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Systematics of zeros in dipole matrix elements for photoionizing transitions: Nonrelativistic calculations

Steven T. Manson
Department of Physics and Astronomy, Georgia State University, Atlanta, Georgia 30303

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The location of the zeros in dipole matrix elements for photoionizing transitions (Cooper minima) in ground states of atoms has been calculated for all subshells over the entire periodic system. These zeros occur for *all* subshells whose wave functions have nodes, except the 2*s* subshell. The behavior of these zeros, as a function of the atomic number Z, has been examined and their "trajectories" are rather complicated. The trajectories are explained primarily in terms of initial-state quantum defects and final-state phase shifts. All atoms with $Z>10$ have at least one zero, and some have as many as five zeros.

I. INTRODUCTION

Over the past two decades our understanding of the atomic photoionization process has increased dramatically.[1-5] The cross sections, photoelectron angular distributions, and spin-polarization parameters are indicators of fundamental interactions within the atom and, thus, are important in their own right. However the cross sections, which are related to optical oscillator strengths in the continuum by a simple multiplicative constant, are of still greater importance since many atomic (and atomic ion) properties are derivable from these oscillator-strength distributions;[16] among these are polarizabilities, Lamb shifts, and stopping power of matter for fast charged particles, to name a few. For these reasons then, an understanding of the details of atomic photoionization cross sections is very desirable.

In the nonrelativistic dipole approximation, which is excellent for photoionization cross sections below $\hbar\nu \sim 10$ keV,[1-5] the cross section is proportional to a weighted sum of the absolute squares of the $l \rightarrow l-1$ and $l \rightarrow l+1$ dipole matrix elements; only a single $l \rightarrow l+1$ in the case of *s*-electron photoionization, of course. An extremely interesting feature of the $l \rightarrow l+1$ dipole matrix element is the existence of zeros in a large number of cases. These occur over the entire Periodic Table for all outer and near-outer subshells whose wave functions have nodes, except for the 2*s* state. These zeros profoundly affect the shape of the cross section, i.e., the spectral distribution of oscillator strength. In addition, they also affect the photoelectron angular distribution and spin polarization very significantly. They were first discovered experimentally in the photoionization of alkali-metal atoms.[7] The basic explanation for the phenomenon in the alkali-metal atoms was given by Bates[8,9] and extended to include relativistic interactions by Seaton.[10] Later Cooper showed that this phenomenon was not limited to the outer shells of the alkali-metal atoms but was a very general occurrence.[11-13] These zeros have, thus, come to be called Cooper minima.

A myriad of Cooper minima have been uncovered both theoretically and experimentally.[1-5] Despite this wealth of data, there has been no systematic study of their behavior across the Periodic Table. This work is a first attempt in that direction. Simple central-field Hartree-Slater wave functions[14] have been used in our calculations; these wave functions have been shown to be good qualitative and fair quantitative predictors of atomic properties. Thus, while we do not expect that the positions of the zeros is terribly accurate, we believe that the systematic trends are correct.

Calculations have been performed over the entire range of the periodic system for all subshells which exhibit Cooper minima, i.e., all but the 1*s*, 2*s*, 2*p*, 3*d*, and 4*f*. Over half of all the elements have been considered. In Sec. II a brief review of the theoretical methodology is given. The results and discussion are given in Sec. III and a summary and conclusions are presented in Sec. IV.

II. BRIEF REVIEW OF THEORY

Within the framework of the central-field approximation, the photoionization cross section for an *nl* subshell of an atom is given in dipole approximation by[1-4]

$$\sigma_{nl}(\epsilon)=\frac{4\pi^2 a_0^2\alpha}{3}\frac{N_{nl}(\epsilon-\epsilon_{nl})}{2l+1} \times\{l[R_{l-1}(\epsilon)]^2+(l+1)[R_{l+1}(\epsilon)]^2\}\ , \tag{1}$$

where a_0 is the Bohr radius, α the fine-structure constant, N_{nl} the occupation number of the subshell, ϵ the photoelectron energy, and ϵ_{nl} the (intrinsically negative) subshell binding energy. The dipole matrix elements are given by

$$R_{l\pm1}(\epsilon)=\int_0^\infty P_{nl}(r)rP_{\epsilon,l\pm1}(r)dr \tag{2}$$

with P_{nl}/r and $P_{\epsilon,l\pm1}/r$, the radial parts of the discrete and continuum wave functions, respectively; the discrete wave function is normalized to unity and the continuum to unit energy in rydbergs, i.e.,

$$P_{\epsilon l}(r)\sim\pi^{-1/2}\epsilon^{-1/4}\sin[\epsilon^{1/2}r-\tfrac{1}{2}l\pi-\epsilon^{-1/2}(\ln 2\epsilon^{1/2}r)+\sigma_l(\epsilon)+\delta_l(\epsilon)] \text{ as } r\rightarrow\infty \tag{3}$$

with $\sigma_l(\epsilon)=\arg(l+1-i\epsilon^{-1/2})$, the Coulomb phase shift, and $\delta_l(\epsilon)$, the non-Coulomb phase shift.

The wave functions used were Hartree-Slater (HS) central-field functions for the initial discrete (ground) state and continuum functions which were solutions to the radial Schrödinger equation in the same central potential as the ground state, i.e., no core relaxation was considered.

III. RESULTS AND DISCUSSION

Calculations were performed from $Z=11$ to $Z=103$ for $ns\,(n=3-7)$, $np\,(n=3-6)$, $nd\,(n=4-6)$, and $5f$ subshells. Before presenting the detailed results, however, it is useful to discuss the general characteristics of Cooper minima. Generally both discrete and continuum states have wave functions which are oscillatory. In addition, the continuum wave function becomes more compact with increasing energy, i.e., its nodes move in towards the nucleus. In the complicated overlap of positive and negative amplitudes which go into the dipole matrix element, it may be that at some energy the positive and negative components just cancel giving a zero in the dipole matrix element. This is the Cooper minimum.

For atoms in their ground state, zeros are found only in the $l\rightarrow l+1$ transitions, never in the $l\rightarrow l-1$. There is, at most, one zero for a given $l\rightarrow l+1$ transition, in addition. Furthermore, the hydrogen atom shows no zeros in any state. This shows that the zeros must be due to deviations of the wave functions from hydrogenlike behavior. The dipole matrix element can be divided into two parts; the first is the contribution of the outer region where the potential has gone asymptotic and is purely Coulomb. In this region, then, the wave functions are hydrogenic with phase shifts (or quantum defects for discrete states) and the contribution of this region to the matrix element can be characterized by the continuum phase shift and discrete quantum defect in a manner much like the Bates-Damgaard method.[15] The second is the contribution of the inner region where the potential is completely non-Coulomb. The contribution of this region to the matrix element is complex and not described by simple parameters. For most Cooper minima, the contributions of both regions are of importance, but the above separation will still be of use in analyzing the systematics of the zeros as a function of Z.

A. *ns* subshells

The calculated "trajectory" of each of the $ns\rightarrow\epsilon p$ Cooper minima is shown in Fig. 1. In each case, the zero is in evidence as soon as the subshell becomes bound in the ground state, i.e., at each of the alkali-metal atoms. As Z increases the zero moves out, away from threshold, in all cases. Increasing Z further, this trend reverses and the zeros move to lower photoelectron energies, eventually moving below threshold (into the discrete spectrum) at high enough Z; this move back to lower energies is not seen for $7s$ because we run out of Z before this happens. Sample calculations for unphysical high-Z atoms ($Z>110$) confirm this point.

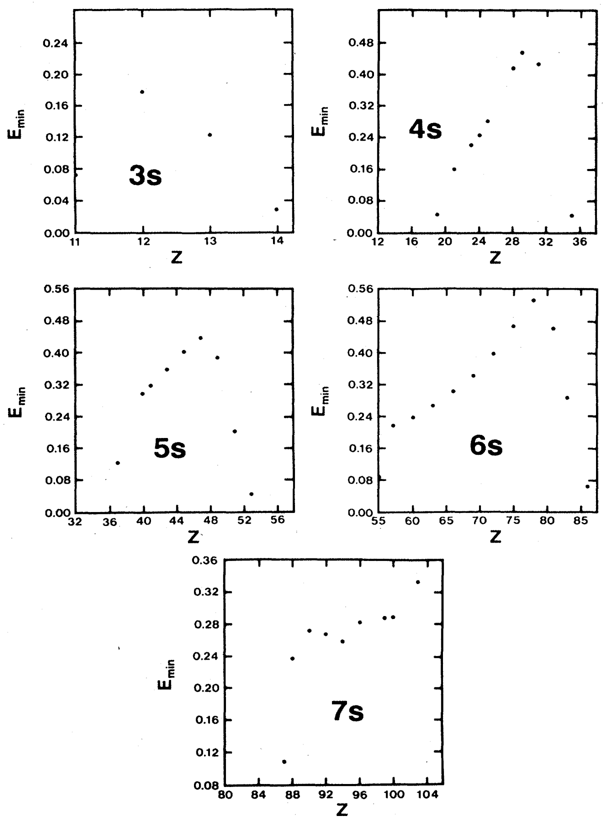

FIG. 1. Location of the zero (Cooper minimum) in the dipole matrix element for $ns\rightarrow\epsilon p$ ionizing transitions in energy in rydbergs above threshold ($E_{\min}$) vs atomic number Z for ground-state atoms.

To account for this behavior, recall, as discussed above, that deviations (phase shifts, quantum defects) of the wave functions from hydrogenic are required to produce zeros. Roughly speaking, if the hydrogenic wave functions are thought of as being "in-phase" sinusoids, then a relative phase shift between the initial- and final-state wave functions of about $\pi/2$ is needed to produce a zero. This, of course, ignores both the fact that the wave functions are not sinusoids and the effect of the inner region, but it is a useful zeroth approximation. The quantum defect of a bound state can be simply calculated from its binding energy[16] and phase shifts for Hartree-Slater continuum states are known.[17] Looking at $Z=11$, the relative phase difference between the bound $3s$ state and the threshold function is just about $\pi/2$. Thus, a zero is expected in the $3s\rightarrow\epsilon p$ matrix element right near threshold, which is just what is seen. Going up to $Z=12$, the relative phase difference increases (the $3s$ quantum defect increases more than the ϵp threshold phase shift) so that the zero occurs at higher energy where the ϵp has moved in enough for just the correct overlap.

For these cases, the $3s$ wave function is quite diffuse (as newly bound wave functions generally are) so that most of the overlap in the dipole matrix element occurs in the region of space where the atomic potential is effectively Coulomb. Going up in Z to $Z=13$, the zero moves to lower energy, towards threshold, despite the fact that the relative phase difference is actually very slightly larger. This is due to the overlap in the inner, non-Coulomb, region. More of the amplitude of the 3*s* wave function has moved to the inner region, with increasing Z, and the phase shifts (quantum defects) are smaller in this region (they are zero at $r=0$) as the phase-amplitude analysis shows.[18] Smaller phases translate to smaller relative phase differences which mitigates against zeros. This effect becomes even more pronounced by $Z=14$ where the zero has moved down almost to threshold; by $Z=15$ the inner region dominates and the zero is below threshold and remains there for all higher Z's. Thus it is the drawing in of the discrete wave function with increasing Z that ultimately causes the Cooper minimum to move below threshold.

This same analysis applies to the other *ns* states as well. The 2*s*, which has no Cooper minima, becomes bound at such a low Z ($Z=3$) that the relative phase difference is only about 0.3π, which is too small to cause a zero; for larger Z, where the relative phase difference is large enough, the 2*s* wave function is quite compact and the inner region is dominant.

The $4s\rightarrow\epsilon p$ Cooper minimum behaves in substantially the same manner as 3*s*, but with two important differences; for the 4*s* the increase from the location of the zero where it is first bound at $Z=19$ occurs over a much larger range of Z, and it moves out much further, ~ 0.5 Ry as opposed to ~ 0.1 Ry. Both of these differences are caused by the binding of the 3*d* subshell in the ground state at $Z=21$. In the 3*s* case, subsequent elements added electrons in the same $n=3$ shell, but in the 4*s* case, the 3*d* electrons that are added are *inside* the major portion of the 4*s* amplitude. Thus, in going up in Z from 20 to 21, the nuclear charge is increased by one and an electron is added in an orbital *inside* the 4*s* which essentially totally screens the 4*s* subshell from the increase in nuclear charge. The 4*s* wave function remains diffuse for all of the elements in which the 3*d* subshell is filling. This, combined with a small steady increase in the relative phase difference, means that the Cooper minimum moves further from threshold with increasing Z, as is seen in Fig. 1. This continues until the 3*d* subshell is filled at $Z=29$; the Cooper minimum moves rapidly towards threshold for still higher Z, disappearing into the discrete above $Z=35$.

The zero in the $5s\rightarrow\epsilon p$ channel virtually duplicates the behavior of the 4*s* Cooper minimum, first appearing at $Z=37$, moving away from threshold until $Z=47$ where the 4*d* becomes filled then moving closer to the threshold and moving into the discrete above $Z=53$.

The $6s\rightarrow\epsilon p$ Cooper minimum is quite similar to the 4*s* and 5*s* except that it is moving away from threshold from $Z=55$ where it first appears (and the 6*s* is first bound in the ground state) until $Z=79$ which is more than double the range of Z's for which the 4*s* and 5*s* Cooper minima move away from threshold. This is the result of not only the 5*d* electrons becoming bound inside of the 6*s* subshell, but the 4*f* electrons as well. Since there are 14 *f* electrons in a filled shell, it would be expected that the range of Z's at which the 6*s* Cooper minimum is moving away from threshold is increased by 14 from the ranges of 4*s* and 5*s*; this is exactly what is found.

Another difference, albeit slight, between the 6*s* Cooper minimum on the one hand, and the 4*s* and 5*s* on the other, is that the highest Z at which the minimum appears in the continuum is the noble gas for the 6*s* ($Z=86$). Since, however, the 6*s* zero is *so* close to threshold for $Z=86$, we would hesitate to make too much of this point since, as discussed above, the position of the zero results from a balance of several effects and more exact calculations could well show that balance altered and obviate the difference.

The trajectory of the $7s\rightarrow\epsilon p$ Cooper minimum looks, at first glance, rather different from the others. This is due primarily to the fact that we come to the end of the known elements while still only on the part of the trajectory that is moving away from threshold. In addition, a finer mesh of Z's is used here than for the other *ns*'s which shows some of the nonmonotonic behavior of the trajectory. Basically, the wiggles are due to nonmonotonic filling of the 5*f* and 6*d* subshells and the completion between them. Calculations assuming only 5*f* electrons showed no structure in the trajectory. It appears that actinide elements having only 5*f* electrons have Cooper minima further away from threshold than those with 5*f* and 6*d* electrons. The 5*f* screens the 7*s* slightly better than the 6*d* does and this screening, as discussed in connection with the 4*s*, is what leads to the movement of the zero away from threshold with increasing Z in the first place. Had such a fine mesh been employed in studying the 6*s* over the range of the lanthanides, it is expected that similar structure would have emerged.

B. *np* subshells

The calculated trajectory of each of the $np\rightarrow\epsilon d$ Cooper minimum is shown in Fig. 2. As in the *ns* cases, the

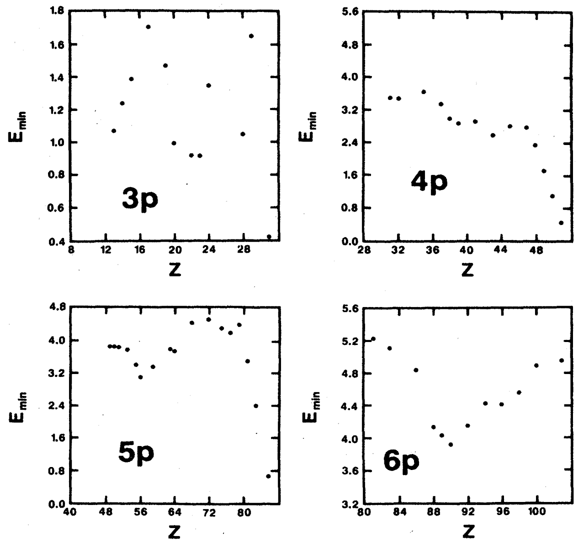

FIG. 2. Location of the zero (Cooper minimum) in the dipole matrix elements for $np\rightarrow\epsilon d$ ionizing transitions in energy in rydbergs above threshold (E_{min}) vs atomic number Z for ground-state atoms.

Cooper minimum are in the continuum just as soon as the *np* orbital becomes bound in the ground state of the atom. However, the trajectories of the *np*'s are rather different from the *ns*'s and, for that matter, from each other. In addition, the *np* zeros are much further from threshold than the *ns*; as high as 5 Ry above threshold for the *np*'s compared with a maximum of about 0.5 Ry for the *ns*'s.

To understand the trajectories of the $np \rightarrow \epsilon d$ Cooper minima, note first that while the *ns* and *np* quantum defects (ϵs and ϵp phase shifts at threshold) increase rather uniformly and monotonically with *Z*, the *d*-wave threshold phase shifts do not; instead they are rather constant with *Z*, jumping by π just above each of the noble gases[17] starting with $Z = 18$. As a result, the relative phase difference for the $ns \rightarrow \epsilon p$ transitions remain in a relatively narrow range between 0.5π and 0.7π, while for the $np \rightarrow \epsilon d$ transitions the phase difference varies between less than π and more than 2π. From this relationship of the relative phase differences to the position of the zeros, discussed in Sec. A, it is clear that the largeness of these differences in the *np* case, as compared to the *ns*, explains why the Cooper minima are much further from threshold for *np*. In addition, the much larger range of variation of the phase differences in the *np* case implies rather different and more complicated trajectories than in the *ns* case.

For the $3p \rightarrow \epsilon d$ Cooper minimum, at $Z = 13$ (where the 3*p* becomes bound in the ground state), the relative phase difference is 1.3π which translates to a zero more than 1 Ry above threshold. The zero moves out further, with increasing *Z*, because the 3*p* quantum defect is increasing, while the *d*-wave threshold phase shift remains relatively constant at zero, so that the relative phase difference increases. by $Z = 19$, the *d*-phase shift jumps to about $\pi/2$ at threshold, with another $\pi/2$ jump at $Z = 20$. These jumps are much greater than the increase in the 3*p* quantum defect here, so the relative phase difference drops and, thus, the zero moves closer to threshold. Increasing *Z* further, the *d*-wave phase remains at about π so that the relative phase difference is increasing, but the "inner effect," discussed in Sec. A, is becoming more important and the balance between the two effects keeps the zero at about the same place until $Z = 31$ where the inner effect dominates and the zero moves much closer to threshold, disappearing below threshold by $Z = 32$.

Two anomalies occur, at $Z = 24$ and 29, where the zero moves markedly further out. These points are caused by anomalous filling of the shells; in each case there are *two* more 3*d* electrons and one less 4*s* than the previous *Z*. This increases the screening and makes the 3*p* wave function less compact, thereby giving more weight to the relative phase difference which is tending to move the zero away from threshold.

The $4p \rightarrow \epsilon d$ Cooper minimum is seen from Fig. 2 to behave rather differently from the 3*p*. At the onset, $Z = 31$, the zero is about 3.5 Ry above threshold, much higher than the 3*p* because the relative phase difference is 1.4π here, greater than in the 3*p* case, and the outer shell is more diffuse than for the 3*p* case since the atom is larger, thus emphasizing the outer region in the dipole matrix element. With increasing *Z*, the zero moves out only a small amount because the 4*p* quantum defect is increasing only about as much as the *d*-wave threshold phase shift, keeping the relative phase difference almost constant. Above $Z = 35$, the jumps in the *d*-phase shift at $Z = 37$ and $Z = 38$ cause the rapid movement of the zero towards threshold, just as in the 3*p* case. Beyond $Z = 38$, the 4*p* trajectory behaves in much the same way as the 3*p* above $Z = 20$, and for precisely the same reasons.

The $5p \rightarrow \epsilon d$ zero behaves, at its onset, just like the 4*p* except that the zero is further out because the atoms here, $Z = 49$ and above, are still larger than the corresponding 4*p* cases. Above $Z = 56$, however, where the minimum has moved closer to threshold owing to the jumps in the *d*-wave phase shift, the zero again moves away from threshold, with increasing *Z*. This differs from the previous *np* zeros discussed and is due to the filling of the 4*f* shell, which fills inside the 5*p*. Since *p*-waves are only slightly penetrating, this means that their quantum defect will increase slightly in passing through the lanthanides; but since *d* waves are almost completely nonpenetrating, the *d*-phase shift remains constant. The net effect is an increase in the relative phase difference which, when combined with the screening effect of the 4*f*'s keeping the 5*p* diffuse, moves the minimum away from threshold, as seen. Above the rare earths, where the 5*d* shell is filling, the 5*p* trajectory behaves just like the lower *np*'s. Note that with the rare earths there are anomalies due to anomalous filling of the 4*f* subshell, just as was found for the filling of the *nd* shells.

The Cooper minimum in the $6p \rightarrow \epsilon d$ channel looks rather different from the other *np*'s but this is mainly because it has not yet moved into the discrete at the highest *Z* considered. When that point is taken into account, it behaves almost exactly the same as the 5*p* and for the same reasons. The 6*p* trajectory ends while still in the actinides. The atoms here are still larger than in the 5*p* case and therefore, still more diffuse, moving the zero still further from threshold, however.

C. *nd* subshells

The behavior of the $nd \rightarrow \epsilon f$ Cooper minima are shown in Fig. 3. It is seen that a zero appears as soon as the subshell becomes bound in the ground state, as with the *ns*'s and *np*'s. It is also seen that with increasing *n*, the Cooper minima move further from threshold, just as the *np*'s did, owing to the increasing diffuseness of the atoms. Furthermore, the *nd* zeros are much further from threshold than the *np*'s, as far out as 14 Ry.

The $4d \rightarrow \epsilon f$ zero starts off at $Z = 39$ at just under 5 Ry above threshold owing to a huge relative phase difference of more than 1.8π. In fact this is a large enough difference to support two zeros in the dipole matrix element, but the inner effect is strong enough to preclude this. In an excited *ns* state of $Z = 39$, where the inner effect is proportionally less important then, it is possible to have two zeros, in fact this has been reported for the excited 5*d* state.[19]

With increasing *Z*, the 4*d* quantum defect increases while the *f*-wave threshold phase shift remains zero, caus-

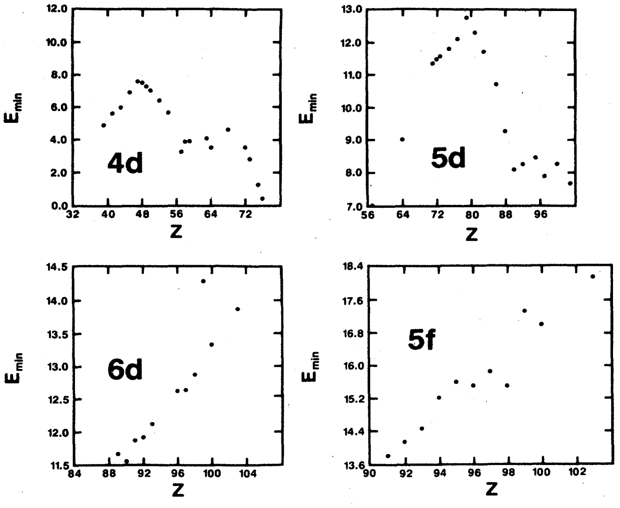

FIG. 3. Location of the zero (Cooper minimum) in the dipole matrix element for $nd \to \epsilon f$ and $5f \to \epsilon g$ ionizing transitions in energy in rydbergs above threshold ($E_{\min}$) vs atomic number for ground-state atoms.

ing the relative phase difference to increase and the zero to move further above threshold, reaching a maximum at $Z=47$. Above this, the zero starts moving back down due to the increasing importance of the inner effect and plummets between $Z=55$ and $Z=56$ where the f-wave phase shift jumps from 0 to π at threshold. At still higher Z, the rare-earth region, where $4f$'s are being bound, the zero moves out slightly. This is because the $4d$ quantum defect is increasing while the f-phase shifts are remaining constant with increasing Z, combined with the screening effect of the $4f$ filling which keeps the $4d$ wave function quite diffuse. The zero does not move out uniformally or monotonically in the rare-earth region owing to the anomalous filling of the $4f$ subshell. Above the rare earths, the inner effect starts to dominate and the zero moves into the discrete above $Z=76$.

The $5d \to \epsilon p$ has a trajectory very similar to the $4d$ with two exceptions. The $5d$ is part of the ground atomic state for $Z=57$, 58, 59, and 64 and for no others below $Z=71$. Further, at the highest Z considered, $Z=103$, the $5d$ zero has not yet moved below threshold. Aside from these, we see the same rise, drop, and subsequent meandering of the $5d$ zero for exactly the same reasons for that behavior in the $4d$ case.

For the $6d \to \epsilon f$ Cooper minima, only the rising portion of the trajectory from the onset is seen before the known Periodic Table ends. This rise is just like the $4d$ and $5d$ cases and occurs for precisely the same reasons. The various anomalies in the $6d$ trajectory, which are seen clearly owing to the fine mesh of Z's considered, are due to the nonmonotonic filling of shells with increasing Z, as discussed previously.

D. $5f$ subshell

The trajectory of the $5f \to \epsilon g$ zero is also shown in Fig. 3 where it is seen that the zero is at 13.8 Ry at the onset, $Z=91$, resulting from a relative phase difference of 2π. Since the Periodic Table runs out, only a small portion of this trajectory is seen. It rises, with increasing Z, since the $5f$ quantum defect is increasing while the ϵg phase shift remains zero at threshold. The anomalous behavior is due to anomalous filling of shells, as in other cases.

IV. FINAL REMARKS

The location of Cooper minima in ground states of atoms has been calculated over the entire periodic system and their behavior, as a function of Z, has been explained primarily in terms of the quantum defects of the discrete states and the phase shifts of the continuum states. It was found that the trajectories of these zeros were generally a very complex function of Z for various reasons as discussed.

It was also found that these Cooper minima are extremely pervasive. At least one subshell is found to have one for all $Z > 10$ and some high-Z elements have as many as *five*, e.g., $Z=92$ has Cooper minima in the $7s$, $6p$, $5d$, $6d$, and $5f$ channels. Clearly, then, this phenomenon is not an isolated curiosity.

These results have been obtained using simple nonrelativistic Hartree-Slater wave functions. Based upon past experience, however, it is expected that using more sophisticated wave functions including exchange and/or correlation might make some quantitative changes, although they are unlikely to alter the overall systematics.[3,20]

Relativistic effects are another matter. This is because a single nonrelativistic Cooper minimum becomes three minima relativistically (two for an initial s state). For example, the $6p \to \epsilon d$ nonrelativistic channel becomes $6p_{1/2} \to \epsilon d_{3/2}$, $6p_{3/2} \to \epsilon d_{3/2}$, and $6p_{3/2} \to \epsilon d_{5/2}$; the $p_{1/2} \to \epsilon d_{5/2}$ transition is forbidden in the electric dipole approximation. Comparing these relativistic zeros with the nonrelativistic, using the same atomic formulation otherwise, provides an extremely sensitive probe of the relativistic interactions. This has been done for $6p$ and $5d$ subshells[21,22] and the results showed huge splitting between the zeros of the relativistic subchannels arising from a single nonrelativistic channel; this emphasizes the sensitivity of the position of the Cooper minima to small interactions such as the spin-orbit effect.

Rich as the phenomenology is for Cooper-minimum ground-state atoms, it is far richer for excited states where zeros exist in $l \to l-1$ transitions[19,23] and multiple zeros (as many as three) in individual channels are found.[19,23,24] It would be interesting to connect the phenomenology of ground- and excited-state atoms by calculating the continuation of the trajectories to Z's below where the orbital is bound in the ground state, e.g., look at the $4p$ transitions for $Z < 31$. We are embarked on this project at present.

ACKNOWLEDGMENTS

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