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LUMINESCENCE AND OPTICALLY DETECTED MAGNETIC RESONANCES OF CLOSE F CENTER PAIRS IN KCl[1)]

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In KCl crystals additively coloured and then quenched or X-irradiated at RT, the complex formed by 2 F centers having a separation distance between 40 and 80 Å is called a "distant F center pair". The luminescence response of this complex to magnetic and microwave fields has led to quite interesting phenomena as among other things the possibility to detect EPR in the F relaxed excited state and a better understanding of the mechanism of the concentration quenching of the luminescence[2)]. An appropriate optical treatment near RT allows to decrease the distance between the 2 members of the F center pair down to 4 interionic distances. The optical and magnetic properties of such a complex, called a "close F center pair" are drastically changed. The increase with magnetic field of the luminescence of distant pairs[2)] gradually diminishes to eventually change to a decrease when the concentration of "close pair" is enhanced. Parallelly the intensity of the EPR peaks ($g = 1.985$, $g^* = 1.981$) observed as a decrease of the luminescence disappears and a new single line appear with the opposite sign.

These new phenomena are related to changes in the population in the ground state caused by the optical pumping. They are very sensitive to the mixing of the spin states produced by the different interactions; in particular an exchange spin-spin interaction $J\ \vec{S}_i\vec{S}_j$ is effective in the relaxed excited state.

The field variations of the luminescence can be calculated for different states of aggregation with a good experimental agreement by assuming an exponential distribution of the J's : $p(J)dJ = \exp(-J/\bar{J})dJ/\bar{J}$ in order to take in account the distribution of the pair separation. The average exchange energy $\bar{J}$ lies between the hyperfine and the Zeeman energy.

The EPR is only observed in the ground state and usually consists of a single line. Its g factor is isotropic but is temperature dependent; it tends asymptotically at high temperature toward $g = 1.998$ with $W_{HW} = 52$ G. However when the exchange constant J is large a splitting in several lines is observed each one corresponding to a particular separation of the "close pair". The relaxation processes are also strongly modified and have been studied as a function of the magnetic field, the optical pumping rate and the state of aggregation.

Moreover ENDOR spectroscopy was detected by a spectral optical technique in order to study the "close pairs" in more details. The optical pumping leads also to an important nuclear polarization due to the spin-lattice relaxation. This new effect shifts the lines of the EPR spectra and also allows an optical detection of the NMR of the "close F center pairs".

A model for the strong field case (electronic Zeeman energy > hyperfine energy > exchange energy) explains quantitatively the observed ratios of the integrated spectra (NMR) : (ENDOR) $\cong$ 1 and (NMR) : (EPR) = 3 with H_x (microwave) = 0.5 G and H_x(RF) = 0.9 G.

REFERENCES

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