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Dzuba, V. A., Flambaum, V. V., Gribakin, G. F., & Harabati, C. (2012). Chaos-induced enhancement of resonant multielectron recombination in highly charged ions: Statistical theory. Physical Review A (Atomic, Molecular, and Optical Physics), 86, [022714]. https://doi.org/10.1103/PhysRevA.86.022714

Published in:
Physical Review A (Atomic, Molecular, and Optical Physics)

Document Version:
Peer reviewed version

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Chaos-induced enhancement of resonant multielectron recombination in highly charged ions: Statistical theory

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(Dated: August 24, 2012)

A statistical theory of resonant multielectron recombination based on properties of chaotic eigenstates is developed. The level density of many-body states increases exponentially with the number of excited electrons. When the residual electron-electron interaction exceeds the interval between these levels, the eigenstates (called compound states or compound resonances if these states are in the continuum) become “chaotic” superpositions of large numbers of Hartree-Fock configurational basis states. This situation takes place in some rare-earth atoms and many open-shell multiply charged ions excited in the process of electron recombination. Our theory describes resonant multielectron recombination via dielectronic doorway states leading to such compound resonances. The result is a radiative capture cross section averaged over a small energy interval containing several compound resonances. In many cases individual resonances are not resolved experimentally (since the interval between them is small, e.g., ≤ 1 meV, possibly even smaller than their radiative widths), therefore, our statistical theory should correctly describe the experimental data. We perform numerical calculations of the recombination cross sections for tungsten ions Wq+ at q = 18–25. The recombination rate for W20+ measured recently [Phys. Rev. A 83, 012711 (2011)] is 103 greater than the direct radiative recombination rate at low energies, and our result for W20+ agrees with the measurements.

PACS numbers: 34.80.Lx, 31.10.+z, 34.10.+x, 32.80.Zb

I. INTRODUCTION

The majority of atoms in highly excited states and many open-shell (e.g., rare-earth) atoms, even in vicinity of the ground state, behave as complex, chaotic many-electron systems. They are characterized by dense level spectra and strong configuration mixing. The corresponding many-electron wave functions are mixtures of large numbers of many-excited-electron basis states (Slater determinants) with nearly random mixing coefficients. Obtaining detailed information about such systems by standard theoretical methods, such as configuration interaction, becomes problematic because of the sheer size of the effective Hilbert space. In fact, the experiment often does not resolve individual energy levels, so a complete description is to some extent pointless.

On the other hand, such complex systems can be described using statistical approaches. Statistical methods for chaotic compound states are widely used in nuclear physics (see, e.g., Refs. [1–5]). Similar methods have been developed for open-shell atomic systems [6–9]. In particular, extensive numerical calculations were performed for the cerium atom to test the assumptions and predictions of the statistical theory [6]. These works examined the properties of the Hamiltonian matrix in chaotic many-body systems, statistics of energy level spacings, dependence of mean orbital occupation numbers on the excitation energy, statistics and mean-squared values of electromagnetic amplitudes between chaotic many-body states, enhancement of weak perturbations in such states, and electronic and electromagnetic widths of chaotic compound resonances. Some aspects of the statistical theory were also tested for multicharged ions [7–9].

In the present paper we want to use the statistical theory to describe the effect of compound resonances on electron recombination with open-shell multicharged ions. Electron recombination is an important process in laboratory and cosmic plasmas, as well as in ion storage rings. Theory and experiment agree very well for relatively simple systems with one or two valence electrons above closed shells (see, e.g., Ref. [10]). For more complex systems theory and experiment often deviate significantly. For example, strong enhancement of the recombination rate were observed for Au25+ [11], U28+ [12] and W20+ [13] at low electron energies. For such ions the observed recombination rate is orders of magnitude greater than that due to direct radiative recombination (RR). In simpler systems the enhancement is due to resonant dielectronic recombination, though even for ions like Fe23+ 3p5 and Fe25+ 3p4 the dielectronic recombination appears to be inefficient [14]. In complex open-shell ions such as Au25+ 4f8 and isoelectronic W20+ the absolute majority of the resonances correspond to many-excited-electron eigenstates which have very high density (exponentially small level spacings). In Refs. [7, 8] use a statistical approach to show that a factor of 200 enhancement over RR observed in Au25+ [11] is due to electron capture in these compound resonances.

In this paper we use the statistical theory to calculate the recombination rates for tungsten ions Wq+, q = 18–25. Our results for W20+, where the measured rate at low (∼ 1 eV) energies is 103 times higher than the radiative rate [13], are in good agreement with experiment. For other tungsten ions we predict similar strong enhance-
The fact that the two-electron-one-hole excitation temporal picture the process looks as a series of electron and the latter at least partially empty. The continuum electronic state plays the role of a excited states and is strongly mixed with them. This di-electronic state which to transfer electrons from orbitals and is not an eigenstate of the highly excited ion. This state is embedded in the dense spectrum of other, multiply excited states and is strongly mixed with them. This di-electronic state captures the electron. The language of strong configuration mixing is more appropriate in this case. With a perfect energy resolution one would see a very dense spectrum of narrow, possibly overlapping resonances. Broad doorway di-electronic states (with width \( \Gamma_{\text{spr}} \)) can only introduce a variation of the average height of these narrow compound resonances on the energy scale \( \Delta \varepsilon \sim \Gamma_{\text{spr}} \).

Compared with the total resonant capture cross section, the recombination cross section

\[
\tilde{\sigma}_r = \omega_f \sigma_c
\]

contains an additional factor \( \omega_f \), known as the fluorescence yield. It accounts for the probability of radiative stabilization of the resonances (as opposed to autoionization), and is given by

\[
\omega_f = \frac{\Gamma^{(r)}}{\Gamma^{(r)} + \Gamma^{(a)}},
\]

where \( \Gamma^{(r)} \) and \( \Gamma^{(a)} \) are the resonance radiative and autoionization widths, respectively. Expressions for these quantities and for the spreading width are presented in the next section.

Note that the capture cross section (1) is not very sensitive to the specific value of the spreading width \( \Gamma_{\text{spr}} \) which for multicharged ions is about 0.5 a.u. [7] (see Table I). After angular reduction of the Coulomb matrix elements in Eq. (1) [Sec. II, Eq. (30)], numerical calculations of the capture cross section are straightforward.

An additional simplification occurs in heavy open-shell ions like Au\(^{25+}\) and W\(^{20+}\), which have almost unit fluorescence yield. Indeed, the compound states in these ions
contain very large numbers of principal basis-state components, \(N \sim 10^4\). Each component contributes to the radiative decay into a large number of states below this compound state. In contrast, only one or few dielectronic (doorway) state components have nonzero Coulomb matrix elements that allow electron autoionization into the continuum (see Fig. 1 and Eq. (1)). Therefore, the autoionization width of the compound resonance is suppressed by the small weight factor \(1/N\) of the dielectronic components in the compound state. This means that the captured low-energy electron cannot escape, i.e., after the capture the radiative process happens with nearly 100% probability. (A similar effect in neutron capture by nuclei is described, e.g., in Ref. [16]). In this situation \(\Gamma_{\text{a}}^{(a)} \ll \Gamma^{(r)}\) and \(\omega_f \approx 1\), so that the electron recombination cross section, Eq. (2), is independent of the radiative width. In this regime one observes maximum chaos-induced enhancement of the resonant multielectron recombination. For example, the electron capture cross section calculated using Eq. (1) for Au\(^{25+}\) [8], was found to be in good agreement with experiment at low energies. On the other hand, in ions with a smaller number of active electrons, the number of components \(N\) may not be so large, leading to \(\Gamma^{(a)} > \Gamma^{(r)}\) and \(\omega_f < 1\).

II. THEORY

A. Resonant recombination cross section

The resonant radiative electron-ion recombination cross section is given by the sum over the resonances \(\nu\) with the angular momentum and parity \(J^\pi\),

\[
\sigma_r = \sum_\nu g(J)\sigma_\nu, \tag{4}
\]

where \(g(J) = (2J+1)/(2(2J+1))\) is the probability factor due to random orientation of the electron spin and angular momentum \(J_i\) of the target ion [17]. The individual resonant contributions \(\sigma_\nu\) are given by the Breit-Wigner formula [17],

\[
\sigma_\nu = \frac{\pi}{k^2} \frac{\Gamma_\nu^{(a)}\Gamma_\nu^{(r)}}{(\epsilon - \epsilon_\nu)^2 + \Gamma_\nu^2/4}, \tag{5}
\]

where \(\Gamma_\nu^{(r)}\) is the radiative decay rate (the total “inelastic width”), \(\Gamma_\nu^{(a)}\) is the autoionization decay rate (the “elastic width”), and \(\Gamma_\nu = \Gamma_\nu^{(r)} + \Gamma_\nu^{(a)}\) is the total width of the level \(\nu\). (We assume here that other inelastic channels, e.g., electronic excitation, are closed, which is correct at low incident electron energies.) The energy of the \(n\)th resonance is given relative to the ionization threshold \(I\) of the final compound ion, \(\epsilon_\nu = E_{\text{ion}} - I\).

For systems with dense compound resonance spectra the recombination cross section displays rapid energy dependence, that may not even be resolved experimentally. Thus it is natural to average the cross section over an energy interval \(\Delta \epsilon\) which is large compared with the small mean level spacing \(D_{J^\pi}\) and the total resonance width, but much smaller than \(\epsilon\). This gives

\[
\frac{1}{\Delta \epsilon} \int \sigma_\nu d\epsilon = \frac{2\pi^2}{k^2} \frac{\Gamma_\nu^{(r)}\Gamma_\nu^{(a)}}{\Delta \epsilon \Gamma_\nu}, \tag{6}
\]

where the integration limits are formally infinite, since the contribution of each resonance to cross section decreases rapidly away from \(\epsilon \approx \epsilon_\nu\). The number of resonances with a given \(J^\pi\) within \(\Delta \epsilon\) is \(\Delta \epsilon/D_{J^\pi}\), and after averaging, the recombination cross section (4) becomes

\[
\bar{\sigma}_r = \frac{\pi^2}{k^2} \sum_{J^\pi} \frac{\Gamma^{(r)}\Gamma^{(a)}}{(2J_i+1)D_{J^\pi}} \left\langle \frac{\Gamma^{(r)}\Gamma^{(a)}}{\Gamma} \right\rangle. \tag{7}
\]

Here \(\langle \ldots \rangle\) means averaging of the width factor at the given energy.

If the fluorescence yield \(\omega_f = \Gamma^{(r)}/\Gamma\) fluctuates weakly from resonance to resonance, the recombination cross section \(\bar{\sigma}_r\) can be factorized, i.e., \(\bar{\sigma}_r = \omega_f\bar{\sigma}_c\). For \(\omega_f \approx 1\) the energy-averaged capture cross section

\[
\bar{\sigma}_c = \frac{\pi^2}{k^2} \sum_{J^\pi} \frac{(2J+1)}{(2J_i+1)} \frac{\Gamma^{(a)}}{D_{J^\pi} \Gamma^{(r)}}, \tag{8}
\]

is the same as the recombination cross section.

In the opposite case of small radiative widths, autoionization dominates \((\Gamma^{(r)} \ll \Gamma^{(a)})\) and \(\omega_f \ll 1\), so Eq. (7) yields the recombination cross section in the form

\[
\bar{\sigma}_r^a = \frac{\pi^2}{k^2} \frac{(\Gamma^{(r)})}{(2J_i+1)} \sum_{J^\pi} \frac{(2J+1)}{\rho_{J^\pi}}, \tag{9}
\]

where \(\rho_{J^\pi} = 1/D_{J^\pi}\) is the level density and \((\Gamma^{(r)})\) is given by Eq. (34). Note that the sum in Eq. (9) is the total density of states, which can be found without constructing states with definite \(J\).

To estimate the recombination cross section in the general case one can use the following formula,

\[
\bar{\sigma}_r \approx \frac{\bar{\sigma}_c \bar{\sigma}_r^a}{\bar{\sigma}_c + \bar{\sigma}_r^a}. \tag{10}
\]

It follows from Eqs. (7)–(9) if \(\omega_f\) does not depend on \(J\).

B. Nature of chaotic compound states

The density of excited states \(\rho(E)\) in a many-electron ion, especially with an open shell, increases rapidly (exponentially) as the number of excited electrons increases. Consider \(n\) electrons that can be distributed among a number of single-electron states \(g = \sum_l 2(2l + 1)\), where \(l\) is the orbital angular momentum of the subshells available. The total number of many-body states that can be constructed is given by

\[
\frac{g!}{n!(g-n)!} \approx \frac{\exp[n \ln(g/n) + n]}{\sqrt{2\pi n}}, \tag{11}
\]
where we used the Stirling formula and assumed $g \gg n$.

Equation (11) indicates exponential increase of the number of many-electron states and the corresponding decrease of the energy interval between them as the number of “active” electrons $n$ increases. For example, Fig. 2 shows how the density of multielectron excited states of Au$^{24+}$ increases with energy $E$. The small level spacings between the states mean that even a small residual electron-electron interaction will cause strong nonperturbative mixing of the many-electron configuration basis states (Slater determinants) $|\Phi_k\rangle$. This occurs when the off-diagonal matrix elements of the Hamiltonian $H_{ij}$ become greater than the energy spacing $D_{ij}$ between the basis states $i$ and $j$ coupled by the residual interaction, $H_{ij} > D_{ij}$.

When the mixing is strong, each eigenstate

$$|\Psi_\nu\rangle = \sum_k C_k^{(\nu)}|\Phi_k\rangle,$$

contains a large number $N$ of principal components $|\Phi_k\rangle$, i.e., basis states for which the expansion coefficients have typical values $C_k^{(\nu)} \sim 1/\sqrt{N}$. (Recall the normalization condition $\sum_k |C_k^{(\nu)}|^2 = 1$.)

The number of principal components can be estimated as $N \sim \Gamma_{spr}/D$, while the spreading width is given by the golden-rule-like formula,

$$\Gamma_{spr} \simeq \frac{2\pi H_{ij}^2}{D},$$

and $D$ is the mean level spacing between the basis states (or eigenstates). Such eigenstates are called compound states, and are well known, e.g., in nuclear physics literature [1]. Owing to the strong mixing, the only good quantum numbers that can be used to classify the eigenstates, are the exactly conserved total angular momentum and parity $J^\pi$, and the energy.

For example, in Au$^{24+}$ the mean spacing between the excited states with a given angular momentum and parity, near the ionization threshold is $D_{J^\pi} \sim 1$ meV and $\Gamma_{spr} \sim 10$ eV, so that $N \sim \Gamma_{spr}/D \sim 10^4$ [7]. Numerical calculations involving a relatively small number of configurations confirm that in this case the eigenstates are indeed chaotic superpositions of the basis states (see Fig. 3).

The energies $E_k$ of the principal basis components lie within the spreading width of the eigenenergy $E_\nu$ of the compound state, $|E_k - E_\nu| \lesssim \Gamma_{spr}$. The components outside the spreading width decrease quickly, so that they do not give much contribution to the normalization. It was tested in Refs. [6, 7] that components of the chaotic eigenstates have the statistics of Gaussian random variables with zero mean (Fig. 3, top). On the other hand, the variation of their mean-squared value as a function of energy (Fig. 3, bottom) is described well by the Breit-
Wigner profile,
\[ |C_k^{(ν)}|^2 = N^{-1} \frac{Γ_{spr}^2}{(E_k - E_ν)^2 + Γ_{spr}^2/4}, \]
with \( N = πΓ_{spr}/2D \) fixed by normalization,
\[ \sum_k |C_k^{(ν)}|^2 \simeq \int |C_k^{(ν)}|^2 dE_k/D = 1. \]

Note that the degree of mixing in such chaotic, compound states is effectively complete, i.e., all basis states that can be mixed (within a certain energy range) are mixed together. These states cannot be described in terms of electronic configurations, as each eigenstate contains significant contributions of all nearby configurations. These properties of chaotic compound states enable one to calculate mean-squared matrix elements of different operators without complete diagonalization of large configuration-interaction Hamiltonian matrices.

C. Mean-squared matrix elements between compound states

Consider a two-body operator (e.g., the Coulomb interaction)
\[ \hat{V} = \frac{1}{2} \sum_{abch} \langle ab|\hat{v}|hc\rangle a_b^\dagger a_h^\dagger a_c a_a. \]

A matrix element of \( \hat{V} \) between two compound states, \(|Ψ_ν⟩\) and \(|Ψ_i⟩\), is given by [see Eq. (12)]
\[ \langle Ψ_ν|\hat{V}|Ψ_i⟩ = \sum_{kk′} C_k^{(ν)*} C_{k′}^{(i)} ⟨Ψ_k|\hat{V}|Ψ_{k′}⟩, \]

or
\[ \langle Ψ_ν|\hat{V}|Ψ_i⟩ = \frac{1}{4} \sum_{abch} \langle ab|\hat{v}|hc\rangle - \langle ba|\hat{v}|hc\rangle \times \langle Ψ_ν|a_b^\dagger a_h^\dagger a_c a_a|Ψ_i⟩, \]

where \( \langle Ψ_ν|a_b^\dagger a_h^\dagger a_c a_a|Ψ_i⟩ \) determines the contribution of the two-particle transition \( ch \rightarrow ab \). Due to the assumption that the expansion coefficients for chaotic compound states are random and uncorrelated \( C_k^{(ν)} = C_k^{(ν)*} = 0 \) for \( ν \neq i \), the value of the matrix element averaged over many compound states \( ν \) is zero,
\[ \langle Ψ_ν|\hat{V}|Ψ_i⟩ = 0, \quad \langle Ψ_ν|a_b^\dagger a_h^\dagger a_c a_a|Ψ_i⟩ = 0. \]

To determine the autoionization width (Sec. II D) we need to calculate the mean-squared matrix element. It is derived using the statistical properties of the expansion coefficients, \( C_k^{(ν)*} C_{k′}^{(ν)} = |C_k^{(ν)}|^2 δ_{kk′} \), so that
\[ \langle Ψ_ν|a_b^\dagger a_h^\dagger a_c a_a|Ψ_i⟩ \langle Ψ_ν|a_b^\dagger a_h^\dagger a_c a_a|Ψ_i⟩ = \delta_{ab} \delta_{bh} \delta_{hc} δ_{ka} \langle Ψ_ν|a_b^\dagger a_h^\dagger a_c a_a|Ψ_i⟩^2 + \ldots, \]

where the dots correspond to three analogous terms with permutations of \( a \) and \( b \), and \( c \) and \( h \). Hence, the mean-squared matrix element is
\[ \langle |Ψ_ν|\hat{V}|Ψ_i⟩|^2 = \frac{1}{4} \sum_{abch} |\langle ab|\hat{v}|hc\rangle - \langle ba|\hat{v}|hc\rangle|^2 \times |\langle Ψ_ν|a_b^\dagger a_h^\dagger a_c a_a|Ψ_i⟩|^2. \]

Let us introduce the strength function
\[ w_i(E_k; E_ν, Γ_{spr}, N) ≡ C_k^{(ν)}|^2, \]
which describes the spreading of the component \( k \) over the eigenstates \( ν \) \( (C_{k′}^{(ν)}\) are assumed to be real). This function depends on the number of principal components \( N \) of the eigenstate, Eq. (12), the spreading width \( Γ_{spr} \), and the difference \( E_ν - E_k \) between the energies of the compound state and component \( k \). In the simplest model \([1]\) \( w_i(E_k; E_ν, Γ_{spr}, N) \) is a Breit-Wigner function, cf. Eq. (14). Using Eq. (15), we then obtain
\[ \langle |Ψ_ν|a_b^\dagger a_h^\dagger a_c a_a|Ψ_i⟩|^2 = \sum_{kk′} \sum_{ll′} C_{k}^{(ν)*} C_{k′}^{(ν)} C_{k′}^{(i)} C_{l′}^{(i)} \times \langle Ψ_ν|a_b^\dagger a_h^\dagger a_c a_a|Ψ_i⟩ \langle Ψ_{k′}^\dagger a_b^\dagger a_h^\dagger a_c a_a|Ψ_i⟩ = \sum_{kk′} w_i(E_k) w_i(E_{k′}) \times \langle Ψ_{k′}|a_b^\dagger a_h^\dagger a_c a_a|Ψ_i⟩ \langle Ψ_k|a_b^\dagger a_h^\dagger a_c a_a|Ψ_{k′}⟩. \]

To obtain the last expression we used the properties of the components and the definition (20), and denoted \( w_i(E_k) ≡ w_i(E_k; E_ν, Γ_{spr}, N_i) \) and \( w_i(E_k) ≡ w_i(E_k; E_ν, Γ_{spr}, N_0) \).

We can assume, without the loss of generality, that the number of principal components \( |Ψ_k⟩ \) in state \( ν \) is greater than or equal to the number of components \( |Ψ_{k′}⟩ \) of state \( i \), i.e., \( Γ_{spr}/D_ν ≥ Γ_{spr}/D_i \). The matrix element \( \langle Ψ_ν|a_b^\dagger a_h^\dagger a_c a_a|Ψ_i⟩ \) does not vanish only if \( |Φ_k⟩ = a_b^\dagger a_h^\dagger a_c a_a|Φ_{k′}⟩ \), so that \( E_k - E_{k′} ≈ ε_a + ε_b - ε_c - ε ≡ ω_{ab, ch} \). Using closure to sum over \( k \) in Eq. (21), we obtain
\[ \langle |Ψ_ν|a_b^\dagger a_h^\dagger a_c a_a|Ψ_i⟩|^2 = \sum_{k′} w_i(E_{k′}) w_i(E_{k′} + ω_{ab, ch}) \langle Ψ_{k′}|n_h n_c (1 - n_a)(1 - n_b)|Ψ_i⟩. \]

In deriving this equation we used the anticommutation relations satisfied by the creation and annihilation operators, and introduced the the occupation number operators \( n_a = a_a^\dagger a_a \). The matrix element \( \langle Ψ_k|n_h n_c (1 - n_a)(1 - n_b)|Ψ_{k′}⟩ \) is equal to unity if the orbitals \( h \) and \( c \) are occupied, while the orbitals \( a \) and \( b \) are vacant in the state \( |Ψ_k⟩ \), i.e., the transition \( ch \rightarrow ab \) is possible.

If one assumes that the single-electron-state occupancies vary slowly with the excitation energy, then the matrix element of the operator \( n_h n_c (1 - n_a)(1 - n_b) \) in
Eq. (22) can be replaced by its expectation value,
\[ \sum_{k'} w_i(E_{k'}) \langle \Psi_{k'} | \hat{n}_i \hat{n}_{c'} (1 - \hat{n}_a) (1 - \hat{n}_b) | \Psi_{k'} \rangle = \langle \hat{n}_i \hat{n}_{c'} (1 - \hat{n}_a) (1 - \hat{n}_b) \rangle, \]  
subject to the normalization condition \( \sum_{k'} w_i(E_{k'}) = 1 \).

The right-hand side of Eq. (23) is the value of the occupancy times “emptiness” in the compound state \( |\Psi_i\rangle \), averaged over a number of neighboring states.

Replacing the matrix element \( \langle \Phi_{k'} | ... | \Phi_{k'} \rangle \) by its average (23) in Eq. (22), and changing summation to integration, one obtains
\[
\frac{1}{D} \int \Psi_{k'} w_i(E_{k'}) w_{i'}(E_{k'} + \omega_{i'b',c'}) |\Psi_{i'}\rangle^2 dE_{k'}. 
\]

This result can be written in the following form:
\[
\sum_{k'} w_i(E_{k'}) w_{i'}(E_{k'} + \omega_{i'b',c'}) |\Psi_{i'}\rangle^2 
\times D_{i'} \delta(\Gamma^{(i)}_{\text{spr}}, \Gamma^{(i')}_{\text{spr}}, \Delta). 
\]

In this expression
\[
\delta(\Gamma^{(i)}_{\text{spr}}, \Gamma^{(i')}_{\text{spr}}, \Delta) \equiv \frac{1}{D} \int \Psi_{k'} w_i(E_{k'} + \omega_{i'b',c'}) |\Psi_{i'}\rangle^2 dE_{k'}. 
\]

where \( \Delta \equiv E_{i'} - E_i - \omega_{i'b',c'} \), is a “spread” \( \delta \) function which was studied in Refs. [2, 3, 6]. It peaks at \( \Delta = 0 \) and describes the approximate energy conservation for the transition between compound states induced by the two-electron transition \( ch \to ab \) and broadened by the spreading widths. For the Breit-Wigner strength functions one has
\[
\delta(\Gamma^{(i)}_{\text{spr}}, \Gamma^{(i')}_{\text{spr}}, \Delta) \equiv \frac{1}{2\pi} \Gamma^{\text{spr}}_{i'} \Delta^2 + \Gamma^{\text{spr}}_{i'} / 4, 
\]

where \( \Gamma^{\text{spr}}_{i'} = \Gamma^{(i)}_{\text{spr}} + \Gamma^{(i')}_{\text{spr}} \). From Eqs. (19) and (25), the mean-squared matrix element of the two-body operator between the compound states is finally obtained as
\[
\frac{1}{4} \sum_{abch} |\langle ab | \hat{v} | hc \rangle - \langle ba | \hat{v} | hc \rangle|^2 
\times \langle \hat{n}_i \hat{n}_{c'} (1 - \hat{n}_a) (1 - \hat{n}_b) \rangle, 
\]

In this expression the summation is carried out over the single-electron states \( a, b, c, \) and \( c' \). Note that if \( |\Psi_i\rangle \) is a simple, unmixed state, there is no sum over \( k' \) in Eqs. (21)–(23). In this case \( \Gamma^{(i)}_{\text{spr}} = 0 \) and \( \Gamma^{(i')}_{\text{spr}} = \Gamma^{(i')}_{\text{spr}} \) in Eq. (27).

For a one-body operator \( \hat{M} = \sum_{ab} \hat{M}_{ab} a^+_b a^+_a \), the mean-squared matrix element is obtained similarly [2, 3, 6]:
\[
\frac{1}{4} \sum_{ab} |\langle ab | \hat{v} | hc \rangle - \langle ba | \hat{v} | hc \rangle|^2 
\times D_{i'} \delta(\Gamma^{(i)}_{\text{spr}}, \Gamma^{(i')}_{\text{spr}}, \Delta, J). 
\]

where \( \omega_{i'a} = \varepsilon_{b} - \varepsilon_{a} \) is the energy of the single-electron transition \( a \to b \).

D. Capture cross section and autoionization width

The autoionization width \( \Gamma^{(i)} = 2\pi |\langle \Psi_i \hat{V} | \Psi \rangle|^2 \) gives the transition rate between the initial state, \( e^- \to A^+ \), and the multiply exited compound resonance of the ion \( \Lambda^{(q-1)+} \) due to the two-body Coulomb interaction \( \hat{V} \).

Unlike the complex multiply excited states \( |\Psi_i\rangle \), the initial state of the recombinaton process is simple. It describes an electron with the energy \( \varepsilon \) incident on the ground (or low-lying excited) state \( |\Phi_0\rangle \), which is often dominated by one configuration. It is clear that the autoionization width averaged over compound resonances is determined by the mean-squared matrix element of the Coulomb interaction between electrons, which is given by Eq. (28). The initial state \( |\Psi_i\rangle = |\Phi_0, c\rangle \) is thus a compound state with negligible spreading width \( \Gamma^{(i)}_{\text{spr}} \ll \Gamma^{(i')}_{\text{spr}} \). The total width of the function \( \delta \), Eq. (27), is dominated by the compound width resonance \( \Gamma^{(i')}_{\text{spr}} \approx \Gamma^{(i')}_{\text{spr}} \).

Nonzero contributions to the sum in Eq. (28), i.e., to \( \Gamma^{(i)} \), arise from the basis states which differ from the initial state \( |\Phi_0, c\rangle \) by the single-particle states of two electrons. Therefore, it is sufficient to sum over the doubly excited basis states,
\[
\Gamma^{(i)} = 2\pi \sum_{d} C_d^{(i)} |\langle d | \hat{V} | \Phi_0, c \rangle|^2. 
\]

Such two-electron excitations \( |\Phi_d\rangle \) play the role of \( \text{doorway states} \) for the electron capture process. Since these states are not the eigenstates of the system they have a finite energy width \( \Gamma^{\text{spr}} \). The wave function of a doorway state can be constructed using the creation-annihilation operators, \( |\Phi_d\rangle = \sum_{ab} a^+_b a^+_a |\Phi_0, c\rangle \), where \( a \equiv n_{a'^j} m_{a'^j} \) and \( b \equiv n_{b} m_{b} \) are excited single-electron states, and \( \hat{c} \equiv n_{b} m_{b} \) corresponds to the hole in the target ground state. Of course, to form the doorway states with a given total angular momentum \( J \), the excited electrons and the ionic residue must be coupled into \( J \). However, the \( 2J + 1 \) factor and summation over \( J \) in Eq. (8) account for all possible couplings. This means that the sum over the eigenstates in Eq. (8) can be replaced by the sum over the one-hole-two-electron excitation, as in Eq. (28), and one obtains the capture cross section in the form of Eq. (1).

Note that when the number of active electrons and orbitals is large, the occupation numbers for different orbitals become statistically independent. In this case, the correlated product of the single-particle occupations, Eq. (23) can be approximated by the fractional occupation numbers of the electronic \( \text{subshells} \) with definite \( j \). The orbital \( c \) is taken as a continuum, \( c \equiv \ell|jm, \) in Eq. (1). Its wave function is normalized to the \( \delta \) function of energy, and it is occupied in the initial state, i.e., \( \hat{n}_c = 1 \).
After summation over the magnetic quantum numbers \( m_a, m_b, \text{etc.} \), and angular reduction of the Coulomb matrix elements, the final expression for the capture cross section is

\[
\sigma_c = \frac{\pi^2}{k^2} \sum_{\alpha h, lj} \frac{\Gamma_{\alpha h l}}{\sum_{\alpha h, lj} (\varepsilon - \varepsilon_a + \varepsilon_b + \varepsilon h)^2 + \Gamma_{\alpha h l}^2/4} \times \sum_{\lambda} \langle a, b || V_{\lambda h l} || h, \ell l j \rangle / 2\lambda + 1 \times \langle a, b || V_{\lambda h l} || h, \ell l j \rangle - (2\lambda + 1) \times \sum_{\lambda', (-1)^{\lambda + \lambda' + 1}} \left\{ \begin{array}{ll} \lambda' & j_a j_b \\ J & 0 \end{array} \right\} \langle b, a || V_{\lambda' h l} || h, \ell l j \rangle \times \frac{n_h}{2j_h + 1} \left( 1 - \frac{n_a}{2j_a + 1} \right) \times \left( 1 - \frac{n_b}{2j_b + 1} \right). \tag{30} \]

Here \( n_a, n_b \) and \( n_h \) are the occupation numbers of the corresponding subshells (ranging from 0 to \( 2j_a + 1, \text{etc.} \)), and \( \varepsilon_a, \varepsilon_b, \) and \( \varepsilon_h \) are their energies. The two terms in square brackets represent the direct and exchange contributions, and \( \langle a, b || V_{\lambda h l} || h, \ell l j \rangle \) is the reduced Coulomb matrix element,

\[
\langle a, b || V_{\lambda h l} || h, c \rangle = \sqrt{(2j_a + 1)(2j_b + 1)(2j_c + 1)} \times \xi(l_a + l_c + \lambda) \xi(l_b + l_h + \lambda) \times \left( \begin{array}{cc} \lambda & j_a j_b \\ 0 & 1/2 \end{array} \right) \left( \begin{array}{cc} \lambda & j_b j_a \\ 0 & 1/2 \end{array} \right) R_\lambda(a, b; h, c), \tag{31} \]

where \( \xi(L) = [1 + (-1)^L]/2 \) is the parity factor, and

\[
R_\lambda(a, b; h, c) = \int \int \frac{r^\lambda}{r^\lambda<1} [f_a(r)f_c(r) + g_a(r)g_c(r)] \times [f_b(r')f_h(r') + g_b(r')g_h(r')drdr'. \tag{32} \]

is the radial Coulomb integral, \( f \) and \( g \) being the upper and lower components of the relativistic orbital spinors.

Once \( \sigma_c \) is known, Eq. (8) allows one to estimate the average ratio \( \Gamma(a)/D \) for a typical \( J^\pi \),

\[
\langle \frac{\Gamma(a)}{D} \rangle = \frac{k^2(2j_1 + 1)}{\pi^2 \sum_{J_1}(2J_1 + 1)} \times \frac{\sigma_c}{k^2(2j_1 + 1)\sigma_c} = \frac{k^2(2j_1 + 1)\sigma_c}{2\pi^2 J_{\text{max}}^2}. \tag{33} \]

where the sum in the denominator is over the angular momentum and parity \( J^\pi \) which contribute effectively to the capture cross section. For example, \( J_{\text{max}} \approx 10 \) and \( J_1 = 6 \) for the recombination of \( \text{Au}^{25+} \) and \( \text{W}^{20+} \). A typical distribution of level densities \( \rho_{J^\pi} \) for different \( J \) is shown on the inset of Fig. 2.

### E. Radiative width

The second step of the recombination process is radiative stabilization. Any excited electron in the compound state \( |\Psi_c\rangle \) can emit a photon. Using Eq. (29), the total photoemission rate \( \Gamma^{(r)} \) can be estimated as a weighted sum of the single-particle rates,

\[
\Gamma^{(r)} \approx \sum_{a,b} \frac{4\omega_{ab}^2}{3\varepsilon_{ab}^3} |\langle a||d||b\rangle|^2 \left( \frac{n_b}{2j_b + 1} \right) \left( 1 - \frac{n_a}{2j_a + 1} \right), \tag{34} \]

where \( \omega_{ab} = \varepsilon_b - \varepsilon_a > 0 \), \( \langle a||d||b\rangle \) is the reduced dipole operator between the orbitals \( a \) and \( b \), and \( \langle \ldots \rangle \) is the mean occupation number factor. The mean subshell occupation numbers for a given energy can be obtained by averaging over the basis states involved, e.g.,

\[
n_a(E) = \sum_k \bar{c}_k^2(E) n_a^{(k)}, \tag{35} \]

where \( n_a^{(k)} \) is the occupation number of the subshell \( a \) in the basis state \( k \).

Since \( |\Psi_c\rangle \) have large numbers of principal components \( N \), the fluctuations of their radiative widths are small, \( \sim 1/\sqrt{N} \). This can also be seen if one recalls that a chaotic multiply excited state is coupled by photoemission to many lower-lying states, and the total radiative width is the sum of a large number of (strongly fluctuating) partial widths. A similar effect is known in compound nucleus resonances in low-energy neutron scattering [1, 16]. In multicharged ions with dense spectra of chaotic multiply excited states, the autoionization widths are suppressed as \( \Gamma(a) \propto 1/N \). Physically this happens because the coupling strength of the two-electron doorways state to the continuum is shared between many complex, multiply excited eigenstates. The radiative width does not have this suppression, since all components of a compound state contribute to the radiative decay. As a result, the radiative width may dominate in the total width of the resonances, \( \Gamma^{(r)} \gg \Gamma(a) \), making their fluorescence yield close to unity. Our numerical results for the recombination of \( \text{Au}^{25+} \) presented in [8], supported this picture.

### III. NUMERICAL RESULTS

In this section we apply our theory to calculate the recombination rate for the tungsten ions from \( \text{W}^{17+} \) to \( \text{W}^{24+} \). Experimental data are available for the recombination of \( \text{W}^{20+} \) forming \( \text{W}^{19+} \) [13]. We will use this system as an example to describe the calculations. Calculations for other ions are similar.

When an electron recombines with \( \text{W}^{20+} \), it can be captured into an excited state of the compound \( \text{W}^{19+} \) ion. Its ground state belongs to the \( 1s^2 \ldots 4f^9 \) configuration. Figure 4 shows the energies of its relativistic orbitals \( mlj \) obtained in the Dirac-Fock calculation. All orbitals below the Fermi level, \( 1s \) to \( 4f \), were obtained in the self-consistent calculation of the \( \text{W}^{19+} \) ground state. Each of the excited-state orbitals above the Fermi level: \( 5s, 5p, \text{etc.} \), was calculated by placing one electron into it, in the field of the frozen \( \text{W}^{20+} 1s^2 \ldots 4f^8 \) core. The energy of the highest orbit occupied (partially)
in the ground state is \( \varepsilon_{4f_{\frac{5}{2}}} = -18.41 \text{ a.u.} \). This value gives an estimate of the ionization potential of \( W^{19+} \): 
\( I \approx |\varepsilon_{4f_{\frac{5}{2}}}| = 18.41 \text{ a.u.} \) This value is in agreement with NIST data, \( I = 18.47 \text{ a.u.} \) [18].

Excited states of the ion are generated by transferring one, two, three, etc. electrons from the ground-state orbitals into the empty orbitals above the Fermi level (Fig. 4), or into the partially occupied \( 4f \) orbitals. We are interested in the excitation spectrum of \( W^{19+} \) near its ionization threshold. This energy (~20 a.u.) is sufficient to push up a few of the nine \( 4f \) electrons, and even excite one or two electrons from the \( 4d \) orbital. However, the preceding \( 4p \) orbital is already deep enough to be considered inactive. Thus, we treat \( W^{19+} \) as a system of 19 electrons above the frozen Kr-like \( 1s^2 \ldots 4p^5 \) core. Note that in constructing the excited-state configurations, we disregard infinite Rydberg series which correspond to the excitation of one electron in the field of \( W^{20+} \). Rydberg states belong to a single-particle aspect of the \( e^- + W^{20+} \) problem and are not expected to contribute much to the recombination cross section in this system.

Assuming that the fluorescence yield is close to unity (see below), we calculate the recombination cross section from Eq. (30). Before using this formula, one needs to obtain a list of two-electron-one-hole excitations of \( W^{19+} \) with energies close to the ionization threshold, which act as the doorway states. One also needs to estimate the spreading width \( \Gamma_{\text{spr}} \). For low-energy electron recombination, we restrict the consideration to the energy interval,

\[
E = I \pm \Delta E/2, \tag{36}
\]

where \( E = 0 \) for the ground state of the final-state ion, and we choose \( \Delta E \sim \Gamma_{\text{spr}} \). In practice, we start from some initial estimate of the spreading width, and subsequently find a more accurate value using an iterative procedure.

The spreading width is obtained from Eq. (13), where the mean-squared off-diagonal Hamiltonian matrix element \( \left[ \overline{H}_{ij} \right] \) is found by averaging over \( N_b \) basis states whose energies \( E_b \equiv \overline{H}_{bb} \) lie within the energy interval (36), and \( D = \Delta E/N_b \). The list of two-electron-one-hole excitations \( h^{-1}ab \) which contribute to the sum (30) includes configurations with basis states in the interval (36). It is known that the spreading width is a robust characteristic of the system. Indeed, we have checked that when more configurations are included, both \( D \) and \( \overline{H}_{ij} \) decrease, whereas \( \Gamma_{\text{spr}} \) does not change much (see also Ref. [9]).

When finding \( \overline{H}_{ij} \) and \( D \) we use basis states with definite projection of the total angular momentum \( J_z \) corresponding to the minimal value of \( J_z \) (0 or 1/2), rather than the states with definite total angular momentum \( J \). This method is significantly simpler than the use of the basis states with definite \( J \) and \( J_z \), and produces the same results for \( \Gamma_{\text{spr}} \), Eq. (13).

Table I shows the spreading widths for the compound ions of tungsten, \( W^{(q-1)+} \), with excitation energies close to the ionization threshold, formed in the process of low-energy electron recombination with \( W^{q+} \). With the exception of the target ion with the smallest number of \( 4f \) electrons (\( W^{25+} \)), the spreading widths are in the range 0.5–0.7 a.u. In fact, the value of \( \Gamma_{\text{spr}} \) does not strongly affect the magnitude of the capture cross section, Eq. (30), since the area under the Breit-Wigner contour corresponding to each doorway \( h^{-1}ab \) is independent of \( \Gamma_{\text{spr}} \).

As discussed in Sec. III.B, strong mixing of the basis states results in the eigenstates with large numbers of principal components, \( N \sim \Gamma_{\text{spr}}/D \sim \overline{H}_{ij}/D^2 \gg 1 \). This occurs when

\[
K \equiv \sqrt{\overline{H}_{ij}/D} \gg 1. \tag{37}
\]

Table I shows that this criterion is fulfilled for all the ions studied, and that the expected number of principal components is indeed large, \( N \sim 10^4 \). As explained in

### Table I: Electron capture cross sections \( \sigma_c \) and rate coefficients \( \alpha_c \) for the tungsten ions \( W^{(q)+} \) with the open \( 4f \) subshell, and properties of the compound ions \( W^{(q-1)+} \) at excitation energies close to the ionization threshold \( I \).

| Target ion | \( I \) | \( D \) | \( K \) | \( \Gamma_{\text{spr}} \) | \( \sigma_c \) | \( \alpha_c \) |
|------------|------|------|-----|-----------------|-------|-------|
| \( W^{25+} \) | 15.5 | 0.2  | 70  | 0.56            | 25    | 1.5   |
| \( W^{24+} \) | 17.0 | 0.1  | 93  | 0.65            | 29    | 1.7   |
| \( W^{23+} \) | 18.5 | 0.1  | 105 | 0.68            | 30    | 1.8   |
| \( W^{22+} \) | 20.0 | 0.1  | 96  | 0.68            | 34    | 2.0   |
| \( W^{21+} \) | 21.8 | 0.2  | 76  | 0.65            | 16    | 0.98  |
| \( W^{20+} \) | 23.5 | 0.4  | 48  | 0.59            | 11    | 0.67  |
| \( W^{19+} \) | 25.2 | 1.3  | 25  | 0.50            | 19    | 1.1   |
| \( W^{18+} \) | 27.0 | 11   | 5   | 0.16            | 12    | 0.7   |

\( ^{4} \)Ionization energy of the final-state ions, Ref. [18].

\( ^{5} \)Capture cross section from Eq. (30) and rate coefficient for incident electron energy \( e = 1 \text{ eV} \).
Sec. II E, in this case one can expect large fluorescence yields, \( \omega_f \approx 1 \). This means that the recombination cross section will be at the limit given by the total electron capture cross section, Eq. (30).

In the present calculations of \( \sigma_c \), Eq. (30), we also include in a semiempirical way the effect of screening of the Coulomb interaction between valence electrons by core electrons. This is done by introducing the screening factors \( f_3 \) in the two-electron Coulomb integrals, assuming that these factors depend on the Coulomb integral multipolarity \( \lambda \) only. The factors were calculated to be \( f_1 = 0.7, f_2 = 0.8, f_3 = 0.9 \) [19]. Coulomb integrals of other multipoles are not modified. The above values of the screening factors were found in the calculations for other atomic systems. However, in practice they change little from one atom to another.

To compare with experiment for \( W^{20+} \) [13], the cross section obtained from Eq. (30) is converted into the rate coefficient \( \alpha_c = \sigma_c v \), where \( v \) is the velocity of the incident electron. The result is shown in Fig. 5 by the solid line. Since the sum in Eq. (30) has a weak dependence on the electron energy, the capture cross section at low energies is proportional to \( 1/\epsilon \), and the corresponding rate coefficient behaves as \( \alpha_c \propto 1/v \). The calculated rate agrees well with the experimental data in the energy range of 0.1–1 eV. At higher energies the experimental rate coefficient tends to drop faster than \( 1/v \).

Figure 5 also shows the rate coefficient for the direct radiative recombination. The latter is estimated using the Kramers formula for the radiative recombination cross section [20] (in atomic units),

\[
\sigma_d = \frac{32\pi}{3\sqrt{3}} \frac{Z_i^2}{k^2} \ln \left( \frac{Z_i}{n_0 k} \right),
\]

where \( Z_i \) is the ionic charge \( Z_i \) (e.g., \( Z_i = 20 \) for \( e^- + W^{20+} \)), and \( n_0 \) is the principal quantum number of the lowest unoccupied ionic orbital (\( n_0 = 5 \) for \( W^{20+} \) [7]. The energy dependence of this cross section is close to \( 1/\epsilon \), and the corresponding rate coefficient (dashed line in Fig. 5) is 3 orders of magnitude smaller than the measurement in the energy range shown.

Below \( \epsilon = 0.1 \) eV the measured recombination rate coefficient can be affected by the velocity distribution of the electron beam, which is characterized by two temperatures, \( T_{\parallel} = 0.15 \) meV and \( T_{\perp} = 10 \) meV [13]. Taking this into account (see Eq. (18) in Ref. [8]) reduces the calculated resonant capture rate below 50 meV (dashed line in Fig. 5), bringing it into closer agreement with experiment. At higher energies, inelastic (electronic excitation) channels open, and the fluorescence yield can drop below unity, reducing the recombination rate relative to that of resonant capture. Also, the ground-state configuration of \( W^{20+} 4d^{10}4f^8 \) contains 293 closely spaced fine-structure levels [13]. A few of these are long lived, which may contribute to reduced values of the fluorescence yield.

As discussed above, the capture cross section has a simple \( 1/\epsilon \) energy dependence at low electron energies. Hence, in Table I we show the cross sections and rate coefficients for \( W^{q+} \) (\( q = 18–25 \)) calculated at one low electron energy, \( \epsilon = 1 \) eV. We see that the largest cross section is predicted for the ion with the half-filled 4f subshell. On the other hand, all the cross sections are within a factor of 3 of each other, and much larger than what one would expect from the direct RR process, Eq. (38).

Of course, one must keep in mind that compared with the capture cross section, the recombination cross section contains an additional factor \( \omega_f \). The fluorescence yield may be significantly smaller than \( \omega_f = 1 \) for ions, in which the degree of mixing is not as large as it is in the compound \( W^{19+} \) ion. In particular, this may be the case for the ions in which the mixing strength \( K \) (see Table I) and the number of principle components \( N \) is not too large. In this case one should regard \( \sigma_c \) as the upper limit, and use Eqs. (9) and (10) to estimate the recombination cross section.

IV. CONCLUSIONS

A detailed derivation of the statistical theory of resonant electron capture by many-electron ions has been presented. Numerical calculations have been performed for a number of tungsten ions with a partially filled 4f subshell. The calculated rate coefficient for \( W^{20+} \) is in agreement with the measurements at low electron energy. The present approach can be used to investigate other processes mediated by chaotic, multielectronic excited states.

Note added: Recently we became aware of the work by Badnell et al. [21]. Their extensive calculation of dielectronic recombination of \( W^{20+} \) underestimates the experimental recombination rate at low energies by a factor of...
3, but it achieves agreement with experiment by “partitioning” the autoionization rates using the Breit-Wigner distribution with a spreading width of 10 eV.

Acknowledgments

This work was funded in part by the Australian Research Council. We thank S. Schippers for providing the experimental data in numerical form, and acknowledge helpful conversations with J. Berengut. G.G. is grateful to the Gordon Godfrey Fund (UNSW) for support.

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