FACULTY OF SCIENCES

EPR STUDY OF LUMINESCENCE-BASED RADIATION DOSIMETERS

K. Maes[1], L. Lowie[1], J. Kusakovskij[1], L. de F. Nascimento[2], H. Vrielinck[1], F. Callens[1]

[1]GHENT UNIVERSITY DEPARTMENT OF SOLID STATE SCIENCES, EMR RESEARCH GROUP
[2]SCK•CEN BELGIAN NUCLEAR RESEARCH CENTRE, RADIATION PROTECTION DOSIMETRY AND CALIBRATION GROUP

Introduction

Electron paramagnetic resonance (EPR) is a physical method to observe resonant absorption of microwave radiation by unpaired electron spins in a magnetic field. It is able to detect, identify and quantify free radicals, such as those present in irradiated materials. This makes it a reliable dosimetric technique for retrospective/accident dosimetry, detection of irradiated food, e.g. using alanine, tooth enamel and sucrose. It finds applications in geology, chemistry, physics, medicine, environmental sciences, archaeology, and industrial irradiations. [1] In the present study, we discuss results on two classes of materials, i.e. LiF:Mg,Ti /LiF:Mg,Cu,P and Al_2O_3:C /Al_2O_3:C,Mg. These materials are used in thermoluminescence dosimetry (TLD) and Optically Stimulated Luminescence dosimetry (OSLD)/Radiophotoluminescence dosimetry (RPLD) respectively.[2-4] In terms of dose sensitivity EPR cannot compete with these luminescence methods, but it can provide complementary insight into the defects and processes leading to luminescence.

Al_2O_3:C /Al_2O_3:C,Mg

X-Band measurements

Both systems were measured before and after X-ray irradiation. Before irradiation, there was no significant EPR signal visible. After irradiation a broad isotropic signal appeared. Although the signal looks similar for both, they are in fact slightly different:

Al_2O_3:C,Mg
g = 2,011 lwpp = 4,3 mT
Al_2O_3:C
g = 2,008 lwpp = 5,3 mT

Their intensities are also different. The Al_2O_3:C,Mg EPR signal is smaller by a factor of around 2 for the same dose received.

Dose dependence

Only Al_2O_3:C is shown, Al_2O_3:C,Mg gave similar results.
Looking at the dose dependence there is a clear increase in intensity in function of dose received for both.

Plotting the EPR intensity in function of dose gives an exponential curve from which we could derive an estimate of the saturation dose:
Fit Al_2O_3:C,Mg:

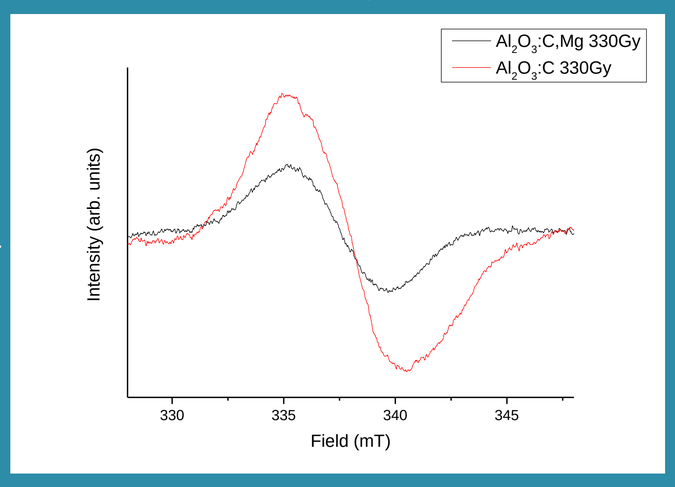
$$I = I_0 * (1 - e^{-\frac{D}{58{,}8\ Gy}})$$

Fit Al_2O_3:C:

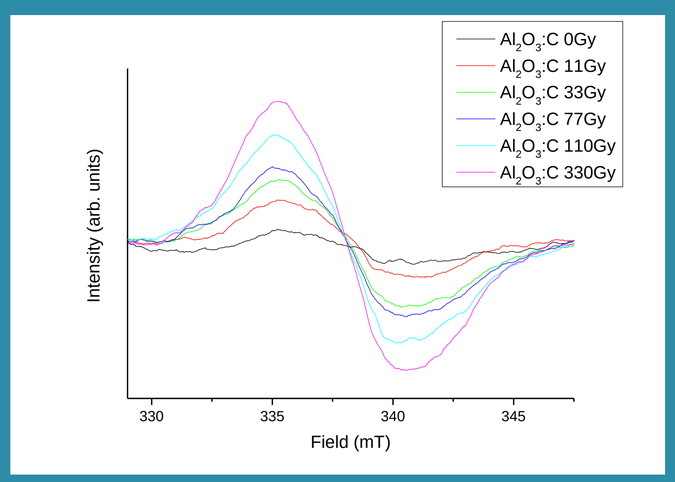
$$I = I_0 * (1 - e^{-\frac{D}{66{,}7\ Gy}})$$

For both samples the saturation dose is around 60 Gy, close to the OSL/RPL saturation dose.

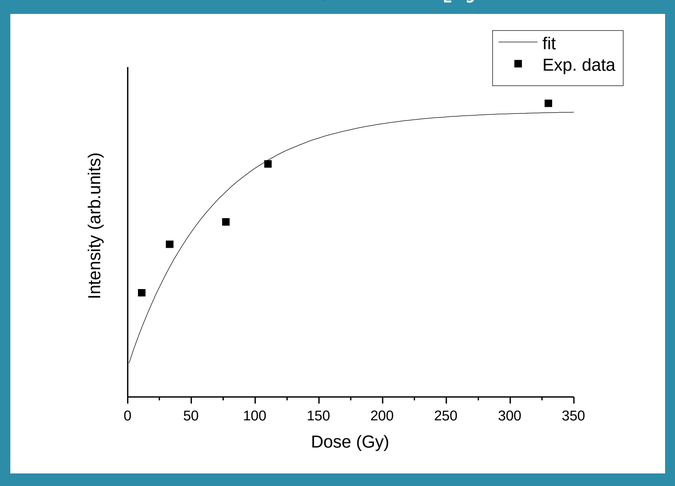
X-band EPR comparison, RT



X-band EPR dose dependence Al_2O_3:C, RT



Fitted dose dependence Al_2O_3:C

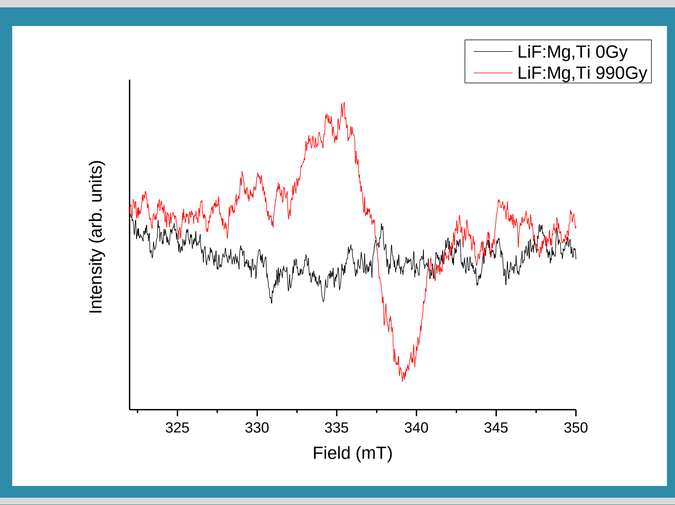


LiF:Mg,Ti

X-Band measurements, RT

Measurements before irradiation did not reveal a significant EPR signal.
After irradiation a signal appears, but even for 1 kGy of dose, the intensity remains very low.

g = 2,014 lwpp = 3,8 mT



LiF:Mg,Cu,P

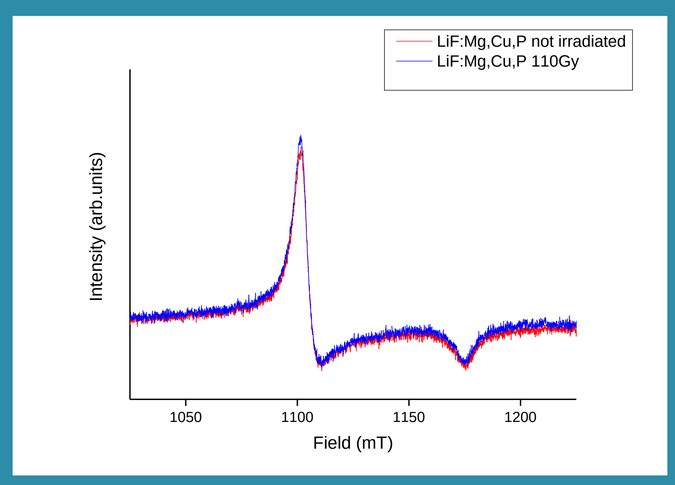
Q-Band measurements

For this system a signal is present before irradiation.
$g_\perp$ = 2,204 $g_{//}$ = 2,068
lw = 8,3 mT (Lorentzian)
Irradiating the sample did not change the intensity of the signal, nor its shape.

Literature

Patil and Moharil 1995 [5]
$g_\perp$ = 2,191 $g_{//}$ = 2,073 (X-band, RT)
The signal is proposed Cu-related.

Q-Band EPR, RT



Prediction g-values Cu^{2+}

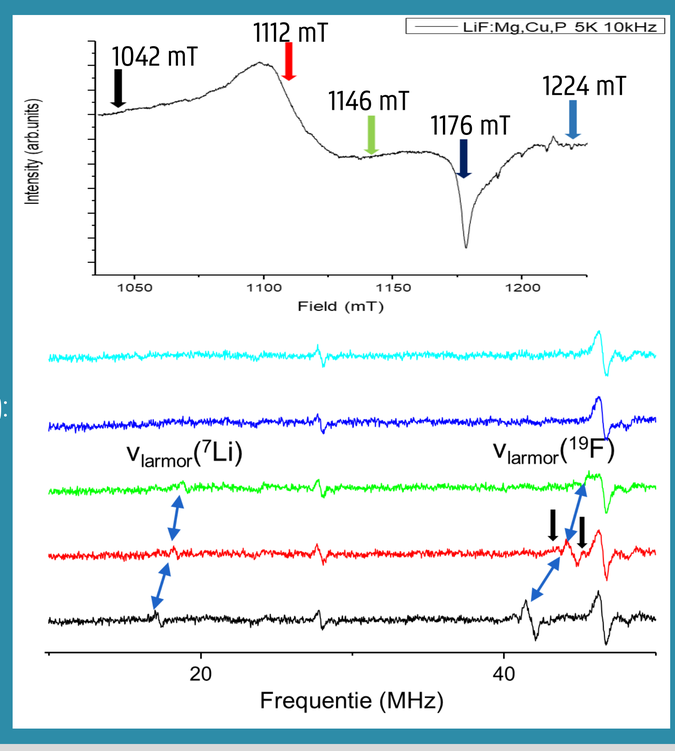
Crystal Field Theory	DFT calculations
$d_{x^2-y^2}$: $g_{//} > g_\perp > 2$	$g_\perp$= 2,115 < $g_{//}$=2,259
$d_{3z^2-r^2}$: $g_\perp > g_{//} \approx 2$	$g_\perp$= 2,219 >$g_{//}$=2,005

CFT predicts that the unpaired electron is either in $d_{x^2-y^2}$ or in $d_{3z^2-r^2}$ orbital. For these orbitals theoretical g-values can be calculated. DFT calculations confirmed the CFT predictions. However, comparing this to the experimentally derived g-values, neither really fit.

Q-band ENDOR measurements

Electron Nuclear Double Resonance (ENDOR): Detecting NMR transitions via EPR.
First results reveal signals at the Larmor frequencies of ^{7}Li and ^{19}F from distant nuclei. Around $\nu_{larmor}(^{19}F)$ a signal is visible with a hyperfine coupling of 0,9 MHz due to a neighbouring F nucleus.

Q-band EPR + ENDOR, 5K



Conclusions

- Al_2O_3:C and Al_2O_3:C,Mg have an EPR signal that is dose sensitive
  - The EPR signal of Al_2O_3:C is more sensitive to radiation compared to the EPR signal of Al_2O_3:C,Mg
  - The saturation dose is around 60 Gy, in agreement with OSL saturation dose
- LiF:Mg,Cu,P has an EPR signal present that is not dose sensitive
  - The signal could be related to Cu^{2+}, however more research is needed
  - The first ENDOR spectra look promising

Acknowledgements

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Contact

Kwinten Maes
PhD assistant
DEPARTMENT OF SOLID STATE SCIENCE
EMR RESEARCH GROUP
E kwinten.maes@ugent.be
T +32 9 264 43 51
www.ugent.be

Ghent University
@ugent
Ghent University

GHENT UNIVERSITY

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