On the Temperature Dependence of Multi-Alkali Photocathodes *

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Measurements of the temperature dependence of the photoemission from multi-alkali cathodes down to 77.4 °K are described. The general shape of the measured characteristics is compared with curves obtained by other authors for cathodes of different origin. It is found that the properties of the cathodes depend on the preparation method. In all measurements, decrease of photoemission with decreasing temperature is a property common to all cathodes if the trivial case of insufficient transverse conductivity can be eliminated. It is shown that this property may be connected with possible emission from acceptor levels in the long wavelength tail range.

Semiconductor photocathodes of the I – V type, which are highly sensitive in the visible spectral range, were first described by Görlich [1]. The most familiar of these is the Cs_3Sb type. Increasing importance is being assumed, however, by multi-alkali cathodes [2] of the $[Cs]Na_xK_{3-x}Sb$ type since the spectral distribution of their quantum yield shifts by about 0.2 eV to the red range. At photon energies of twice the activation energy $h\nu_0$ the quantum yields increase to close on 12% in very much the same way as the photoemission of all the other materials in the far UV range. This is equivalent to a valence band photoemission [3] and explains the high degree of sensitivity as compared with alkali-metal or Cs_2O cathodes. The first experimental results on the quantum yields vs temperature characteristics of multialkali cathodes were published in 1960 [4, 5] and later [6]. The special properties of such cathodes were shown by results of Spicer [7] and by measurements of the temperature dependence of the electrical conductivity and the vector effect [8]. Other data on the temperature characteristics of EMI multi-alkali cathodes were published by Young [9].

In one case highly pronounced maxima of the photoemission were observed in the vicinity of 110 °K at photon energies of about 2 eV [4]. This effect can be used to construct highly sensitive radiation detectors [10] based on thermal sensitization of the photoemission. The differences between results [5] to [9] are not surprising in view of the differences in the temperature dependence of Cs_3Sb cathodes already pointed out by Frischmuth et al. [4] and Miyazawa [11]. By virtue of their composition and preparation method, however, these cathodes should differ less from one another than multi-alkali cathodes.

In the following an account is given of measurements of the temperature dependence of the photoemission from various multi-alkali cathodes of different origin. The purpose is to provide a basis, in keeping with the usual experimental approaches, for further investigations of anomalies in temperature dependence [4]. The measurements were made between 77.4 and 293 °K and in the energy range from 1.4 to 3.1 eV.

I. Method

In the course of this investigation 8 cylindrical photocells with transparent multi-alkali cathodes supplied by Physikalisch-Technische Werkstätten Prof. Heimann, Wiesbaden, and 3 similar cells from Telefunken, Ulm, were studied. The sensitivity of the cathodes was consistently close to the usual maximum value of approximately 200 μA/lumen. The vacuum of the cells was checked by measuring the photocurrent saturation with increasing anode voltage. Two different cryostat arrangements were used for the measurements.

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In the first system the cells were mounted in a transparent-glass DEWAR vessel, the cathode surface being perpendicular. A tungsten lamp and monochromator were used in each case for optical excitation. The exit slit of the monochromator was imaged on the cathode surface with vertical incidence by means of an optical system. The intensity was almost uniform over the entire sensitive cathode surface. The DEWAR vessel and the optical path were shielded from disturbing light by a metal container including sockets for the electrical connections and tubes for the supply of liquid N_2. Prior to each measurement the level of the liquid N_2 was set just below the cathode. Free evaporation of the liquid brought about a very low heating rate for the cathode. The temperature was determined by means of a thermocouple mounted on the glass envelope with the cathode film on the inside.

The second arangement consisted of a stainless-steel cryostat with two concentric cooling systems. This cryostat is normally used for thermoluminescence measurements [12]. The inner system contained the cell with the cathode surface in the horizontal position. Through a tube system comprising a lens and prism the exit slit of the monochromator was imaged directly on the cathode surface. The almost monochromatic light bundle therefore illuminated only a few mm² of the cathode surface with nearly vertical incidence. The outer cooling system served as an almost completely closed thermal shield at a constant temperature of 77.4 °K. The inner system with the reflecting films on its surface was also supplied with liquid N_2. Combining the two cooling systems resulted in extremely low heating rates, which were also measured with a thermocouple.

The investigation procedure for each cell was as follows: First the cathode was cooled down to 77.4 °K. The subsequent heating process was extremely slow since it was produced by retarded evaporation from the inner N_2 system. During this time the spectral distribution of the photoemission current was measured at many temperatures T and at photon energies between 1.4 and 3.1 eV. Because of the low heating rates these cathode temperatures T only increased by less than 0.2 °K during each spectral run.

II. Results

For comparison with previous results [4] the measured quantum yields $Q(\nu, T)$ at each temperature T and each light frequency ν were divided by the quantum yield $Q_{293\,°K}(\nu)$ and plotted in a 3-dimensional diagram. Figs. 1 and 2 give the results for the cathodes H4 (Heimann) and T1 (Telefunken)

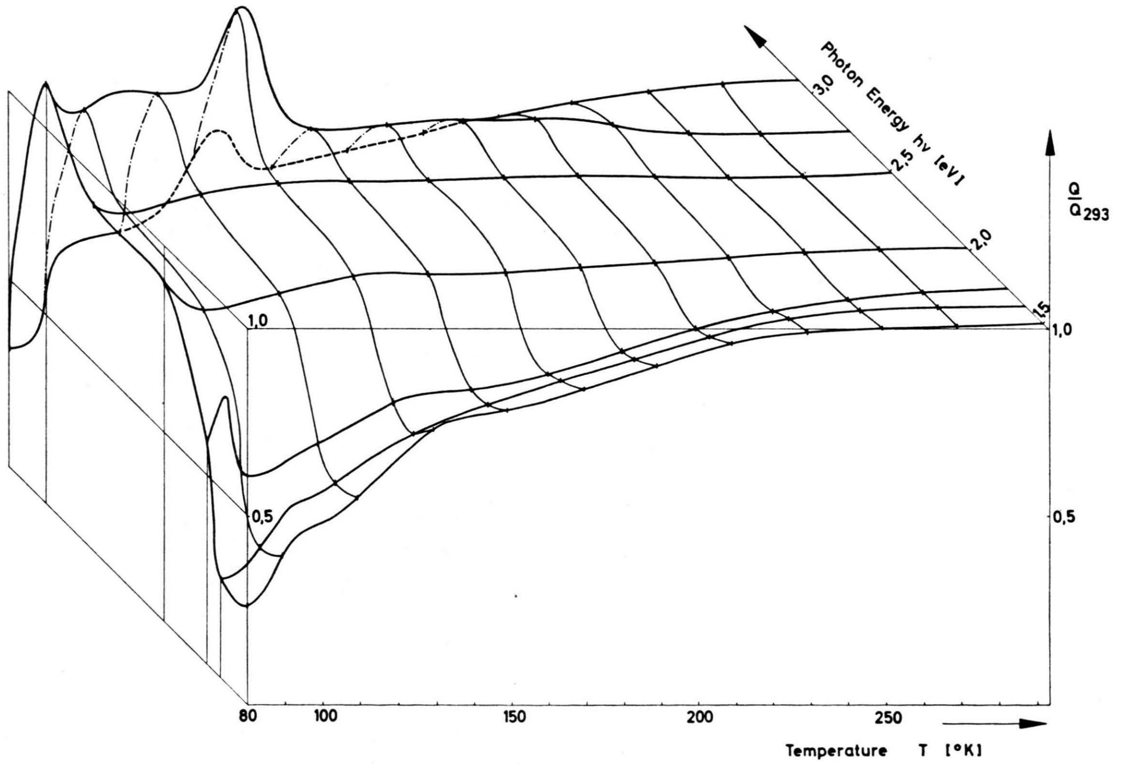

Fig. 1. Spectral and temperature dependence of the quantum yield Q of the Heimann cathode H4 related to the corresponding values at room temperature $Q_{293\,°K}$.

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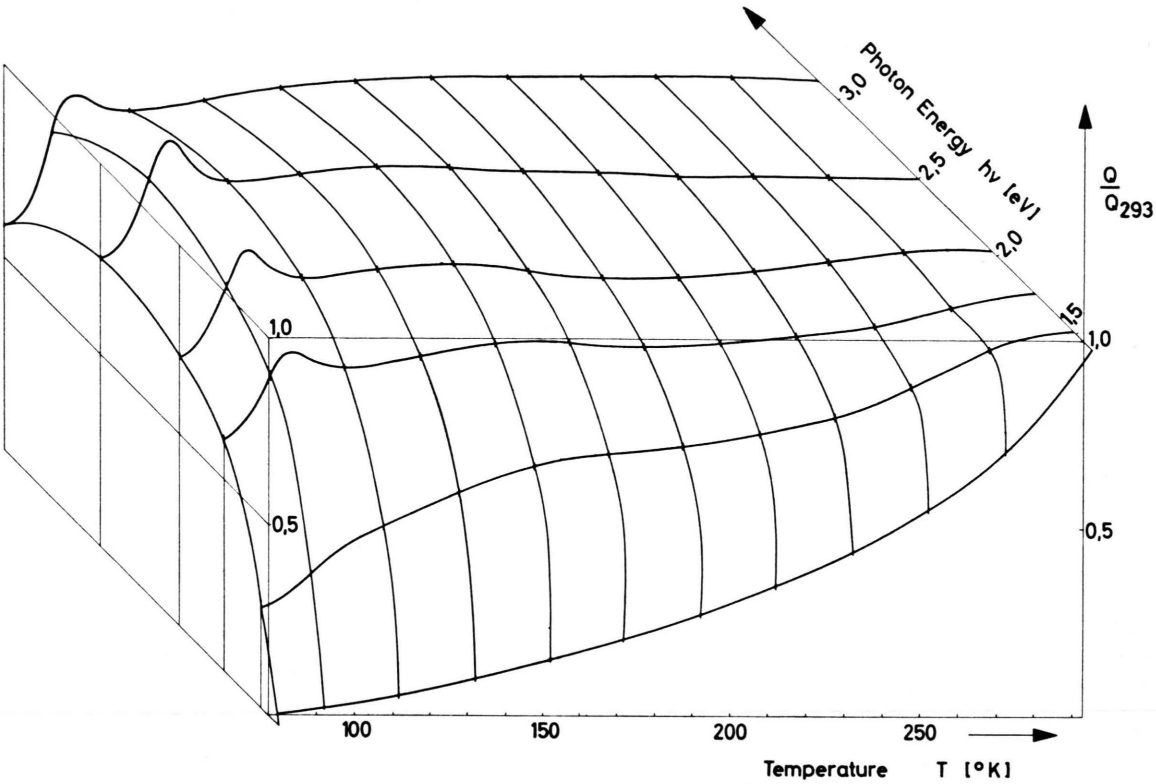

Fig. 2. Spectral and temperature dependence of the quantum yield Q of the Telefunken cathode T1 related to the corresponding values $Q_{293\,^\circ\mathrm{K}}$ at room temperature.

respectively. For the sake of clarity the points in the diagram are plotted at the intersections of the curves of equal frequency with the curves of equal temperature. The individual test points located along the curves of equal frequency are not indicated in these diagrams because they deviate slightly from the plotted curve values in all ranges. This deviation was given by the relation $\pm\Delta(Q/Q_{293\,^\circ\mathrm{K}}) < 8\%$. The scattering here is due to fluctuations of the lamp current. At temperatures of about 80 °K these fluctuations are subject to even more pronounced deviations from a smooth characteristic. This can be seen in Fig. 3, which shows the results obtained with the cathode H5. Repeated measurements showed that all experimental values are reproducible at temperatures above 100 °K within the normal scattering range defined above. The peaks observed previously [4] around 110 °K did not appear. At somewhat lower temperatures, however, similar peaks were observed under the more difficult conditions created by intensity variations. But such variations proved unnecessary in view of experience with the law of linearity between photoemission current and light intensity which was observed by LENARD and formulated by EINSTEIN in his photon theory of light. The results of a more exact study of such peaks encountered in the course of this work are dealt with in a previous paper [13].

III. Discussion of Results

The marked difference between the Heimann H4 cell and the Telefunken T1 version is obvious even from a superficial comparison of the two sets of results. At photon energies between 1.7 and 3 eV and temperatures exceeding 120 °K the temperature dependence of the Heimann cell (Fig. 1) is almost completely constant. At 130 °K and photon energies around 3 eV the Telefunken cell (Fig. 2), on the other hand, gives maximum photoemission, while with decreasing temperature the photoemission is

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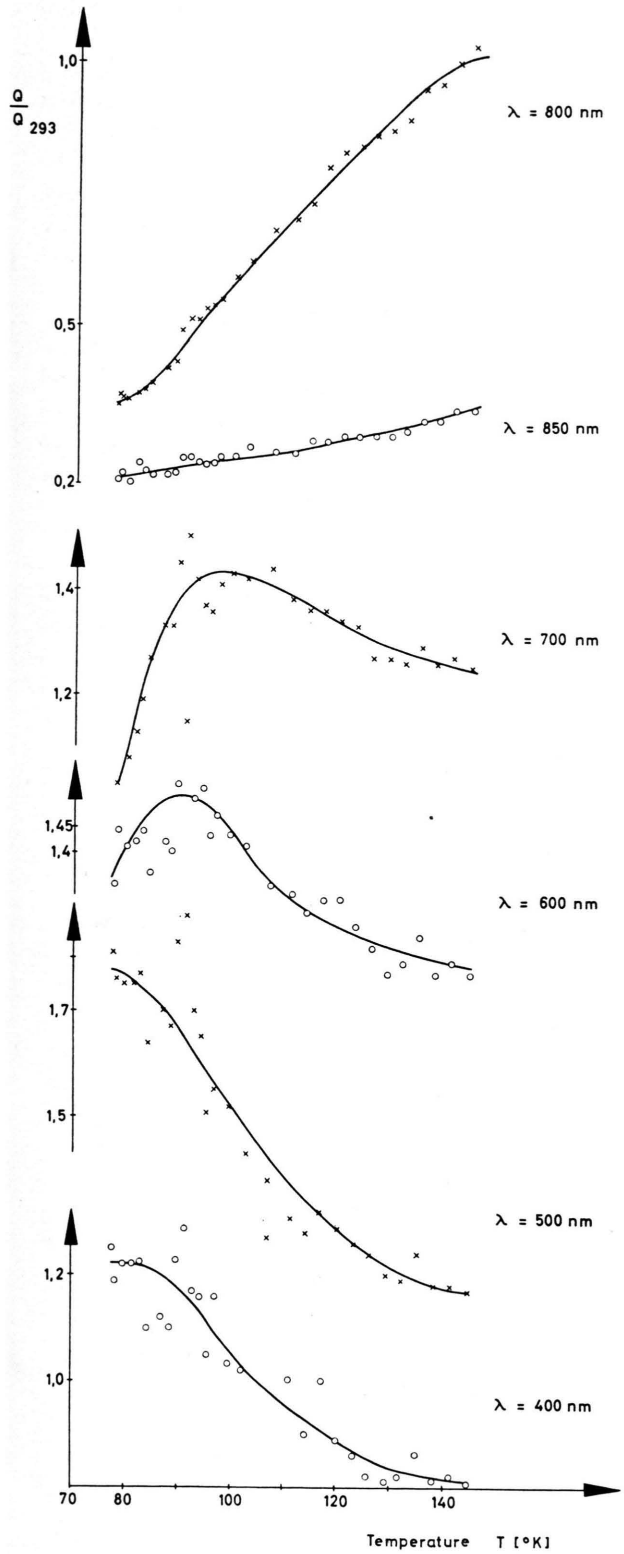

Fig. 3. Spectral and temperature dependence of the quantum yield Q of the Heimann cathode H5 related to the corresponding values $Q_{293\,°K}$ at room temperature.

reduced. The constant temperature dependence affords good agreement with previous results [4] for temperatures above the peaks. OSHEROVICH et al. [6] also obtained a constant temperature dependence of the quantum yields from their second multi-alkali cathode at photon energies around 3 eV and temperatures of down to 170 °K. In the remaining spectral range, however, the temperature behaviour is completely different from that of the H4 and T1 cathodes. The different results for the first and second cells of OSHEROVICH et al. [6] are not surprising when compared with our results. Measurements on EMI multi-alkali cathodes [9] at photon energies exceeding 1.7 eV also contradict the cited results of the present authors and others. In this case an increase of $Q/Q_{293\,°K}$ to a value of 2 is obtained with decreasing temperature. This seems to indicate that there are appreciable differences in the structure of multi-alkali cathodes of different origin. On the other hand, the slight differences between the cells of one particular manufacturer observed in the course of our investigation point to a consistent preparation method for the individual make. Properties common to all cells examined are found for wavelengths exceeding 780 nm. At a wavelength λ of 850 nm an increase of $Q/Q_{293\,°K}$, which is equal to 0.2 at $T = 80$ °K, to values of 1 at $T = 293$ °K was observed in all cases. It is worth noting that the curves of the Heimann cathode are inflected downwards with a slight break at about 170 °K, whereas those of the Telefunken cathodes are inflected upwards. At $\lambda = 800$ nm similar behaviour is observed, but the curves are inflected upwards in all cases. In the longer wavelength range (less than 1.7 eV) EMI multi-alkali cathodes also give the expected decrease of emission with decreasing temperature, in agreement with our results. A similar result was obtained with other I – V type cathodes and, in particular, with a Cs_3Sb cathode in the long wavelength tail range of the photoemission [4]. This is also the case for the results of MIYAZAWA [11] and OSHEROVICH et al. From the observed increase $Q/Q_{293\,°K}$ of the relative quantum yields with increasing temperature it can be concluded that this property obeys a law of general validity.

Altogether different is the behaviour of Rb_3Sb cathodes at the long wavelength limit. With decreasing temperature these show a sharp increase of the quantum yield to values greater by a factor of 10 than that at room temperature [14]. The structure of these cathodes is so different from the type investigated here that a similar response should not necessarily be expected. On the other hand, these measurements on Rb_3Sb cathodes contradict the results of SPICER [7], who observed a decrease of the quantum yield of Rb_3Sb and other I – V and multi-alkali cathodes with decreasing temperature. This agrees with our results.

Some comment should be made on the extent to which reduced transverse conductivity at low temperatures impairs the photoemission. This phenomenon, which was observed in Cs_3Sb cathodes at 80 °K by OSHEROVICH et al. [6] and probably also in the second multi-alkali cathodes of these authors, is known to exist in standard I – V cathodes. In our case and cases [6, 7] and [9], however, the temperature response and the magnitude of the quantum yields prevent such impairment because the transverse conductivity is reduced. In addition, it was experimentally verified that cell saturation was also caused by variation of the anode voltage. From temperature vs conductivity measurements on a Heimann cell [6] it can be seen that the transverse conductivity may be near the critical limit in this particular case. It can be assumed, however, that in the cases under investigation here and in the others cited a sufficiently high defect concentration created sufficient transverse conductivity, in agreement with the experimental result. In the case of the Cs_3Sb cathodes without sufficient transverse conductivity the purity of the cathode materials was obviously too high, although defect concentrations of $10^{19}\ cm^{-1}$ and more are not unknown for the necessarily p-type conducting materials [15]. Because of the complicated methods of preparing multi-alkali cathodes a sufficiently high defect concentration can be expected in most cases.

The decrease of photoemission with decreasing temperature at the long wavelength limit makes it clear that in this narrow range the photoemission does not originate in the valence band, but in the electron states 0.05 to 0.25 eV above the valence band. This is true for both Cs_3Sb and multi-alkali cathodes. Confirmation is provided by the fact that not only does the temperature dependence deviate, but also that matching of the normal spectral distribution of the quantum yield following from a phenomenological theory to corresponding experimental values gives an activation energy $h\nu_0$ assigned to the valence electrons [16]. This energy is found to exceed by the above-mentioned small differences the energy corresponding to the measured frequency limit of the cathodes [17]. Moreover, in the region of this limiting frequency the vector effect of Cs_3Sb decreases from the high values corresponding to the excitation of valence electrons [18] to smaller values corresponding to activity in the long wavelength tail [19]. If the photoemission in the long wavelength tail were to occur at acceptor levels, its decrease with decreasing temperature could not be accounted for in terms of the density of these populated levels because this density increases with decreasing temperature. A complicated excitation mechanism involving interaction with the lattice is responsible for the decrease. This mechanism may then possibly be of a different kind from that for valence electrons [20].

Acknowledgments

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