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Autor: Azzoni, C.B. / Paleari, A.
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EPR STUDY OF X-IRRADIATED YTTRIA STABILIZED ZIRCONIA

C.B.Azzoni and A.Paleari, Dipartimento di Fisica A.Volta dell'Universita' di Pavia, via Bassi 6, 27100 Pavia, Italy

ABSTRACT: The EPR spectrum of X-irradiated crystals of yttria stabilized zirconia is put in relation with the disordered rearrangements of the anion sublattice around the defects.

INTRODUCTION

The cubic fluorite phase is stabilized in ZrO_2 by the presence of trivalent yttrium ions in the cation sites, producing a large concentration of oxygen vacancies. The study of the structure of the defect centers at the oxygen vacancies is fundamental for many technological applications⁽¹⁾.

RESULTS

We have performed ESR measurements on oriented single crystal of X-ray irradiated yttria stabilized zirconia; the anisotropic spectra, generally consisting of four signals, are analyzed in terms of a single paramagnetic defect with local axial symmetry in four equally probable orientations with respect to the crystal axes, as already proposed⁽²⁻³⁾. The observed signals have a variable linewidth, whose angular dependence has a π periodicity. The minimum width (0.7mT) is detected when the magnetic field B is parallel to a $\langle 111 \rangle$ axis, while the maximum (6mT) is reached for $B \perp \langle 111 \rangle$. Decreasing the linewidth, the lineshape becomes asymmetric.

DISCUSSION

The axially of the defect is accounted for by supposing that the EPR center experiences an axial crystal field: the cation site in proximity of an anion vacancy may constitute an electron trap subjected to the crystal field of seven coordinated oxygens; the unpaired spin should be a 4d electron of a Zr^{3+} in a distorted cubic electric potential. In such a system one should observe $g_{\parallel} > g_{\perp}$, where g_{\parallel} depends on the crystal field only at the second order while g_{\perp} at the first one.

We have simulated the experimental asymmetry of the signal allowing an angular distribution of the directions of the defect axis, indicating a detectable deviation (between 0° and $4^{\circ} \pm 2^{\circ}$) from the $\langle 111 \rangle$ directions. This feature of the signal may constitute an evidence of the disordered distribution of the rearrangements of the oxygen positions around the anion vacancies, causing a spread in the direction of the symmetry axis of the defects.

Taking into account the existence of distortions in the anion lattice around the oxygen vacancies, the model involving an oxygen vacancy near a zirconium in a $4d^1$ configuration, may also suggest a possible explanation of the angular dependence of the linewidth. In fact the different dependence of the principal values of the g-tensor from the crystal field parameters results in an inhomogeneous broadening of the orthogonal component only (deriving from a spread of the g_{\perp} -value), the g_{\parallel} -value being nearly unaffected by variations of the crystal field induced by the different anions environments.

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