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Plasmon Excitation in the Interaction of Slow Singly Charged Argon Ions with Magnesium

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Abstract: We report angle-resolved energy spectra of electron emitted in the interaction of slow singly charged heavy ions with Mg surface. The work is focused mainly on the excitation of plasmons of Mg under Argon impact. Potential excitation of plasmons occurs when incoming ions are neutralized at the expense of the potential energy carried by incoming ions. The process competes with the known mechanisms of neutralization via Auger transitions. Differently from Al samples, our results show that the neutralization of Ar^{+} ions at Mg is dominated by the excitation of surface plasmons by the potential energy released in the electron capture process that neutralizes incoming ions. Bulk plasmon excitation is observed at higher impact energy and is ascribed to fast electrons excited by the transfer of the kinetic energy of incoming particles. The data show that bulk plasmon excitation occur inside the bulk, while the theoretically predicted excitation by potential energy transfer of incoming projectiles is not observed.

Keywords: plasmons; electron emission; ion beam analysis; ion scattering

Citation: Riccardi, P. Plasmon Excitation in the Interaction of Slow Singly Charged Argon Ions with Magnesium. *Solids* **2024**, *5*, 321–332. https://doi.org/10.3390/solids5020021

Academic Editor: Joaquim Carneiro

Received: 14 April 2024
Revised: 17 May 2024
Accepted: 29 May 2024
Published: 1 June 2024

1. Introduction

Charge exchange and electronic excitations in the interaction of slow ions with solids are currently the subject of an intense investigation [1–14]. These processes are important in charge fraction formation, in energy deposition in solids and in several applications such as microscopy and characterization of materials. Most of these recent investigations have focused on electronic excitations that occur in local interactions through binary atomic collisions. However a detailed description of charge exchange and electronic excitation processes involves also non local interactions that occur at distance from the surface as well as delocalized excitations of the solid like plasmons. The knowledge of these phenomena is not readily accessible through scattering techniques and charge fraction measurements. On the other side, electron emission is one of the main outcomes of these processes, making electron spectroscopy techniques very well-suited to obtain detailed information on electronic processes that occur in ion-solid interaction [13–16], which can complements the information obtained with other techniques.

Electron emission processes are grouped in the two main categories of Potential Electron Emission (PEE) and Kinetic electron emission (KEE). PEE occurs at the expense of the potential energy carried by incoming ions [17–19], while KEE occurs from the conversion of the kinetic energy of incoming projectiles [19–24]. Studies of PEE dates back to the 50s, starting with the pioneering work of Hagstrum [17,25–28], who discussed PEE in terms of the conversion of the potential energy carried by incoming projectiles into electronic excitation and emission through Auger transitions. Hagstrum provided a taxonomy of these Auger transitions. The Auger Neutralization (AN) involves two electrons of the solid. In AN, the hole in the incoming singly charged ion is neutralized by electron capture from the solid. The energy released in this transition is transferred to a second electron of the solid which is revealed in vacuum as an Auger electron with a characteristic energy. On the other side, Auger processes that involve one electron of the solid and one of the ion are termed Auger Deexcitation (AD). The incoming ion can be neutralized by electron capture from the solid releasing energy to an electron in an higher energy level of the projectile. Alternatively, the hole in the incoming ion can be neutralized by an electron excited in an higher energy level, releasing energy that is transferred to an electron of the solid. This last process is usually preceded by a Resonant Neutralization (RN) process, in which incoming ions are neutralized by a resonant capture of an electron of the solid into an excited state. Therefore, RN processes do not produce electron emission directly, but can be important as precursors of subsequent Auger transitions.

Another mechanism of neutralization of incoming ions involves plasmons excitation [19,29–33]. Plasmons of energy E_{pl} can be excited if the potential energy trasformed in the electron capture process that neutralizes incoming ions is sufficient. This process has been studied theoretically. Calculations have predicted either the excitation of monopole surface plasmons of high momentum q or the excitation of bulk plasmons [34,35]. Experimentally, the excitation of plasmons can be identified by the electrons that are emitted when they decay transferring their energy to a valence electron (interband transition) [19,29–33]. This emission produces a characteristic structure in the spectra of emitted electrons. The plasmon structure is characterized by a high energy edge determined when the plasmon energy is transferred to an electron at the Fermi level. Therefore, the spectrum of electrons emitted by decay of plasmons have a maximum energy $E_m = E_{pl} - \phi$, where E_{pl} is the plasmon energy and ϕ is the metal work function. Our recent experimental investigations on potential excitation of plasmons are consistent with the excitation of either the predicted monopole or the multipole surface plasmons [13,14]. So far, this latter collective excitation has not been considered in theoretical calculations.

On the other hand, spectra of electrons emitted in the interaction of low energy ions with metal surfaces reveal the excitation of bulk plasmons, that has been observed at higher impact energies in the keV range. The energy dependence of plasmon excitation in Al samples indicated the existence of a threshold incident energy for bulk plasmon excitation. This threshold appears to be consistent with the threshold for excitation of the LMM Auger signal of the target. Moreover, plasmon excitation has been observed as loss satellites in the Al-2p Auger spectra excited by electron impact as well as by electron promotion in violent atom-atom collisions in our energy range [14]. These observation are consistent with the idea that, at the threshold, plasmon excitation results primarily by fast Auger electrons traveling inside the solid.

Plasmons in the free electron metals Al and Mg have been extensively studied since the 50's with several techniques, including electron energy loss spectroscopy (EELS) and electron induced secondary electron emission (SEE) [36–39]. Plasmon have been also observed as losses and gain satellites in Photoemission and Auger electron spectra [40]. In the field of ion-solid interactions, most of the investigations of plasmon excitations have been performed on Al targets and found nicely consistent with experiment of electron induced secondary electron emission and electron energy loss spectroscopy [33,39]. Investigations of plasmon excitation in the interaction of low-energy ions on Mg targets are sparse and in some studies of PEE using He projectile the excitation of plasmons was not identified [41]. Very recently we have performed an investigation of electron emission from Mg under the impact of Neon projectiles [13]. The study has focused on plasmon excitation and projectile autoionization. The spectra revealed under Ne impact on Mg show unambiguous signatures of potential excitation of plasmons. In this case, the structures due to electron emission from potentially excited plasmons are clearly separated in energy from the AN ones. For Neon we observed that at low impact energy E_i of the incident ions surface plasmons are excited by PEE during the neutralization of incoming ions, while at higher E_i bulk plasmons are excited by the transfer of the kinetic energy of incoming projectiles.

This work is focused on the excitation of plasmons in the interaction of slow singly charged Ar ions with Mg. Earlier PEE spectra by Ar impact on Mg [29] showed a feature that could correspond either to an Auger Neutralization process or to a plasmon assisted neutralization. The measurements reported in this work allow to resolve this long standing issue. The electron emission spectra acquired in this work show that the neutralization behaviour of argon ions at Mg surfaces is dominated by the potential excitation of surface plasmons.

At higher impact energy the spectra provide evidence for bulk plasmon excitation. Angle resolved measurements show a cosine dependence of the intensity of emission from bulk plasmon decay on the observation angle of emitted electrons, indicating that bulk plasmons are excited at depth inside the solids. The results are consistent with the idea that these plasmons are excited by fast electrons excited by the transfer of the kinetic energy of the projectiles.

2. Experiments

The experiments have been performed at university of Calabria using a setup that has been described elsewhere [14]. It consisted in a UHV chamber with a base pressure in the low 10^{-10} Torr range. Neon and Argon ions in the 0.5–6 keV energy range were produced in an electron impact source. The source was operated at a discharge voltage lower than the second ionization potential of the atomic projectiles, to prevent the formation of a significant amount of doubly charged ions. The Ne and Ar ion beams were characterized with a movable Faraday cup at the target position. At any of the energies used in this work the beams had a gaussian spatial profile with a width of less than 1 mm in both the vertical and horizontal directions and their currents were kept in the low 10^{-9} A range.

Samples were mounted on a manipulator whose rotation allowed for variation of the ion incidence angle. Electrons emitted from the sample were collected by an hemispherical energy analyzer that was mounted on a goniometer, so that the incidence angle Θ_i and the observation angle Θ_e could be varied independently. The ion beam direction, the axis of the spectrometer and the surface normal were coplanar and throughout the paper Θ_i and Θ_e will be measured with respect to the surface normal.

The spectrometer had an acceptance angle of 1.5° and was operated at a pass energy of 50 eV and a constant transmission function over the measured energy range. The energy scale of emitted electrons was calibrated by the high energy edge of the LVV Auger spectrum of Mg and by comparing the energy of electrons from autoionization lines of Ne^{**} $2p^4 3s^2$, with published value [13]. The UHV chamber was shielded with μ-metal to reduce the effect of stray magnetic fields on the trajectory of emitted low energy electrons. Comparison with published results shows that the spectra are reliable down to energies lower than 1 eV.

The samples were polycrystalline Mg surfaces, 99.999% purity, that were sputter cleaned by 6 keV Argon impact. The cleaning was monitored by checking for oxygen and carbon contaminants in electron induced Auger electron spectra and was continued until the ion induced spectra became constant. In some cases small traces of carbon remained in Auger electron spectra which had no significant effects on the shape of ion induced spectra.

3. Results and Discussion

Figure 1 reports energy distributions $N(E)$ of electrons emitted from a Mg surface under the impact of 5 keV Ar^+ ions as a function of observation angle Θ_e and for fixed incidence angle Θ_i = 30° (angles are measured with respect to surface normal). At this impact energy electron emission is mostly determined by the kinetic emission process. The spectra are dominated by a broad feature due to the emission of secondary electrons excited during the cascade of electronic collisions inside the solid generated by incoming projectiles. The emission of secondary electrons produces a feature with a peak at about 3 eV followed by a high energy tail. The secondary peak and its continuous tail toward higher energies constitutes the background on which are superimposed discrete features originating from other specific emission mechanisms. In the electron energy range of Figure 1, the spectra show a shoulder that can be attributed to electron emission from the decay of bulk plasmons of Mg [29], as will be discussed in the following. The visibility of the plasmon structure it is commonly enhanced taking the first derivative of the spectrum dN(E)/dE. The numerical derivatives of the spectra in Figure 1 are shown in Figure 2. For Argon projectiles impacting with 5 keV kinetic energy on Mg, the plasmon shoulder results in the minima at energy E_m~6.9 eV, which corresponds closely to the q = 0 bulk plasmon of Mg ($E_m = E_{pl} - \phi$ = 10.6–3.75 eV), consistently with earlier results on polycristallyne Al and Mg target. The energy of the bulk plasmon is also consistent with EELS measurement.

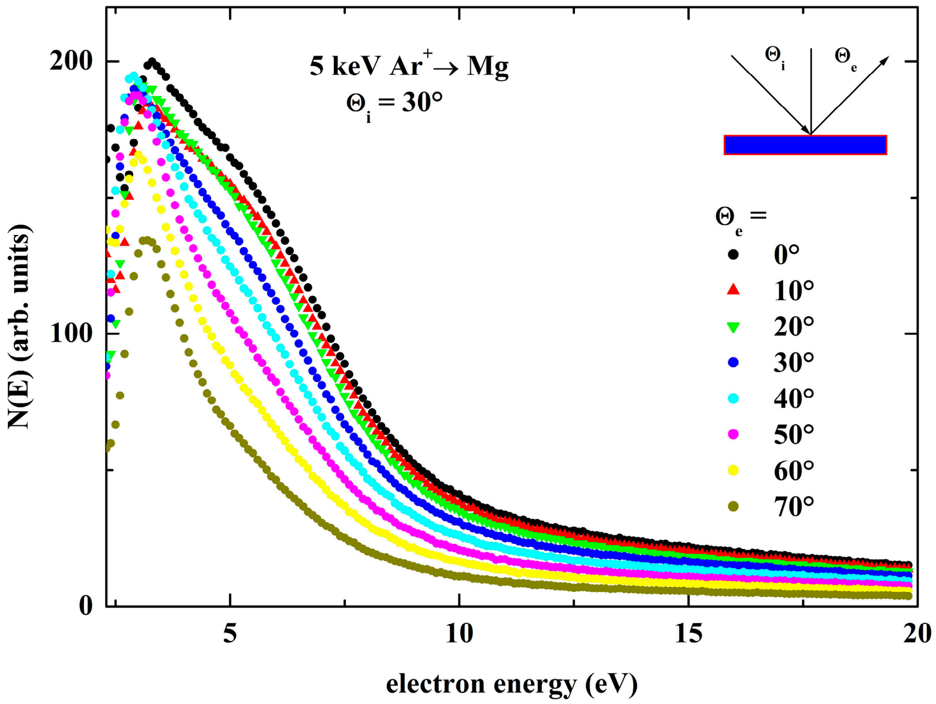

Figure 1. N(E), the energy distributions of electrons emitted from Mg surfaces by 5 keV Ar^+ at varying the emission angle Θ_e and for fixed incidence angles Θ_I = 30°.

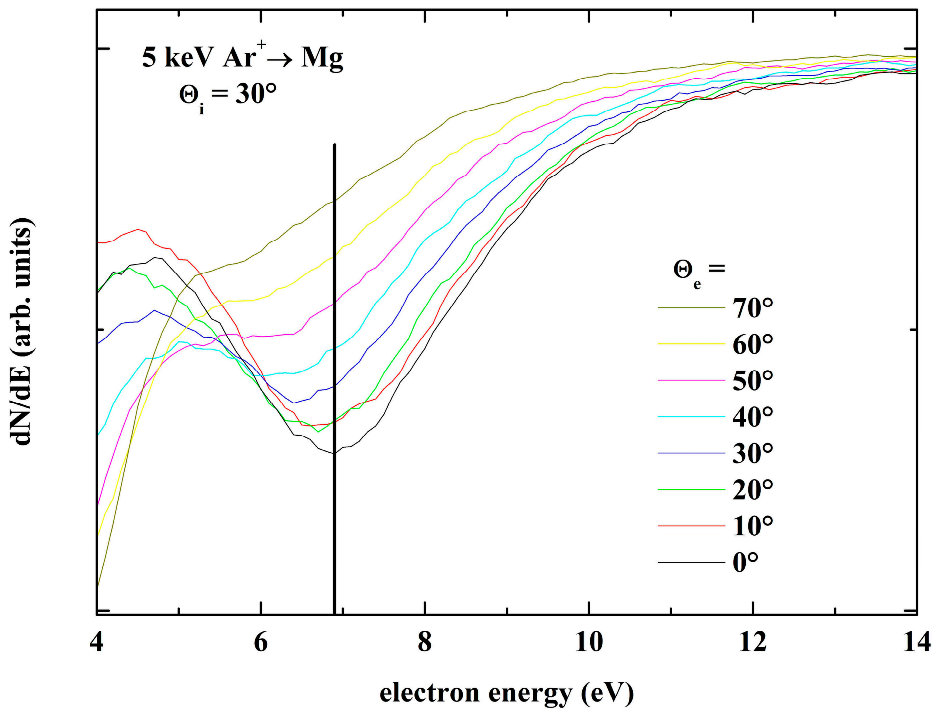

Figure 2. Derivatives dN(E)/dE of the spectra in Figure 1 showing the minima due to the bulk plasmon decay feature. The vertical line marks the position of the bulk plasmon of Mg.

Figures 1 and 2 show that the plasmon shoulder grows above the background of secondary electrons as the observation direction is moved toward the normal to the surface (as Θ_e decreases). The intensity of electron emission from plasmon decay can be estimated from a simple analysis of the derivatives [30,31] and are reported in Figure 3 as a function of the observation angle Θ_e. These intensities have been evaluated by subtracting a smooth polynomial background from the derivatives of the spectra. This subtraction results in a negative peak whose integral is proportional to the intensity of emission. We observe that the intensity of emission from decay of bulk plasmons exhibits a cosine behaviour as a function of emission angle. This behaviour is explained considering that the decay of plasmon is an isotropic source of electrons, so that electrons excited at a certain depth are more attenuated when they travel inside the solid and are emitted in directions closer to the surfaces.

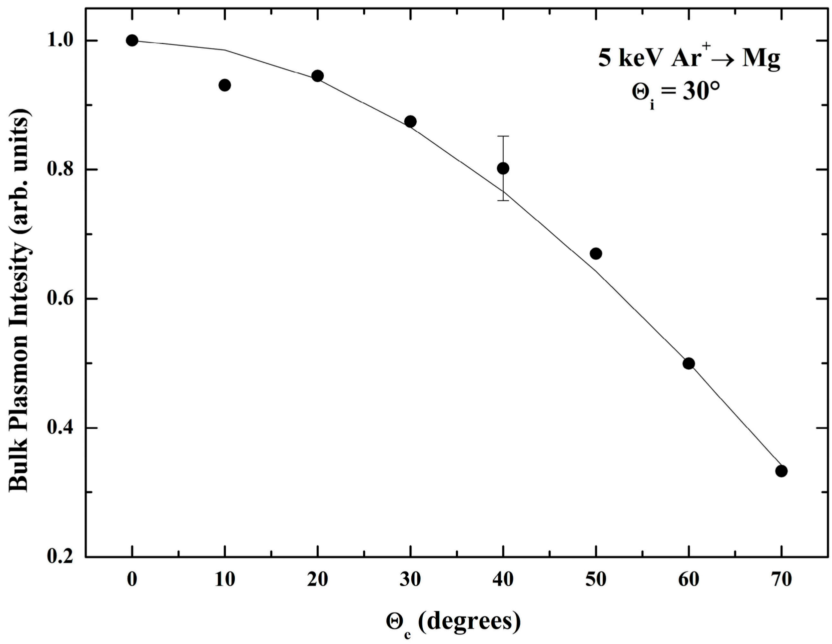

Figure 3. Cosine dependence of the intensity of the plasmon decay feature in the spectra in Figures 1 and 2. The intensities have been normalized to their maximum value. An error bar of 10% has been added to the graph.

The current results are consistent with previous research performed on Al surfaces [14]. The dependence on the impact energy of the projectiles of the intensity of emission from plasmon decay also indicated the existence of a threshold energy for bulk plasmon excitation by keV Ar^+ ions that could be correlated to the threshold for Al L-shell ionization in Ar-Al collisions [14,33]. The results we obtained in this investigation of Argon impact on Mg are displayed in Figure 4, which reports the spectra for the impact of Ar^+ ions on the Mg surfaces at incident energies ranging from 1 keV to 3 keV. The derivatives of the spectra in Figure 4 are reported in Figure 5. At 3 keV impact energy the spectra are already dominated by KEE and the plasmon feature in the spectra appears at an energy closely matching that of the bulk plasmon of Mg. With decreasing ion energy the spectra show the transition from the kinetic emission to the potential regime.

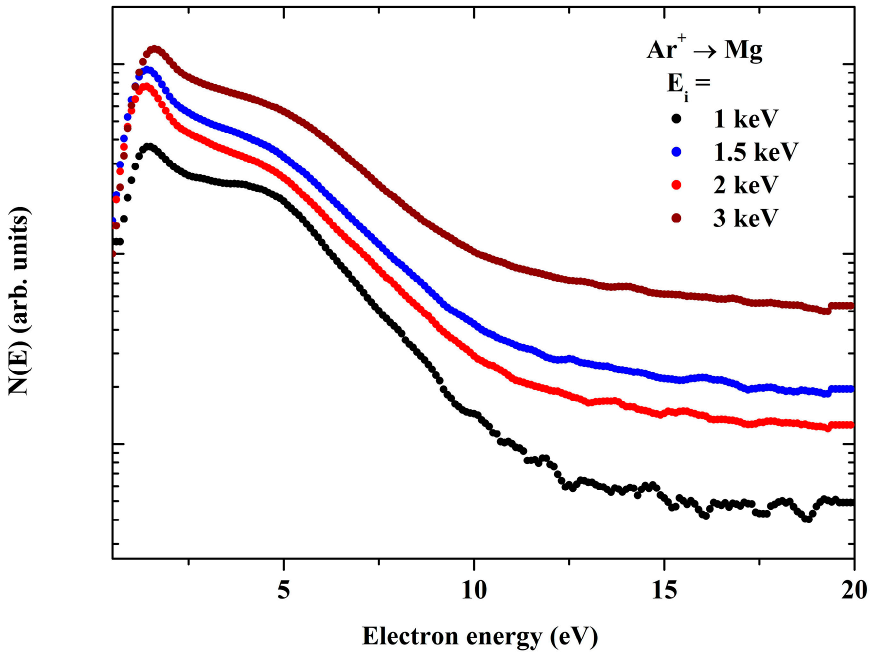

Figure 4. Energy spectra of electrons emitted from Mg surface under the impact of Ar^+ ions as a function of the ion incident energy E_i, for fixed Θ_i = 60° and Θ_e = 0°. The spectra have been normalized to the beam current and have been displaced on the vertical scale for clarity.

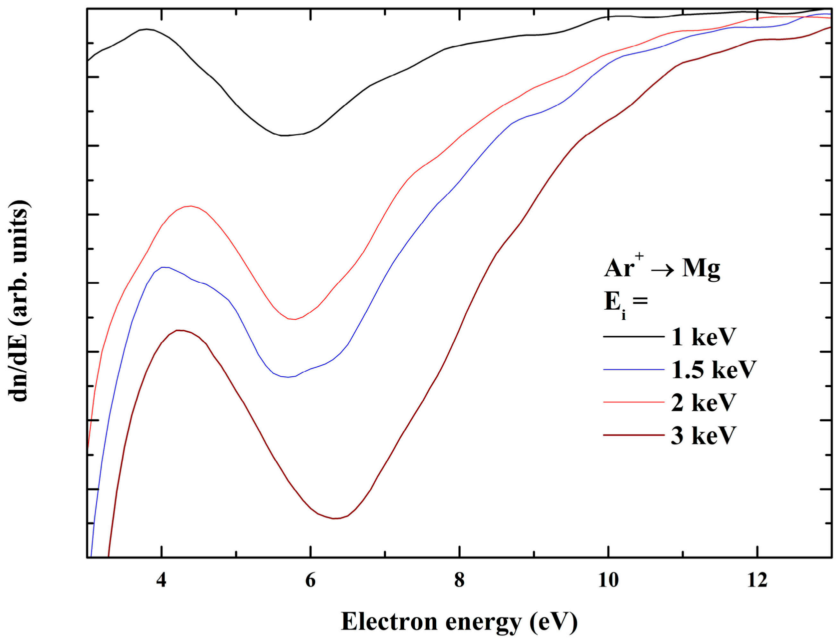

Figure 5. Derivative of the spectra in Figure 4.

In the derivatives of Figure 5, we observe that, with decreasing ion energy, a second shoulder appears on the low energy side of the bulk plasmon, which become dominant as the projectiles' energy is reduced. These observations are fully consistent with earlier investigations [14] and also observed in the case of Neon and Helium impact on Mg [13,14]. Therefore, the excitation of bulk plasmons of Mg under Argon impact is assigned to the energy loss suffered by fast secondary electrons, primarily 2p Auger electrons of Mg, excited in binary atomic collisions inside the solid.

On the other side, the spectrum acquired at 1 keV in Figures 4 and 5 shows a minimum in the derivative at an energy that is not consistent with that of a bulk plasmon. Bulk plasmon excitation by potential energy transfer from incoming ions when they are above the surface has been theoretically proposed [35]. We conclude that in the PEE regime the spectra do not reveal the proposed excitation of bulk plasmons. Moreover, The results of the angular measurements show that bulk plasmon excitation occurs at the expense of the kinetic energy of incoming projectiles and requires penetration inside the bulk of incoming particles.

The investigation of the potential electron emission regime in the interaction of Argon ions on Mg is discussed in the following. The results for Argon will be compared with those recently obtained in the case of Neon projectiles incident on Mg [13]. Figure 6 shows the spectra acquired in the case of 500 eV Ne^+ and Ar^+ ion impact on Mg. At this impact energy the spectra are dominated by PEE and show its characteristic features. As above, to help in discerning spectral features superimposed to the background of secondary electrons, Figure 7 shows the numerical derivatives of the spectra in Figure 6. In this case, a slight smoothing has been applied to the derivatives, taking care that this procedure did not introduce artifacts. In the case of Neon projectiles, the spectra show also features due to kinetic emission: the low energy peak due to electrons excited in the cascade of electronic collisions inside the solid, and two lines at energies around 20–25 eV due to the autoionization of Ne $2p^4\,3s^2$ excited by electron promotion in violent binary atomic collisions with surface atoms [42–45]. These last peaks have been recently investigated for the information they provide in the electronic processes that occur in ion-solid interactions [46–49] and are not correlated to plasmon excitation [32].

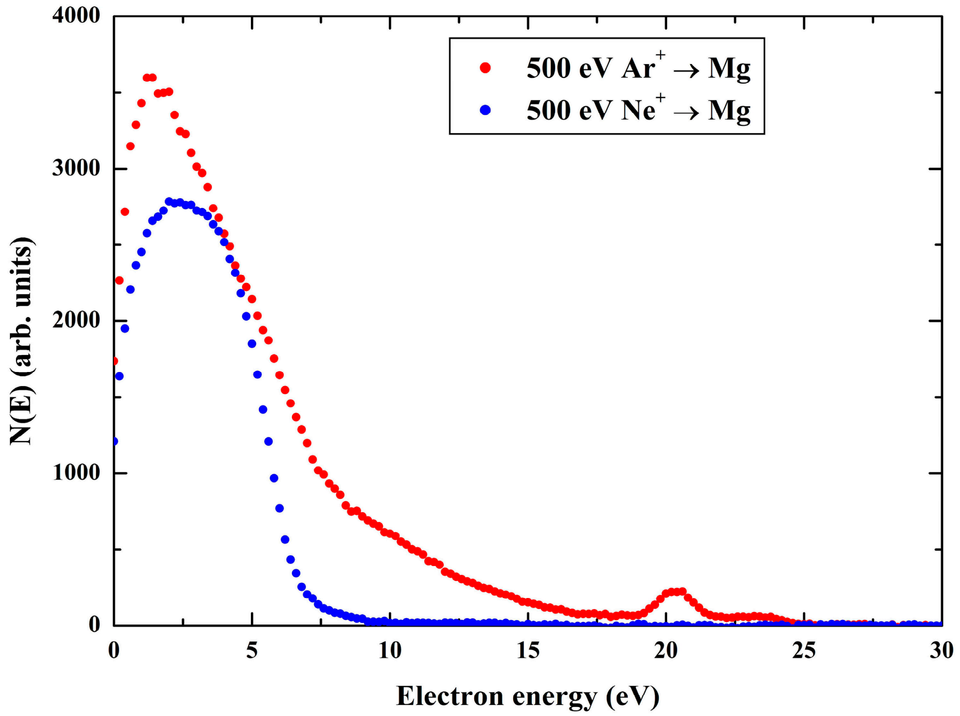

Figure 6. N(E), the energy distributions of electrons emitted from Mg surfaces by 500 eV Ar^+ and Ne^+ ions.

In the case of Neon, PEE produces two broad features in the spectra of emitted electrons, due respectively to the Auger Neutralization and the Plasmon assisted neutralization processes. Both these features can be identified by their characteristic high energy edge, which produces minima in the derivatives of the spectra. The potential energy transferred in the Auger Neutralization of singly charged Neon projectiles is related to the ionization potential of the incoming projectile and can be written as $\Delta E = \mathrm{I}' - \phi$, where I' is the ionization potential of the incoming projectile, which is reduced by an energy Δ due to the image interaction with the surface, and ϕ is the metal work function. The maximum energy of emitted electrons will occur when this energy is transferred to an electron at the Fermi level of the surface. Therefore, the spectrum of electrons emitted in an Auger neutralization process have a maximum energy $E_b = I' - 2\phi$ [17,19]. In the case of Neon, the ionization potential of an isolated atom is I = 21.6 eV. Given that the work function of Mg is $\phi = 3.75$ eV, the emission of electrons by Auger neutralization is therefore identified in the feature revealed in the spectrum excited by 500 eV Ne^+ ions in the energy range 8–14 eV, which results in the minimum in the derivative dN(E)/dE in Figure 7 at the energy E_b =12 eV, which implies a shift of the ionization potential $\Delta = 2$ eV, consistent with earlier estimations [17]. On the other hand, the feature revealed at 4–8 eV energy in the spectrum of electrons emitted by Neon in Figures 6 and 7 can be assigned to emission of electrons from the decay of plasmons [13]. The spectrum of electrons emitted by plasmon decay produces a feature with a maximum energy $E_m = E_{pl} - \phi$, which corresponds to the minimum observed in the derivatives of Figure 7 at about 6 eV. This energy is about 1 eV lower than the energy expected for the bulk plasmon of Mg but matches the expectation for the excitation of a multipole surface plasmon [13,14].

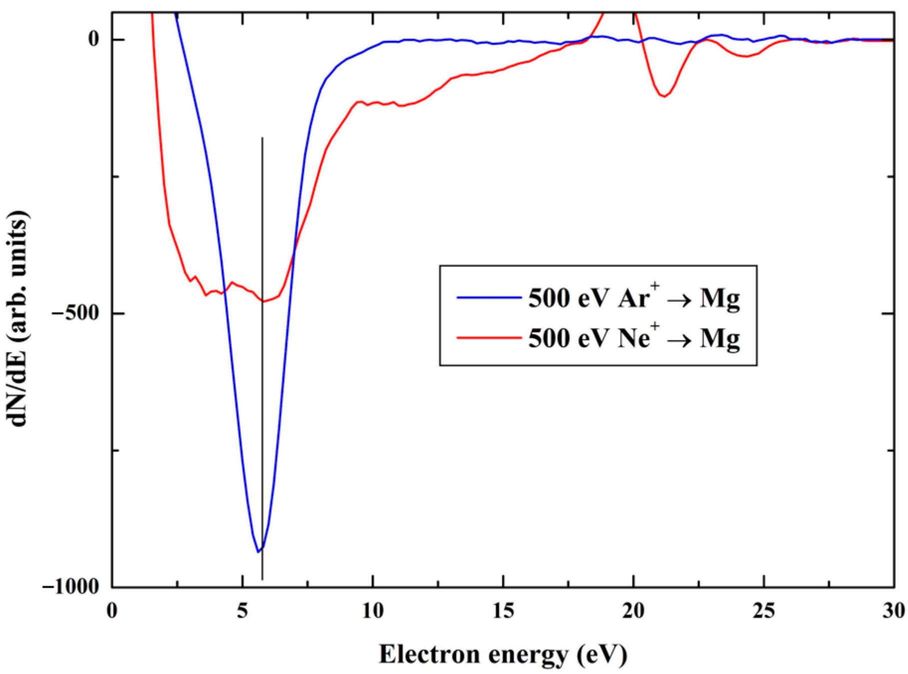

Figure 7. Derivative dN/dE of the spectra excited in Figure 6. The line mark the position of the plasmon dips for both Neon and Argon projectiles.

While in the spectra of electrons emitted by Neon projectiles the spectral feature of Auger neutralization is clearly separated in energy from that due to the decay of plasmons, this is not the case of Ar projectiles. In this case, with a ionization potential I = 15.6 eV, the spectral features due to Auger neutralization and that due to the decay of potentially excited plasmons are expected to be strongly overlapped. Indeed, in the spectra acquired under 500 eV and 1 keV Argon impact on Mg we observe only a minimum in the derivative that could correspond to both the AN and the plasmon processes. To gain insight into the origin of this feature, we performed measurements at varying the incidence angle of the ion beam. These measurements were performed to reveal the broadening behaviour of the spectra with changes in the component of the velocity perpendicular to the surface. These spectra, reported in Figure 8, have been acquired at a fixed observation angle of 30°. To compare lineshapes, the spectra in Figure 8 have been normalized to the same area, which correspond to an electron yield independent of perpendicular velocity [32], which applies to the case of Potential electron emission.

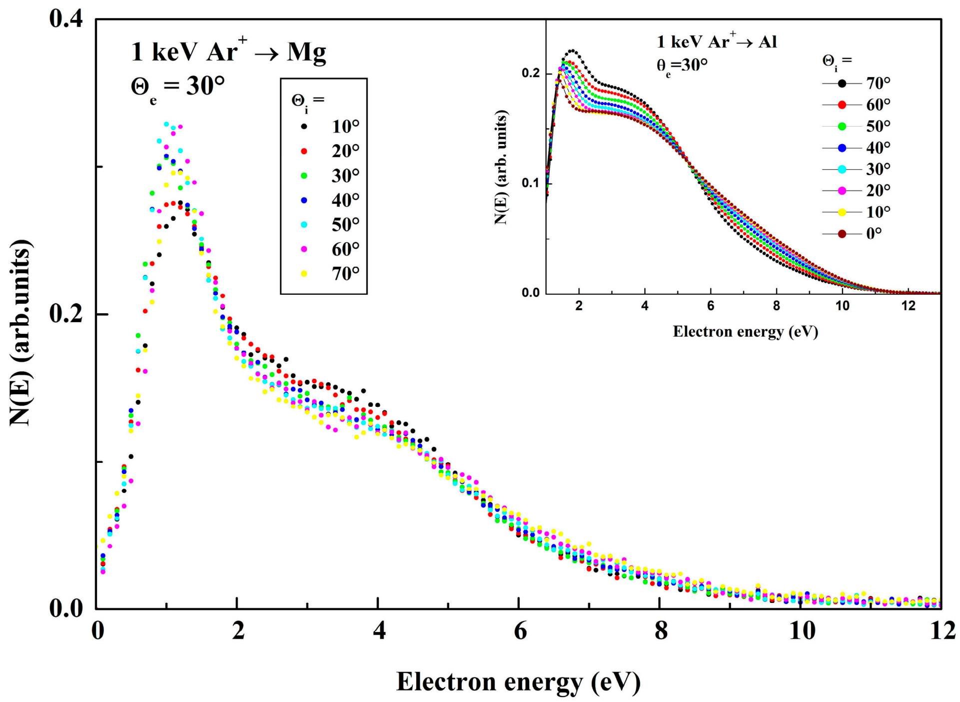

Figure 8. Energy spectra of electrons emitted by the Mg surface by 1 keV Ar^+ as a function of ion incidence angle, for a fixed observation angle of 30°. The inset shows the results obtained in similar experiments on Al [32]. Note the different broadening for Mg and Al samples under the same experimental geometries and impact energy.

The spectra show a peak at low energy that it is assigned to emission of secondary electrons excited by the transfer of the kinetic energy of incoming ions. Superimposed to the secondary electron spectrum we observe the broad feature we want to investigate. If this broad feature superimposed to the KEE peak were due to AN, we should observe the characteristic broadening of the high-energy edge of the spectra, as the incidence direction of the ion beam approaches the surface normal, that is, as the velocity of the ion normal to the surface increases [28,50]. This broadening has been observed in the spectra excited by 1 keV Ar^+ on Al surfaces [32], which are reported for comparison in the inset of Figure 3. The broadening of the electron energy distribution observed for Argon impact on Aluminum is typical of Auger neutralization. It is due to the distance dependent shift of atomic energy levels near the surface of the solids and to the non-adiabaticity of the process because of the finite ion velocity normal to the surface. A typical characteristic of this broadening is that the spectra crosses at a point, as clearly observed on Al surface [50]. Therefore, the spectra in the inset of Figure 8 show that the neutralization of incoming Argon projectiles at Al surfaces proceed via Auger neutralization. This must be evidently attributed to the fact that the energy released in the neutralization of incoming ions is not enough to excite Al plasmons. In contrast, the spectra of electrons emitted by 1 keV Ar^+ on Mg do not show such a broadening. This means that the electron spectrum is not affected by the ion velocity and therefore we assign the spectral feature to a plasmon assisted neutralization. In this case, in fact, the emission of the electrons results from the decay of an elementary excitation of the solid, which is broadened only by the finite plasmon lifetime. Therefore, unlike the Auger spectra excited at Al sample shown in the inset in Figure 8, the plasmon decay feature in Figure 8 excited by Ar ions does not broaden by the finite projectile velocity [32].

4. Conclusions

Analysis of the energy distributions of electrons emitted by Mg under slow noble gas ions impact reveals clear plasmon structures and provide therefore an unambiguous identification of both the AN and the plasmon structures, particularly the last one, that was not resolved in previous studies on Mg using He projectiles. Comparison between electron spectra excited by Neon and Argon reveal similarities and differences that deepen the basic understanding of plasmon excitation in slow ion-solids interactions. In the case of Ne^+ impact on Mg, the plasmon structure is well separated from the high-energy edge of Auger neutralization. The angle-resolved spectra acquired under Ar^+ impact on Mg, allow to clearly identify the structure due to electron emission from plasmon decay, which is more important than that of AN. This lead to the conclusion that that potential plasmon excitation should dominate the neutralization behavior whenever energetically allowed. Consistently with other studies, our experiments reveal the excitation of bulk plasmons in the KEE regime, most likely due to fast electrons travelling inside the solid. The angle resolved spectra in the interaction of 5 keV Ar ions with Mg show that bulk plasmon excitation is determined by the penetration inside the bulk of incoming particles, while the theoretically predicted excitation of bulk plasmon by the transfer of potential energy from projectiles above the surface is not revealed.

Funding: This research received no external funding.

Data Availability Statement: Data are available upon reasonable request.

Conflicts of Interest: The author declares no conflict of interest.

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