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PUBLICAÇÃO IEA N.º 286
Fevereiro — 1973

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(*) - Separata do J.Appl.Phys., Vol.43, Nº 7, July 1972.

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R.W. Christy and M.R. Mayhugh

Summary

The high-temperature dosimetry glow peaks in LiF : Mg are thought to result from thermal release of electrons from traps associated with the Mg impurity. There is also evidence, however, that the visible-light emission in the same glow peaks is due to recombination of mobile holes. These observations were reconciled by a mechanism proposed by Mayhugh in which the thermally ionized electron frees a V_3 -center hole, which then emits a photon at another recombination center. We find that this model is supported by new measurements of Podgorsak, Moran, and Cameron on glow peaks which occur below room temperature.

The thermoluminescence (TL) peaks observed above room temperature in LiF which contains of the order of 100 ppm of Mg are important because of the application of this material in thermoluminescence dosimetry. These peaks, numbered 1–5, are seen when material irradiated at room temperature is heated to about 300°C. The higher-temperature peaks 3–5 have been attributed to the thermal release of trapped electrons (rather than holes), on the basis of changes induced by optical and thermal bleaching in the F band and in impurity-absorption bands associated with the TL peaks.¹ The light emission, however, was ascribed not to recombination of these thermally ionized electrons but to recombination of indirectly released holes: On the basis of changes induced in an absorption band² at 113 nm, identified with the V_3 multiple-hole band,³ we suggested that electrons thermally released from the trapping centers were captured at V_3 centers, freeing a hole which then emitted the observed light at another recombination center. The TL-emission spectrum,⁴ which peaks at about 400 nm, was consistent with emission observed on the destruction of V_k hole centers at low temperature.⁵

Podgorsak, Moran, and Cameron⁶ have investigated TL peaks occurring in dosimetry LiF below room temperature. These peaks, numbered –4–0, are seen when the material is irradiated at liquid-nitrogen temperature and heated to room temperature. During further heating above room temperature the same peaks 1–5 are seen as after irradiation at room temperature. Optical bleaching in the F band at low temperature showed that all the TL peaks except –4 and –1 are repopulated, in agreement with our assignment of peaks 3–5 to trapped electrons. Because peak –4 was not repopulated and because of its TL-peak temperature, it was thought to be due to untrapping and recombination of V_k -center holes. The emission spectrum of peak –4 had a maximum at 250 nm (with a tail extending to 400 nm). The difference between the emission-spectrum maxima of the V_k center and the high-temperature electron traps led Podgorsak et al. to reject our indirect mechanism of emission by hole recombination, since our model "predicts the same emission spectrum for the glow peaks due to electrons as for glow peaks due to V_k centers".

The purpose of this communication is to argue that the emission spectra of Podgorsak et al. are not inconsistent with our TL mechanism, and to suggest that in fact their experiments can be interpreted to lend further support to our model. Our proposed emission mechanism does not require that the recombination emission of V_k -center holes near 150 K should have the

same spectrum as that of subsequently freed holes above 400 K: First, it is possible that the recombination centers giving the V_k -hole emission at 250 nm could be used up before the indirect holes are released subsequently, so that only the 400-nm recombination centers are left for the high-temperature TL emission. Second, it is possible that the recombination trapping or emission mechanisms could themselves be temperature dependent, so that the emission spectra at 150 and 400 K are different even though the recombination-trap densities were the same. Indeed some such explanation is supported by their emission spectrum for peak - 1, which is probably also due to trapped holes because, like peak - 4, it was not repopulated by F-band bleaching. Peak - 1 has its strongest emission maximum at 400 nm, like the high-temperature TL peaks, but it also has weaker maxima in the uv, at the same wavelengths as the strong maxima of peak - 4. Thus the spectrum of peak - 1 (270 K) is clearly intermediate between those of - 4 (150 K) and 5 (460 K), as would be expected if the hole emission spectrum depends on temperature or availability of recombination sites. In fact, peak - 1 may be due to V_k centers stabilized by a cation vacancy. ⁷

We believe that the TL mechanism in dosimetry LiF is still best explained by the following model, which reconciles the various evidence that the high-temperature glow peaks originate from the untrapping of electrons but that the corresponding light emission comes from the recombination of mobile holes. During room-temperature irradiation electrons are trapped at centers associated with the Mg impurity (and also in F centers), and the corresponding holes are trapped in V_3 centers. On heating through 200°C the Mg-center electrons are released (though not the more tightly bound F-center electrons). The mobile electrons annihilate one of the holes in a V_3 center, thus freeing the other hole which finds itself in an unstable single-hole center. This mobile hole causes the visible luminescence when it is captured at a recombination site associated with another impurity, probably Ti. ^{8,9} This impurity is present only in trace amounts and possibly functions as an intermediary in the ultimate recombination of the hole with an F-center electron (perhaps by a tunneling process). ¹⁰ The optical absorption identified with the Ti center ^{8,9} is little affected by either irradiation or TL emission. ¹⁰ On heating to 400°C the crystal is restored to its preirradiation state. This rather complicated picture appears to be the simplest one which accounts for all these features of the dosimetry process.

Resumo

Os picos dosimétricos de alta temperatura do LiF : Mg parecem resultar da liberação térmica de elétrons de armadilhas relacionadas com a impureza Mg. Por outro lado, há também evidências de que a emissão de luz visível desses picos é devida à recombinação de lacunas móveis. Essas observações foram conciliadas pelo mecanismo proposto por Mayhugh no qual o elétron liberado termicamente liberta uma lacuna de um centro- V_3 , que por sua vez emite um fóton quando capturada pro outro centro de recombinação. Aachamos que este modelo é sustentado pelas novas medidas da TL dos picos de emissão que ocorrem abaixo da temperatura ambiente, realizadas por Podgorsak, Moran e Cameron.

Résumé

Les pics d'émission dosimétrique à haute température du LiF : Mg paraissent résulter de la libération des électrons des pièges associés à l'impureté Mg. Il est également évident, cependant, que l'émission de lumière visible des mêmes pics d'émission est due à la recombinaison des "trous" mobiles. Ces observations ont été conciliées par un mécanisme proposé par Mayhugh dans lequel l'électron ionisé thermiquement libère une lacune de centre V_3 , qui alors émet un photon sur un autre centre de recombinaison. Nous pensons que ce modèle est confirmé par des mesures récents de Podgorsak, Moran et Cameron sur les pics d'émission qui se produisent à des températures inférieures à la température ambiante.

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