ar⁺ 3s²3p²P½/3½, and positions of dashed bars and circles to those from the Ar⁺ 3s²3p²P½/3½. Numbers are taken from level numbers of table 2. Thin solid curves centred at 21.4 and 21.65 eV express the FEL spectrum averaged over 50 shots.

The 3p→3d excitation of Ar⁺ has attracted considerable attention from theorists for more than 30 years [21, 22], because the 3d orbital is partially collapsed in the singly charged ion, and therefore electron correlation plays the inevitable role to understand the resonance state. The 3d electron that resides in the vicinity of the 3p⁴ electrons in Ar⁺ 3p⁴3d mediates the interaction between the 3p⁴ sub-shell terms. As described in [22], the configuration p⁴d includes three ²D basis states, which are lead from three p⁴ parents 3P, 1D and 1S. Following this theoretical interpretation, the 3p⁴3d resonance state can be expressed by the multi-configurations with ²D symmetry:

\[
|3p⁴ 3d ²D⟩ = a|3p⁴(⁴P) 3d ²D⟩ + b|3p⁴(⁴D) 3d ²D⟩ + c|3p⁴(⁴S) 3d ²D⟩, \tag{4}
\]

where a, b and c are the mixing coefficients of the configurations.

To obtain precise and quantitative information on the multi-configuration character of the resonance states, we have made a large-scale multi-configuration Dirac–Fock (MCDF) calculation using GRASP92 [23] and RATIP [24] programs for the states of Ar⁺ that have 1s²2s²2p⁶3s²3p³d as their leading configuration. For the Ar⁺ 3p⁴3d configuration, there are a number of theoretical studies in the literature [25–28]. They have demonstrated the importance of the strong configuration interactions between the 3p⁴3d and the excited state configurations. The Ar⁺ 3p⁴3d configuration state consists of 28 multiplets [20], and they spread almost 6.5 eV in level energies. The 3d orbital may be expected to be loosely bound for higher energy components of the multiplets compared to the ones for lower energy components. To describe these features properly, one can consider the inclusion of the atomic orbitals with higher principal quantum numbers in the procedure of the MCDF optimizations. Then, we have carried out an MCDF calculation under the extended optimized level option including all the orbitals with the principal quantum number n = 1, 2, 3 and 4. In the MCDF optimization procedure, all the possible single- and double-electron excitations from the 3s, 3p and 3d sub-shells are taken into account. The total number of the configuration state functions (CSF) was 26141. We have made a similar MCDF calculation for the Ar⁺ ground state 3p⁴ ²P½/3½ doublet considering the possible single- and double-electron excitations from the 3s, 3p and 3d sub-shells to 3d, 4s and 4f orbitals. Subtracting the ground state 3p⁴ ²P½/3½ energy from the excited state energies, we have obtained all the 28 excitation energies for the levels that belong to the Ar⁺ 3p⁴3d multiplets. The differences of the calculated values from the experimental ones [20] are 0.09 eV for the lowest energy term 3s²3p⁴ (⁴P) 3d ²D½/₁½, and 1.06 eV for the highest energy term 3s²3p⁴ (⁴D) 3d ²S½/₁½. The results of the present MCDF calculations are shown in table 2. In this table, the leading 4 configurations of the 11 major terms out of the total 28 multiplets are listed together with the term designations and their level energies in [20]. In the entry designated as 3p⁴ (⁴D) 3d ²D½/₁½ at 21.428 eV in [20], for example, we find that the level has substantial contributions from all of the 3p⁴ ¹S, ¹D and ³P sub-shell terms although the leading configuration remains to be 3p⁴ (⁴D) 3d with 38.7% in percentage fraction.

We have further made the calculation of dipole oscillator strengths for excitations from the 3s²3p²P½/₁½ ground states. Figure 3 shows the calculated distribution of weighted oscillator strength gf values, which are the oscillator strengths.
f multiplied by the statistical weights \( g \) of the lower state of transitions. In the present case, \( g = 4 \) and 2 for excitations from \( 3s^23p^52p_2 \) and \( 3p_1 \), respectively. As can be seen from figure 3 and table 2, at the photon energy of 21.4 \( \pm \) 0.2 eV, the four resonance states, \( 3s^23p^4(1D)3d^2D \) and \( 3s^23p^4(1D)3d^2P \) of \( Ar^+ \), are contributing to the resonant two-photon process. Considering the term mixing in these four excited states, numbered from 5 to 8, as found in table 2, we may naturally expect the simultaneous creation of the 3s23p4 core configurations, \( 1 \) and \( 3 \) states through a sudden removal of the 3d electron in the third step (3) from the \( Ar^+ \) 3p43d resonance to the resonant two-photon process (see figure 3 and table 2), as well. Then, we may also expect the simultaneous creation of the final Ar+ 3p5 1S, 1D and 3P states, which is consistent with the observation of three photoline from \( Ar^+ \) in figure 1(a). This scenario will be verified if the sudden approximation works for the photoionization of the intermediate \( Ar^+ \) resonance states. At the photon energy of 21.65 \( \pm \) 0.2 eV, the four resonance states of \( 3s^23p^4(1D)3d^2D \) and \( 3s^23p^4(1D)3d^2P \) of the intermediate \( Ar^+ \) are contributing to the resonant two-photon process (see figure 3 and table 2), as well. The authors in [11] have argued that \( Ar^+ \) 3p5(1D) 3d2P is realized by Ar+ 3p5(1D) 3d2D whereas Ar+ 3p5(S) and 1D are realized by Ar+ 3p5(1D) 3d2P resonances by investigating their mean photon energy dependence of the photoelectron intensities. However, it may be worthy to point out that the substantial contribution of the Ar+ 3p5(1D) 3d2P resonance to the creation of Ar+ 3p5(S) might not be probable because, as found in table 2, the Ar+ 3p5(1D) 3d2P states cannot have any 3p5(S) components in the CSF expansion of the states due to the symmetry constraint.

Generally speaking, the MCDF method is one of the variational calculations to only the energy eigenvalues, and therefore it is quite insensitive to the change of the mixing coefficients of the core electron configurations, i.e. the calculated percentage fraction of the \( 3s^23p^5 \) core configurations of \( Ar^+ \) could contain some error even though we obtain a variational convergence out of the numerical calculation. And further on, we have to point out that this percentage distribution depends strongly on the 3d orbital shape, which may vary depending on the way of orbital optimization. Nevertheless, based on the results by the MCDF calculations we can conclude that the intermediate \( Ar^+ \) 3s23p4(1D) 3d2D and 3s23p4(1D) 3d2P states excited by the photon energy of 21.4 \( \pm \) 0.2 and 21.65 \( \pm \) 0.2 eV contain appreciably different \( 3s^23p^4 \) core configurations and that the experimental observation of split photoelectron spectral peaks is the reflection of the multi-configuration nature of the resonance states.

It should be noted that the present normalized intensity ratio of three photoline at 21.4 eV, \( I(1S) = 67\% : I(1D) = 24\% : I(2P) = 9\% \), is quite different from that of the recent results: 32\% : 40\% : 28\% [11]. The disagreement may be due to the different photoelectron angular distribution for each photoline of the three [9], because the former is the differential measurements of photoelectrons along the polarization vector of an EUV-FEL light but the latter is the integrated measurements of photoelectrons by a magnetic bottle spectrometer.

The number of photoelectrons \( I(1) \) generated per light pulse in the first step (1) is expressed by an ordinary exponential saturation function [29]

\[
I(1) = N(1 - \exp(-\sigma(1)H_{ph})),
\]

where \( N \) is the initial number of \( Ar \) atoms within the interaction volume, \( H_{ph} \) is the photon number per pulse and unit area and \( \sigma(1) \) is the photoionization cross section for the first step (1). Here we used the obvious relation that the number of photoelectrons \( I(1) \) is equal to the number of \( Ar^+ \) ions. The relevant experimental result is shown in figure 4. The light intensity \( I_{ph} = H_{ph} \omega / \Delta t \) was controlled by the pressure of the \( Ar \) gas attenuator installed in the beam line of SCSS. Equation (5) is well fitted to the result \( I(1) \) at 21.65 eV, which is the number of \( Ar \) 3p photoelectrons detected at the parallel direction to the electric vector of the incident light. This supports that the photoelectron angular distribution relative to the electric vector is independent of the \( I_{ph} \) over the whole regime of the present light intensity. Moreover, the good description of the intensity dependence by equation (5) implies.
that the change of the interaction volume on the intensity was not detectable in the present intensity regime, because the constant volume was assumed in equation (5) [29]. On the other hand, according to lowest-order perturbation theory, the ion yield ratio of \( \frac{N(\text{Ar}^2\text{+})}{N(\text{Ar}^+)} \), in which \( \text{Ar}^2\text{+} \) is produced through the two-photon single ionization of \( \text{Ar}^+ \), is written by

\[
\frac{N(\text{Ar}^2\text{+})}{N(\text{Ar}^+)} = \frac{\sigma^{(2)}}{2 \Delta \tau},
\]

where \( \sigma^{(2)} \) is the two-photon single-ionization cross section [3]. Figure 5 shows the experimental results of \( \frac{I(e_2)}{I(e_1)} \) at 21.4, 21.65 and 23.8 eV together with fits according to the power law of \( I^{\alpha}_{\text{ph}} \). The ratio should depend on the square of \( I_{\text{ph}} \), if the lowest-order perturbation theory is applicable to the present case. Contrary to this prediction, the experimental results, \( I_{\text{ph}}^{0.56} \) for 21.4 eV, \( I_{\text{ph}}^{0.74} \) for 21.65 eV and \( I_{\text{ph}}^{0.41} \) for 23.8 eV, strongly deviate from the power law of \( I_{\text{ph}}^{2} \). Furthermore, as can be seen from figures 5(a)–(c), the light intensity dependence of the resonant two-photon single-ionization rate, \( I(e_2)/I(e_1) \), depends on not only the light intensity but also the inherent oscillator strength for the excitation into the relevant resonance of \( \text{Ar}^+ \); the power \( \alpha \) value, the slope in the log–log plot, changes from 0.56 and 0.74 for the \( 3p \rightarrow 3d \) transition to 0.41 for the weaker \( 3p \rightarrow 4d \) Rydberg transition. This implies that Rabi oscillation between the ground and resonance states of \( \text{Ar}^+ \) during the EUV-FEL pulse duration plays the essential role to determine the resonant two-photon single-ionization rate, because the period of the Rabi oscillation frequency depends on both the electric dipole transition moment and electric field strength of the EUV-FEL pulse. Although time-independent non-perturbative theory such as the Floquet approach [12–16] cannot describe a time-dependent dynamical picture like the Rabi oscillation, it may explain the observed deviation from the \( I_{\text{ph}}^{2} \)-scaling.

It should be noted that Sorokin et al [3] measured the two-photon single-ionization rate of \( \text{Ne}^+ \) by the ion TOF technique at 38.4 eV and found the power law of \( I_{\text{ph}}^{2} \) for the process under the light intensity \( \sim 3 \times 10^{13} \text{ W cm}^{-2} \). And they suggested a two-photon single-ionization process of \( \text{Ne}^+ \) via the \( \text{Ne}^+ \) 2p–4s resonance to explain the enhanced ionization rate, although the photon energy of 38.4 eV was detuned by 0.2 eV from the resonance. After this work, Hamonou et al [16] explained theoretically the enhanced ionization rate due to the \( 2p^3(1P^o)nd 3D^o \) resonances, which significantly enhance ionization through the emission of a 2s electron. Recently the same group found that at 38.4 eV, the two-photon single-ionization rate of \( \text{Ne}^+ \) between \( 2 \times 10^{13} \) and \( 10^{14} \text{ W cm}^{-2} \) deviates from the \( I_{\text{ph}}^{2} \)-scaling by the time-dependent \( R \)-matrix theory, and pointed out the significant influence of resonances on the ionization process [30]. It can be mentioned in the analogue of the present work that by the ion TOF technique, Martins et al [31] measured the resonant two-photon single-ionization rate of \( \text{Ne}^+ \) ion via \( \text{Ne}^+ 2s^22p^3(1D^o)4n^2 \text{F}_2 \) Rydberg states and found that a resonance enhancement of the \( 2s^2 \) to \( \text{Ne}^+ \) ratio increases by a factor of 2, although they did not report the laser intensity dependence of the ratio.

In summary, we have presented the photoelectron spectra of \( \text{Ar} \) due to the three-photon double ionization. When
the photon energy of the EUV-FEL has been tuned to the resonances of the intermediate Ar+, photoelectron peaks due to the resonant two-photon single ionization of Ar+ have been observed. And we have revealed that the appearance of the peaks reflects the multi-configuration character of the resonance states. Furthermore, we have shown that the laser intensity dependence of the photoelectron signals due to the resonant two-photon single ionization of Ar+ deviates significantly from the $I^2$-scaling, and then suggested that such non-perturbative phenomena might be caused by the Rabi oscillation between the ground and resonance states of Ar+.

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