EJECTION OF EXCITED SLOW ATOMS AND MOLECULES OF HELIUM FROM A CARBON FILM PRODUCED DURING BOMBARDMENT OF SOLID TARGETS BY A BEAM OF FAST HELIUM IONS

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Radiation due to slow atoms and molecules of helium was observed when different solid targets were exposed to a beam of 20 keV He^+ ions. It was established that this radiation was emitted by slow helium atoms and molecules ejected from the carbon film formed on the surface of the target as a result of the interaction between incident ions and adsorbed hydrocarbon molecules. A mechanism for this phenomenon is proposed.

INTRODUCTION

WHEN solid targets are exposed to an ion beam there are, in principle, three types of radiation that can be observed: 1) radiation due to excited particles on the surface of the target, 2) radiation due to fast excited particles produced either as a result of momentum transfer to particles on the surface of the target as a result of single collisions with the incident ions or as a result of scattering of the incident ions by the surface of the target, which is accompanied by the excitation of the scattered particles, and 3) radiation due to slow excited particles ejected from the target as a result of cascade momentum transfers from incident ions penetrating the target to particles in the crystal lattice of the target or to particles of the bombarding beam initially imbedded in the target.

The radiation emitted by fast excited particles ejected from the crystal lattice of the target was investigated in[1-3]. Radiation due to the incident beam ions excited as a result of scattering by the surface of the target was observed in[2-3]. Finally, the emission from fast excited atoms produced as a result of the scattering of the incident ion beam by the surface of the target accompanied by the capture of an electron to an excited state was observed in[4-5]. The experimental material on the emission by ejected slow particles is very scanty. Emission by slow excited particles from the crystal lattice of the target during the bombardment of aluminum by Na^+ ions was observed in[5]. Emission by slow Cs particles under the bombardment of Ta, Mo, and W targets by Cs^+ ions was investigated in[3]. Emission by slow excited particles initially imbedded in the target by the bombarding beam was first observed in[7] where it was established that prolonged bombardment of solid targets (C, Ni, Pt, Pd) by an He^+ ion beam (ion energy 20 keV, beam intensity 160 $\mu A/cm^2$) resulted in an emission which was localized very near the target surface. The spectrum of this emission was found to contain molecular helium bands. This unusual result has led us to a more detailed investigation of the conditions for the emission of radiation by slow helium particles from solid targets bombarded by fast He^+ ions. The results of this investigation are reported below.

EXPERIMENTAL RESULTS AND DISCUSSION

An important result of our previous studies of the molecular helium emission[7] is that this emission appears after a long period of time following the initial exposure of the target to the He^+ ions.[1)] This was difficult to understand, especially since the saturation of the surface layer of the target with helium particles should take place in a relatively short time. In fact, if we assume that saturation corresponds to a density of imbedded helium particles approaching the density of the target atoms ($\sim 10^{22}$ cm^{-3}), then the time to saturate the target of thickness equal to the range of the 20 keV (~ 1000 Å) in the particular material should be of the order of 2 min for an ion beam density of $\sim 10^{15}$ cm^{-2} sec^{-1}[2)]. It is found (see below) that the large difference between the time intervals indicated above is due to the fact that the molecular helium emission is due to molecules ejected from the film formed on the surface of the metal as a result of dissociation of adsorbed hydrocarbon molecules under ion impacts, and not by molecules emitted from the bombarded metal itself.[3)] This conclusion was confirmed by experiments in which solid targets (C, Ta, W) were bombarded for long periods of time by He^+ ions in vacuum chambers from which hydrocarbon vapor was efficiently frozen out by liquid nitrogen traps. The carbon film was not present on the target surface under these conditions and, consequently, molecular helium emission was not observed. On the other hand, when the hydrocarbon vapor was not frozen out, a carbon film was found to appear on

[1)]This time was found to depend on the type and temperature of the target, the density of the incident beam, and a number of other factors, and lay between 2 and 10 h.

[2)]The time for saturation was determined experimentally from the time dependence of the current I(t) of secondary He^+ ions ejected from a tantalum target by fast primary helium ions. The experiments were carried out with the apparatus described in [8]. It was found that the time for the current of the secondary He^+ ions to saturate was 2.5 min. This was of the same order as the calculated result.

[3)]The formation of such films under electron or ion bombardment of metal targets in vacua produced by oil diffusion pumps has been observed by many workers. [9,10] We shall refer to such films as carbon films although they probably contain a certain amount of hydrocarbon radicals in addition to the carbon.

the target surface and the molecular helium emission was found to originate in its neighborhood. It can therefore be considered as reliably established that the molecular helium emission reported in[7] does, in fact, originate in the carbon surface film. It follows that the time between the beginning of the bombardment and the appearance of the molecular emission is none other than the time necessary for the formation of the carbon film on the target surface.

Further studies of this helium emission by particles ejected from the carbon film and, in particular, studies of the spectrum of this radiation and the conditions under which it appears and disappears thus become especially interesting.

It is noted in[7] that the very small extent of the radiation producing the molecular helium spectrum indicates that the excited helium molecules have relatively low velocities. It is desirable to establish whether the helium particles ejected from the carbon film include slow excited helium atoms. To show this, it is necessary to record the spectrum not in the direction lying in the plane formed by the axis of the incident beam and the normal to the target surface, which is done in[7], but in a direction perpendicular to this plane.

A number of short bright bands and lines is observed in the spectrum recorded in this direction. Analysis of this spectrum shows that these bands and lines are due to molecular and atomic helium. It follows that when the carbon film is bombarded by fast He^+ ions this results in the ejection of both slow helium molecules and slow helium atoms.

We have carried out certain additional experiments to investigate the properties of the slow helium particles ejected by He^+ ions from the carbon film.

If the target and, consequently, the carbon film are rapidly heated to 400°C at 100°C/sec during the intensive emission by slow helium particles, and without interrupting the ion bombardment, it is found that this emission disappears. As the carbon film cools down from 400°C to the original temperature under continuing ion bombardment, the emission due to slow helium atoms is found to reappear at temperatures of the order of 200°C and this is followed by the appearance of the molecular helium spectrum. A few minutes of ion bombardment is necessary in the case where the carbon film is allowed to cool in the absence of ion bombardment in order to ensure that the emission of slow helium particles will reappear for an initial target temperature of 100°C. Under these conditions, the emission of slow helium atoms is found to appear first and that of molecular helium ions second.

It is also found that if bombardment of the target with He^+ ions is used to produce intensive emission by slow helium particles and then the bombardment of the carbon film by the ions is terminated but the target is kept in the vacuum at room temperature for different intervals of time (between 1 sec and 100 h) the emission of slow helium atoms reappears practically instantaneously as soon as the ion bombardment is continued again. As far as the emission of slow helium molecules is concerned, this is found to appear immediately after the beginning of the ion bombardment if the target is held in the vacuum for not more than 3 h. For longer periods there is a delay in the appearance of emission by the slow molecules which lies between 5 and 60 sec.

The experimental data summarized above lead us to a possible mechanism for the emission by slow helium particles ejected from a carbon film by a beam of fast He^+ ions. As noted above, the emission will appear if the following two conditions are simultaneously satisfied: 1) formation of a carbon film with definite properties on the surface of the target and 2) a high concentration of imbedded helium particles in the surface layer of this film. It follows from the above experimental data that the first condition is satisfied after a substantial interval of time. To satisfy the second condition the carbon film formed on the target surface must be continuously bombarded by He^+ ions for several minutes.[4)] When the target is bombarded by He^+ ions both conditions are simultaneously satisfied because the ion beam does produce a carbon film on the surface and saturates the surface layer with imbedded helium particles. The time necessary for the emission of radiation by the imbedded helium particles under these conditions is determined by the time necessary for the formation of the carbon film on the target surface.

When the intensive emission by impacted slow helium particles is observed there is a dynamic equilibrium in which the current of particles introduced by the He^+ beam into the carbon film is equal to the sum of two currents, namely, 1) the current of imbedded helium particles into the body of the film and 2) the current of helium particles leaving the carbon film for the vacuum. If the second current contains slow excited helium particles, the radiation originating above the carbon film consists of photons emitted by these particles.

The fact that the emission by particles ejected from the solid body can be observed is connected with the probability of the transfer of excitation from the departing particle to the solid body. When this probability is high, emission by the ejected particles cannot be observed. In the opposite case, the density of excited particles in the overall current of ejected particles may be sufficiently high for the emission to become observable.

The probability of excitation transfer from the departing particle to the solid body increases with decreasing velocity of the particle. This is confirmed by the fact that, when metal atoms are ejected under ion beam bombardment, the only particles among the ejected particles which are excited are atoms with sufficiently high velocities.[1-3] This would also appear to explain the absence of radiation by slow ejected helium particles under the bombardment of a pure metal surface by He^+ ions. The presence of this emission during bombardment of the hydrocarbon film by He^+ ions is connected with the particular properties of this film, which ensure a substantially lower probability of excitation transfer between the departing particle and the film as compared with the corresponding probability for the departure of the particle from the pure metal surface. The structure of the hydrocarbon film which ensures a low probability of excitation transfer from the departing particle to the film is quite critical. It turns out that this particular structure will appear only in a relatively narrow base-temperature range (40—150°C), which is

[4)] This time corresponds to a helium-ion beam density of ~150 $\mu A/cm^2$.

indicated by the experiments carried out in this research.[5)] Moreover, the carbon film must have a certain optimum thickness in order to ensure that this structure is such that the excitation transfer probability is a minimum. This follows from the fact that a long bombardment time is necessary to ensure the appearance of the ejected helium particle emission, and from the fact that this emission disappears after a subsequent sufficiently prolonged bombardment. The presence of excited helium molecules among the slow excited particles ejected from the carbon film into the vacuum is obviously explained by the fact that some of the atomic helium particles in the current leaving for the vacuum succeed in recombining while they reside on the surface of the carbon film. As a result, a proportion of the current of helium particles entering the vacuum will consist of excited helium molecules.

There are two possible mechanisms for the ejection of excited helium molecules from the carbon film.

1) Helium atoms in the field of adsorption forces form a stable He^2 molecule in the electronic ground state on the surface of the carbon film. The incident ion beam communicates to the He^2 molecule the necessary excitation energy and the momentum necessary for its escape from the film surface.

2) Helium particles passing through the surface of the carbon film into the vacuum are in the atomic state in the film. Some of these atoms are excited by the ion beam and then the recombination of the excited and unexcited atoms results in the appearance of excited helium molecules.[6)] The momentum communicated to these molecules at the end of a collision cascade due to the penetration of an incident ion into the carbon film enables them to leave the surface.

The proposed mechanism for the emission by slow ejected helium particles can be used to explain experiments in which this emission appears and disappears during the heating and cooling of the carbon film under continuous bombardment by the He^+ ion beam.

The coefficient of thermal diffusion for the helium particles in the carbon film at room temperature is quite small. This follows directly from experiments with carbon films with imbedded helium particles (see above). In fact, if such films are kept in vacuum at room temperature for long periods of time, the emission by slow helium molecules imbedded into them is found to reappear instantaneously as soon as the bombardment starts. Consequently, the number of helium particles imbedded in the carbon film does not decrease substantially with time, i.e. the coefficient of thermal diffusion is low. However, at higher film temperatures the ther thermal diffusion of the helium particles imbedded in the film is no longer negligible. The equilibrium density of imbedded helium particles in the carbon film begins to decrease after termination of bombardment. The reduction in the helium-particle density can be deduced from the delay in the appearance of slow helium-particle emission following the introduction of target bombardment after heating. Maintenance of the carbon film in vacuum at ~300°C for a few minutes is sufficient to ensure that the delay time reaches 2–2.5 min. This means that a few minutes are necessary to ensure that all the helium particles imbedded in the carbon film will escape into the vacuum through its surface while the film is being kept at about 300°C.

The above process of thermal diffusion of helium particles imbedded in the carbon film enables us to explain the disappearance of slow helium particles as the film temperature is increased. As noted above, the equilibrium current of helium particles escaping from the film into the vacuum consists of two components, namely, the thermal diffusion current and the radiation diffusion current.[7)] If the incident beam current is constant, then since the thermal diffusion current increases with increasing temperature there will be a corresponding reduction in the radiation diffusion current. If we now recall that the helium particles transported into the vacuum by the thermal diffusion current have thermal velocities, and assume that all the excited particles in this current transfer their excitation energy to the film as they leave it, then it becomes clear why the increase in the film temperature is accompanied by a reduction in the emission of helium particles entering the vacuum and, finally, why the emission disappears altogether at a certain temperature.

We note in conclusion that the above mechanism for the emission by slow excited helium particles ejected from the carbon film are, of course, only preliminary, and further experimental studies of this phenomenon are desirable. In particular, there is considerable interest in experiments on the relation between the structure and composition of the carbon film and the character of the emission by slow helium particles. One would expect that the phenomenon investigated in this research will also occur when solid targets are bombarded by other ions. We shall perform these experiments in the near future.

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[7)] The phrase "radiation diffusion" is meant to represent the escape of helium particles from the carbon film into the vacuum as a result of the momentum flux introduced into the film by the incident beam.

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