Secondary threshold laws for multiple ionization of atoms

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We propose a physical mechanism that leads to the emergence of secondary threshold laws in processes of multiple ionization of atoms. We argue that the removal of n electrons ($n>2$) from a many-electron atom may proceed via intermediate resonant states of the corresponding doubly charged ion. For atoms such as rare gases, the density of such resonances in the vicinity of subsequent ionization thresholds is high. As a result, the appearance energies for multiply charged ions are close to these thresholds, while the effective power indices μ in the near-threshold energy dependence of the cross section, $\sigma \propto E^{\mu}$, are lower compared to those from the Wannier theory. This provides a possible explanation of the recent experimental results of B. Gstir *et al.* [Nucl. Instrum. Methods Phys. Res. B **205**, 413 (2003)].

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I. INTRODUCTION

In this paper, we propose a physical mechanism that may lead to the emergence of secondary threshold laws in processes of multiple ionization of atoms. Among the processes leading to production of multiply charged fragments, multiple ionization of an atom by electron impact,

$$e^- + A \rightarrow A^{n+} + (n+1)e^-, \tag{1}$$

is probably the one most easily accessible experimentally (see, e.g., [1]). Another widely studied process is multiple photoionization. Below we discuss both reactions in parallel, denoting by $(n+1)$ the number of continuum electrons in the final state. In general, the near-threshold energy dependence of the cross sections of such processes is determined by the so-called Wannier mechanism [2,3].

Originally, Wannier developed a threshold theory for the simplest case of only two electrons receding from a charged core ($n=1$) [2]. His treatment was based on the idea that as the energy threshold is approached from above, the kinetic energies of all the electrons tend to zero simultaneously. *Slow* electrons spend a significant time in the region of space where the influence of the *long-range* Coulomb interactions is important. This results in a highly correlated electron motion. A detailed development of this idea leads to a power-law energy dependence of the ionization cross section $\sigma_n(E)$ near the threshold,

$$\sigma_n(E) = C_1 E^{\mu_n}, \tag{2}$$

where E is the excess energy above the multiple ionization threshold. The threshold index μ_n depends on the basic parameters of the final state of the system [the number of receding electrons $(n+1)$ and the charge of the residual ion Z], and bears a signature of strong electron correlation. Evaluation of the constant C_1 is usually beyond the threshold theory [4].

The case of multiple ionization with more than two outgoing electrons in the final state was studied by a number of authors, starting from Klar and Schlecht [5] and Grujić [6] ($n=2$) and Grujić [7] ($n=3$). A comprehensive bibliography on the subject can be found in the paper by Kuchiev and Ostrovsky [8], where a general threshold theory for the break-up of a particle into an arbitrary number of fragments with different masses and charges was developed. Further references can be found in recent experimental and theoretical works [9–12].

Wannier himself considered *multiple* ionization ($n>1$) neglecting electron correlation [13]. In this approximation, he obtained a simple estimate,

$$\mu_n \approx n, \tag{3}$$

which follows from a statistical (phase space volume) argument for the $n+1$ continuum electrons. Wannier remarked that the actual value of the threshold index μ_n was "probably slightly larger" than n due to electron correlation, although he did not go further. Equation (3) was obtained independently by Geltman [14,15]. It does in fact underestimate the exact threshold indices μ_n obtained when electron correlation is properly accounted for.

Establishing the energy range where the cross section behaves according to a threshold law is a difficult question. In most cases, it is beyond the threshold theory. An attempt to describe a deviation from Eq. (2) *intrinsic* to the Wannier mechanism for the three-particle fragmentation ($n=1$) was made in Ref. [16]. Alternatively, the range of validity of the threshold law can be limited if a different reaction mechanism becomes operational and dominant when the excess energy E is greater than some finite (albeit small) value $E_{\rm sec}$. In this situation, the mathematically rigorous primary threshold law valid in the limit $E \rightarrow 0$ can be masked and effec-

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TABLE I. Theoretical and experimental Wannier threshold indices.

Index	$n=1$	$n=2$	$n=3$	$n=4$	$n=5$	$n=6$	$n=7$
μ_n [a]	1.12689	2.27043	3.41938	4.62066	5.80400	7.05504	8.31522
$\mu_n^{(\mathrm{expt})}$ [b]	1.127 ± 0.05	2.11 ± 0.2	2.75 ± 0.5	3.15 ± 0.9	4.01 ± 1.4	3.89 ± 1.4	

[a]Wannier-type theory for $n=1$ [2], $n=2$ [5,6,8], $n=3$ [7,8], $n=4$ [8], and $n=5-7$ [22]. Note that for an unknown reason, some theoretical values cited in the experimental papers [9–11] differ somewhat from those provided in the original publications.

[b]Experimental values for Ar [9] are typical for all rare-gas atoms.

tively replaced by a different *secondary* threshold law at $E>E_{\mathrm{sec}}$. In this case, an experimental observation of the primary threshold law in the range $0<E<E_{\mathrm{sec}}$ may prove to be difficult or impossible due to the smallness of E_{sec} and/or a weak reaction yield.

It seems that historically the existence of a secondary threshold law was inferred for the first time empirically from experiments on triple photoionization of O and Ne atoms by Samson and Angel [17]. However, no physical mechanism to underpin the observation was put forward. A theoretical justification of this particular law was claimed in Ref. [18] but no actual theory has ever been published. The assertion of Ref. [18] that the secondary threshold law arises within the Wannier mechanism due to a previously unaccounted for unstable mode in the configuration of the receding electrons is disproved by a simple count of the modes [8].

In principle, these developments do not rule out the existence of secondary threshold laws based on different reaction mechanisms, as discussed above. The Wannier mechanism itself is highly universal, in the sense that the threshold index μ_n does not depend on either the reaction type and reactants (e.g., ionization by electron or photon impact) or the initial state and the structure of residual ion A^{n+} (a bare atomic nucleus or a multicharged many-electron ion in the ground or excited state). However, some of these features may play a crucial role when secondary threshold laws are concerned.

It seems that the first clear indication of a secondary threshold law based on a well-defined physical mechanism was obtained only recently in experiments on triple photoionization of Li atoms [19]. Here the new reaction pathway arises due to a strong nonequivalence of the electron orbitals in the ground state of Li. It was described as a two-step process where a double photoionization of the $1s^2$ inner shell is followed by the shake-off of the weakly bound outer $2s$ electron. The authors argue that this separation is meaningful when the excess energy is of the order of, or larger than, the $2s$ binding energy (5.39 eV) which provides a characteristic value of E_{sec} [20].

II. RESONANCES AND SECONDARY THRESHOLD LAWS

In this paper, we put forward another mechanism which leads to secondary threshold laws due to a many-electron nature of the residual ion A^{n+}. The inspiration comes from the recent experiments on near-threshold multiple ionization of rare-gas atoms by electron impact [9–11]. The empirical threshold indices μ_n were extracted by fitting experimental data for n from 1 to 8. In Table I, the experimental values for Ar are compared with the predictions of Wannier-type threshold theories for reaction (1) [21].

The error bars for high n are large and the procedure of fitting the experimental data to a power law could possibly be improved [23]. Nevertheless, the trend is clear. There is a good agreement with the Wannier theory for $n=1$ and 2, but starting from $n=3$ the experimental values are systematically lower than the predictions of both the Wannier theory and the statistical independent-electron model (3) [24]. In contrast with the monotonic increase of the theoretical values of μ_n, the experimental data exhibit a maximum for $\mu_n^{(\mathrm{exp})}$ at $n=5$. This feature looks even more prominent when one examines the whole sequence of rare-gas atoms from Ne to Xe. This disagreement cannot be attributed to the structure of the initial state for the active electrons, since, unlike in Li [19], all these electrons are in the equivalent p states.

As discussed above, the Wannier-type threshold law is governed by the long-range Coulomb interaction of the escaping electrons. It does not depend on the details of the interaction in the inner region of configurational space where the electrons are close to each other and to the ionic residue. In particular, the effects of resonances in the compound system are completely disregarded in the rigorous mathematical derivation of the threshold law in Wannier-type theories. However, such resonances may be of importance for a secondary threshold law, since they provide a different reaction mechanism with a distinct intermediate state.

Let us first consider the effect of an isolated resonance of the doubly charged ion, A^{2+*},

$$e^- + A \to A^{2+*}(\nu) + 3e^- \to A^{n+} + (n+1)e^-, \qquad (4)$$

where ν denotes the set of quantum numbers specifying the resonant state, and we assume that $n\geqslant 3$. Of course, for $n\geqslant 4$ one can also consider intermediate resonant states of the triply charged ion A^{3+} ($n\geqslant 4$) along the same lines, as well as a generalization to higher charge states.

In order to discuss energetics of the process, we introduce some notation. Let $I_{0\to n}$ be the nth ionization potential of the neutral atom A, i.e., the minimum energy required to remove n electrons from the atom initially in the ground state. Let $I_{2\to n}$ be the minimum energy necessary to remove $(n-2)$ electrons from the ground state of the doubly charged ion A^{2+}. By virtue of energy conservation, we have $I_{0\to n}=I_{0\to 2}+I_{2\to n}$, which is the energetics of sequential n-fold ionization. Let the energy of the resonance involved in the sequential process (4) be E_ν with respect to the ground state of A^{++}. For this process to occur, the resonance must lie above the

*n*th ionization threshold. A secondary threshold law is likely to be observed if the difference $\Delta E_\nu \equiv E_\nu - I_{2\to n}$ is small, i.e., the resonance lies only slightly above the threshold of $(n-2)$-fold ionization of A^{2+}. The energy of the resonance with respect to the ground state of the neutral atom is $E'_\nu = E_\nu + I_{0\to 2}$. Hence, one can also define ΔE_ν as the resonance energy excess above the *n*th ionization threshold, $\Delta E_\nu = E'_\nu - I_{0\to n}$.

Let ε be the kinetic energy of an incident electron in the multiple ionization process (1). The excess energy is then defined as $E = \varepsilon - I_{0\to n}$. If $E > \Delta E_\nu$, then the resonance state might be excited and the pathway (4) is energetically allowed.

If we neglect the unstable nature of the resonance (i.e., assume that its width Γ_ν is small), the process of double ionization by electron impact leading to the "final" state $A^{2+*}(\nu)$,

$$e^- + A \to A^{2+*}(\nu) + 3e^-, \tag{5}$$

will be well defined. Its threshold behavior is described by the Wannier-type power law,

$$\sigma_{2\nu}(E) \propto (E - \Delta E_\nu)^{\mu_2}\theta(E - \Delta E_\nu). \tag{6}$$

Here $\theta(x)$ is the step function: $\theta(x)=0$ for $x<0$, and $\theta(x)=1$ for $x>1$. Since the resonance state eventually decays, $A^{2+*}(\nu) \to A^{n+} + (n-2)e^-$, its excitation cross section cannot be defined completely rigorously. Instead one may say that $\sigma_{2\nu}(E)$ provides a contribution of the resonance to the observable yield of multicharged ions A^{n+}. Bearing in mind that the direct contribution is given by Eq. (2), we conclude that the observable yield will have the following energy dependence close to the threshold:

$$\sigma_n(E) = C_1 E^{\mu_n} + C_{2\nu}(E - \Delta E_\nu)^{\mu_2}\theta(E - \Delta E_\nu). \tag{7}$$

The constants C_1 and $C_{2\nu}$ determine the weights of the two contributions and are governed by details of the ionization dynamics. Since $\mu_n > \mu_2$ for $n \geqslant 3$, the first term is heavily suppressed close to threshold and the second term can quickly overtake it at $E > \Delta E_\nu$.

In Eq. (7), we neglect the interference between the two physically distinct pathways since only one of them dominates at any given energy E. Another reason which allows one to neglect interference is that the final states corresponding to the two mechanism are sufficiently different. The direct mechanisms assumes a simultaneous strongly correlated recession of all $(n+1)$ electrons, while in the resonant mechanism the emission of the $n-2$ electrons is delayed. Therefore, Eq. (7) displays a secondary threshold behavior, with the secondary threshold at $E_{\rm sec} = \Delta E_\nu$. The primary Wannier-type threshold behavior is operational only in the excess energy interval $0 < E < \Delta E_\nu$.

In a realistic experimental situation, one has to account for the energy spread of the electron beam, which introduces an uncertainty into the excess energy E. As a result, the Wannier-type law may be completely masked by the secondary threshold law. This is likely to happen if a resonance is available just above the $(n-2)$-fold ionization threshold of A^{2+}, i.e., the value of ΔE_ν for a given atom is small.

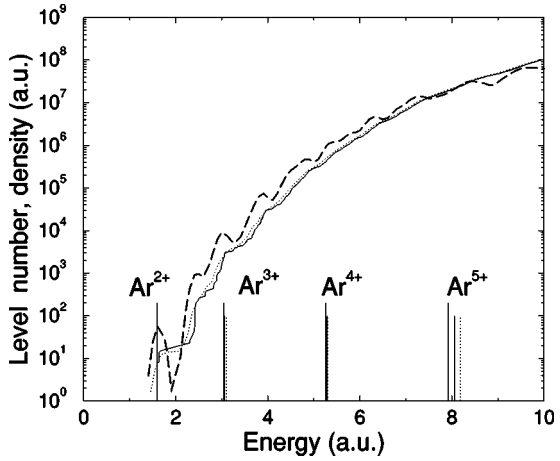

FIG. 1. The cumulative level number [Eq. (A4)] and density of multiply excited states of Ar^{2+}, as functions of energy above the Ar ground state. Dashed curve, smoothed level density; solid curve, $N(\mathcal{E})$; dotted curve, $N(\mathcal{E})$ obtained by integrating the smoothed level density. Vertical bars show successive ionization thresholds, as obtained from our calculations (long solid lines) and spectroscopic data (shorter solid lines), together with experimental appearance energies (dotted lines) [9–11].

Because of the closed-shell structure, the excitation spectra of neutral rare-gas atoms near the ionization threshold are relatively simple. In contrast, their doubly charged ions possess an open np^4 shell. The energies of the excited-state orbitals of the ion are lowered due to the higher charge of the core, reducing the relative size of the gaps in the single-particle excitation spectrum. Due to both of these factors, the spectrum of multiple excitations of the ion becomes quite complex and dense (see below). This means that the occurrence of a single or even several resonances in the immediate vicinity of the higher ionization thresholds is almost unavoidable, leading to a multitude of densely spaced secondary thresholds.

To verify this picture, we have carried out a calculation of spectra of multiply excited states of rare-gas atoms and their ions (see the Appendix). The typical results for Ar^{2+} are shown in Fig. 1. The main conclusion that one can immediately draw from the graph is that there are plenty of multiply excited states of Ar^{2+} in the vicinity of each subsequent threshold. Thus, near the Ar^{3+} threshold the density of multiply excited states is about 10^4 levels per a.u. After accounting for the $2J+1$ degeneracy of the levels with angular momenta J (with typical $J \sim 3$), we estimate that the spacing between neighboring resonances is of the order of 20 meV.

This means that as the energy E increases, not one but *many* autoionizing resonances will contribute to the secondary threshold law term. Their total contribution to the ionization cross section, $\sigma_n^{(2)}$, can be estimated as follows:

$$\sigma_n^{(2)}(E) = \sum_{\Delta E_\nu < E} C_{2\nu}(E - \Delta E_\nu)^{\mu_2} \approx C_2 \int_0^E (E - \Delta E_\nu)^{\mu_2}\rho(E_\nu)d(\Delta E_\nu) \tag{8}$$

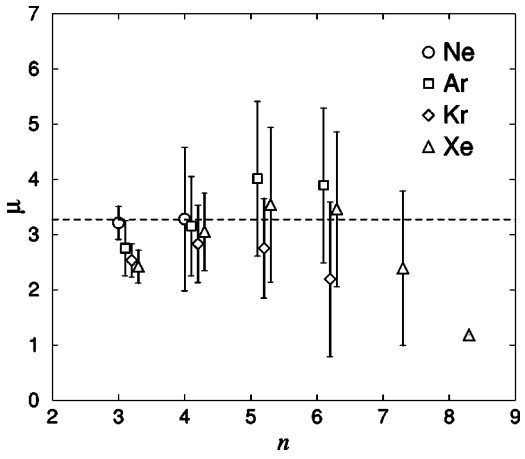

FIG. 2. Empirical threshold indices $\mu_n^{(\mathrm{expt})}$ with error-bars for various degrees of ionization n of rare-gas atoms [11]. The horizontal dashed line shows the value of $\mu^{(\mathrm{eff})}$, Eq. (10), predicted by the present theory.

$$\simeq \frac{C_2\rho}{\mu_2+1}E^{\mu_2+1}, \tag{9}$$

where in Eq. (8) we replaced $C_{2\nu}$ by its mean value C_2 and converted the sum into an integral, with $\rho \equiv \rho(I_{2\to n})$ being the density of resonant states at the threshold.

Equation (9) gives a power threshold law with an effective index

$$\mu^{(\mathrm{eff})} = \mu_2 + 1 = 3.27. \tag{10}$$

This is the key quantitative result of the present study. The value of $\mu^{(\mathrm{eff})}$ is universal since it does not depend on n ($n \geqslant 3$) or on the atomic species, as long as A^{2+} supports a dense spectrum of autoionizing states at higher ionization thresholds. Figure 2 shows that almost all experimental data listed in Ref. [11] agree with this result (Ref. [11] reports apparently more realistic error bars, compared to the previous publications [9,10] from the same group).

In a more general case, when multiple ionization proceeds via the near-threshold resonant states of the A^{q+} ion, the secondary threshold law would read $\sigma_n^{(q)} \propto E^{\mu_q+1}$. Comparison with experiment suggests that $q=2$ charge states play a major role. What could be the dynamical reason for this? In the context of the present work, it seems important that the doubly ionized rare-gas atoms have a sufficiently open-shell structure which promotes dense excitation spectra (Fig. 1). A paradigmatic example of open-shell systems with complex spectra is given by *d* elements, such as Fe and its ions. Rare-earth atoms exhibit an even greater degree of complexity related to the presence of several open shells and strong relativistic effects. (Thanks to the latter, neither the total spin nor the total orbital angular momentum is conserved, and the eigenstates can only be classified by their energy and total angular momentum.) Numerical studies in Ce indicate that configuration mixing in such systems reaches the extent described as *many-body quantum chaos* [25]. More recently, the effect of a dense spectrum of strongly mixed multiply excited states was invoked to explain huge enhancement of the low-energy electron recombination rates in Au^{25+}, which has a $4f^8$ ground state [26–28]. In this system, the resonances cannot be resolved experimentally due to extremely small level spacings between them. However, their energy-averaged contribution exceeds that of direct radiative recombination by a factor of 150.

In the above, we have neglected the effect of resonance widths on the threshold law. Owing to a finite resonance width Γ_ν, the transition to the secondary threshold law in Eq. (6) becomes smoother as the threshold energy ΔE_ν is smeared. Strictly speaking, the unstable nature of the resonance makes it impossible to separate the formation of the resonance from its final break-up. A similar physics manifests itself in the problem of the threshold behavior of a cross section for the formation of an unstable particle. In the case of short-range interactions, the latter was established by Baz' [29] and appeared recently in the problem of positron annihilation in positron-atom collision near the positronium formation threshold [30]. In the context of multiple ionization, it is worth mentioning the works on the post-collision interaction (PCI) between the receding Wannier pair and the autoionizing decay of a resonance in the ionic residue [31]. They show that PCI may change the shape and position of the autoionizing electron line. However, the total intensity of the process remains unchanged (provided the width is sufficiently small) and the cross section follows the Wannier law as if the residual ion were stable.

III. SUMMARY AND DISCUSSION

The present work indicates that non-Wannier threshold law indices observed in multiple ionization of rare-gas atoms can be a consequence of "invisible" unresolved resonances underpinning the two-step ionization process. The key element of this picture is the assumption of a strong interaction between several electrons in the open valence shells and excited-state orbitals, which allows such resonances to be populated.

It is customary to consider various processes in an approximation where only few electrons are treated as active, while all the rest play the role of spectators. Such an approach proved to be extremely fruitful in many cases. However, it may fail when a large amount of energy is deposited into an atomic system. In multiple fragmentation processes, the energy is brought in by the incident electron or photon. It is expended on ejecting two electrons, leaving a doubly charged ion in an autoionizing state lying high in the multielectron continuum. The high density of such states is essentially a many-body effect emerging due to a large number of ways in which the excitation energy can be distributed among the remaining electrons. In this compound state, no valence or subvalence electron remains a spectator.

Thus the nature of the present secondary threshold mechanism is of a statistical origin, but it differs substantially from earlier statistical approaches. Within the proposed mechanism, all the electrons are active electrons while previously only *n* atomic electrons were included in an analysis [32,33] within a statistical energy deposition model [34]. It also differs from the most recent classical trajectories Monte Carlo

calculations [35], where only single and double electron impact ionization was considered and multielectron effects were included only within a rather crude two-electron model. In contrast to the previous schemes, the present resonant mechanism cannot operate at all if the atom has only n electrons. It requires the number of valence and subvalence electrons (which can participate in the formation of resonances), n_{val}, to be greater than n. Moreover, for a small number of residual electrons, $n_{\mathrm{val}}-n$, the resonant mechanism is expected to give only a minor correction, while for multielectron atoms with large $n_{\mathrm{val}}-n$, the resonance contribution may become overwhelming.

Compare this situation with that in multiple ionization in a laser field, where a discussion of sequential and nonsequential mechanisms has continued for more than a decade. The present study suggests that in near-threshold multiple ionization by an electron or single-photon impact, the sequential mechanism dominates in many-electron processes (n-fold ionization with large n and $n_{\mathrm{val}}-n$). A similar resonant mechanism may be important in other processes, such as collisional multiple ionization by particles other than electrons (e.g., positrons or ions). Finally, we should mention that Koslowski *et al.* [24] detected the presence of metastable excited states of Ar^{2+} formed in near-threshold electron-impact ionization. This is an experimental indication that a similar process where *autoionizing* states of Ar^{2+} are populated can be behind the production of higher charged ions. The role of this mechanism could be verified by a careful measurement of the ejected electron spectrum.

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APPENDIX

Ar^{2+} has 16 electrons and its ground state belongs to the $1s^2\cdots 3p^4$ configuration. To determine its ionization potential, we start from a Dirac-Fock calculation of $Ar^{2+}3p^4$. A configuration interaction (CI) calculation which includes all relativistic $4p^4$ configurations shows that the ground state is characterized by the total angular momentum $J=0$ and total energy $\mathcal{E}_{\mathrm{tot}}=-527.175$ a.u. A similar calculation for the $J=\frac{3}{2}$ ground state of $Ar^{3+}3p^3$ gives $\mathcal{E}_{\mathrm{tot}}=-525.733$ a.u., yielding the ionization potential of Ar^{2+}, $I_{2\to 3}=1.442$ a.u. $=39.3$ eV. Similar calculations have also been performed for other ions of Ar.

In this work, we are interested in the spectra of multiply excited states of Ar ions (in particular, Ar^{2+}). A set of excited single-electron orbitals nlj ($3d_{3/2}$, $3d_{5/2}$, $4s_{1/2}$, etc., up to $6g_{9/2}$) was obtained from the Dirac-Fock calculation in the field of the $Ar^{+}3p^5$ residue. For the purpose of the present study, we consider Ar^{2+} as a system of six active electrons above the frozen Ne-like core. Excited-state configurations are obtained by distributing these six electrons among the 30 orbitals from $3s_{1/2}$ to $6g_{9/2}$. The basic structure of the spectrum of multiply excited states of Ar^{2+} is found by calculating the energies of the configurations in the mean-field approximation,

$$\mathcal{E}_c=\mathcal{E}_{\mathrm{core}}+\sum_a \epsilon_a n_a+\sum_{a<b}\frac{n_a(n_b-\delta_{ab})}{1+\delta_{ab}}U_{ab}, \tag{A1}$$

and evaluating the numbers of many-electron states N_c in each configuration (multiplicity),

$$N_c=\prod_a \frac{g_a!}{n_a!(g_a-n_a)!}, \tag{A2}$$

where n_a are the orbital occupation numbers of the relativistic orbitals in a given configuration. In the equations above, $\epsilon_a=\langle a|H_{\mathrm{core}}|a\rangle$ is the single-particle energy of orbital a in the field of the core, $g_a=2j_a+1$, and U_{ab} is the average Coulomb matrix element for the electrons in orbitals a and b (see Refs. [26,28] where similar calculations were performed to estimate the level density in Au^{24+}).

Using this procedure, we have generated a list of about 80 000 configurations within 38 a.u. of the Ar^{2+} ground state, and calculated their energies and multiplicities. They can be used to evaluate the level density,

$$\rho(\mathcal{E})=\sum_c N_c\delta(\mathcal{E}-\mathcal{E}_c), \tag{A3}$$

and the cumulative level number,

$$N(\mathcal{E})=\int_{-\infty}^{\mathcal{E}}\rho(\mathcal{E}')d\mathcal{E}'. \tag{A4}$$

To obtain a smooth level density we simply replace the δ functions in Eq. (A3) with unit-area Gaussians, $\exp[-x^2/(2\sigma^2)]/\sqrt{2\pi\sigma^2}$, using $\sigma=0.1$ a.u. Good agreement between the result of numerical integration of the smoothed density and "raw" $N(\mathcal{E})$ in Fig. 1 confirms the validity of the smoothing procedure. Note that the level density has a characteristic $\exp(a\sqrt{\mathcal{E}_{\mathrm{exc}}})$ energy dependence on the excitation energy $\mathcal{E}_{\mathrm{exc}}$ above the Ar^{2+} ground state, where a is constant related to the single-particle spectrum density [36].

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