# Electro-luminescence

H. K. HENISCH

# ELECTROLUMINESCENCE

*by*

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# PREFACE

THIS introductory account is intended primarily for those who have a general background of semiconductor physics and are about to make a special study of electroluminescence as a phenomenon. I hope that it will also be of value to engineers and technologists whose interests in the field are essentially practical and that it will fulfil the dual function of a "status report" and a textbook. The materials which occupy the wide borderland between insulators and orthodox semiconductors are receiving increasing attention and their investigation now constitutes one of the most interesting branches of solid state physics. A great deal has been written about electroluminescence, as the appended bibliography will show, and a framework of theoretical interpretation is now available. On the other hand, much of our knowledge is still empirical and this has largely governed the arrangement of material in the present volume. Many urgent questions of detail remain unanswered and it is hoped that the development of the subject has been served by the definition and isolation of these problems.

During the preparation of this volume I have enjoyed the generous support of many friends to whom grateful thanks are due. In particular, I should like to acknowledge the help and encouragement of P. C. Banbury, M. E. Bell, J. L. Birman, C. V. Bocciarelli, E. A. Davis, D. H. Frankl, S. T. Henderson, G. T. Hodge, R. Kaufman, A. Lempicki, E. E. Loebner, B. R. Marathe, H. M. O'Bryan, J. Tauc, W. A. Thornton and A. J. Young.

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Chapter 1

# GENERAL SURVEY

THIS Chapter provides an empirical and descriptive account of electroluminescence in the multiplicity of forms in which the phenomenon has been observed on different materials. Some of the observations are not yet well understood and will have to be the subject of further experimentation for some time to come. Many others have been extensively studied and successfully interpreted. These and their corresponding theoretical models receive more detailed treatment in later Chapters.

## 1.1 ELECTRO-PHOTOLUMINESCENCE

### 1.1.1 GUDDEN–POHL AND DÉCHÊNE EFFECTS

It has long been known that the application of high electric fields, whether constant or alternating, can have important effects on the behaviour of photoluminescent substances subjected to ultraviolet stimulus. These effects can also be observed during the decay period which follows the cessation of radiative stimulus, and it is in this form that they were originally discovered. In quite general terms, we distinguish between an enhancement of light intensity by the applied field, called the *Gudden–Pohl Effect* (1920:**1**),† and a partial quenching, called the *Déchêne Effect* (1935:**1**, 1938:**1** and 1943:**2**). The Gudden–Pohl effect can be observed when the phosphor is well insulated from the metal electrodes which supply the field, whereas conditions for the Déchêne effect appear to be more favourable when the phosphor passes an appreciable current. The applied fields must be of the order of

† These designations refer to the bibliography at the end of the book.

several kilovolts per centimetre. Both phenomena decay either to zero or, at any rate, to a lower stable value. They can occur together, the Gudden–Pohl effect being normally associated with shorter time constants.

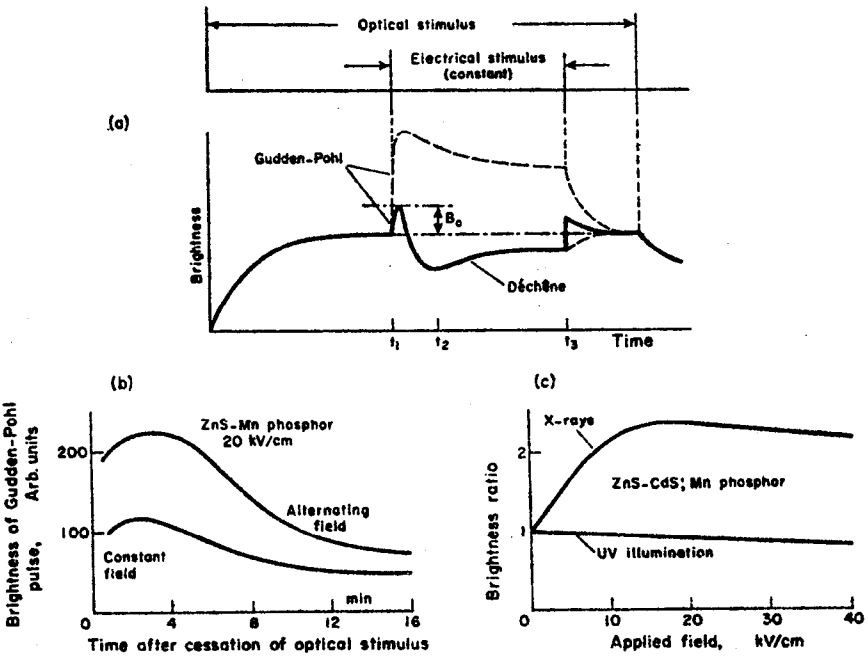


FIG. 111.1. Effect of electric fields on photoluminescence. (a) Summary of electro-photoluminescence phenomena. After Matossi (1957:31). (b) Intensity of Gudden–Pohl light pulses as a function of time during photoluminescent decay. After Hinderer (1931:1). (c) Dependence of the Gudden–Pohl and Déchêne effects on the mode of excitation. Brightness levels recorded after the initial transient. After Destriau and co-workers (1955:78).

Figure 111.1a gives a composite picture of the forms which these phenomena can take when the applied field is constant. The relative prominence of the various features may vary a great deal from specimen to specimen. When the field is alternating, the light intensity has a superimposed ripple which, as a rule, has a complicated wave shape. For a sufficiently high applied field, the ripple occurs with twice the

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applied frequency. The broken line illustrates a case in which the Déchêne effect is absent. A small enhancement occurs normally at the instant when the external field is removed but in some cases it is suppressed. This particular relaxation can be very rapid as, for instance, reported for one of the phosphors investigated by Steinberger, Low and Alexander (1955:**36**). The detailed features of these effects bear complicated relationships to the magnitude and character of the field as well as the instant of its application relative to the timing of the optical stimulus. Figure 111.1b, for instance, shows how the magnitude of Gudden–Pohl light pulses excited during photoluminescent decay diminishes with time. There is an interesting and, indeed, perplexing increase before the main decay sets in, which illustrates one out of many complicated and interconnected forms of behaviour [e.g. see (1923:**1** and 1949:**1**) and Sub-section 1.1.2]. No corresponding effects are observed when magnetic fields are applied (1924:**2** and 1955:**85**).

It is well known that the decay processes in excited phosphorescent substances can be accelerated by infra-red radiation. In this case the *total* light output is always the same, no matter whether the decay is accelerated or allowed to occur spontaneously. The present effects are fundamentally distinguished from this form of optically accelerated decay, since the intensity–time integral of the light output may be changed substantially by the action of the electric field. The Gudden–Pohl effect, for instance, can increase the total light output during the decay period. During the irradiation period electrons are excited by the absorption of photons, and when the optical stimulus ceases, only a finite number of electrons can be in excited states. An applied field which increases the total light emission must thus act in one of two ways (or in both): it could increase the relative probability of radiative as opposed to non-radiative recombination or it could give rise to additional excitation on its own account. This last possibility seems inherently more likely and establishes the principal link between the more familiar phenomena of electro-photoluminescence and those which are the main subject of this book (see particularly Sub-sections 2.3.3 and 6.2.2). However, Matossi (1957:**31**) has reviewed these problems and, contrary to the above suggestion, has ascribed the quenching effect to an increase in the probability of non-radiative transitions. The detailed manner in which these particular changes could be brought about is still not clear.

It is possible to study the field effects not only during normal phosphorescence but also during infra-red stimulated emission from

phosphors previously irradiated by ultraviolet light. Such experiments should, in principle, yield information on the role played by *trapping processes*, which control the delay between the excitation of a charge carrier and its radiative recombination (Sub-section 2.7.4). The results are, however, complicated and have not yet led to a consistent theoretical picture (1957:**56**).

### 1.1.2 Recent Experiments on Field Effects

Electro-photoluminescence effects as described above have been the subject of numerous investigations and though many of the interpretations are still doubtful, the principal experimental results are of interest in the present context.

The broken line on Fig. 111.1a shows a case in which a permanent enhancement of brightness remains after the initial transient. Destriau and co-workers (1955:**78**) have observed this to occur in certain ZnS–CdS and some ZnS phosphors under X-ray stimulus using an alternating field. For test purposes these powder phosphors were suspended in a transparent dielectric. With constant external fields, the effect was only temporary owing to the high conductivity of the phosphor. The enhancement factor was sometimes found to be as high as three. In these cases, the usual form of Déchêne quenching occurred when electric fields were applied during irradiation by ultraviolet light instead of X-rays. The field effects are thus sensitive to the mode of optical excitation. Figure 111.1c gives typical results on the field dependence of enhancement and quenching effects after the initial transient. The existence of the maximum followed by a decrease has also been reported by Steinberger and co-workers (1955:**36**). No luminescence at all could be observed when the external field was applied in the absence of incident radiation. In this way, the enhancement effect is observationally (but not fundamentally) distinguished from the phenomenon of *photo-electroluminescence* described in Subsection 1.3.6. Gobrecht and Gumlich (1960:**35**) have described an interesting phosphor containing manganese which responded to electric fields by exhibiting simultaneously an enhancement of the yellow and a diminution of the blue photo-luminescent bands. [See also (1959:**63**).]

The complicated nature of the present group of phenomena is further demonstrated by the observation that the consequences of electrical excitation can persist over long periods of time (e.g. several hours) and that they can manifest themselves also by their effects on

secondary optical stimulation (1954:**49**). As far as is known, systematic experiments on electro-photoluminescence have not yet been carried out on single crystal specimens of zinc sulphide, though some have been reported for cadmium sulphide (1959:**66**). Such experiments are certainly needed to complete our picture of these phenomena. Their interpretation should become a straightforward matter when the electrical luminescence phenomena which occur *without* optical stimuli are more thoroughly understood. Further references to electro-photoluminescence can be found in an extensive bibliography prepared by Ivey (1959:**29**).

## 1.2 BASIC OBSERVATIONS OF ELECTROLUMINESCENCE

### 1.2.1 General Considerations

In the above Section we have noted some of the effects which strong electric fields can have on photoluminescent solids under optical (u.v. or X-ray stimulus). In this context, the electric field fulfils only a modulating function. It has been found, however, that such fields can in certain substances stimulate light emission by themselves, even in the absence of optical irradiation. This is the phenomenon of *electro-luminescence* which is the principal concern of the present volume. Different forms of the effect were discovered by Lossew in 1923 and by Destriau in 1936. It involves the direct conversion of electrical energy into radiation, without recourse to any intermediate energy form, such as heat. Whereas the emission from incandescent bodies is determined by the temperature alone, "cold" emission from an electroluminescent substance can be shown to depend on its chemical and physical constitution. It belongs to the category of properties known as "structure sensitive". This leads to the possibility of control which is important in many practical ways. It also provides an opportunity for using the phenomenon itself to further our understanding of electronic processes in these materials.

Electroluminescence in the present sense, is normally observed only in solids. However, certain liquids are known to behave electrically as orthodox semiconductors (e.g. liquid selenium), and this leads us to expect that they may conceivably be electroluminescent as well. Indeed, instances have been recorded in which contaminated oils have given light emission under high voltage d.c. excitation, though the mechanism of this process has not yet been identified (1959:**37**). Solids

which exhibit electroluminescence may be polycrystalline or single crystals; it is not definitely known whether the phenomenon occurs in non-crystalline solids. Since the energy conversion cannot be one hundred per cent efficient, electroluminescent light sources do in fact heat up during normal operation. They remain "cold" sources, however, in the sense that no part of their emitting surface even approaches the temperature range of incandescence as the emitted radiation might otherwise suggest. The high field strengths required are normally provided by applying a moderately high voltage across a phosphor layer of small thickness. In general, this involves the use of at least one semi-transparent electrode which serves as electrical contact and permits the radiation to be observed. Such an arrangement is called an *electroluminescent cell*. One of the electrodes, though not transparent, may be very much smaller than the other, and light emission can then be observed in the neighbourhood of its boundaries. When one of the electrodes is so small as to constitute a "point contact", the operative field may be that which exists across the electrical spreading resistance.

It is not of primary importance in the present context whether the radiation emitted from an electroluminescent substance is visible to the eye or not. In many important cases which are described in Chapter 4 the wavelengths fall into the infra-red region of the spectrum. Moreover, the term "electroluminescence" is here phenomenologically defined, without any implication that it refers to a single physical mechanism. There is, on the contrary, no doubt that electrically stimulated light emission can be brought about in several different ways. In the remainder of this Section we are concerned with some of the early observations which are still of historical interest, and with the characterization of electroluminescence as a *primary* phenomenon.

Photoluminescent phosphors used for experiments under electrical excitation normally contain a variety of deliberate additives, e.g. copper, manganese, chlorine, lead, etc., which may be built into the lattice or may occur in interstitial positions. The detailed manner in which they are accommodated controls the mechanism of the emission processes. The principal additives are called *activators* and the localities in which they occur are referred to as *activator centres* or *luminescence centres*. In some cases, an additive is effective only in the presence of a particular second additive which is then called a *co-activator*. Charge carriers excited by the absorption of ultra-violet light return to the ground state within the immediate environment of luminescence

centres. The centres also play an important role in determining the *electro*luminescent response of these phosphor materials, though their electrical excitation involves additional complications. In Sub-section 1.2.3 and in Section 1.3 we shall be concerned only with phosphors of this kind. There are, however, electroluminescent materials in which some of the principal features of the emission characteristics are not controlled by activators but by the parent lattice itself. The corresponding recombination processes receive more detailed consideration in Section 2.7.

It is possible for electric currents to promote chemical reactions (dry electrolysis) at solid phase boundaries, and in some cases these involve a radiative energy release. This phenomenon is a form of *chemiluminescence* and is outside the scope of the present work.

### 1.2.2 Early Investigations on Detector Contacts

Silicon carbide was the first solid reported as being an electroluminescent material, at a time when it was most commonly used as the basis of crystal detectors. In the earliest investigations described by Lossew (1923:**2**) and by Claus, the luminescence was always confined to the immediate neighbourhood of the contacts. The phenomenon thus came to be called "Detektorleuchten". Many unsuccessful attempts were made to discover signs of external arcing to which the luminescence might have been ascribed. In the early experiments the luminescence disappeared when the polarity of the applied voltage was reversed. There are, however, discrepancies in respect of the polarities involved between the reports of Lossew and those of Claus (1931:**2**, 1930:**1** and 1932:**1**), possibly due to the use of crystals of different character. It was soon found that the use of metallic contacts is not essential (1931:**3**). Electroluminescent patches of the same kind could be produced with electrolyte contacts, using aqueous solutions of sodium carbonate or sodium bicarbonate. The emitted light had a continuous spectrum.

Tetzner (1948:**1**) has given a detailed description of various experiments carried out on commercial silicon carbide of low purity (97 per cent) and indifferent crystal development. The results are still interesting in a qualitative way. Confirming some of Lossew's later observations (1928:**1**), Tetzner noted that luminescence could appear at both cathode and anode, the two emission patterns being somewhat different in character. At the cathode, the light appeared to come from a region of large area and about $10^{-2}$ cm thickness. At the anode, the light

occurred in the form of very small spots which appeared to be situated deeply inside the crystal. Cathodic luminescence was normally yellowish-green and sometimes red, anodic luminescence normally bluish-green. The results of probe measurements suggested that the thin region in which cathodic luminescence occurred was characterized by a high conductivity. Again, on certain crystals only one of the two forms of luminescence could be observed. In these particular experiments the spectral composition of the emitted light was independent of the applied voltage, contrary to earlier reports on this point by Lossew and Claus. As far as could be ascertained by a coarse test, the radiation was not polarized. The silicon carbide used in these experiments is likely to have been of the hexagonal ($\alpha$) form. In all probability, the specimens were non-uniform. Indeed, Lossew (1940:4) had already suggested that the observed light emission was, at least in some cases, associated with the presence of $p$–$n$ junctions within the material. The full significance of this suggestion could not be appreciated at the time, since the characteristic behaviour of $p$–$n$ junctions was not yet properly understood.

### 1.2.3 First Observations of Electroluminescence in Powder Phosphors

The first observations of this kind were reported by Destriau in 1936 in the context of various studies on electro-photoluminescence (1936:1). A comprehensive report of the experiments appeared in 1947 (1947:1). The phenomena differ from those discussed above in a number of ways. The materials concerned generally have a much higher resistivity and, indeed, the phosphor particles need not be in direct contact but can be suspended in an insulating medium. In this case, continuous light emission can be excited only by the application of *alternating* fields. Various test configurations used for the exploration of electroluminescent powder phosphors are described below. Using average field strengths of the order of $10^5$ V/cm, Destriau was able to excite luminescence in sulphides, tungstates, germanates, silicates, platinocyanides and other substances. It is not possible now to assess in which of these cases (if any) the luminescence was excited by, or at any rate modified by, "secondary" processes. This possibility is always present and is further discussed in the following Sub-section.

The zinc sulphide test specimens used by Destriau were prepared without the sodium chloride flux which is commonly added to

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