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Thermodynamic models of radiation-induced processes in solids

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Abstract. A thermodynamic model is proposed to qualitatively describe the radiation-induced processes in solids: temperature dependence of the X-ray radio luminescence output, dependence of these processes on the excitation density, energy accumulating in a solid under exposure to ionizing radiation and its temperature dependence. The proposed model and the formula derived can be used to develop radiation-resistant and radiation-sensitive materials.

1. Introduction

Mathematical and computer modeling of radiation defects in solids has been widely researched in a number of papers, among which we particularly mention the papers [1–3] which provide an extensive bibliography. Nevertheless, the development of high-quality analytical models of radiation-induced defect formation is advantageous for both theoretical considerations and practical applications.

In [4], we addressed the issue of the response of the defect subsystem in solids to external effects (temperature, pressure, light, etc.) using a nonequilibrium statistical thermodynamics approach.

Crystal defects (color centers, etc.) were considered as a non-interacting particle system immersed in a thermostat. Quantum transitions caused by the interaction of defects with the thermostat are dissipative (with probability of P_0) in contrast to those occurring during the interaction with an external field (with probability of P_1). The dissipative processes always make the secondary field (system feedback) smaller than the primary one that causes defects formation.

For the response function $F = P_0/P_0 + P_1$, the obtained expression is

$$F = \frac{1}{1 + \frac{\tau_p}{\tau} \frac{2\Delta S}{k} \exp\left(-\frac{E_m - G^0/N}{kT}\right)}, \quad (1)$$

where τ is radiative lifetime, τ_r is relaxation time, ΔS is the entropy change during the radiationless transition, k is the Boltzmann constant, E_m is the energy of the defect ground state, G^0 is the thermostat Gibbs energy, N is the number of defects and T is temperature.

In this paper, we attempt to show how to use thermodynamic model (1) to analyze the radiation-induced processes in solids.

2. Temperature dependence of the radio luminescence yield

The simplest pattern of the temperature dependence of the luminescence yield can be observed for sufficiently pure crystals (Fig. 1).

The nature of this luminescence is well-established: the emission appears during radiative annihilation of the self-trapped exciton caused by the electron recombination with the V_K -center. This recombination may occur at different points of the lattice without migration losses. The shape of the dependence $I(T)$ is characteristic of the intra-center processes, and it can be well described by the Mott formula:

$$I(T) = \frac{I_0}{1 + A \exp(-q/kT)}, \quad (2)$$

where q is the activation energy of the nonradiative transition, A is a constant, T is temperature and k is the Boltzmann constant.

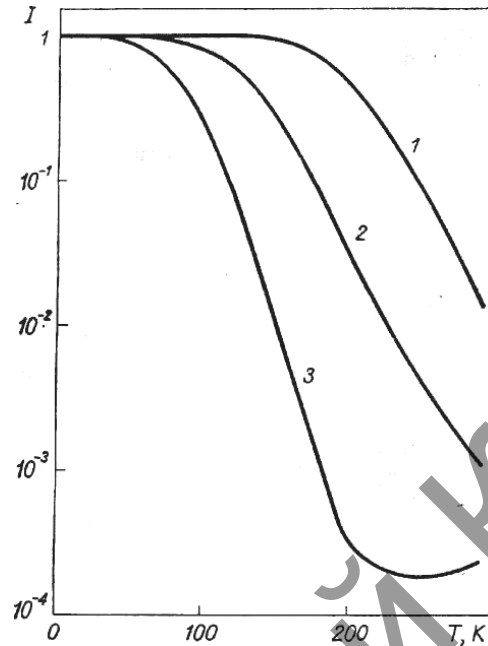


Figure 1. Temperature dependence of the luminescence intensity for pure NaBr (1), LiCl (2) and KI (3) crystals excited by X-irradiation [5].

The Mott formula coincides in its form with formula (1), where the response function $F=I/I_0$. However, in this case, the activation energy

$$q = \frac{E_m - G^0 / N}{kT} \quad (3)$$

is related to both the luminescence center parameters (E_m and N), and the crystal properties, the matrix (G^0 and T).

Equations (1) and (3) enable interpretation of the difference in the X-ray luminescence quantum yield and its temperature behavior for different crystals that cannot be directly derived from (2).

Equation (3) shows the dependence of the activation energy q for temperature quenching on the number of luminescence centers N even in the absence of coagulation processes. This does not correlate with the conventional model of the luminescence center potential curves.

We found out this effect experimentally long time ago [6], and it is illustrated in Fig. 2.

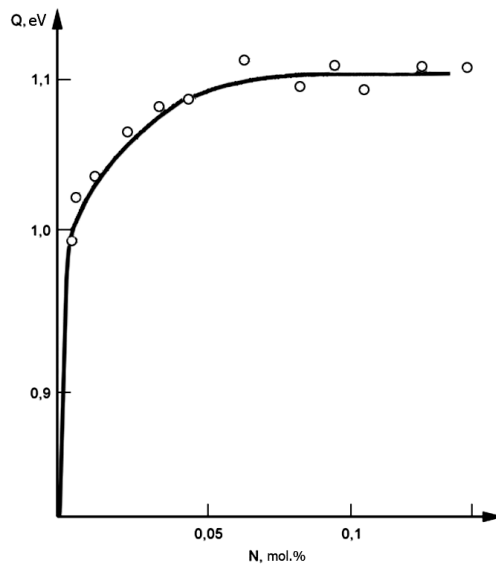


Figure 2. Dependence of the activation energy Q (eV) on the luminescence center concentration N (mol.%) for KBr-Tl [6].

3. Dependence of the crystal x-ray luminescence quantum yield on excitation density

In case of one type of defects present in the solid, the defect concentration N is proportional to the excitation intensity, and the dependence for the recombination luminescence yield obtained for the AHC series by (1) is shown in Fig. 3.

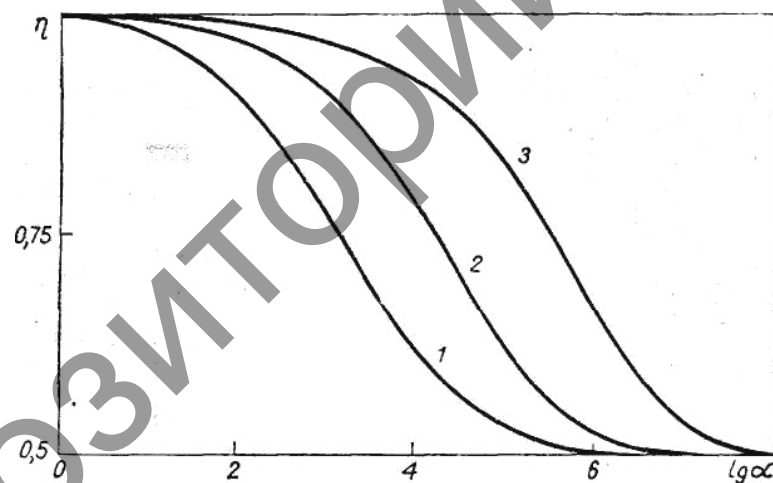


Figure 3. Dependence of the luminescence yield on the excitation density for KCl (1), KBr (2) and KI (3) [5].

The assumptions are rough, and in practice the dependence $\eta(e)$ is much more complex, however, the pattern presented in Fig. 3 indicates that the greater the Gibbs energy of the host crystal, the greater the excitation intensity required for recombination luminescence quenching.

4. Storage of energy in dielectrics under the exposure of ionizing radiation

Equation (1) can be reduced to a linear form under certain assumptions. Then, taking the stored energy S as a response function, we get:

$$S = b \frac{kT}{G^0} \cdot N, \quad (4)$$

where b is the constant.

At low irradiation doses, the energy storage is a linear function of the irradiation time (Fig. 4).

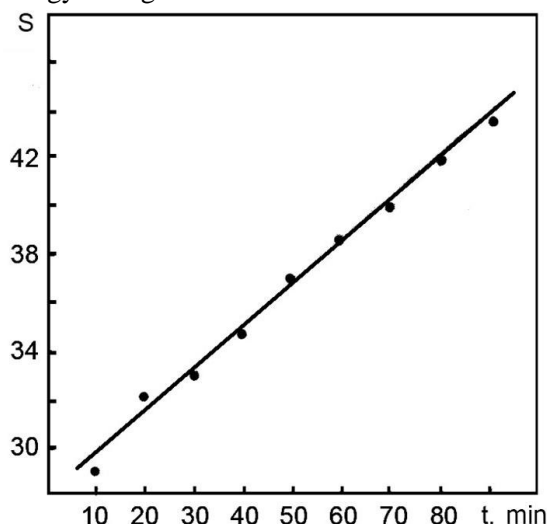


Figure 4. Dependence of the stored light sum in the TSL peak $T=200$ K for the crystal BiOCl on the X-raying time.

The rate of the defect accumulation is determined by the slope of the curve (4):

$$v_d = t g \alpha = a \frac{kT}{G^0} \quad (5)$$

Table 1 shows the values v_d for the AHC series obtained from the experimental data.

Table 1. – The defect accumulation rate and Gibbs energy for the AHC series.

Crystal	Defect accumulation rate, v_d , $\text{cm}^{-3} \text{c}^{-1}$	ΔG^0 Gibbs energy, kJ/mol
NaCl	$0.7 \cdot 10^{-13}$	-356.2
KCl	$3 \cdot 10^{-13}$	-409.05
KBr	$2 \cdot 10^{-13}$	-380.7
KI	$0.3 \cdot 10^{-13}$	-323.24

Table 1 shows that the greater absolute value of Gibbs energy results in the greater rate of defect accumulation due to the negative Gibbs energy. Since the defect accumulation rate defines the crystal radiation resistance, the crystals with greater absolute values of Gibbs energy will be radiation resistant. For example, oxides of most of the elements (MgO, Al₂O₃, etc.) have greater absolute values of Gibbs energy and, hence, these are radiation resistant. Note that $G^0 = \mu$ (chemical potential) for metals.

The temperature dependence of the F-center generation can be divided into three stages [5]: the first stage is linear dependence, the second stage is temperature independence, and the third stage is the decrease with the increased defect concentration. The complexity of the dependence of the F-centers generation is considered to be caused by the change in the lifetime of various halogen defects complementary to F-centers. We offer another interpretation of this effect based on formula (4). In fact, the dependence of Gibbs energy on the temperature is of the form:

$$G^0 = A + BT + CT^2, \quad (6)$$

where A, B, C are constants.

According to formula (4), we have a linear dependence of energy storage on temperature at insignificant $T - A > BT + CT^2$. If $G^0 \approx BT$, S does not depend on temperature. At greater T, Gibbs energy $G^0 \approx CT^2$ and $S \sim 1/T$, that is the energy decreases hyperbolically with the increased temperature grows. A qualitative pattern of the temperature dependence S is shown in Fig. 5.

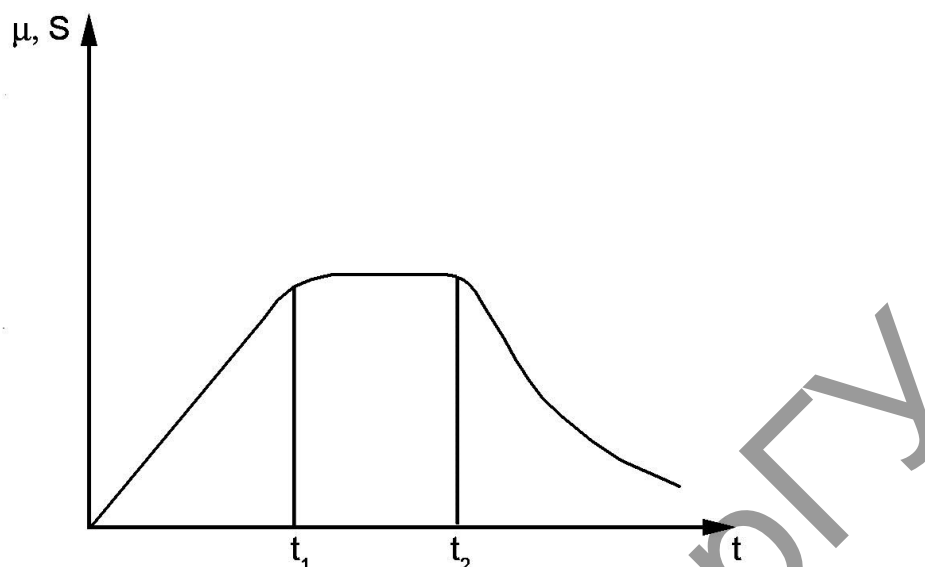


Figure 5. Three stages of temperature dependence of the optical absorption coefficient (μ) of F-centers and stored energy (S) on temperature (t C).

5. Conclusion

The study results show that the proposed thermodynamic model enables qualitative description of numerous radiation-induced processes in solids under exposure to ionizing irradiation. Considering that Gibbs energy $G^0=U-TS+PV$ (U is internal energy, T is temperature, S is entropy, P is pressure and V is volume), formulas (4) and (5) can be used to perform the analysis of the effect of pressure (P), phase transitions (S) [7-9] and other effects in solids exposed to ionizing irradiation.

Acknowledgements

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