Isotope shift and search for metastable superheavy elements in astrophysical data

V. A. Dzuba,[1] V. V. Flambaum,[1,2] and J. K. Webb[1]
[1]*School of Physics, University of New South Wales, Sydney 2052, Australia*
[2]*Helmholtz Institute Mainz, Johannes Gutenberg University, 55099 Mainz, Germany*
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We explore a possibility that astrophysical data may contain spectra of metastable nuclei belonging to the island of stability where the nuclei have a magic number of neutrons of $N = 184$. The laboratory-produced nuclei have a significantly smaller number of neutrons. To identify spectra of the $N = 184$ isotopes of these nuclei and their neutron-rich superheavy decay products in astrophysical data we calculate the isotope shift which should be added to the laboratory-measured wavelengths. The results for the isotope shifts in the strongest optical electromagnetic transitions in No, Lr, Nh, Fl, and $Z = 120$ elements are presented.

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The possible existence of a hypothetical *island of stability* for superheavy elements (atoms with a nuclear charge of $Z \geqslant 104$) is an important problem of modern nuclear physics. All known nuclei with $Z > 98$ have short lifetimes, varying from fractions of seconds to hundreds of days. On the other hand, the theoretically estimated lifetime of the Fl nucleus with $Z = 114$ and $N = 184$ is 10^7 yr [1,2]. The nuclear shell model states that nuclei are usually more stable when both protons and neutrons fully occupy closed shells (doubly magic nuclei). For superheavy elements with spherical nuclei, the magic neutron number is believed to be 184, while magic proton numbers are $Z = 114$, $Z = 120$, and $Z = 126$ (see, e.g., Refs. [1,2]). (Magic numbers $Z = 122$ and $N = 172$, 178, 182, and 194 are also mentioned in the literature [1–4].) Therefore, the prospective candidates for the island of stability include $^{298}_{114}$Fl, $^{304}_{120}$Ubn, $^{310}_{126}$Ubh, and some other isotopes.

Superheavy elements are not found in nature but are produced in laboratories by colliding lighter atoms. All elements up to $Z = 118$ have been synthesized so far [2]. However, all superheavy elements synthesized in a laboratory are neutron-poor elements, with the number of neutrons being significantly smaller than what is required to make the most stable isotopes. For example, the heaviest isotope of Fl, produced in Dubna [1], $^{292}_{114}$Fl, is six neutrons short of the magic number $N = 184$. This is a common problem because the Coulomb repulsion energy for protons increases as Z^2, and to compensate for this by the attractive strong interaction energy, the number of neutrons N should increase with Z faster than the number of protons. Therefore, the very large number of neutrons, $N = 184$, needed for a more stable superheavy element, cannot be obtained by colliding any pair of lighter elements where the N/Z ratio is smaller than that in the island of stability.

There is an alternative way of searching for the island of stability by looking for traces of superheavy elements in astrophysical data (see, e.g., Ref. [5]). For example, optical lines of many actinide atoms and ions up to einsteinium (Es, $Z = 99$) have been possibly found in the spectra of the Przybylski's star [6] (the detection still needs to be further confirmed, as stressed, for instance, in Refs. [7,8]). Since in the laboratory these lines belong to short-living isotopes the natural question is how the isotopes were produced. Possibly, they were produced via decay of long-living elements with the magic number of neutrons $N = 184$ or have the magic number of neutrons themself (the production of superheavy elements with the magic number of neutrons $N = 184$ by supernovae is discussed, e.g., in Ref. [9]).

Heavy elements may be dispersed into the interstellar medium during supernova explosions and neutron star mergers. The neutron flux may be sufficiently high for the production of the $N = 184$ and other neutron-rich isotopes. If they are close to the island of stability they may have longer lifetimes and higher probability to be detected. So far this scenario has no solid observational or theoretical proof. To test a possibility of the existence of neutron-rich superheavy elements in nature one needs to know frequencies of the strong electric dipole transitions for superheavy elements to search for them in astrophysical spectra. The heaviest element for which one such frequency has been measured is No ($Z = 102$) [10]. Work is under way for similar measurements in Lr ($Z = 103$) [11–13]. There are good prospects for further progress in this field.

There are many high-quality atomic spectra calculations for superheavy elements (see, e.g., Refs. [14–21]). However, the accuracy of the calculations is not sufficiently high to reliably identify spectral lines in astrophysical data. A possible solution involves the following three-stage process.

(i) Measure the frequencies of strong electric dipole transitions in a laboratory-produced superheavy element. This will be a neutron-poor isotope.

(ii) To find the frequencies for the more stable neutron-rich isotope, the isotopic shifts are calculated and added to the laboratory frequencies.

(iii) The results are used to search for the spectral lines from the more stable neutron-rich isotope in astrophysical data (the methods and problems of such a search are discussed in Ref. [7]).

One needs to know the isotope shifts for superheavy elements to follow this path. The heaviest elements for which isotope shift experimental data are available are Pu, Am, Cm [22], and No [10]. These data can be used for searching heavier isotopes of the elements. However, no experimental data are available for the superheavy elements in the vicinity of the island of stability, such as $_{114}$Fl, $_{120}$Ubn, etc.

In the present work we calculate the isotope shift for some elements within the nuclear charge range $102 \leqslant Z \leqslant 120$, including candidates for the island of stability, Fl and Ubn. The isotope shift in superheavy elements is strongly dominated by the field (volume) shift [23]. Therefore, we ignore the mass

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shift and calculate only the energy shift due to the change of the nuclear charge radius. We assume a Fermi distribution for the nuclear charge with the radius given by $R_N = 1.1A^{1/3}$ fm with a skin thickness of 2.3 fm. Here A is the number of nucleons in the nucleus.

We use a combination of the configuration interaction (CI) method with the many-body perturbation theory (MBPT) to perform the calculations (the CI + MBPT method [24–26]). The field shift is obtained by repeating calculations with different values of nuclear radius and taking differences between the results. We consider only the strongest optical transitions from the ground state, which are the s-p electric dipole transitions to states of opposite parity having the same total electron spin (the change of the spin leads to a partial suppression of the $E1$ transition probability since the electric dipole operator conserves the spin).

Since $R_N \propto A^{1/3}$ it is convenient to approximate the calculated isotope shift ν by an analytic formula:

$$\nu = a\left(A_1^{1/3} - A_2^{1/3}\right). \tag{1}$$

The coefficient a in Eq. (1) is found by fitting to the results of the CI + MBPT numerical calculations. The values of a for No, Lr, Nh, Fl, and Ubn are presented in Table I.

For No there is an additional source of the information about the isotope shift. We may use the experimental value of the isotope shift: $\nu(^{252}\mathrm{No}) - \nu(^{254}\mathrm{No}) = 0.32\,\mathrm{cm}^{-1}$ (This value was presented in Ref. [10] without error bars. The paper with more detailed data is in preparation by the same group.) Using Eq. (1) for ^{252}No and ^{254}No leads to the value $a = 19\,\mathrm{cm}^{-1}$, which is about 1.8 times smaller than the calculated value $a = 34\,\mathrm{cm}^{-1}$ (see Table I). This illustrates a well-known fact that the formula $R_N \propto A^{1/3}$ generally does not work for neighboring isotopes (see, e.g., Ref. [27]). Indeed, the formula $R_N \propto A^{1/3}$ gives an average trend in the dependence of R_N on A while the actual change of the nuclear radius depends on what orbitals are occupied by the additional neutrons. Such deviations from the average trend are common in the studies of the isotope shifts. Thus, we may trust the dependence of $R_N \propto A^{1/3}$ only for a significant change in A when the shell model fluctuations are relatively suppressed in comparison with the average trend. This means that using experimental isotope shift for the ^{252}No and ^{254}No isotopes cannot give a reliable value for the constant a in Eq. (1). Instead, one should use atomic calculations to extract the change of nuclear radius from the isotope measurements. A corresponding paper is in preparation by experimentalists and theoreticians. Theoretical uncertainty of the present calculations comes mostly from electron correlations and does not exceed 20%. This has been found by comparing the results based on different methods of calculations as well as by comparing calculations with the measurements for other atoms.

The ^{254}No isotope of nobelium is the heaviest element for which an experimental frequency is available. The isotope with the magic neutron number $N = 184$ is ^{286}No. The difference in neutron numbers for these two isotopes is large, $\Delta N = 286 - 254 = 32$. Therefore, the $R_N \propto A^{1/3}$ trend should hold to a high precision. Using the calculated value $a = 34\,\mathrm{cm}^{-1}$ we get a prediction for the transition frequency in the ^{286}No isotope:

$$\begin{aligned}\nu(^{286}\mathrm{No}) &= 29\,961.457 + 34(254^{1/3} - 286^{1/3}) \\ &= 29\,952.8\ \mathrm{cm}^{-1}.\end{aligned} \tag{2}$$

It is important to have an independent way of estimating the isotope shift. We use analytic solutions for the isotope shift problem in the single-electron approximation. The accuracy of this approach is significantly lower than the accuracy of numerical many-body calculations. However, it helps to avoid mistakes and it can be used to extrapolate the isotope shift from lighter atoms (where experimental data are available) to heavier elements with a similar electron structure. Such an extrapolation makes sense since the relative value of the many-body corrections to the single-electron formula is approximately the same in atoms with similar structures of external shells.

In the nonrelativistic approximation the field isotope shift is not zero for the s orbital only. It is given by the Racah-Rosental-Breit formula presented, e.g., in Ref. [23]. This formula is derived in the first-order perturbation theory assuming that the finite nuclear size effect is small. The matrix element of the finite nuclear size perturbation is taken using the relativistic Coulomb wave functions for the pointlike nucleus. However, this is not correct when Z approaches 137. The finite nuclear

TABLE I. Isotope shift for strong electric dipole transitions from the ground state of some heavy elements. The shift is given by $\delta E = a(A_1^{1/3} - A_2^{1/3})$, where A_1 and A_2 are the atomic numbers of the two isotopes, the values of the parameter a are presented in the last column. $A_0 = Z + 184$ is the atomic number of the more stable isotope with the neutron number $N = 184$; A_s is the atomic number of the heaviest synthesized isotope. The "Frequency" column presents experimental (E) or theoretical (T) values for the frequency of the transition found in literature. Theoretical uncertainty is presented in parentheses.

Atom							Frequency		a
Z	Symbol	A_0	A_s	Transition			(cm^{-1})	Reference	(cm^{-1})
102	No	286	259	$7s^2\ {}^1S_0$	–	$7s7p\ {}^1P_1^o$	$29\,961.457^{+0.041}_{-0.007}$[a]	E [10]	34
103	Lr	287	266	$7s^27p\ {}^2P_{1/2}^o$	–	$7s^28s\ {}^2S_{1/2}$	20 253(500)	T [19]	–19
113	Nh	297	286	$7s^27p\ {}^2P_{1/2}^o$	–	$7s^28s\ {}^2S_{1/2}$	36 041(440)	T [21]	–17
114	Fl	298	292	$7p^2\ {}^1S_0$	–	$7p8s\ {}^1P_1^o$	43 876(310)	T [21]	–2.6
120	Ubn	304		$8s^2\ {}^1S_0$	–	$8s8p\ {}^1P_1^o$	27 559(200)	T [20]	148

[a]Experimental values for the $^{254}_{102}$No isotope. The theoretical value is 30 200(300) cm^{-1} [19].

size effect is not a small perturbation here because of the singularity of the relativistic wave functions and apparent collapse of the pointlike nucleus spectrum for $Z > 137$.

The formula for the isotope shift for p waves is not presented in the textbooks. Indeed, in the nonrelativistic approximation, the single-particle isotope shift for p orbitals is zero since the p orbitals vanish at the origin $r = 0$. However, the relativistic $p_{1/2}$ wave density near the nucleus is proportional to the s-wave density and has the same order of magnitude in heavy atoms. Below we present a formula for the single-particle isotope field shift which describes all waves including the more accurate treatment of the finite nuclear size (the latter leads to a difference with the Racah-Rosental-Breit formula for s waves).

If we model nuclear charge distributions as the electric charge homogeneously distributed inside the sphere of the radius R_N, the electrostatic nuclear potential inside is

$$U_N = -\frac{Ze^2}{2R_N}\left(3 - \frac{r^2}{R_N^2}\right).$$

The potential near the nucleus is the unscreened Coulomb potenatial $U_C = -\frac{Ze^2}{r}$. The relativistic wave functions in the small distance area $r \ll a_B/Z$ are proportional to the Coulomb wave functions and are presented in Eq. (4.5) in Ref. [28]:

$$f_{n\kappa} = \frac{\kappa}{|\kappa|}(\kappa - \gamma)\left(\frac{Z}{a_0^3\nu^3}\right)^{1/2}\frac{2}{\Gamma(2\gamma+1)}\left(\frac{a_0}{2Z}\right)^{1-\gamma} r^\gamma, \quad (3)$$

$$g_{n\kappa} = \frac{\kappa}{|\kappa|}Z\left(\frac{Z}{a_0^3\nu^3}\right)^{1/2}\frac{2}{\Gamma(2\gamma+1)}\left(\frac{a_0}{2Z}\right)^{1-\gamma} r^\gamma. \quad (4)$$

Here $\kappa = (-1)^{j+1/2-l}(j+1/2)$, $\gamma = \sqrt{(j+1/2)^2 - (\alpha Z)^2}$, j is the electron angular momentum, a_0 is the Bohr radius, ν is the effective principal quantum number (single-electron energy ϵ is given by $\epsilon = -1/(2\nu^2)$, and Γ is the gamma function. The electron charge density $\rho_e(r)$ calculated using these wave functions can be presented as $\rho_e(r) = Br^{2\gamma-2}$, where B is the normalization constant [see Eqs. (3) and (4)]. For $s_{1/2}$ and $p_{1/2}$ orbitals this density tends to infinity at $r = 0$. If we use this singular charge density for s waves to calculate the effect of the perturbation $U_N - U_C$ we obtain the Racah-Rosental-Breit formula. This approach overestimates the shift in the relativistic case of $Z > 100$.

A more accurate approach is to treat as the perturbation the small change of the nuclear potential $\delta U_N(r)$ due to the small change of the nuclear radius δR_N (instead of the large difference between the finite-size and the zero-size potentials $U_N - U_C$). The parameter of our perturbation theory is $\delta R_N/R_N \ll 1$.

For the nonsingular potentials the charge density $\rho_e(r) = Dr^{2j-1}$ for $r \ll R_N$. To estimate the normalization constant D we can extend this charge density to the nuclear surface and match it with the solution outside: $\rho_e(R_N) = DR_N^{2j-1} = BR_N^{2\gamma-2}$. This approach gives us the continuation of the electron charge density inside the nucleus, $\rho_e(r) = Dr^{2j-1}$, which approximates the true electron charge density inside the nucleus significantly better than the singular Coulomb density $\rho_e(r) = Br^{2\gamma-2}$. Integration of the perturbation $\delta U_N = \frac{3Ze^2}{2R_N}(1 - \frac{r^2}{R_N^2})\frac{\delta R_N}{R_N}$ with the electron charge density $\rho_e(r) = Dr^{2j-1}$ gives the following result for the single-particle isotope field shift:

$$\delta\epsilon = \frac{12\kappa(\kappa-\gamma)}{(2|\kappa|+1)(2|\kappa|+3)\Gamma(2\gamma+1)^2} \times \frac{|\epsilon|^{3/2}}{(Z_i+1)(\mathrm{Ry})^{1/2}}\left(\frac{2ZR_N}{a_B}\right)^{2\gamma}\frac{\delta R_N}{R_N}. \quad (5)$$

Here Z_i is the ion charge ($Z_i = 0$ in neutral atoms), R_N is the nuclear radius, ϵ is the energy of the orbital ($|\epsilon|$ is the ionization energy), and $\mathrm{Ry} = e^2/(2a_B) = 109\,737\,\mathrm{cm}^{-1}$. Note the simple relation between the isotope shifts for $p_{1/2}$ ($\kappa = 1$) and $s_{1/2}$ ($\kappa = -1$) orbitals in heavy atoms:

$$\delta\epsilon_p = \frac{1-\gamma}{1+\gamma}\left(\frac{\epsilon_p}{\epsilon_s}\right)^{3/2}\delta\epsilon_s. \quad (6)$$

Thus, in the superheavy atoms, where $(1-\gamma) \sim 1$, the $p_{1/2}$ shift is not suppressed significantly.

The "exact" analytical solution of this problem is very cumbersome and gives only small corrections to Eq. (5), $\sim$1%. These corrections as well as the detailed derivation of the single-particle isotope field shift will be presented in a future publication [29].

Equations (5) and (6) give $a = 160\,\mathrm{cm}^{-1}$ for Ubn in good agreement with the calculated value $a = 148\,\mathrm{cm}^{-1}$(see Table I). The difference can be attributed to the many-body effects neglected in Eqs. (5) and (6). The same formulas give $a = 9.3\,\mathrm{cm}^{-1}$ for Ra, which is a lighter analog of Ebn. On the other hand, the fitting of the experimental data [30] gives $a = 12\,\mathrm{cm}^{-1}$.

Note the very rapid increase of the isotope shift when the nuclear charge is approaching 120. Such an increase is clearly seen in both the analytic formulas and the numerical many-body calculations. This is the result of the relativistic effects mentioned above.

Another feature of the isotope shift is its smaller value in $7p_{1/2}$-$8s$ transitions in Lr, Nh, and Fl (see Table I). This is the result of the cancellation of the shifts of the $7p_{1/2}$ and $8s$ states. For example, for Nh the formulas give nearly the same shifts of the energies of the lower $7p_{1/2}$ and upper $8s$ states: $\delta\epsilon(7p_{1/2}) = 1.11\,\mathrm{cm}^{-1}$, while $\delta\epsilon(8s_{1/2}) = 0.97\,\mathrm{cm}^{-1}$. When such a cancellation occurs, the accuracy of the analytic formulas is low: the formulas give $a = -4.5\,\mathrm{cm}^{-1}$ for Nh in apparent disagreement with the result of the many-body calculations $a = -17\,\mathrm{cm}^{-1}$ (see Table I).

Thus, Eqs. (5) and (6) give a reasonable accuracy when an s state is lower than a p state and indicate cancellation when a $p_{1/2}$ state is lower than an s state.

We hope that this work provides motivation for further progress in the measurement of the transition frequencies for superheavy elements, the calculation of the isotope shifts, and the search for the corresponding transitions in astrophysical spectra.

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