Electron Correlations Observed through Intensity Interferometry

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Intensity interferometry was applied to study electron correlations in doubly ionizing ion-atom collisions. In this method, the probability to find two electrons emitted in the same double ionization event with a certain momentum difference is compared to the corresponding probability for two uncorrelated electrons from two independent events. The ratio of both probabilities, the so-called correlation function, is found to sensitively reveal electron correlation effects, but it is rather insensitive to the collision dynamics.

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One of the basic problems underlying almost all research efforts in physics is to understand how a system of particles develops in space and time under the influence of the mutual interactions within that system. Here a particularly interesting aspect is the question to what extent the interactions lead to correlation effects like, e.g., collective modes of motion, interferences, or an interdependence of the state of motion between several particles of the system. Such correlation effects are well known and still extensively studied in various areas of physics, such as giant resonances in nuclear physics [1] or superconductivity in solid state physics [2].

For time-dependent atomic systems it is rather difficult to observe clean signatures of electron correlations. Significant research efforts have been invested for their identification in many-electron transitions induced by photon or charged particle impact [3]. These studies were frequently hampered by two requirements which work against each other. On one hand, the transition probabilities for many-electron processes usually decrease rapidly with increasing number of active electrons. If one wants to measure multiply differential cross sections with sufficient accuracy, one is therefore forced to use highly charged projectile ions with relatively small energies, where the cross sections maximize. On the other hand, in order to identify correlation effects which result from the mutual interaction between the electrons, it is important to keep the interaction with and between other particles of the collision system as small as possible. This latter requirement favors projectiles with small or no electric charge at relatively large energies.

In this Letter we introduce a new method of analyzing electron correlations in time-dependent atomic systems. It is based on kinematically complete experiments on double ionization in ion-atom collisions where the complete two-electron final continuum state in momentum space is recorded. Intensity interferometry is applied, which originally has been invented in astrophysics to determine the sizes of distant stars [4] and more recently was also used to analyze effects due to the symmetry of the many-particle wave function in nuclear systems [5] and in two-dimensional electron gases in semiconductor devices [6]. Here we demonstrate that for ion-atom collisions intensity interferometry reveals electron correlations very sensitively, but at the same time it is remarkably insensitive to the kinematics and dynamics of the collision. It thus resolves the above-mentioned problems and makes it possible to analyze electron correlations in a very clean manner even for highly charged projectile ions.

The experimental techniques have been reported previously [7] and only the salient points are repeated here. Two independent experiments were performed. In the first, a 3.6 MeV/amu Au⁵³⁺ beam from the UNILAC at GSI Darmstadt was used, in the second a 100 MeV/amu C^{6+} beam was produced at GANIL in Caen. The projectiles collided with very cold (< 1 K) Ne and He gas target atoms from a supersonic jet. The recoiling target ions were extracted by a weak electric field and their charge state and complete momentum vector were determined using position sensitive detectors and time of flight techniques. With the same field the ionized electrons were extracted in the opposite direction. A uniform magnetic field of 20 G confined the transverse electron motion so that all electrons with a transverse momentum of less than 3.5 a.u. were guided onto the detector. Up to three electrons from the same collision were detected simultaneously with a single detector using a multihit technique. For each electron the complete momentum vector was obtained. The projectiles which did not change charge state in the collision were measured in coincidence with the doubly charged recoil ions and both ionized electrons. The multihit dead time, i.e., the minimum time delay between two electrons hitting the detector required to identify them as separate particles, was 10 nsec.

Contrary to what the name may suggest, intensity interferometry is not related to conventional interference phenomena due to well defined phase relations between various waves. Rather, it is based on analyzing the probability of finding two ionized electrons with a specific momentum difference vector relative to each other. These spectra are generated for two cases: In the first case, the momentum difference $\Delta p = |\mathbf{p}_1 - \mathbf{p}_2|$ is taken from two electrons emitted in the same collision and measured in coincidence. This yields an intensity distribution as a function Δp which we call I_{cor} . In the second case exactly the same set of double ionization events as in the first case is used. However, this time one of the two ionized electrons is randomly picked from the *n*th and one electron from the (n + 1)st recorded double ionization event (event mixing). The momentum difference is then calculated for these completely independent electrons emitted in two different, completely unrelated collisions. This leads to a second intensity distribution as a function of Δp , for entirely unrelated electrons, which we call I_{unc} .

In Fig. 1 the spectra for $I_{\rm cor}$ and $I_{\rm unc}$ are shown for double ionization in 100 MeV/amu C⁶⁺ + He collisions as closed and open symbols, respectively. Some differences between these spectra are apparent: at small Δp $I_{\rm cor}$ lies systematically below $I_{\rm unc}$, for Δp between 1.5 and 3 a.u. it is above $I_{\rm unc}$ before it drops again below $I_{\rm unc}$ for $\Delta p > 3$ a.u. If the two electrons emitted in the same collision would be completely independent, then $I_{\rm cor}$ and $I_{\rm unc}$ should be identical for all Δp . The discrepancies between these spectra are thus a measure for the interdependence between the two electrons ionized in the same collision. We therefore define the correlation function $R = I_{\rm cor}/I_{\rm unc} - 1$. This notation is chosen so that R = 0 refers to completely independent electrons, R > 0to a correlation (i.e., the electrons favor the corresponding Δp), and R < 0 to an anticorrelation (i.e., the electrons avoid the corresponding Δp).

In Fig. 2 *R* is shown for 3.6 MeV/amu Au⁵³⁺ colliding with He and Ne target atoms (closed circles and crosses, respectively) and for 100 MeV/amu C⁶⁺ + He collisions (open circles). For the sake of clarity we show error bars only for the C⁶⁺ projectile data. For the other two collision systems the error bars are generally smaller. Extensive numerical tests have been performed to ensure that the shape of *R* is not artificially influenced by the limitations in the experimental momentum acceptance of the electrons (see experimental description). For all three collision systems pronounced structures are observed. The very negative values near $\Delta p = 0$ show that the emission of two electrons with equal momentum vectors is strongly suppressed. Likewise, the positive maximum





FIG. 1. Momentum difference spectra for 100 MeV/amu C^{6+} + He collisions. The closed symbols show coincidence data between the two electrons ionized in the same collision. The open symbols show data for two independent electrons emitted in different collisions and not measured in coincidence with each other.

FIG. 2. Correlation functions $R = I_{cor}/I_{unc} - 1$, where I_{cor} and I_{unc} are the momentum difference spectra as shown in Fig. 1, for the collision systems indicated in the legend. The dashed curve shows a calculation of R only based on the correlated initial target wave function. The full curve includes a correction for the Coulomb repulsion in the continuum by multiplying by a Gamow factor.

around $\Delta p = 2$ a.u. means that these intermediate momentum differences are favored. In any case the data clearly indicate that the behavior of one electron is significantly influenced by the other electron. In this sense we ascertain that the correlation function reveals clear signatures of electron correlations.

A second important and very surprising feature which can be seen in Fig. 2 is that the data qualitatively look very similar for all three collision systems. This is quite remarkable considering that the projectile velocities v_p and charge states Q_p differ by a factor of 5 and 9, respectively. For the perturbation Q_p/v_p this even corresponds to a difference of a factor of 45. It is well known that the collision dynamics depend sensitively on the perturbation [8] leading to significant differences in the spectra of various quantities, e.g., the momentum distributions of the ionized electrons and the recoil ions. In particular, for a perturbation as small as for 100 MeV/amu C⁶⁺ ($Q_p/v_p = 0.1$), the postcollision interaction (PCI) between the outgoing projectile and the ionized electrons is rather insignificant. For a perturbation as large as for 3.6 MeV/amu Au $^{53+}$ $(Q_p/v_p = 4.5)$, in contrast, the PCI has a dominating effect on the momentum distributions [9]. Furthermore, the reaction mechanism leading to double ionization is known to sensitively depend on the perturbation [3,10]. For very small perturbations, double ionization proceeds predominantly through processes involving a single interaction of the projectile with only one electron. The second electron is ionized through a correlation with the first electron. For large perturbations the dominant double ionization mechanism involves two independent interactions of the projectile with both electrons.

To summarize, the similarity in the data of Fig. 2 for very different perturbations clearly shows that the correlation function is surprisingly insensitive to the collision dynamics and to the specific reaction mechanisms that predominantly contribute to double ionization at different perturbations. At the same time, the pronounced structures demonstrate that R sensitively displays the correlation between two electrons. Thus, the correlation function represents a quantity which should be ideally suited to study electron correlations in a dynamical, timedependent situation, being essentially free of complications due to kinematic and dynamic effects of the two-center potential generated by the projectile and target nuclei, like, for example, the PCI. This is very important since at present it appears to be a virtually impossible task to theoretically describe both the dynamics of the two-center potential and electron correlations simultaneously with sufficient accuracy. For the correlation function, however, it may suffice to describe the two-center potential dynamics with a relatively simple model. The major efforts could then be invested in a sophisticated description of the electron-electron interaction both in the initial bound state and in the final continuum state.

In the following we discuss those factors which influence (or which one might expect to influence) the shape of the correlation function. First, from measurements of the correlation functions for photons emitted from distant stars [4], for particles produced in nuclear collisions [5], and for electrons in semiconductor devices [6] it is well known that the symmetry of the manyparticle wave function strongly shapes R if the particles are emitted from an incoherent source: Indistinguishable particles with a symmetric space wave function have a particularly large probability to have similar momentum vectors, which leads to R = 1 at $\Delta p = 0$. Likewise, an antisymmetric wave function leads to R = -1at $\Delta p = 0$. Second, the Coulomb repulsion between the two ionized electrons strongly suppresses identical momentum vectors for both electrons leading to R < 0 near $\Delta p = 0$. Third, and not established up to now, a correlation between the two electrons in the initial target state could potentially produce structures in the correlation function. Fourth, the kinematics and collision dynamics could conceivably influence the shape of R as well. For example, due to the PCI the two ionized electrons could be "pulled" to the same side of the target nucleus as where it is passed by the projectile. However, this factor is readily ruled out by the similarity of the present results for very different collision systems.

The strong drop of R with decreasing Δp below 1.5 a.u. could be due to either the final state Coulomb interaction between the two electrons or to an antisymmetric two-electron wave function in momentum space. However, for He it is clear that the electrons are in a symmetric two-electron space state (ionization processes involving spin flips are negligible). Therefore we are led to conclude that at least for He the negative values of Rfor small Δp are mainly due to the Coulomb repulsion between the electrons. For a Ne target, where both singlet and triplet states can be produced, statistics would favor an antisymmetric space state with a ratio of 3:1. The distinct similarity of the He and Ne results therefore suggests that the correlation function is not very sensitive to symmetry properties.

In order to analyze the role of an initial state electron correlation in R, we calculated the probabilities of two electrons in the He ground state to have a specific momentum difference Δp . Again, this was done for two electrons bound to the same He atom and for two completely independent electrons bound to two different He atoms. For two electrons bound to the same atom, this probability can be calculated according to

$$[dP/d(\Delta p)]_{\rm cor} = \int |\phi(\mathbf{p}_1, \mathbf{p}_1 + \Delta \mathbf{p})|^2 d\mathbf{p}_1.$$
(1)

For two independent electrons bound to different atoms the corresponding probability is given by

$$[dP/d(\Delta p)]_{\rm unc} = \int \left[\int |\phi(\mathbf{p}_1, \mathbf{p}_2)|^2 d\mathbf{p}_2 \int |\phi(\mathbf{p}_1 + \Delta \mathbf{p}, \mathbf{p}_2)|^2 d\mathbf{p}_2 \right] d\mathbf{p}_1.$$
(2)

For the two-electron momentum wave function $\phi(\mathbf{p}_1, \mathbf{p}_2)$ of the He ground state we used the 16-term correlated wave function given by Regier and Thakkar [11]. In accordance with the definition of the experimental correlation function we define $R_i = [dP/d(\Delta p)]_{cor}/[dP/d(\Delta p)]_{unc} - 1$. The calculated $R_i(\Delta p)$ is shown in Fig. 2 as the dashed curve. Indeed, some influence of the initial state on R can be seen: According to this calculation, the initial state alone would lead to a positive correlation for $\Delta p < 2.2$ a.u. and to a negative correlation for $\Delta p > 2.2$ a.u.

With the suppression of small Δp by the Coulomb repulsion in the continuum and the calculated R_i it may now be possible to interpret the maximum in the experimental correlation function as due to a combination of both factors. In order to test this hypothesis we incorporated the Coulomb repulsion in the calculation in an approximate manner. The calculated $dP/d(\Delta p)_{cor}$ were multiplied by the Gamow factor, which is given by $G(\Delta p) = a\pi/\{\Delta p[\exp(a\pi/\Delta p) - 1]\}$. For a point source, the parameter *a* would be equal to 2. However, for an extended source it is known that the slope of the Gamow factor is significantly weaker than for a = 2 [12]. In our calculation we used a = 0.5. In order to conserve flux, after multiplying by the Gamow factor, $dP/d(\Delta p)_{cor}$ was renormalized to unity.

The calculated R_i corrected for the Coulomb repulsion in the continuum is shown in Fig. 2 as the full curve. Some qualitative agreement with the data can be seen. In particular the sharp decrease of R toward $\Delta p = 0$ and the maximum at about $\Delta p = 2$ a.u. is reproduced by the calculation. Quantitative discrepancies are probably mainly due to the simplified description of the Coulomb repulsion in the continuum. As mentioned above, the Gamow factor in the form given is valid only for a point source. A more realistic description of the Coulomb repulsion would therefore also have to incorporate the position distribution of the electrons in the initial state. Strictly speaking, it is thus not even possible to separate the Coulomb repulsion from the initial state.

For Ne a sophisticated correlated two-electron wave function is not available to us. However, one may not necessarily expect significant differences in the shape of R to a He target. Since the double ionization energies for He and Ne are quite similar and the nodal structure of a 2pstate is the same as for a 1s state, the momentum difference distribution could be fairly similar for both cases.

In summary, we have measured correlation functions for double ionization in kinematically complete experiments by analyzing the momentum difference spectra of the two ionized electrons. The data sensitively reveal correlation effects while they appear to be very insensitive to the collision kinematics and the dynamics of the two-center potential. Therefore, the correlation function represents a very appealing quantity for a theoretical analysis of correlation effects because it may be possible to treat the difficult two-center potential problem in a simplified manner without significant loss of accuracy in the description of electron correlations. In future experiments the correlation function will be determined with sufficient statistics to investigate quantitative differences between various targets, which in the present data are only indicated, but cannot be confirmed within the error bars. By comparing to theoretical calculations, it should then be possible to obtain information about correlated many-particle bound states.

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