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Who Named the Quantum Defect?

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Abstract

The notion of the quantum defect is important in atomic and molecular spectroscopy and also in unifying spectroscopy with collision theory. In the latter context, the quantum defect may be viewed as an ancestor of the phase shift. However, the origin of the term "quantum defect" does not seem to be explained in standard textbooks. It occurred in a 1921 paper by Schrödinger, preceding quantum mechanics, yet giving the correct meaning as an index of the short-range interactions with the core of an atom. We present the early history of the quantum-defect idea, and sketch its recent developments.

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Prologue

A character in Molière's *Le Bourgeois Gentilhomme*¹ expresses astonishment that "for more than forty years, I have been speaking prose without knowing it." In sciences, too, concepts and ideas evolve, even coming into common practice before being formally named. Subsequently, the name becomes so familiar to later generations that its origins are forgotten, it being almost unthinkable to dissociate the name from the concept, or even to imagine that they were not born full-blown together. This is especially true of felicitous names that seem well-tailored to the concept that they label.

The Rydberg formula and the quantum defect

One such name in physics is the "quantum defect." Every physicist is familiar today with its role as parametrizing a Rydberg series of atomic energy levels E_n below an ionization limit (taken as the zero of the energy scale),

$$E_n = -Ry / (n - \delta_\ell)^2, \quad (1)$$

where $Ry = 109,737 \text{ cm}^{-1}$ is the Rydberg constant, and δ_ℓ is the quantum defect, depending on the angular momentum ℓ of an excited electron in a simple "hydrogenic" spectrum such as that of an alkali. The quantum defect arises because of the ionic core of the other electrons in the atom. Depending on its ℓ value, the excited electron samples this distribution to a varying degree; only electrons with the lowest ℓ values penetrate appreciably into the core, making δ_ℓ negligibly small for $\ell > 3$ even in heavy atoms.

The above formula itself, although not the names, dates back, of course, to Rydberg's 1890 paper.² For a historical account of the Rydberg formula, see El'yshevich *et al.*³ The slightly earlier Balmer⁴ formula without the δ_ℓ , applicable to the hydrogen atom, which has a bare point-nucleus, became the basis of Bohr's seminal paper that launched the quantum theory of atomic structure, n then getting the name of a (principal) quantum number. Today, atomic and molecular physicists use an elaborate "quantum-defect theory" applicable to Coulombic systems including ones more complicated than alkali atoms and containing open shells in the core. In such

"multichannel" situations, with energy dependencies more complicated than the simple expression (1), "multichannel quantum-defect theory" (MQDT) has been extensively developed and is now widely applied for the analysis of complex spectra.⁵⁻⁷

Effects of the electron penetration into the ion core

A key physical point initiating these developments underlies Eq. (1): when an electron moves in a combined long-range Coulomb field (due to the net positive charge of the core) and short-range multiparticle interactions (confined to the region of the core), the two kinds of interactions have different energy scales, and therefore different sensitivities to the energy. The short-range region involves strong potentials, and therefore cannot influence drastic changes in small energy steps. Thus, parameters such as the quantum defect which have their seat in this region vary smoothly with energy, and are, to a first approximation, constant from one energy level to the next in the upper reaches of a Rydberg spectrum. Indeed, they continue across the ionization limit into the continuum to connect with similar smoothly varying parameters in the scattering domain. One of the key and early such connections is that the quantum defect δ_ℓ is the zero-energy phase shift η_ℓ (in units of π), additional to the Coulomb phase shift, for the scattering of an electron of that same angular momentum ℓ from the core,⁵⁻⁷ viz.,

$$\delta_\ell = \eta_\ell / \pi . \quad (2)$$

Sensitive energy dependencies, on the other hand, can only arise from regions of weak potential, where the kinetic energy of the electron becomes significant. That is, such sensitivities in the vicinity of the ionization limit, such as the dependence on n in Eq. (1), can only arise from the long-range field.

The modern literature^{6,7} uses the symbol μ_ℓ for the quantum defect, and the symbol δ_ℓ for the phase shift; in this usage, Eq. (2) is written as $\mu_\ell = \delta_\ell / \pi$. Throughout the present article, however, we adhere to the old tradition of using δ_ℓ for the quantum defect.

In view of the physical meaning sketched above and from the vantage point of today, a term such as "parameter of core penetration" or "index for short-range interactions" would be more descriptive than "quantum defect".

The first appearance of the term "quantum defect"

So far as we have been able to trace its origin, Schrödinger was the first in 1921 to use this term in its German form "Quantendefekt." This paper⁸ is remarkable for standing alone among Schrödinger's papers in the period (on statistical mechanics) as a contribution to the quantum theory of atomic structure. (See Moore's commentary⁹ for details.) Schrödinger did not return to quantum matters until four years later in the now famous papers establishing wave mechanics. But the 1921 paper presents a fairly sophisticated treatment of penetrating orbits and an estimate of the quantum defect for s-waves in sodium, along with the use of Δ as the symbol for it [Eq. (16)], followed by the words "Nennt man Δ den 'Quantendefekt'," which was the formal christening of the term.

In the papers of Bohr, Sommerfeld, and their co-workers before and around 1921, the Rydberg formula appears with the symbol α used consistently for the quantum defect but referred to only as "eine Konstante." Interestingly, in unpublished notes and in publications in the early 1920s, Bohr uses the symbol n^* for the related concept of "effektive Quantenzahl" (effective quantum number) $n^* = n - \delta_l$. Also, a "screening constant, "subdivided into "inner" and "outer", occurs in Bohr and Coster.¹⁰ Prior to this, in unpublished notes¹¹ for the 1921 Solvay conference and in seven lectures¹² at Göttingen in 1922, Bohr models the perturbation on outer electrons due to the inner-core electrons as an additional dipole potential superposed on the Coulomb field of the positive charge and estimates α for s, p, and d electrons in alkalis; see especially pp. 141-144, p. 384, and pp. 395-396 of *Collected Works, Vol. 4*. These appear again in the Coster and Bohr paper¹⁰ and in a 1923 paper¹³, where a footnote refers to Schrödinger's 1921 work in the following words, "At the same time as the general features...were developed by the author (of *Nature*, March 24, 1921), Schrödinger independently emphasized that in the s-terms of the alkalis...the electron during its revolution penetrates into...." Bohr goes on to refer to papers by Fues¹⁴ and by van Urk¹⁵, which followed Schrödinger's, but does not adopt the term "quantum

defect". Schrödinger sent Bohr a copy of his 1921 paper, as a " 'note' which may be of some little interest to you", in a letter dated 7 June 1921, which was acknowledged on 15 June.¹⁶

Altogether we feel that Schrödinger and Bohr arrived independently and simultaneously in the years 1920 and 1921 at the concept of core penetration modifying the Bohr formula to the Rydberg formula. A slightly different flavor is given by Pais,¹⁷ who states that Bohr talked of core penetration leading to a stronger electron binding in a December 1920 Copenhagen lecture and that "the idea appeared first in print in a paper by Schrödinger (submitted in January 1921)". In any case, the name "quantum defect" was certainly Schrödinger's, Bohr failing to adopt it even as late as 1925. Two authors who did adopt immediately Schrödinger's term were Fues¹⁴ and Wentzel,¹⁸ who refer to Schrödinger's 1921 paper, using the term "quantum defect" freely. van Urk,¹⁵ on the other hand, also begins by referring to Schrödinger but presents his estimate of 0.74 ± 0.01 for the s-quantum defect in Na without using the name.

It is also interesting to follow the evolution in Sommerfeld's publications regarding the term "quantum defect." Early papers¹⁹ developed Rydberg-type formulae, with a non-integer contribution to the denominator in Eq. (1) and correctly attributed it to potential terms that fall off as higher powers of r^{-1} , without naming it. The 1919 and 1921 editions of his classic text *Atombau und Spektrallinien* also fail to mention the term. The 1922 third edition²⁰ (submitted January 1922 in Munich) includes a footnote on p. 404 to Schrödinger's 1921 paper, present on p. 329 in the Methuen English translation²¹ in 1923, citing the estimate of 0.74 without giving it a name. The word "Quantendefekt" and symbol Δ occur from the 1924 editions on, but, curiously, when they are introduced, as on p. 389 of the 1934 English translation,²² there occur the words "The expression 'quantum defect' for Δ is obviously justified", but there is no reference to Schrödinger at that point. Only in the next section stands the reference to the 1921 paper with the words "Schrödinger was the first to show with Na as an example that the s-orbits penetrate into the atomic core...", giving the value $\Delta = 0.74$, followed by references to the Fues, Wentzel, and van Urk papers. In the later editions of the Sommerfeld book, the term quantum defect is well established, even acquiring several index entries.

We feel fairly confident, therefore, that we have traced the original usage of the term "quantum defect" to the 1921 paper of Schrödinger's, predating the term "quantum mechanics"

whose origin is instead clear in a 1924 paper²³ of Born's with the title *Über Quantenmechanik*," which puts the words within quotation marks later in the text. It is, therefore, curious that Schrödinger returns to the subject in a 1925 paper²⁴ with the following on p. 63, "It is this that Sommerfeld in the above referenced place in his book (whose stimulus I thank for my investigations of these questions) chooses as an example, and illustrates with a figure, for exhibiting the quantum defects of individual terms as functions of the quantum number of the orbits." He also refers to the papers of Fues,¹⁴ Wentzel,¹⁸ and van Urk,¹⁵ rather than to his own earlier paper! He also makes mention in a note added in proof of "negativen Quantendefekts" in a Table (for Si) of Fowler's Bakerian lecture.²⁵ Fowler himself in this 1925 paper uses only the term "effective quantum number" but not quantum defect!

Incidentally, one finds the term "Rydberg correction", meaning the same as quantum defect, in the early literature, e.g., in Sommerfeld¹⁹, and also after the advent of wave mechanics. In our views, the term is uninformative and uncommendable; fortunately, it is practically obsolete now.

Epilogue: Current Status of the Quantum Defect Theory

The quantum defect, or equivalently, the phase shift in Eq. (2), is a key parameter of atomic structure. Schrödinger's seminal estimate of $\delta_0 = -0.74$ is a precursor of extensive tabulations of δ_ℓ for atoms and ions across the periodic table. His result, expressed as $\delta_0 = 1.26$ in a modern convention, closely agrees with recent values²⁶: $\delta_0 = 1.359$ from a Hartree-Slater calculation, and $\delta_0 = 1.35$ from analysis of spectroscopic data.

In evaluating δ_0 by fitting spectroscopic data to Eq. (1), its integer part depends on the convention adopted for n . It is now usual to employ the hydrogenic quantum number, regarding, for instance, the first excited s-state of Na as 4s, with $n = 4$, $n = 2.64$ and thereby $\delta_0 = 1.36$. A different choice, such as $n = 2$ for this state (regarding the ground 3s state as $n = 1$), would give instead $\delta_0 = -0.64$. This ambiguity in the integer part of the quantum defect is connected, of course, to the ambiguity of any phase as in Eq. (2) to integer multiples of π . Definitions of asymptotic energies or phase shifts at infinity are further complicated by the long-range Coulomb field with its finite zero-energy phase shift. All these complications and ambiguities are skirted by

the modern point of view to be sketched further in this section, wherein the short-range origins of δ_ℓ are emphasized by integrating outwards from the origin over the finite radial distance of short-range interactions.

Soon after its advent, the Schrödinger equation began to be applied to the evaluation of the quantum defect.²⁷⁻³³ Spectroscopic data or *ab initio* numerical calculations give energy levels and wave-function properties which are best summarized in terms of δ_ℓ . Examples of early systematic studies are seen in the papers by Fermi³² and Fano³³. Figure 1 presents a recent example showing the variation of δ_ℓ as a function of the nuclear charge Z for neutral atoms³⁴. This plot carries a wealth of information basic to atomic structure and also to molecular structure. Not only are gross features evident such as the periodicity of elements that reflects the potential for the electron motion in the valence layers of the atom, but also more subtle variations, sometimes monotonic and sometimes otherwise, between neighboring atoms can be seen. The trends along the transition elements, i.e., lanthanides and actinides, including a jump near the complete or half filling of the d and f shells, reflect the influence of sub-surface layers of the atom and of the prevailing core potential over those ranges of the radial distance. Furthermore, the derivative of δ_ℓ with respect to energy, which characterizes the weak residual energy dependence, reflects even finer details of the electron motion in the atom.³⁴ Physically, the derivative represents³⁵⁻³⁷ the time delay of the electron motion due to short-range interactions, and is important in the interpretation of resonances. Plots of δ_ℓ for atomic ions, as functions of both the nuclear charge Z and the total number N of electrons are also available.²⁶

The quantum defect is the principal among several parameters in the modern quantum defect theory⁶ for atoms. The basic idea, i.e., the characterization of the long-range Coulomb interaction in terms of the n^{-2} dependence of Eq. (1) and of the short-range interaction in terms of δ_ℓ , is subsumed under the following themes, which form the basis of a powerful and general analysis,⁷

1. The configuration space is divided into long and short ranges with respect to the motion of one electron of interest from the rest of the atomic system. The two regions have very different sensitivities to the energy. At small r , the radial co-ordinate of the electron, a strong potential prevails; as a result, modest or even substantial changes in the electron's asymptotic kinetic energy

pale into insignificance. Therefore, different bound states, especially at high n , and continuum states, of modest energy at least, are described on a common footing. At large r , the asymptotic behavior of the electron wave function is chiefly determined by its kinetic energy, viz., its total energy minus the potential energy, which is small at large r .

2. In general, different basis sets of functions are appropriate for representing electron wave functions efficiently. At small r , an energy-independent basis set, defined through an energy-independent boundary condition at the origin, proves appropriate, applying equally to bound and continuum states. Distinctions between these states arise at large r , being established by the boundary condition for $r \rightarrow \infty$.

3. The connection between the two regions involves both geometrical and dynamical elements. The geometrical elements, dealing with all the angular momenta, orbital and spin, also differ for the two regions. For instance, the LS coupling scheme is generally appropriate at small r , whereas the jj coupling scheme is more suitable at large r . Passage between these alternative geometrical elements is accomplished by unitary transformations, often obtainable analytically and called frame transformations.⁷

4. The dynamical elements in this connection pertain to radial wave functions. The motion at large r in the Coulomb potential is described by well-known analytical solutions, common to all atoms. The motion at small r , specific to each atom, depends instead on the nature of the core. Dependable calculations for small r will require numerical work. However, owing to points 1 and 2, a coarse energy mesh will often suffice, making for economy in numerical work. The finiteness of the core volume also makes for economy.

5. The wave functions at large r and small r are connected at a boundary, as in the Wigner R-matrix theory.³⁸⁻⁴⁰ For a single-channel problem, the connection involves the logarithmic derivative of the radial function leading to the determination of the phase shift or the quantum defect. (A channel here refers to a whole set of states differing only in energy but sharing all discrete quantum numbers of angular momenta, orbital and spin.) For multichannel problems, e.g., in an atom with open-shell structure, the phase shift η_ℓ is replaced by a matrix generalization; technically, $\exp(i\eta_\ell)$ is replaced by the Jost matrix.⁴¹ The elements of the Jost matrix are therefore the key parameters for describing bound and continuum states of the atom.

6. The powerful and attractive approach sketched above lies in the judicious separation of the short-range and long-range dynamics, and can be generalized to situations in which the long-range interaction is other than Coulombic, e.g., the dipole interaction between an electron or an ion with a polar molecule, the polarization interaction between an electron or an ion with an atom, or the van der Waals interaction between atoms or molecules. The short-range dynamics involve many electrons and nuclei (or cores) and are necessarily complicated, but need to be treated in a finite volume only. Thus, the generalized quantum defect theory has been developed⁴²⁻⁴⁵ and applied successfully to many problems in atomic and molecular physics.⁷

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Figure Captions

Figure 1. The quantum defect, or the phase shift at zero energy, for neutral atoms as a function of the atomic number Z for $\ell = 0, 1, 2,$ and 3 . This figure is reproduced with permission from Fano *et al.*³⁴

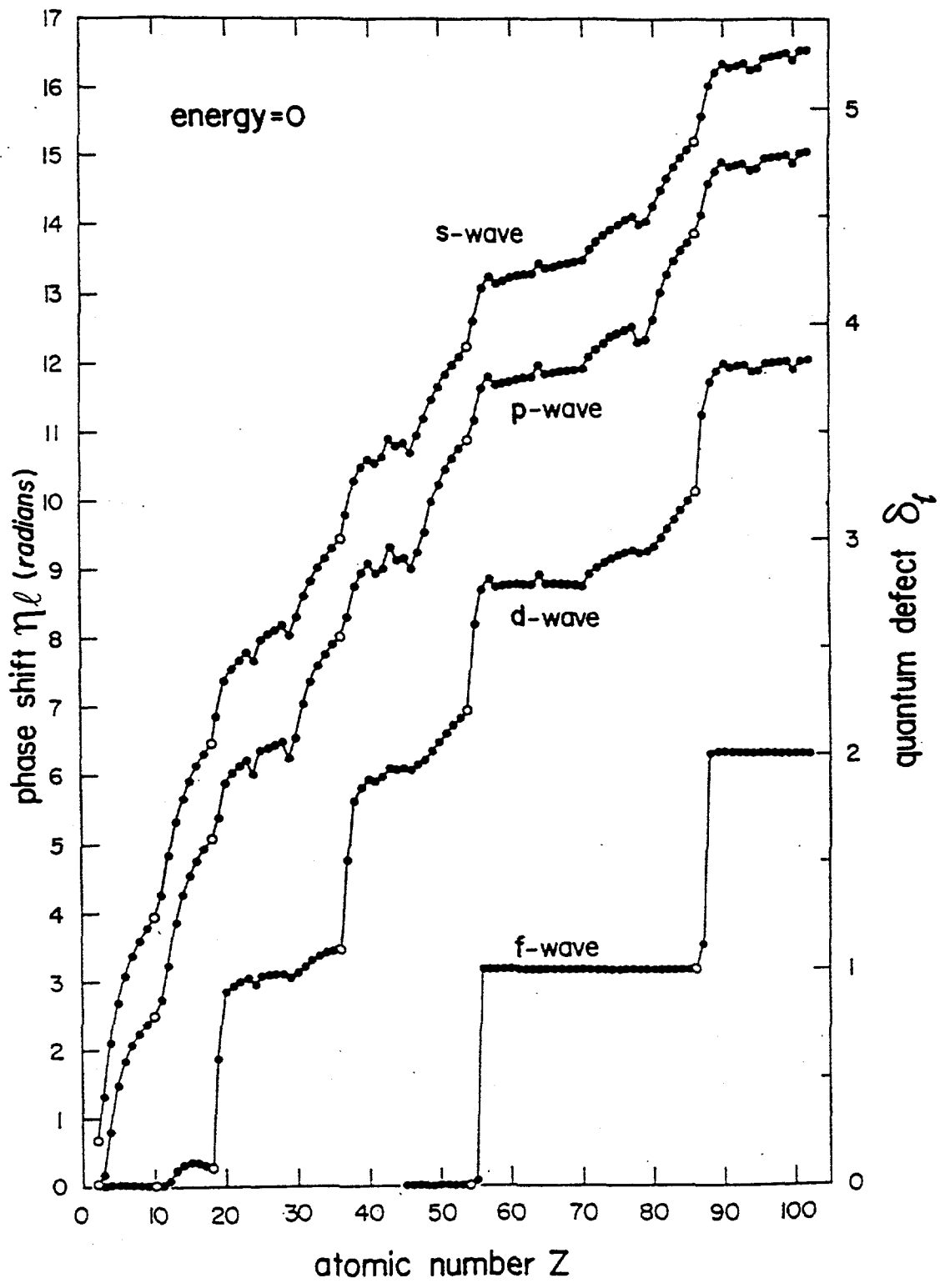


Fig. 1