

Stability conditions for the solutions of the half-projected Hartree-Fock scheme. The lithium-hydride ground state

Yves Gabriel Smeyers and Gerardo Delgado-Barrio

Instituto de Estructura de la Materia del Consejo Superior de Investigaciones Científicas, Serrano 119, Madrid-6, Spain

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The stability conditions for the solutions of a two-unrestricted-determinant function (the half-projected Hartree-Fock function) are deduced from the necessary requirements for the minimization of the energy functional. The theory is applied to the case of the LiH ground state, in order to investigate the various solutions encountered in the variational problem. It is found that the two lowest solutions are local minima. The number of these solutions is estimated and their significance discussed as a function of the nuclear separation.

I. INTRODUCTION

The stability conditions for the solutions of the Hartree-Fock equations were first formulated by Thouless for a single determinant function.¹ This author deduced the expression that ensures the minimization of the wave function from the necessary conditions for minimizing the energy functional. Adams reformulated the stability conditions from the first-order density-matrix formalism and compared the relative stability of the restricted (RHF) and unrestricted Hartree-Fock schemes (UHF).² Similarly, Cizek and Paldus studied the instability of the RHF solutions with respect to variations, which release the double occupancy conditions, in cyclic polyenes, linear polyacenes, and odd linear polyenic radicals.³ Furthermore, from the variational conditions, the multiple solutions to the RHF problem were investigated by Stanton.⁴ On the other hand, the stability of a RHF function with respect to symmetry breakdowns was studied by Kaplan *et al.* from another point of view based on an upper-bound theorem for the RHF ground energy.⁵

In the present work, we extend the above investigations and deduce the stability conditions for the solutions of a two-unrestricted-determinant function, i.e., the half-projected Hartree-Fock function (HPHF).⁶ The latter has been found to be very useful in the study of bond-breaking process,⁷ and was successfully employed in calculations on small molecules.⁸

As is well known, the HPHF function has the form

$$\begin{aligned} \psi^{\text{HPHF}} &= \hat{A}(S) \psi^{\text{UHF}} \\ &= \frac{1}{2} [| \varphi_1 \bar{\varphi}_1 \varphi_2 \bar{\varphi}_2 \cdots | \\ &\quad + (-1)^{n+1} | \bar{\varphi}_1 \varphi_1 \bar{\varphi}_2 \varphi_2 \cdots |], \end{aligned} \quad (1)$$

where $\hat{A}(S)$ is the half-projection operator which projects on the subspace of quantum numbers S even or odd.^{6,7}

When S is even, the HPHF function may be successfully used for the study of singlet ground states, because it does not contain triplet contaminants. In fact, the HPHF function in this case only has one shell which is clearly split. Therefore this function is shown to be approximately equivalent to two natural configurations of closed shells: the fundamental and a second doubly excited one.⁹

Some difficulties arise in the determination of the HPHF function because various solutions, lower than the RHF one, are often encountered in the variational procedure.^{6,8} In this work, we examine this problem from the stability point of view. For this purpose we have investigated the two lowest solutions, found for the LiH ground state, as a function of the nuclear separation.

II. HPHF STABILITY CONDITIONS

A way for determining if the HPHF solution corresponds to a minimum (or a saddle point) is to study the second variation of the energy functional. If this second variation is positive definite the solution corresponds to a real minimum. This may be, however, of local type because of the range of the variation.

Let us express the variation of the occupied orbitals φ_p and φ_p^* in terms of the virtual ones. Thus,

$$\delta\varphi_p = \sum_i c_i^p \varphi_i, \quad \delta\varphi_p^* = \sum_i d_i^p \varphi_i^*, \quad (2)$$

where the coefficients c_i^p and d_i^p are infinitesimal. The first variation of the HPHF function may then be written as a linear combination of singly ex-

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