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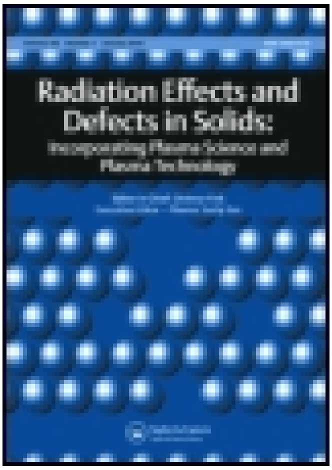

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Defect formation and VUV luminescence in BaF 2

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DEFECT FORMATION AND VUV LUMINESCENCE IN BaF_2

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Nominally pure BaF_2 single crystals were investigated at 77 K with optical absorption and electron paramagnetic resonance to understand the mechanism of radiation damage. We find that X-irradiation at 77 K of undoped BaF_2 produces V_k- and F-centres having absorption bands at 3.4 and 2.3 eV respectively.

Keywords: Barium fluoride; Absorption; EPR

1. INTRODUCTION

BaF_2 single crystals are applied as scintillation materials for gamma ray detection in many high-energy physics experiments. Fast timing is possible with these crystals because the very short decay time of about 0.8 ns of luminescence at 195 and 220 nm which is due to cross-luminescence transitions. BaF_2 also has an intense, slow emission component at 300 nm with a decay time of about 620 ns at room temperature.

In recent years most of the attention has been focused on the scintillation mechanism in this crystal. Nevertheless, there are many outstanding questions on the processes of the radiation defect creation in undoped and doped barium fluorides. The basic part of the studies has been made thirty years ago (see for example [1]) and interpretation of the results was complicated by the problem of sample purity. Frankly, this problem always remains unsolved.

Alkaline earth fluoride crystals which have not been deliberately doped with impurities are much less susceptible to coloration at room temperature by X-rays than most alkali halides. Undoped CaF_2 and SrF_2 crystals may be colored by X-rays much more readily at 77 K than at room temperature but the coloration efficiency, especially in the case of CaF_2 is still much slower than in most alkali halides. Only a weak spectrum of self-trapped holes was visible after the X irradiation for 11 h at 20 K of pure CaF_2 crystals; no trace of F-centers could be found [2]. This small concentration of V_k-centers may arise from residual concentrations of electron trapping impurities in the crystals.

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The sole exception is the crystals of pure BaF_2. After X-irradiation of undoped BaF_2 crystals at 77 K intensive absorption bands at 3.4 and 2.3 eV are found. In the early works it was suggested that the absorption responsible for the peak at 3.4 eV is associated with the presence of a broad EPR line-with $g = 1.98$ in the irradiated crystals which arises from trapped electrons. Authors concluded that the centers responsible for this band may be associated with an unknown chemical impurity in the crystals [3].

This paper contains results of an investigation of radiative point defects in undoped BaF_2 crystals. We find that X-irradiation at 77 K of undoped BaF_2 produces V_k- and F-centres having absorption bands at 3.4 and 2.3 eV respectively. Therefore, this absorption is associated with intrinsic defects rather than with an unknown chemical impurity.

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2. EXPERIMENTAL TECHNIQUE

The crystals of undoped BaF_2 were grown using the combined Shteber-Stocbarger method. The samples were of high optical quality and no indication of oxygen contamination. The absorption spectra were taken with a "Specord" spectrophotometer. The crystals were irradiated at 77 K by X-rays from a Pd tube operating at 40 kV and 50 mA for 30 min.

3. EXPERIMENTAL RESULTS

X-irradiated BaF_2 crystals at 77 K show optical absorption having two maxima at 3.4 (365 nm) and 2.3 eV (540 nm) (Fig. 1). Mutual annihilation of the centers related with these absorption bands occurs below 130 K that correlates with thermal destruction of the V_k-centers.

When the undoped barium fluorides are X-irradiated at 77 K the V_k center is predominant hole center formed and the TL spectrum exhibits the intense glow peak at about 110 K, which

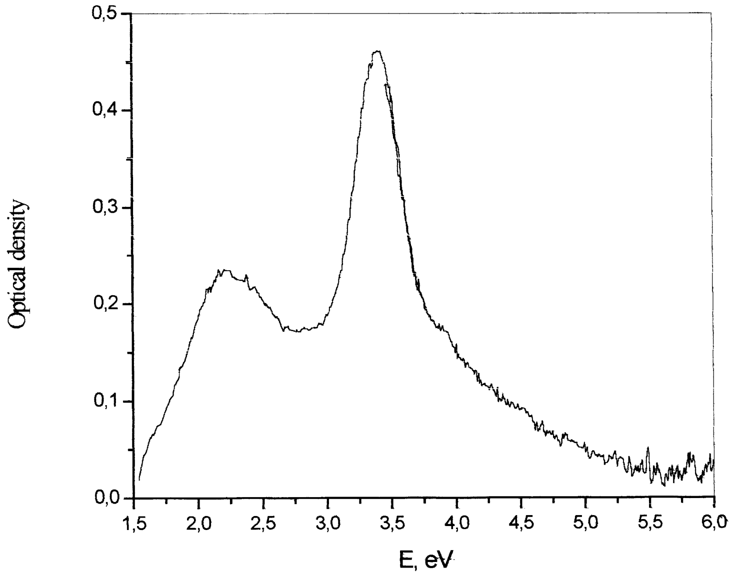

FIGURE 1 Optical absorption of pure BaF_2 crystal after X-irradiation at 80 K.

is associated with the destruction of V_k centers. Aside from this there are additional weaker features above 200 K [4].

The EPR spectrum of pure BaF_2 crystals after X-irradiation at 77 K is shown in Figure 2. It can be seen that X-irradiation of undoped BaF_2 at 77 K produces V_k- and F-centers. The EPR spectrum of V_k-centers in our X-irradiated undoped BaF_2 has the same g and A values and general appearance as that of V_k-centers in BaF_2-Tm^{3+} crystals [5]. However, no absorption bands at 3.7 eV, which, in authors view [5] was associated with V_k-centers are produced by X-irradiation at 77 K of the undoped BaF_2 crystals.

The weak EPR F-centre lines in BaF_2 are superimposed on intensive EPR lines of V_k centers. No absorption band at 2.03 eV (611 nm), which in authors view [3] was associated with F-centers, is produced by X-irradiation at 77 K of the undoped BaF_2 crystals. The absorption band at 2.03 eV (611 nm) is only found in the BaF_2 crystals doped with lanthanum X-irradiated at 77 K. Thermal and optical destruction of the EPR spectrum correlates with thermal and optical destruction of the centers responsible for the absorption bands at 3.4 and 2.3 eV. From this it may be concluded that X-irradiation of undoped BaF_2 at 77 K produces V_k-centers with absorption band at 3.4 eV and F-centers with absorption band at 2.3 eV. The absorption band at 2.3 eV probably arises from an F-centre which is associated with negatively charged fluorine interstitials in distinction from the absorption band at 2.03 eV (611 nm).

In BaF_2 crystals a fast ultraviolet emission with two main maxima at 6.3 and 5.6 eV under x-ray excitation or particle irradiation is due to cross-luminescence transitions. In our previous work [6] we found that in the BaF_2 crystal doped with lanthanum a weak additional high-energy emission appears and its intensity grows with increasing La-doping. The emission arises from a radiation transition between the electron state of the interstitial F_i^--ion, its electron state is located in the forbidden band at low concentration of lanthanum, and the outermost Ba^{2+} 5p core band.

In the BaF_2 crystals doped with yttrium the high-energy emission band is also detected. As in the case of La-doping, a slow component of the luminescence at 4.0 eV, which is caused by self-trapped excitons, is suppressed by doping with Y. In both cases the high-energy emission, as the main cross-luminescence, is temperature independent at least from 77 to 300 K.

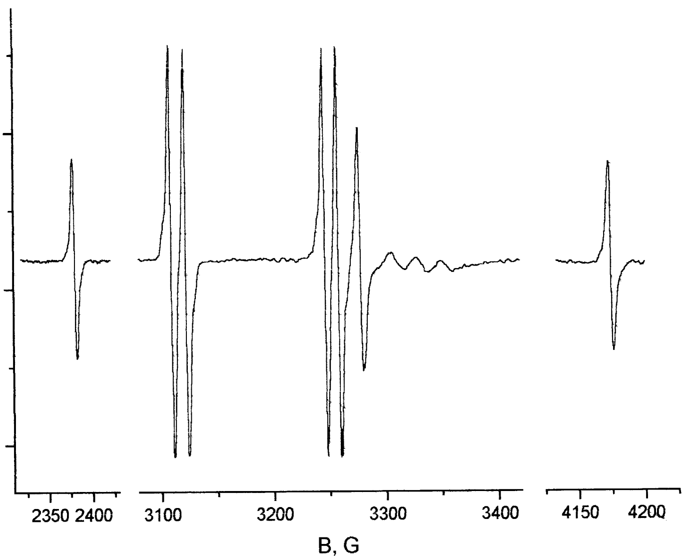

FIGURE 2 EPR spectrum of pure BaF_2 crystal after x-irradiation at 80 K, B ∥ ⟨100⟩.

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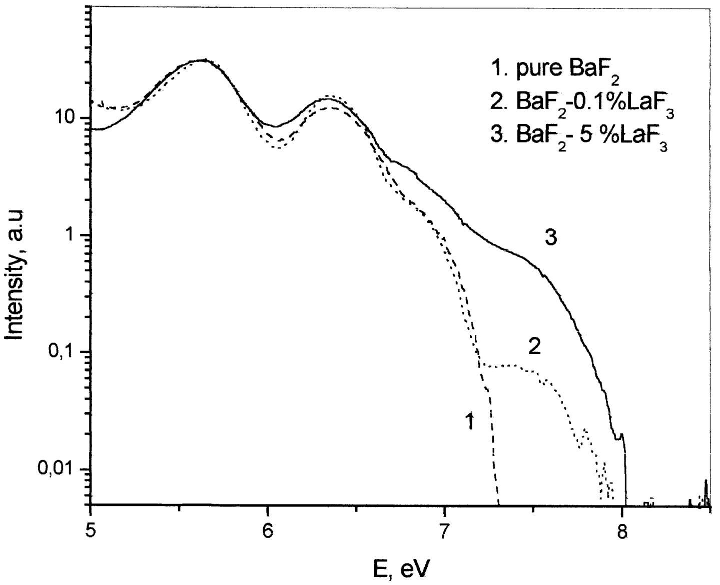

FIGURE 3 X-ray excited luminescence spectra at 80 K of La-doped BaF_2 crystals.

Unfortunately, in our previous work [6] the wave-length scale of our monochromator was in error. Now we present the revised spectra (Fig. 3) of the high-energy emission band which has maximum at about 7.5 eV.

In pure BaF_2 crystals under X-ray excitation at both 77 and 300 K the high-energy emission band is not observed. However, in some cases the high-energy emission band with maximum at about 7.5 eV was detected using 6 keV electron excitation [7] or synchrotron irradiation [8] of pure BaF_2 crystals. The emission is observed in the temperature range below 120 K. This temperature behavior is not similar to that of the cross-luminescence. This is due to the fact that the negatively charged fluorine interstitials can be created not only by doping with trivalent ions, but also by radiation damage.

4. CONCLUSION

X-irradiation of undoped BaF_2 at 77 K produces V_k-centers and F-centers. The V_k-centers and F-centres begin to disappear due to mutual annihilation at 100 K and have completely disappeared by 130 K. The strong resistance of undoped BaF_2 to coloration by X-rays at room temperature is apparently due to recombination of electrons and holes created by the irradiation.

The production efficiency of V_k- and F-centers by X-rays in our undoped BaF_2 at 77 K is at least an order of magnitude greater than in undoped CaF_2. This suggests, in contrast to CaF_2, that interstitial anion-vacancy pairs with sufficient separation to trap electrons may be created by X-rays in BaF_2.

$$e^0 \rightarrow e^0(F - H) \rightarrow \alpha - I$$

The anion vacancy can then trap an electron to become an F-center.

$$\alpha - I + e^- + e^+ \rightarrow F + F_i^- + V_k$$

The distinctions between the absorption spectrum of the X-ray induced *F*-centers in BaF_2 and that of the *F*-centre in additively colored BaF_2 could be due to anion interstitials.

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