Properties of luminescence radiation excited in a small volume by an electron beam

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We describe a stroboscopic procedure in a scanning electron microscope (SEM) operating in the cathode luminescence (CL) regime. The use of a definite phase of the CL pulse to construct the image has made it possible to increase the spatial resolution by more than one order of magnitude. The method was used to investigate a crystal of molybdoscheelites.

Stroboscopic electron optics makes it possible to receive a luminescence signal from a small volume. The linear dimensions of the volume should be much smaller than the length of the light wave. This makes it possible to increase the locality of the optical information obtained from a bulky object.

The optoelectronic signal can be received with a scanning electron microscope (SEM) operating in the cathode luminescence (CL) regime. This optical signal serves (a) to obtain the image[1] or (b) to extract the light for spectral diagnostics with an optical monochromator.[2] The resolution (which is determined by the dimensions of the luminescent region) is usually of the order of several tenths of a micron, i.e., the same as in an optical microscope.[3] In the SEM operating in a secondary electron emission (SEE) regime, resolution better than optical has been attained long ago.[4]

We consider in this paper the mechanism whereby the cathode luminescence is produced and the conditions that make it possible to approach the resolution attainable with SEE. These conditions are determined by the following: (a) the radius of the "illuminating" electron beam, (b) the character of the local electron excitation, (c) by the times of the line scanning τ_p and of the luminescence emission τ_s. Condition (a) requires that the dimensions of the luminescent region be small in comparison with the light wavelength. This is done by using an electron probe of small cross section. Usually the radius of the probe ranges from 5 to 200 Å. This condition is necessary, but not sufficient, since the region of the recombination emission, which determines the resolution, increases with time as the result of the diffusion of the electrons and holes. It will be shown in connection with the discussion of condition (c) how this difficulty can be overcome. We proceed to condition (b). To prevent superposition of the luminescence from individual spots, it is necessary to excite the light not simultaneously, but in sequence. V.K. Arkad'ev[5] called attention to this possibility of suppression of optical diffraction phenomena. In the SEM the points of the object are excited in sequence, but it is still necessary to take into account the time ratio τ_p/τ_s, which can be much smaller (first case, fast scanning) or much larger (second case, slow scanning) than unity. In the

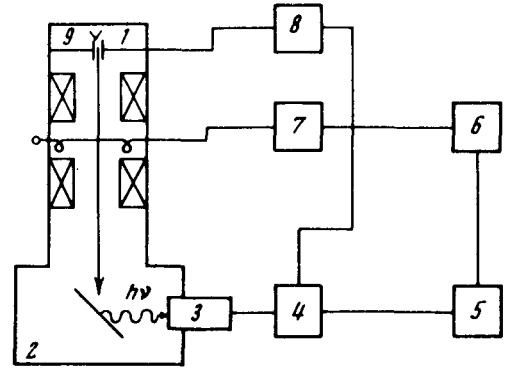

FIG. 1. Functional diagram of CL-stroboscopic system.

FIG. 2. Cathode-luminescence image of scheelite with improved resolution.

usual use of CL and SEM for observation,[3] an error of the "loop" type appears for the first case (see Fig. 10 of[6]), since the probe has already left the point at which the luminescence was excited, but its emission is "ascribed" to another point. In the second case, there is a considerable loss of the resolution. Let us consider our assumptions with respect to condition (c). Knowing that the times τ_s lie in the range $10^{-8} \leq \tau_s \leq 10^{-3}$ sec[3] and assuming a suitable value for τ_p, we can expect the CL to run through the high-resolution phase. The resolution is then lost because of the delay of the recombination of the diffusing particles.

Thus, to separate this phase it is necessary to use in the SEM a stroboscopic device[4] with an electronic gate (Fig. 1). The sequence of the periodically repeating operations is the following: (a) a pulsed electron beam generating electrons and holes is turned on for a definite time, (b) the gate is at first closed and then opens for the required time with a possible shift relative to the start of the particle generation pulse. Multiple automatic repetition of operations (a) and (b) enables us to separate the phase of the phenomenon of interest to us, and also to ensure a sufficient intensity of the CL. Figure 1 shows the observation system (1-SEM column, 2-working chamber, 3-photomultiplier, 4-gate circuit, 5-amplifier, 6-video control device, 7-sweep generator, 8-generator of short strobing pulses, 9-device that cuts off the illuminating electron beam).

Other parameters of the system are the duration of the electron-beam "illuminating" pulse, which is set by the rise time of the luminescence pulse, and the frequency f of the repetition of these pulses. It is clear that it should be given by the condition $1/f \geq \tau_s$. The point is that although the gate circuit does not pass a strongly "delayed" luminescence, it is necessary nevertheless to await the deexcitation of the object before the next pulsed beam of electrons is turned on. This causes a loss of the average CL intensity, which is partially offset by using the stroboscopic regime. Figure 2 shows a luminescent image of a scheelite crystal, obtained by the described method. The useful magnification is $M = 11\,400$. When compared with the secondary-emission image (Fig. 3), Fig. 2 shows good agreement. The described procedure was used by us to observe the structural details of a number of crystals, previously unobservable by optical means. We note that a luminescence spectrum obtained with high resolution (Fig. 2) is of interest when it comes to revealing the chemical composition of the object. An x-ray microanalysis, which yields information only on the presence of various elements, has so far a resolution on the order of several tenths of a micron. To check on the assumption that separation of the luminescent radiation can ensure high resolution, we have obtained calculated estimates based on the one-dimensional diffusion equation. The calculations confirmed the obtained value of the experimentally observed resolution.

It may happen that large deexcitation times do not limit the CL resolution. This occurs when luminescence of small particles (compared with the diffusion length) is excited. We have observed this by exciting the luminescence of lunar regoliths in the color CL regime.[7] The boundaries of these particles are rapidly reached by the produced pairs, which recombine and radiate, or else give up their energy at the boundaries to nonradiative processes. The emission region is then strictly limited, resulting in a better spatial resolution of the CL.[1,3]

The method proposed by us can be regarded as an artificial limitation of the emission region, which improves the spatial resolution of a continuous object. We note that waiting for the "delayed" luminescence to attenuate, so as to eliminate the "loop," turns out to be a useful factor in the observation of CL of dielectrics (scintillators, biological objects, minerals, etc.). Then, owing to the long wait, there is time for the charge due to the electron beam to leak off.[8]

Thus, the stroboscopic electron microscopy previously proposed for the study of temporal processes[4] is also effective for an increase, by approximately one order of magnitude, of the spatial resolution of optical systems.

FIG. 3. Image of the same section (Fig. 2) of the sample in SEE.

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