

## EFFECT OF ELECTRIC FIELD ON LUMINESCENCE. A MINIREVIEW

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*Abstract.* This review gives an account of a few experiments made to study the effect of external electric field on photoluminescence and thermoluminescence. The explanations of varied nature as offered by different researchers are put forth in the review. The case of thermoluminescence is however unique. Here possible effects of internal electric field due to impurity-vacancy pairs created in a lattice are also important and envisaged through changes in trap depth values reflected by observed TL emissions. The proposed theoretical basis for electric field effect of I-V pair in TL emission is also quoted in this review.

*Key words:* Photoluminescence, thermoluminescence, electric field effect, impurity-vacancy pair, internal electric field.

### INTRODUCTION

Some mysterious phenomena fascinated the people in early days. These may be enumerated as: St. Almo fire and Aurora Australis due to radioluminescence, emission on rubbing a crystal due to triboluminescence, ephemeral blue haze from a glass of gin in sun light due to fluorescence, enchanting radiation from fireflies due to chemiluminescence. The early records of Chinese literature (1500–1000 BC) mention about these [37, 38]. Thousand years later, Aristotle (384–322 BC) stated that these mysterious phenomena had nothing to do with any heating effect [6]. The first scientific publication in this area of bio-organism, however, emerged only in 1555 AD through a book by Gesner [15]. This was a time, when non-scientific verses were written by Nostradamus in France. A history of luminescence from early times to 1900 AD was given by Harvey [17].

Our present interest is on electric field effects on photoluminescence (PL) and thermoluminescence (TL) and details for the two are given as follows.

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## PHOTOLUMINESCENCE

In PL, light is made to fall onto a sample. The light is absorbed by the sample and the electrons (shown by black circles) in the material reach the excited state. The excited electrons subsequently jump to the conduction band, while relaxing through non-radiative processes. The electrons in the conduction band then spontaneously drop to the valence band to emit light of appropriate wavelength. The simple energy level representation for PL is given below in Fig. 1.

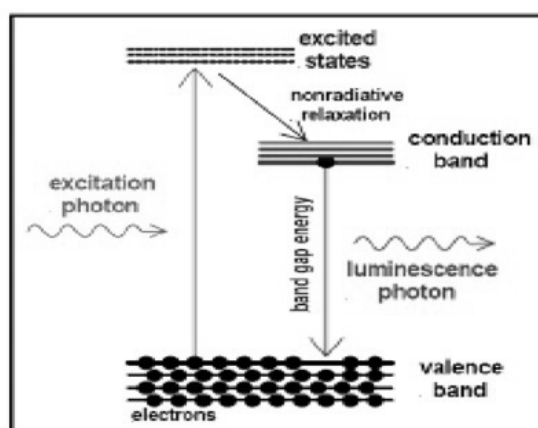


Fig. 1. Basic features of PL emission.

More elaborative representation for PL is as per Jablonski diagram given by Atkins *et al.* [4] which involves various processes for a general PL system *viz.* absorption, fluorescence, phosphorescence, vibration relaxation, internal conversion, external conversion, and inter-system crossing.

## IMPORTANCE OF PL

PL provides non-destructive contact-less method of probing a material [16, 40]. Its applications are in many photonic devices for example in LED/phosphor screens. In the context of semiconductors, it is used in semiconductor lasers and solar cells. PL is an important technique for knowing purity and crystalline character of semiconductor for knowing disorder present in the system. PL is also sensitive to surface effects and is used as a probe for electron-hole surface processes in a material.

PL studies help in estimation of band gap energies. In nanostructured material, the band gap is changed remarkably in contrast to its normal form [9]. The band gap measurements thus help in estimating the nano-crystalline content in the material.

As described in the following paragraphs, the PL technique has been found quite useful in development of dyes, coated textile materials, knowing the nature of materials in antiques, study of nanomaterials, in biotechnology research and in gemology.

Sagar *et al.* [34] have established PL enhancement in an organic dye by graphene quantum dots. Through studies in Han blue and Han purple pigments, Pozza *et al.* [31] have further proposed the use of PL in works of art and culture. Murai *et al.* [26] have established how enhanced PL emission at a specific angle could be achieved by absorption and subsequent PL emission from a dye in thin polymer film form on a plasmonic array of metallic nano-cylinders.

In the area of textile development, Yan *et al.* [43] have studied the effect of inorganic pigments on the luminescence properties of colored luminous fiber. The technique of photoluminescence to identify materials in modern Avant-garde paintings has been developed by Artesani *et al.* [3]. This would be helpful in dating science and to understand ongoing degradation of painting material.

Rodriguez-Rodriguez *et al.* [33] showed TL to occur by radiative energy transfer from optically trapped colloidal quantum dots through fluorescence dyes. This produces a light source with a great spatial adjustability useful to understand nano-structures.

Persistent luminescence phosphors (PLNP) are a group of materials having glow even after stoppage of excitation. Wu *et al.* [42] have focused how PLNP could be manipulated in bio sensing, cell tracking, bio-imaging and cancer therapy. Ibarra-Ruiz *et al.* [20] have presented synthesis, surface functionalization and biomedical application of photoluminescence nano-particles. A method for production of photoluminescent carbon nano-tags from harmful cyanobacteria for drug delivery and imaging in cancer therapy has been developed by Lee *et al.* [23].

PL is also an essential tool used for distinguishing treated and synthetic diamonds [13].

#### ELECTRIC FIELD EFFECT ON PL STUDIES

PL spectra of  $\text{Al}^{3+}$  doped ferroelectric guanidium chromium sulfate hexahydrate were reported by Morita *et al.* [25] from liquid helium to liquid nitrogen temperature. It is seen that  $\text{Al}^{3+}$  diluted crystals manifest extra recombination lines caused by internal electric field of the ferroelectric host.

Stampor and Kalinowski [36] have reported luminescence quenching due to electric field in thin films of aluminum3a-hydroquinone. Luminescence quenching is found to depend upon excitation wavelength and electric field. The quenching is attributed to field assisted hopping of charges to localized excited states.

Intensity and frequency studies have been undertaken in PL of CdSe crystals by Park *et al.* [30] and the PL intensity appears to be modulated due to electric

field. The results could be attributed to a model which imagines modulation of energies of surface charge trapping sites by electric field, causing modulation in the range of exciton quenching by nano-crystalline sites in the sample.

Electric field assisted PL intensity changes are also reported as a function of incident light frequency by Chiang and Ohta [10] for pyrene solution in alcohol. At high concentration of pyrene, for which excimer fluorescence is observed along with monomer fluorescence, it is seen that non-radiative decay, the process for excimer emitting state is decelerated on application of electric field.

#### MECHANISM OF ELECTRIC FIELD EFFECT ON PL

The main theme of PL studies is the bulk emission of electrons accumulated in the conduction band by dropping into the valence band. It is presently proposed that the electric field incident on a sample checks electron getting accumulated by providing alternative paths. The electrons may thus be forced to occupy several alternative stable sites in the lattice which finally lead to the appearance of new structure lines through recombination processes. The electron may also be forced to occupy localized excited states through electric field assisted hopping mechanisms.

#### THERMOLUMINESCENCE

Thermoluminescence is a process in which a mineral emits light while it is being heated. It is a stimulated emission process occurring when the thermally excited emission of light follows the previous absorption of energy from (an ionizing radiation) X-ray or  $\gamma$ -ray.

TL is a convenient technique to access the life of a rock, the process being termed as dating. The dating process is used in archeology and earth sciences to date artifacts and rocks. The dating results have been found to be accurate to within one percent.

As far as thermoluminescence (TL) emission is concerned, the electrons in the traps are supposed to get excited to the conduction band upon external heating and then fall down to the hole traps or recombination centers as per Srivastava [35] and Bos [8]. The recombination for an electron is also possible without reaching the conduction band. The traps have an important role in the TL process. The electrons situated in traps do not spontaneously lose energy to drop to the lower available states.

TL studies in a number of alkali halides have been reported by Deshmukh and Moharil [12]. These studies do not, however, yield detailed information about the structure of traps involved. The TL studies in gamma-irradiated Harshaw KCl crystals are also reported by Ausin and Alvarez Rivas [5] who observed that they are a under the TL curve is always proportional to F-center concentration in the sample

before heating. They further attribute the role of F-centers to the recombination centers in the annealing process. The studies to explore the spectroscopy of traps in powdered KCl are also undertaken by Ranita *et al.* [32] and it was found that the trap depth is proportional to the glow peak temperature ( $T_m$ ).

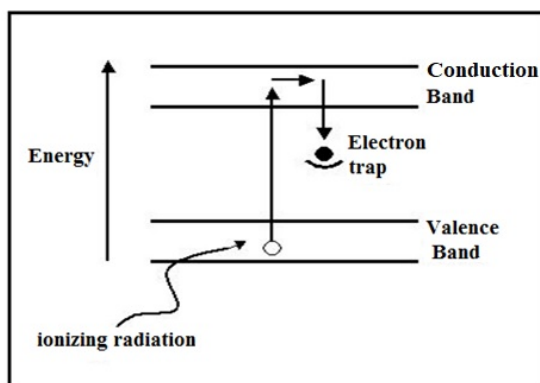


Fig. 2. Trapping of electrons for TL emission.

The TL studies were further conducted in NaCl and NaCl:  $\text{Ca}^{2+}$  crystals by Joshi *et al.* [21] and the role of the latter system in dosimetry has been discussed.

In a simple TL process, the ionizing radiation excites the electrons from valence band and these are held in traps near the conduction band (Fig. 2). Thermal excitations then lead to the shift of electrons from traps and subsequent emission through recombination process.

#### IMPORTANCE OF TL

There have been numerous studies which characterize the electron traps and their applications in dosimetry, but the nature and the physical structure of such traps in alkali halides is not yet fully established. The TL technique is also used elaborately in dating process for determining the life of rocks and minerals. This technique is also a non-destructive technique.

There have further been many studies related to development of new TL phosphors with increased TL efficiency.

Poly-crystalline powders of  $\text{BaSO}_4$  were prepared by Annalakshmi *et al.* [2]. These samples exhibited very high TL efficiency following gamma radiation compared to other phosphors. These phosphors are found suitable to make dosimeters for detecting very small exposers of low energy X-rays.

TL properties of pure and europium doped hydroxyapatite (HAp) have been derived by Mendoza-Anaya *et al.* [24] by exposing these to different doses of

gamma radiation. It is further seen that europium doping in a compound increases monoclinic phase at the cost of usual hexagonal phase.

Alkaline earth silicates are appropriate compounds for TL materials due to their water resistance and chemical stability. Recently, Alajlani *et al.* [1] have reported results of TL studies in  $\text{Sm}^{3+}$  doped barium silicate and observed two glow peaks at 91 °C and 193 °C. They also studied the dose and heating rate response of TL curves.

#### EXTERNAL ELECTRIC FIELD EFFECTS ON THERMOLUMINESCENCE

Nouailhat *et al.* [27] made TL studies in KI:Tl samples irradiated with  $\beta$  rays at liquid nitrogen temperature and observed two peaks at 105 K and 170 K. However for experiments performed with samples under static electric field of nearly 40kV/cm, the intensity of second glow peak was strongly reduced. These observations were attributed to retrapping of electrons responsible for second peak at some new traps.

The changes in intensity of TL output from ZnS phosphors on application of external DC field were studied by Kiveris and Pipinis [22] and the results are attributed to tunnelling mechanism of release of electrons with phonon participation.

Enakshi and Rao [14] have reported the effects of high AC electric field on TL of X-ray irradiated NaCl and KCl crystals. They observed decrease in F-center concentration and light output from crystals with the effect of AC field.

#### THE BI-VACANCY

For TL to occur an electron must be captured at a suitable trap. Fig. 3(a) represents an electron trapped at a secluded anion vacancy. Such TL traps should not be quite stable and thus have a little role in TL process. Fig. 3(b) inherits the bondage of anion vacancy with cation vacancy (bi-vacancy) and shows an electron trapped to this combination. The electron gets trapped at an anion vacancy site and accompanied cation vacancy provides increased lifetime of the trap. The bi-vacancy may thus be termed as anion vacancy accompanied by life guard.

As per a theoretical model by Herman and Barnett [18], it has been estimated that the 2s level of secluded F-center has energy close to the conduction band in NaCl. The 2s energy level thus fits as an ideal candidate for possible electron traps near conduction band in NaCl crystal. It may be mentioned here that the energy of 2s level has also been estimated by Hoya *et al.* [19] to be close to the conduction band in KCl crystals, however, in a different context.

The electron traps are supposed to be situated at dislocation sites in a crystal lattice, this being supported by the reported works of Benia *et al.* [7] and Wosinski [41].

A vacancy pair, also called a bi-vacancy, is a suitable trap for electrons in NaCl lattice and helps in the formation of a cation vacancy coupled F-center as shown in Fig. 3(b). The electrons trapped at anion vacancy end of bi-vacancy cannot fall to the lower energy states as in a secluded F-center, because the potential at the electron site in a bi-vacancy is neither pure Coulombic, nor like a perfect spherical well. Therefore, a 2s energy level of such a bi-vacancy center in pure NaCl is likely to act as an ideal trap for electrons.

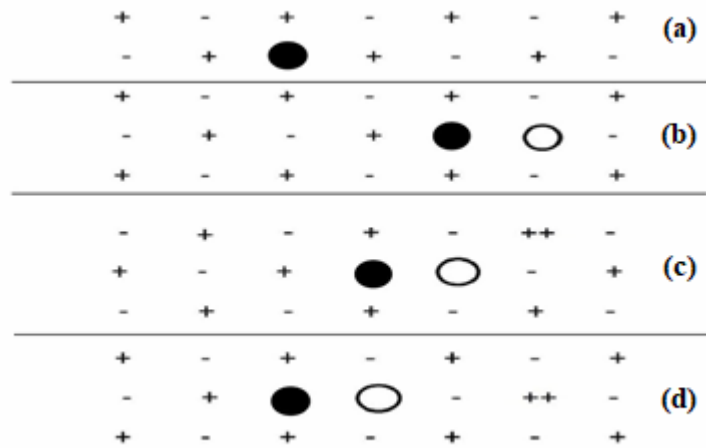


Fig. 3. Basic defects in an alkali halide crystal.

#### Theoretical support for bi-vacancy

A spherical well model was developed by Herman and Barnett [18] to estimate the energy level values of 2s-trap for a usual secluded F-center in NaCl lattice. They could estimate the total number of bound states as per solution of Schrodinger 3d wave equation, using the well depth  $V_0$  and width of the well ' $a$ '. Here,  $V_0$  is determined by the positive charges surrounding the anion vacancy.

Pandey *et al.* [28] have recently extended the above model to estimate the energy of 2s-traps in a bi-vacancy. This is done by assuming the electron to occupy distorted spherical well appropriate to bi-vacancy with reduced well depth ( $5/6 V_0$ ) and increased well width ( $a$ ). It may be stated that the values of  $V_0$  and ' $a$ ' determine the position of 2s trap level in the solution of 3d Schrodinger wave equation for the electron in the bi-vacancy well.

Of course, as per Pandey *et al.* [28], in the above proposed distorted well for the bi-vacancy trap, the well depth value is reduced nearly by a factor of 1/6 as one of six nearest cation neighbors of anionic vacancies is removed, and the increased

value of the well width ( $a$ ) is determined by the more available space in a bi-vacancy, in contrast to secluded anion vacancy.

Pandey *et al.* [28] have justified suitability of bi-vacancy trap in KCl and successfully explained the observed trap depth value of 0.9 eV for 2s level in KCl lattice using the above proposition.

#### INTERNAL ELECTRIC FIELD EFFECTS ON THERMOLUMINESCENCE

Results presented in the earlier text highlight the role of external electric field on TL system. In a crystal like that of an alkali halide wherein the divalent cation may exist as impurities, impurity vacancy (I-V) pairs would also be found in a lattice. As per Pandey *et al.* [28], These I-V pairs maybe linked to anion vacancy (Fig. 3(c)) or bi-vacancy (Fig. 3(d)) to form nearest neighbor (nn) and next nearest neighbor (nnn) I-V pair complexes respectively. The internal electric field of such I-V pair could influence the energy level of such F-center and may shift the energy of the trap and affect the TL emission.

#### ROLE OF I-V PAIRS

In a divalent cation doped KCl, there would be plentiful I-V (impurity-vacancy) pairs in the lattice. Now if a doped KCl crystal is exposed to ionizing radiations to create F-centers, some of these would also be under the influence of the electric field of I-V pairs, whether nn (nearest neighbor) or nnn (next nearest neighbor) type. The Two types of I-V pairs have been identified by Watkins [39], while investigating EPR of  $Mn^{2+}$  in alkali halides.

#### SHIFTING OF 2S AND OTHER F-CENTER LEVELS WITH I-V PAIR COUPLING

As described in the following, the I-V electric field could be responsible to shift the 2s energy levels of F-centers under quadratic Stark effect as per Davydov [11]. This effect makes the traps at 2s levels shallower.

If there is an F-center perturbed by an electric field of nearby I-V pair (Fig. 3(c)/3(d)) the effect of the electric field on F-center energy levels has to be sorted out.

Assuming an F-center is exactly like a hydrogen atom, the degeneracy of 2s, 2p levels combination should also split into three levels under electric field of neighboring I-V dipoles. However, as per the real situation, an F-center in KCl host is not exactly akin to the hydrogen atom. In the case of a hydrogen atom, the electron is strictly under the influence of a spherically symmetric electric field. For a unique F-center in KCl, the electron which occupies the secluded anion vacancy site is, further, under the influence of an octahedral internal field which is not exactly spherically symmetric but was assumed to be close to it by Herman and



Barnett [18]. Thus, 2s and 2p levels in F-center are not degenerate and the effect of electric field of I-V dipoles on these levels must be dealt with uniquely.

Pandey [29] has proposed the role of quadratic Stark effect on F-Center levels under second order perturbation theory. For 2s level, such an energy contribution  $\Delta E$  is given by Eq. 1.

$$\Delta E = \frac{\langle 2s | e z \varepsilon | 2p_0 \rangle \langle 2s | e z \varepsilon | 2p_0 \rangle}{E(2s) - E(2p_0)} \quad (1)$$

where,  $\varepsilon$  is the electric field of I-V dipoles and  $e$  is the charge of electron. The  $2p_0$  is the 2p level component for  $m = 0$  and as in above expression this contributes to energy shift. The  $m = \pm 1$ , 2p level components do not contribute to above energy change due to selection rules. On solving the integrals using hydrogen atom wave functions, one finds that 2s energy level gets shifted upwards by an amount  $\frac{4 (e z \varepsilon)^2}{9 \Delta E}$  where  $\Delta E$  represents separation between 2s and  $2p_0$  levels.

It may be mentioned that the effect of second order perturbation theory on 2p ( $\pm 1$ ) levels came out to be zero. The  $2p_0$  levels were however affected by the electric field. The shift in their energy involves two terms and as per different energy level values of F-center this came out to be negative. The  $2p_0$  levels were thus shifted downwards under the I-V field in a KCl crystal. In that work Pandey [26] was however interested to know the shift in energy of only the 2s level traps to explain observed TL emission changes.

## CONCLUSIONS

The observed effects of electric field in both PL and TL studies have been elaborated in earlier paragraphs.

So far TL studies are concerned, these have been found to be also affected by internal electric field caused by impurity-vacancy dipoles within the crystal. Such electric fields are of the order of 1000 V/cm and may cause lifting of trap levels and thus affecting the TL emission. As per an earlier paragraph, the shift of trap level is proportional to the square of electric field; thus, both AC and DC fields could cause similar shifting effects. Again, the I-V pairs may occupy mn or nnn positions with respect to anion vacancy which traps the electron. Thus, corresponding different electric field values at the anion site could be estimated and quantitative shift of trap levels may be obtained to understand the electric field effect on TL emission. It would be interesting if, future workers are interested to make such studies. No such I-V pair electric field effects are possible for PL emission.

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