

Projectile Coherence Effects in Simple Atomic Systems

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Recent studies of projectile coherence effects in ion-atom collisions are presented. For intermediate-energy proton collisions an extensive literature provides strong support for the importance of such effects. In this regime coherence effects are now used as a tool to study the few-body dynamics very sensitively. In contrast, for high-energy ion impact the literature is much sparser and here an important role of coherence effects cannot be regarded as being established. In this context, a recent claim that in COLTRIMS experiments the coherence properties are determined only by the target beam is rebutted.

Introduction

Kinematically complete experiments on atomic collisions between charged particles and simple atoms and molecules have been performed for several decades (for reviews see e.g. [1,2]) in order to study the fundamentally important few-body problem (FBP). The essence of the FBP is that the Schrödinger equation is not analytically solvable for more than two mutually interacting particles even when the forces acting within the system are precisely known. Therefore, theory has to resort to numerical modelling efforts and the assumptions entering in these models have to be tested by detailed experimental data. One process that is particularly suitable to study the few-body dynamics is ionization because there the final state involves three unbound particles.

For electron impact a rich literature on kinematically complete experiments on ionization was developed over the last five decades [e.g. 1,3-10] since the pioneering work of Ehrhardt et al. [11]. A qualitative understanding of the fully differential cross sections (FDCS) extracted from such experiments emerged relatively soon. Usually, the FDCS are dominated by two structures known as the binary and recoil peaks. The binary peak is due to a direct collision between the projectile and the electron and, because of momentum conservation, occurs at or near the direction of the momentum transfer \mathbf{q} . The recoil peak also involves a projectile-electron interaction, however, here the electron subsequently backscatters off its parent nucleus so that it appears at or near the direction of $-\mathbf{q}$. At large projectile energies and for light target atoms experimental data could to a large extent be well described by theory even quantitatively [e.g. 12], however, for small projectile energies it took several decades before a (nearly) complete understanding emerged when several non-perturbative approaches were developed [e.g. 12-14].

Studies on ion-impact ionization are significantly more challenging, both from an experimental and theoretical point of view. The difficulties are introduced by the much larger

projectile mass compared to electron impact. This leads to extremely small scattering angles and projectile energy losses (relative to the initial energy), which can be determined from a direct projectile measurement only for light ions at small or intermediate projectile energies [e.g. 15,16]. Only with the advent of cold target recoil-ion momentum spectroscopy (COLTRIMS) [17,18] kinematically complete experiments on ionization by ion impact became feasible [e.g. 2,15,16,19-24]. Theoretically, the large projectile mass means that in non-perturbative approaches an enormous number of angular momenta has to be accounted for to describe the final projectile state. Only relatively recently were non-perturbative models developed [25-27]. However, for large perturbation parameters (projectile charge to speed ratio η) either large discrepancies to experiment were found [25] or the model has not been tested yet by comparison to experiment on a fully differential level [27].

Nevertheless, it was very surprising that significant and qualitative discrepancies were found between experiment and theory even at very small η [19], where good agreement with perturbative and non-perturbative calculations was expected and routinely found in the case of electron impact [e.g. 28,29]. Even more surprising, very good agreement was achieved with a rather simple model, based on the first Born approximation (FBA), in which the nucleus-nucleus (NN) interaction (not included in the FBA) was accounted for retroactively by convoluting the FBA with classical Rutherford scattering [30].

This success led to the hypothesis that the problem with the fully quantum-mechanical models could be that they all assume a fully coherent projectile beam [31], i.e. that the intrinsic momentum spread is zero. This is not a realistic assumption for fast heavy ions and as a result any interference predicted by theory may not be observable experimentally because fast heavy projectiles tend to be incoherent. In contrast, even very fast electrons usually have a much narrower intrinsic momentum spread due to their much larger deBroglie wavelength. A first indication that indeed measured cross section can be significantly affected by the projectile

coherence properties were found in double differential cross sections (DDCS) for ionization of H₂ by 75 keV proton impact [31]. Later, numerous further experimental [e.g. 32-36] and theoretical [e.g. 37-40] studies supported this hypothesis.

In this article some of the most recent studies on the effect of the projectile coherence properties on FDCS are presented. While for intermediate energy proton collisions a large body of data and theoretical analysis lend strong support to the importance of coherence effects, the literature is much sparser for fast and heavy ion impact. In this regime, the hypothesis that the projectile coherence properties could be responsible for large discrepancies to theory is still awaiting ultimate evidence and did not go completely unchallenged. These critical views are also analyzed.

Experimental Set-Up

Kinematically complete experiments were performed on ionization of He and on dissociative capture (DC) from H₂ in collisions with 75 keV protons. Protons were generated with a hot cathode ion source and accelerated by a high voltage platform. The ions are usually generated with a large intrinsic momentum spread, i.e. with a small coherence length. However, in analogy to classical optics, the coherence length increases during the propagation of the projectile wave. Therefore, the coherence length Δr can be manipulated by placing a collimating slit with width a at a distance L from the target and is given by $\Delta r = L\lambda/2a$, where λ is the deBroglie wavelength. The experiments were performed for $a = 150 \mu\text{m}$ and for two distances of 50 cm, corresponding to $\Delta r = 3.5 \text{ a.u.}$, and 6.5 cm. For the smaller distance the local collimation angle at the target is not restricted by the collimating slit and here $\Delta r = 1 \text{ a.u.}$, which is larger than $L\lambda/2a$.

The collimated projectile beam was crossed with a very cold target beam from a supersonic jet at a temperature of approximately 1 – 2 K. The recoiling target ions were extracted by a

weak electric field of about 5 V/cm (40 V/cm) in the experiment on ionization (DC). The recoil ions (He^+ for ionization and the charged molecular fragment for DC) were detected by a two-dimensional position-sensitive multi-channel-plate (MCP) detector. After the collision, the projectiles were charge-state analyzed by a switching magnet. The beam component not charge-changed in the collision (ionization experiment) was decelerated by 70 keV and then energy-analyzed by an electrostatic parallel plate analyzer [41] and detected by a second MCP detector. For the DC experiment a third MCP detector was mounted at the 0° port of the switching magnet to detect the neutralized projectiles. The recoil-ion detector was set in coincidence with the respective projectile detector.

From the position information from each detector two momentum components of the corresponding particle were obtained. The third momentum component was obtained from the time-of-flight information contained in the coincidence time in the case of the recoil ion and from the energy loss in the case of the scattered protons. The momentum of the electrons ejected in ionization and the neutral molecular fragment in DC was determined from momentum conservation. In the case of projectiles neutralized in DC the third momentum component could not be measured directly. However, due to four conservation laws (energy in addition to momentum) only five of the nine momentum components are independent so that the measurement of 3 recoil-ion and 2 projectile components manifests a kinematically complete experiment. However, the resolution was not sufficient to determine the final state of the captured electron.

Results and Discussion

a) Ionization

For the analysis of the FDCS for ionization we use a coordinate system defined by the projectile momenta: the positive x-direction is given by the direction of the transverse component of \mathbf{q} and the z-direction by the initial projectile momentum. This choice of

coordinate system implies that q_x is always positive and q_y is always zero (within the experimental resolution). The FDCS were generated for an energy loss of $\varepsilon = 30$ eV, various fixed recoil-ion momentum components in the x -direction ranging from $p_{\text{rec}x} = 0.2$ to 1.25 a.u., and fixed polar electron emission angles ranging from $\theta_{\text{el}} = 15^\circ$ to 85° (measured relative to the projectile beam axis). The data were then analyzed as a function of the azimuthal electron emission angle ϕ_{el} . Here, $\phi_{\text{el}} = 90^\circ$ is defined by the direction of q_x and $\phi_{\text{el}} = 270^\circ$ by the direction of $-q_x$. At these angles, the binary peak and recoil peak, respectively, are expected in a first-order picture.

In analogy to classical optics the FDCS measured for a coherent beam $d\sigma_{\text{coh}}$ can be expressed as the one measured for an incoherent beam $d\sigma_{\text{inc}}$ multiplied by the interference term I . Therefore, if the coherence properties of the projectiles are indeed determined by the distance of the collimating slit to the target, the interference term can be extracted as the ratio R between the FDCS measured for large L relative to small L . These ratios are shown as a function of ϕ_{el} in Fig. 1 for a subset of the data, where the columns represent data for θ_{el} fixed at (from left to right) 25° , 45° , and 65° , and the rows data for $p_{\text{rec}x}$ fixed at (from top to bottom) 0.2 a.u., 0.7 a.u., and 1.25 a.u. Note that the regions in which no data are shown are kinematically prohibited because there q_x is negative, which is not possible in our coordinate system. The solid curves show a time-dependent calculation [42,43] in which the projectile is described by a wave packet with a width reflecting the coherence length. R was evaluated between FDCS calculated for a width of 3.5 a.u. and 1.0 a.u., respectively.

Theory predicts that for small $p_{\text{rec}x}$ and θ_{el} coherence effects are rather weak. Indeed, this prediction is confirmed by experiment, where R is nearly constant with a value close to 1. In contrast, for increasing $p_{\text{rec}x}$ and θ_{el} increasingly pronounced structures are found in R . Again, this finding is in very good qualitative accord with the experimental data, although there are some quantitative discrepancies. For the other kinematic settings similarly good agreement

was found. These structures show that interference is present in the coherent FDCS. Here, different (non-observable) impact parameters leading to the same scattering angle interfere with each other, which is analogous to single-slit interference in classical optics and to which we refer as single-center interference.

Further theoretical analysis showed that the FDCS are very sensitive to the coherence length and that even at $\Delta r = 3.5$ a.u., which is substantially larger than typical impact parameters, the collision cannot be viewed as fully coherent [44]. This sensitivity makes the good qualitative agreement between theory and experiment the more significant. Overall, the strong departure of R from unity (except for small p_{recx} and θ_{el}) along with the reproduction of the experimental data by theory strongly support the hypothesis that cross sections for ion-atom collisions can be strongly influenced by the projectile coherence properties.

b) Dissociative Capture

In dissociative capture an electron from the target molecule is captured to the projectile and at the same time the residual molecular ion is excited leading to its fragmentation. There are two fragmentation channels which can be distinguished: in the first, the second electron residing with the molecule gets excited to a repulsive electronic state (i.e. the overall electronic process is transfer plus excitation). This channel leads to kinetic energy releases (KER, the sum kinetic energy of both fragments) ranging from approximately 5 to 25 eV. In the second fragmentation channel, the second electron remains in the ground state of the molecular ion, however, the nuclear motion is excited to a vibrational continuum state. This channel is known as ground state dissociation (GSD). Here the KER is significantly smaller (less than 2 eV). Therefore, the two fragmentation channels can be distinguished based on the KER.

From the momentum components of the molecular fragments extracted from the experiment the KER and the molecular orientation at the instant of the collision can be determined. In the following we will focus on GSD, which was selected by setting a condition on $\text{KER} < 2 \text{ eV}$ in the data analysis. Two molecular orientations were analyzed, both of which were perpendicular to the projectile beam axis. The first was also perpendicular to q_x while the second was parallel to q_x . As in the ionization experiment, the FDCS were measured at large and small L for each orientation and the ratios R between the corresponding $d\sigma_{\text{coh}}$ and $d\sigma_{\text{inc}}$ were analyzed [45].

The interference term for molecular two-center interference is given by

$$I_2 = 1 + \cos(\mathbf{p}_{\text{rec}} \cdot \mathbf{D}) = 1 + \cos(\mathbf{q} \cdot \mathbf{D}), \quad (1)$$

where \mathbf{D} is the internuclear separation vector of the molecule. In this expression the dot product $\mathbf{q} \cdot \mathbf{D}$ is constant at 0 for the perpendicular orientation. Therefore, the ratio R for this orientation reflects single-center interference. For the parallel orientation the total interference term results from a combination of single- and two-center interference. Under the assumption that both are independent of each other we extracted the two-center interference term I_2 by dividing the ratio R by the single-center interference term obtained from the data for the perpendicular orientation.

The experimentally determined two-center interference term R_2 is plotted in Fig. 2 as a function of projectile scattering angle θ_p . A pronounced oscillating pattern can clearly be seen. For two reasons these data are particularly significant relative to the question whether the departure from $R = 1$ is a manifestation of projectile coherence effects or merely due to experimental artifacts. First, in contrast to the ionization experiment the measurements for the large and small slit distance were performed simultaneously in the same experiment. This was accomplished by placing the x-slit at a small L and the y-slit at a large L and then

selecting scattering in the x- and y-direction in the data analysis to obtain the incoherent and coherent FDCS, respectively. Thereby, the data for small and large L were taken under otherwise identical experimental conditions, i.e. differences between them cannot be explained by experimental artifacts. Second, the oscillation extrema in R_2 occur at exactly the same θ_p as in the absolute FDCS. This makes it very unlikely that the structures in R_2 are just artificial, which could result for example from inaccuracies in the calibration of θ_p affecting scattering in the x- and y-directions differently.

The two-center interference term of equation (1) predicts maxima where minima are found in the experimental data and vice versa. However, if a phase shift of π is added to the argument of the cosine function, I_2 (solid line in Fig. 2) is in nearly perfect agreement with the data. Such a phase shift was observed previously in electron capture to the dissociative $2p\sigma_u$ state of the projectile in $H_2^+ + He$ collisions [46] and in dissociative ionization of H_2 by electron impact [47]. In [46] this shift was convincingly explained as a consequence of parity conservation: the switch in symmetry in the electronic wave function (from gerade in the ground state of He to ungerade in the final molecular state) has to be compensated by a corresponding switch in symmetry in the projectile wave function. However, the existing literature suggests that there are other factors which also need to be considered to fully understand the phase shift: Schmidt et al studied dissociative transfer excitation in $p + H_2$ collisions [48]. Although they also considered a case where the symmetry of the electronic wave function switches, they did not find any phase shift in the interference term. On the other hand, no switch in symmetry of the electronic wave function is involved neither in the present study nor in the one reported in [47], but yet in both cases a phase shift of π was found. At present, no explanation can be offered as to which factors apart from parity conservation may decide whether or not a phase shift occurs. However, we note that so far a phase shift was only observed in dissociative processes.

c) Coherence in Fast Collisions

For intermediate collision energies the presence of projectile coherence effects is supported by a large body of experimental data [e.g. 31,32,35,36,42,45]. However, for fast ion impact the literature is much sparser [33,34]. The aforementioned discrepancies between theory and experiment in FDCS for ionization in 1.2 GeV $C^{6+} + He$ collisions were blamed on the very small transverse coherence length of the projectiles [31] and indeed data obtained for $p + He$ collisions at the same perturbation and a coherence length larger by orders of magnitude were much better reproduced by theory [33]. On the other hand for somewhat slower proton collisions no significant differences between the FDCS for two different transverse coherence lengths were found [24]. However, there even the smaller coherence length was orders of magnitude larger than for 1.2 GeV C^{6+} impact and larger than the size of the target atom¹. Under these conditions only weak coherence effects are expected [38] and the results of [24] are thus not inconsistent with the interpretation that the data for 1.2 GeV collisions are significantly affected by projectile coherence effects. Furthermore, the comparison between FDCS for ionization and transfer-ionization in 16 MeV $O^{7+} + He$ also provided some strong indications for pronounced coherence effects [34].

Nevertheless, the interpretation that the failure of theory to reproduce measured FDCS for ionization in 1.2 GeV $C^{6+} + He$ collisions is due to coherence effects was challenged by Kouzakov [49]. He argued that in an experiment where only the recoil ions and the ejected electrons are momentum-analyzed the FDCS can only be affected by the coherence properties of the target beam, but not the projectile beam. In the following, we argue that this conclusion is not consistent with what is generally known for coherence and interference effects.

¹ In the original publication the coherence length was calculated incorrectly. The correct value is at least 1.3 a.u. instead of 0.8 a.u.

First, it should be noted that the coherence properties of the collisions system are determined by the spread in the relative momenta of the two collision partners. The collision is coherent if this spread is narrow and incoherent if it is broad. But if the momentum distribution is broad for one of the collision partners, it must be broad for the relative momentum distribution because if an essentially constant value is added to a broad distribution the resulting distribution remains broad. Therefore, for interference structures to be observable (i.e. for the collision system to be coherent) *both* beams must be coherent. This is true regardless of which particle is actually momentum-analyzed in the experiment. To illustrate this important point consider the following scenario: suppose an experiment is performed in which both the projectile and the recoil-ion (and the ejected electrons) are momentum-analyzed and the coherence length of the projectiles is very small. In this scenario the analysis of Kouzakov yields that due to the incoherence of the projectiles (and therefore the collision system) no interference is observable. Then the experiment is repeated, but this time the projectiles are not detected. However, due to the constraints imposed by the conservation laws the experiment is nevertheless kinematically complete, i.e. exactly the same final state is observed in the experiment. In this case the analysis of Kouzakov yields that interference becomes observable. However, not detecting the projectiles obviously has no effect at all on the projectile coherence length, i.e. the collision system is still incoherent and no interference can be observed, contrary to the conclusion of Kouzakov. To put it in casual language: it is not possible to create an interference structure out of nothing simply by not looking at any incoherent collision partner in the experiment.

Finally, we note that this analysis also demonstrates one important difference between incoherence and beam divergence, which constitutes part of the experimental angular resolution. Both are theoretically often treated by the same mathematical approach, namely by convoluting the transition amplitude for the coherent case with the momentum spread (either due to incoherence or experimental resolution) of the collision partners [e.g. 38-40,50].

However, the beam divergence obviously will not have any effect on measured cross sections if the corresponding particle is not even detected, in which case it should not enter in the convolution. In contrast, the coherence properties are completely independent on whether or not the corresponding particles are detected and thus always need to enter in the convolution.

Conclusions

Over the last decade numerous studies have been performed demonstrating that the projectile coherence properties can have a major effect on measured cross sections, not because of imperfections in the experiment, but rather due to the inherent properties of the projectile wavefunction. This important point was overlooked for decades of ion-atom collision theory. The reason for this oversight is probably that FDSC on ionization were initially measured for electron impact, where the coherence properties do not play an important role. When such measurements became possible for ion impact, it seemed reasonable to apply the same theoretical techniques that worked well for electron impact to collisions with ions.

With the importance of coherence effects uncovered the literature on ion-atom collision studies should be revisited for previously unexplained discrepancies between experiment and theory. While it is unlikely that the coherence properties can offer a complete explanation for all of these cases, they may provide at least partial explanations in some cases. Furthermore, coherence effects can be used as a tool to study the few-body dynamics in collisions in greater detail. For example, the dependence of cross sections on the projectile scattering angle tends to be very steep. As a result, an interference structure superimposed on this steep dependence may be very difficult to identify. However, if the cross sections are measured for a coherent and an incoherent beam the interference term can be extracted as the ratio between these cross sections, in which the steep scattering angle dependence is removed. Thereby, a much more accurate analysis of the interference term is possible.

For fast ion impact the role of coherence effects is not completely settled yet. Although the claim that only the coherence properties of the target beam matter if the projectiles are not detected is unsubstantiated, this does not mean that the interpretation that discrepancies between experiment and theory are due to the coherence properties can be regarded as established. Further studies are needed, both theoretical and experimental.

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Figure Captions

Fig. 1: Ratio between fully differential cross sections for ionization of He by 75 keV proton impact measured at large and small distances between the collimating slit and the target as a function of the azimuthal electron ejection angle. The projectile energy loss was fixed at 30 eV. For data within a column θ_{el} was fixed at (from left to right) 25° , 45° , and 65° and for data within a row p_{recx} was fixed at (from top to bottom) 0.2, 0.7, and 1.25 a.u. The solid lines show time-dependent calculations in which the transition amplitude was convoluted with a wave packet describing the projectile with the width representing the transverse coherence length (3.3 and 1.0 a.u., respectively).

Fig. 2: Ratio between fully differential cross sections for dissociative capture in 75 keV p + H₂ collisions measured at large and small distances between the collimating slit and the target

as a function of the projectile scattering angle. The KER was fixed at 0 to 2 eV and the molecular orientation was perpendicular to the projectile beam axis and parallel to the transverse momentum transfer. The solid line represents equation (1) with a phase shift of π added to the argument of the cosine function.

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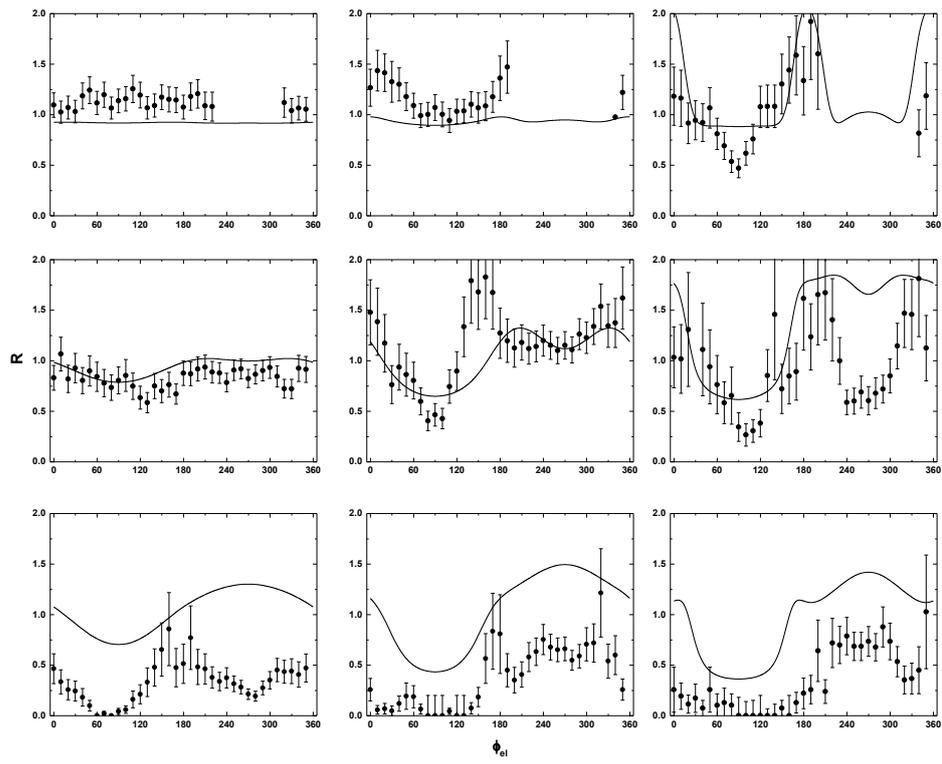


Fig. 1

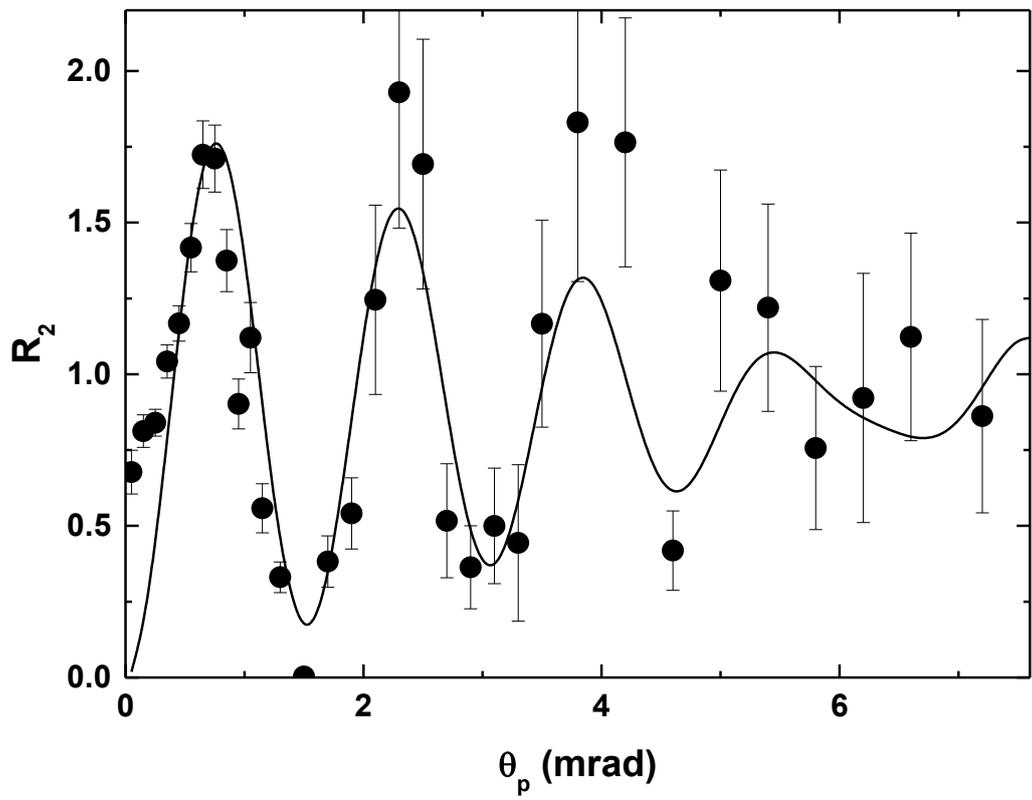


Fig. 2