Many-particle fragmentation processes in atomic and molecular physics – new insight into the world of correlation

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Abstract

Correlated many-particle dynamics in Coulombic systems, which is one of the unsolved fundamental problems in AMO-physics, can now be experimentally approached with so far unprecedented completeness and precision. The recent development of the COLTRIMS technique (COLd Target Recoil Ion Momentum Spectroscopy) provides a coincident multi-fragment imaging technique for eV and sub-eV fragment detection. In its completeness it is as powerful as the bubble chamber in high energy physics. In recent benchmark experiments quasi snapshots (duration as short as an attosecond) of the correlated dynamics between electrons and nuclei have been made for atomic and molecular objects. This new imaging technique has opened a powerful observation window into the hidden world of many-particle dynamics.
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1. Introduction

When calculating the total binding energy of an atomic many-particle system, multi-configuration interaction (MCI) atomic structure theory counts all those contributions to e–e-correlations which cannot be described in the Hartree–Fock (HF) approach [1,2]. To obtain excellent agreement between experimental and theoretical binding energies so called "off-shell" or "pseudo" states have to be included into the wave function basis set. The need for such virtually excited "off-shell" states reflects the non-local behavior of electrons in atomic or molecular states, i.e. electrons can have such large momenta even at large distance from the nucleus that classically these electrons would be in a continuum state and should escape from the bound state. Already Bethe [2,3] predicted that in ground state He the electron momenta in such an "off-shell" state can exceed MeV/c. The non-local behavior allows that these fast electrons "tunnel" to large distances from the nucleus. Thus one finds these very high momenta even far away from the nucleus, too. "Tunneling" means, the electrons can move to large distance and the Coulomb force cannot slow them down. Therefore, far outside one should find only the high momenta, just opposite to classical expectations.

Instead of presenting here a profound theoretical discussion of the e–e correlation physics, we will "explore" the secrets of correlation in the "simple" view of an experimental physicist. Nevertheless, we hope to convince the reader that these correlations might or even must be to a large extent due to perfectly entangled many-particle dynamics. What do we know about this entangled dynamics, i.e. what do we know about the intra-atomic or molecular motion of such closed quantum-mechanical bound many-particle systems? How does a non-local electron move with respect to the other electrons and the nucleus in an atom or molecule, i.e. how is their motion correlated?

From basic laws of physics and also quantum mechanics we know that in case of no external force both the sum of the momenta and the sum of angular momenta of all particles belonging to the closed system must be at any moment perfect constants of motion. Since in a bound many-parti-cle system momentum and angular momentum are conserved at any time, even for the non-local regime there must be an instantaneous entanglement between all particles. Since the electronic motion is non-local, the momentum exchange by the local $1/r^2$-Coulomb forces alone cannot explain the secrets of many-particle dynamics in atoms and molecules. Instantaneous means, virtual photons contribute to momentum and angular momentum conservation, beyond the classical force concept. Thus, if we cool a He atom down to nano-Kelvin temperature, and when the atom stays such cold for hours or days or years, then at any moment the three-particle motion in the He with respect to its center-of-mass must be so perfectly "synchronized", i.e. so "precisely" correlated, as nature defines a fundamental constant (infinite precision).[1]

How do electrons and the nucleus contribute to momentum and angular momentum conservation? Often the nucleus is considered as infinitely heavy particle. But since the nuclear mass is finite and about 10^4 times heavier than the electron mass, the contribution of the nucleus to the internal angular momentum with respect to the atomic center-of-mass is indeed negligibly small. Thus the two He electrons, when they are in a non-s^2 "off-shell" state, carry practically all internal angular momentum. Since the He ground state is a $^1S_0$-state, the angular momentum vectors of both electrons are always pointing in opposite directions and their vector sum is at any moment zero. In such a non-s^2 "off-shell" state both electrons need, however, the nucleus to ensure momentum conservation. In the language of solid state physicists one needs strong phonon coupling of the electronic motion to the nucleus to ensure at any moment simultaneously internal momentum and angular momentum conservation. It is easy to imagine that now all momentum and angular momentum vectors are correlated and they must move fully entangled like the wheels of a railway steam locomotive (RSL). The wheels of a RSL are coupled or let's say entangled by the "piston rod". Consider-

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[1] In classical physics this statement is well accepted, however, since in the quantum world the total energy is not conserved at any moment, even physicists often mistakenly suppose that the total momentum is not conserved at any moment.

ing only the total energy of the moving wheels, the wheels of the RSE could even rotate in opposite directions or having different radii or rotation frequencies. But it is clear that such an "uncorrelated" wheel system would never be able to pull a train. Therefore, like the wheels in a RSE, the intra-atomic motion of all particles must be fully entangled even for the non-local, i.e. tunneling electronic motion.

Unfortunately, mathematics does not provide the tools for exactly solving on a microscopic and fully time dependent basis the many-particle motion. Also it is even questionable whether the Schrödinger or Dirac equations are adequate to ensure in many-particle systems this full entangled motion (at any moment!). To entangle momentum and angular momentum, the operators in the correct quantum mechanical many-particle equation should already correlate at any moment the total momentum and total angular momentum. Therefore, presently it seems to be beyond feasibility that by means of present theoretical approaches the secrets of e–e-correlation in many-particle systems can be revealed. Thus experimental physics must take the lead and go ahead even without the guidance of theory to explore these fundamentally so important aspects of entangled many-particle motion, and in particular to explore the highly correlated long range non-local behavior of such motion.

Recently experimental physics has developed indeed very powerful tools to study and reveal this correlated motion. The COLTRIMS (COLd Target Recoil Ion Momentum Spectroscopy) reaction microscope [4–6] can directly image the secrets of many-particle dynamics in atoms. Through a very sudden ionization process (attosecond duration) the atom or molecule can be fragmented into free electrons and ions in such a way that the intra-atomic momentum structure is nearly unperturbedly projected onto the final momentum state. By using the high resolution COLTRIMS multi-coincidence momentum-imaging technique, the momenta of all emitted charged fragments can be determined. This method measures simultaneously the trajectories of all charged fragments like the bubble chamber in high energy physics. Using selected coincidence requirements between frag-

ments, this technique can even "zoom" into special features of the many-particle wave function, e.g. viewing only the long range tails of the wave function.

The fragmentation process can be induced by ion, electron or photon impact. Depending on the detailed kinematics of the reaction process the final-state momentum pattern, however, can reflect the initial-state correlation or the final-state interaction. To view initial-state correlation the total momentum transferred from the impacting projectile to the whole atom or molecule should be very small even if the transferred energy is high. This is only the case in a photoionization process or fast ion impact at large impact parameters, where high-energy virtual photons are inducing the fragmentation. Furthermore, the fragments should have a very small post-collision interaction. This is the case when the emitted electrons are fast and they leave in opposite directions. The fast ion impact at large impact parameters is the ideal collision process to reveal initial-state correlation. Its duration can be shorter than one attosecond, and the momentum as well as energy transfer in this process are like in photoionization. Furthermore, from the nuclear momentum exchange one can deduce the nuclear impact parameter (classical nuclear trajectories) and thus obtain information on the spatial and the timing coordinates of the fragmentation process.

Also a new attosecond pump-and-probe technique is provided by ion–molecule collision processes. Let's consider a non-linear molecule with three atoms: A, B and C. In case of known nuclear trajectories the impacting ion could react first with atom A (pumping) and some attosecond later with atom B or C (probing). The ion trajectory with respect to the molecular system can be deduced from the final-state momentum pattern, if the momenta of all ionic fragments are measured. For very fast ion impact the electronic motion is slow compared to the projectile motion and thus one could explore how initial-state correlation entanglement influences the fragmentation process in atom B or C. For relativistic ion impact in such a triangle-shaped molecule the ion flight time from A to C can be shorter than the information transport inside the molecule with speed of light.

2. Experiment: COLTRIMS – the dynamics microscope

The principle of the method, namely measuring the trajectory of all fragments, i.e. the momentum of the emitted charged particles from an atomic or molecular fragmentation process is as simple as determining the trajectory of a thrown stone. From knowing the positions, from where the stone was slung and where it hits the target, as well as measuring its time-of-flight, the trajectory of the stone and thus its initial velocity vector can precisely be determined. Furthermore, in order to achieve good precision we have to know whether the person, who throws the stone, was at rest in the frame of observation or we have to know the relative velocity this person was moving with. Thus to obtain optimal momentum resolution for the exploding fragments one has to bring the fragmenting object to a complete rest in the frame of measurement before the reaction occurs, i.e. if the object is a gas atom or molecule, one has to cool it down to sub-milli Kelvin temperatures.

In Fig. 1 the scheme of the COLTRIMS reaction microscope is presented. In a well designed electric field configuration (static or pulsed with a superimposed magnetic field [4–6]) the positively and negatively charged fragments are projected (typically with 4π solid angle) on two position-sensitive detectors. Measuring the impact position on the detector (with a resolution of typically less than 0.1 mm) and the time-of-flight of the fragment (TOF) from the moment of fragmentation until hitting the detector, the particle trajectory, i.e. the particle momentum after fragmentation can be determined. To improve the momentum resolution, electrostatic lenses can be incorporated into the projection system, thus the influence of the unknown size of the target region, from where the fragments originate, can completely be eliminated [4–6]. Magnetic fields can be applied in order to detect fragments of higher energy. Pulsed electric fields can be used to improve the multi-coincidence efficiency. If particle detectors based on fast delay-line position read-out are used, multi-hit detection is possible. Even two or more particles hitting the detector at the ''same'' moment ($\Delta t < 1$ ns) can simultaneously be detected. The number of detected multi-hits is practically only limited by the electronics needed to store in event mode all information. In the future even up to 100 particles per microsecond might be detectable if fast transient recorder units with channel resolution of about 0,1 ns become available. Thus the COLTRIMS method is indeed powerful like an advanced bubble chamber system or even comparable with modern TPC (time projection chamber) systems used in high energy physics. Furthermore, the rate of
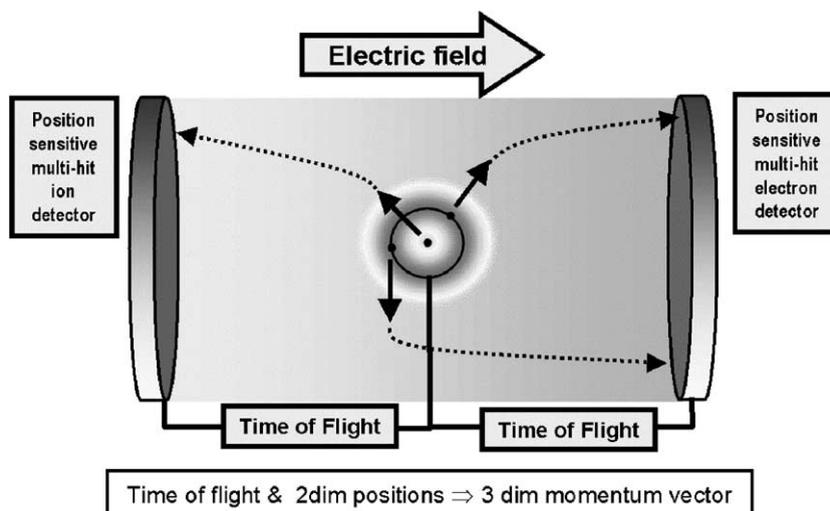

Fig. 1. Scheme of the COLTRIMS imaging system [5].

detected fragmentation processes per second can exceed several 100 kHz.

In numerous benchmark experiments [4–6] quasi snapshots (duration as short as one attosecond) of the correlated dynamics between electrons and nuclei had been made for atomic and molecular objects. In the following a few selected examples will be presented which explore the dynamical correlations in the long-range contributions of ground state wave functions. The data presented below were measured (a) for the transfer ionization process in fast proton on He collisions at the 2.5 MV Van-de-Graaff-accelerator at the Institut für Kernphysik der Universität Frankfurt, and (b) for the photoionization of Ne dimers at BESSYII in Berlin.

3. Data and discussion

3.1. Highly correlated non-s^2 contributions in He ground state

The correlated tunneling transfer ionization (TuTI) channel in fast 4-body processes $p + He \Rightarrow H^0 + He^{2+} + e$ is utilized [7,8] to probe the highly correlated asymptotic parts of the He ground state momentum space wave function. In this reaction, predominantly at large nuclear impact parameters, one electron in the He ground state is captured by the proton by tunneling through the two-center barrier when the electron and proton velocity vectors resonantly match (a Brinkman–Kramers type capture). Due to the coincidence condition only those events can be viewed where one electron is captured, while events from direct single and double ionization characterized by many orders of magnitude larger cross sections can uniquely be distinguished and separated. The coincidence requirement allows watching the long range part of the wave function like viewing the solar corona in a solar eclipse. The TuTI process competes with other transfer ionization (TI) channels [7–15]. Each channel can be identified by a characteristic pattern in the H^0 and He^{2+} final-state momentum space [8,9]. This is achieved by measuring, in coincidence, at least five final-state momentum components (three of

the He^{2+} recoil momentum and two of the H transverse momentum or recently three of the He^{2+} recoil momentum and three of the emitted electron). An extremely high resolution (better than 10^{-5} of the projectile momentum, equivalent to 100 μeV in recoil-ion energy) is required. A very small fraction of the He ground state occupation probability is projected onto the final continuum state due to initial-state correlation by this electron capture reaction in which one electron is picked up by a fast proton and the second electron is left in the continuum in a first order process. The correlation can be seen directly in the momentum distribution of the continuum electron rather than having to be deduced indirectly from measured cross section ratios. The momentum vectors of the two electrons and the nucleus in the initial state are determined in a very sudden fragmentation process which takes a nearly unperturbed snapshot of the highly correlated three-particle motion in the He ground state. We show that this fragmentation process proceeds most likely via the non-s^2 components of the He ground state. These components make up only a very small fraction of the He ground-state and are, therefore, difficult to probe with standard techniques such as spectroscopy.

To reveal the characteristic influence of the non-s^2 components on the final-state momentum distributions, the measured data must be plotted in different presentations:

(a) Plotting the ratio of TI to single capture cross-sections differential in the H^0-scattering angle, a puzzling peak is observed at about 0.65 mrad [8,16]. Measuring the longitudinal energy loss of the projectile with high precision we showed [7–9] that this peak is due to the TuTI process. Obviously at large impact parameters TuTI capture proceeds more and more via the non-s^2 components. According to MCI calculations [8,9] the relative importance of higher angular momentum components increases towards larger distance from the nucleus, and due to their "off-shell" nature their momenta increase with the angular momentum in contradiction to HF predictions. Capture from these

non-s^2 contributions will thus lead with increasing projectile velocity to higher shake-off probabilities (even up to 100%). The observed high shake-off probability (larger than 25%) at large impact parameters (i.e. small H^0 deflection angle) is in perfect agreement with these predictions.

(b) Plotting the fully differential cross sections projected on the projectile scattering plane, i.e. the measured momentum distributions of the shake-off electrons, more surprising features are observed. These features are quite different from those expected for s^2 shake-off electrons (i.e., isotropic emission and peaking at zero energy) [7–9,11]:

   (i) The captured electron (called electron 1), the He^{2+} ion and shake-off electron (electron 2) always share comparable momenta. None of these particles in the final-state Lab system shows a momentum distribution peaking at zero. For TuTI process the angular emission pattern of electron 2 varies strongly with the projectile deflection angle and shows characteristic structures.

   (ii) The momentum vector distribution of the He^{2+} ion and that of electron 2 show a dipole emission pattern in the azimuthal direction (in the plane perpendicular to the H^0-scattering plane).

   (iii) Electron 2 is predominantly emitted into the backward direction and always opposite to the scattered H^0.

Very recent calculations for the TuTI process [17,18] prove that the TuTI process is indeed a shake-off from the non-s^2 components. This calculation is a first-order theory and all TuTI contributions are due to initial-state correlation. The initial-state wave function is obtained in a state-of-art MCI calculation. In Fig. 2 the final-state polar angular correlation of the shake-off electron is shown.

With decreasing H^0-scattering angle (increasing impact parameter) only one amplitude (proton interacts with the captured electron) remains. The observed agreement between data (step function) and theory proves that indeed at large dis-
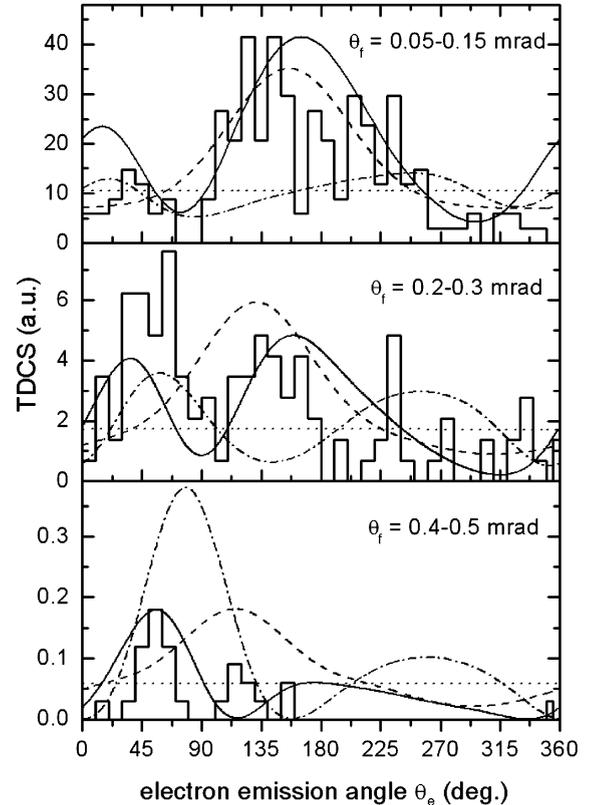

Fig. 2. Polar angular correlation of the free electron shown for different H^0-scattering angles. Several theoretical calculations are shown. Dotted line: for pure s^2 shake-off a fully isotropic emission is predicted in clear contradiction to the data. Solid line: in this first order theory the impacting proton interacts only with one electron, either the captured electron 1 or with the emitted electron 2. The other electron participates in the TuTI process only by initial-state correlation. Thus these two amplitudes do interfere. Including the non-s^2 components the measured data are in excellent agreement with theory (solid line). In particular, the strong H^0-scattering dependence (impact parameter dependence) is well reproduced for $\theta = 0.05$, 0.25 and 0.45 mrad. Projectile energy is 630 keV. Dashed-dotted and dashed lines: also shown are calculated probabilities for cases where the proton interacts only with the captured electron 1 or only with the continuum electron 2 (no interference between these amplitudes). It can be seen that at larger scattering angle both amplitudes are of similar strength and their interference has a significant influence on the TuTI distributions, in nice agreement with the data.

tance from the nucleus the TI process proceeds only via capture of fast non-s^2 electrons, and the electron 2 is emitted due to shake-off from the highly correlated initial state. In Fig. 3 the azi-

muthal dependence is shown. Also here the data are in accordance with the theory showing strong polarization of the non-s^2 contributions.

As pointed out in [7,8], the TuTI process reveals indeed the virtually excited non-local part of the He ground-state wave function, and it displays in a direct way the highly correlated dynamics in the He ground state [17]. The here presented good agreement in the e–e angular correlation between data and the first order theory gives the impression that present quantum theory can perfectly describe the correlated dynamics of the non-s^2 contributions. However, this is not correct. This approach can only describe the angular momentum correlation part, but not the momentum distribution between all three particles. For the angular correlation distributions (see Figs. 2 and 3) the absolute values of the momenta are not so critical. Since the nucleus does not contribute to the internal angular momentum, the angular correlation is still a two-electron effect and thus the angular correlation does not depend on the nuclear mass and the nuclear motion. To predict, however, the absolute values of the correlated three particle momenta and to explain that the final-state momenta of all three particles are of comparable size, the nuclear momentum plays a crucial role. So far no theoretical approach was able to predict reasonably well the measured final-state three-particle momentum distributions, i.e. to predict the three particle correlated entangled momenta. It is to notice that the TuTI process at a given projectile

velocity and H^0 transverse momentum is directly related to a fixed virtual excitation energy and thus to a given QED self energy of the electron pair system. Since we can measure the absolute probability for such a given virtual excitation energy and even measure the momenta of that correlated e–e pair, we can reveal by the TuTI process QED effects highly differential and with a precision orders of magnitude higher than done by highest resolution Lamb shift measurements.

3.2. Long range correlation in Ne dimers

Recently Cederbaum et al. [19] predicted a new decay channel of excited states in molecules, the so-called interatomic Coulombic decay. In this process one atom in a diatomic molecule is ionized by photoionization. The photon energy is specifically chosen with respect to the atomic energy levels, that intra-atomic Auger decay in the single ionized atom is energetically not allowed. However, the molecular neutral partner atom can undergo an Auger decay, if the excitation energy of the ionic partner is fast enough transferred to the neutral partner, before the excited ionic partner can decay by photon emission. Such a molecule with the ideal energy level structure is the Ne dimer. Furthermore, the interatomic distance in Ne dimer is 3.2 a.u. This is of particular interest, since here the long range van der Waals interaction is crucial for this energy transfer (long range correlations). If the transfer time, i.e. the decay time,
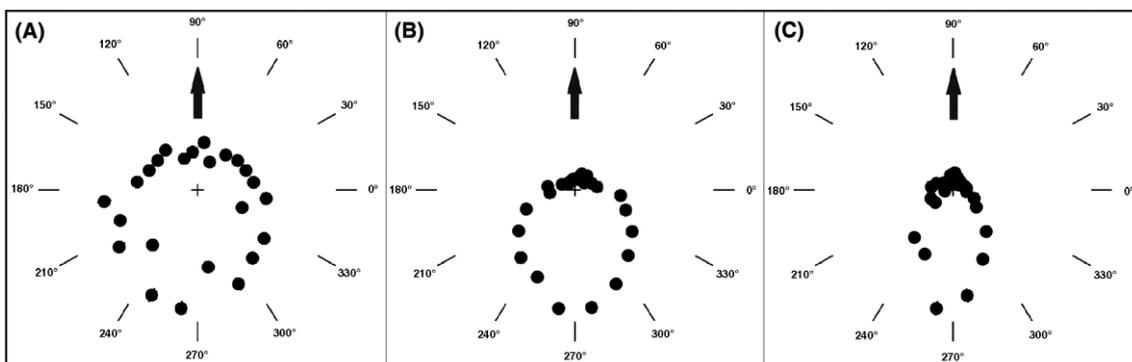

Fig. 3. Azimuthal angular correlation of the free electron for different H^0-scattering angles (A = 0.1 mrad/B = 0.25 mrad/ C = 0.55 mrad). Proton energy is 630 keV. Arrow indicates direction of the scattered H^0 ejectile.

could be measured, important information on the long range interaction via van der Waals forces can be obtained.

COLTRIMS provides the experimental tools to investigate this long range energy transfer process in complete detail [20]. In this "interatomic Coulombic decay" two electrons (the prompt photoelectron and the delayed Auger electron) together with the two Coulomb exploding Ne^{1+} fragments are emitted into the final state. The kinetic energy of the photoelectron yields the information on the electron initial state, the energies of the emitted ionic fragments (kinetic energy release, KER) provide the information on the internuclear distance at the moment of the Auger decay. Thus from the internuclear distance the time-delay after the photon absorption can be deduced. From the Auger electron energy distribution as a function of KER the time for energy transfer from the ionic partner to the neutral partner can be determined with the help of theory.

Using a specially designed COLTRIMS system [21] the two electrons and the two ionic fragments were measured in a three to four particle coincidence. In Fig. 4 the electron energy distribution is shown as a function of KER of the two ionic fragments. From comparison the experimental
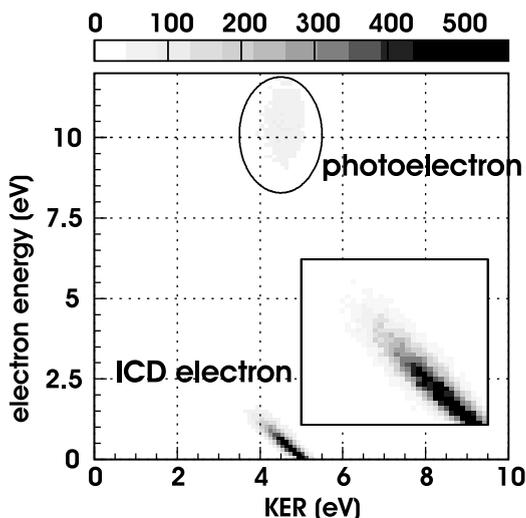
Fig. 4. Kinetic energy release (KER) of the ionic neon fragments in relation to the measured energy of one detected electron.

and theoretical energy distribution of the Auger decay channel [22], an inter-dimer transfer time of the excitation energy due to van der Waals forces of about 10 fs is derived. This transfer time is unexpectedly short. It shows that the dynamics in the long range tail of the wave function due to virtual photon processes or kinematics of virtually excited off-shell electrons is orders of magnitudes faster than that of the tunneling mechanism.

4. Conclusion and outlook

This paper reviews the talk given by HSB as keynote speaker at this Debrecen conference. It was the intention of the speaker to point out in a provocative manner where the basic challenges of many-particle dynamics in atomic and molecular physics may be encountered. Progress can only come from recognizing serious discrepancies between experiment and theory and not by celebrating reasonable agreement with data and unsufficient theoretical models. The coauthors are not responsible for these statements, however, they are added to the author list since they have made substantial contributions to experiment and theory. The provocative statements are speculative, but the data and theory presented here are absolutely reliable results. Some of the coauthors contributed in numerous discussions to the questions raised here, in trying to point down the puzzles of the unknown secrets of correlated many-particle dynamics presented here. Therefore, any reader of this paper, who likes to attack the statements given here, he should write to HSB and blame him.

The presented data and theory show that new imaging techniques allow to reveal the dynamics of the non-local part of the He ground state wave function. Interesting properties of long range van der Waal forces with virtual photon exchange can be experimentally explored. Good agreement with present quantum theory is obtained where two-particle interaction dominates (like angular correlation), but theory fails, where all three particles play equal roles (like in linear momentum exchange). Nevertheless the motivation for

expressing these statements here is to push atomic and molecular physicists in order to question, whether our stationary tools are appropriate to solve the dynamics of the many-particle problem. Since in a bound many-particle system momentum and angular momentum are conserved at any time even for the non-local regime, there must be an instantaneous entanglement between all particles. Instantaneous means, virtual photons contribute to momentum and angular momentum conservation, beyond the classical force concept.

This instantaneous interaction between the particles (or knowing from each other independent on the particle position) is the most beautiful feature of a quantum mechanical system. When the system gets more complex, i.e. is made of more and more particles like in biomolecules, these systems become entangled over large distances and can probably exchange information over 1000 a.u. and more. Therefore it might be not surprising that biological transition rates are often 10^9 times larger than those of "classical" dynamical systems, since the dynamics in such systems is not just statistical. The structure of the molecules alone cannot be the reason for these miracles; entangled many-particle dynamics must play an important role in exchanging and storing that information. Also when forming chemical molecules by van der Waals forces the dynamical entanglement between the atoms is crucial and must be ensured at any moment. To make progress in exploring the dynamics of bound many-particle systems it is crucial to develop also a language between experimentalists and theorists for the physics of the non-local electronic motion. New experimental techniques have opened new observation windows into the world of dynamical correlation. We hope that this paper may help to raise more questions on the dynamics of bound many-particle systems and that theory finds new ways to solve the secrets of entangled correlated motion and guides experimental physics which direction to go.

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