Observation of Electric Octupole Emission Lines Strongly Enhanced by an Anomalous Behavior of Cascading Contribution

Hiroyuki A. Sakaue,1 Daiji Kato,1,2 Izumi Murakami,1,3 Hayato Ohashi,4 and Nobuyuki Nakamura5

1National Institute for Fusion Science, Toki, Gifu 509-5292, Japan
2Department of Advanced Energy Engineering Science, Kyushu University, Fukuoka 816-8580, Japan
3Department of Fusion Science, The Graduate University for Advanced Studies (SOKENDAI), Tokyo, Gifu 509-5292, Japan
4Institute of Liberal Arts and Sciences, University of Toyama, Toyama 930-8555, Japan
5Institute for Laser Science, The University of Electro-Communications, Tokyo 182-8585, Japan

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We present extreme ultraviolet spectra of Ag-like W$^{27+}$ observed with an electron beam ion trap. In the spectra, the $4f_{7/2,5/2} - 5s$ transitions are identified as the first observation of spontaneous electric octupole emission. Our theoretical investigation shows that the emission line intensity is strongly and specifically enhanced at the atomic number 74 by an anomalous behavior of cascading contribution to $5s$ via $5p \leftrightarrow 5d$.

I. INTRODUCTION

Studies of electric dipole ($E1$) forbidden transitions are important not only for testing atomic physics theory describing the interaction of atoms or ions with multipole radiation fields, but also for several applications, such as plasma diagnostics [13] and atomic clocks [4,5]. The transition probability of forbidden transitions is generally too small for the decay to be observed in emission spectra of neutral atoms and low charged ions. However, the probability increases rapidly with the atomic number $Z$ with a strong power law dependence. As a typical example, the transition probability of the $1^{1}S_{0} - 2^{3}S_{1}$ magnetic dipole ($M1$) transition in the He-like iso-electronic system has $Z^{10}$ dependence [6]. Thus observations of forbidden transitions have often been performed for highly charged ions to date. For example, $M1$ transitions in highly charged heavy ions, such as iron and tungsten, are often used for the diagnostics of astrophysical and fusion plasmas [7–9]. Many $M1$ transitions have thus been observed and identified over a wide range of wavelengths so far [8,10]. Electric and magnetic quadrupole ($E2$ and $M2$, respectively) transitions are also often observed in laboratory and astrophysical plasmas. A typical example of $E2$ transition is $[3d^{10}]_{J=0} - [3d^{9}4s]_{J=2}$ in Ni-like ions [11,12] whereas that of $M2$ transition is $1^{1}S_{0} - 2^{3}P_{2}$ (the line often indicated as “$x$”) in He-like ions [13,14].

In contrast, observation of multipole transitions beyond quadrupole is quite limited even for highly charged ions. There is only one example, which is the $[3d^{10}]_{J=0} - [3d^{9}4s]_{J=3}$ magnetic octupole ($M3$) transition in Ni-like ions. It was first observed in the x-ray region for Th$^{62+}$ ($Z=90$) and U$^{64+}$ ($Z=92$) with an electron beam ion trap (EBIT) at Lawrence Livermore National Laboratory (LLNL) [15]. In an EBIT, trapped highly charged ions interact with a low-density (typically $10^{10} - 10^{12}$ cm$^{-3}$) electron beam. The collision frequency is typically in the order of 10 Hz, so that weak forbidden transitions with a transition probability down to $\sim 10$ s$^{-1}$ can be observed [16,18]. The observations with the LLNL EBIT showed that the $M3$ transition in Ni-like ions is directly excited through radiative cascades and can have an intensity comparable to $E1$ transitions depending on electron density. The observation of the $M3$ transition was also conducted for Ni-like Xe$^{26+}$, Cs$^{27+}$, and Ba$^{28+}$ [19]. Time resolved measurements with a microcalorimeter mounted to the LLNL EBIT enabled acquisition of the decay lifetime of the metastable $[3d^{9}4s]_{J=3}$ level.

Observations of an electric octupole ($E3$) transition in an atomic system have been reported for the $2^{3}P_{1/2} - 2^{3}P_{3/2}$ transition in Yb$^{+}$ [20,22]. In their observations, the transition was detected as an excitation driven with a laser. The transition probability is so small ($\sim 10^{-9}$ s$^{-1}$ [21]) that it is practically impossible to detect the emission. There is no observation of spontaneous electric octupole ($E3$) emission so far even for highly charged ions. In this paper, we present the first direct observation of spontaneous $E3$ emission lines performed for Ag-like W$^{27+}$. In an emission spectrum of Ag-like W$^{27+}$ in the extreme ultraviolet (EUV) range observed with an EBIT, two lines are assigned to $4f_{7/2,5/2} - 5s$ transitions, which can be realized by $E3$. We also present the analysis based on collisional radiative (CR) model calculations. The calculated result shows that an anomalous cascading contribution enhances the $E3$ emission intensity strongly and specifically at the atomic number $Z=74$, and hence enables us to observe the emission.

II. EXPERIMENTS

The present experiments were performed using a compact EBIT, called CoBIT [23]. CoBIT mainly consists of an electron gun, an ion trap (drift tube), an electron collector, a superconducting coil, and a liquid nitrogen tank. A high critical temperature superconducting Helmholtz-like coil, which can be used at the liquid nitrogen temperature, is mounted around the drift tube. An electron beam emitted from the electron gun is accelerated (or decelerated) toward the drift tube while being compressed by a magnetic field produced by the superconducting coil. After passing through the drift tube, the electron beam is
collected by the electron collector. In the present study, tungsten was injected into CoBIT as a sublimated vapor of tungsten hexacarbonyl W(CO)₆ through a gas injection system.

Emission from the trapped tungsten ions in the EUV region was observed with a flat-field grazing incidence grating spectrometer with a 1200 grooves/mm aberration-corrected concave grating (Hitachi 001-0660). A back-illuminated charge coupled device (CCD) detector (Princeton Instruments PyLON: X0-2KB) was mounted at the focal position for detecting the diffracted EUV photons. The CCD was cooled at -120 °C by liquid nitrogen for suppressing the dark current. Either aluminum or zirconium foil was placed in front of the grating for examining and removing the contribution of the second-order diffraction. Furthermore, these metal filters cut the stray visible light from the cathode of the electron gun. Wavelength calibration was carried out using well-known emission lines of highly charged Ar and O [24]. The uncertainty in the wavelength calibration was estimated to be less than ±0.02 nm.

III. COLLISIONAL RADIATIVE MODELING

To analyze the experimental spectra, we performed collisional radiative (CR) model calculations. The line intensity of radiative transitions is expressed as the product of the transition probability and the fractional population of the upper level. In the present CR model calculations, the dimension-less fractional population \( n_i \) of the level \( i \) was calculated by the following CR equilibrium equation,

\[
n_i = \frac{n_e \sum_{j \neq i} C_{ij} n_j + \sum_{j > i} A_{ij} n_j}{n_e \sum_{j \neq i} C_{ji} + \sum_{j < i} A_{ji}} = \frac{C_{in} + R_{in}}{F_{in}} = \frac{F_{in}}{F_{out}},
\]

where \( C_{ij} \) and \( A_{ji} \) are the electron impact (de)excitation rate coefficient and the radiative transition rate for the \( i \leftarrow j \) transition, respectively, and \( n_e \) electron density. The electron collision rate coefficients were obtained by assuming the delta function distribution for the electron beam energy in CoBIT. \( F_{in} \) stands for the inflow rate of populations to the level \( i \), and consists of the collisional \( (C_{in} = n_e \sum_{j \neq i} C_{ij} n_j) \) and radiative \( (R_{in} = \sum_{j > i} A_{ij} n_j) \) inflow. \( F_{out} \) stands for the rate of collisional \( (c_{out} = n_e \sum_{j \neq i} C_{ji}) \) and radiative \( (r_{out} = \sum_{j < i} A_{ji}) \) depopulations from the level \( i \), which equals to the corresponding outflow rate divided by \( n_i \). Energy levels, radiative transition probabilities, and distorted-wave excitation and ionization cross sections were calculated with the Hebrew University Lawrence Livermore Atomic Code (HULLAC) [25]. 21,530 fine-structure levels of W⁷⁺ below the ionization threshold (881.4 eV [26]) were obtained by calculations with ⁴d¹⁰f₈, ⁴d¹⁰nl (n = 5 – 6, l < n), ⁴d⁸f², ⁴d⁶f₅l, ⁴d⁸f₃, and ⁴d⁸f₂⁵l configurations.

IV. RESULTS AND DISCUSSION

Figure 1 shows EUV spectra obtained at electron energies of 770, 800, and 870 eV. The lines observed in the wavelength region 9.5 to 10.6 nm and a line at 12.8 nm in the 800eV spectrum were not observed at 770 eV. Thus they should be assigned to W²⁶⁺ considering that the ionization potential of W²⁷⁺ is 784 eV [26]. Comparison with the calculated transition wavelengths shown as the blue vertical lines in the figure indicates that these lines should correspond to ⁴f² – ⁴f₅s and ⁴f₅p – ⁴f₅d transitions. The ⁴f² – ⁴f₅s transitions are strictly forbidden in a single configuration approach, but strongly enhanced by the configuration interaction between the ⁴f₅s and ⁴f₅d levels as discussed by Jonauskas et al. [27]. By increasing the electron energy further to 870 eV, three lines were additionally observed at around 8.9, 9.2, and 12.6 nm. These lines should be assigned to W²⁷⁺ considering the ionization potential of W²⁶⁺ (833 eV) [20]. To identify these lines, the CR model spectrum is shown in the top panel of the figure. As seen in the figure, in this wavelength region, the CR model calculation pre-

![EUV spectra](image_url)
The definition of the intensity is the same as that in the present and available calculations [28, 29]. It is confirmed in the table, the wavelength values by Safronova lists the three lines in W$^{27+}$ observed in the present study with the present and available calculations [28, 29]. It is noted that the 5s by the product of the 5s decay can be realized not only by E3 but also by M2. However, as shown in the table, the transition probability by M2 (1.1 × 10$^{-9}$ s$^{-1}$) is much smaller than that by E3 (83 s$^{-1}$), thus the transition is considered to be realized dominantly by E3. As confirmed in the table, the wavelength values by Safronova show the best agreement with the present experimental values.

Although the transition probabilities of these E3 decays are much larger than that of, for example, the $2^2$F$_{7/2}$ – 2S$_{1/2}$ E3 transition in singly charged Yb$^+$. (~ 10$^{-9}$ s$^{-1}$) owing to their large transition energies, they are still much smaller than the transition probability of E1 allowed transitions. However, the transition probability of forbidden transitions often has a strong Z dependence, as described earlier. The blue squares in Fig. 2 show the atomic number dependence of the calculated transition probability of the $4f_{5/2}$ – 5s E3 transition. As expected, the transition probability increases quickly with Z. The dependence on Z is indeed about Z$^{40}$ at around Z = 74. This drastic dependence on Z is considered to be caused by the fact that the level crossing between 4f and 5s exists at Z ≈ 60. In the vicinity of the level crossing, the transition probability is almost zero as the transition energy is nearly zero. When Z is increased from the crossing, the energy interval ΔE between 4f and 5s rapidly increases. This results in the steep rise in the transition probability, which is approximately proportional to ΔE$^5$ for this E3 transition. Although the dependence becomes weaker as Z increases, it is still Z$^{40}$ at Z ≈ 74. One may thus expect that the intensity of the E3 lines should also increase when Z is increased beyond 74. Figure 2 also plots the intensity of the $4f_{7/2}$ – 5s E3 emission, calculated by the present CR model. In each calculation, the electron energy was assumed to be just below the ionization energy of the Ag-like ion, and the electron density was fixed at 10$^{10}$ cm$^{-3}$, which is the typical value in CoBIT. The transition probability and the intensity of the $4f_{5/2}$ – 5s E3 emission have almost the same Z-dependences although they are not shown in the figure. As seen in the figure, in contrast to the expectation, the $4f_{7/2}$ – 5s E3 emission line intensity has a sharp maximum at Z = 74, and decreases quickly as Z increases when Z exceeds 74.

In order to understand this strong Z-dependent behavior of the E3 intensity, population kinetics for 5s (the upper state of the E3 transition) is considered. Direct collisional excitation rate from the ground state to 5s is negligibly small, and thus feeding by radiative cascades from upper states is required for populating 5s. The importance of radiative cascades was also confirmed for other multipole transitions, such as E2 [11] and M3 [12] in low-density plasmas. Fig. 3(a) shows theoretical values for inflow and depopulation rates (see Eq. (1)) calculated for the 5s level. The collisional inflow $C_{in}$ is not shown because it is negligibly small (less than 10$^{-1}$ s$^{-1}$) in this Z region. As shown in Eq. (1), the fractional population of the 5s level is determined by the ratio between the inflow rate $F_{in}$ (~ $R_{in}$) and the depopulation rate $f_{out}$. The numerator $R_{in}$ decreases as Z increases for a whole range of Z shown in Fig. 3(a) because collisional excitation rates from the ground state to the higher levels decrease as Z increases. For Z ≤ 68, the denominator $f_{out}$ also decreases with a similar Z dependence because $f_{out}$ is dominated by the collisional depopulation $c_{out}$, whose collisional excitation rates also decreases as Z increases. Thus, the fractional population of the 5s level, which is shown by the solid black squares in Fig. 3(a), is almost constant for Z ≤ 68. However, $f_{out}$ starts to increase from Z ≈ 70 due to the contribution of the radiative depopulation $r_{out}$. The increase of $f_{out}$ shows the stronger Z dependence than the decrease of $R_{in}$. As a result, the population is steeply decreased when Z exceeds 70. The resultant dependence is about Z$^{-37}$ as shown by the dashed line in the figure. On the other hand, the E3 transition probability has a Z dependence with a ~ 37 at Z ≈ 75, whereas a > 37 for Z < 75 and a < 37 for Z > 75. Thus, the E3 intensity, which is determined by the product of the 5s population and the transition probability, should have a maximum at Z ≈ 75. However, such a maximum should be rather gentle, thus another mechanism should be needed to explain the sharp maximum at Z = 74.

![FIG. 2. Atomic number dependence of the transition probability (blue) and the intensity (red) of the $4f_{7/2}$ – 5s E3 transition. The definition of the intensity is the same as that in Fig. 1. It is noted that the ground state is 5s and the upper state is 4f for Z < 62, whereas the ground state is 4f and the upper state is 5s for Z ≥ 62.](image)
TABLE I. Experimental and theoretical wavelengths of the emission lines in Ag-like W$^{27+}$ observed in the present study. Calculated transition probabilities are also given in the last column.

| label | transition | Exp. | Theory | Wavelength (nm) | A (s$^{-1}$) |
|-------|------------|------|--------|-----------------|-------------|
|       | upper      | lower | type   | present         | present     | RMBPT$^{[28]}$ | MCDF$^{[29]}$ | present       |
| a     | 5s         | 4f$_{5/2}$ | E3    | 8.91            | 8.982       | 8.905        | 9.040        | 83            |
| b     | 5s         | 4f$_{5/2}$ | M2    | -               | -           | -            | -            | 1.1 × 10$^{-3}$ |
| c     | 5d$_{3/2}$ | 5p$_{1/2}$ | E1    | 9.14            | 9.225       | 9.146        | 9.285        | 96            |
|       | 5s         | 4f$_{7/2}$ | E3    | 12.59           | 12.533      | 12.589       | -            | 1.7 × 10$^{11}$ |

![Figure 3](image)

**FIG. 3.** (a) Inflow and depopulation rates for the metastable 5s level (left axis). Fractional population of the 5s level determined by $F_{in}/f_{out}$ is also plotted (right axis). See also Eq. [1]. (b) Branching ratios to 5p in the decay of the 5d$_{3/2}$ (open diamond) and 5d$_{5/2}$ (closed diamond) levels.

Figure 3(b) shows the branching ratios to 5p in the decay of the 5d levels. As seen in the figure, they show anomalous Z dependence with almost unity at Z = 73 but almost negligible at Z = 74. This behavior is due to the anomalous minimum at Z = 74 in the Z-dependence of the 4f − 5d transition probability. The minimum can be understood as follows. Figure 4(a) shows energy levels assigned to 5d$_{3/2}$ and (4d$^9$4f$^2$)$_{3/2}$ states as a function of Z. The 5d$_{3/2}$ level becomes quasi-degenerate with the highest level of the (4d$^9$4f$^2$)$_{3/2}$ state in between Z = 72 and 73, where the two levels have a strong configuration mixing. Reduced matrix elements of the E1 transition between 5d$_{3/2}$ and 4f$_{5/2}$ levels exhibit a local irregularity at the level-crossing as shown in Fig. 4(b). The same irregularity has already been found in relativistic many-body perturbation calculations by Safronova et al. (see Fig. 2(c) of Ref. [30]). Here we investigate it in more detail to elucidate mechanisms of the anomalous minimum at Z = 74. The transition matrix element can be decomposed into two components: the primary component of 5d$_{3/2}$ state and the complementary component which results from the configuration mixing of (4d$^9$4f$^2$)$_{3/2}$ state. The irregularity is caused by the (4d$^9$4f$^2$)$_{3/2}$ component which increases resonantly and change the sign at the crossing. The magnitude of the (4d$^9$4f$^2$)$_{3/2}$ component decreases as Z becomes distant from the level-crossing, and becomes almost the same magnitude of the 5d$_{3/2}$ component at Z = 74. The two components of opposite signs cancel each other, akin to Cooper minima in photoionization cross sections, which results in the local minimum of the 4f$_{5/2}$ − 5d$_{3/2}$ transition probability. As a result of the minimum in the 4f − 5d transition probability, almost all the 5d population decays to 5p, and thus has a large contribution to the 5s population via 5p specifically at Z = 74. The enhancement in the 5s population, which can be confirmed as a deviation from the general trend (Z$^{-37}$ dependence shown by the dashed line in Fig. 3(a)), is specifically large at Z = 74, but rapidly decreases as Z increases, and almost negligible at Z = 77. This behavior results in the sharp Z dependence of the emission line intensity shown in Fig. 2.

Figure 5 shows the energy levels of Ag-like W$^{27+}$ and the breakdown of the population flows calculated for an electron energy of 870 eV and a density of 10$^{10}$ cm$^{-3}$. The number associated with each arrow represents the flow in s$^{-1}$ defined by the product of the fractional population $n_i$ of the initial level $i$ and the rate of the transition to the levels $j$ ($n_i \sum_j C_{ij}$ for collisional transitions and $\sum_j A_{ji}$ for radiative transitions). As understood from the figure, for the 4d$^{10}$5s level, which is the upper state of the E3 transitions, the collisional outflow to the 4d$^{10}$nln$l'$ levels (1.8 s$^{-1}$) and the radiative inflow from the 4d$^{10}$nln$l'$ levels (2.0 s$^{-1}$) are almost balanced. The E3 flow is thus realized by the radiative inflow from the 4d$^{10}$5p levels (5.1 s$^{-1}$), which exceeds the collisional outflow to the 4d$^{10}$5p levels (1.3 s$^{-1}$). The excess amount (3.8 s$^{-1}$) is predominantly due to the radiative inflow from the 4d$^{10}$5d levels (3.3 s$^{-1}$). If the anomalous minimum in the 4f − 5d transition probability did not exist,
the greater part of the radiative flow from the $4d^{10}5d$ levels should fall into the $4d^{10}4f$ ground state, and thus the prominent $E3$ emission could not be obtained.

V. SUMMARY AND OUTLOOK

In summary, we have observed extreme ultraviolet spectra of Ag-like $W^{27+}$ with an electron beam ion trap, and identified the $4f_{7/2,5/2} - 5s$ transitions in the spectra as the first observation of spontaneous electric octupole ($E3$) emission lines. Our collisional radiative model calculation has shown that the line intensity is strongly enhanced at $Z = 74$ due to an anomalous behavior of the $4f - 5d$ transition probability. If it had not been for the anomalous behavior, we might have failed to observe the emission.

![Energy Level Diagram](attachment:energy_level_diagram.png)

**FIG. 4.** (a) Energy level diagram of $5d_{3/2}$ (connected blue bars) and $(4d^9 4f^2)_{3/2}$ (red bars) states. The $(4d^9 4f^2)_{3/2}$ state has 15 energy levels. (b) Reduced matrix elements of E1 transition between $5d_{3/2}$ and $4f_{5/2}$ states. Open circles show $5d_{3/2}$ (blue) and $(4d^9 4f^2)_{3/2}$ (red) components, respectively. The blue solid circles are the sum of the two components.

Tungsten is an important element for the spectroscopic diagnostics in the future ITER plasmaz [3]. The present $E3$ transitions should thus contribute to the future fusion plasma study. For a fundamental atomic physics aspect, transition life time measurement for this $E3$ emission is desirable for understanding the interaction of ions with multipole radiation fields in more detail.

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