Evidence for strong configuration mixing in $n=3$ excited levels in neonlike ions

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Wavelengths have been measured for x-ray transitions from the excited states with strong configuration mixing between $(2p_{1/2}^{-1}3s)_{J=1}$ and $(2p_{3/2}^{-1}3d_{5/2})_{J=1}$ to the ground states in neonlike I^{43+}, Cs^{45+}, and Ba^{46+}. The measurements were made with a flat crystal spectrometer on the Tokyo electron beam ion trap. The experimental wavelengths are compared to theoretical values calculated with the multiconfigurational Dirac-Fock method and other existing theoretical results. Using the present results, the degree of mixing in the wave functions between the two excited electronic configurations is investigated. The present investigation gives a clear demonstration for strong configuration mixing in highly charged ions through observation of an avoided crossing.

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I. INTRODUCTION

From a basic atomic physics point of view, accurate measurements of energy levels of highly charged ions are important for testing the atomic structure theory. In this paper we present systematic measurements of transition wavelengths for neonlike highly charged ions. There is a great deal of interest in excited states in the neonlike isoelectronic sequence, because they are expected to show distinguished features due to strong mixing of excited configurations.

Generally, an electronic state in an atom or an ion should be represented not by a single electronic configuration, but by a linear combination of many configurations that have the same symmetry. This configuration mixing becomes important especially when two or more configurations have comparatively close energy. For example, strong mixing between nd and $(n+1)s$ is generally found for the ground state in the transition elements. O'Malley and Beck [1] also found strong mixing between the $5d$ and the $6s$ configurations in the excited states of the xenonlike ions, Cs^{+} and Ba^{2+}. These nd-$(n+1)s$ mixing characteristics are strong and almost independent of the atomic number Z. However, apparent features which appear in $n=3$ excited states for neonlike ions are different from the above. In the neonlike sequence, strong configuration interaction can be found for the particular values of Z around which the order of energy levels changes in the course of the change of the coupling scheme from LS to jj. As an example, strong configuration mixing between $(2p_{1/2}^{-1}3s)_{J=1}$ and $(2p_{3/2}^{-1}3d_{5/2})_{J=1}$ at $Z\sim 55$ is shown by Kagawa *et al.* [2]. In contrast to the nd-$(n+1)s$ mixing in the transition elements or the xenonlike ions, this mixing is due to local degeneracy of their levels at $Z\sim 55$ in strong variation of the respective configuration energies. The local degeneracy is attributed to large fine-structure splitting of the $2p$ orbit in highly charged ions, which compensates and eventually becomes larger than energy difference between the $3s$ and the $3d_{5/2}$ orbit with passing over $Z=55$. Since each of these states cannot be represented by a single configuration for this Z-region, in the following text, we refer to the state having the largest mixing coefficient for $(2p_{3/2}^{-1}3d_{5/2})_{J=1}$ as $|3D\rangle$, and that having the largest mixing coefficient for $(2p_{1/2}^{-1}3s)_{J=1}$ as $|3F\rangle$, following the notations used by Loulergue and Nussbaumer [3].

The effect of the strong configuration mixing results in anomalous behavior in the Z-dependence of the oscillator strengths. Kagawa *et al.* [2] and Ivanova and Grant [4] showed in their theoretical investigations that the oscillator strength for the $2p^6\rightarrow|3F\rangle$ transition is enhanced, while that for the $2p^6\rightarrow|3D\rangle$ transition is suppressed at $Z\sim 55$ as a result of the strong mixing. On the other hand, the effect of the strong configuration mixing also appears as a so-called avoided crossing in the energy-level change with Z. In this paper, we present characteristics for the avoided crossing between $|3D\rangle$ and $|3F\rangle$, which have been investigated through measurements of the transition wavelengths in the x-ray region. We have also made a multiconfigurational Dirac-Fock (MCDF) calculation in which the self-energy has been estimated with taking the strong configuration mixing into account. The present experimental results are compared with previous experimental and theoretical results as well as the present theoretical results.

II. EXPERIMENTS

The experimental setup is shown in Fig. 1. Neonlike ions were produced and trapped in the Tokyo electron beam ion trap (Tokyo-EBIT) [5–7]. The x-ray transition excited by a 60-μm-diam electron beam was observed with a flat crystal spectrometer. Since the radiation source in an electron beam ion trap is a line source whose width is about 60μm, it is possible to use wavelength dispersive spectrometers without an entrance slit [8]. The spectrometer consisted of a flat LiF(200) crystal with an area of 100×50 mm^2 and a position sensitive proportional counter (PSPC) with a backgammon-type cathode [9]. In the present observations, the crystal was placed at 620–730 mm away from the center of the trap and the PSPC at 460–740 mm away from the crystal. The effective volume of the PSPC was $100\times 30\times 4$ mm^3 in which 4 atm pressure of PR gas (90% Ar + 10% CH_4) was filled. Typical resolution in position for the present PSPC was

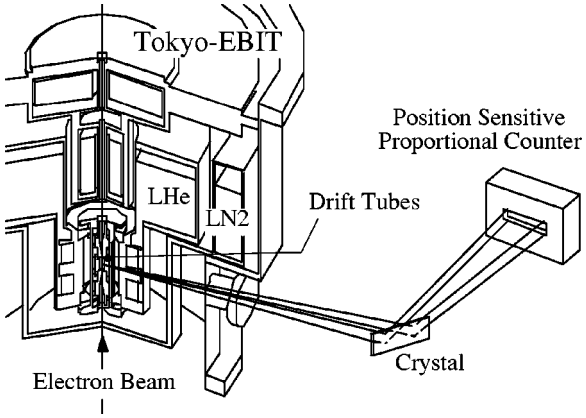

FIG. 1. Diagram of the experimental arrangement. Neonlike ions were produced and trapped at the center of the drift tubes. The spectrometer was operated *in vacuo*, although the vacuum chambers are not shown.

about 300 μm. Although the vacuum chambers are not shown in the figure, the spectrometer was operated in vacuo ($\sim 10^{-7}$ torr) to avoid absorption by air. A beryllium foil with a thickness of 50 μm was used to separate the vacuum of the EBIT ($\sim 10^{-9}$ torr) from that of the spectrometer.

Figure 2 shows X-ray spectra from neonlike I^{43+}, Xe^{44+}, Cs^{45+} and Ba^{46+}. It is clearly demonstrated in the figure that $3D$ (corresponding to the $|3D\rangle \rightarrow 2p^6$ transition) and $3F$ ($|3F\rangle \rightarrow 2p^6$) become close, change positions, and then become far again as the atomic number increases. For I^{43+}, Cs^{45+} and Ba^{46+}, since the electron energies were just below the ionization energies of the neonlike ions, no lines from the fluorinelike and other higher charge-state ions were not observed in the spectra. A few lines from the sodiumlike ions were observed. From the intensity ratios between the sodiumlike and the neonlike ions (e.g., $3D/3D_{\mathrm{Na}}$ in the Cs spectrum) it is suggested that the abundance of the sodiumlike ions in the trap was much smaller than that of the neonlike ions [10].

It seems that the intensity ratio between $3F$ and $3D$ has Z dependence. It may be compared with theoretical transition probabilities. However, the intensity of radiation from an EBIT is dominated by the electron-impact excitation rate rather than the transition probability because the electron density is low enough. Furthermore, indirect excitation processes, such as radiative cascades, are possible for the present electron energies. The intensity of radiation is thus determined by complicated processes, and cannot be explained only by transition probabilities. Since the present paper is focused on wavelength measurements, the intensity of the radiation will be discussed elsewhere.

Wavelengths for neonlike I^{43+}, Cs^{45+}, and Ba^{46+} were determined using the transition wavelengths in neonlike Xe^{44+} which were determined experimentally in the previous work [11]. Since there was no entrance slit in the present spectrometer, the radiation source should keep the same position during the observations. Therefore, the same values of the EBIT parameters were used in the observations for both the objective ions (I, Cs, and Ba) and the reference (Xe). The measurements of the objective ions and the reference were

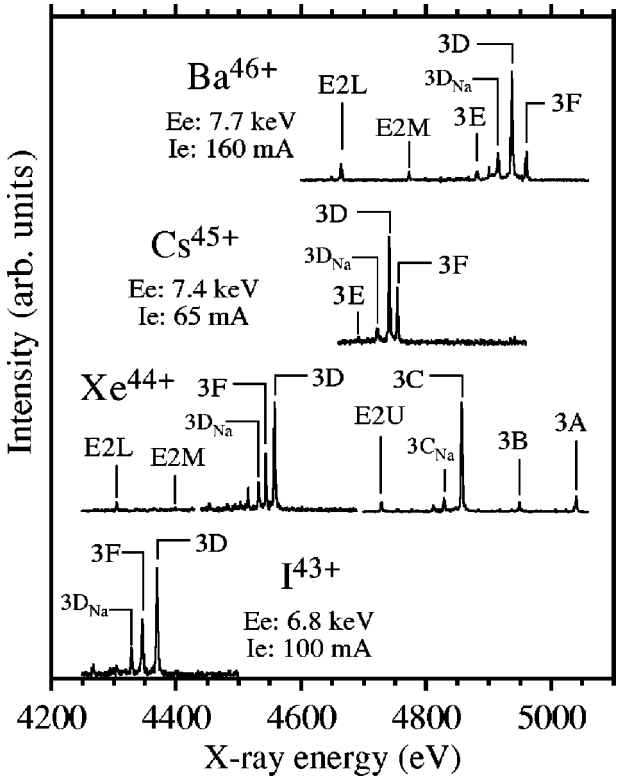

FIG. 2. X-ray spectra from neonlike I^{43+}, Xe^{44+}, Cs^{45+}, and Ba^{46+}. The notations used by Loulergue and Nussbaumer [3] and Beiersdorfer *et al.* [11] are indicated in the figure. $3D_{\mathrm{Na}}$ and $3C_{\mathrm{Na}}$ represent satellites to $3D$ and $3C$ respectively. Ee and Ie represent the electron energies and the currents, respectively, at which the spectra were obtained. For neonlike Xe^{44+}, three spectra (4250–4430, 4440–4690, and 4700–5060 eV) were observed individually. It is noted that intensity ratio between different spectra cannot be compared.

performed alternately for the uncertainty arising from the drift in the power supplies and other electronics to be removed. Actually, for example, data for Ba^{46+} were accumulated for about 1 h acquisition time and then Xe^{44+} were used as a reference for the same accumulation time. This combination of observations is called ''one run'' of the measurement. More than ten runs were accumulated for each objective ion to obtain the final results of the wavelengths in the present measurement.

For neonlike Ba^{46+}, two sets of measurements were done using different positions of the crystal and the detector. These measurements allowed us to check the systematic uncertainties arising from the position dependence on the crystal and the detector. Since the two sets of measurements gave consistent results, the systematic error coming from the crystal spectrometer is considered to be negligible. Although only one set of measurements was done for neonlike I^{43+} and Cs^{45+}, it is considered that the systematic errors for these ions are also negligible since the crystal and detector used in their measurements were the same.

A line center was determined by fitting each spectrum to a Voigt spectral shape. A wavelength of each line was determined by interpolation using two reference lines. Exceptions

TABLE I. Wavelengths of several $n=3$ to 2 transitions in Ne-like I^{43+}, Cs^{45+}, and Ba^{46+}. All values are given in Å. The numbers in the parentheses represent the experimental uncertainties, e.g., 2.8527(6) represents 2.8527 ± 0.0006.

Element (Z)	Line[a]	Upper level	Ne-like Xe Reference[a]	Present (Expt)	Present (MCDF)	Aglitskii (MP) [16]	Quinet (MCDF) [19]	Avgoustoglou (MBPT) [17,18]
I (53)	$3F$	$(2p_{1/2}^{-1}3s_{1/2})_{J=1}$	$E2L$-$E2M$	2.8527(6)	2.8526	2.8517	2.8544	2.8522
	$3D$	$(2p_{3/2}^{-1}3d_{5/2})_{J=1}$	$E2L$-$E2M$	2.8373(6)	2.8365	2.8360	2.8379	2.8371
Cs (55)	$3E$	$(2p_{3/2}^{-1}3d_{3/2})_{J=1}$	$3C$-$E2U$	2.6424(7)	2.6433	2.6430	2.6447	
	$3D$	$(2p_{3/2}^{-1}3d_{5/2})_{J=1}$	$3C$-$E2U$	2.6149(6)	2.6151	2.6147	2.6166	
	$3F$	$(2p_{1/2}^{-1}3s_{1/2})_{J=1}$	$3C$-$E2U$	2.6078(6)	2.6080	2.6075	2.6093	
Ba (56)	$E2L$	$(2p_{3/2}^{-1}3p_{1/2})_{J=2}$	$3C$-$E2U$	2.6582(7)	2.6592		2.6589	
	$E2M$	$(2p_{3/2}^{-1}3p_{3/2})_{J=2}$	$3C$-$E2U$	2.5978(5)	2.5978		2.5977	
	$3E$	$(2p_{3/2}^{-1}3d_{3/2})_{J=1}$	$3B$-$3C$	2.5396(4)	2.5396	2.5394	2.5409	2.5395
	$3D$	$(2p_{3/2}^{-1}3d_{5/2})_{J=1}$	$3B$-$3C$	2.5110(3)	2.5109	2.5106	2.5121	2.5113
	$3F$	$(2p_{1/2}^{-1}3s_{1/2})_{J=1}$	$3A$-$3B$	2.4995(4)	2.4998	2.4993	2.5012	2.4995

[a]The notations used by Loulergue and Nussbaumer [3] and Beiersdorfer *et al.* [11] are used.

are $(2p_{3/2}^{-1}3p_{1/2})_{J=2} \rightarrow 2p^6$ (denoted $E2L$) of neonlike Ba^{46+} and $(2p_{3/2}^{-1}3d_{3/2})_{J=1} \rightarrow 2p^6$ (denoted $3E$) of neonlike Cs^{45+}, for which extrapolation was used.

The uncertainties in the present experiments were estimated from the quadrature sum of four contributions: the statistical errors for the reference lines (Xe^{44+}) and the objective lines (I^{43+}, Cs^{45+} and Ba^{46+}), the uncertainties in the reference lines, and the uncertainties arising from the nonlinear positional response of the detector. Among them the last two contributions were dominant.

III. THEORY

To calculate the transition wavelengths, we used the GRASP92 code developed by Grant and co-workers [12]. A set of subshell wave functions for the ground configuration $1s^2 2s^2 2p^6$, and that for 36 excited configurations $2l^{-1}3l'$ (representing that one of the $2l$ core-subshell electrons is excited to the $3l'$ subshell) were optimized independently through separate self-consistent field (SCF) calculations. For the ground configuration, a set of radial wave functions for all occupied subshells was obtained by solving the Dirac-Fock equation. For the excited configurations, we solved the multiconfigurational Dirac-Fock equation in the (extended-) averaged-level (EAL) mode. In the EAL mode calculation, radial wave functions are optimized so that the average energy of all the configurations weighted by their respective statistical weights becomes stationary with respect to variations of the radial wave functions. As a result, the subshell wave functions for the excited configurations become different from those for the ground configuration; the core-subshell wave functions for the excited configurations shrink more tightly around the nucleus, since the net screening effect of the nuclear attraction field becomes weakened after one of the core electrons is excited to the outer subshell.

Both the transverse-photon exchange effect and the vacuum polarization effect were treated as perturbations in the Dirac-Coulomb Hamiltonian. The transverse-photon exchange effect was taken into account through the frequency-dependent Breit operator [12]. For the vacuum polarization effect, expressions for the second- and the fourth-order perturbation potential by Fullerton and Rinker [13] were used. For the ground atomic level, these corrections were treated as the first-order perturbation to the Dirac-Coulomb energy of the ground configuration, while a set of excited levels was obtained by the configuration-interaction (CI) calculations for all the excited configurations including the above corrections. Thus, the perturbations were treated to all orders for the excited levels within a space spanned by the 36 excited configurations. The mass polarization effect was also taken into account as a perturbation, though it has little contribution for heavy ions. To take the finite nuclear size into account, the nuclear charge distribution was represented by the Fermi model throughout the present calculations.

The self-energies for the subshell electrons which move in their respective effective nuclear fields were obtained by interpolating the hydrogenic self-energies tabulated by Mohr [14] and Johnson and Soff [15] for the $1s$, $2s$, and $2p$ subshells. For the $3l'$ subshells, the n^{-3} scaling law was used to estimate their self-energies. Then the self-energy for each configuration may be obtained by summing up the self-energies for all occupied subshells weighted by their respective occupation numbers q_{nl}^j, where nl represents each subshell and j each configuration. However, when configurations involved in an atomic level are strongly mixed, it is impossible to make a unique correspondence of the atomic level with one of the configurations. For estimation of the self-energy in such an atomic level, it may be more reasonable to use weighted sums of occupation numbers for all configurations involved in the atomic level,

$$\tilde{q}_{nl}^i = \Sigma_j [q_{nl}^j |c_j^i|^2], \tag{3.1}$$

where $\{c_j^i\}$ represents a set of the CI coefficients for the ith atomic level. A weighted occupation number for each subshell can be a fractional number, while the total number of electrons, Q, is always conserved for every atomic level, i.e., $\Sigma_{nl} \tilde{q}_{nl}^i = Q$ as long as $\Sigma_j |c_j^i|^2 = 1$. As a typical example, the self-energy for the $|3D\rangle$ level of neonlike Cs^{45+} was estimated to be $+121.7699$ eV with a set of the weighted occu-

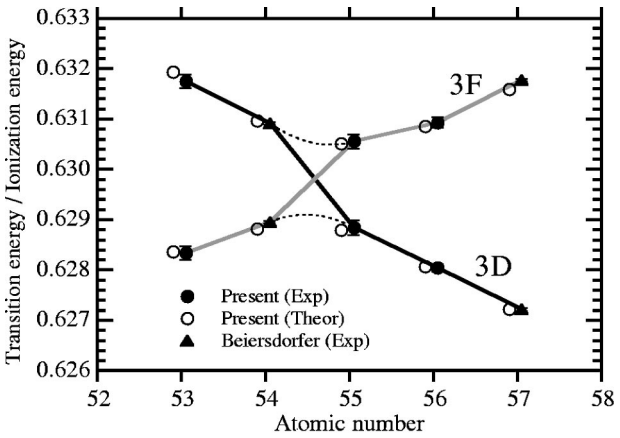

FIG. 3. Experimental and theoretical transition energies for $3D$ and $3F$. Solid and open circles represent the present experimental and the theoretical values, respectively. Triangles represent the experimental values obtained by Beiersdorfer *et al.* [11].

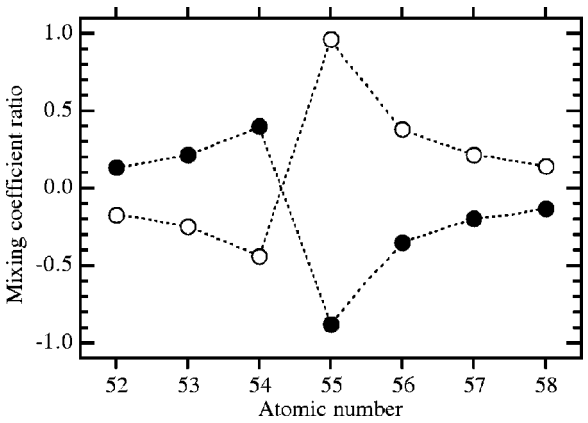

FIG. 4. Ratios of the CI coefficients. Open circles represent the ratio of the $|2p_{1/2}^{-1}3s\rangle$ component to the $|2p_{3/2}^{-1}3d_{5/2}\rangle$ component for the $|3D\rangle$ level, and solid circles the ratio of the $|2p_{3/2}^{-1}3d_{5/2}\rangle$ component to the $|2p_{1/2}^{-1}3s\rangle$ component for the $|3F\rangle$ level.

pation numbers, whereas that was estimated to be $+120.7096$ eV with a set of occupation numbers for the configuration whose mixing coefficient has the largest magnitude for the level. The transition wavelength for $3D$ was reduced by 0.0006 Å with the former self-energy compared to that with the latter self-energy. However, the modification is insignificant in the present case; the change in the transition wavelength is no more than the experimental error.

IV. RESULTS AND DISCUSSION

In Table I the results for the present experiments and the calculations are listed with the corresponding values in previous investigations. The reference lines that were used for each line are also listed in the table. It is noted that the wavelengths which have a similar value are listed in the same row regardless of the assignment in the original papers. Aglitskii *et al.* [16] determined experimental wavelengths for several transitions listed in the table. However, these wavelengths were not directly observed, but obtained by the least square method from the experimental values for other neonlike ions with different Z. Therefore, these values are not included in the table. As seen in the table, agreement between the present experimental results and the theoretical values is quite well for all the lines. The model potential (MP) method calculations by Aglitskii *et al.* [16] and the many-body perturbation (MBPT) calculations by Avgoustoglou *et al.* [17,18] also predict wavelengths to be totally consistent with the present measurements. All the values listed in the table coincide within three significant digits, though the MCDF values by Quinet *et al.* [19] are somewhat larger than the present experimental results and the other theoretical values for all the lines.

The present experimental and the theoretical results for $3D$ and $3F$ are plotted in Fig. 3. For neonlike Xe^{44+} and La^{47+}, the experimental values obtained by Beiersdorfer *et al.* [11] are used in the plot. It is noted that both of the experimental and the theoretical results clearly indicate that the two levels get close to each other around $Z=55$ but avoid to degenerate. It suggests that they are coupled through strong mixing of electronic configurations. To see the degree of their mixing, the ratios of the CI coefficients for the two configurations are plotted in Fig. 4; the ratio of the $|2p_{1/2}^{-1}3s\rangle$ component to $|2p_{3/2}^{-1}3d_{5/2}\rangle$ and that of the $|2p_{3/2}^{-1}3d_{5/2}\rangle$ component to $|2p_{1/2}^{-1}3s\rangle$ are plotted for the $|3D\rangle$ and the $|3F\rangle$ level respectively. Thus, magnitudes of the ratios indicate the degree of the configuration mixing. In the figure, an enhancement of the degree of the mixing is clearly seen at $Z=55$ for both the levels.

In addition, the figure shows the ratios change their signs with passing over $Z=54$. It means that configuration mixing patterns in the levels interchange somewhere between $Z=54$ and 55. This finding suggests an alternative representation for the electronic states of the levels. In general, a system with two independent states which are mixed through some interactions has two eigenstates: one in which phases of the mixing coefficients coincide, and the other in which their phases differ by π. We refer to these eigenstates as $|+\rangle$ and $|-\rangle$ respectively. In the present case they are represented by,

$$\begin{pmatrix} |+\rangle \\ |-\rangle \end{pmatrix} = \begin{pmatrix} a & b \\ -b & a \end{pmatrix} \begin{pmatrix} |2p_{3/2}^{-1}3d_{5/2}\rangle \\ |2p_{1/2}^{-1}3s\rangle \end{pmatrix}, \tag{4.1}$$

where a phase convention is set to be $a,b>0$. The present results showed that for $Z\leqslant 54$ $|+\rangle$ and $|-\rangle$ correspond to $|3F\rangle$ and $|3D\rangle$, but for $Z\geqslant 55$ they correspond to $|3D\rangle$ and $|3F\rangle$ respectively. Therefore, the $|+\rangle$ level is always lower in energy than the $|-\rangle$ level. They would alter their energies smoothly if the atomic number could vary continuously. Then a set of energy curves with an avoided crossing around $Z=55$ would be obtained; one may associate their behavior with atomic energy levels in a strong external field. When atomic levels involve two configurations mixed strongly, it becomes essential to represent the levels in terms of $|\pm\rangle$.

The $|\pm\rangle$ representation is also convenient to understand their interactions with electro-magnetic fields. The $2\ ^1P^o$ doubly excited levels of helium make three series converging

to the He^+ $(N=2)$ manifold. Cooper *et al.* [20] represented electronic states for two of them by $|2snp\rangle+|2pns\rangle$ and $|2snp\rangle-|2pns\rangle$, and clearly explained their respective propensity rules for the intensities of electric-dipole transitions from the ground level $1\ ^1S^e$. In the present case the electric-dipole moment between the ground state and the $|-\rangle$ state is represented by $\langle 0|\hat{D}|2p_{1/2}^{-1}3s\rangle a-\langle 0|\hat{D}|2p_{3/2}^{-1}3d_{5/2}\rangle b$, where $\langle 0|$ represents the ground state and $\hat{D}$ the electric-dipole operator. This electric-dipole moment could vanish when the first and the second term cancel each other. That is the case for the electric-dipole transition $3F$. Theoretical calculationsby Ivanova and Grant [4] predicted that the transition rate falls steeply and vanishes around $Z=58$.

In summary, we have measured and calculated the transition wavelengths for $3D$ and $3F$ in neonlike I^{43+}, Cs^{45+}, and Ba^{46+}. The calculated wavelengths reproduce the experimental values quite well. The systematic investigation made it possible to clearly demonstrate the strong configuration mixing between $(2p_{1/2}^{-1}3s)_{J=1}$ and $(2p_{3/2}^{-1}3d_{5/2})_{J=1}$ in this Z region through the observation of the avoided crossing. Further investigations, including excitation cross section measurements [21], are ongoing.

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