

NEW MEASUREMENT INSTRUMENTS AND METHODS

The quasi-chromaticity method in field-ion microscopy

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The main trends in the use of the quasi-monochromaticity method in field-ion microscopy are considered, as well as the existing means of its practical realization for the separation of the investigated phenomena. Certain difficulties of the method are considered. New trends in the application of the method are reported, particularly for the investigation of the kinetics of continuous field evaporation, for the study of the initial stage of image formation in the field-ion microscope, in the analysis of surface particles, and others.

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The real field-ion image of the surface of a metallic sample—a field emitter with curvature radius $\sim 500 \text{ \AA}$ —consists of about 10^5 points, each of which corresponds to the image of a single atom projected on the fluorescent screen of the field-ion microscope.^[1] A visual (without some additional technique) determination of a non-correspondence between the atomic details of two or more field-ion images, for example those representing different stages of the investigated process, is therefore not feasible in many cases. Of great importance in the comparison and interpretation of a series of field-ion images of the changing field-emitter surface is the use of quasi-chromaticity—a method of artificially coloring the images with the aid of color filters.^[1] The main purpose of this method is to separate the investigated effects from the overall spectrum of phenomena that cause the appearance of some contrast in the black-and-white field-ion image. This method is developing in two directions:

1) Superposition of two or more black-and-white microphotographs of field-ion images (foreign workers use negatives as a rule) projected on a common screen through different filters.

2) Production of single-color (but still colored) microphotographs through filters of various colors directly from the field-ion microscope screen coated with a colored luminor. In the second method photographs are taken of different stages of the process. It presupposes also subsequent superposition of one-color microphotographs and production of quasi-chromatic field-ion images—in three or more colors.

The first trend was first proposed by the developer of the field-ion microscope, Müller,^[3,4] and realized later in a number of studies of radiation defects in metals^[5] (of particular importance is the use of this method when the same field-emitter surface is recorded with irradiation directly in the field-ion microscope^[6,7]), self-diffusion,^[8] adsorption, desorption, and surface migration of gas molecules in a strong electric field,^[9-11] and in the study of alloys.^[1] The second came into use relatively recently in the studies of van Oostrom^[12] and Schubert^[13] (the latter used different imaging gases to investigate formation of films on the surface of the field emitter). Its application is described in a paper by Brandon and Melmed^[14] on the analysis of the structure of the dislocation core in tungsten.

Brandon and Melmed, analyzing the possibilities and the main difficulties of the quasi-monochromaticity method, distinguished also one more trend, different from the first one only in that motion pictures are first taken

of the field-ion images; their superposition is effected not by a double projector, but sequentially. Among the methodological elements pointed out in that paper, notice is taken of the following:

a) mechanical stability of the apparatus used to superimpose the field-ion images;

b) drying of the film and its distortion both in the course of photographic development and following possible heating in the projector;

c) establishment of a coordinate system common to two field-ion images;

d) change of the local magnification for the atoms when their ambient changes;

e) displacement of the atoms during their evaporation by the field.

The first three points are clear; even if they cannot be fully allowed for in the experiment, they can in any case be estimated with sufficient accuracy. The last two phenomena are interesting not only from the point of view of the method, although allowance for them is highly necessary in many field-ion microscopy investigations (for example, in the study of the distribution of particles of various components by sites^[15]). One can expect an analysis of the change in the dimensions of the images of individual atoms (or complexes) as a function of their position (atomic surrounding) on the surface of the field emitter not only to increase the reliability of the interpretation of real field-ion images and reveal some new possibilities in this direction, but also help understand better how the image itself is produced in the field-emission microscope. To carry out this analysis we used, besides photometry of black-and-white field-ion images, also the quasi-chromaticity method (Fig. 1). Its use in this case, while not offering obvious advantages, yields very illustrative pictures and permits an estimate of the possible contribution of this effect in the observed distortion of color contrast (the appearance of "halos" of different color around the points) of quasi-chromatic field-ion images. Figure 1 was obtained by superposition of two field-ion images of a section of the surface of a protuberance on a tungsten field emitter, recorded in the course of continuous field evaporation.^[16] The photography, as in most cases considered below, was with an RFK-5 motion picture camera. To increase the brightness we used a three-chamber electron-optical converter of type UM-92.^[17] The exposure time t_e and the sample potential U_0 are indicated in the figure caption; the approximate magnification in all cases was $\sim 5 \times 10^6$. The imaging gas was helium, and the temperature of the field emitter during the analysis was

¹⁾The terminology used here is borrowed from [2].

78°K. The green spot on Fig. 1 corresponds to the image of a certain atom *a* in the case when another atom, *b*, produces no image (the spot indicated by arrow 2 is not seen on the first image). The transition of the atom to a position where its image can be obtained (during the continuous field evaporation of the surface atoms of the field emitter) causes a sharp decrease in the dimensions of the image of atom *a* (black spot against a green background—indicated by arrow 1). We note that in addition to the change in the dimensions of the image of the atom *a* a change takes place also in the position of its (image) "center of gravity."

The use of the quasi-chromaticity procedure to determine the relative adsorptivity a' in different crystallographic faces is illustrated by Fig. 2. The initial field-ion images corresponded to a surface purified by an electric field (first) and the same surface coated with atoms and molecules of residual gases (second). The adsorption was effected by lowering the sample potential, for a short time, by a certain amount ΔU_c . Variation of the exposure time Δt of the emitter at $U = U_0 - \Delta U_c$ made it possible to vary in a wide range the number of adsorbed particles, while variation of the difference ΔU_c made it possible to limit the type of adsorbed particles. The method of calculating a' is described in [10]. The quasi-monochromaticity method makes it possible to separate in this case unambiguously the images of the adsorbed particles from those of the matrix atoms, thus leading to a noticeable increase in the accuracy of the sought values of a' , and also facilitates greatly the very procedure of processing the images.

Another quite effective, in our opinion, application of the quasi-chromaticity method is in the analysis of the type of surface particles. [18] It is known [19] that one of the most important problems in field-ion microscopy is the determination of the type of particle responsible for the appearance of a particular spot or group of spots on the image. The exceptionally effective and sufficiently accurate probe analysis method [19, 20], which requires among other things rather complicated and expensive equipment, cannot determine simultaneously the nature of several spots (for example, all the spots with increased brightness) in different positions on the analyzed surface of the emitter. The reason is that the evaporating pulse removes not only the "probed" (taken to be the probing aperture) atom or complex. At the same time, in a number of cases such information concerning one and the same surface is greatly needed. The gist of the analysis method proposed in [18] is to superimpose on the emitter potential short-duration pulses of ever increasing amplitude $n \cdot \Delta U_b$ (n is the number of the pulse) over and above the value U_0 corresponding to the conditions of best surface-image production, and taking motion pictures of the field-ion images during the intervals between the pulses. The obtained series of $N = [(U_{\text{evap}} - U_0)/U_b] + 2$ black-and-white field-ion images are used to prepare by the quasi-chromaticity method $(N - 1)$ quasi-chromatic field-ion images (slides), the pair for each quasi-chromatic field-ion image being made up of neighboring exposure frames (Fig. 3). The slides are then measured with semi-automatic measuring apparatus [11] in which the coordinates of the points of definite color (green in the case of Fig. 3) are punched on cards. The method calls for the performance of calibration experiments (their number depends on the purpose of the investigation and may be small), wherein the indicated information is accumulated for many known substances adsorbed

by (sputtered on) the surfaces of the materials. The final calculations are with a computer in accord with a special program.

We wish to call attention also to the quality of the field-ion quasi-chromatic images shown in Figs. 2 and 3 from the point of view of their interpretation (measurement with the apparatus, visual analysis). The quasi-chromatic field-ion image of Fig. 2 was obtained by superposition with negatives, whereas in the case of Fig. 3 previously prepared positives were used. It appears that the second method yields more satisfactory results. In the case of Fig. 3, by suitable choice of filters, it was furthermore possible to attenuate considerably the contrast from the matrix, as a result of which the final contrast permitted identification of the crystallographic faces but did not interfere with the data reduction.

The described applications of the quasi-chromaticity method (with the exception of the case corresponding to Fig. 1) have one common feature: the analyzed field-ion images (both black-and-white and single-color) are obtained in a stationary regime—at $U = U_0$. All the manipulations that change the surface (with the exception of the irradiation of the emitter during its ion-microscopy analysis) are carried out in the intervals between the exposure. We report here the use of this procedure in a study of the kinetics of field evaporation in a continuous regime. As shown in [16, 21], such investigation not only enable us to determine certain important parameters of the process itself, but also to establish the relative binding energies of the atoms with different coordinations on the surface of the field emitter. The initial information in this case consists of the time dependence of the diameters d (more accurately, the dimensions in two mutual perpendicular directions) of the displayed edges of definite crystallographic planes. Besides the difficulties connected with the performance of the experiment itself, it is necessary here to cope with one more difficulty—in the measurement of the field-ion images and the analysis of the result. The point is that when motion pictures are taken of the field-ion image, images of atoms that are displaced from their sites in the lattice, but not yet fully removed, can appear on the frames. They can then be represented on the image not only by bright spots shifted relative to the initial ones, but also by smeared spots. It is precisely to this effect that an appreciable fraction (depending on the ratio of the rate of field evaporation and the rate of image registration) of the points of the inclined sections of the experimental plots of $d = f(t)$ are due (t is the time). [16] The quasi-chromaticity method makes it indeed possible to account for this effect, and also to carry out more accurate measurements of the difference Δd for two images (not necessarily from two successive frames). Examples of two quasi-chromatic field-ion images obtained for three successive black-and-white field-ion images are shown in Fig. 4. We investigated the kinetics of continuous field evaporation of pure tungsten. The prints were obtained from positives of field-ion images through different (for Figs. 4a and 4b) filters. "Single" colors correspond on the figures to atoms evaporated in the intervals between exposures, and to atoms that turned out to be as a result in image positions. A "combination" color indicates displacement of the atoms during the course of the field evaporation. Without going into details, we note that in the analysis of such quasi-chromatic field-ion images (Fig. 4) account is taken also the possible effect of afterglow of the lum-

FIG. 1. Change in dimensions of image of single surface atoms with changing atomic environment ($t_e=1/33$ sec, $U_0=9.9$ kV).

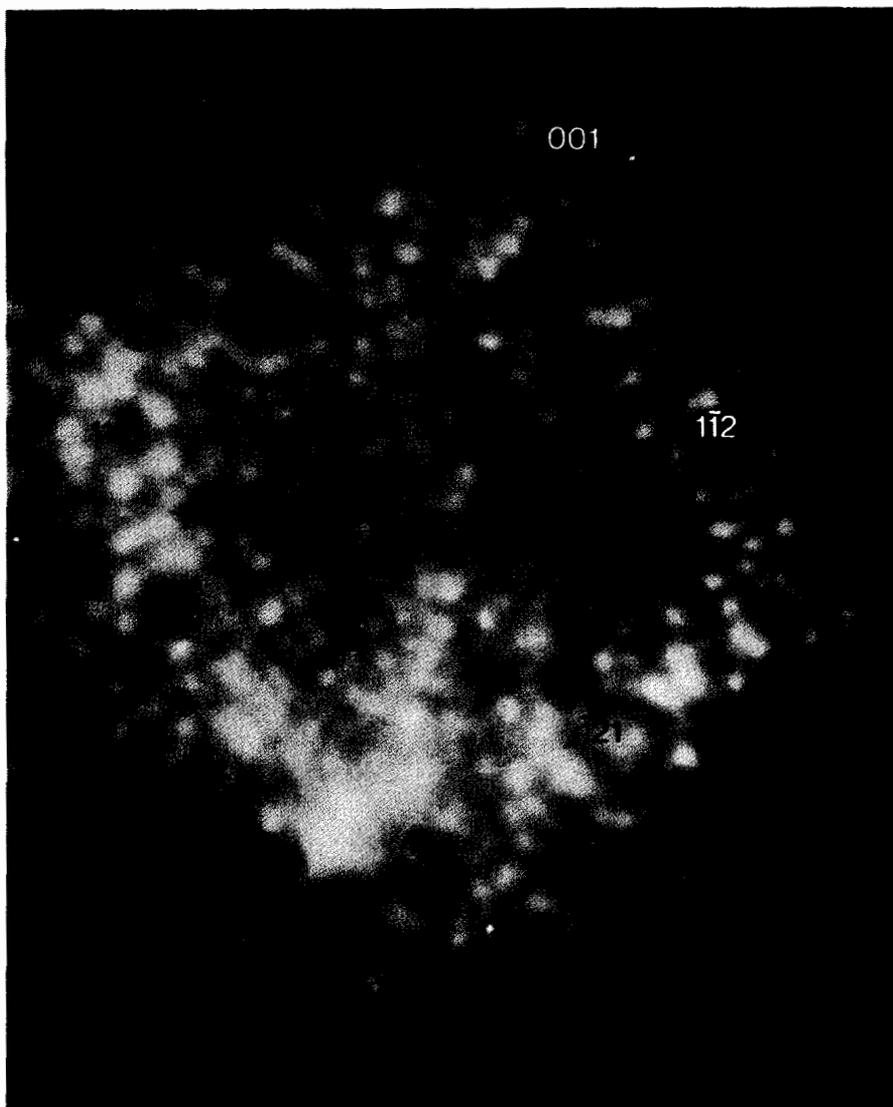
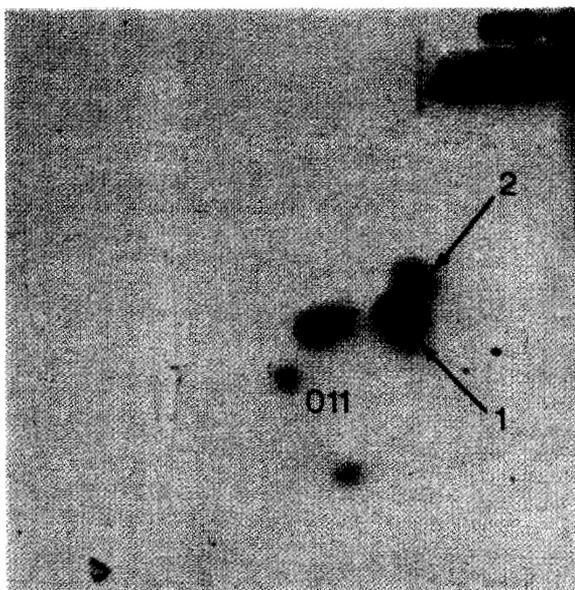


FIG. 2. Field-ion image of tungsten surface covered with adsorbed residual-gas atoms (green points) ($t_e=1.0$ sec, $U_0=9.4$ kV).

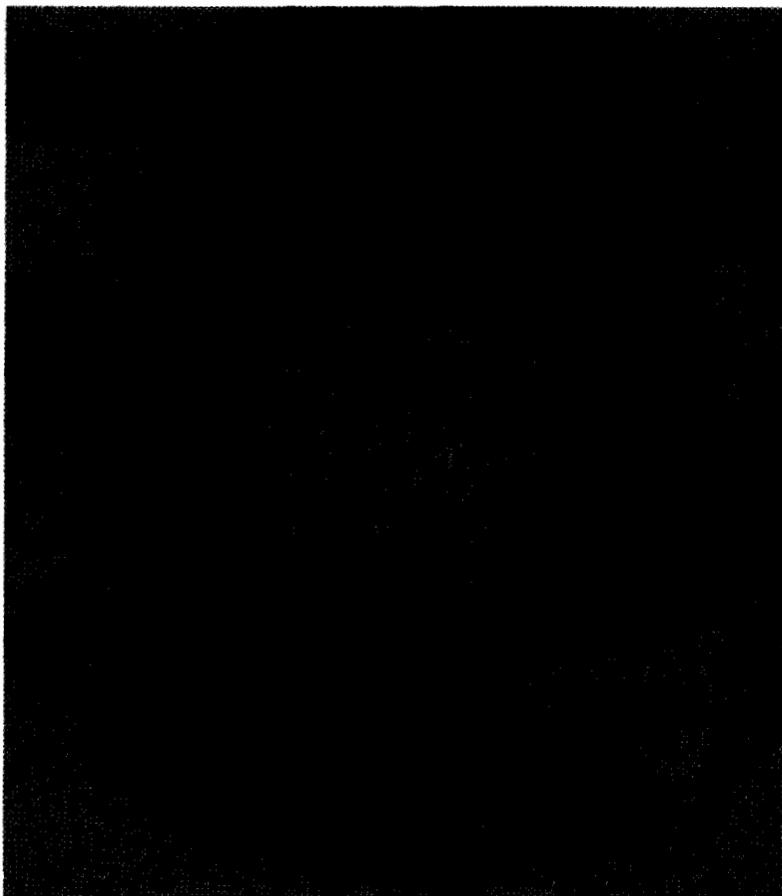


FIG. 3. Change of surface as a result of a pulsed increase of the sample potential. The green points correspond to remote foreign atoms and the red ones to the remaining atoms ($U_0=11.5$ kV).

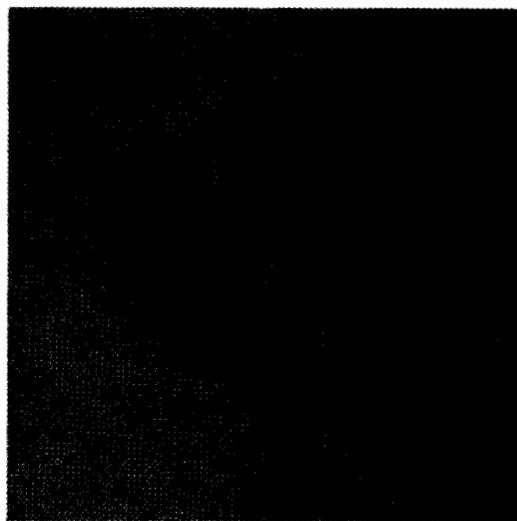


FIG. 5. Initial stage of formation of field-ion image of tungsten sample ($t_e=0.5$ sec, $U_0=14.0$ kV).

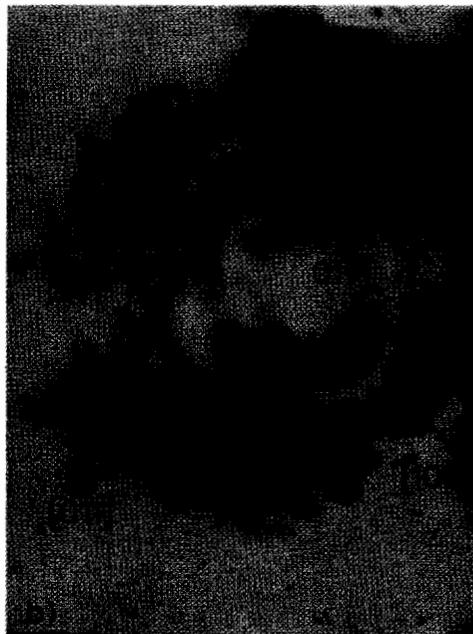
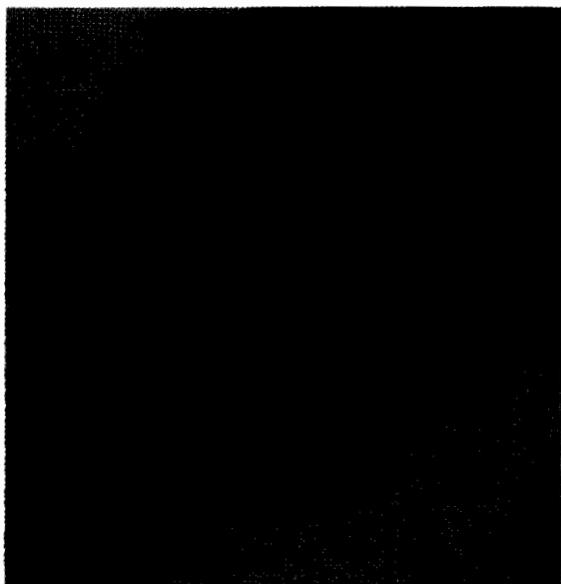


FIG. 4. Kinetics of continuous field evaporation of central (011) face of tungsten sample ($t_e=0.5$ sec, $U_0=9.9$ kV). Superposition of successively exposed frames 1 and 2 (a) and 2 and 3 (b) through different filters.

inors of the microscope and of the electron-optical converter, and also the possible superposition of images of atoms from neighboring parallel planes.

Another interesting, in our opinion, use of quasi-chromaticity method is for the study of the initial stage of the formation of the field-ion image (Fig. 5). We have in mind the fact that the sample potential is "instantaneously" raised from zero to U_0 (which is determined beforehand for the given field emitter) and remains constant, while the photography is carried out continuously with the motion-picture camera. Depending on the degree of surface contamination by the adsorbed particles of the residual and image gases (i.e., depending on the degree of preliminary rarefaction and the time interval elapsed after the removal of the voltage U_0 from the sample during the preceding stage of the experiment), one contrast or another is gradually formed on the microscope screen. If the contamination is negligible, then the crystal structure of the sample begins to "show" through the characteristic "flicker" of the picture on the screen; an analysis of the kinetics of its development will provide a better understanding of certain essential details of the field-ion image formation.

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