A GEOMETRIC APPROACH TO CORRELATED SYSTEMS

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1. INTRODUCTION

Over the past few decades there has been an impressive and a steady progress in computational material science [1, 2, 3]. This development is fueled by the ever growing computational resources and by the demand for yet more precise information on technologically relevant material properties, such as the optical, transport and magnetic characteristics. A microscopic description of these properties entails the knowledge of the quantum spectrum of the system under study. Thus for real materials one has to deal with the notoriously difficult many-body problem in a computationally acceptable manner. For this purpose remarkably successful and efficient conceptual schemes have been developed where the multi-particle system is mapped onto a one body problem for a particle moving in an effective (non local) field created by all the other constituents of the system [1, 2, 4]. Usually, this effective field is further simplified according to certain recipes such as the local density approximation within the density functional theory [5]. These computationally manageable concepts have rendered possible the routine and accurate calculation of a wealth of static material properties, such as the ground state energies. On the other hand, however, it has been observed that the ground-state of certain compounds, e.g. transition metal oxides, is not described adequately within a single particle picture [2]. In addition, for the theoretical description of the excitation spectrum [6] and for the treatment of dynamical processes, such as many particle reactive scattering, methods have to be envisaged that go beyond the effective single particle model.

For example, correlated many-body states are a prerequisite for the theoretical formulation of recent measurements of the double and triple electronic

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excitation of localized or delocalized electronic compounds by one ultraviolet photon [7, 8, 9]. Recent technological advances have made it even possible to explore in full details the many-body continuum spectrum of four and more interacting particles [9, 10, 11, 12, 13, 14, 16] where numerical calculations are absent. Thus, it is desirable to develop, for interacting systems, a genuine many-body theory (in a sense specified below) that is conceptually sound and allows for systematic approximations that can be implemented numerically. This is particularly important in view of the current trend in miniaturization techniques that ultimately aim at the fabrication of atomic-size systems whose features are controlled primarily by the quantal behaviour of a finite number of correlated particles.

In this theoretical work a strategy is presented that enables us to derive approximate expressions for the spectrum of many-body, interacting finite systems by utilizing the knowledge of the properties of other reference systems with a less number of interactions. The treatment is not perturbative, i.e. it does not rely on pre-assumptions regarding the strength of a certain interaction or characteristic parameter in the system. Furthermore, the method provides a systematic and mathematically sound scheme for approximations that are computationally tractable and allows, when desirable, to incorporate well established single particle and perturbative approaches.

2. FORMAL DEVELOPMENT

The fundamental quantity from which we derives the microscopic properties of N body quantum systems is the Green operator $G^{(N)}$ which is the resolvent of the total Hamiltonian. It is defined by the Lippmann-Schwinger integral equation $G^{(N)} = G_0 + G_0 U^{(N)} G^{(N)}$ where G_0 is the Green operator of a reference N particle system which is usually chosen as a noninteracting system. U stands for total interaction and is given formally by $U^{(N)} = G^{-1} - G_0^{-1}$. Equivalently one may determine the dynamical behaviour of a system by means of the transition operator $T^{(N)}$ which satisfies the integral equation $T^{(N)} = U^{(N)} + U^{(N)} G_0 T^{(N)}$. These integral equations for $G^{(N)}$ and $T^{(N)}$ provide a natural framework for perturbative treatments, as they can be expanded in terms of interaction $U^{(N)}$ or some parts of it which are deemed small. However, for $N \geq 3$ the application of the above Lippmann-Schwinger equations (and those for the state vectors) is hampered by mainly two difficulties: 1.) as shown in Refs.[17, 18] the Lippmann-Schwinger equations for the state vectors do not have a unique solution, and 2.) as shown by Faddeev [19, 20] the kernel of these integral equations $K = G_0 U^{(N)}$ is not a square integrable operator

for $N \geq 3$, i.e. the norm $\|K\| = [\mathrm{Tr}(KK^\dagger)]^{1/2}$ is not square integrable. The kernel K is also not compact. The origin of these obstacles lies in the presence of the so-called disconnected diagrams that correspond to processes in which one of the N particles is a spectator, i.e. not correlated with the other $N-1$ particles. Therefore, for three-particle systems Faddeev and others [19, 22, 23] suggested an alternative set of equations for $G^{(N)}$ and $T^{(N)}$ that are free from disconnected diagrams (the behaviour of these equations for infinite range potentials, such as Coulomb potentials, is still the subject of current research [21]).

The scope of this study is three fold: (a) We aim at deriving a recursive procedure that expresses the Hamiltonian of N interacting body systems in terms of Hamiltonians of systems with a reduced number of interactions.
(b) This reduction scheme should not be a perturbative one, and the diagrams should be arranged in a way that disconnected terms are excluded. The procedure should be linear in N, i.e. it should relate $G^{(N)}$ and $T^{(N)}$ to $G^{(N-M)}$ and $T^{(N-M)}$ (where $M = 1, 2, \cdots, N-2$) in a linear manner in N.
(c) The method should provide a systematic and mathematically sound scheme for approximations that could be computationally realized (here we evaluate the continuum spectrum of four interacting Coulomb particles and two particle excitations at surfaces).

To this end we consider a nonrelativistic system consisting of N interacting particles. Total potential is assumed to have the form $U^{(N)} = \sum_{j>i=1}^{N} v_{ij}$, where v_{ij} stands for a two-particle interaction without any further specification of its analytical form.

Based on geometrical considerations (cf. Fig.1) one can express the potential $U^{(N)}$ exactly in terms of recurrence relations:

$$U^{(N)} = \frac{1}{N-2} \sum_{j=1}^{N} u_j^{(N-1)}, \tag{1}$$

$$u_j^{(N-1)} = \frac{1}{N-3} \sum_{k=1}^{N-1} u_{jk}^{(N-2)}, \quad j \neq k. \tag{2}$$

In these equations $u_j^{(N-1)}$ is the total potential for a system of $N-1$ interacting particles, i.e. $u_j^{(N-1)}$ is obtained from $U^{(N)}$ by detaching particle j from the N particle system. In terms of the pair interactions v_{mn} the following relation applies $u_j^{(N-1)} = \sum_{m>n=1}^{N} v_{mn}$, $m \neq j \neq n$. Since the kinetic energy operator is an additive single particle operator the expansions (1,2) are reflected in similar recursion relations for the total Hamiltonian. Fig. 1 illustrates how, according to Eqs.(1,2), the potential of a system of six interacting particles is expressed

in terms of potentials of five correlated particles. The latter potentials can be further expressed in terms of four body potentials (cf. Eq.(2)). This procedure is repeated until a potential with a desired number of interactions is reached.

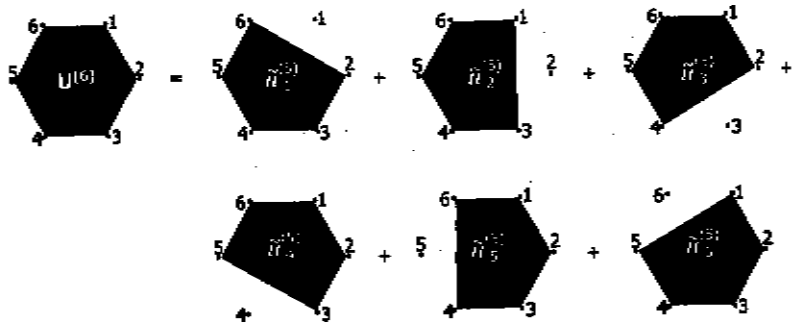

Figure 1: A pictorial interpretation of the total potential expansion (1) for six interacting particles enumerated and marked by the full dots at the corners of the hexagon. The hexagon indicates the full potential $U^{(6)}$ of the six correlated particles. Each pentagon symbolizes the full five body potential $\tilde{u}_j^{(5)} = \left(u_j^{(5)}\right)/4$ of those five particles that are at the corners of the pentagon. The particle being not at a corner of a pentagon is free (disconnected).

From Fig. 1 it is clear that this "minimal geometric reduction" scheme (Eqs.(1,2)) treats all interactions on equal footing and provides maximal flexibility to reduce systematically the N body potential (Hamiltonian) to sums of $N-M$ potentials (Hamiltonians) with $M = 1, 2, \cdots, N-2$.

The geometric expansion (Eqs.(1,2)), as visualized in Fig.1, has a wide ranging consequences in that the transition and the Green operators can be expanded along the same lines!.

This can be seen as follows: According to the decomposition (1), the integral equation for the transition operator can be written as

$$T^{(N)} = \sum_{j=1}^{N} T_j^{(N-1)} \tag{3}$$

$$T_j^{(N-1)} = \tilde{u}_j^{(N-1)} + T^{(N)} G_0 \tilde{u}_j^{(N-1)}, \; j = 1, \cdots, N. \tag{4}$$

Here we introduced the scaled potentials $\tilde{u}_j^{(N-1)} = \left(u_j^{(N-1)}\right)/(N-2)$. The physical meaning of the operators (4) is illustrated in Fig. 2 for the system depicted in Fig. 1.

The transition operator $t_j^{(N-1)}$ for a system of $N-1$ particles interacting via the scaled potential $\tilde{u}_j^{(N-1)}$ is

$$t_j^{(N-1)} = \tilde{u}_j^{(N-1)} + \tilde{u}_j^{(N-1)} G_0 t_j^{(N-1)}.$$

With this relation Eq.(4) can be reformulated to yield

$$\begin{aligned} T_j^{(N-1)} &= t_j^{(N-1)} + t_j^{(N-1)} G_0 T^{(N)} - t_j^{(N-1)} G_0 \left(\tilde{u}_j^{(N-1)} + \tilde{u}_j^{(N-1)} G_0 T^{(N)}\right) \\ &= t_j^{(N-1)} + t_j^{(N-1)} G_0 \left(T^{(N)} - T_j^{(N-1)}\right) = t_j^{(N-1)} + t_j^{(N-1)} G_0 \sum_{k \neq j}^{N} T_k^{(N-1)}. \end{aligned} \tag{5}$$

Eq.(5) can be expressed compactly in the matrix form

$$\begin{pmatrix} T_1^{(N-1)} \\ T_2^{(N-1)} \\ \vdots \\ T_{N-1}^{(N-1)} \\ T_N^{(N-1)} \end{pmatrix} = \begin{pmatrix} t_1^{(N-1)} \\ t_2^{(N-1)} \\ \vdots \\ t_{N-1}^{(N-1)} \\ t_N^{(N-1)} \end{pmatrix} + [\mathbf{K}^{(N-1)}] \begin{pmatrix} T_1^{(N-1)} \\ T_2^{(N-1)} \\ \vdots \\ T_{N-1}^{(N-1)} \\ T_N^{(N-1)} \end{pmatrix}. \tag{6}$$

The kernel $[\mathbf{K}^{(N-1)}]$ is a matrix operator and is given by

$$[\mathbf{K}^{(N-1)}] = \begin{pmatrix} 0 & t_1^{(N-1)} & t_1^{(N-1)} & \cdots & t_1^{(N-1)} \\ t_2^{(N-1)} & 0 & t_2^{(N-1)} & \cdots & t_2^{(N-1)} \\ \cdots & \cdots & \cdots & \cdots & \cdots \\ t_{N-1}^{(N-1)} & \cdots & t_{N-1}^{(N-1)} & 0 & t_{N-1}^{(N-1)} \\ t_N^{(N-1)} & \cdots & t_N^{(N-1)} & t_N^{(N-1)} & 0 \end{pmatrix} G_0. \tag{7}$$

From Eq.(2) we deduce that $t_j^{(N-1)}$ is expressible in terms of transition operators of the $N-2$ interacting subsystems as $t_j^{(N-1)} = \sum_{k \neq j}^{N-1} T_k^{(N-2)}$. The operators $T_k^{(N-2)}$ are deduced from Eq.(6) with N being replaced by $N-1$.

From the relation $G^{(N)} = G_0 + G_0 T^{(N)} G_0$ we conclude that the Green operator of the interacting N particle system has the form

$$G^{(N)} = G_0 + \sum_{j=1}^{N} G_j^{(N-1)}.$$

The reduced operators $G_j^{(N-1)}$ are related to the Green operators $g_j^{(N-1)}$ of systems in which only $N-1$ particles are correlated by virtue of $\tilde{u}_j^{(N-1)}$. This relation is readily deduced from the above derivation for transition operator and reads

$$\begin{pmatrix} G_1^{(N-1)} \\ G_2^{(N-1)} \\ \vdots \\ G_{N-1}^{(N-1)} \\ G_N^{(N-1)} \end{pmatrix} = \begin{pmatrix} g_1^{(N-1)} - G_0 \\ g_2^{(N-1)} - G_0 \\ \vdots \\ g_{N-1}^{(N-1)} - G_0 \\ g_N^{(N-1)} - G_0 \end{pmatrix} + [\tilde{\mathbf{K}}^{(N-1)}] \begin{pmatrix} G_1^{(N-1)} \\ G_2^{(N-1)} \\ \vdots \\ G_{N-1}^{(N-1)} \\ G_N^{(N-1)} \end{pmatrix}, \tag{8}$$

where the kernel is now redefined as $[\tilde{\mathbf{K}}^{(N-1)}] = G_0[\tilde{\mathbf{K}}^{(N-1)}]G_0^{-1}$.

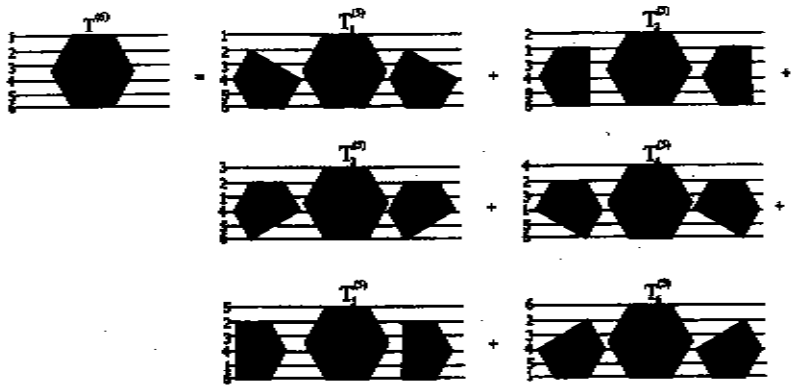

Figure 2: A diagrammatic representation of Eq.(4) for a system of six correlated particles (cf. Fig. 1). The hexagons and the pentagons (with a specific orientation) label the same potentials as explained in Fig. 1. Each of the pictures stands for a transition operator of the six body system (the particles are labeled by straight lines). E. g., the diagram $T_1^{(5)}$ means that the five particles 2, 3, 4, 5 and 6 interact first, propagate and then all 6 particles interact with each other. The system then propagates and finally the five particles 2, 3, 4, 5 and 6 interact again.

From Eqs.(6,8) we conclude that if the Green operator of the interacting $N-1$ body system is known the Green operator of the N particles can then be deduced by solving a set of N linear, coupled integral equations (namely Eqs.(6,8)). According to the above equations, if only the solution of the $N-M$ problem is known where $M = 1, 2, \cdots, N-2$ we have to perform a hierarchy of calculations starting by obtaining the solution for the $N-M+1$ problem and repeating the procedure to reach the solution of the N body problem.

It is straightforward to show that any iteration of the kernels of Eqs. (6,8) is free of disconnected terms since the disconnected terms occurs only in the off-diagonal elements of $[\mathbf{K}^{N-M}]$ and $[\tilde{\mathbf{K}}^{N-M}]$. For $N = 3$ the present scheme reduces to the well established Faddeev equations.

Figure 3: A diagrammatic representation of the approximation (9). The square represent the four-body total potential whereas the triangles are the scaled, total three-body potentials of those three particles whose lines cross the respective triangle.

3. EXCITATION SPECTRUM OF FOUR CHARGED PARTICLES

To demonstrate the applicability of the present approach we consider the four charged-particle problem. This is of a particular interest, since a substantial amount of knowledge on the three particle problem has been accumulated over the years whereas theoretical studies on the four body problem are still scare. On the other hand, in recent years an impressive progress has been made on the experimental side [9, 10, 11, 12, 13, 15] which renders possible a detailed insight into the four body continuum spectrum.

Using the method outlined above of the four body Green operator can be expressed in terms three body Green operators for which approximate solutions may be employed. For $N = 4$ the first iteration of Eq.(8) yields

$$G^{(4)} = \sum_{j=1}^{4} g_j^{(3)} - 3G_0, \tag{9}$$

where $g_j^{(3)}$ is the Green operator of an interacting three body system (particle j is noninteracting). For the latter Green function we utilize the method developed in Ref.[24] (it is important to note in this context that the *three-*

body Green function defined here is called in quantum field theory the *two-particle* Green function which is defined through the Bethe-Salpeter equation). According to the theory in Ref.[24] the Hamiltonian of a general three body system reduces to a sum of three commuting Hamiltonians $h_k^{(2)}$ in which only two particles are interacting (particle k is free). The Green operators $g_j^{(3)}$ in Eq.(9) can therefore be written as

$$g_j^{(3)} \approx G_0^{-2} \prod_{k=1}^{(3)} g_k^{(2)}, k \neq j \in \{1,2,3,4\},$$

where $g_k^{(2)}$ is the resolvent of $h_k^{(2)}$ (this approximation for the three-body Green functions corresponds to summing all the diagrams in the ladder approximation which is done numerically). Thus we obtain from Eq.(9) $G^{(4)} = \left[\sum_{j=1}^{4} G_0^{-2} \prod_k^3 g_k^{(2)}\right] - 3G_0$, $j \neq k$.

Now we employ this four-body Green function for the description of three excited, interacting electrons (or two electrons and a positron) that moves in the Coulomb field of a residual positively charged ion. Such a state is achieved following the electron and positron impact double ionization.

Figs. 3(a,b) show the results for the electron and the positron impact double ionization of the ground state of atomic helium along with the experimental data [10, 15] and a full numerical evaluation of the first order perturbation series (first Born term) within a convergent close coupling (CCC) method [25]. The first Born approximation (FBA) corresponds to one term in Eq.(9) where the projectile motion is decoupled from the rest of the system. As a result of the perturbative treatment the calculations within the FBA are insensitive to the projectile charge state.

As seen from Fig. 4, the present model is capable of describing adequately the experimental cross sections in shape and magnitude. The origin of the peaks observed in the spectrum has been discussed in Refs. [15, 26]. The present model and the FBA theory yield similar results since the experiments are performed in the validity range of the perturbation theory. However, the differences between the positron and the electron impact case indicate that the Born limit is not yet fully reached.

5. THE TWO-PARTICLE EXCITATION AT SURFACES

Now let us turn to the application of the theory to extended systems. In particular the method will be applied to evaluate the simultaneous two-particle excitation at surfaces upon the absorption of a single VUV photon. Such experiments have been done recently [8] on Ni(001) and Cu(001). In the experiment one resolves the wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ (i.e. the energies E_1, E_2

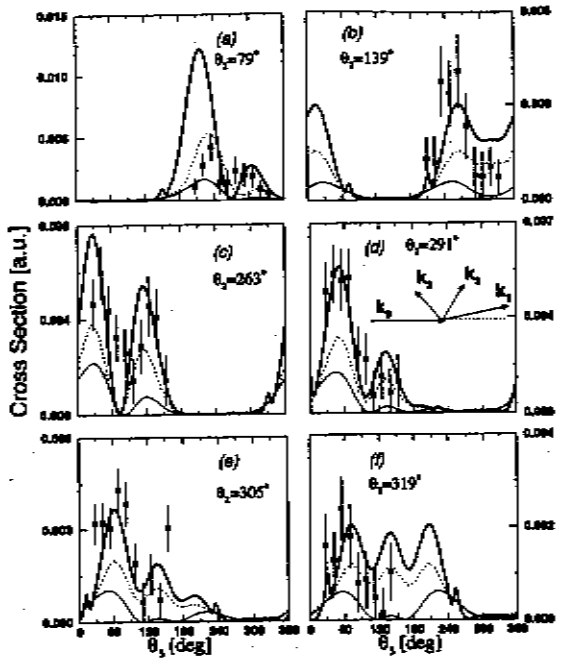

Figure 4: The fully resolved double ionization cross section of $He(^1S^e)$ following electron (solid lines) or positron (dotted line) impact. The scattering geometry is shown by the inset in (d). $\mathbf{k}_0$ and $\mathbf{k}_1$ are the initial and final state momenta of the projectile while $\mathbf{k}_2$ and $\mathbf{k}_3$ refer to the momenta of the two ejected electrons. The incident energy is $5.6\,keV$ and $k_f^2/2 = k_3^2/2 = 10\,eV$. All angles are measured with respect to $\hat{\mathbf{k}}_0$. The projectile is scattered through an angle of 0.45°. The emission angle θ_2 of one of he electron is fixed at the value indicated on the figures while the cross section is scanned as function of the emission angle θ_1 of the second electron. The thick solid (dotted) line is the result of the present model for electron (positron impact) whereas the light solid curve is the outcome of the CCC method within the first Born approximation [25]. The data (full square [15]) are on absolute scale.

and emission angles $\hat{\mathbf{k}}_1, \hat{\mathbf{k}}_2$) of the two photoelectrons emitted simultaneously from the sample upon the absorption of a single VUV photon with energy $\hbar\omega$. The probability for such a reaction is given by [28, 27] ($E = E_1 + E_2$)

$$W(\hat{\mathbf{k}}_1, E_1, \hat{\mathbf{k}}_2, E_2, \hbar\omega) \propto \langle \mathbf{k}_1, \mathbf{k}_2 | \Pi_{12}^- \, \Delta \, \Im\Pi_{12}(E - \hbar\omega) \Delta^\dagger \, \Pi_{12}^- | \mathbf{k}_2, \mathbf{k}_1 \rangle. \quad (10)$$

Here $\Pi_{12}^{\pm}$ is the particle-particle (p-p) propagator [29] appropriate for the description of the two interacting photoelectrons in the presence of the surface.

The method presented in this paper can be utilized upon the following assumptions: 1. We treat the two electrons as independent ones in which case the single particle Green function for each electron can be deduced by conventional methods, such as density functional theory (we employ the layer Korringa Kohn Rostoker technique). 2. The electrostatic interaction between the two electrons is assumed local and is renormalized according to the Thomas-Fermi theory of screening. With this information we are now in a position to write down the p-p propagator as sum of three quasi-particle propagators. More details concerning the range of validity of the approximations will be given elsewhere. In Fig.5 the two-electron energy correlation function is shown along with the experimental results on Cu(001). As evident, the emission probability of one of the photoelectrons with certain energy depends strongly of the properties of the second photoelectrons which endorses the importance of a realistic treatment of the coupling between the electrons.

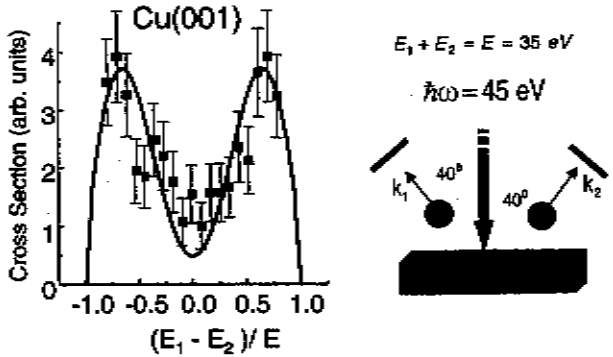

Figure 5: The one-photon two-electron emission from Cu(001). The photon with energy $\hbar\omega = 45$ eV is linear polarized and its wave vector is normal to sample. The two photoelectrons are detected co-planar with the photon wave vector and under emission angles of 40° to the left and to the right of the incident photon direction (cf. inset). The total energy of the two escaping electrons is $E = 35$ eV. The emission cross section is scanned as function of sharing of the total energy E between the two photoelectrons

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References

[1] A. Gonis *Theoretical materials science : tracing the electronic origins of materials behavior*, (Warrendale, Pa : Materials Research Society, 2000)

[2] I. Turek, V. Drchal, J. Kudrnovský, M. Šob, and P. Weinberger, *Electronic Structure of Disordered Alloys, Surfaces, and Interfaces*, (Kluwer Academic Pub., Boston, London, Dordrecht, 1997)

[3] K. Ohno, K. Esfarjani, and Y. Kawazoe, *Computational Material Science*, (Springer, Berlin, 1999)

[4] P. Fulde, *Electron Correlation in Molecules and Solids*, Springer Series in Solid-State Sciences, Vol. 100, (Springer Verlag, Berlin, Heidelberg, New York, 1991)

[5] R. M. Dreizler and E. K. U. Gross *Density Functional Theory* (Springer Verlag, Berlin, Heidelberg, New York, 1990)

[6] L. Hedin, J. Phys. C 11 R489 R528 (1999)

[7] O. Schwartzkopf, B. Krässig, J. Elmiger, and V. Schmidt Phys. Rev. Lett **70**, 3008 (1993)

[8] R. Herrmann, S. N. Samarin, H. Schwabe, and J. Kirschner, Phys. Rev. Lett.**81**, 2148 (1998)

[9] R. Wehlitz et al Phys. Rev. Lett. **81**, 1813 (1998)

[10] I. Taouil, A. Lahmam-Bennani, A. Duguet, L. Avaldi Phys. Rev.Lett. **81**, 4600 (1998)

[11] A. Dorn et al Phys. Rev. Lett. **82**, 2496 (1999)

[12] M. Unverzagt et al Phys. Rev. Lett. **76**, 1043 (1996)

[13] B. El-Marji, J. P. Doering, J. H. Moore, M. A. Coplan Phys. Rev. Lett. **83**, 1574 (1999)

[14] R. Moshammer et al Phys. Rev. Lett. **84**, 447 (2000)

[15] A. Lahmam-Bennani et al Phys. Rev. A **59**, 3548 (1999)

[16] A. Dorn et al Phys. Rev. Lett. **86**, 3755 (1999)

[17] B. A. Lippmann, Phys. Rev. **102**, 264 (1956)

[18] L.L. Foldy and W. Tobocman, Phys. Rev. **105**, 1099 (1957)

[19] L.D. Faddeev, Soviet Phys. JETP **12**, 1014 (1961)

[20] L.D. Faddeev *Mathematical Aspects of the Three-Body Problem* (Davey, New York, 1965)

[21] A. M. Mukhamedzhanov, E. O. Alt, G. W. Avakov, Phys. Rev. C **61**, 064006 (2000)

[22] O.A. Yakubovsky, Sov. J. Nucl. Phys. **5**, 937 (1967)

[23] S.P. Merkuriev and L.D. Faddeev *Quantum Scattering Theory for Systems of few Particles* (Nauka, Moscow, 1985)

[24] J.Berakdar, Phys. Lett. A **277**, 35 (2000)

[25] A. Kheifets et al J.Phys.B **32**, 5047 (1999)

[26] J. Berakdar, Phys. Rev. Lett. **85** , 4036 (2000)

[27] N. Fominykh, J. Henk, J. Berakdar, P. Bruno, H. Gollisch, R. Feder , Solid State Commun. **113**, 665 (2000)

[28] J. Berakdar, Phys. Rev. B. **58**, 9808 (1998)

[29] E. K. U. Gross, E. Runge, *Vielteilchentheorie* (Teubner, Stuttgart, 1986)