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Structured KCl:Tl Emission Detected by Electric Field: A Dynamical Jahn-Teller Effect Interpretation

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The temperature dependence of the triplet structure detected by electric field on the 3000-Å emission of KCl:Tl has been studied. The splitting follows a $\sqrt{T}$ law, in agreement with a model based on the dynamical Jahn-Teller effect.

Stark-effect measurements on the emission of KBr:Tl phosphors were performed at liquid nitrogen temperature by Giorgianni, Grasso, and Perillo,[1,2] and showed a well-resolved triplet structure in both high- and low-energy emission; in spite of the novelty of this result and the lacking of any adequate interpretation, this subject was not investigated any further.

In this Letter we report the results of similar experiments performed at various temperatures on the 3000-Å emission of KCl:Tl. This phosphor is well suited for this kind of research as the intensity of its prominent 3000-Å emission is fairly constant through the range of temperature considered. We found that the triplet structure evidenced by the electric field is present at all temperatures between 10 and 300 K and that the splitting increases with temperature. These facts are here interpreted in terms of the dynamical Jahn-Teller effect.

Single crystals of KCl:Tl (thallium concentration ~30 ppm, sample size $10\times10\times1$ mm^3) were mounted in a cryostat between two electrodes (one of which was semitransparent), and a sinusoidal electric field (50 kV_{rms}/cm, 500 Hz) was applied in the same direction as the exciting light. Emission was observed at right angle. The experimental setup was similar to the one described in Ref. 2.

Our results are displayed in Fig. 1. We note that the relative change of the emission, $\Delta I/I$, shows a nearly symmetric triplet structure at all temperatures; the splitting increases with temperature from ~0.1 eV at 10 K to ~0.2 eV at 300 K. With increasing temperature the structure becomes less prominent since the maximum-to-minimum ratio decreases considerably as a result of a larger negative variation of the zero moment of the emission band.

A similar electric-field–induced structure was observed in the absorption bands[2,3] and was related to Toyozawa and Inoue's model[4] which accounts for the triplet shape of the absorption bands observed with no external field. Here an explantion of the electric-field–induced structure of the *emission* bands is proposed and is based on the hypothesis that the transitions arise from the three branches of the triplet state as split by coupling to the trigonal vibrational modes, and that consequently each emission band is actually composed of three different sub-bands. In other words, we think that the Jahn-Teller effect—responsible for the structure of the no-field absorption bands—also engenders the structured emission as revealed by the electric field. The application to the phosphor of such field is expected

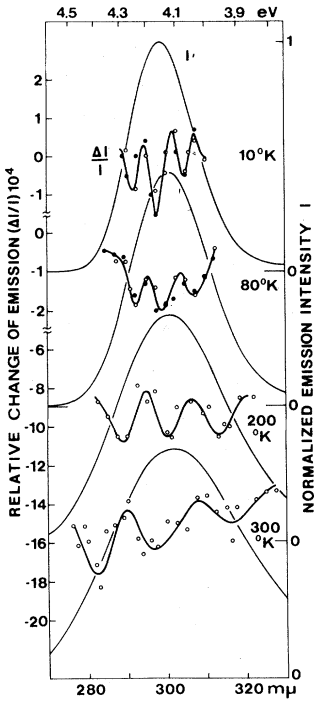

FIG. 1. Relative change of KCl:Tl emission excited in the A band, as a result of an external electric field, versus energy at various temperatures. The corresponding emission bands are also reported.

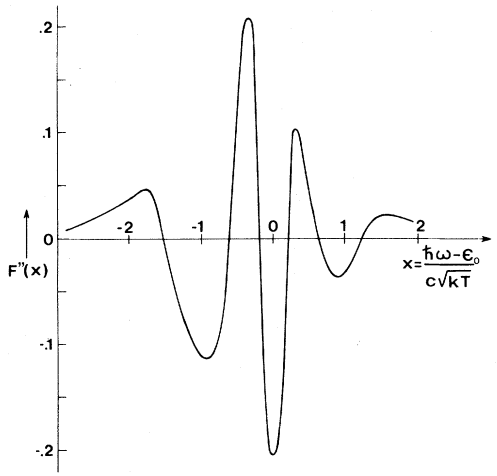

FIG. 2. Second derivative of the theoretical emission band shape, computed for $x_0 = -0.2$, versus normalized energy. ϵ_0 is the energy difference between the ground and excited states, neglecting the Jahn-Teller effect.

to produce a small but detectable increase in the second moment of each sub-band, which results in a fractional change in each peak intensity, $\Delta I/I \sim 10^{-4}$, with a field of the order of 10^5 V/cm.[5] The observed effect is just of this order of magnitude. A first-moment variation should also be induced, but too small to be observed ($\langle \Delta E \rangle / \langle E \rangle \sim 10^{-6}$).

In order to check our model we have computed the emission band shape $F(x)$ by adapting to emission Toyozawa and Inoue's formula[4] for the absorption band shape, and assuming, for simplicity, thermal equilibrium among the three excited states. The computed bands show triplet structure[6] provided that $0 \leq |x_0| \lesssim 0.5$, where $x_0 = Q_0/(kT)^{1/2}$ and Q_0 is the coordinate of the Jahn-Teller minimum. Since the variation $\Delta F(x)$ due to a second-moment increase is nearly proportional to the second derivative $F''(x)$, we have computed this latter quantity which is shown in Fig. 2 for $x_0 = -0.2$.

The similarity between this diagram and the experimental $\Delta I/I$ spectra of Fig. 1 is self-evident, especially at low temperature. At higher temperature a zero-moment decrease is superimposed on the triplet structure, and can be accounted for by the analysis of Ref. 3. Another feature of Fig. 2 is the $\sqrt{T}$ dependence of the splitting,[7] which is in excellent agreement with the experimental behavior if the effective temperature, $T_{\text{eff}} = \theta \coth(\theta/T)$, $\theta = 62.5$ K,[8] is used (see Table I). The best fit between experiment and theory is obtained by assuming the electron-lattice coupling parameter $c = 1.37$ (eV)$^{1/2}$. Given the plausibility of this value, one could conclude that the model accounts not only for the temperature behavior of the splitting, but also for its order of magnitude.

However, we are aware that our model works only provided that the two upper branches of the triplet are appreciably populated; in our semiclassical treatment this means $|x_0| \lesssim 0.5$. This is not easily justified as the observed splitting (~0.1–0.2 eV) is much larger than the vibrational energy. In order to try the plausibility of this

TABLE I. Theoretical and experimental splittings for KCl:Tl.

T (K)	$T_{\text{eff}} = \theta \coth(\theta/T)$, $\theta = 62.5$ K (K)	$\Delta E \simeq 0.9c(kT_{\text{eff}})^{1/2}$ Theor. (10^{-2} eV)	Expt. (10^{-2} eV)
10	62.5	9	9
80	95	11	10
200	207	16	17
300	305	20	23

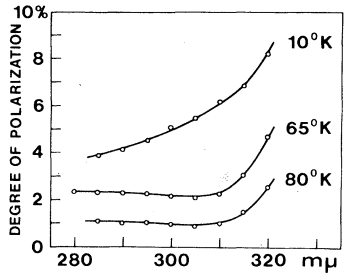

FIG. 3. Degree of linear polarization of the 3000-Å emission of KCl:Tl under linearly polarized excitation in the *A* band, as a function of the emission wavelength.

assumption we excited the phosphor by linearly polarized light and measured the degree of linear polarization of the emission, as a function of the emission energy. The results are displayed in Fig. 3 and show that the degree of polarization decreases considerably on the high-energy side of the spectrum. As the linear polarization is due to the presence of tetragonal minima, which can lie only on the lowest branch of the excited state, it seems reasonable to attribute the high-energy side of the spectrum to transitions from the two upper branches, which is consistent with our assumption.

The fact that no structure is observed in the emission bands without field apparently contradicts our mechanism, but there are several factors which mask the nonelementary character of the band, such as totally symmetrical and tetragonal vibrational modes. It is, however, still to be clarified as to how the electric field can discriminate among vibrational modes of different symmetries. On the other hand we would like to remark that the magnitude of the electric-field-induced structure on the absorption bands is practically independent of the presence of visible structure in the absence of field.[2,3]

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Divide all the four-wave interactions by 2 in Eqs. (10)–(13). The right-hand side of Eq. (14) and the left-hand side of Eqs. (15) and (16) should read $2[\gamma - (M^+ + M^-)N^{\pm}/\hbar]$ instead of $2[\gamma - 2|M^-N^{\pm}/\hbar|]$. The final result remains unchanged.