Candidates for laser cooling of atomic anions: La^- versus Os^-

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This brief report is a follow-up to the recent proposal to use La^- as another candidate, in addition to Os^-, in laser cooling of anions, which can then be used to cool antiprotons sympathetically. Using the relativistic configuration interaction formalism, we calculate the photodetachment cross sections of the upper laser cooling state La^- $5d6s^26p\,^3D_1$ and Os^- $5d^66s^26p\,^6D_{9/2}$. Our results show that La^- has a very similar two-photon detachment loss as Os^-, retaining it as another promising candidate for cooling antiprotons sympathetically.

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Ultracold antihydrogen atoms are of great scientific interest [1]. For example, they can be efficiently trapped for precise laser spectroscopy, enabling spectroscopic comparison with hydrogen. They are also ideal systems for direct measurement of gravitational acceleration of antimatter. To form antihydrogen below 1 K, it is crucial that the antiprotons be precooled before combining with the positrons [2]. It was proposed [3] that antiprotons can be sympathetically cooled by transferring their energy to some precooled anions. These anions are cooled via laser cooling, where they absorb a laser photon and undergo electric dipole ($E1$) transitions to a bound state of opposite parity.

In an earlier paper [4], our research group proposed a new candidate for laser cooling, La^-, in addition to Os^- [3,5]. Comparisons were made [4] between these two anions. For example, the $E1$ transition in La^- has a lower transition energy than that in Os^- (Fig. 1), but the upper state in La^- does not need to be repumped using a laser as will be necessary for Os^-. An ideal candidate anion would have only two bound states connected by an $E1$ transition. This is essentially true for La^-, whose upper state decays almost entirely back to the ground state even though there are other lower states. The Os^- upper state, however, can decay into another state, necessitating the need to repump it to maintain efficient laser cooling.

One important consideration in laser cooling is the two-photon detachment loss. The upper state of the anion may absorb a second photon, which kicks off the excess electron, and become neutralized. La^- would not be a competitive candidate to Os^- for laser cooling if its loss due to photodetachment is much larger. For this reason, we decided to compare the upper state photodetachment cross sections of La^- and Os^- at their corresponding photon energies.

As shown in Fig. 1, the possible detachment channels for La^- and Os^- in laser cooling are as follows.

$$\begin{aligned} La^- &: 5d6s^26p\,^3D_1 \rightarrow 5d6s^2\,^2D_{3/2} + \epsilon l_j, \\ Os^- &: 5d^66s^26p\,^6D_{9/2} \rightarrow 5d^66s^2\,^5D_{2,3,4} + \epsilon l_j \\ &\rightarrow 5d^76s\,^5F_5 + \epsilon l_j, \end{aligned}$$

where ϵl_j denotes $\epsilon s_{1/2}$ and $\epsilon d_{3/2,5/2}$. The multiple detachment thresholds in Os^- are due to its high laser photon energy, 1.067 eV. The Os I $5d^66s^2\,^5D_0, ^5D_1$ states are omitted because neither can make a total J value that is accessible by an $E1$ transition after coupling with a free s or d electron.

In computing the cross sections for the above channels, we employ the relativistic configuration interaction (RCI) methodology. The wave functions for the anion bound state and the neutral thresholds were obtained by doing RCI calculations at the valence stage. The basis set consists of the reference configurations (Fig. 1) and correlation configurations that are one- or two-electron replacements of the reference configurations. The radial functions for the spinors occupied in the reference configurations are generated by the multiconfigurational Dirac-Fock code of Desclaux [8]. For those not occupied in the reference configurations, called virtual orbitals and denoted vl, their radial functions are represented by the relativistic screened hydrogenic functions (RSHs). The only adjustable parameter in an RSH function, the effective charge, Z^*, is determined via energy minimization.

One merit of the RCI formalism is that the basis set can be tailored to the atomic property under study. In a cross-section calculation, the energy values of the anion states and the neutral states are taken from the measurements or calculations in the literature (refer to the caption to Fig. 1). With energies being thus accounted for, in cross-section calculations it is adequate for the wave function to be able to yield the converged LS compositions for an anion state or a Landé g value that is in good agreement with the measurement in case of a neutral state. In addition, the radial space of the important configurations may need to be saturated by adding another virtual orbital of the same symmetry. In La^-, introducing a second set of virtual orbitals lowers the total cross section by $\sim$7% while maintaining excellent gauge agreement ($\leqslant$1%). The additional virtual p orbital helps saturate La^- $5d6s^2vp$, which is the only big contributor to the cross section other than the reference configuration. In Os^-, the addition of the second virtual orbital changes the cross section very little (<3%). This is consistent with the observation that the big contributors to the Os^- cross sections are exclusively the reference configurations.

Unlike the previous work [4], the $5d^2$ correlation was explicitly introduced in the basis set in this work. The near-degeneracy between the $5d$ and the $6s$ electron manifests itself as the mixing of $5d^26s6p$ ($\sim$18%) in La^- $5d6s^26p\,^3D_1$ and the mixing of $5d^76s6p$ ($\sim$6%) in Os^- $5d^66s^26p\,^6D_{9/2}$. A full *ab initio* treatment would explicitly correlate both configurations by applying the same one- and two- electron replacements to them. For Os^-, as discussed in the previous paper [4], it was thought that this would cause extensive second-order effects and lead to overcorrelation of $5d^76s6p$. The solution

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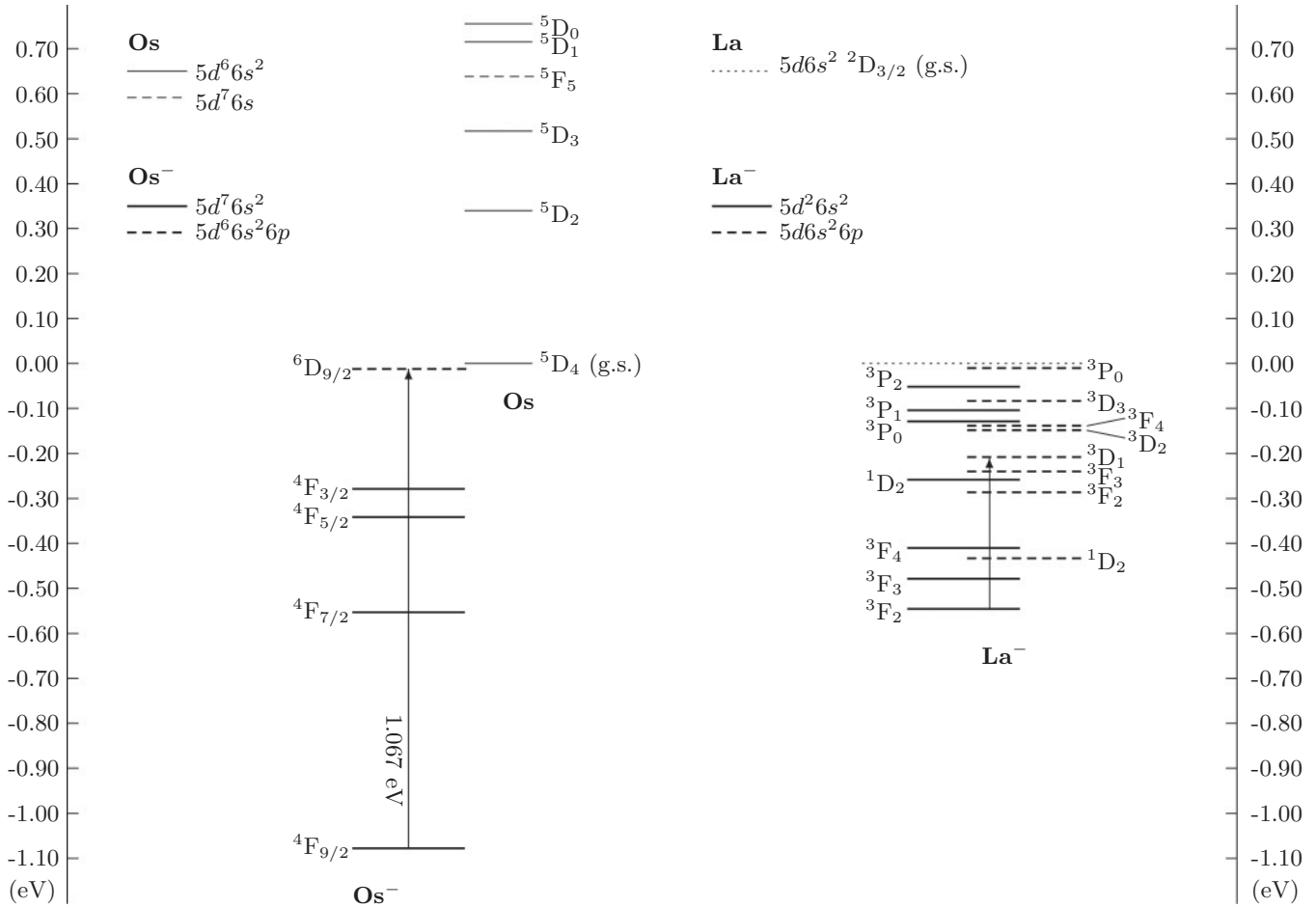

FIG. 1. Bound states of Os^- (Table I in Ref. [4]) and La^- (Tables VI and VII in Ref. [6]), together with neutral thresholds [7] that can be accessed by the laser photon. Arrows show laser cooling transitions. The energy of the transition in La^- is 0.337 eV.

then was to omit the $5d^2$ correlation but incorporate its effect to the wave function by shifting the related diagonal matrix elements. However, for the Os^- $^6D_{9/2}$ odd state studied here, including the $5d^2$ correlation does not seem to cause problems. While the $5d^2$ correlation in $5d^6 6s^2 6p$ does introduce to $5d^7 6s 6p$ the triple replacements $5d^3 \rightarrow 6sxl^2 + 6sxlx'l'$ (xl denotes either a Dirac-Fock orbital, nl, or a virtual orbital, vl), these replacements are going to be small. Experience has shown that large triple replacements are usually products of the large one- and two-electron replacements. The preceding $5d^3$ correlation can be viewed as a product of $5d \rightarrow 6s$ and $5d^2 \rightarrow xl^2 + xlx'l'$, but $5d \rightarrow 6s$ is small (e.g., contributes only several tens of milli–electron volts to $5d^7 6s$ in Os I). Therefore, we do not expect this $5d^3$ correlation to contribute much to $5d^7 6s 6p$. In contrast, these $5d^3$ replacements are not applicable to $5d^6 6s^2 6p$, whose $6s$ subshell is already full. In support of the foregoing argument, after explicitly incorporating the $5d^2$ correlation in the basis set, we obtain LS compositions almost identical to those in the previous calculation [4]. For the neutral states, the $5d^2$ correlation is also explicitly included and good agreements with the experimental Landé g values are obtained.

The $5d^2$ correlation produces a large number of basis functions, so we use the REDUCE method [9] to keep the basis size within the default 20 000 limit. Briefly, this method rotates the basis functions of a correlation configuration to maximize the number of functions that have zero matrix elements with the reference functions. These functions are then discarded, but those that have nonzero matrix elements are kept. With the REDUCE method, a reduction of a factor of 20 is achievable.

The continuum-state wave function is constructed by coupling the wave function of a free electron to that of a neutral state [10]. Assuming that the angular part of the free electron's wave function takes the same form as that of a bound electron, its radial function is numerically generated in a frozen-core Dirac-Fock potential, using a modified version [10] of the relativistic continuum wave solver code of Perger *et al.* [11,12].

The cross section is calculated using [13]

$$\sigma = 4\pi^2 \alpha\, a_0^2 \frac{df}{dE} = 8.067 \frac{df}{dE} \quad \text{(Mb)}, \tag{1}$$

where α is the fine-structure constant, a_0 is the radius of the first Bohr orbit, and $\frac{df}{dE}$ is the differential oscillator strength for the $E1$ transition from the anion bound state to the continuum state. $\frac{df}{dE}$ is evaluated using a modified version [14] of our code for bound states. This modified version has been used to reproduce the experimental photoelectron spectrum of Ce^- [15].

It is known that the presence of resonances may produce pronounced features in the photoelectron spectrum. Therefore,

it is important to mix the important resonance states into the wave function for the continuum state. An important resonance state, according to Fano's theory [16], should posses at least two properties: (1) it should be connected to the initial bound state by a strong *E1* transition; and (2) it should have a large mixing in the continuum state. In a previous work on Hf^- [17], we implemented Fano [16] and Mies's [18] theory to incorporate big resonance states. For La^- and Os^-, however, analysis shows no big resonance state to be present. In the La^- case, given the initial bound state $5d6s^26p\ ^3D_1$, the potentially important resonance in a $6p$ detachment would be $5d^26s^2$. RCI calculations have shown (see Fig. 1) that all the $5d^26s^2$ states lie below the neutral ground state except for the $^1S_0$, $^1G_4$ state. Since the initial $5d6s^26p$ state is dominantly $^3D_1$, transition to either $^1S_0$ or $^1G_4$ is not *E1* allowed. Similarly, for Os^-, the initial $5d^66s^26p$ state is dominated by sextuplets ($^6D_{9/2}$, $^6F_{9/2}$), but the potentially important resonance $5d^76s^2$ can make at most a quartet. Due to this lack of big resonance states, no mixing of resonances into the continuum was made.

The calculated cross sections for La^- and Os^- are summarized in Table I. The cross section to each neutral threshold is the sum of all the relevant relativistic channels. As can be seen, we obtained very good gauge agreement for La^- due to the careful selection of the $6p$ radial functions [6] for La^- $5d6s^26p$. The gauge agreement for Os^- is not at the same level. However, our results for Os^- are consistent with the order of estimates, 10^{-17} cm^2 (tens of megabarns) [5] and 5×10^{-17} cm^2 [19], both made by experimental data fitting.

TABLE I. Photodetachment cross sections (in both Babuskin gauge and Coulomb gauge) of the upper state of La^- and Os^- in laser cooling.

	Anion upper bound state	Neutral threshold	Cross section (Mb)	
			Babu. gauge	Coul. gauge
La^-	$5d6s^26p\ ^3D_1$	$5d6s^2\ ^2D_{3/2}$	**34.4**	**34.9**
Os^-	$5d^66s^26p\ ^6D_{9/2}$	$5d^66s^2\ ^5D_4$	30.1	36.6
		$5d^66s^2\ ^5D_2$	~0	~0
		$5d^66s^2\ ^5D_3$	0.6	0.5
		$5d^76s\ ^5F_5$	1.5	1.0
		Total	**32.2**	**38.1**

In conclusion, our calculations show that the upper states in laser cooling of La^- and Os^- have very similar photodetachment cross sections. In other words, La^- will have two-photon detachment loss very similar to that of Os^- if used in laser cooling. Combined with its other merits [4], La^- does make a promising candidate for laser cooling. It is our hope that this work will stimulate more experimental explorations of laser cooling of La^-, which can then be used to cool antiprotons to very low temperatures.

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