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2018

REELS measurements and MC simulations of electrons backscat-tered from graphite surface at energies between 500 and 5000 eV have been presented. The electron scattering angle was 130◦; the angle of the incident electron beam was 50◦ and the detection an-gle was 0◦ with respect to the surface normal, respectively. Both elastic and inelastic collisions were considered during the simu-lation. The incident angle of the electron beam is relatively large, so the bulk dielectric function for graphite was used. Our results show that intensities and shapes of the simulated backscattered electron spectra are in good agreement with the measurements. In all cases, there was a strong elastic peak as well as significant inelastic contributions. The existing discrepancy between measure-ments and theoretical calculations can be attributed partially to the surface roughness and contamination of the surface sample. The calculated energy spectra were very sensitive to the applied dielectric functions and better agreement is obtained for higher energies, probably because different surface effects are dominant in the lower energy region. Overall, we can conclude that there is a good accordance between the experiment and the calculations. This suggests that our theoretical model can be used to explain backscattered electron energy distribution for carbon and for different materials

We presented *ab initio* calculations of the electronic differential energy-transfer cross sections for antiprotons with energies between 3 keV and 1 MeV interacting with helium. By comparison with simulations employing the mean-field description based on the single-active electron approximation we were able to identify electron correlation effects in the stopping and straggling cross sections. We found that straggling exceeds the celebrated Bohr straggling limit when correlated shake-up processes were included.

We have investigated the radiative decay of double *K*-shell vacancy states produced in solid Ca, V, Fe, and Cu targets by impact with about 10 MeV/amu C and Ne ions. The *K*-hypersatellite x-ray lines were measured by means of highenergy-resolution spectroscopy using a von Hamos curved crystal spectrometer whose FWHM energy resolution varied between 1.6 eV for Ca and 3.4 eV for Cu. The main objective of this work was to determine the single-to-double*K*-shell ionization cross sections for the eight investigated collisions. The cross-section ratios were deduced from the corrected relative intensities of the hypersatellites. The results were found to be on one hand consistent with the theoretical predictions provided by the SCA model based on Dirac-Hartree-Fock wave functions but, on the other hand, systematically bigger than the latter. This was explained by the fact that the electron capture process which is not considered in the SCA model plays an important role in the investigated collisions as shown by CTMC calculations and leads to bigger cross-section ratios. However, when the EC process is taken into consideration, the theoretical cross-section ratios become significantly bigger than the experimental ones because, as shown in the literature, the EC cross sections are strongly overestimated by theory. The difficulty was circumvented by correcting the EC contribution with a scaling factor α which was determined by a least-squares-fit method. Finally, using the SCA DHF model for the determination of the DCI cross sections and the CTMC predictions corrected by the scaling factor $\alpha = 0.11$ for the EC cross sections, a quite satisfactory agreement between theory and experiment was found for the $\sigma KK/\sigma K$ cross-section ratios.

The combined experimental and theoretical studies of the optical properties and excitation energies of iridium were presented. REELS spectra were measured on clean Ir at 500, 1000 and 2000 eV primary energies and the RMC method was applied to calculate the energy loss functions in the 2–200 eV range. The high accuracy of our results was justified by using the ps- and f-sum rules. We found that our data are much closer to the nominal theoretical values than any others. The average of the three loss functions was further used to determine the refractive, n, and extinction coefficients, k. The peak positions in the optical constants derived from the REELS spectra partly agree well with Weaver's data in the low energy below 40 eV. Our calculation of k slightly differs from the previous results, but for n we found larger differences. Moreover, we identified for the first time in the optical data the 5p3/2, 4f5/2 and 4f7/2 shell excitation of solid Ir sample. Furthermore, using the derived ELF the IMFP of Ir was presented from 1 eV up to 10 keV.

The transmission of low-energy electrons through a macroscopic steel capillary has been investigated both experimentally and theoretically. The length of the steel capillary was L = 19.5 mm and the inner diameter was d = 0.9 mm. The kinetic energy distribution of electrons transmitted through the steel capillary was recorded for a tilt angle of $\psi = 2.6°$ of the incident electron beam with respect to the capillary axis. Accompanying simulations based on classical transport theory reproduce the experimental data to a high degree of agreement. Transmission for other tilt angles has also been simulated to investigate the influence of the tilt angle on the guiding efficiency.

We have calculated the Energy Loss Functions ELFs and the optical onstants of three transition metals, Cr, Co, and Pd, in the photon energy range of 0–120 eV from the analysis of three Reflected Eelectron Energy Loss Spectroscopy (REELS) spectra at three primary energies. A recently developed RMC method was applied to extract the absolute ELF values from each REELS spectrum, where a Monte Carlo simulation of electron scattering in the REELS experiment and a global optimization technique for oscillator parameter fitting were employed. With implementation of a spatially varying differential inverse inelastic mean free path (DIIMFP), derived under the semi-classical framework for electron inelastic scattering and Mott's cross-section for electron elastic scattering, an accurate description of electron transport was established for the REELS spectral analysis. The validity of the obtained ELF of the reverse Monte Carlo (RMC) t method has been confirmed by the fairly good performance of the f- and ps-sum rule checks and by good consistency of the ELFs obtained at different primary energies. This work then proves that the RMC method with REELS spectra enables establishing a database of optical constants for many materials at a higher accurate level than the previous databases.

We presented a theoretical description of the spectra of electrons elastically scattered from various samples. The analysis was based on very large scale Monte Carlo simulations of the recoil and Doppler effects in reflection and transmission geometries. Besides the experimentally measurable energy distributions the simulations give many partial distributions separately, depending on the number of elastic scatterings (single, and multiple scatterings of different types). Furthermore, we presented detailed analytical calculations for the main parameters of the single scattering, taking into account both the ideal scattering geometry, *i.e*. infinitesimally small angular range, and the effects of the real, finite angular range used in the measurements. The effect of the multiple scattering on intensity ratios, peak shifts and broadening, were shown. We have shown results for multicomponent and double layer samples. Our Monte Carlo

simulations were compared with experimental data. We found that our results are in good agreement with the experimental observations.

2019

The spatial interference effects appearing during the ionization of atoms (H, He, Ne, and Ar) by few-cycle laser pulses using single-electron *ab initio* calculations were studied. The spatial interference is the result of the coherent superposition of the electronic wave packets created during one half cycle of the driving field following different spatial paths. We have shown that this spatial interference pattern can be interpreted as the hologram of the target atom. With the help of a wave-function analysis (splitting) technique and approximate (strong-field and Coulomb Volkov) calculations, we have directly shown that the hologram is the result of the electronic-wave-packet scattering on the parent ion. On the He target we demonstrated the usefulness of the wave-function splitting technique in the disentanglement of different interference patterns. Further, by performing calculations for the different targets, we have shown that the pattern of the hologram does not depend on the angular symmetry of the initial state and it is strongly influenced by the atomic species of the target: A deeper bounding potential leads to a denser pattern.

We presented a combined experimental and theoretical study of the transmission of single charged 1-keV Ar ions through a cylindrical glass capillary of macroscopic dimensions. From quantitative measurements of the incoming and transmitted ion currents, combined with a detailed analysis, the amount of beam entering the capillary was determined. This, combined with the measured transmitted currents, was used to determine the amount of charge deposited on the inner wall of the capillary which produces the guiding electric field. We have shown experimental results for fully, and partially, discharged conditions of the time evolution of the guided beam intensity following a wide range of times during which the capillary was allowed to discharge in order to provide information about the insulating surface charging and discharging rates. Combining our recent theoretical model describing the charge patch dynamics with these data, it is shown that the model is consistent with the experimental transmission curve data measured after the capillary was allowed to discharge for times ranging from 5 to 1000 s or longer and for injected currents that differed by a factor of 50. In contrast, models which do not include a dynamic rearrangement of charge along the surface prior to decay were found to be in consistent with our experimental measurements. Additional data about the time dependences of the fraction of the injected beam which is transmitted as a function of injected beam current when transmission through the capillary is inhibited due to blocking are also presented. These data have a temporal dependence consistent with our model predictions that blocking occurs when the total capillary charge, i.e., the capillary potential, reaches a certain value.

The state-to-state (exchange) interference of the autoionizing resonances of helium is studied in (e,2e) experiments. These studies are disturbed by the coincidence events caused by the direct ionization, so their decrease is desirable. For this reason, to mimic the experimental observation, we performed four body classical trajectory Monte Carlo calculations. The calculations were done for 93.15 eV primary energy, where the exchange interference of the 2s2(1S) and 2p2(1D) autoionizing states of helium is expected. The yields of non-coincidence and coincidence events detected in various combinations of scattering geometry were calculated and compared with the experimental observations.

We presented a combined experimental and theoretical study of the electron elastic differential cross sections of triethyl phosphate molecule (C2H5)3PO4 (TEP). The experimental setup was based on a crossed beam technique comprising of an electron gun, a single capillary gas needle and a detection system with a channeltron was used to measure differential cross sections. The absolute scale for the cross sections was obtained by relative-flow method using argon gas as a reference. For the interpretation of the measured data we applied the partial expansion method to calculate the elastic cross sections of TEP. We found excellent agreement between the shapes of measured and calculated data.

The energy loss functions (ELFs) of three transition metals (Cr, Co and Pd) have been derived from reflection electron energy loss spectroscopy spectrum with a theoretical analysis of the measured data. In this work, we updated our previous ELFs in a wider photon energy region (0-200 eV) with a better accuracy, which is verified by sum rules and a root-mean-square deviation. The electron inelastic mean free paths (IMFPs) of Cr, Co and Pd have been calculated with the obtained ELFs by adopting a dielectric response theory. We employed both the single-pole approximation and full Penn algorithm for the calculation of IMFPs, and the calculated results are compared with previous works.

We presented an analysis of electron incident angle-dependent reflected electron energy loss spectroscopy spectra of silver by using a Monte Carlo simulation method with which the separated contributions from surface and bulk excitations can be identified. The simulations were performed at several different incident electron energies and with various incident angles with respect to the surface normal. We found that the surface plasmon excitation plays a dominant role in the loss peak at around 3.7 and 7.5 eV.

We extended the semiclassical two-step model for strong-field ionization that describes quantum interference and accounts for the Coulomb potential beyond the semiclassical perturbation theory to the hydrogen molecule. In the simplest case of the molecule oriented along the polarization direction of a linearly polarized laser field, we predicted significant deviations of the two-dimensional photoelectron momentum distributions and the energy spectra from the case of atomic hydrogen. Specifically, for the hydrogen molecule the electron energy spectrum falls off slower with increasing energy, and the holographic interference fringes are more pronounced than for the hydrogen atom at the same parameters of the laser pulse.

The photoionisation of rubidium in strong infra-red laser fields based on ab initio calculations was investigated. The bound and the continuum states are described with Slater orbitals and Coulomb wavepackets, respectively. The bound state spectra were calculated with the variation method and we found it reproduced the experimental data within a few percent accuracy. Using the similar approach, ionization of Rb was also successfully investigated. The effects of the shape and the parameters of the pulse to the photoionisation probabilities and the energy spectrum of the ionized electron are shown. These calculations may provide a valuable contribution at the design of laser and plasma based novel accelerators, the CERNAWAKE experiment.

The collisional radiative models used in the modeling of beam emission spectroscopy diagnostics neglect atom-atom collisions because of a lack of sufficiently detailed atomic data. Filling this scantiness we performed a classical trajectory Monte Carlo simulations to calculate the cross sections for various channels in collisions between $H + H2$ and $Li + H2$ for a wide range of projectile energies. Based on the calculated cross sections, a simplified version of the

collisional radiative model has been derived. We have shown that the model is suitable to obtain the beam attenuation in neutral gases outside of the confined plasma region. A strong density dependence has been found for each beam species.

The K-shell ionization cross sections of copper and silver atoms by muons and pions with negative and positive charge have been calculated with the classical trajectory Monte Carlo method and the plane wave Born approximation with corrections for the Coulomb-deflection and binding-energy effects. Both results are in good agreement with each other. The obtained cross sections for muons on copper are also compared with the coupled-channels calculations.

We have presented combined experimental and theoretical works of the study of the absolute single and double electron loss (stripping) and single and double electron capture cross sections for O+ projectiles colliding with water molecule. The measured projectile energy range was from 0.2 to 1.2 MeV. The objectives are to provide new data of interest not only to applications in hadron therapy and to contribute the available absolute data for testing theoretical models describing many-body processes of collisions between many-electron projectiles colliding with multielectronic systems. The present data have been compared with an isoelectronic system, i.e. with the methane molecule. All measured cross sections (single and double electron loss and single and double electron capture) are virtually the samefor both targets. The present data were also compared with the results of classical trajectory Monte Carlo calculations. We found very good agreement between the present results for water and the previous data for the single and double electron loss and single and double electron-capture cross sections by O+ projectiles in methane.

We developed a new computer simulation code that calculates trajectories of photoelectrons emitted from nanoparticles by laser excitation. The code uses the pre-calculated electric field obtained by finite-difference time-domain simulations as input. The photoelectron trajectories emitted from silver nanoparticles were calculated using the classical trajectory Monte Carlo method, where the image force towards the surface is taken into account. We show that our present code is suitable to describe the recent experimental findings reasonably well. Significant effect of the image acceleration to the calculated electron spectra was observed. We found that the calculated energy distributions of photoelectrons are in agreement with the recent experiments.

We presented comparisons between quantum and classical calculations for above threshold ionization of an Argon atom interacting with a strong laser pulse. The time-dependent complete-active-space self-consistent field (TD-CASSCF) method and the classical trajectory Monte Carlo (CTMC) method were used for the cases of quantum and classical calculations, respectively. We found differences between the results obtained by the TD-CASSCF and the CTMC simulations.

We presented theoretical investigation of the temporal evolution of the energy spectrum of ions passing through an insulating capillary. In our simulation we used 1 MeV proton microbeam, and a single, cylindrical shaped polytetrafluoroethylene (PTFE) macrocapillary. In order to prevent the simple geometrical transmission the beam axis was tilted with 1° compared to the capillary axis. According to our simulations, we found that the beam first hit the inner wall of the capillary and build a positive charge patch at a well localized place of the inner capillary surface. As a result, a repulsive electric field is generated for protons enter later into the capillary and a collision-less transmission occurs, which is called ion guiding. We have

shown that our simulated energy spectra are in good agreement with our previous experimental findings.

Experimental observation of the dynamics of a micro-focused, 1 MeV proton beam guided through a single, insulating macrocapillary was presented. The micrometer sized proton beam was center injected into a poly(tetrafluoroethylene) capillary with macroscopic dimensions. The capillary was tilted to 1° with respect to the beam, i.e. no geometrical transmission was possible, but the beam first hit the inner capillary wall. After a sufficient electric charge-up of the insulator material due to the interaction with the charged particle beam, guiding appeared thanks to the beam deflection caused by the formed electrostatic field. With an increasing amount of the accumulated charge on the wall, the deflection of the beam also increased, resulting in the dynamics of the transmission. When a dynamical equilibrium between the charges being deposited and flowing away in form of leakage current was set in, the deflection of the beam saturated at a certain position. Here, the transmitted fraction of the beam reached 90% relative to the incident beam. At this position the angular distribution of the transmitted beam was determined. In the sample tilting plane focusing effect was observable, while in the perpendicular plane, the beam was defocused.

The interaction of charged particles with materials can always be associated with the energy transfer process that results in the change of the energy of the particles. We presented examples when projectiles suffer energy loss either in ion-atom, or electron-surface and ion-surface collisions. We presented classical trajectory Monte Carlo results to calculate energy losses of the projectiles in proton-hydrogen atom collisions. The obtained results verify that high order effects should be included for a proper description of electronic stopping power. Energy loss of charged particles near surfaces pose several interesting problems, among them the separation of surface from bulk effects. We analyzed the possible way of the separation in electron-surface and ion-capillary collisions. We have shown that the correlation between the angular distribution and the energy loss of ions passing through capillaries can be used to probe the surface loss functions without the contribution of the bulk one.

2020

We presented a detailed analysis and comparison of four models describing the extension of the electron-energy loss function from the optical limit of $q \rightarrow 0$ into the (q,ω) plane to obtain the bulk and surface terms of differential inverse inelastic mean free paths. We found that the best model that describes accurately and times efficiently the calculation of the energy loss function of free-electron-like materials is the combination of the Penn algorithm [Phys. Rev. B **35**, 482 (1987)] with the Ritchie-Howie method [Philos. Mag. **36**, 463 (1977)]. Applying this model in our reverse Monte Carlo method, we determined, with high-precision, electron-energy loss functions of silicon and germanium based on the theoretical analysis of the high-energy resolution reflected electron energy loss spectroscopy (REELS) spectra, measured at 3, 4, and 5 keV incident electron energies. The refractive index n, the extinction coefficient k, and the complex dielectric function ($\varepsilon = \varepsilon 1 + i\varepsilon 2$) were calculated from the obtained energy loss function in a wide energy loss range of 0–200 eV. The accuracy of the obtained results is justified with various sum rules. We found that the calculated optical data of Si and Ge fulfill the sum rules with an average accuracy of 0.11% or even better. Therefore, the use of these optical data in materials science and surface analysis is highly recommended for further applications.

A Monte Carlo method has been applied to the simulation of the secondary electron emission from a SiO2 macro-capillary due to electron irradiation. For the analysis of the present charging problem, a simplified approach was employed to deal with the charging of internal wall of the capillary. These calculations on the secondary electron yields of the internal wall and the top surface found that, with a small tilt angle of 2◦, the top surface that dominates the secondary electron emission for the SiO2 macrocapillary.

Cross sections for ionization, charge exchange, and low-level excitation channels were simulated for a Be4++H(1s) collision system using the classical trajectory Monte Carlo method (CTMC) and quasiclassical trajectory Monte Carlo method of Kirschbaum and Wilets (QTMC-KW). We draw the conclusion that the classical treatment can describe the calculated cross sections reasonably. While there is no experimental data for the mentioned collision system, we compared our results with the previous literature based on other methods, such as AOCC, MOCC, adiabatic superpromotion model, and Symmetric Eikonal approximation. We found that our calculations are in good agreement with previous results.

We presented a four-body classical trajectory Monte Carlo simulation of collisions between two ground state hydrogen atoms. The total cross sections for the dominant channels—namely, the net single ionization of the target and ionization of the projectile, resulting from direct ionization and electron transfer (capture or loss) processes for 2.0–100 Kev/amu incident energy of the hydrogen atom impact—have been presented. We also presented cross sections for the complete break of the system, resulting in the final channel for four free particles. While the pure direct ionization channel is a one-step process, the ionization channel in combination with electron capture and loss is a two-step process. Our results were compared with theoretical and experimental data for hydrogen–hydrogen collision systems with the same energies. Our recent cross sections for projectile ionization (CTMC EL) show excellent and close agreement with the experimental data above 20 keV incident energies and also show good agreement with the CCC+B2e approximation. However, for the case of direct target ionization, our calculated cross sections above 30 keV are higher than the previous theoretical results.

We have presented a Monte Carlo simulation of the electron backscattering coefficient of Be, B and C (glassy, graphite and amorphous) at impact energy range between 0.1 keV and 100 keV. Both the high energy cascade secondary electrons and Auger electrons produced during the transport of the incident electrons inside the bulk beryllium were taken into account. This up-to-date CTMC-SEM modelling uses the Mott's cross section calculated with more accurate scattering potential and a relativistic dielectric functional formalism with full Penn algorithm and experimental optical data. Sum rule checks indicate that, beryllium and glassy carbon have good sum rule values and the simulation of backscattering coefficients for them should be reasonable; while boron and amorphous possess an under-estimated sum rule and graphite has overestimated ones, hence, the calculation of backscattering coefficients for them would tend to be overestimated and underestimated, respectively. The simulation results show that the Auger electrons contribute negligibly to the backscattering coefficient. For beryllium the simulation results are found well below experimental data at low energies and approaches the lower limit of the experimental data distribution range at high energies above 10 keV. Further simulations for carbon or water absorbed on beryllium surfaces indicate that a contamination with a thickness of one or two atomic/molecular layers can largely increase the backscattering coefficient of beryllium below 1 keV. Therefore, the early experimental data measured either in bad vacuum conditions and/or without previous ion-sputter cleaning of the surface are likely to be over-estimated. The very low values of the backscattering coefficients for extreme low-Z elements (Be and B) are partly attributed to the strong elastic forward scattering, while

previously it was assumed that the backscattering coefficient is dominated by the total cross sections of elastic and inelastic scattering. We believe we have derived the presently most accurate backscattering coefficients for beryllium and glassy carbon among all available theoretical and experimental data. We highly recommend to use these data in applications at least until further accurate experimental data for clean Be surface become available. Both reliable experimental and theoretical benchmark results need to be established in future.

Works under publication

Lihao Yang, Bo Da, Károly Tőkési and Zejun Ding, Mixing-free individual separation of surface, bulk and Begrenzungs effect components in the surface electron energy spectra, Scientific Reports to be published.

We presented the first theoretical recipe for the clear and individual separation of surface, bulk and Begrenzungs effect components in surface electron energy spectra. The procedure ends up with the spectral components originated from surface and bulk excitations without any mixing between the components. As an example, the model is applied to the reflection electron energy loss spectroscopy spectrum of Si. The electron spectroscopy techniques can directly use the present calculation schema to identify the origin of the electron signals from a sample. Our model provides the possibility for the detailed and accurate quantitative analysis of REELS spectra.

L.H. Yang, A. Sulyok, M. Menyhárd, G. Sáfrán, K. Tőkési, B. Da and Z.J.Ding, Optical properties of amorphous carbon derived from reflection electron energy loss spectroscopy spectra, Carbon to be published.

We presented the combined experimental and theoretical investigations of optical properties of amorphous carbon. The reflected electron energy loss spectroscopy (REELS) spectra of carbon were measured with a cylindrical mirror analyzer in ultrahigh vacuum conditions at primary electron energies of 750, 1000 and 1300 eV. The energy loss function and thereby the refractive index n and the extinction coefficient k were extracted from these REELS spectra in a wide loss energy range of 2-200 eV by applying our reverse Monte Carlo method. The high accuracy of the obtained optical constants is justified with the ps- and f-sum rules. We found that our present optical constants of amorphous carbon fulfill the sum rules with the highest accuracy compared with the previous published data. Therefore, we highly recommend to replace the previous data with the present ones in a practical application. Moreover we present the atomic scattering factors of amorphous carbon obtained based on the present dielectric function to predict optical constants of amorphous carbon with a given density.