

MANIFESTATION OF STRUCTURAL FEATURES OF GARNETS IN THE RELAXATION KINETICS OF PHOTOINDUCED PHENOMENA

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The relaxation kinetics of a long-lived addition to optical absorption, caused by the irradiation of the garnets $\text{NaCa}_2\text{Mn}_2\text{V}_3\text{O}_{12}$, $\text{Ca}_3\text{Mn}_2\text{Ge}_3\text{O}_{12}$ and $\text{Ca}_3\text{Ga}_{2-x}\text{Mn}_x\text{Ge}_3\text{O}_{12}$ with visible light, is explored and connected with the structure of garnets. A nontrivial character of the relaxation kinetics, common for all the garnets examined, is associated with their common structure element, viz., the anion group O_{12} forming the sublattice of oxygen ions O^{2-} . The relaxation kinetics observed mirrors the creation and motion of oxygen hole polarons in the oxygen sublattice, while the cation composition of garnets determines the hole hopping barrier varying in rather narrow limits. The role of the oxygen sublattice is convincingly corroborated by energy considerations.

Introduction

Since the seventies, long-lived photoinduced phenomena in garnets were observed and studied [1–14]; photoinduced changes in optical and magnetic properties were generally associated with the transfer of photoproducted charges. There exists also another class of oxide crystals subject to a long-lived effect of illumination, viz., copper-oxide superconductors extensively explored in the nineties [15–25]; it was disclosed that illumination affects their transport and magnetic properties through the creation and rearrangement of holes in the oxygen sublattice of the conduction plane. In 1999, the authors [25] draw attention to some related features of electronic processes in these two classes of oxide crystals. One could suppose that the oxygen sublattice of garnets is also involved in the mechanism of photoinduced phenomena through creating oxygen holes. The present paper is aimed to investigate the role of the oxygen sublattice in photoinduced phenomena in garnets.

A large variety of garnets with the same anion group O_{12} and different cation composition provides a favorable possibility to trace the role of the oxygen sublattice. To adequately realize this possibility, it is necessary to divide the entire variety of garnets into two subclasses as follows.

1. The garnets with two (or more) magnetic sublattices, whose interaction results in a non-compensated magnetic moment, are ferrimagnets. They have a rather high temperature of magnetic ordering exceeding the upper temperature boundary of the existence of long-lived photoinduced phenomena. In such crystals, interrelated photoinduced changes in optical and magnetic properties make up a complicated physical picture difficult for microscopic interpretation. For example, the illumination of yttrium iron garnet $\text{Y}_3\text{Fe}_5\text{O}_{12}$ with linearly polarized light induces a change in magnetic anisotropy [1–3], linear dichroism [3] and domain structure [4]. Unpolarized light affects magnetic permeability and susceptibility [5,6], coercivity and mobility of domain walls [7,8], magnetostriction [9], and optical absorption [10,11]. The garnets of this type are inconvenient for exploring the role of the oxygen sublattice: it can be masked by the influence of magnetic ordering of the cation magnetic sublattices.

2. The garnets with a single magnetic sublattice (formed by a rare-earth or transition metal) are, as a rule, antiferromagnets with a low temperature of magnetic ordering [26]. Their advantage is a broad temperature region above the Neel temperature where long-lived changes in optical properties are observed in the absence of magnetic ordering. In this temperature region, the mechanism of photoinduced optical phenomena can be explored without making

allowance for their complicated interconnection with photoinduced changes in the magnetic structure.

In the case of antiferromagnetic garnets, photoinduced phenomena can be explored in a convenient logical sequence: at first, the mechanism of pure optical phenomena is studied above the Neel temperature [12–14], which can strongly facilitate the subsequent investigation of the influence of magnetic ordering on optical properties below the Neel temperature.

In the present paper, we restrict ourselves to the variety of antiferromagnetic garnets; it is broad enough to provide a possibility to trace the role of the invariable anion group O_{12} , forming the oxygen sublattice, in the mechanism of photoinduced optical phenomena.

The purpose of this paper is to elucidate the connection between the structural features of garnets and photoinduced phenomena in the temperature region above the Neel temperature. This connection is mirrored by the relaxation kinetics of photoinduced absorption that is determined by the motion of photoproduced charges sensitive to structure.

To achieve this aim, we will use our previous experimental results, related to photoinduced absorption in the garnets $Ca_3Mn_2Ge_3O_{12}$ and $Ca_3Ga_{2-x}Mn_xGe_3O_{12}$ ($x \leq 0.02$), together with the disclosed mechanism of its formation [12–14]. To elucidate the connection between structure features and relaxation kinetics, it is helpful to expand the variety of examined garnets. To this end, in addition to the above garnets, we will study in this paper also the garnet $NaCa_2Mn_2V_3O_{12}$ with an essentially different chemical composition. After a brief description of the experimental technique (Sec. 2), we will present in Sec. 3 new data on the relaxation kinetics in $NaCa_2Mn_2V_3O_{12}$ and compare them with the kinetics of photoinduced absorption in other garnets. When doing so, the general mechanism of photoinduced phenomena in garnets [12–14] will be concisely summarized. On the base of this material, the connection between structural features and relaxation kinetics will be established in Sec. 4.

1. Experimental

Single crystals of the garnet $NaCa_2Mn_2V_3O_{12}$ were grown in a platinum crucible from a melt solution by the method of spontaneous crystallization on a seed at a slow cooling. Melted salts of potassium, sodium vanadates, and their mixtures were used as solvents. The solubility of the garnet forming oxides in the salt melt was 25 to 35 weight % at a temperature of 1100 °C. The

crystallization region of the garnet $NaCa_2Mn_2V_3O_{12}$ was 800 to 980 °C or 750–960 °C for the solvents KVO_3 or $NaVO_3$, respectively. Single crystals of the garnet with the size of 3 to 8 mm and mainly faceted by the plane $\{211\}$ have been obtained.

The samples for optical studies were prepared in the form of 25–50 μm thin plates cut perpendicularly to the $[100]$ axis. The plates were mechanically polished.

The methods for growing single crystals of the $Ca_3Mn_2Ge_3O_{12}$ and $Ca_3Ga_{2-x}Mn_xGe_3O_{12}$ garnets have been described in [14, 27].

The study of photoinduced phenomena was carried out with an optical double-beam setup. A helium-neon laser with the wavelength $\lambda = 633$ nm and flux density of 0.13 W/cm^2 was used as a source of pumping. As a probe light, we applied a stable wide-band emission of an arc xenon lamp having a maximum intensity at $\lambda = 590$ nm after dispersing through the diffractational monochromator. The intensity of the probe beam was low enough to cause no photoinduced phenomena. After passing through the sample, the probe beam was run through a second monochromator tuned to the wavelength of the probe light in order to cut off the scattered pumping light. After that, the probe beam was registered by a photomultiplier. The electric signal from the photomultiplier was amplified, transformed to digital form by an analog-digital converter, and transmitted to a computer.

2. Experimental Results. Relaxation Kinetics of Photoinduced Absorption

Photoillumination of the examined crystals $Ca_3Mn_2Ge_3O_{12}$, $Ca_3Ga_{2-x}Mn_xGe_3O_{12}$, and $NaCa_2Mn_2V_3O_{12}$ results in an increase of optical absorption in different spectral regions [12–14]. Photoinduced augmentation of absorption is conditioned by an enhancement of the oscillator strengths of different electronic transitions by photoinduced electric fields. In the case of $Ca_3Mn_2Ge_3O_{12}$, the forbidden transition $^5E_g - ^5T_{2g}$ between even states of Mn^{3+} ions is enhanced by the photoinduced electric field via adding an odd admixture to each of the even states. As a result, the absorption coefficient grows by a value of 20 to 90 cm^{-1} in the spectral region $550 < \lambda < 700$ nm [12]. In the case of $Ca_3Ga_{2-x}Mn_xGe_3O_{12}$, photoinduced enhancement of absorption (by roughly the same value) was observed in the region $300 < \lambda < 450$ nm corresponding to the allowed transition with charge transfer between O^{2-} and Mn^{4+} ions (photoinduced field shifts this transition from high frequencies, where absorption is stronger, to

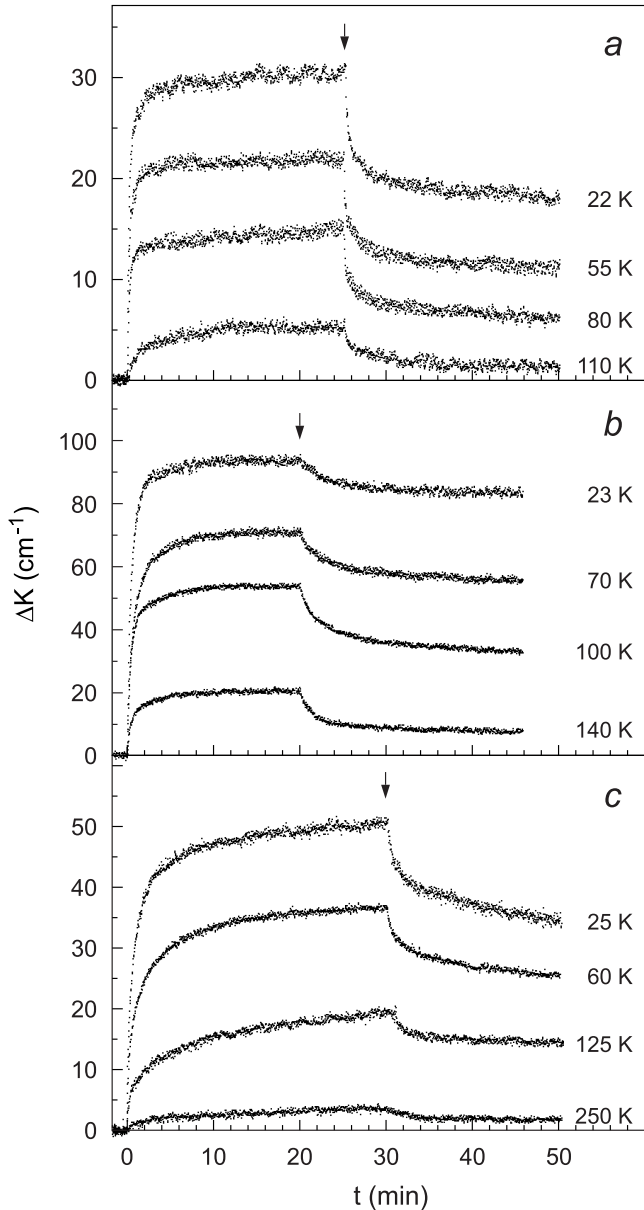


Fig. 1. *a* – time dependences of the photoinduced addition to absorption measured for the garnet $\text{NaCa}_2\text{Mn}_2\text{V}_3\text{O}_{12}$ at different temperatures indicated in the figure. *b*, *c* – the corresponding data for $\text{Ca}_3\text{Mn}_2\text{Ge}_3\text{O}_{12}$ [13] and $\text{Ca}_3\text{Ga}_{2-x}\text{Mn}_x\text{Ge}_3\text{O}_{12}$ with $x = 0.02$ [14] are shown for comparison. Arrows indicate the moment of switching off irradiation

the region of lower frequencies where absorption was registered) [14]. In the case of $\text{NaCa}_2\text{Mn}_2\text{V}_3\text{O}_{12}$, the photoinduced absorption was observed in the region $570 < \lambda < 800$ nm; the corresponding optical transition has not been established yet but it can

hardly be assigned to intraatomic d-d transitions in manganese ions entering $\text{NaCa}_2\text{Mn}_2\text{V}_3\text{O}_{12}$ in the form of Mn^{2+} (such transitions are twice forbidden by both spin and parity).

Thus, for the three garnets under examination, photoinduced augmentation of absorption ΔK is related to different optical transitions. Nevertheless, the relaxation kinetics of photoinduced absorption has essential common features.

For the three garnets, Fig.1 shows the time dependence of ΔK under pumping ($t \leq t_0$) and during the subsequent relaxation without illumination ($t > t_0$), measured at various temperatures (the moment, t_0 , of switching off illumination is indicated in the figure by arrows). Let us concentrate on the relaxation kinetics that bears the most important information.

Attention should be paid to the fact that the slope of the relaxation curves is great immediately after switching off irradiation and diminishes with time at $t > t_0$ very rapidly. It follows that the optical centers, responsible for the photoinduced addition to absorption, have a very broad distribution over relaxation time τ . The quantity τ , defined from the relaxation curve slope, varies in the range from a minute to at least several hours. At low temperatures, a predominant part of photoinduced absorption is produced by long-lived centers with the lifetime not less than several hours. A special analysis showed that the entire interval of τ is continuously filled in by relaxation components [14].

In view of such a broad continuous set of the relaxation decay times, one has to discard the usual mechanism of photoinduced absorption assigned to some new absorption centers produced by photoillumination (e.g., through creating lattice defects or a photostimulated chemical reaction involving impurities).

The total set of experimental data is naturally explained by the model of photoproduced hole polarons whose electric field plays a double role. First, it enhances the mentioned optical transitions causing a photoinduced augmentation of the absorption coefficient ΔK (estimations show that the number of holes, produced by the pumping intensity used, is sufficient to cause the observed values of ΔK [12,13]).

Second, the random electric fields of photoproduced holes condition the character of relaxation kinetics shown in Fig.1. Photoproduced holes in a polaronic state move over the lattice via hopping between adjacent sites. The hopping rate of hole polarons, Γ , is essentially enhanced by the random field, \mathbf{F} , of their charges:

$$\frac{1}{\tau} \equiv \Gamma = \Gamma_0 \exp\left(-\frac{Q - e\mathbf{F}\mathbf{a}}{2T_{\text{eff}}}\right) \quad (1)$$

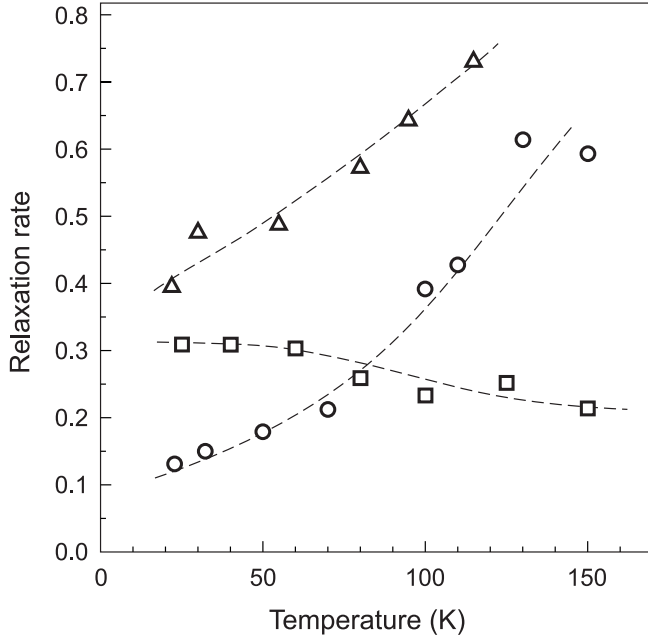


Fig. 2. Temperature dependences of the relaxation rate (2) for the garnets $\text{NaCa}_2\text{Mn}_2\text{V}_3\text{O}_{12}$ (triangles), $\text{Ca}_3\text{Mn}_2\text{Ge}_3\text{O}_{12}$ (circles), $\text{Ca}_3\text{Ga}_{2-x}\text{Mn}_x\text{Ge}_3\text{O}_{12}$ with $x = 0.02$ (squares) derived from the relaxation kinetics observed. Lines are guides to eye

(T_{eff} differs from the true temperature T by making allowance for the zero lattice vibrations at low temperatures). Eq. (1) describes the hopping of a hole with a charge e by a distance \mathbf{a} to an adjacent site in the direction of the applied field \mathbf{F} [13]. This random field, lowering the hopping activation energy Q by $e\mathbf{F}\mathbf{a}$, is the sum of the statistically independent electric fields of all holes. The distribution of such a sum over its magnitude is described by a broad Gaussian, which entails a broad continuous distribution of holes over lifetimes and the relaxation kinetics presented by Fig.1. The distribution of hole polarons over lifetimes, given by Eq.(1), coincides in main features with the expansion of the observed relaxation curve (Fig.1) over exponential components $\exp(-t/\tau)$ [14].

It is convenient to define the relaxation rate R with the use of experimental data as

$$R = \frac{\Delta K(t_0) - \Delta K(t_0 + 20\text{min})}{\Delta K(t_0)} \quad (2)$$

(t_0 is the moment of switching off illumination).

Fig.2 presents the temperature dependence of the relaxation rate (2) derived from the data of Fig.1. Except the case of garnet $\text{Ca}_3\text{Ga}_{2-x}\text{Mn}_x\text{Ge}_3\text{O}_{12}$, that

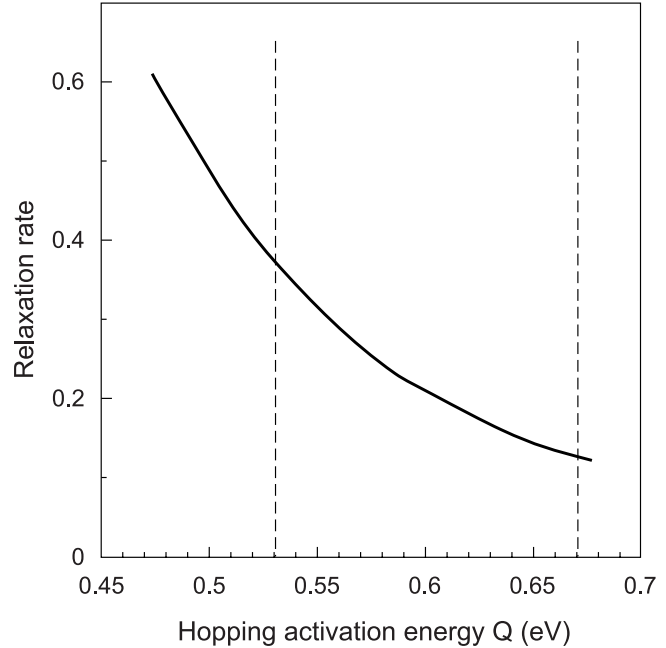


Fig. 3. Dependence of the relaxation rate (2) on the hopping activation energy Q obtained via solving the kinetic equation [12, 13] at a temperature of 23 K. Dashed vertical lines indicate the boundary of the region of the relaxation rates observed for all the three garnets

will be analyzed at the end of Sec. 3, the relaxation rate monotonously increases with temperature.

The observed kinetics of photoinduced absorption is quantitatively described by a kinetic equation with taking into account dependence (1) of the relaxation time on random electric fields and their time variation [12, 13].

The kinetic equation enables one to connect the relaxation rate (2) with the hopping activation energy Q appearing in (1). The calculated dependence of the relaxation rate R on activation energy, related to a temperature of 23 K, is shown in Fig. 3. In the figure, the region of the R values, observed for all the three garnets (Fig. 2), is indicated as the space between dashed vertical lines. This region is rather narrow: the corresponding values of Q vary by $\pm 12\%$ relative to the mean value of 0.6 eV.

3. Role of Anion and Cation Sublattices in the Formation of Relaxation Kinetics

Taking into account the different cation compositions of the examined garnets $\text{Ca}_3\text{Mn}_2\text{Ge}_3\text{O}_{12}$, $\text{Ca}_3\text{Ga}_{2-x}\text{Mn}_x\text{Ge}_3\text{O}_{12}$ and $\text{NaCa}_2\text{Mn}_2\text{V}_3\text{O}_{12}$, it is

natural to assume that a similar relaxation kinetics of photoinduced absorption, observed in these garnets, is conditioned by the motion of photoproduced holes in the oxygen sublattice that has a practically identical structure in all these garnets. The separation between adjacent O^{2-} ions in the oxygen sublattice, close to 2.8 Å, is sufficient for the hole motion over the sublattice.

This notion is convincingly corroborated by the following. Experiment shows [12, 13] that photoinduced absorption can be produced by red light with photon energy of about 2 eV. This energy is much less than the ionization potential of cations in garnets $NaCa_2Mn_2V_3O_{12}$, $Ca_3Mn_2Ge_3O_{12}$, and $Ca_3Ga_{2-x}Mn_xGe_3O_{12}$ (the ionization potential of a free Na^+ ion equals 47 eV and that of the rest of cations is even greater). Although the ionization potentials of cations can be lowered in the lattice, they indicate the scale of energy required for creating a hole in the cation sublattice. A photon energy of 2 eV is obviously insufficient for that. As regards the oxygen sublattice, the hole creation energy is equal to about 2 eV (such an estimate follows from the energy position of the lowest density peak of hole states in the oxygen hole band of $YBa_2Cu_3O_{6+x}$ [20]). Thus, energy considerations confirm that holes, responsible for photoinduced absorption, are created and move in the oxygen sublattice of garnets.

Based on this notion, let us consider the structure of a hole polaron in the sublattice of O^{2-} -ions with the closed-shell configuration of neon, $1s^22s^22p^6$. The structure of the stable state of a hole polaron is well known for the crystals of Ne, Ar, Kr and Xe, formed by atoms with a closed valence shell ns^2np^6 . The valence np -hole, distributed between two adjacent atoms separated by a distance r , realizes a strong exchange binding (of the scale of 1 eV), that sharply grows with decreasing r and makes two adjacent atoms strongly approach to each other [28]. Similar two-site holes (called V_k -centers), formed in the anion sublattice of alkali

Valence p -state radius, ρ , and the interatomic distance in the ideal lattice, b , whose ratio determines the interatomic exchange interaction, forming a two-site hole polaron state in some crystals

| Crystal | ρ , Å | b , Å | ρ/b |
|----------------------|------------|---------|----------|
| $Ca_3Ga_2Ge_3O_{12}$ | 1.54 | 5.25 | 0.293 |
| Ne | 0.99 | 5.97 | 0.166 |
| Ar | 1.74 | 7.10 | 0.245 |
| Kr | 2.00 | 7.54 | 0.265 |
| KCl | 1.62 | 8.40 | 0.193 |
| KI | 2.36 | 9.42 | 0.250 |
| NaI | 2.36 | 8.63 | 0.273 |

halides, are also well known as a stable polaronic state (see, e.g., [29, 30]). In all cases, the exchange binding mainly depends on the ratio ρ/b (ρ is the valence p -state radius and b is the corresponding interatomic distance in the ideal lattice). This ratio is given in Table 1 for the O^{2-} -sublattice of the $Ca_3Ga_2Ge_3O_{12}$ garnet and, for comparison, for some of the mentioned crystals. Since the ρ/b ratio for the garnet O^{2-} -sublattice is not less than that for other crystals, where the existence of two-atom hole polarons has been reliably established, they must exist also in the O^{2-} -sublattice.

Thus, a similar relaxation kinetics of photoinduced absorption, observed in the garnets $Ca_3Mn_2Ge_3O_{12}$, $Ca_3Ga_{2-x}Mn_xGe_3O_{12}$, and $NaCa_2Mn_2V_3O_{12}$, is formed by the motion of two-site hole polarons in the oxygen sublattice (built from the common anion group O_{12}). The role of cation composition consists in influencing the activation energy of hole hopping in rather narrow limits shown in Fig.3.

In addition, let us take a look at the effect of a non-stoichiometric cation composition. Incorporation of manganese into the Ga-sublattice of the garnet $Ca_3Ga_{2-x}Mn_xGe_3O_{12}$ causes lattice strains and distortions manifesting themselves in a low solubility of Mn in $Ca_3Ga_2Ge_3O_{12}$. These structure inhomogeneities obviously are responsible for a decreasing temperature dependence of the relaxation rate observed only for $Ca_3Ga_{2-x}Mn_xGe_3O_{12}$ (see Fig. 2). Indeed, structure inhomogeneities result in a straggling of potential wells, serving as traps for hole polarons, over depth. The existence of holes, populating shallow and deep traps, manifests itself in the following way. At low temperatures, all the hole traps participate in forming the relaxation kinetics; shallow traps are predominant in number and dictate a large relaxation rate. But, at high temperatures, only holes with a long lifetime, trapped in very deep wells, persist and entail a long relaxation time.

The straggling of traps over depth manifests itself also in a significant delay of the saturation of photoinduced absorption observed after switching on illumination for $Ca_3Ga_{2-x}Mn_xGe_3O_{12}$ only (see Fig. 1). Very deep traps, occupied by holes, cannot be depopulated by varying random electric fields (contrary to the rest of traps) and make an accumulated contribution to ΔK . However, this accumulation takes a long time because of a small number of very deep traps.

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ПРОЯВ СТРУКТУРНИХ ОСОБЛИВОСТЕЙ
ГРАНАТІВ В КІНЕТИЦІ РЕЛАКСАЦІЇ
ФОТОІНДУКОВАНИХ ЯВИЩ

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Резюме

Досліджено кінетику релаксації персистентного додаткового оптичного поглинання, спричиненого опроміненням гранатів $\text{NaCa}_2\text{Mn}_2\text{V}_3\text{O}_{12}$, $\text{Ca}_3\text{Mn}_2\text{Ge}_3\text{O}_{12}$ і $\text{Ca}_3\text{Ga}_{2-x}\text{Mn}_x\text{Ge}_3\text{O}_{12}$ видимим світлом. Показано, що кінетика релаксації пов'язана зі структурою гранатів. Нетривіальна особливість кінетики, спільна для всіх досліджуваних гранатів, зумовлена спільним елементом структури — аніонною групою O_{12} , що утворює підґратку кисневих іонів O^{2-} . Релаксаційна кінетика відбиває зародження і рух діркових поляронів у кисневій підґратці, а від катіонного складу залежить стрибковий бар'єр поляронів, що змінюється в не дуже широких межах. Роль кисневої підґратки переконливо підтверджено енергетичними міркуваннями.