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Two-center and path interference in dissociative capture in $p + H_2$ collisions

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We have measured and calculated fully differential cross sections (FDCS) for dissociative capture in 75-keV $p + H_2$ collisions. FDCS were analyzed in the kinetic energy release (KER) ranges 0 to 2.1 eV and 4 to 7 eV for two different molecular orientations. In the latter range, dissociation is dominated by electronic excitation to the $2p\pi_u$ state. Here, we observed two-center interference for an orientation in the plane perpendicular to the initial beam axis and parallel to the transverse momentum transfer. The interference pattern is afflicted with a constant phase shift of π. In the range KER = 0 to 2.1 eV, dissociation is dominated by vibrational excitation. Here, we observed structures in the KER dependence, which we interpret as interference between two different paths of the molecular fragments during the dissociation.

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I. INTRODUCTION

One of the basic fragmentation processes of molecules is dissociation in collisions with photons, electrons, or ions caused by accompanying capture from or excitation or ionization of the molecule (e.g., Refs. [1–3]). Such processes were observed already many decades ago. However, the interest in them was renewed when two-center interference effects were found in the spectra of electrons ejected in (initially nondissociative) ionization of H_2 by highly charged ion impact [4]. From first principles, one cannot distinguish from which atomic center of the molecule the incoming projectile wave gets diffracted. Therefore, both contributions have to be added coherently which can give rise to an observable interference pattern. The phase angle in the interference term is given by the scalar product between the momentum vector of the recoiling H_2^+ ion $\mathbf{p}_{\mathbf{rec}}$ and the internuclear separation vector $\mathbf{D}$ of the molecule [5]. In Ref. [4], neither of these quantities could be determined. As a result, the interference structure was heavily "washed out" because the phase angle was not well determined.

Later, two-center interference was studied for ionization of H_2 using cold-target recoil-ion momentum spectroscopy [6,7]. In another experiment, ionization of H_2 by electron impact was studied by measuring the complete momentum vectors of the scattered and the ejected electrons [8]. Since in these experiments $\mathbf{p}_{\mathbf{rec}}$ was either directly measured [6,7] or deduced from the kinematic conservation laws [8], the phase angle was better determined than in Ref. [4]. This resulted in more pronounced interference effects. However, $\mathbf{D}$ was not measured in these experiments and the observed interference effects represented an average over this parameter still leading to a significant washing out of the structure.

In several kinematically complete experiments on dissociative capture or excitation, $\mathbf{D}$ could be experimentally determined by momentum analyzing the molecular fragments [9–11]. In all three cases a very pronounced interference structure was found. However, in two of them [9,10] the interference pattern was afflicted with a π-phase shift compared to what was initially expected based on the interference term given in Ref. [5]. This phase shift was convincingly explained by the symmetries of the molecular states involved in the transition [9]: in both experiments, molecular transitions from the gerade ground state to an ungerade repulsive state were analyzed. This switch in symmetry must be compensated by a corresponding switch in symmetry of the wave function of the atomic collision partner corresponding to a π-phase shift. In Ref. [11] the symmetry of the dissociating state could not be determined; however, the experiment was performed in a range of kinetic energy releases (KER) which should be dominated by ungerade states. Therefore, based on the symmetry argument presented in Ref. [9] a π-phase shift should have been observed in Ref. [11] as well. Instead, the authors reported an interference pattern which was consistent with a zero-phase shift. This was a hint that the phase of the interference term is more complex. Apart from symmetry considerations there may be other aspects which need to be considered in order to determine under what conditions a phase shift occurs.

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Further indications that the molecular symmetry is not the only factor that can lead to a phase shift came from two other kinematically complete experiments measuring fully differential cross sections (FDCS) for dissociation of H_2. In one, dissociative capture in collisions with protons was studied [12] and in the other dissociative ionization by electron impact [13]. In both experiments dissociative molecular states with gerade symmetry were analyzed. Based on the symmetry argument, a phase shift should have thus not been observed, but was actually found. This clearly showed that apart from a switch of the molecular symmetry there are other scenarios which can also lead to a phase shift. So far, conclusive evidence for such an alternative explanation has not been reported yet. However, in a follow-up study to Ref. [12] a hypothetical explanation was suggested [14] (in the following referred to as "our hypothesis"). There, it was also found that the phase shift is actually not constant at π (as in Refs. [9,10]), but rather it varied with projectile scattering angle θ_p between π at small θ_p and zero at large θ_p. The hypothesis is based on the classical analogy of a reflection of a traveling wave from a fixed end which results in a π-phase leap. Quantum mechanically, this corresponds to a reflection of a de Broglie wave from an infinite potential step. The potential curves in the H_2^+ molecule are not exactly stepping up to infinity at sharp locations; however, they are very steep and asymptotically go to infinity at $D = 0$. Thus, they bear some resemblance to an infinite potential step. The authors in Ref. [14] attempted to explain the phase shift varying with θ_p in terms of two different paths leading to dissociation: in one, the fragments immediately depart from each other (direct path) while in the second they first approach each other and then get reflected from the potential wall approaching infinity at $D = 0$ (reflected path). The phase shift is expected to be zero for the direct path and π for the reflected path. Based on classical considerations it was argued that small θ_p should favor the reflected path and large θ_p the direct path, which would explain the observed dependence of the phase shift on θ_p.

While this hypothesis may seem plausible, conclusive evidence is still lacking. Further studies are needed to either further support it or to dismiss it. In the present work we pursued this goal using two approaches: in the first, we analyzed the two-center interference pattern for a kinetic energy release of 5.5 ± 1.5 eV. In this KER range dissociation is dominated by electronic excitation to the $2p\pi_u$ state [15] and vibrational dissociation does not play any role. Therefore, our hypothesis should not hold in this case, and one would expect a constant phase shift of π. In the second approach, we measured multiple differential cross sections for fixed molecular orientations as a function of KER in the range from 0 to 2 eV. The motivation is born out of the notion that if indeed dissociation can proceed through the direct or the reflected path both channels would be indistinguishable. Their contributions would then have to be added coherently. The phase difference between the corresponding amplitudes would depend on the momentum of the molecular fragments and thus on the KER. Therefore, apart from two-center interference, an additional interference, to which we refer as path interference, might be observable in the KER spectra. In our previous work on this topic [12,14] we neither studied KER values larger than 2 eV nor the KER dependence of the cross sections for KER < 2 eV.

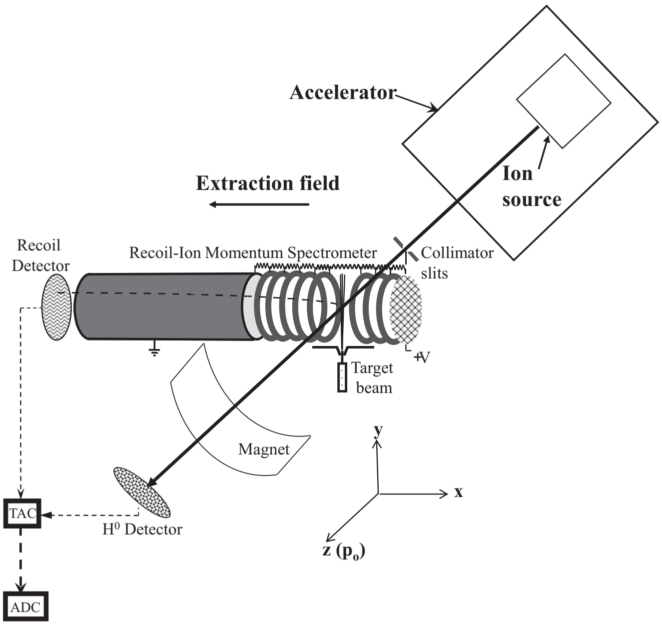

FIG. 1. Schematic sketch of the experimental setup and of the coordinate system in which the momenta of the collision fragments are given.

II. EXPERIMENT

The experiment was performed at the medium-energy ion accelerator of the Missouri University of Science and Technology. A schematic setup is shown in Fig. 1. A proton beam was generated using a hot-cathode ion source, extracted at 5 keV, and further accelerated to an energy of 75 keV using a high-voltage platform. The projectile beam was collimated to a size of about 0.15×0.15 mm^2 and crossed with a very cold ($T \approx 1.5$ K) H_2 beam from a supersonic gas jet. The scattered projectiles neutralized by capture from the target molecule were selected by a switching magnet and detected by a two-dimensional position-sensitive microchannel plate detector (MCP). From the position components x and y the corresponding components of the momentum transfer $\mathbf{q}$ were determined with a resolution of 0.5 atomic units (a.u.) full width at half maximum (FWHM). It mainly results from the beam divergence and the position resolution of the detector. The polar scattering angle is given by $\theta_p = (q_x^2 + q_y^2)^{1/2}/p_o$ and was obtained with a resolution of about 0.2-mrad FWHM. The azimuthal projectile angle is determined by $\varphi_p = \text{atan}(q_y/q_x)$ with a resolution of 3° FWHM. The longitudinal component of the momentum transfer from the projectile to the target q_z is obtained from the relation $q_z = -Q/v_p + v_p/2$ [16]. Here, Q is the Q value of the reaction which depends on the final state of the captured electron. However, the variation of Q/v_p with the final state is small compared to $v_p/2$. Furthermore, the capture goes predominantly to the ground state of the projectile. Therefore, q_z is known with sufficient accuracy (about 0.1 a.u.).

The positively charged fragments from the molecular dissociation were extracted by a weak electric field of 7.8 V/cm and detected by a second two-dimensional position-sensitive MCP. Both detectors were set in coincidence. The two momentum components in the plane of the detector p_y and p_z are obtained from the position information, where y and z are defined by the directions of the target and projectile beams, respectively. In the x direction, pointing along the extraction field, the momentum p_x is determined by the time of flight from the collision region to the detector, which in turn is obtained from the coincidence time. In the y direction, the resolution of about 0.5-a.u. FWHM is mostly due to the temperature of the target beam. In the x- and z directions the target temperature is much smaller. In the z direction the resolution is dominated by the finite size of the reaction volume (i.e., the spatial overlap between the projectile and target beams). It can be directly obtained from the width of the line in the momentum spectrum representing nondissociative capture to the ground state of the projectile and is found to be 0.15-a.u. FWHM. In the x direction, the effects of the size of the reaction volume are minimized by time focusing realized by choosing the Wiley-McLaren geometry of the acceleration and drift regions of the momentum spectrometer [17]. Here, the resolution is estimated to be somewhat smaller than in the z direction (approximately 0.1-a.u. FWHM). The corresponding angular resolutions depend on the KER and on the angles themselves. For the molecular orientation analyzed in this work (see the next section) and $\mathrm{KER} = 0.2$ eV, corresponding to a momentum of the detected fragment of about 3.7 a.u., the resolutions in the polar and azimuthal angles were 2° and 8° FWHM, respectively. With increasing KER, i.e., for most of the KER spectrum which we analyzed, the resolution improves.

III. DATA ANALYSIS

We analyzed FDCS for dissociative capture for two molecular orientations. Both of them are perpendicular to the initial projectile momentum p_o (within $\pm 10^\circ$). One is also perpendicular to the transverse component of $\mathbf{q}$ (q_{tr}) while the second is parallel to q_{tr}. We refer to them as the perpendicular and parallel orientations, respectively. The orientation is obtained from the measured momentum of the charged molecular fragment $\mathbf{p_{fr}}'$ and from the measured momentum transfer. $\mathbf{p_{fr}}'$ is a vector sum of the momentum resulting from the dissociation in the center-of-mass frame of the molecule $\mathbf{p_{fr}}$ and $\mathbf{q}/2$ ($\mathbf{q}$ is equally shared by both fragments). Therefore, in order to obtain the molecular orientation at the instant of the dissociation $\mathbf{p'_{fr}}$ was corrected for $\mathbf{q}/2$ [14]. In the data analysis a fixed molecular orientation along the x axis was selected ($\varphi_{fr} = 0^\circ \pm 10^\circ$). This has the advantage that in this direction we obtained the best momentum resolution, and the orientation can be obtained nearly unaffected by the relatively poor resolution in the y direction. The perpendicular (parallel) orientation is then selected by setting a condition on the azimuthal projectile angle $\varphi_p = 90^\circ \pm 10^\circ$ ($0^\circ \pm 10^\circ$).

The KER, i.e., the sum kinetic energy of both fragments, was calculated as $\mathrm{KER} = \mathbf{p}_{\mathbf{fr}}^{\mathbf{2}}/M$, where M is the mass of one fragment. Two KER regions were analyzed. First, we studied the θ_p dependence of the FDCS in the range KER = 4 to 7 eV to select a region which is dominated by dissociation due to electronic excitation to the $2p\pi_u$ state of the H_2^+ molecule. Second, we analyzed the KER spectrum in the region KER = 0 to 2.1 eV, which is dominated by dissociation due to vibrational excitation.

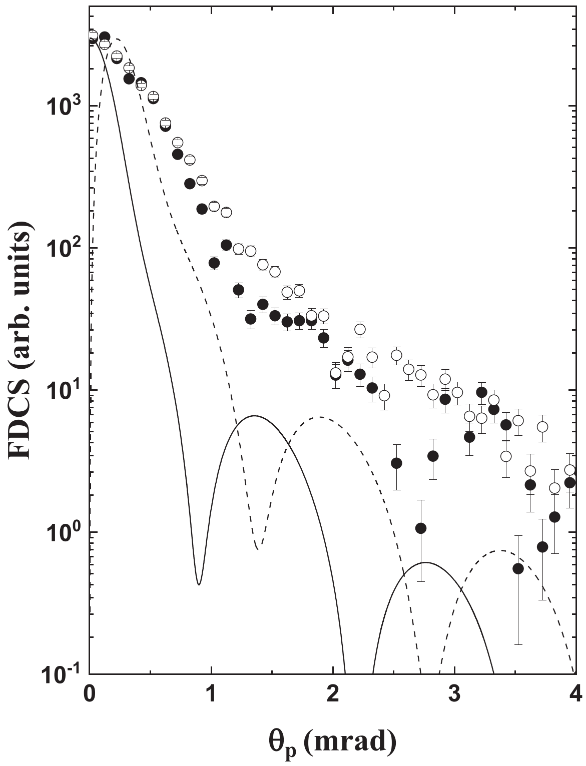

FIG. 2. Fully differential cross sections (FDCS) for dissociative capture for $\mathrm{KER} = 5.5 \pm 1.5$ eV as a function of θ_p. Open symbols: perpendicular orientation; closed symbols: parallel orientation. Curves: distorted wave calculations for dissociation for the parallel orientation from the $2p\pi_u$ state (solid curve) and from the $2p\sigma_u$ state (dashed curve).

IV. RESULTS AND DISCUSSION

A. Projectile scattering-angle dependence for KER = 5.5 eV

In Fig. 2 the FDCS are plotted as a function of θ_p for $\mathrm{KER} = 5.5 \pm 1.5$ eV. The closed symbols represent the data for the parallel orientation and the open symbols represent those for the perpendicular orientation. For the parallel orientation a structure with a minimum at 2.6 mrad and a maximum at 3.2 mrad can be seen. Furthermore, a plateau is observed between 1.3 and 2 mrad, which adumbrates further interference extrema in this region. In contrast, no such structures can be discerned for the perpendicular orientation. This is the expected behavior for two-center molecular interference. The phase angle α is given by the dot product between the recoil-ion momentum $\mathbf{p_{rec}}$ (where in the case of capture $\mathbf{p_{rec}} = \mathbf{q} - \mathbf{v_p}$) and the internuclear separation vector $\mathbf{D}$ (plus possibly a phase shift δ). Therefore, α is constant for the perpendicular orientation and no oscillating interference pattern is expected.

The solid line in Fig. 2 shows our calculation for dissociative capture through excitation to the $2p\pi_u$ state using a distorted wave approach [18,19]. This model includes the interaction between the projectile and the nuclei of the H_2 target. The magnitude was adjusted to the one of the measured

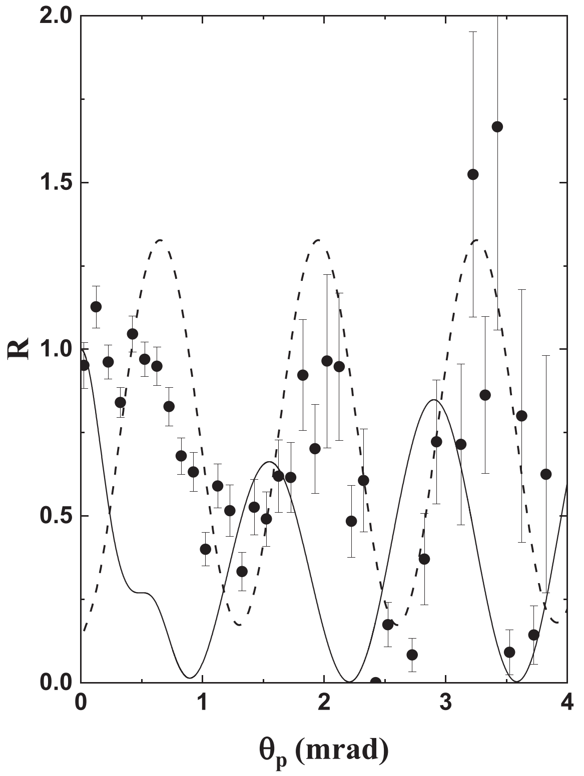

FIG. 3. Ratio between the FDCS of Fig. 1 for the parallel and perpendicular orientations. Dashed curve: two-center molecular interference term from Ref. [5] with a phase shift of π. Solid curve: distorted wave calculations for dissociation from the $2p\pi_u$ state.

FDCS (which could not be normalized on an absolute scale) at 0 mrad. Significant discrepancies between experiment and theory are quite apparent. The calculated cross sections fall off with increasing θ_p much more rapidly than in the data. Furthermore, the position of the extrema in the measured cross sections is not reproduced at all. Rather, the theoretical minima seem to be closer to the experimental maxima and vice versa. For comparison, the dashed curve shows our calculation of the FDCS for excitation to the $2p\sigma_u$ state. Both calculations are phase shifted relative to each other by π, i.e., where one calculation exhibits minima the other shows maxima. Furthermore, the $2p\sigma_u$ is qualitatively in better agreement with the data in that it does not fall off as quickly with θ_p as the $2p\pi_u$ calculation and the position of the extrema is better reproduced. We currently cannot offer an explanation as to why the data are more closely reproduced by the calculation for the $2p\sigma_u$ state.

In the measured cross sections, the oscillating structure is not very pronounced, partly due to the steepness of the θ_p dependence and partly due to the resolution. The effect of the steepness can be minimized by analyzing the ratio R between the FDCS for the parallel and perpendicular orientations. Since for the perpendicular orientation the phase angle is constant, R would represent the interference term if the incoherent part of the FDCS would have the same shape for both orientations. The data of Fig. 2 suggest that for the perpendicular orientation the θ_p dependence is actually slightly flatter. However, the position of the interference extrema is not affected much by this difference. Therefore, in the following analysis, as a good approximation, we use R as if it did represent the interference term.

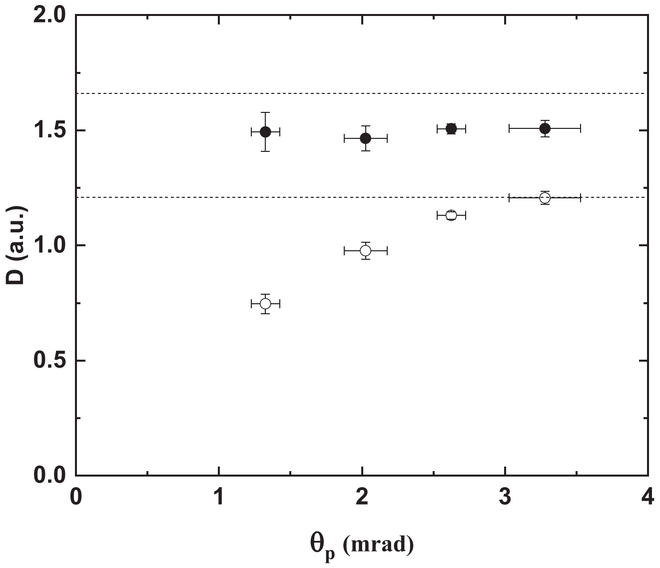

FIG. 4. Internuclear distance D at the instance of the dissociation extracted from the position of the extrema in the data of Fig. 2 as a function of θ_p, under the assumption that the phase shift is 0 (open symbols) or π (closed symbols). The dotted lines show the positions of the classical inner and outer turning points of the ground-state vibration.

In Fig. 3 R is plotted as a function of θ_p. Indeed, the oscillating interference pattern in R is much more pronounced than in the cross sections of Fig. 2. Clear minima can be seen at about 1.3 and 2.6 mrad and maxima at about 2.1 and 3.4 mrad. Between 0 and 0.6 mrad R is rather flat. These positions of the extrema are determined by the condition for constructive (even n) and destructive interference (odd n) $\mathbf{q} \cdot \mathbf{D} - \delta = n\pi$, which for the parallel orientation becomes

$$mv_p \tan(\theta_p) D - \delta = n\pi. \qquad (1)$$

D can then be calculated using Eq. (1) for the θ_p at which the extrema occur if we assume some value for δ. They are plotted in Fig. 4 as a function of θ_p as open symbols assuming $\delta = 0$ and as closed symbols assuming $\delta = \pi$. The dashed lines show the Ds for the classical inner and outer turning points of the vibrational ground state. This is the Franck-Condon region in which a transition can occur [20] (although in Ref. [20] a breakdown of the Franck-Condon principle was reported for double capture to very slow highly charged ions). For $\delta = \pi$ the Ds deduced from the position of the extrema are constant at about $D = 1.5$ a.u. This value is within the Franck-Condon region and close to the equilibrium distance of 1.4 a.u., as expected for dissociation by electronic excitation. In contrast, for $\delta = 0$ D increases with θ_p and, except for the data point at the largest θ_p, is well outside the Franck-Condon region. Therefore, the data of Figs. 2 and 3 are consistent with $\delta = \pi$, and thus with the symmetry argument of Ref. [9], but not with $\delta = 0$.

With regard to our hypothesis one important question is whether δ is constant or varies with θ_p. We therefore extracted δ from Eq. (1) assuming $D = 1.5$ a.u., as plotted in Fig. 5.

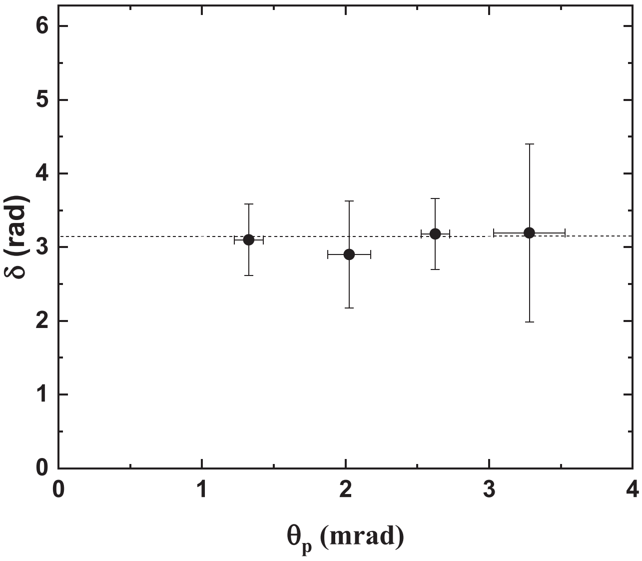

FIG. 5. Phase-shift δ extracted from the position of the extrema in the data of Fig. 2 as a function of θ_p, assuming a constant internuclear distance D of 1.5 a.u.

Within error bars the data are constant at $\delta = \pi$ and clearly inconsistent with δ approaching 0 for large θ_p, as observed for vibrational dissociation [14]. If we use the equilibrium distance for D instead of 1.5 a.u., δ is still constant, but slightly larger than π. Since the present data and those for vibrational dissociation were taken in the same experiment under identical conditions, an experimental artifact leading to a different θ_p dependence of δ in both channels is unlikely. The qualitatively different behavior of δ suggests that a different mechanism is responsible for the phase shift in vibrational dissociation.

The dashed curve in Fig. 3 shows the theoretical interference term from Ref. [5] for $\delta = \pi$ and $D = 1.5$ a.u. The positions of the extrema in the experimental data are very well reproduced for $\theta_\mathrm{p} > 1$ mrad. On the other hand, there are qualitative discrepancies for $\theta_\mathrm{p} < 0.7$, where the data are constant at 1, while the theoretical interference term predicts a minimum at $\theta_\mathrm{p} = 0$. However, it should be kept in mind that at this angle the molecular orientation (relative to q_tr, which is 0) is not defined and thus the FDCS must be equal for the perpendicular and parallel orientations. Therefore, with infinitely good resolution R would have to sharply step up at $\theta_\mathrm{p} = 0$ to $R = 1$. With a finite resolution this leads to an artificial maximum which cannot be resolved from the maximum near 0.5 mrad (with a natural width of about 0.8-mrad FWHM), which could explain the flat behavior at small θ_p.

The solid line in Fig. 3 shows the ratios between our calculations for the parallel and perpendicular orientations for the $2p\pi_\mathrm{u}$ state. There is not even qualitative agreement with the experimental data. Most notably, there is a large phase shift between the theoretical and experimental ratios as seen by the shift of the position of the extrema. Also, the pronounced maximum at $\theta_\mathrm{p} = 0$ is incompatible with $\delta = \pi$ found in the measured ratios. It should be noted that this maximum is not merely due to the fact that at $\theta_\mathrm{p} = 0$ the orientation is not defined. Since the calculation is not afflicted with any resolution, the ratios should sharply spike up from 0 to 1 if $\delta = \pi$. The finite width of the maximum shows that it is a true interference maximum.

In summary, the qualitatively different behavior of the θ_p dependence of δ from the data for vibrational dissociation reported in Ref. [14] shows that a different mechanism is responsible for the phase shift and the data for both dissociation channels are thus not inconsistent with our hypothesis.

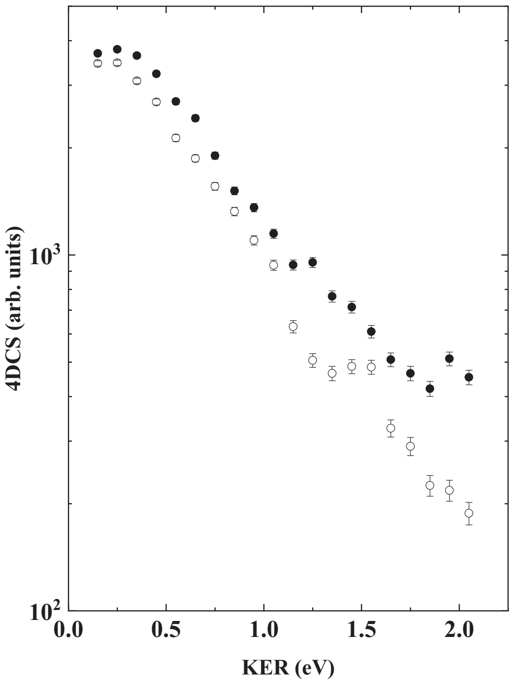

FIG. 6. Fourfold differential cross sections 4DCS (FDCS integrated over θ_p) for dissociative capture for the perpendicular (open symbols) and parallel orientations (closed symbols) as a function of the KER.

B. KER dependence of vibrational dissociation

In the previous section we provided evidence that the phase shift in vibrational dissociation has a different cause from the one for electronic excitation to an ungerade state. As a next step we need to investigate which mechanism does lead to the phase shift in vibrational dissociation. More specifically, in this section we will attempt to find further support for or to dismiss our hypothesis. To this end, we analyzed KER spectra in the range 0 to 2.1 eV, where dissociation by electronic excitation does not play any role. As mentioned in the Introduction, an interference between the direct and reflected paths should be observable in these spectra if our hypothesis was correct. In Fig. 6, we show the KER spectra for the perpendicular (parallel) orientation as open (closed) symbols. We found that the shape of the KER spectra is not sensitive to θ_p. The data in Fig. 6 thus represent FDCS integrated over θ_p (i.e. fourfold differential cross sections, 4DCS) in order to optimize the statistics.

The most prominent behavior of the 4DCS for both orientations is, as expected [15], a steep decline with increasing KER. However, for the perpendicular orientation we find, superimposed on this general trend, a maximum at about 1.5 eV. In contrast, for the parallel orientation we do not observe any

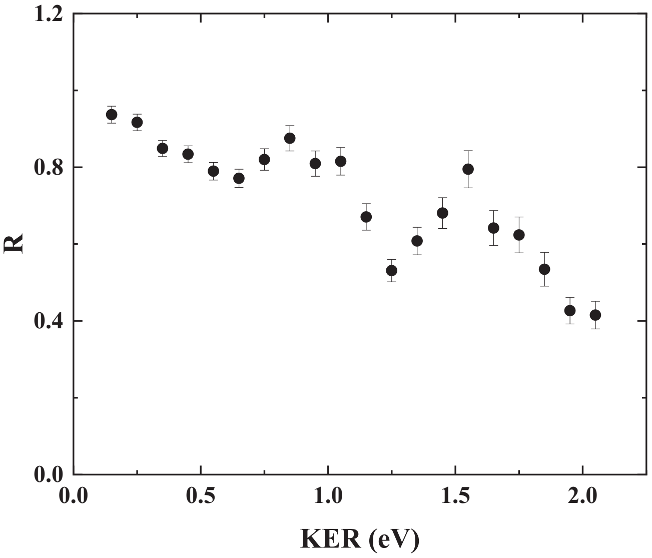

FIG. 7. Ratio between the 4DCS of Fig. 5 for the perpendicular and parallel orientations as a function of the KER.

statistically significant structure. On the other hand, we cannot rule out that there could be maxima at about 1.2 and 2 eV, but if these do not just reflect statistical fluctuations, they appear to be less pronounced than the maximum observed for the perpendicular orientation. Generally, the 4DCS tend to fall off faster with the KER for the parallel orientation; however, both datasets approach each other at about KER $= 0.8$ eV and then depart from each other once again for larger KER. This could be a signature of another structure which is blurred by the steep decline of the 4DCS with the KER.

In order to minimize any blurring effect on potential structures due to the steepness of the 4DCS, we analyzed the ratios R between the 4DCS for the perpendicular and parallel orientations. These are shown as a function of the KER in Fig. 7. Indeed, the maximum at 1.5 eV is now more pronounced and a second maximum at 0.8 eV, where the 4DCS for both orientations approach each other, is now clearly visible. If interference would only occur for one orientation and the incoherent part of the 4DCS had the same shape for both orientations, R would reflect the interference term. However, if the interference was indeed due to coherent contributions from the direct and reflected paths in our hypothesis, it would not be immediately clear why the path interference should only be present for certain orientations. Therefore, R should be regarded as a quantity which qualitatively reflects the ratio between the interference term for the perpendicular and parallel orientations. Unfortunately, this means that no quantitative information about the interference can be extracted from R. For example, the maximum at 0.8 eV could either reflect destructive interference in the parallel orientation or constructive interference in the perpendicular orientation. Nevertheless, the structures in R show that some kind of interference is present for at least the perpendicular orientation and possibly for the parallel orientation. The fact that the structures occur in the KER dependence shows that the phase angle depends on the KER, which matches the expectation for path interference between the direct and reflected paths.

While we do not claim conclusive evidence that the structure in R is due to such a path interference, we do believe that our interpretation is supported by a similar observation reported by Bryant *et al.* [21]. They studied photodetachment from H^- in a uniform external electric field. The total potential energy seen by the active electron is a combination of the atomic potential and the linear potential due to the external field. This potential has a qualitatively similar r dependence as the potential-energy curve for a binding molecular state: There is a maximum on one side of the nucleus (i.e., a potential barrier) due to the external field, a minimum at $r = 0$ due to the internal potential, and a maximum on the other side of the nucleus due to a combination of the decreasing external potential and the internal potential. Just like the fragments in vibrational dissociation, the ejected electron in photodetachment has two options: it can be ejected away from the potential barrier (direct path), or it can initially be ejected towards the potential barrier, from where it will get reflected (reflected path). Both contributions are indistinguishable and can thus lead to an oscillating interference pattern in the photoelectron spectrum. Such an interference was predicted by theory (Refs. [22,23]) and later observed in Ref. [21]. The interpretation of the authors was further supported by their own calculations as well as by theoretical work by Wang and Starace [24].

V. CONCLUSIONS

We studied dissociative capture in 75-keV $p + H_2$ collisions in the KER range 0 to 7 eV. Fully differential cross sections were analyzed for two molecular orientations in the plane perpendicular to the initial beam axis: one is perpendicular and the other parallel to the transverse momentum transfer q_{tr}. In the range KER $= 4$ to 7 eV dissociation is dominated by electronic excitation to the repulsive $2p\pi_{\mathrm{u}}$ state. Here, a pronounced two-center molecular interference pattern was observed in the projectile scattering-angle dependence for the parallel orientation. This pattern is afflicted with a constant phase shift of π. In contrast, for the KER range 0 to 2.1 eV, which is dominated by vibrational dissociation, earlier we reported a phase shift which strongly depended on θ_{p} [14]. Since the data for both KER ranges were taken in the same experiment under identical conditions, this different behavior of the phase shift cannot be explained by experimental artifacts. It is well established that a constant phase shift of π in dissociation by electronic excitation to an ungerade molecular state can be explained by symmetry considerations. The present data reconfirm that a nonzero phase shift can be realized by mechanisms not related to the molecular symmetry.

In the KER range 0 to 2.1 eV, i.e., for vibrational dissociation, we analyzed fourfold differential cross sections (FDCS integrated over θ_{p}) as a function of KER. For the perpendicular, and possibly for the parallel orientations we observe structures, which become more pronounced in the ratios between the 4DCS for both orientations. These structures are not inconsistent with path interference between direct and reflected vibrational dissociation, in which the phase angle should depend on the KER. Indeed, this type of interference has been observed in the energy spectra of electrons emitted in photodetachment in a uniform external electric field

[21], where the underlying potential is quite similar to the one underlying vibrational dissociation. Since in the case of photodetachment this path interference is well established, it seems to offer a realistic and plausible explanation for the structure we observed in the KER spectra as well. If confirmed, this would also provide strong support for our hypothetical explanation for the phase shift in the interference term for vibrational dissociation and the observation that this shift varies with θ_p. However, further studies, both experimental and theoretical, are needed to obtain conclusive evidence or to dismiss this hypothesis.

As an outlook, we plan to measure FDCS for vibrational dissociative capture of D_2 by proton impact. The electronic states are practically identical for H_2 and D_2 so that no significant differences in the FDCS for dissociation by electronic excitation are expected. In contrast, the vibrational states, as well as the conversion from momentum (which enters in the path-interference term) to the KER, are mass dependent. Consequently, the KER spectra should look quite different for both molecular species. More specifically, if indeed path interference leads to the structures observed in the KER spectra, the pattern should be sensitive to the mass. If observed, such a mass dependence would provide strong support that the KER spectra are afflicted by path interference and thus for our hypothesis.

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