EXPERIMENTAL INVESTIGATION OF THE TEMPERATURE DISTRIBUTION IN THIN FILMS HEATED BY AN ELECTRON BEAM

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UDC 536.12:537.533.7

The temperature distribution in thin carbon films irradiated in an electron microscope has been measured. The experimental curve is in good agreement with the result calculated by assuming that heat is removed from the object mainly by conduction.

In various electron-optical devices the state of the object under study is changed by the electron beam. One of the important factors is the heating of objects as a result of the absorption of part of the electron energy. Thus under certain conditions the high current densities in electron microscopes may raise the temperature of thin objects several hundred degrees [1-3]. This effect has been used for the direct observation of phase transformations [2, 4].

Data on the temperatures of thin films irradiated by electrons reported by various investigators [3, 5-7] are approximate and generally widely divergent. Accurate measurements of the temperature are difficult because of the nonuniform distribution of electrons in the beam and because the temperatures are different at various points of the object under different heat removal conditions. To a greater or lesser degree earlier measurements of the temperature are averages. The temperature distribution must be known to explain the pattern of thermal changes in the object.

The temperature distribution in objects was calculated in [8-10] by making various assumptions about the relative importance of mechanisms of heat removal. The method of successive approximations appears to be the most accurate [10] but it requires complicated calculations and a knowledge of a number of quantities such as the thermal conductivity which in most cases cannot accurately be estimated for this films.

We have measured the temperature distribution in objects heated by the beam of an electron microscope. Previously the very possibility of making such measurements had been in doubt [10].

Let us consider the radiation conditions characteristic of electron microscopes. The object 1 in the form of a thin film is fastened to a metal screen 2 with holes of radius R (Fig. 1). The electron beam passes close to the center of the hole and does not hit the edge. The screen is fixed in a massive holder which is hardly heated; its temperature remains close to room temperature $T_R \approx 290°K$. After the electron beam is turned on, it typically requires $\sim 10^{-2}$ sec to establish a steady state [10], and therefore the temperature of the object can be regarded as independent of time. If the center of the beam coincides with the center of a hole in the metal screen the temperature distribution in the object depends only on the distance r from the center. The increase in temperature $\Delta T_0 = T_0 - T_R$ is maximum at the center. The problem is to determine the ratio $\Delta T_r/\Delta T_0$ as a function of r.

It is known that electrons emitted by a hot cathode have a Gaussian distribution at the crossover of the electron beam. It was shown in [9, 11] that the form of the distribution function is preserved even after the electrons pass through the diaphragms and condenser lenses. Therefore the current density j_r at a distance r from the center of the beam in the object plane is related to the current density j_0 at the center by the expression

$$j_r = j_0 \exp\left(-\frac{r^2}{a^2}\right), \tag{1}$$

Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 21, No. 6, pp. 1084-1090, December, 1971. Original article submitted November 2, 1970.

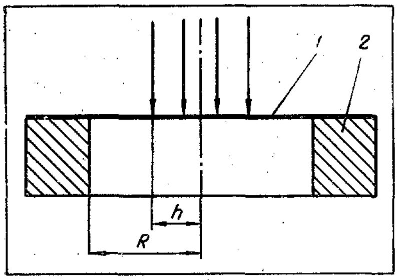

Fig.1

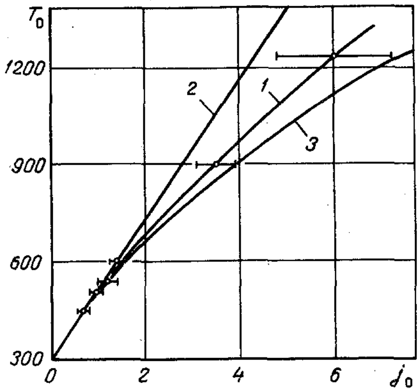

Fig.2

Fig.1. Schematic diagram of irradiation: 1) object; 2) screen.

Fig.2. Temperature at the center of the irradiated portion of an object as a function of the current density at the center: 1) experimental; 2) linear; 3) calculated by Eq. (3). T_0, °K; j_0, rel. units.

where a is a parameter characterizing the half-width of the distribution h ($h = 2a\sqrt{\ln 2}$). Since 94% of the electrons pass through a circle of radius h, the half-width of the current density distribution is simultaneously the effective radius of the beam [11]. The quantities j_0 and h are measured by the method discussed in [11]. The objects were carbon films 20 nm thick fastened to copper screens with circular holes 50 μ in radius. Thin layers of indium, tin, bismuth, lead, antimony, and silver were deposited on these by vacuum evaporation. The melting points of these metals are respectively 429, 505, 544, 600, 903, and 1233°K. For an average layer thickness of about 10 nm all these metals form isolated islands in the deposit on the substrate. Only for thicker layers is a connected film formed as a result of coalescence of the islands and recrystallization of the layer. The metal layers consisting of individual small crystals are convenient indicators of the temperature of irradiated objects since the temperatures of the various films in the free state and with small metallic crystals deposited on them differ slightly under fixed irradiation conditions [3, 6].

A gradual increase in the beam intensity leads to melting of the small crystals, recorded by the disappearance of the diffraction reflections in the dark-field image. In order to prevent the evaporation of the antimony and silver layers, which occurs quite rapidly at temperatures below their melting points, a carbon film about 5 nm thick was deposited on them. It was first established that depositing a thin carbon film on the other metals did not affect the value of the current at which the metal melted.

Because of the spread of the geometric parameters of the small crystals they melt at different values of the current density. It was assumed that the melting point of the layer corresponds to the current density at which the last diffraction reflections vanish in the center of the field of view. The largest crystal sizes were 50-100 nm. For such sizes the thermodynamic effect of lowering the melting point of small particles should be small [12]. Therefore the melting point of the layer determined by the instant of transition of the largest crystals to the liquid state can be considered equal to the melting point of massive samples.

The proposed method of measuring the temperature distribution in a specimen requires a knowledge of the temperature at the center of the irradiated portion as a function of the current density. This relation, obtained by measuring the current density producing melting of layers of various metals, is shown in Fig.2. In performing the measurements, the half-width of the current density distribution remained constant at 8 μ and the accelerating voltage at 80 kV. The initial portion of the experimental curve is nearly linear, and for $T_0 \lesssim 900$°K the deviation from linearity does not exceed 20%. In this temperature range the results are very reliable. At higher temperatures there is a greater spread in the measured values of the current density, leading to melting of the metals, possibly because of the beginning of graphitization of the carbon films [3]. Henceforth we consider temperatures below the melting point of antimony (903°K). In most practical cases the temperatures of irradiated objects are lower than this.

To test the relation found for the temperature as a function of the current density $T_0 = f(j_0)$ we made multilayered objects by depositing layers of tin and antimony on opposite sides of a carbon film. The presence of the second metal has only a small effect on the current at which the first metal melts. This confirms

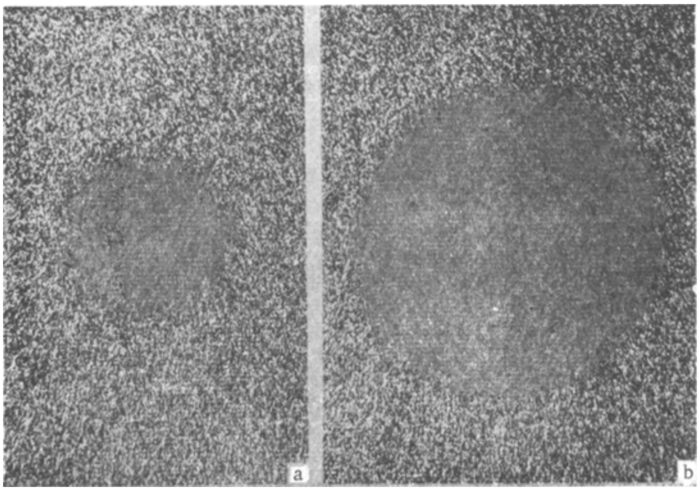

Fig. 3. Enlargement of melting zone of tin with increasing current density. The zone boundary is the isotherm $T = 505°K$; a) $j_0 = 0.40\ A/cm^2$; b) $j_0 = 0.43\ A/cm^2$. Magnification (×9000).

the previous assumption [3] that very thin metal layers deposited on various objects do not significantly affect their temperatures in an electron beam. The ratio of the current densities $j_0(Sb)/j_0(Sn)$ leading to melting of antimony and tin for a constant beam diameter is about 3.7, which is 1.3 times as large as it should be for a linear dependence of the temperature on the current density. This value agrees with the results shown in Fig. 2 within the limits of experimental error.

The deviation of the experimental relation $T_0 = f(j_0)$ from linearity can be explained by the increased importance of radiation as a heat removal mechanism at higher temperatures (3). For a steady state the amounts of heat gained and lost by the object are related by [6]

$$j_0 = A\Delta T_0 + B(T_0^4 - T_R^4), \tag{2}$$

where A and B are factors determining the contributions of conduction and radiation to the heat loss. These factors cannot be calculated because the parameters characterizing the thermal properties of a thin film are indeterminate. However their ratio in the present case can be estimated from the measurements. Let us assume $j_0(Sb)/j_0(Sn) = 3.7$. This condition can be satisfied by setting $B = 4 \cdot 10^{-10}\ A \cdot deg^{-3}$. Then

$$j_0 = A\left[\Delta T_0 + 4 \cdot 10^{-10}(T_0^4 - T_R^4) \cdot deg^{-3}\right]. \tag{3}$$

It is clear from Fig. 2 that the experimental curve for $T_0 = f(j_0)$ lies between the straight line and the curve determined by Eq. (3). In calculating the temperature distribution in the object we assume as a first approximation that $j_0 = A\Delta T_0$ and later correct for the deviation from linearity at higher temperatures.

The temperature distribution was measured directly in layers of tin, chosen because of its low melting point, small evaporation rate, and relatively high oxidation resistance. A current density $j_0^{(0)}$ causes melting of small tin crystals at the center of the irradiated portion. This value of the current density corresponds to an object temperature of $T_0 = 505°K$ or a temperature rise $\Delta T_0^{(0)} = 215°K$. An increase in the current density causes an enlargement of the melting zone of the metal (Fig. 3), the boundary of which is obviously the isotherm $T_r = 505°K$. Suppose the current density $j_0^{(r)}$ corresponding to the radius r of the melting zone of tin leads to an increase in the temperature at the center by $\Delta T_0^{(r)}$. The small ellipticity of the zone is due to the astigmatism of the condenser and can be taken into account in measuring the average value of r. For a linear dependence of the temperature on the current density we have

$$\Delta T_0^{(r)} = \Delta T_0^{(0)} \frac{j_0^{(r)}}{j_0^{(0)}}. \tag{4}$$

Taking into account that the temperature rise ΔT_r at the zone boundary is equal to $\Delta T_0^{(0)}$, and omitting the superscript on $\Delta T_0^{(r)}$ referring to an arbitrary value of r we rewrite Eq. (4) in the form

$$\frac{\Delta T_r}{\Delta T_0} = \frac{j_0^{(0)}}{j_0^{(r)}}. \tag{5}$$

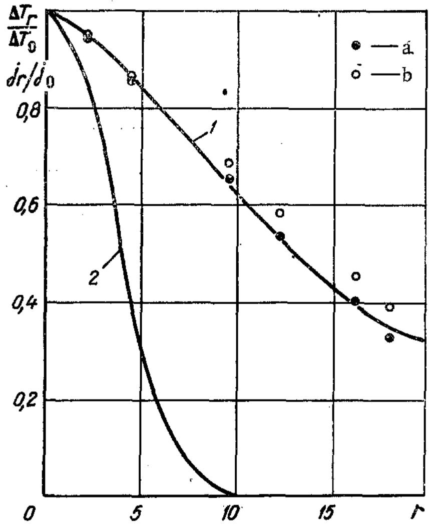

Fig. 4. Temperature distribution (1) and current density (2). The solid curves are constructed by using Eqs. (6) and (1). The points are values of the relative temperature drop at a distance r from the center calculated from the experimental results: a) by assuming that ΔT_0 varies linearly with j_0; b) by taking account of the deviation from linearity determined by Eq. (3). $\Delta T_r/\Delta T_0$ and j_r/j_0 are dimensionless ratios; r, μ.

The left-hand side of this expression represents the required temperature distribution under the assumption that the relation $T_0 = f(j_0)$ is linear. By determining the breadth of the melting zone of the metal it is possible to find the distance r from the center at which the temperature rise of the irradiated object is $j_0^{(0)}/j_0^{(r)}$ times smaller than the temperature rise at the center. The current density $j_0^{(r)}$ was measured with a Faraday cup and an electrometer amplifier, and the corresponding value of r was determined directly by dark-field photographs.

Equation (3) was used to take account of the deviation of the relation $T_0 = f(j_0)$ from linearity. The correction was found by numerical calculations. For example, suppose tin melts at the center for a certain current density $j_0^{(0)}$, i.e., $\Delta T_0^{(0)} = 215°K$. For $j_0^{(r)} = 2j_0^{(0)}$ when tin has melted in a circle of radius r, the temperature rise at the center should be $\Delta T_0^{(r)} = 393°K$, as is easily seen from Eq. (3). This value is approximately 9% smaller than the value calculated by assuming a linear increase in temperature with current density, and this means that the ratio $\Delta T_r/\Delta T_0$ for a given value of r is correspondingly larger.

Gale and Hale [9] found an expression for the radial temperature distribution in an object irradiated in an electron microscope by assuming that conduction is the only mechanism of heat loss from the object. This expression for the current density distribution, given by Eq. (1) for R > h, when the electrons do not strike the metal screen, has the form

$$\frac{\Delta T_r}{\Delta T_0} = \frac{2\ln\frac{R}{r} + \mathrm{Ei}\left(-\frac{r^2}{a^2}\right)}{2\ln\frac{R}{a} + C}, \tag{6}$$

where $\mathrm{Ei}(-r^2/a^2)$ is the exponential integral $\mathrm{Ei}(-x) = -\int_0^x \exp(-t)t^{-1}dt$ tabulated in [13]; C is Euler's constant, $C \approx 0.58$.

$\Delta T_r/\Delta T_0$ as a function of r, given by Eq. (6), is evaluated for $R = 50\ \mu$ and $h = 8\ \mu$ (or $a = 4.8\ \mu$) (solid curve of Fig. 4). Figure 4 also shows two sets of points obtained from the experimental results by assuming a linear relation between ΔT_0 and j_0 and using Eq. (3). These points mark the upper and lower limits of the possible values of $\Delta T_r/\Delta T_0$, since the true values of the temperature lie between curves 2 and 3 of Fig. 2. It is evident from Fig. 4 that the experimental points lie close to the theoretical curve, and therefore the temperature distribution in an object can be estimated with sufficient accuracy by using Eq. (6). The error in determining ΔT_r is no more than a few percent of ΔT_0. The measurements were performed in a temperature range up to $\Delta T_0 \lesssim 600°K$. Equation (6) becomes more accurate for lower temperatures since $T_0 = f(j_0)$ approaches a linear relation as the temperature falls.

We note that the temperature falls off from the center to the edge of the beam much more slowly than the current density does. Thus for $r = h = 8\ \mu$, $j_r \approx 0.06\ j_0$ and $\Delta T_r \approx 0.7\Delta T_0$. Therefore parts of the object not directly irradiated may also experience appreciable heating. This fact can be used to distinguish between thermal damage to the object and radiation damage produced by the ionizing effect of electrons and occurring only in the radiation zone.

Gale and Hale [9] considered the use of Eq. (6) only for solid metal foils having high thermal conductivities. We have established that this relation can also be used for thin carbon films having a thermal conductivity estimated in [14] to be about two orders of magnitude smaller than those of metals. It follows from [3, 5, 6] that the temperatures of various dielectric films (carbon, collodion, quartz, etc.) are nearly the same under conditions of constant irradiation. Therefore the temperature distributions in them must be similar. Thus the temperature distributions in various thin films, including both metals and dielectrics, can be calculated with sufficient accuracy by Eq. (6), at least for temperatures $T_0 \lesssim 900°K$.

NOTATION

R	is the radius of a hole in the screen supporting the object;
r	is the distance from the center of the irradiated portion;
T_R	is the temperature of the screen;
T_0	is the temperature at the center of the irradiated portion;
T_r	is the temperature at a distance r from the center of the irradiated portion;
ΔT_0	is the rise in temperature of the object at the center when irradiated;
ΔT_r	is the rise in temperature of the object at a distance r from the center when irradiated;
α	is a parameter related to h;
h	is the half-width of the current density distribution in the object plane;
A, B	are factors determining the contributions of conduction and radiation to the heat loss from the object (Eq. (2));
C	is Euler's constant.

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