Dominant Correlation Effects in Two-Electron Atoms

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Abstract

Two-electron atoms have been investigated near threshold of double escape within the framework of hyperspherical coordinates. A particularly useful set of hyperspherical angles has been used. It is well known for many years that the hyperradial motion is nearly separable from the hyperspherical angular motion. Therefore, the Born-Oppenheimer separation method should be useful. However, the success of that method in molecular physics is based on the small mass ratio, electron mass to nuclear mass. In the atomic application such a small parameter does not exist. Nevertheless the method works surprisingly well in the lower part of the spectrum. For increasing excitation energy the method becomes shaky. Near ionization threshold, it breaks even down. The author will present elsewhere an improved Born-Oppenheimer method. First pilot developments and comparison with the experimental situation are presented already here. Inclusion of a momentum-momentum radial coupling delivers an improved basis. We show that our extended Born-Oppenheimer approach leads to a deformation of the whole potential energy surface during the collision. In consequence of this deformation we outline a quantum derivation of the Wannier threshold cross section law, and we show that (e, 2e) angular distribution data are strongly influenced by that surface deformation. Finally, we present a mechanism for electron pair formation and decay leading to a supercurrent independent of the temperature. Our framework can be extended to more than two electrons, say 3 or 4. We conclude that our improved Born-Oppenheimer method [1] is expected not only to deliver better numerical data, but it is expected to describe also the Wannier phenomenon. The idea of the new theory together with first qualitative results is presented in this paper.

Keywords

Born-Oppenheimer Approximation, Wannier Theory, Electronic Correlation, 3-Body Systems, Hyperspherical Coordinates

1. Introduction

The theory of multi-electron atomic spectra is far from being well understood. Models by Bohr [2], and Hartree-Fock [3] deal with single electron configurations, and neglect correlation. These models are based on the simple and well understood hydrogen atom.

There is, however, strong evidence in the case of many-electron atoms that correlation becomes more and more important at increasing excitation energy. Near thresholds of multiple escape correlation become even dominant. A typical example is the threshold ionization of a hydrogen-like atom by electrons. The two-electron atom (or ion) shows already all difficulties of a many-electron atom. Therefore He-like atoms constitute a good candidate for theoretical work. Many years ago Wannier [4] has calculated using classical mechanics the total threshold cross-section σ for the ionization process $H + e \to H^+ + 2e$. He came to the surprising result

$$\sigma \propto \left(E - E_0 \right)^{1.127\cdots} \tag{1}$$

There is strong experimental evidence for the fractional exponent in (1), see for instance [5] and references therein. The present paper outlines a quantum derivation of (1).

Since the early days of quantum theory highly accurate variational calculations of the *He* ground state have been performed. The ground state is, however, not a suitable candidate to study correlation because correlation becomes important only at double excitation.

Theoretical progress has been achieved using hyperspherical coordinates. Macek [6] has used a channel expansion method analogous to the Born-Oppenheimer approximation [7]. Klar *et al.* [8] have presented similar calculations employing more suitable angular coordinates. But both works have one common disadvantage. Their results are very good in the lower part of the spectrum. They become, however, shaky at increasing excitation energy, and their method breaks down near the threshold of double escape.

It is the aim of the present paper to work out the reason for that shortcoming, see also [1]. We show that the familiar Born-Oppenheimer approximation contains one fundamental mistake. That method employs a countable set of collision channels represented by static potentials. That description breaks down near threshold of double ionization where an infinity of channels converges to it. The present paper goes therefore beyond the usual Born Oppenheimer method, and takes a momentum-momentum coupling into account too. The latter was disregarded by Born and Oppenheimer [7].

The paper is organized as follows. §2 treats geometrical aspects, §3 develops an improved Born-Oppenheimer treatment for simplicity restricted to *S* states, §4 analyzes novel motions of the whole complex and compares results with experimental work, §5 presents a novel supercurrent based on the extended Born-Oppenheimer model, and §6 outlines possible generalizations to more

than two electrons. §7 summarizes our conclusions. An Appendix lists and explains all used symbols.

2. Geometrical Aspects

Subject of the following investigation is the S-state two-electron atom, like He. That constitutes the simplest candidate of atoms suitable for correlation studies. For three-body Coulomb systems (nucleus + two electrons) are analytical solutions neither of the classical equations of motion nor of the wave equation available. A key ingredient for the successful treatment of the equation of motion is the choice of appropriate coordinates. Basically there are two possibilities. First, single electron coordinates, or second collective coordinates. Single electron coordinates are useless in correlation studies. An example for collective coordinates is hyperspherical coordinates in the case of few-electron atoms. See Wannier [4], Macek [6], Klar [8] and others, in the case of two electrons.

The key idea of hypersphericals is to replace the single electron position vectors r_1 and r_2 by one six-dimensional vector given by

$$\boldsymbol{R} = \begin{pmatrix} \boldsymbol{r}_1 \\ \boldsymbol{r}_2 \end{pmatrix} \tag{2}$$

with R

$$R = \sqrt{r_1^2 + r_2^2} \tag{3}$$

where r_1 and r_2 are the electron positions. The hyper-radius R describes the size of the whole atom. In addition to R we need five angles on the hyperradius S_5 to fix the electron positions. Macek [6] used the familiar polar angles $\{\vartheta_i \varphi_i\}$ $i = 1, 2$ plus the pseudo angle given by $\alpha = \tan^{-1}\dfrac{r_1}{r_2}$. The disadvantage of these angles is that they don't provide a clear separation between overall rotations and intrinsic motion. A more convenient choice to map the five sphere S_5 uses Euler angles (α, β, γ) to describe overall rotations plus two body-fixed angles ψ, φ for intrinsic motions, see for instance [8] and references therein. Both [6] [8] employed a Born-Oppenheimer [7] approximation to solve the wave equation where the hyperadius is the adiabatic coordinate.

A look to the potential surface of the electron pair in the nuclear field shows immediately why these Dragt [9] angles $(\alpha, \beta, \gamma, \psi, \varphi)$ are superior. The electrostatic potential of the two-electron atom reads

$$V = -Z\left(\frac{1}{r_1} + \frac{1}{r_2}\right) + \frac{1}{r_{12}} \tag{4}$$

where r_{12} is the electron-electron separation, and Z is the nuclear charge. Because of overall rotational invariance the potential is independent of α, β, γ and reads in terms of the Dragt coordinates simply

$$V(R, \psi, \varphi) = \frac{C(\psi, \varphi)}{R} \tag{5}$$

with the charge function given by

$$C(\psi,\varphi) = -Z\left[\frac{1}{\sqrt{1-\cos 2\psi \sin\varphi}} + \frac{1}{\sqrt{1+\cos 2\psi \sin\varphi}}\right] + \frac{1}{\sqrt{1-\cos 2\psi \cos\varphi}} \quad (6)$$

The physical meaning of the angles ψ, φ is as follows. ψ describes the ratio of two moments of inertia whose principal axes lie in the particle plane, *i.e.*

$$\psi = \frac{1}{2}\tan^{-1}\sqrt{\frac{\Theta_{xx}}{\Theta_{yy}}} \quad (7)$$

For instance the value $\psi = 0$ describes all collinear configurations. The potential is an even function of ψ. It is therefore sufficient to consider only one hemisphere; in this paper we use $0 \le \psi \le \frac{\pi}{4}$ to map the electron positions onto the northern hemisphere including the equator.

The angle φ was already introduced long ago by Sommerfeld [10] as

$$\varphi = \tan^{-1}\frac{r_1^2 - r_2^2}{2r_1 r_2 \cos\Theta} \quad (8)$$

which is the hyperspherical azimuth angle ($0 \le \varphi < 2\pi$). The electron position vectors in the particle plane are then given by [8]

$$\boldsymbol{r}_1 = R\begin{pmatrix} \cos\psi \cos\frac{1}{2}\left(\varphi+\frac{3\pi}{2}\right) \\ \sin\psi \sin\frac{1}{2}\left(\varphi+\frac{3\pi}{2}\right) \end{pmatrix} \quad (9)$$

$$\boldsymbol{r}_2 = R\begin{pmatrix} \cos\psi \cos\frac{1}{2}\left(\varphi-\frac{3\pi}{2}\right) \\ \sin\psi \sin\frac{1}{2}\left(\varphi-\frac{3\pi}{2}\right) \end{pmatrix} \quad (10)$$

The two-body coincidences are located at the angles $\psi = 0$, $\varphi = \frac{\pi}{2}$ ($r_1 = 0$); $\psi = 0$, $\varphi = \frac{3\pi}{2}$ ($r_2 = 0$); $\psi = 0$, $\varphi = 0$ ($r_{12} = 0$).

Wannier [4] has stressed the importance of a saddle point at $\psi = 0, \varphi = \pi$. We will see below that this point plays a key role for high double excitation.

3. Zero-Energy Wave Function

This section presents an exact solution of the three-body system valid in the low energy Coulomb zone near the saddle point. We proceed as follows:

At low energy we expect the three particles in consequence of the electron-electron repulsion in a collinear configuration (electron-nucleus-electron). The potential surface shows there two attractive Coulomb zones separated by a barrier. The top of this barrier corresponds to equal electron-nucleus distances, *i.e.* $r_1 = r_2$, and its height coincides with the threshold energy for double escape.

The present paper treats for simplicity only S states. The Hamiltonian for S states of the whole atom reads then in terms of Dragt coordinates [8]

$$H = -\frac{1}{2}R^{-5}\frac{\partial}{\partial R}R^5\frac{\partial}{\partial R} - \frac{1}{2R^2}\left\{\frac{1}{\sin 4\psi}\frac{\partial}{\partial \psi}\sin 4\psi\frac{\partial}{\partial \psi}\right.$$
$$\left. + \frac{4}{\cos^2 2\psi}\frac{\partial^2}{\partial \varphi^2}\right\} + \frac{C(\psi,\varphi)}{R} \tag{11}$$

with C given by (6). Unfortunately, (11) is not separable. Nevertheless we present here an exact solution in the Coulomb zone near the saddle point of $C(\psi,\varphi)$ which controls high double excitation. To this end we generate the wave function in product form as follows.

For the intrinsic motion we employ oscillator functions, $\exp\{-\kappa\psi^2\}$ for the stable bending motion and $\exp\{i\lambda(\varphi-\pi)^2\}$ for the unstable anti-oscillator of the radial correlation. For the hyper-radial motion we use an Eikonal description because the potential surface shows due to Coulomb forces only smooth variation. Thus we arrive at the following wave function of the atom

$$\Psi(R,\psi,\varphi) = R^{\frac{5}{2}}\exp\left\{i\int^R K(R')\,dR'\right\}\cdot\exp\left\{-\kappa\psi^2 + i\lambda(\varphi-\pi)^2\right\} \tag{12}$$

The function (12) appears suitable for the Hamiltonian near the saddle point. Its Taylor expansion reads there

$$H = -\frac{1}{2}\frac{\partial^2}{\partial R^2} - \frac{1}{2R^2}\frac{1}{\psi}\frac{\partial}{\partial \psi}\psi\frac{\partial}{\partial \psi} - \frac{2}{R^2}\frac{\partial^2}{\partial \varphi^2} + \frac{15}{8R^2}$$
$$- \frac{C_0}{R} + \frac{C_1}{2R}\psi^2 - \frac{C_2}{2R}(\varphi-\pi)^2 \tag{13}$$

where also the charge function $C(\psi,\varphi)$ has been Taylor expanded around the saddle point with the following coefficients:

$$C_0 = \frac{4Z-1}{\sqrt{2}} \tag{14}$$

represents the net charge on the top of the ridge in the (R, φ) subspace, and

$$C_1 = \frac{1}{2\sqrt{2}} \tag{15}$$

is the frequency of the bending vibration, and

$$C_2 = \frac{12Z-1}{4\sqrt{2}} \tag{16}$$

is the curvature across the ridge.

Our problem, however, is still not separable. The key point is that the coordinate R is not a constant but changes during the e + He⁺ collision. Remember that the Born-Oppenheimer approximation [7] works with constant values of R; that shortcoming has been removed now [1]. Because we describe the electron-electron interaction in a moving frame, the frequencies κ and λ must be treated as functions of R. That motion proceeds along the evolution coordinate R. Substitution of (12) into (13) yields then the following relations

$$K^2 = 2\left(E + \frac{C_9}{R} - \frac{\frac{15}{8} + 2\kappa - i\lambda}{R^2}\right) \tag{17}$$

$$\kappa^2 = \frac{RC_1}{4} - \frac{iKR^2}{2}\frac{d\kappa}{dR} \tag{18}$$

$$\lambda^2 = \frac{RC_2}{16} - \frac{KR^2}{8}\frac{d\lambda}{dR} \tag{19}$$

We treat (18), (19) as follows: Beginning with (19) we use the zero-energy Coulomb zone approximation by putting $E = 0$ in (17), and conclude $K \cong \pm\sqrt{\frac{2C_0}{R}}$, since at zero energy the Coulomb zone extends to infinity. Further we conclude from (19) the R-dependence $\lambda(R) \propto \sqrt{R}$. Therefore the derivative term on the rhs of (19) is $\propto R$, and may be combined with the static curvature $C_2 \to C_2 + D$ where D is independent of R. We observe that the derivative term has deformed the static curvature of the antioscillator. Dominant correlation has manifested itself by a deformation of the electrostatic potential energy surface. The analysis of (18) runs along the same lines.

Equations (18) and (19) constitute nonlinear Riccati differential equations. Long ago Peterkop [11] assumed in a WKB treatment constant values for κ, λ. That corresponds to an adiabatic approximation, equivalent to a standard Born-Oppenheimer approximation. We avoid here such an approximation, and solve exactly the Riccati equations. That step corresponds to the inclusion of a new momentum-momentum coupling in the Hamiltonian [1]. Actually the derivative terms on the rhs of (18, 19) change the frequency of the bending vibration, see (18), and the curvature of the antioscillator, see (19). Moreover, the total energy E enters through the radial momentum K. We stress that K carries two alternative signs describing incoming and outgoing waves. Therefore we expect different wave propagations in the two cases of motion, see the next section.

So far we have treated singlet events (spin zero). The wave function is then even with respect to electron exchange. In terms of our coordinates exchange is provided by the transformation $\varphi \to 2\pi - \varphi$, R and ψ unchanged. The odd wave function

$$\Psi = R^{5/2}\exp\left\{i\int^R K(R')dR'\right\}(\varphi - \pi)\exp\left\{-\kappa\psi^2 + i\lambda(\varphi - \pi)^2\right\} \tag{20}$$

describes therefore triplets in the collinear configuration. Substitution into the wave Equation (13) delivers then the same relations (17)-(19) as for singlets. *I.e.* the potential deformation is spin-independent.

4. Analysis of the Potential Deformation

In Equations (17)-(19) appear three unusual quantities. In (17) appears the term $\frac{15}{8R^2} = \frac{\frac{3}{2}\left(\frac{3}{2}+1\right)}{2R^2}$. That looks like a squared orbital angular momentum divided by a moment of inertia. That conclusion, however, is wrong. We study here pure S states; there is no rotation at all. The term under consideration stems from the six dimensional description of the electrons. Only in the space $\mathbb{R}_3$ occurs no

pseudo centrifugal term. That term has therefore purely geometrical origin. However, we will see below that our angles are useful and necessary because this term is essential for the description of the formation of electron pairs and their decay, see §5 of this paper.

Equation (19) has the following interpretation: If we would assume a constant value λ the solution would be the adiabatic one given by $\lambda = \pm \frac{1}{4}\sqrt{RC_2}$. We stress, however, that we describe here the electron-electron interaction in a moving frame. The motion runs along the evolution coordinate R. It is well known that in moving frames fictitious forces may occur. The R-dependence of λ may be attributed to such a fictitious force. We compare now the exact solution of (19) with the adiabatic one. To this end we remember that the dependence of λ on R is simply $\lambda(R) = \bar{\lambda}\sqrt{R}$ with $\bar{\lambda}$ independent of R, see above. This reduces (19) to an algebraic equation with two real solutions.

In (18) we find an analogous situation. The adiabatic solution $\kappa = \pm \frac{1}{2}\sqrt{RC_1}$ has been corrected by a complex value of κ.

The dynamical contributions from correlation presented here are not negligible. We will see below that they may be quite important, and depend explicitly on the momentum vector K. Therefore, they are different for shrinking and expanding modes of motion of the whole complex.

Equation (19) is closely related to the escape of two slow electrons after ionization near threshold. For small values of ψ this angle is equal to the solid angle Θ between the two escaping electrons. In consequence of the electrostatic repulsion between the electrons we should expect a peak in the angular distribution at $\psi \cong \Theta \cong \pi$, *i.e.* the electrons escape into opposite directions.

In the triply differential cross section measurements the data by the Erhardt group [12] however don't show such a peak. The angular distribution is more or less flat. This is not a contradiction to the Wannier theory, but it is a wrong interpretation of the theory. To clear the situation we look into the exact solution of (18). To this end we put $\kappa(R) = \bar{\kappa}\sqrt{R}$, $\bar{\kappa}$ is then independent of R, and we arrive at the algebraic equation

$$\bar{\kappa}^2 - i\bar{\kappa}\sqrt{\frac{C_0}{2}} - \frac{C_1}{4} = 0 \tag{21}$$

This equation has exclusively imaginary roots. The distribution

$$\left| \exp\left\{ -\kappa\psi^2 \right\} \right|^2 \tag{22}$$

is therefore uniform rather than peaked at $\psi = \pi$ in agreement with Erhardt's observation. This is a convincing manifestation of a dominant correlation effect due to a fictitious force in electron-atom scattering.

Also the quantity $\lambda(R)$ shows the simple behavior $\lambda(R) = \pm\bar{\lambda}\sqrt{R}$ with $\bar{\lambda}$ independent of R, see above. The equation analogous to (21) has now two real solutions. That implies that incoming and outgoing flux are different, a phenomenon entirely foreign in two-body collisions. The terms "incoming" and "outgoing" have now in the three-body system the meaning of "shrinking" and "expanding" modes of the whole complex. The derivative term in (19) is respon-

sible for electron-electron attraction and for pair formation analogous to Cooper [13] pairs, see §5 of this paper. Moreover, the squared ratio of the radial expanding current divided by the shrinking one delivers the Wannier Formula (1).

5. Supercurrent

We present here a vision study of a linear cluster whose ingredients are atoms with two valence electrons irrespective if such a target is available or not. In that situation the wave function (12) is no longer applicable. Due to the confinement we must replace (12) by

$$\Psi\left(R,\psi,\varphi\right) = R^{5/2} \exp\left\{-\int K\left(R'\right)\mathrm{d}R'\right\} \exp\left\{-\kappa\psi^2 - \lambda\left(\varphi - \pi\right)^2\right\} \qquad (25)$$

Putting (25) into (13) we rediscover except for an imaginary factor of i Equations (17)-(19). We conclude that the confinement does not change correlation contributions.

Therefore a current in a linear arrangement of atoms may run as follows. In consequence of the electrostatic repulsion the three bodies (core + two valence electrons) enter into a collinear configuration (electron-core-electron). In that configuration the fictitious force attracts the electrons. That force transports the electrons to the top of the antioscillator. This part of the process was referred to by Wannier [4] as converging trajectory. The pair is now born, and is electrostatically attracted by the core. The electron pair, however, does neither penetrate into the core nor do the electrons fall into the nucleus. The pair as a whole will be reflected by the pseudo centrifugal barrier given by $\dfrac{15}{8R^2}$. At this step of the electrons motion the force between the core and the pair changes sign. During the electrons were on the ridge top they jump now onto Wannier's diverging trajectory. The pair motion is now unstable. The electron pair falls down from the ridge top, *i.e.* the pair dies because the correlation-induced force between the electrons is now repulsive, see (19). Nevertheless the electrons don't escape to the universe because of the confinement. One electron will be trapped into a Rydberg orbital of the positive core. The other electron experiences a repulsive force with respect to the mother core. But in the frame of the next neighbour atom that force is attractive. A neighbor core attracts the electron, and the whole procedure repeats. Along these lines one electron is transported along the whole chain of atoms and constitutes a current. On that path there is no inelastic collision in which the electron would lose energy. The charge transport is provided here exclusively by electron wave diffraction. That implies we have a current without any resistance. Finally we stress that the pair presented here is not a Cooper pair [13] because in our case the binding does not result from an electron-phonon interaction but from dominant correlation at threshold for double ionization.

6. Generalization to More Than Two Electrons

Recently the author has shown that also atoms with $N = 3$ or 4 electrons possess

unstable equilibrium configurations [14]. Three highly excited electrons may form an equilateral triangle configuration with one electron in each corner. Instead of a pair formation we predict now the production of an electron triple. To this end we consider for instance a high doubly excited resonance He[**] located slightly below the double ionization threshold (ca. 79 eV). A further slow electron will be attracted to that target, and the complex will enter into a triangular equilibrium configuration. After reflection from a pseudo centrifugal barrier now given by $6/R^2$ the triple decays into three individual electrons. One of them is ejected, and the other two fall down into a lower doubly excited helium state He[**]. The situation of $N = 4$ electrons runs along similar lines.

7. Conclusion

For nearly a century the atomic and molecular community relies on the Born Oppenheimer approximation [7]. Unfortunately the BO method has inadvertently disregarded one important portion of the Hamiltonian, namely a momentum-momentum coupling. The present paper has tried to figure out what the rejected energy term may deliver. The result of the present pilot investigation is encouraging. Therefore a forthcoming paper will be dedicated to a systematic representation of an amended Born-Oppenheimer theory [1].

Conflicts of Interest

The author declares no conflicts of interest regarding the publication of this paper.

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Appendix

List of Symbols

σ = Total ionization cross section

E = Total Energy

E_0 = Ionization threshold

R = Hyperradius

$\boldsymbol{r}_1, \boldsymbol{r}_2$ = Elctron positions

V = Potenialenergy

r_{12} = Electron-electron separation

Z = Nuclear charge

$C(\psi, \varphi)$ = Charge function

ψ, φ = Body-fixed hyperspherical angles

Θ_{ii} = Moment of inertia around the x-axis

H = Hamiltonian

Ψ = Wavefunction

K = Radial momentum

C_0 = Electron net charge on the top of the ridge

C_1 = Bending frequency

C_2 = Curvature of the potential ridge