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EPR SPECTRUM OF MANGANESE-DOPED
ZINC SELENIDE SINGLE CRYSTALS**

G. Hartung, et al

**Foreign Technology Division
Wright-Patterson Air Force Base, Ohio**

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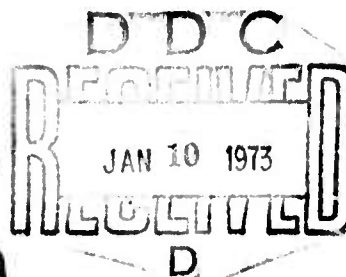
FOREIGN TECHNOLOGY DIVISION



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by

G. Hartung and W. Graf



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ZnSe single crystals, doped with 1% Mn show, besides 30 lines of the main EPR spectrum, several groups of 6 lines on both ends of the main signal. The main signal is explained by replacement of Zn^{2+} by Mn^{2+} ; the satellite lines result from a partial replacement of Se^{2-} by a single valent anion, like Cl^- . This causes in addn. to the cubic structure a trigonal structural element. A theoretical soln. for the trigonal structure is very complex and till now solved just for several different orientations of the crystal in the magnetic field.
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**ELUCIDATION OF SATELLITE LINES
IN THE EPR SPECTRUM OF MANGANESE-
DOPED ZINC SELENIDE SINGLE CRYSTALS**

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In the EPR spectrum of ZnSe:Mn^{2+} , in addition to the main lines, a great number of additional lines were found, which must be attributed to the Mn^{2+} -ions. The intensity of the satellites is 100 to 1000 times less than that of the main spectrum, which is explained through a pure cubic symmetry of the environment of these Mn^{2+} -ions. The symmetry of the environment of each of the Mn^{2+} -ions produced by the satellite spectrum is lower than cubic. Investigations have proven that this lower symmetry which occurs does not arise from defective structures - such as in similar appearances in the ZnS:Mn^{2+} - but rather foreign substitutes in the close vicinity of these Mn^{2+} -ions are responsible for this.

Experimental Data and Preparation

All investigations were carried out with an X-band-spectrometer, with which the specimen could be rotated around a vertical axis with reference to the magnetic field. The temperature at the specimen site amounted to about 300°K and 200°K in the experiments. The single crystals were grown in accordance with the sublimation process according to Piper/Pollich by Certified Chemist Huettich in the Phytel Section of the Ilmenau Technical High School. The appropriate manganese doping of 10^{-2} g Mn/lg ZnSe was admixed as a chloride to the beginning materials before the sublimation. The single crystals were supplied in pear shaped pieces. Through repeated splitting, a cubic habitus of the measuring specimen could be prepared, which made possible an oriented mounting in the cavity resonator.

Experimental Findings

During the exposure of angle dependences of the spectral lines, the expected Mn^{2+} spectrum was found to be in agreement with the statements published in literature. With the aid of this well known spectrum, encompassing 30 lines, the final positioning of the angle position of the crystal with the required accuracy resulted. At a 100 to 1000 times increase of the spectrometer sensitivity, several groups of six lines each appeared at magnetic fields above and below the just mentioned main spectrum. The six lines have an equal spacing among themselves, which additionally agrees with the spacing of the hyperfine structure lines in the main spectrum. The groups have differing intensities and display an angle dependence. In the four orientations of the crystal, in accordance with $[111]$, $[\bar{1}11]$, $[1\bar{1}1]$ or $[11\bar{1}]$, parallel to the applied magnetic field, the satellite spectra are equal and display extreme positions. A better resolution

of the individual lines could be achieved in that the crystal was cooled to a temperature of about -100°C , since thereby the line width diminished. The quantitative determination of the characteristic parameter of the satellite spectrum, which is attributed to Mn^{2+} -ions with an environment of axial symmetry, is presently taking place.

Model Conception

The examined ZnSe crystals have blende structures. The Mn^{2+} -ions replace the Zn^{2+} -ions. One such Mn^{2+} -ion is then found in the center of a tetrahedron, on which corners four Se^{2-} ions are arranged. This type of installed Mn^{2+} -ions cause the main spectrum. If any one of the selenium ions is replaced by a foreign anion, the existant cubic symmetry of the electrostatic potential, in which such a Mn^{2+} -ion resides, is destroyed. The new situation is thereby characterized in that an axial potential component is added to the cubic potential component, which henceforth is responsible for a trigonal symmetry of the environment. The connection line between the foreign anion and the Manganese ion is represented by the trigonal, which simultaneously has the direction of a space diagonal in the unit cell of the host lattice (Figure 1).

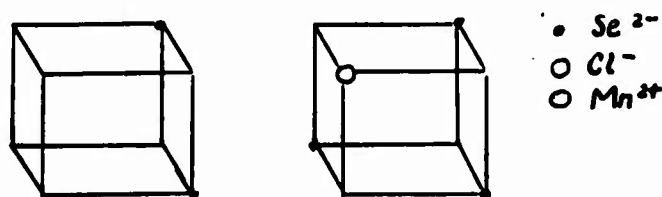


Fig. 1. Paramagnetic Mn^{2+} - center in cubic (left) and trigonal (right) environment.

The available Cl^{-} -ions are assumed to be the foreign anions. Because all four selenium positions have equal rights, the Cl^{-} -ions

here in question are statistically distributed among the four possibilities. In the iso-structural ZnS, similar satellite spectrums are also explained by axial potential component in the environment of Mn^{2+} -ions. The model based thereupon brought disorder manifestations into the host lattice. No preferenced space diagonal of the unit cell and no sign of twin formation in the main spectrum could be proven, however, in the investigated ZnSe crystals.

Theoretical Principle

For mathematical treatment of the problems, the usual methods of the Spin-Hamilton-operator is utilized. In paramagnetic centers in the S-condition, as is here appropriate for Mn^{2+} -ions, the Spin-Hamilton-Operator assumes the following form:

$$\begin{aligned}
 H = & g\beta(H_x S_x + H_y S_y + H_z S_z) + D[S_z^2 - \frac{1}{3}S(S+1)] + AS_z I_z + \\
 & + B(S_x I_x + S_y I_y) + \frac{A}{6}[S_1^4 + S_2^4 + S_3^4 - \frac{1}{3}S(S+1)(3S^2+3S-1)] + \\
 & + \frac{F}{180}[35S_z^4 - 30S(S+1)S_z^2 + 25S_z^2 - 6S(S+1) + 3S^2(S+1)^2] + \\
 & + Q[S_z^2 - \frac{1}{3}I(I+1)] - \gamma A_N I
 \end{aligned}$$

For the special case of the cubic symmetry, the constants D, B, F and Q disappear. To indicate the axial case, they must be brought in and receive the values determined by the experiments. For the practical computations for determining the energy level and resonance transition, the Operator, as given above, must be subjected to a few more transformations and recalculations. In the trigonal case, because of the high computation effort, the solution of this problem is found in literature for only a few distinct angle positions of the crystal in the magnetic field.