Impact-parameter dependence of proton-induced K-shell ionization of carbon and aluminum

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The impact-parameter dependences of the ionization probability of the carbon and aluminum K shells by proton impact have been measured in a proton–x-ray coincidence experiment. Influences of the fluorescence yield and of the multiple scattering of the projectiles inside the target foils on the evaluation of the ionization probabilities have been investigated. The experimental results are compared with existing theories. Whereas the aluminum data can be explained by Coulomb ionization if adiabatic relaxation of the $1s$ state is taken into account, the experimental results for carbon exceed the theoretical values, especially at small impact parameters. Several possible reasons for this result are discussed.

I. INTRODUCTION

In recent years an increasing number of single-differential cross-section measurements of inner-shell excitation have been published[1-12] and thereby compared with current impact-parameter dependent ionization theories.[13-19] The present status of understanding of atomic inner-shell excitation may be divided into two categories: (i) The incoming Coulomb field of the projectile is only a small perturbation compared to the Coulomb field of the target nucleus ($Z_1/Z_2 \ll 1$). Simultaneously the ratio of the projectile velocity v to the orbital velocity u of the excited inner-shell electron should not be too small in order to prevent any adiabatic relaxation of the inner-shell wave function ($v/u \not\ll 1$). Both conditions limit the validity of theories like the plane-wave Born approximation (PWBA),[20] the binary encounter approximation (BEA),[15-17] or the semiclassical approach (SCA).[13] (ii) The Coulomb field of the projectile is a strong distortion of the target field ($Z_1 \cong Z_2$) and the inner-shell electronic state is allowed to adjust adiabatically as a function of the internuclear distance of both colliding particles. Thus the inner-shell excitation will be dominated by processes like electron pro- and demotion and corresponding couplings between molecular-orbital states (MO theories).[21]

Whereas the considered cases apply to the extreme regimes of inner-shell excitation, there exists—both experimentally and theoretically—a transition region. Starting from the pure case of Coulomb ionization the restriction for Z_1/Z_2 and v/u of (i) may be weakened thus allowing for adiabatic readjustment effects of the inner-shell states.[22] These effects may either be incorporated by a higher-order perturbation theory[23] or by approximating the first-order transition amplitude between molecular states within the perturbed stationary state method (PSS).[24] It is the aim of this work to fill this transition regime with experimental data by measuring the impact-parameter dependence of the K-shell ionization of carbon and aluminium by protons. Figure 1 shows the location of our experiments within the frame of existing ionization theories.[20] Whereas Z_1/Z_2 ratios equivalent to ours could be obtained also by other projectile-target combinations, we feel that the use of protons and low Z_2 targets has the advantage of excluding charge-state effects of the projectile and charge-exchange processes as competitive ionization mechanisms which may occur when higher Z_1 projectiles[11,12] are used.

II. EXPERIMENTAL METHOD

The differential cross sections have been measured by a proton–x-ray coincidence experiment. Magnetically selected proton beams with 100-μm diameter and collimated within 0.03° have been directed on self-supporting carbon foils of 3-μg/cm^2 thickness and 3-μg/cm^2 aluminium layers on 5-μg/cm^2 carbon backing. Scattered protons have been detected by a 50-mm^2 surface-barrier detector equipped with an annular diaphragm selecting scattering angles within a 2π geometry. An angular range from 0.5° to 20° with a relative angular acceptance of ±5% could be covered by moving the detector assembly along the beam axis. The cri-

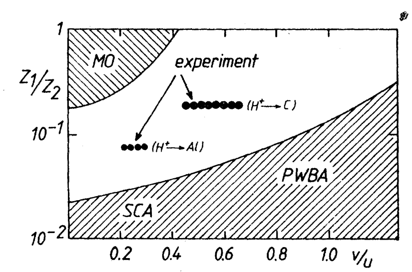

FIG. 1. Location of the experiments within a diagram demonstrating the limits of various ionization theories.[20]

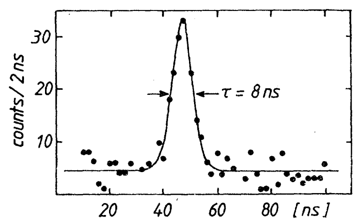

FIG. 2. Time spectrum of coincidences between C(*K*) x-ray and 200-keV protons scattered through 2.5°.

tical detector for the coincidence circuit is the x-ray detector. Usual Si (Li) detectors or proportional counters have time resolutions of the order of μsec, at least for the *K* radiation of carbon. This would require measuring times in the order of weeks for each data point. Instead we used a 0.5-μm thick CsI film as a photon-electron converter[25] followed by a channeltron. Since such an assembly is not energy dispersive, appropriate filter foils in front of the photocathode have been used. They consisted of a 5-μm thick Al foil for the Al target and a 750-$\mu g/cm^2$ carbon foil for the C experiment. The transmittivity of these foils provided at the absorption edge a photon window of 200-eV FWHM for Al(*K*) radiation and of 15-eV FWHM for C(*K*) radiation. The absolute photon efficiency of such an assembly has been carefully calibrated by comparing photon yields of Al(*K*) and C(*K*) radiation when the photocathode assembly and a proportional counter of well-known efficiency were alternatively used as photon detectors in a Bragg spectrometer. The absolute efficiency changes smoothly from 32% for Al(*K*) radiation to 40% for C(*K*) radiation. Figure 2 shows a typical time spectrum [specifically it was obtained for C(*K*) radiation and 200-keV protons at a scattering angle of $\theta = 2.5°$]: the time resolution of 8 nsec is about 2 orders of magnitude better than for Si (Li) detectors or proportional counters.

The target composition could be obtained by measuring backscattered protons with an annular surface-barrier detector placed upstream. This turned out to be necessary for a correction of impurity scattering inside the foil which was especially important for oxygen content in case of Al foils. With an additional particle detector in the forward direction, the multiple scattering distribution for each target foil was measured in order to correct for multiple scattering effects at small scattering angles. This will be treated in more detail in Sec. IV. Impact parameters and scattering angles were connected by the momentum approximation using the Molière potential.[26] Deviations from Rutherford scattering due to nuclear interference effects can be neglected even for carbon below 200 keV.[27] The Bohr parameter $\kappa = 2Z_2e^2/\hbar v$ lies in the range $4 \leqslant \kappa \leqslant 12$, thus indicating a classical path picture for the proton.[28]

III. FLUORESCENCE YIELDS

The large scatter of experimental data on the fluorescence yield ω_K for *K* radiation of carbon in the literature[29,30] and the probable influence of multiple ionization due to the use of proton projectiles revealed the necessity of finding an appropriate fluorescence yield which corresponds to the specific situation of our experiment. Thus we measured the total x-ray production cross section σ_X with our photon detector and compared it with the Auger cross section σ_A of Stolterfoht *et al.*[31] which was obtained with CH_4. Assuming that the ionization cross section of the C(*K*) shell is the same in a foil and in CH_4 (Ref. 32) we obtained the fluorescence yield by $\omega_K = \sigma_X/\sigma_A$. Figure 3 shows ω_K as a function of the proton energy. With increasing energy, i.e., with decreasing relative amount of multiple ionization, ω_K tends to reach the limiting value for the (K^1L^0) configuration.[33] The influence of multiple ionization on ω_K can be twofold: (a) it *increases* ω_K due to changes of the electronic configuration[34] and (b) it shifts the center of gravity of the *K*-emission band to higher photon energies due to the appearance of high-energy satellites.[35] This is an effect which is especially important for carbon since even a K^1L^1 configuration might shift a considerable part of the emission band into the absorption edge of carbon.[36] Since most x-ray detectors for low photon energies use C containing windows this effect may influence the photon yield considerably. The net result is an effective fluorescence yield which is *smaller* than $\omega_K(K^1L^0)$.[32] This we assume is probably the reason for the decrease of the experimentally determined ω_K towards small energies (Fig. 3). The combined error of ω_K due to the experimental Auger and x-ray data is estimated to be 20%. The use of thus obtained ω_K values for the evaluation of ionization cross sections from experimental photon yields seems to be justified,

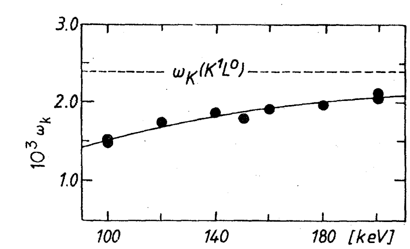

FIG. 3. Fluorescence yield ω_K of carbon as a function of proton energy. In addition the limiting value of the (K^1L^0) configuration is inserted.

since in the independent electron approximation the vacancy production due to standard Born predictions are valid for the removal of an inner-shell electron without specification of the fate of any other electron.[37] It should be emphasized that the effective fluorescence yield of Fig. 3 is confined to the specific situation of our experiment and transforms therefore x-ray data into ionization probabilities correctly.

Effects like these for C do not occur for Al since even the K^1L^5 satellite line lies below the absorption edge. Since on the other hand $\omega_K(K^1L^1)$ and $\omega_K(K^1L^0)$ deviate only by a few percent[38] we took for ω_K(Al) at all proton energies the value $\omega_K = 0.038 \pm 0.002$.[39]

IV. MULTIPLE SCATTERING

The assumptions, which normally are accepted for the evaluation of impact-parameter dependent ionization probabilities from photon-particle coincidence experiments, are that the relevant scattering angles lie well inside the single-scattering region and that the photon yield from all other encounters except the one which is responsible for the large-angle scattering event can be neglected. In order to investigate these questions quantitatively we looked for a solution of the following question: What is the distribution of impact-parameters of all elastic collisions inside the foil which scatters a particle to a preset total scattering angle for a given thickness of a scattering foil? The convolution of this distribution with the ionization probability will determine the coincidence yield. Clearly, for two extreme situations the result can be foreseen: if the total scattering angle θ (which is identical with the detector angle) is large compared to the HWHM angle $\theta_{1/2}$ of the multiple scattering function, the distribution will be a δ function at that impact parameter ρ which relates ρ with θ via the single scattering law. The δ function is placed on a background which results from statistical scattering events. In contrast, for $\theta \ll \theta_{1/2}$, the distribution $d\nu/d\rho$ will completely resemble that of statistical scattering: $d\nu/d\rho = 2\pi\rho N \Delta x$, where N is the target atom density and Δx the foil thickness. Adding up statistical single scattering events to a resulting scattering angle as in usual multiple scattering theory,[40,41] it can be shown that the impact-parameter distribution $(d\nu/d\rho)_\theta$ for a given total scattering angle θ and any foil thickness Δx is given by[42]

$$\left(\frac{d\nu}{d\rho}\right)_\theta = 2\pi\rho N\Delta x\,[\pi f(\theta;\Delta x)]^{-1} \times \int_0^\pi d\varphi f([\theta^2 + \vartheta^2(\rho) - 2\theta\vartheta(\rho)\cos\varphi]^{1/2};\Delta x),$$

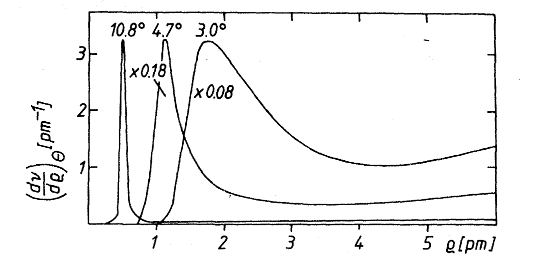

FIG. 4. Impact-parameter distribution of 200-keV protons in an aluminum foil for three different total scattering angles. The HWHM of the multiple scattering distribution is 0.65°.

where $f(\theta;\Delta x)$ is the multiple scattering function and $\vartheta(\rho)$ the single scattering angle for a given impact parameter ρ. Thus the coincidence yield resulting from excitations in a foil will be proportional to $\int_0^\infty (d\nu/d\rho)_\theta P(\rho)\,d\rho$, where $P(\rho)$ is the desired ionization probability.

Multiple scattering functions $f(\theta;\Delta x)$ have been measured for each scattering foil. Comparison with theoretical estimates of $f(\theta;\Delta x)$ (Ref. 41) shows an almost perfect agreement if a 15% uncertainty of the foil thickness Δx is taken into account. Based on the experimentally determined multiple scattering function, impact-parameter distributions $(d\nu/d\rho)_\theta$ for 200-keV protons on an aluminium foil for which $\theta_{1/2}$ equals 0.65° have been calculated and are shown in Fig. 4. For total scattering angles 10.8° and 4.7° the distribution is more like a δ function; however, for an angle of 3° it is rather broad in spite of the fact that this angle is roughly a factor of 5 larger than $\theta_{1/2}$. Introducing multiple scattering as a perturbation the influence on the differential cross section will be to shift the ionization probability to larger impact parameters. For our experimental results given in the next section the influence of multiple scattering is strongest for the 3° example of Fig. 4. A deconvolution of the photon yield coincident with 3° scattered protons showed that the extracted ionization probability is shifted to an impact parameter which is 40% larger than the one from a single scattering law. A detailed treatment of this multiple scattering effect and the resulting deconvolution will be published elsewhere.

V. EXPERIMENTAL RESULTS AND DISCUSSION

Figure 5 shows the ionization probability $P(\rho)$ as a function of impact parameter. The data points are for 200-keV protons on aluminum. In Fig. 6,

$$P = \frac{d\sigma^I/d\Omega}{d\sigma^S/d\Omega}$$

is given for a fixed scattering angle of 3° as a func-

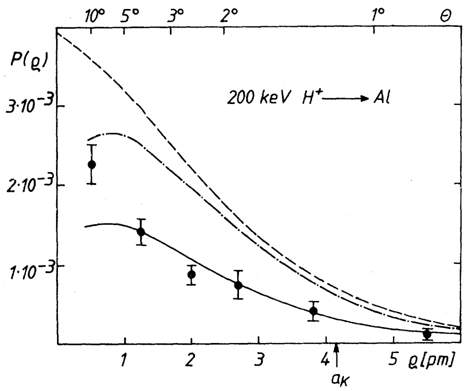

FIG. 5. Comparison of the experimental ionization probability $P(\rho)$ with different forms of SCA; dashed curve: noncorrected SCA; dash-dotted curve: corrected for deflection and retardation; solid curve: additionally binding has been incorporated.

tion of proton energy where $d\sigma^I = P(\rho)\, d\sigma^S$ with $d\sigma^S = 2\pi\rho\, d\rho$ is the differential ionization cross section. The error bars in Fig. 5 and the following ones reflect mainly the error due to counting statistics. The ratio of true to accidental coincidence counts ranged from 1 to 5. Additionally the error resulting from the described correction procedure has been added properly if necessary. Here an important contribution is the above mentioned background of statistical scattering events. For this correction the absolute target thickness must be known which introduces the main error source at large impact parameters. Not included is a systematic error which results from the uncertainty in the fluorescence yield (see Sec. III).

In Fig. 5 the experimental points are compared with different versions of the semiclassical approach (SCA). Whereas the dashed line represents the noncorrected SCA, in the dash-dotted curve retardation and Coulomb deflection and, in addition, in the full drawn curve the binding effect are included.[24] For the latter effect a Ritz-like variational principle for the adiabatic adjustment of both the binding energy and the K-shell wave function has been applied.[12] For the SCA ionization probability no scaling has been adopted, but the original matrix elements of Bang and Hansteen[13] have been calculated including s and p states for the electron continuum wave function in case of aluminium K-shell excitation. If $P(\rho)$ is calculated according to the binary encounter approximation (BEA)[15] including adiabatic relaxation effects and path integration due to the true Kepler motion of the projectile the theoretical curve is nearly the same as the SCA curve of Fig. 5, except for impact parameters smaller than 1 pm. Here the BEA curve is favored by the experimental points. This may indicate that the SCA corrections which were taken at the point of closest approach are slightly too strong.

Figure 6 demonstrates that the energy dependence of the ionization probability at a fixed scattering angle is reproduced by both theories if the corrections cited above are introduced. The numerical integration of $P(\rho)$ of Fig. 5 yields a total cross section of the Al K ionization at 200 keV of 530 b. The simultaneously measured total cross section was 600±90 b. In comparison the theoretical cross sections including all corrections are $\sigma(\text{SCA}) = 540$ b and $\sigma(\text{BEA}) = 520$ b. Basbas *et al.*[43] have found experimentally 470±110 b.

Figure 7 shows similar results for carbon K-shell ionization at three different proton energies. Here the comparison with theory is mainly made with the BEA concept. SCA calculations for 200-keV protons ($v/u = 0.62$) showed that both theories coincided within 15% (in the impact-parameter region of interest) if s, p, and d scattering were included. Since the SCA calculations were about a factor of 10^4 as time consuming as the BEA calculations only the latter ones will be compared with the experiment. The full lines in Fig. 7 are the BEA curves obtained after a correction for binding and the Kepler motion. One can clearly see that at small impact parameters the experimental points deviate from the theory.

In Fig. 8 this deviation is again shown by plotting the ratio $P_{\text{exp}}/P_{\text{theor}}$ as a function of the impact parameter. For very small impact parameters the deviation reaches a factor of 4. It should be emphasized at this stage that this deviation from theory will hardly be seen in the total cross section[31,44] since the contribution from impact parameters in the range $0 \leq \rho \leq 3$ pm can be neglected

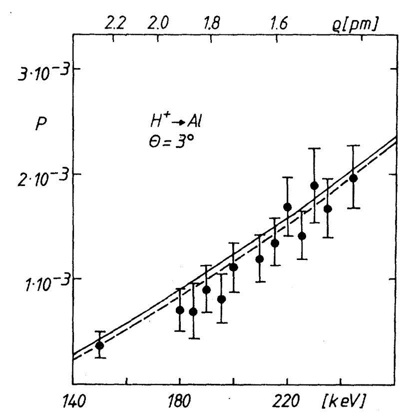

FIG. 6. Energy dependence of the ionization probability of aluminum at 3° compared with the full corrected SCA and BEA results (solid and dashed curve, respectively).

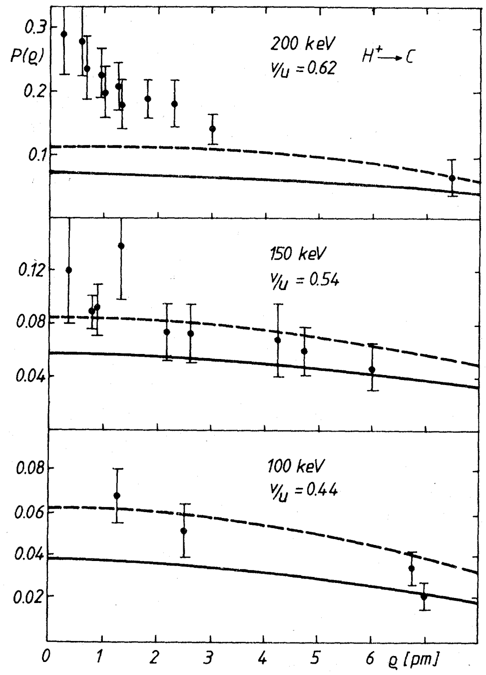

FIG. 7. Carbon K-shell ionization probability for three different proton energies. Comparison is made with the noncorrected (dashed) and corrected (solid) BEA.

with regard to the rest of the ionization function which extends at the experimental proton velocities up to the $C(K)$-shell radius a_K of 9.3 pm. Figure 8 indicates that the deviation increases with decreasing impact parameters at a fixed proton energy and at a fixed impact parameter with increasing energies. This behavior excludes the possibility of explaining the deviation by any kind of a recoil effect. The recoil velocity increases with decreasing impact parameter at a fixed projectile energy but decreases with increasing energy at a fixed impact parameter. Therefore, if there would be any effect which increases the photon yield beyond that of the BEA predictions and which depends on the recoil velocity in a unique fashion, it would not be consistent with the experimental data of Fig. 8. Besides this it should be mentioned that for instance for 200-keV protons the primary recoil energy at $\rho = 0.5$ pm is about 120 eV which is below the 1s binding energy of carbon. Thus any K x-ray production by C-C collisions is prohibited.

Deviations from the SCA model at very small impact parameters have also been found by Andersen *et al.*[45] and Chemin *et al.*[46] In the former work the enhancement of $P(\rho)$ at very small ρ values is attributed to a variation of the ionization probability with scattering angle θ due to interference between the excitation amplitudes corresponding to the projectile trajectory before and after the collision (they differ due to the rotation of coordinate system by an angle θ). A detailed analysis of the SCA theory for the in- and outgoing part of the particle path shows that $P(\theta)$ can be written as $P(\theta) = P_0(1 + B\cos\theta)$, if only s and p scattering is considered. Since the absolute value of the anisotropy coefficient $|B|$ is almost of the order of 1 (Ref. 45) and the scattering angle varies in the case of the C target only between 0.5° and 10°, this effect would account for an additional variation of the ionization probability of only a few percent and can therefore be ruled out. The same holds for the polarization of the initial state by the fast projectile,[47] an effect which increases the ionization probability at large projectile velocities ($v/u \rightarrow 1$) but is operative preferentially at large impact parameters ($\rho/a_K \gtrsim 1$). Andersen and co-workers[12] looked for deviations in the SCA amplitude by going beyond the stationary phase approximation. By regarding the time variation of the transition frequency they have been able to show that this time dependence increases the ionization amplitude especially at large impact parameters. Thus again, this effect does not give any increase of $P(\rho)$ at small distances from the nucleus. The same holds for charge exchange.

At the energies under consideration protons are believed to behave like ions passing through solids.[48] Therefore only charge exchange to the projectile continuum should be regarded. Estimates by Doolen *et al.*[49] on the basis of the Brinkman-Kramers theory show that this can lead to appreciable ionization of target K electrons at high velocities. Since the adiabatic impact parameters for both electron loss and capture do not significantly deviate[13,50] and are, in our case, of the order of the K-shell radius, the differential charge exchange cross section would nearly have the same flat shape as the SCA curve and therefore does not explain the additional increase of $P(\rho)$ at small values of ρ. There exists—at least in principle—

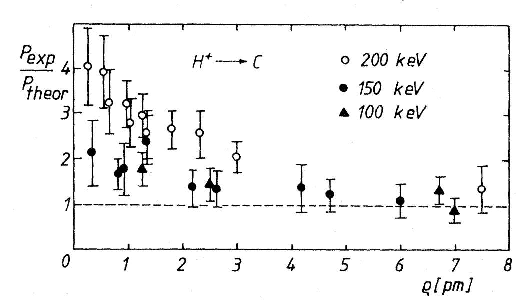

FIG. 8. Ratio of the experimental and theoretical ionization probabilities as a function of the impact parameter. The theoretical P_{theor} values are those of the full corrected BEA.

the possibility of an impact-parameter dependent effective fluorescence yield. In order to simulate the results of Fig. 8 it has to *increase* at small impact parameters. It is hard to believe that any additional L-shell ionization could affect the fluorescence yield especially at these ρ values and it would as well contradict the finding of Sec. III which has shown that any additional ionization tends to *decrease* the fluorescence yield.

Similar results as those discussed here have been found by Chemin *et al.*[46] They measured the ionization probability at zero impact parameter for a variety of targets and different energies. If $P(0)$ is compared with the noncorrected BEA[17] it is found that the experimental values get increasingly larger than theoretical ones if v/u approaches unity. Since for $v/u \rightarrow 1$ any of the described adiabatic corrections become negligible, this result is comparable with our findings for protons on carbon. The cited authors attribute this deviation to a possible difficulty of adequately describing K-shell electrons of atoms with low atomic number by a hydrogenic model. While such a deviation might explain an overall discrepancy between experiment and the BEA model [including $P(0)$-values], it is hard to see how for instance Hartree-Fock wave functions enhance the ionization probability especially at small ρ/a_K values.

The previous discussion was based on theories like the SCA or BEA since both have been tested in many cases successfully by measurements of both total and differential cross sections. In contrast, Kaminsky *et al.*[51] have shown that $P(\rho)$ evaluated by a method given by Schiff[52] decreases remarkably steeper with increasing ρ than ionization probabilities given by either SCA or BEA theories. This could be in accord with our experimental findings of carbon K-shell ionization.

Finally it should be mentioned that the energy ω_{rot}, where $\omega_{\text{rot}} = v/\rho$ is the rotation frequency of the intermediately formed molecule, reaches values at $\rho \cong 1$ pm which are comparable with the ionization energy for aluminum but exceeds that for carbon by an order of magnitude. Thus the adiabatic correction procedure might be questionable especially for carbon at small impact parameters.

VI. SUMMARY

The ionization probability of the Al(K) and C(K) shell as a function of impact parameter has been measured and compared with Coulomb excitation theories. Whereas the K-shell ionization of aluminum can be well described if the Kepler path of the incident proton and an adiabatic relaxation of the K shell due to the proton are taken into account, the carbon data exceed the theoretical prediction especially at small impact parameters. A variety of additional effects such as recoiling of the target nucleus, interference of the ingoing and outgoing excitation amplitude, polarization, and charge exchange fail to explain the experimental data. An explanation for such a deviation cannot be given at the moment. It should be noted that at the velocities under discussion $P(0)$ is of the order of 0.1. This might be too large for a successful application of first-order perturbation theory.[53]

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