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THE CUT OFF OF LOW-FREQUENCY RADIATION OF EXCITED HELIUM IONS SCATTERED ON METALLIC SURFACES

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A considerable weaking of low-frequency components of the Pickering series has been observed when 3.8 keV helium ions were scattered on metallic surfaces. This effect, brought about by the interaction between excited ions and solid-state plasma, was checked for Na, Al, Cu, Ag and Fe targets, the latter being treated only qualitatively. The magnitude of the phenomenon depends monotonically on electron concentration in the metal. There is a similar effect in the case of intensity ratios in multiplets of atoms sputtered from a target. For the AgI doublet a ratio of 1:1 was observed instead of the expected 2:1 ratio.

1. *Introduction*

Scattering ions of plasmatic energies on solid surfaces gives rise to the generation of a series of products, among which there are photons emitted by the scattered ions and atoms and ions sputtered from the target surface. The spectrum of such radiation has been the object of research in over ten experimental and theoretical studies, and the acquired data primarily have the character of preliminary results. In particular, this is true of the cut off of the long-wave part of the spectrum mentioned in some papers published hitherto, for example, the papers of Kistemaker and associates [1, 2]. These authors put forth the thesis that the interaction of excited ions and atoms with the plasma formed by the electric charge carriers in the target is responsible for this effect. It is the aim of this work to study this effect more closely.

2. *The interaction of plasma with excited atoms and ions*

Due to Coulomb interaction valence electrons in a solid may perform collective oscillations which in character correspond to the vibrations of electrons in a gaseous plasma [3]. The characteristic frequency of these oscillations is expressed by the formula $\omega_p = \sqrt{\frac{4\pi ne^2}{m}}$, where n is electron density, and m and e are electron mass and charge,

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respectively. In the case of interaction between an electromagnetic wave emitted by an atom or ion and a solid-state plasma it can be shown that it is the electrons which bear the main effect on the phenomenon, while the positive ions, because of their large mass, may be treated as a positively charged stationary gas [4]. The solution to Maxwell's equations for the wave interacting with the electronic component of the plasma gives its phase velocity value as being $v_p = c\Big/\sqrt{1-\frac{\omega_p^2}{\omega^2}}$, where ω is the wave's frequency, and c is the speed of light in vacuum. This dispersion formula immediately provides the value of refractive index of the wave in the plasma relative to vacuum, *viz.* $\frac{c}{v_p} = \sqrt{1-\frac{\omega_p^2}{\omega^2}} = \sqrt{1-\frac{\lambda^2}{\lambda_p^2}}$. This in turn shows that radiation of wavelength greater than λ_p will have an imaginary refractive index, which means that the wave will undergo total reflection off the plasma. Hence, if in the process of scattering ions on the surface of a solid excited secondary particles (scattered ions, sputtered atoms) appear, then the radiant relaxation of these particles may encounter some difficulty should they be within range of the target's electronic plasma interaction at the instant of transiting to the lower energy level. In such case, non-radiant relaxation will be more probable. Assuming the plasma to be homogeneous, this would mean a cut-off of the spectrum for waves longer than λ_p. The values of the parameters characterizing the electric plasma in Al, Cu, Ag and Na targets, with which measurements have been performed, are arranged in Table I.

TABLE I

Target material	n [cm^{-3}]	$\gamma = \frac{\omega_0}{2\pi}$ [s^{-1}]	$\lambda_p = \frac{c}{\gamma}$ [Å]
Al	1.809×10^{23}	3.817×10^{15}	785
Cu	0.845×10^{23}	2.613×10^{15}	1 147
Ag	0.586×10^{23}	2.173×10^{15}	1 380
Na	0.254×10^{23}	1.428×10^{15}	2 099

In the calculations it was assumed that the electronic plasma has a density just above the target surface identical with that inside the crystal and that all of the radiating particles are immersed in it. Hence, the boundary should be treated as one in intensity terms. This means that for the luminescing helium atoms in the experiments the intensity of lines for waves longer than λ_p drops with increasing n.

3. *Experimental*

The scheme of the measuring arrangement is shown in Fig. 1. A target adequately purified by chemical means, *T*, was bombarded with a monoenergetic beam of 3.8 keV He$^+$ ions. The ions were supplied by a Penning type source IS with a cold cathode, featuring high stability in time (the change in beam intensity was of the order of 2% per hour) and low operating pressure (of the order of 2×10^{-5} mm Hg). This is of special importance when performing long duration measurements. In this experiment the irradiation time was about 3 hours. The beam of ions was shaped into its final form by means of a quadrupole lens QL and two grounded diaphragms. The ion current at the target was about 0.3 mA/cm^2.

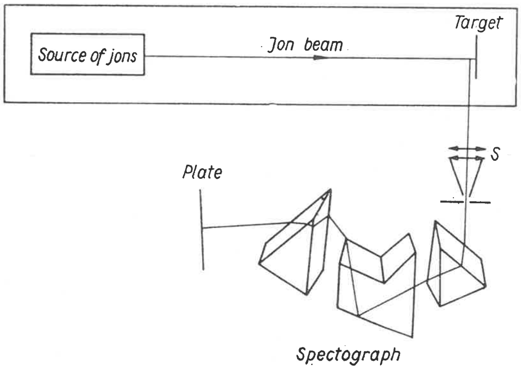

Fig. 1

Observations were made at an angle of $\pi/2$ relative to the direction of the initial beam incidence, which in turn made angles of $\pi/4$ and $\pi/2$ with the plane of the bombarded target in the successive exposures. Each exposure was repeated three times in order to

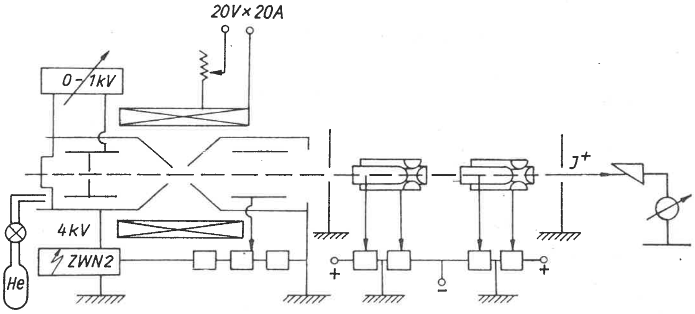

Fig. 2

exclude any chance results. The light emitted by the scattered particles immediately above the target surface was gathered by means of lens L onto the slit S of the Zeiss triple-prism spectrograph incorporating a low-dispersion Raman camera. Detection was carried out with ORWO NP 27 DIN panchromatic film, and every successive exposure giving a spectrum of the particles scattered on the target was recorded on separate pieces of film together with the hydrogen reference spectrum recorded through a Hartman diaphragm. The recordings on the films were performed by means of a self-recording "Lirepho" microphotometer, manuf. Carl Zeiss–Jena. An illustrative example of a microphotogram showing the photometered spectrum of He$^+$ ions scattered on an Al target is given in Fig. 2.

4. *Results*

The films had recordings of the lines of scattered ions and those of secondary ions and atoms sputtered from the target. An analysis of four HeII lines, of which three belonged to the Pickering series, was made. The affiliation of the lines to the same series considerably facilitates discovery of any tendency of changes in the intensity ratios in the spectrum when the type of target is changed. In order to get a strong effect three different targets of very different electronic plasma density were used. Since the intention was rather to demonstrate the general regularities which exist, the authors resigned from applying the tedious method of heterochromatic spectrophotometry and accepted as a measure of line intensity the area of the surface bounded by the line profile recorded by means of the microphotometer. This procedure is well-founded especially in this case, when the intensities of the same line but with a change in target material are compared. The area under the line was found with a planimeter, the intensity of the HeII $\lambda = 4339$ Å line taken as the arbitrary unit.

Table II gives the intensities of the Pickering series components corresponding to transitions from $ng\,^2G$ levels to the $4f\,^2F^o$ level. In the experiment the first member of the Pickering series of wavelength 1.012 μ was not observed as it lies beyond the spectrograph's range. Also, the line corresponding to the transition from $7g\,^2G$ to $4f\,^2F^o$ at a wavelength of 5412 Å was unobserved because the sensitivity of the panchromatic emulsion has a quite deep minimum in this region; apart from this region the sensitivity is fairly constant. Table II also contains the results of measurements for the line of wavelength 4686 Å, which does not belong to the series under consideration and corresponds to the transition from $4f\,^2F^o$ to $3d\,^2D$. Its intensity is also normalized to the intensity of the NeII $\lambda = 4339$ Å line. The inclusion of this line in the data being analyzed is reasonable only when it is assumed that the experimental conditions remain unchanged. Other

TABLE II

Target material	$n \times 10^{-23}$ cm^{-3}	Intensity			
		6560 Å $6g^2G-4f^2F^o$	4859 Å $8g^2G-4f^2F^o$	4686 Å $4f^2F^o-3d^2D$	4339 Å $10g^2G-4f^2F^o$
Na	0.254	9.9	2.7	—	1
Cu	0.845	5.4	1.9	0.9	1
Al	1.809	2.1	1.2	0.4	1

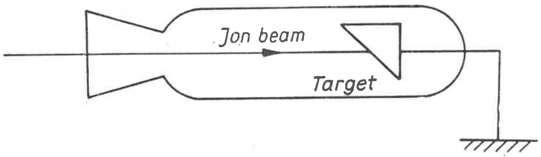

Fig. 3

He II lines recorded in the spectrograms have not been included in Table II because in their immediate neighbourhood there are perturbing lines which made it impossible to compute the area of the profile with a planimeter.

The diagram in Fig. 3 presenting the relative intensities of the HeII lines shows that they distinctly depend on the type of target. It should be supposed that this is directly linked with the density of the electronic plasma in the metal of the target. This dependence becomes clearer as the wavelength increases, that is, as the difference becomes greater between the wavelength emitted by the atoms and the theoretically predicted spectrum cutoff by the solid-state plasma.

This dependence is even more evident in Fig. 4, where the change in relative intensity of each line is presented separately as a function of electron density n in the various targets.

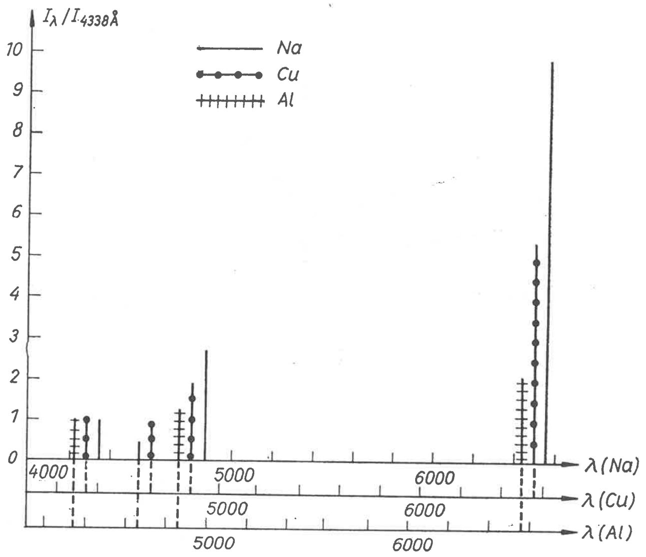

Fig. 4

Excited ions which emit a photon once they are quite distant from the target surface make a contribution to the spectrum identically as free ions would. On the other hand, ions which undergo relaxation within the reach of the solid-state plasma's range of action give a smaller contribution to the long-wave part of the spectrum, and gets smaller as n increases and as the wavelength is longer.

Of the lines of atoms sputtered from the target as analysis was made for the doublet of silver corresponding to transitions from the level $6d\,^2D_{5/2}$ to $5p\,^2P_{3/2}$ and from $6d\,^2D_{5/2}$ to $5p\,^2P_{1/2}$ giving off waves of lengths 4211 Å and 4055 Å, respectively. This was the only multiplet of atoms sputtered from the target recorded within the range of the spectrograph which had lines intense enough and spread apart from the perturbing lines. The ratio of intensities of this doublet's components determined by Terpstra and Smit [5] for arc excitation is 1.8; this is close to the ratio of statistical weights of the lower levels of 2 : 1. In our experiments it was found that the areas under the profiles of the doublet's components are identical within the limits of experimental error (Fig. 5). Since the sensitivity of the panchromatic emulsion slightly increases with wavelength in the wavelength range encompassing the silver doublet being considered, it should be concluded that in this case ratio of the doublet's components surely does not exceed 1 : 1. From the point of view of the theory outlined here this means that the relaxation of the excited states *via* radiant emission is subject to strong prohibition which grows with increasing wavelength. In the case of atoms sputtered from the target this effect of interaction with the solid-state plasma is all the more vivid that the energies of these atoms do not exceed several electron-volts, which makes them stay for some time in the near-surface zone.

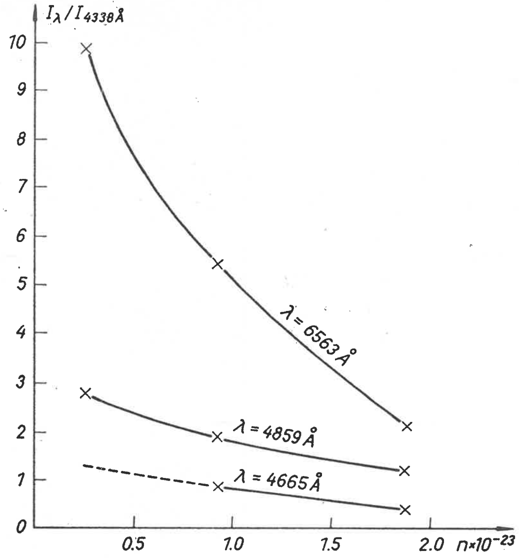

Fig. 5

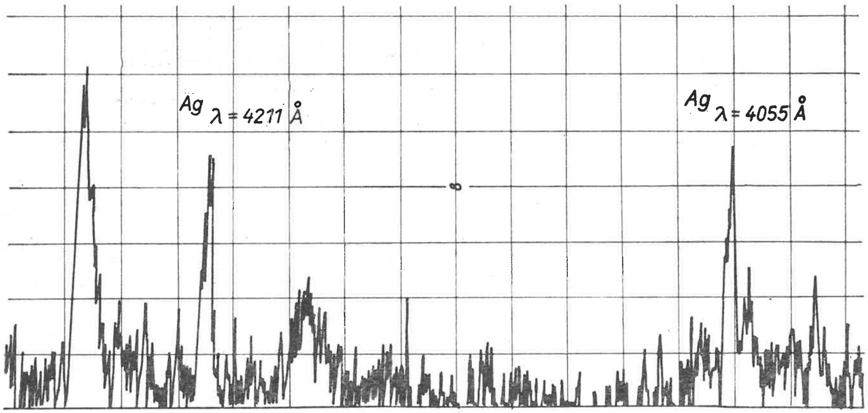

Fig. 6

Finally, it should be mentioned that an analogous experiment was carried out with an iron target. Due to the multitude of multiplets in the entire spectral region this should have provided a lot of data for analysis. The necessity of using a Raman camera of high transmission and relatively low resolving power, however, made it impossible to carry out with sufficient accuracy an analysis of intensities in the obtained spectrum by determining the areas for the various components of the multiplets. Notwithstanding, a rough estimation of the intensity ratios implies that there occur relationships which confirm the findings for the remaining targets.

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