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研究課題名(和文) Post Collision Interaction and photoelectron recapture upon atomic and molecular photoionization

研究課題名(英文) Post Collision Interaction and photoelectron recapture upon atomic and molecular photoionization

研究代表者

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研究成果の概要(和文)：内外の放射光実験施設において真空紫外線およびソフトx線を用いて希ガス原子の内殻光イオン化過程におけるオージェ電子分光測定実験を行った。平行して理論計算を進めた。特に光イオン化閾値近傍における衝突後相互作用と光電子再捕獲がオージェ電子スペクトルの位置及び形状に大きな変化を与える現象を追求した。従来衝突後効果は動径方向の相互作用によってのみ生ずると考えられていたが本研究によって角度相関が非常に重要なケースがあることが明らかになった。また多段階オージェ過程における段階の時間依存性についても衝突の効果を利用して解析することができた。

研究成果の学術的意義や社会的意義

衝突後効果および光電子再捕獲は、量子力学の最も重要な未解決問題の1つである3体クーロン連続状態問題の顕著かつ典型的な例であるといえる。その意味で新たな知見を得る事は物理学の基礎研究として非常に重要である。また、今回100アト秒オーダーの時間分解測定に相当する結果を自由電子レーザーや超超高検出技術を用いることなく在来技術による実験データから衝突の効果解析することによって得ることが可能であることが示された。これは、今後の超高速原子分子過程の研究のために有用である。

研究成果の概要(英文)：Investigations of post collision interaction effects were pursued with experiments conducted at synchrotron radiation facilities including SOLEIL in France and SPring-8 in Harima. Measurements were performed by auger electron spectroscopy with high-resolution hemispherical electron analyzers. Very interesting results were obtained on three fronts including 1. The discovery of conjugate auger recapture peaks dominating the spectrum, indicating the exchange of angular momentum between auger electron and photoelectron through postcollision interaction effect. 2. Large variations of angular distributions of auger electrons depending on the recapture Rydberg orbital. 3. New insight into the time-dependent dynamics of multistep auger processes including fluorescence, in effect leading to an effective clock mechanism. In addition, new photoelectron correlation set lights were discovered and analyzed in the case of krypton.

研究分野：Atomic and molecular physics

キーワード：PCI Synchrotron Radiation Photoionization Inner-shell Photoelectron recapture

様式 C-19、F-19-1、Z-19 (共通)

1. 研究開始当初の背景

- (1) The phenomenon of atomic postcollision interaction was discovered and pursued in the 1980s through electron impact ionization studies of atoms. Although it was a very basic subject without any immediate applications, it was recognized as an important research theme relating to the three-body coulomb continuum problem in quantum mechanics, one of the most important unsolved problems in physics. Research in this area gained momentum particularly in the atomic photoionization context with the advent of high brilliance synchrotron radiation facilities. However, by the early 2000s, research activities entered a somewhat dormant stage after the leveling off of the experimental capabilities and the successful establishment of theoretical interpretation based on radial interactions.
- (2) In 2015, we published our result from a chance discovery of beautiful Rydberg series structures manifested on the Auger spectrum profiles made at Berkeley ALS [reference below]. This was due to the recapture of photoelectrons into Rydberg orbital's of the Ionic core caused by postcollision interactions. The spectral pattern was found to resemble the postcollision interaction profile much better than the shakeup/shakedown patterns observed in Auger resonant Raman effects. Thus we produced the following publication and also decided that this theme warrants much further investigation.

“PCI effects and the gradual formation of Rydberg series due to photoelectron recapture, in the Auger satellite lines upon Xe $4d^{-1/2}$ photoionization”

Satoshi Kosugi, Masatomi Iizawa, Yu Kawarai, Yosuke Kuriyama, A L David Kilcoyne, Fumihiro Koike, Nobuhiko Kuze, Daniel S Slaughter and Yoshiro Azuma
2015 *J. Phys. B: At. Mol. Opt. Phys.* **48** 115003

2. 研究の目的

- (1) To elucidate the near threshold atomic photoionization process involving postcollision interaction and photoelectron recapture. Particular attention paid to effects that transcend the previous radial interaction model. Also investigate the role of multi step dynamics and fluorescence involved in the process.
- (2) Provide interpretation for the above utilizing theoretical calculations.

3. 研究の方法

- (1) Angle resolved Auger electron and photoelectron spectroscopy measurements utilizing high resolution beamlines at synchrotron radiation facilities including the Photon Factory, UVSOR, Spring8, and SOLEIL.
- (2) Theoretical calculations utilizing the multi-configuration Dirac-Fock method.

4. 研究成果

- (1) “Fluorescence time delay in multi-step Auger decay as an atomic clock”

S. Kosugi, F. Koike, M. Iizawa, M. Oura, T. Gejo, K. Tamasaku, J. R. Harries, R. Guillemin, M. N. Piancastelli, M. Simon, and Y. Azuma

Phys. Rev. Lett. 124, 183001 (2020) - Published 8 May 2020

Differences in post-collision interaction (PCI) effects on Kr $L_3M_{4,5}M_{4,5}$ Auger electron spectra were observed, depending on whether the initial photoionization occurred slightly above the K threshold or slightly above the L_3 threshold. For the former, KL fluorescence emission most likely happens and then Auger processes due to the L_3 hole follow. The time delay due to fluorescence causes a reduced shift of the Auger peak and tailing toward lower energy, since the Auger overtaking of the photoelectron happens later in time and at a location farther away from the ionic core, compared to the case for the simple 1-step $L_3M_{4,5}M_{4,5}$ Auger decay after L -shell photoionization. Time-dependent theory for PCI in multi-step processes agrees well with experiment, illustrating the effect as an atomic clock for the time-sequence of the dynamical process.

(2) “Dominance of angular momentum exchange in the PCI recapture of photoelectrons revealed by high resolution Auger electron measurements of Kr”

S Kosugi, N Suzuki, N Kumagai, H Iwayama, E Shigemasa, F Koike and Y Azuma
2019 *J. Phys. B: At. Mol. Opt. Phys.* **52** 245002

An unexpectedly prominent angular momentum exchange effect in the double photoionization continuum was identified through the photoelectron recapture process induced by post-collision interaction (PCI). The Kr ($M_5 N_{2,3} N_{2,3}$) Auger electron spectrum originating from $3d^{-1} 5/2$ photoionization was measured with the photon energy tuned very close above the ionization threshold. As the photon energy approached the $3d^{-1} 5/2$ photoionization threshold, Rydberg series structures including several angular momentum components were formed within the Auger peak profile by the recapture of the photoelectrons into high-lying final ion orbitals. Our spectrum with line-width much narrower than the lifetime width of the corresponding core excited state allowed us to resolve detailed structures due to the orbital angular momentum states very clearly. Unexpectedly, conjugate peaks originating from the exchange of angular momentum between the photoelectron and the Auger electron through PCI were found to dominate the spectrum. Comparison with valence photoelectron spectrum of Kr via direct photoionization confirms the new assignment. Conjugate peaks were also found to be in accord with the quantum defect values obtained for the high Rydberg series $Kr^+4p^{-2}(^1S_0, ^1D_2)ml$. Extension of the new assignments to photon energies below threshold may call for the revision of previous interpretations of resonance Auger processes.

(3) “Conjugate photoelectron recapture peaks in the high resolution Auger electron spectra following near-threshold Ar 2p photoionization”

S Kosugi, J Martins, F Hosseini, T Marchenko, O Travnikova, J D Bozek, K Ito, E Sokell, M N Piancastelli, M Simon, F Koike and Y Azuma
2020 *J. Phys. B: At. Mol. Opt. Phys.* **53** 125001

We have observed formation of Rydberg series superimposed on the Ar ($L_3 M_{2,3} M_{2,3}$) Auger electron spectral profile with photoexcitation just above the $2p^{-1} 3/2$ photoionization threshold. This is due to the recapture of photoelectrons induced by post-collision interaction between the Auger electron and photoelectron: $Ar^+ 2p^{-1} 3/2 + e^- \rightarrow Ar^+ 3p^{-2} ml + e^-$. The intensity distribution of the angular momentum state resolved Rydberg peaks revealed prominent conjugate peaks due to angular momentum exchange between photoelectrons and Auger electrons in the process. In addition, we

measured the $2p^{-1} \text{ }_{3/2} \text{ } nl \rightarrow 3p^{-2} \text{ } ml + e^{-}$ resonant Auger spectra with high resolution below the $2p^{-1} \text{ }_{3/2}$ threshold. Our results showed that conjugate shake-up/down $2p^{-1} \text{ }_{3/2} \text{ } nd \rightarrow 3p^{-2} \text{ } mp + e^{-}$ processes are stronger than the direct $2p^{-1} \text{ }_{3/2} \text{ } nd \rightarrow 3p^{-2} \text{ } ms, (m-1)d + e^{-}$ processes in the spectator Auger processes, implying a strong angular momentum exchange effect between photoelectron and Auger electron.

(4) “Strong configuration interaction in the 3p photoelectron spectrum of Kr”

S. Kosugi, F. Koike, T. Nagayasu, F. Hosseini, J. Martins, T. Marchenko, O. Travnikova, M. Oura, T. Gejo, J. R. Harries, J. D. Bozek, K. Ito, E. Sokell, S. Fritzsche, M. N. Piancastelli, M. Simon, and Y. Azuma Phys. Rev. A 101, 042505 – Published 8 April 2020

We measured the Kr photoelectron spectrum in the region close to the 3p ionization threshold. Our high-resolution measurements allowed a clear observation of spectral structures due to electron correlation effects. Analysis based on relativistic multiconfiguration calculations could explain these observed peaks as due to strong configuration interactions between the $3p^{-1}$ state and $3d^{-2}nl$ states. Calculated and experimental data for peak assignments and intensity distributions are in good agreement. In addition, we measured the anisotropy parameter β , which also agreed well with theory. These findings provide a detailed view of strong configuration interactions between the $3p^{-1}$ and $3d^{-2}nl$ inner-shell hole states.

(5) “Angle dependent PCI shifts in the Xe 4d Auger electron spectrum with photoelectron recapture”

S. Kosugi, N. Kumagai, N. Suzuki, F. Hosseini, J. Martins, T. Marchenko, O. Travnikova, R. Guillemin, M. N. Piancastelli, M. Simon, H. Iwayama, E. Shigemasa, J. D. Bozek, K. Ito, E. Sokell, F. Koike, and Y. Azuma

(Submitted to Phys Rev A)

Angle-dependent shift was found in the Xe $N_5 O_{23} O_{23} (1S_0)$ Auger electron spectral profile close to the $4d_{5/2}$ photoionization threshold. High resolution measurements allowed the observation of Rydberg series with peak intensity distribution manifesting shift and tailing towards higher energy due to PCI (post-collision interaction). Very pronounced differences in this pattern were observed depending on whether the incident photon polarization was perpendicular or parallel with respect to the detector. Our measurements also yielded the anisotropy parameter β_{ml} for $Xe^+ 5p^2(1S_0) ml$ high Rydberg ions differential with respect to the ion angular momentum state. The result challenges previous theoretical studies assuming that PCI does not depend on the angle with respect to the photon polarization.

(6) “PCI effects with multi-step Auger transitions following Ar 1s photoionization”

S. Kosugi, R. Guillemin, O. Travnikova, R. Püttner, D. Céolin, L. Journel, M. N. Piancastelli, F. Koike, S. Sheinerman, Y. Azuma, and M. Simon

(Submitted to Phys Rev A)

The Post-Collision interaction (PCI) effect, involving multi-step decay processes following Ar 1s photoionization has been studied by Auger electron spectroscopy. High resolution synchrotron beam was scanned over the Ar 1s photoionization threshold. Processes studied and compared included 1) $Kr^+ 2p^1 \rightarrow Kr^{2+} 3p^2 LMM$ Auger process due to a single L hole created by KL fluorescence, 2) $Kr^{2+} 2p^2 \rightarrow Kr^{3+} 2p^1 3p^2 LMM$ Auger process following double L shell hole states

produced by *KLL* Auger processes and 3) the subsequent single *L* hole state leading to $\text{Kr}^{3+} 2p^1 3p^2 \rightarrow \text{Kr}^{4+} 3p^4$ *MMM* Auger transitions. Particularly pronounced PCI shift and unusual line shape compared to ordinary one step PCI process were found in the spectrum of Auger process following the *KLL* Auger process. In addition, we have conducted theoretical calculations corresponding to these PCI effects in multi-step processes. Good agreement was found between the measured and calculated PCI shifts (and shape?).

5. 主な発表論文等

〔雑誌論文〕 計4件（うち査読付論文 4件/うち国際共著 3件/うちオープンアクセス 0件）

1. 著者名 S. Kosugi, F. Koike, M. Iizawa, M. Oura, T. Gejo, K. Tamasaku, J. R. Harries, R. Guillemin, M. N. Piancastelli, M. Simon, and Y. Azuma	4. 巻 124
2. 論文標題 Fluorescence Time Delay in Multistep Auger Decay as an Internal Clock	5. 発行年 2020年
3. 雑誌名 Physical Review Letters	6. 最初と最後の頁 183001
掲載論文のDOI (デジタルオブジェクト識別子) https://doi.org/10.1103/PhysRevLett.124.183001	査読の有無 有
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1. 著者名 S. Kosugi, F. Koike, T. Nagayasu, F. Hosseini, J. Martins, T. Marchenko, O. Travnikova, M. Oura, T. Gejo, J. R. Harries, J. D. Bozek, K. Ito, E. Sokell, S. Fritzsche, M. N. Piancastelli, M. Simon, and Y. Azuma	4. 巻 101
2. 論文標題 Strong configuration interaction in the 3 p photoelectron spectrum of Kr	5. 発行年 2020年
3. 雑誌名 Physical Review A	6. 最初と最後の頁 42505
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3. 雑誌名 Journal of Physics B	6. 最初と最後の頁 125001
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1. 著者名 S Kosugi, N Suzuki, N Kumagai, H Iwayama, E Shigemasa, F Koike and Y Azuma	4. 巻 52
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1. 発表者名 小杉 聡
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3. 学会等名 第32回日本放射光学会年会放射光科学合同シンポジウム
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1. 発表者名 Satoshi Kosugi
2. 発表標題 Angular dependence of the PCI following inner-shell photoionization
3. 学会等名 International Workshop on Post-collision Interaction (招待講演) (国際学会)
4. 発表年 2018年

1. 発表者名 Fumihiro Koike
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3. 学会等名 International Workshop on Post-collision Interaction (招待講演) (国際学会)
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1. 発表者名 Yoshiro Azuma
2. 発表標題 Electrons playing tag upon photoionization
3. 学会等名 7th Topical Conference of the Indian Society of Atomic and Molecular Physics (招待講演) (国際学会)
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〔図書〕 計0件

〔産業財産権〕

〔その他〕

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6. 研究組織

	氏名 (ローマ字氏名) (研究者番号)	所属研究機関・部局・職 (機関番号)	備考
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