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Photon emission from Ar^+–Ag(111) interaction

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Abstract. An attempt to find the radiation from Ag atoms sputtered during ion bombardment as a result of Silsbee's effect was made. In spite of strong corpuscular emission along ⟨110⟩, ⟨101⟩, ⟨011⟩, ⟨112⟩, ⟨121⟩ and ⟨211⟩ directions no photon emission from these atoms was observed within the limits of experimental error. On the other hand, the Ag atoms sputtered from the Ag(111) surface due to random atomic collisions appeared to be a source of intensive radiation. The linear polarization rate of the light equals zero within the limits of error. Some conclusions on the mechanism of excitation are drawn.

1. Introduction

The origin of photon emission from ion–surface interactions is an essential problem of inelastic collision processes and a complex question as well. There exist a number of concepts which can explain how the continuous or linear spectra arise. The particular mechanisms of the excitation are more or less probable depending on experimental conditions. A critical review of existing excitation models was given in [1] and recently in [2]. The present experiment seems to be a good criterion for the reality of particular models. This was the main reason why our experiment was undertaken.

Particularly unclear circumstances concern the excitation of atomic particles sputtered from a surface by an energetic ion beam. Usually, the place at which the sputter originates as well as the events occurring along the particle trajectories inside a solid are not known exactly. This has caused numerous sophisticated, often contradictory concepts to be proposed. However, an exception does exist which enables one to examine the history of atoms during the sputtering process. This regards atoms sputtered from a single crystal to result from a sequence of collisions with focusing of the momenta along the close-packed directions. This phenomenon is widely known as Silsbee's [3] effect. In the present paper an experiment with an Ag(111) surface bombarded perpendicularly by focused Ar^+ ions of energy 9 keV is described. An attempt to find the radiation from Ag atoms sputtered during ion bombardment as a result of Silsbee's effect was made. Preliminary results have already been announced at a workshop on this subject [4]. Measurements of the polarization rate of the radiation were also performed.

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2. Apparatus and experimental technique

The argon ions produced in the Nielsen source were accelerated to an energy of 9 keV and focused on a Ag(111) surface with an electrostatic system of a lens and a quadrupole triplet. The target was mounted on the oven, heated with a tungsten filament to a temperature of a few hundred degrees Celsius and could be rotated around its ⟨111⟩ axis. An ion beam current density of about 0.2 mA cm^{-2} was attained. Because the ion beam usually has a bell-shaped current density profile, a silver diaphragm with an orifice of 2 mm diameter was placed in front of the target to obtain a constant current density within the bombarded spot. This reduced significantly the so-called edge effect, i.e. enhanced photon emission from areas bombarded by the beam of low density.

The base pressure of the residual gases in the collision chamber was maintained at around 10^{-7} Torr. Before insertion into the collision chamber, the surface of the target was thoroughly cleaned and chemically etched. It has been found that the cleaning process during ionic bombardment at an increased temperature is more efficient than the deposition of molecules of the residual gases.

The photon flux from sputtered Ag atoms was measured along an axis perpendicular to the beam direction. Both analogue measurements and photon-counting techniques were used depending on the type of experiment. Photometric measurements of the Ag I, 338.29 nm resonance radiation were performed with a narrow-band interference filter of half-width 13 nm tuned to a 335 nm, photomultiplier (Thorn EMI 9789 QA) followed by an analogue measurement or photon-counting system. The former was based on a lock-in amplifier while the latter consisted of a fast preamplifier, shaping amplifier, amplitude discriminator and pulse counter. The photon-counting measurements were normalized to the same electric charge deposited on the target. For this purpose the target current was processed by means of an amplitude-to-frequency converter and pulse counter.

3. Directional emission of silver atoms from the Ag(111) surface

The angular distribution of the atoms sputtered from single-crystal targets under ion bombardment is quite different from that from polycrystalline targets. Apart from random collision processes resulting in isotropic sputtering, one can observe successive collisions within close-packed directions with momenta focused along a line. When the thus-focused energy packet exceeding the binding energy reaches the surface, the atom which rests at the end of the line can be ejected in the same direction (figure 1). Hence the directional emission of atoms overlaps the isotropic sputtering. Wehner [5] was the first to give experimental evidence for collisions related to crystallographic directions. A number of workers repeated such experiments under various conditions [6], but none of them considered the excitation of directionally emitted atoms. This aspect of preferential sputtering was just what the present authors decided to investigate.

Prior to completion of the experimental set-up for the spectroscopic measurement, the correctness of the sputtering set-up was checked. In order to reveal the directional emission of silver atoms from the Ag(111) surface a typical set-up presented in figure 2 was used. At a distance of 5 mm from the surface a glass plate with a hole 4 mm in diameter was mounted in a parallel manner. This polished plate served as a substrate for condensation of sputtered Ag atoms. As mentioned above, the beam was formed by

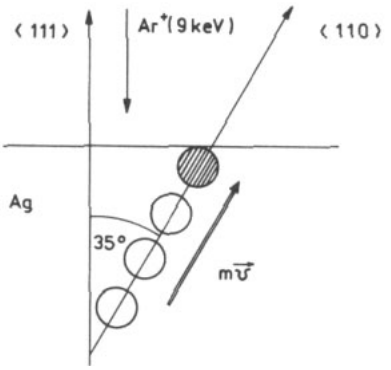

Figure 1. Directional emission from the Ag(111) surface.

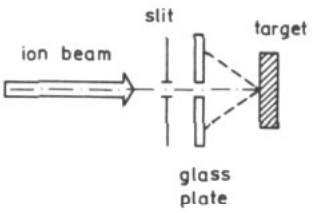

Figure 2. Schematic diagram of the sputtering set-up for the directional emission tests.

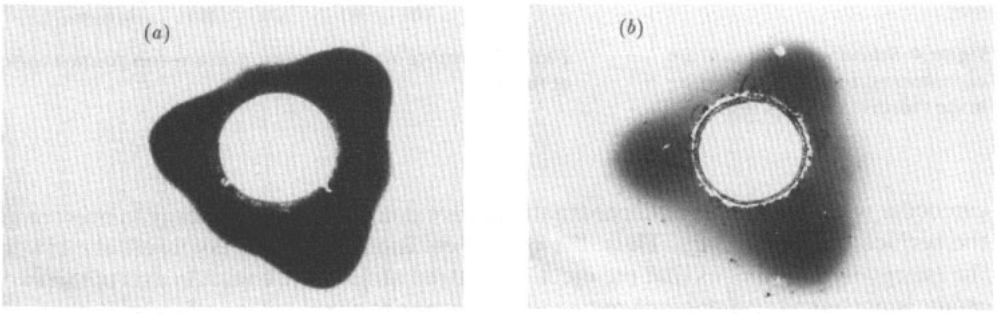

Figure 3. Wehner's spots from Ar^+ bombardment of Ag(111): (*a*) dose density of 8.2×10^{17} ions cm^{-2}; (*b*) dose density a factor of six less.

a silver diaphragm of 2 mm diameter, placed before the glass collector. Thus, the sputtering of glass components over the Ag(111) surface was avoided.

The result of bombardment with a dose of density 8.2×10^{17} ions cm^{-2} is shown in figure 3. Two sets of Wehner's spots are visible, imaged at the ⟨110⟩, ⟨101⟩, ⟨011⟩ and ⟨112⟩, ⟨121⟩, ⟨211⟩ sets of directions in the Ag single crystal. The stronger three spots correspond to the first set of directions which are distributed over the cone with a ⟨111⟩ axis and a vertical semi-angle of 35°. The weaker spots are due to the second set of directions and are probably overlapped by the spots from the ⟨114⟩, ⟨141⟩ and ⟨411⟩ directions. Both sets are rotated by 60° to each other and form a star-like pattern of threefold symmetry. This distinct pattern shows that the molecules adsorbed on the target surface do not perturb the movement of the terminal Ag atoms ejected directionally from the ends of closely packed arrays. The details of the energy transport in the sequence of focused collisions were thoroughly discussed in [7, 8]. It was indicated there that the collision chains in the ⟨110⟩ direction focus at kinetic energies below approximately 30 eV and defocus at higher energies. Assuming another type of interatomic potential, Brinkman [9] has evaluated this critical energy as 87 eV.

Finally, one should remark that another explanation of the occurrence of the spot patterns was given by Lehmann and Sigmund [10]. The concept is based on the assumption that ejection of a surface atom as a consequence of a low-energy collision cascade

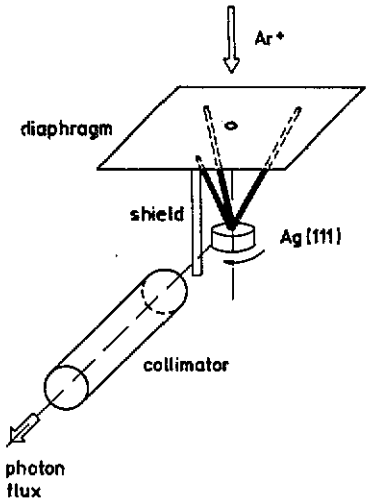

Figure 4. Schematic diagram of the experimental set-up for recording in the radiative emission test.

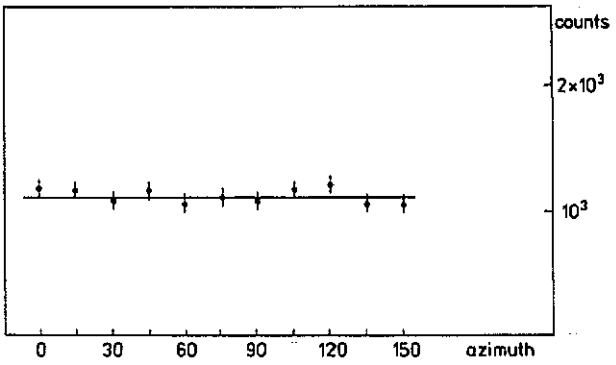

Figure 5. Number of photons per ion dose versus position angle of the target.

can occur when the atoms underneath the surface deliver sufficient energy to overcome the surface binding energy. Thus, the central collisions are more favoured, the lower the energy is. This means that the ejection centred around the common axis of the two atoms is preferred and the spot patterns arise owing to the regular surface structure.

Independently of the point of view, one can conclude that directionally ejected atoms originate from the surface and do not participate in violent collisions induced by primary ions.

Apart from the existing controversy on the character of the focusing mechanism the basic idea of the present experiment as well as the conclusions drawn from it remain the same.

4. Photon emission from sputtered atoms

An attempt to find the radiation from directionally emitted atoms was undertaken by means of the experimental set-up presented in figure 4. The photon flux from the sputtered atoms was directed through a cylindrical collimator to a photomultiplier to eliminate scattered light. In front of the collimator a narrow shield parallel to the incident ion beam was mounted. This was a silver strip 2.2 mm wide stiffly jointed with the diaphragm which formed the ion beam. Thus, neither the beam itself nor the atoms ejected in the plane defined by the optical axis and ⟨111⟩ direction were visible to the photocathode. In this way it was possible to eliminate the photons emitted from particular atomic streams according to the position of the target.

The rotation of the target around the ⟨111⟩ axis should result in periodic amplitude modulation of the photon flux. Taking into account the facts considered in section 3, one should expect less than 33% modulation with an angular period of $\pi/3$. The intensity of Wehner's spots shows that the modulation should still be possible to detect in spite of the presence of isotropic sputtering.

The results of photometric measurements are presented in figure 5. The numbers of single photon counts normalized to constant ion dose deposited on the target were measured at various positions of the Ag single crystal. The target position angle was varied within a 150° range in steps of 15°. It was not possible to extend the range of rotation for technical reasons. The background radiation did not exceed 6% of the ion-induced signal. Against expectation, a constant optical signal within the limits of experimental error, independently of target position, was recorded. It was attributed to the atoms sputtered as a result of random atomic collisions. One can conclude that the focused energy packets transmitted in a series of head-on collisions with almost full efficiency do not contribute to the excitation of the terminal atoms ejected from the ends of the close-packed arrays.

In addition to the above photometric measurements, the linear polarization rate of the bombardment-induced radiation was also evaluated. The experimental set-up was quite similar to that presented in figure 5 with the exception of the shielding system. Instead of a narrow strip a cubic case made of sheet iron foil was used in which the target holder with a Ag single crystal was contained. The inner surface of the case was blackened to eliminate scattered light. Only two small apertures were made in the walls of the case. The first was used to form the ion beam. The second was for the radiation output just in front of the collimator.

Between the filter and the photomultiplier a rotating polaroid was arranged. The polaroid could work in a static or kinetic regime. The first was based on photon counting at two consecutive positions of the polarization plane perpendicular to each other. In the second case the polaroid rotated with a constant frequency that enabled us to detect a modulated photocurrent by means of a lock-in amplifier. Both kinds of measurement showed that the linear polarization of the Ag I, 338.29 nm spectral line, if it exists, does not exceed the rate of 3% that was the limit of experimental error. The present result is analogous to that obtained for other metallic targets [11, 12].

5. Conclusions

The question arises of why the atoms ejected directionally from a Ag single-crystal surface do not emit radiation contrary to the atoms sputtered isotropically due to random collision cascades. Neither of these two groups is distinguished by their energies. They have the following differences.

(i) Directionally ejected atoms do not participate in violent collisions induced by primary ions. Energy packets of less than 30 eV are transferred to the terminal atoms in a series of head-on collisions.

(ii) Chemical interaction between adsorbed molecules and Ag atoms participating in isotropic sputtering can occur as opposed to directionally ejected atoms which apparently are not included in outer molecular systems perturbing the Ag(111) structure.

The experimental data obtained so far do not allow us to decide which mechanism plays the essential role. In any case the models of excitation that are restricted to neutralization or other elementary processes caused by the ejection itself from the solid phase to vacuum are not adequate for the description of photon emission in our experiment.

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