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NOISE AND MODULATION EXPERIMENTS ON SATURATED CATHODOLUMINESCENCE FROM WILLEMITE

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Synopsis

Measurements of the spectral noise density and a.c. modulation response of saturated cathodoluminescence from polycrystalline Zn_2SiO_4–Mn (Willemite), are compared with a theory developed by Fijnaut and Zijlstra for the case of characteristic luminescence involving one relaxation time. It was found that, with increasing degree of saturation, the crossover frequencies of the excess noise spectrum and a.c. modulation response are shifted towards higher values, while the plateau value of the excesss noise spectrum decreased with increasing degree of saturation. This behaviour was in qualitative agreement with the theory. A quantitative comparison was not possible because of the occurrence of more than one relaxation time in the luminescence under study. The degree of saturation of the luminescence was varied by varying the focussing of the electron beam incident on the phosphor, while keeping the total beam current constant.

1. *Introduction.* In this paper we are dealing with characteristic cathodoluminescence[1]) under saturation conditions. If the density of the exciting electron flux is increased all available luminescence centres may become excited. Further increase in the electron flux density does not change the luminescence intensity: we have full saturation. The degree of saturation may be indicated by the ratio of the number of N excited to N_c available centres: N/N_c.

The influence of saturation on fluctuations in cathodoluminescence has already been described by Fijnaut and Zijlstra[1]). They have found that the spectral noise density in the luminescence radiation, in excess of full shot noise, should decrease as $(1 - N/N_c)^3$. Some remarks, however, have to be made concerning the assumptions leading to their result. They have assumed that the saturation of luminescence is uniform over the light emitting volume of the phosphor. Bethe[2]), however, has derived theoretically that the excitation density (number of excitations per cm^3) within the phosphor is a function of space. This implies that also the degree of saturation is space dependent. In first approximation, however, we shall deal with the simplified case of uniform saturation.

It must be noted here that saturation of luminescence can be experimentally achieved in two ways (at constant primary electron energy): first by increasing the incident electron flux, without change of focus; secondly by focussing the electron beam while the total flux is kept constant. In our noise measurements we preferred the second method, since in this case the beam current in the cathode-ray tube as well as the photo-electric current in the photo-detector can be kept at a low level. In this way, undesirable contributions of the $1/f$ component to the noise in the two currents can be avoided.

Experiments were made on a polycrystalline Zn_2SiO_4–Mn phosphor (Willemite) deposited on the screen of a MG13–38 Philips projection television tube. This phosphor appeared to be suitable for saturation experiments, because its luminescence is characteristic[4]) and its relaxation time[4]) is large enough to reach saturation easily and small enough to perform noise measurements with reasonable precision in the audio-frequency range.

2. *Theory.* For the actual number N of excited luminescence centres we can write:

$$\frac{\mathrm{d}N}{\mathrm{d}t} = g - r, \tag{1}$$

where t is time and g and r are the excitation and de-excitation rates respectively. For characteristic luminescence the de-excitation is monomolecular, so that $r = \alpha N$, where α^{-1} is the lifetime of the excited state. Following Fijnaut and Zijlstra[1]) the excitation rate can be described by $g = \gamma\phi(1 - N/N_c)$, where ϕ is the incident electron flux on the phosphor and γ the number of excitations per primary electron for $N/N_c \ll 1$.

The spectral noise density $S_{\Delta r}(f)$ of fluctuations in r around a steady-state value $\bar{r}$, is according to ref. 1 given by:

$$S_{\Delta r}(f) = \frac{2\bar{r}(1 - \overline{N}/N_c)^2}{1 + \omega^2\tau^2}\left[\langle\gamma\rangle(1 - \overline{N}/N_c) - \frac{2\overline{N}/N_c}{1 - \overline{N}/N_c}\right] + 2\bar{r}, \tag{2}$$

where $\tau = \alpha^{-1}(1 - \overline{N}/N_c)$, $\omega = 2\pi f$, f stands for frequency, while the brackets denote an ensemble average and the horizontal bars the time average. It must be noted here that downward transitions induced by photons as well as by electrons and self-absorption are neglected, while it is assumed that $\langle\Delta\gamma^2\rangle = \langle\gamma\rangle$[3]).

If, on the average, a fraction p of the downward transitions is radiative and only a mean fraction λ of the emitted photons is converted into photoelectrons in a phototube, the spectral noise density of the photocurrent is:

$$S(f) = e^2\lambda^2p^2S_{\Delta r} + 2ei_a(1 - \lambda p), \tag{3}$$

where i_a is the mean photocurrent ($i_a = e\lambda p\bar{r}$), and $-e$ the electron charge. If $S_{\Delta r}(f) = 2\bar{r}$, eq. (3) reduces to the well-known shot noise formula:

$$S_0 = 2ei_a. \tag{4}$$

From eqs. (2), (3) and (4) we now find for the so-called relative excess noise $[S(f) - S_0]/S_0$:

$$\frac{S(f) - S_0}{S_0} = \frac{p\lambda(1 - \bar{N}/N_c)^2}{1 + \omega^2\tau^2}\left[\langle\gamma\rangle(1 - \bar{N}/N_c) - \frac{2\bar{N}/N_c}{1 - \bar{N}/N_c}\right], \tag{5a}$$

with

$$\tau = \alpha^{-1}(1 - \bar{N}/N_c). \tag{5b}$$

Inspection of eqs. (5) tells us that for $\omega\tau \ll 1$ the low-frequency value of the relative excess noise for $\bar{N}/N_c \ll 1$ is $p\lambda\langle\gamma\rangle$. The plateau value, however, decreases with increasing value of $\bar{N}/N_c$. Considering only the crossover frequency $f = 1/(2\pi\tau)$, where the excess noise is half its plateau value, it can readily be seen that with increasing value of $\bar{N}/N_c$, the crossover frequency shifts towards higher values [see eq. (5b)].

To calculate the squared a.c. response of the luminescence to sinusoidal modulated excitation we refer to Fijnaut[3]). It can be proved that the frequency dependences of squared modulation response (SMR) and relative excess noise are the same. Consequently the SMR should also show a shift in the crossover frequency with increasing saturation coefficient. In addition, if the saturation is achieved by focussing the primary electron beam, while the other experimental conditions are not changed, also the plateau value of the SMR should decrease.

3. *Experimental arrangement and results.* The experimental setup for measuring excess noise and modulation response has been described earlier[1]). In the experiments reported here, an EMI vacuum phototube, type 9715 with S 20 photocathode, was used as photodetector, instead of a photomultiplier, in view of the light intensity from our phosphor. No optical filter was placed between phototube and phosphor. As mentioned in the introduction saturation was achieved by focussing the electron beam incident on the phosphor. The experiments reported were performed at primary electron energy of 10.5 keV, a beam current of about 1 μA, and a spot size of a few square millimeters in the unfocussed situation. Starting from this situation, the luminescence intensity initially remains constant with increasing degree of focus. A further increase brings about a decrease in the luminescence intensity, while the incident electron flux is strictly kept constant. The spot size is varied over two orders of magnitude. It appeared that the focussed electron beam in the cathode-ray tube was extremely sensitive to magnetic fields. In order to avoid disturbances from a.c. magnetic stray fields (mainly

of line frequency) cathode-ray and phototube were placed within an 8 mm thick cast-iron pipe.

Since we did not know N or N_c we introduced a saturation parameter Sat by:

$$\text{Sat} = 1 - L/L(\infty), \tag{6}$$

where L is the observed luminescence intensity at a certain degree of focussing while $L(\infty)$ is the luminescence intensity in the non-focussed situation. From the steady-state solution of eq. (1): $\langle r \rangle = \langle g \rangle$, it could be found (see ref. 1) that for our theoretical model Sat $= \overline{N}/N_c$.

The following experimental results were found:

i) a shift of crossover frequency towards higher values with increasing saturation coefficient. This is presented in fig. 1 for the SMR, while fig. 2 shows the same behaviour in the case of the relative excess-noise spectrum. The theoretical relation between crossover frequency and Sat in our model is quite simple. In our experiments we observed that deviations between theory and experiments increase from 2% for Sat = 0.3, 10% for Sat = 0.5 to 60% for Sat = 0.75. The observed frequency shift was lower than predicted by theory;

ii) squared modulation response and excess-noise power have the same frequency dependence. This is illustrated for two values of Sat in fig. 2.

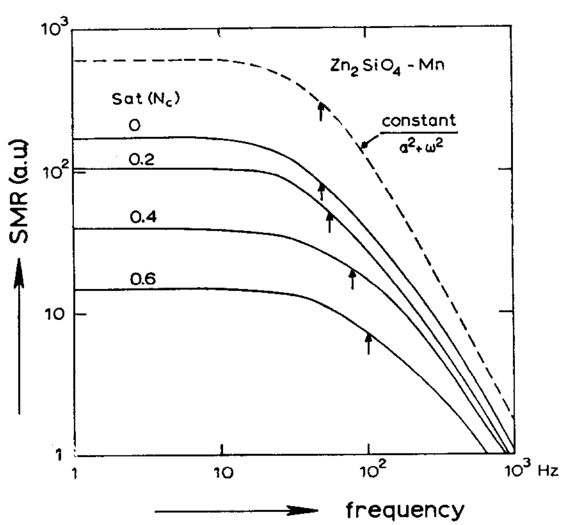

Fig. 1. Squared modulation response (SMR), in arbitrary units, as a function of frequency for different values of the saturation parameter Sat. For comparison a theoretical curve with frequency dependence predicted by eq. (5) and crossover frequency of the experimental SMR at Sat = 0 is given (dashed curve). The arrows indicate crossover frequency.

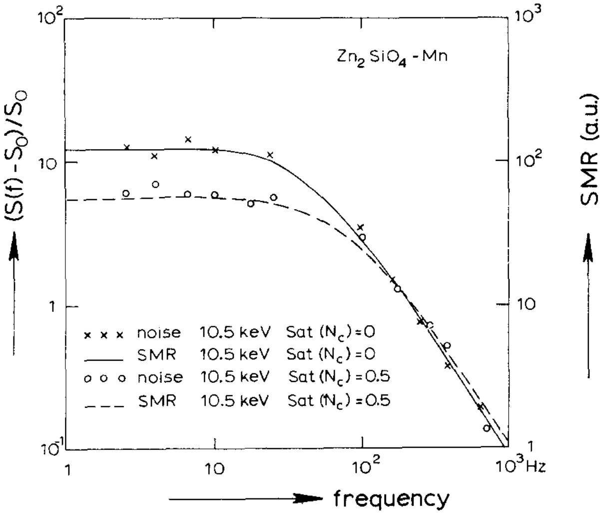

Fig. 2. Relative excess noise and squared modulation response as a function of frequency for two values of Sat.

Within the statistical precision[1]) of the measurements no difference was found between the frequency dependence of these quantities at corresponding values of the saturation parameter;

iii) there is a discrepancy between theoretical and experimental frequency dependence of SMR as well as excess noise. Comparing the dashed curve in fig. 1, presenting the function: constant $/(\alpha^2 + \omega^2)$ (where α is chosen such that its crossover frequency corresponds with the one of the SMR for Sat = 0), with the experimental ones, we note a smearing out of the experimental frequency dependence in the high-frequency range. This indicates perhaps the existence of more than one relaxation time in the luminescence;

iv) the low-frequency plateau of relative excess noise decreases with increasing value of the saturation parameter, as was predicted by theory. No quantitative agreement, however, exists between theory and experiment. In order to compare theory with experiment the relative excess noise for $\omega = 0$ was estimated through eq. (5). The quantum efficiency of the S20 photocathode used for the green radiation from Willemite was estimated from the EMI data sheet to be $(15 \pm 2)\%$. In view of the very close position of the phototube with respect to the screen of the cathode-ray tube, we estimated that about 50% of the emitted photons reached the photocathode. The detection efficiency λ is thus estimated to be roughly 0.07. The experimental value of the relative excess noise in absence of saturation (Sat = 0) was found from fig. 3 to be 13 ± 1. This implies that $p\langle\gamma\rangle \simeq 2 \times 10^2$ [see eq. (5)]. Since the value of $p\langle\gamma\rangle$ is large with respect to the second term of the right-hand side of eq. (5a), its accuracy is not relevant to the results

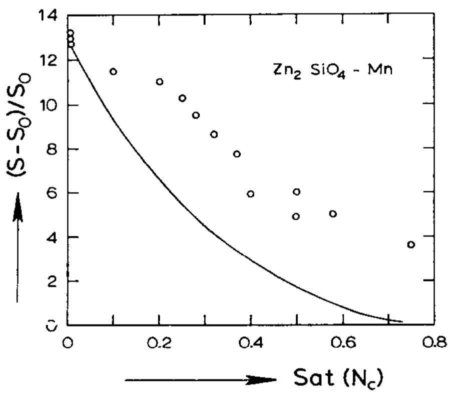

Fig. 3. Relative excess noise in the limit of low frequency as function of Sat; o experiment; ——— theory.

of calculation of the relative excess noise as a function of $\overline{N}/N_c$. The result is shown in fig. 3. The quantitative discrepancy with experiments may be ascribed also to the occurrence of more than one relaxation time in our phosphor. The simple theoretical curve in fig. 3 should then be replaced by a superposition of similar curves with several τ's, thus lowering the discrepancy.

As has been pointed out before [1]) the energy efficiency (ratio of the emitted photon and incident electron energy) of the phosphor can be found from the values of the relative excess noise, the mean photon energy, the primary electron energy, and the detection efficiency. Using $(S - S_0)/S_0 = 13 \pm 1$, a mean photonenergy of 2.5 eV, a primary electron energy of 10.5 keV and a detection efficiency $(15 \pm 2)\%$ for photons leaving the phospor at the eyeside, we found for the energy efficiency of our phosphor at the eyside of the screen: $(2.3 \pm 0.3)\%$. This is a reasonable value [4]).

We tried to extend our theory of the influence of saturation on the luminescence noise in order to find other explanations of the quantitative discrepancy between theory and experiment. First the stimulation of downward transitions by electron impact was taken into account. It was found that in this case the spontaneous character of the de-excitations is affected which should give rise to a luminescence noise density above full shot noise in the high-frequency limit [see eq. (2)]. This was not found, however. We also tried to develop a theory, in which the saturation coefficient is space dependent [2]). From this it appeared that the relative excess noise as function of $\overline{N}/N_c$ differs only by a few percent from the behaviour predicted by the simpler eq. (5) and could not explain the discrepancies found in fig. 3.

4. *Conclusions.* In accordance with the theoretical predictions based on our simple model [see eq. (5)] the crossover frequency of relative excess noise

spectrum and squared modulation response shifts towards higher values with increasing value of the saturation parameter [eq. (6)]. The experimental crossover frequency as a function of Sat could not unambiguously be compared with the theoretical predictions, since in our experiments more than one relaxation time occurred, whereas our simple theory only takes into account one relaxation time.

The agreement in frequency dependence of relative excess noise and squared modulation response (see fig. 2) is in accordance with our theoretical expectations.

The difference between theoretical predictions and experimental results concerning the relative excess noise at low frequencies might be ascribed to the occurrence of more than one relaxation process in the luminescence.

According to eq. (2) we have: $\tau = \alpha^{-1} (1 - \bar{N}/N_c)$ where α^{-1} is the lifetime of the excited state.

For $\bar{N}/N_c \ll 1$, *i.e.* for low saturation τ becomes identical to the lifetime of the excited luminescence centre (manganese). From the value of 50 Hz for the crossover frequency at low saturation (see fig. 1) this lifetime is (3.0 ± 0.2) ms. Bril and Klasens[4]) have found in the case of Zn_2SiO_4–(0.004 Mn) 13 ms while Bril[5]) has found a lifetime of about 4 ms in the case of Zn_2SiO_4–(0.08) Mn. The Mn concentration in our case was 5% and therefore the observed lifetime is reasonable. The distribution of lifetimes with upper limit of 3 ms is presumably due to a distribution of Mn concentration over the grains of the phosphor.

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