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Absorption of Femtosecond Laser Pulses by Optical Centers in the Presence of Strong Electron-phonon Coupling

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ABSTRACT: In this paper, absorption of femtosecond laser pulses by optical centers in a solid in the presence of strong electron-phonon coupling is calculated. The 4f-5d electron transition in a Eu^{2+} europium ion put in the $SrGa_2S_4$+$MgGa_2O_4$ multiphase phosphor is considered. The probability of absorption of a laser pulse as a function of its duration for different system parameters is analyzed.

Key words: femtosecond laser pulse, optical centers in solids, strong electron-phonon coupling.

In the present work, absorption of ultrashort electromagnetic pulses (USP) by optical centers in solids in view of electron-phonon coupling is studied theoretically. By optical centers (OC) are meant impurity or defect centers put in a solid-state matrix and capable of absorbing radiation in the visible and near-IR regions. An example of OC can be F-centers in alkali halide crystals, nitrogen-vacancy centers (NV centers) in diamond, ions of rare-earth elements in phosphors, etc.

A distinguishing feature of absorption of electromagnetic radiation in the case under consideration is the presence of strong electron-phonon interaction that modifies significantly the OC absorption spectrum. The spectrum of absorption of radiation in a phonon wing without considering phonon dispersion is described by the following formula (in the system of units in which the Boltzmann constant and the Planck constant are assumed to be equal to one) [1]:

$$G(\omega) = \exp\left[-\frac{a}{2}\coth\left(\frac{\omega_1}{2T}\right)\right]\sum_{p=-\infty}^{\infty}\exp\left(\frac{p\,\omega_1}{2T}\right)I_p\left(\frac{a}{2\sinh\left(\omega_1/2T\right)}\right)\delta\left(\omega-\omega_0-p\,\omega_1\right), \quad (1)$$

here p is the number of phonons emitted/absorbed as a result of absorption of a light quantum, ω_1 is the phonon energy, a is the heat-release constant, T is the temperature, ω_0 is the frequency of a zero phonon line, I_p is the modified Bessel function.

Hereafter we assume that the electron-phonon transition is homogeneously broadened with the phonon relaxation constant g. Then the delta functions in the right-hand side of the formula (1) should be replaced by Lorentzians:

$$\delta(x) \to \frac{1}{\pi}\frac{\gamma}{x^2+\gamma^2}. \quad (2)$$

As an example, let us consider absorption of radiation at the 4f-5d transition in a Eu^{2+} ion put in the $SrGa_2S_4$+$MgGa_2O_4$ multiphase phosphor that is promising for development of high-efficiency LEDs. For this case we have the following numerical values of parameters: $\omega_0 = 2.536$ eV, a = 8, $\omega_1 = 34.5$ meV [2].

The absorption spectrum calculated by the formulas (1)-(2) for this phosphor is presented in Fig. 1 for room temperature and different values of the phonon relaxation constant γ.

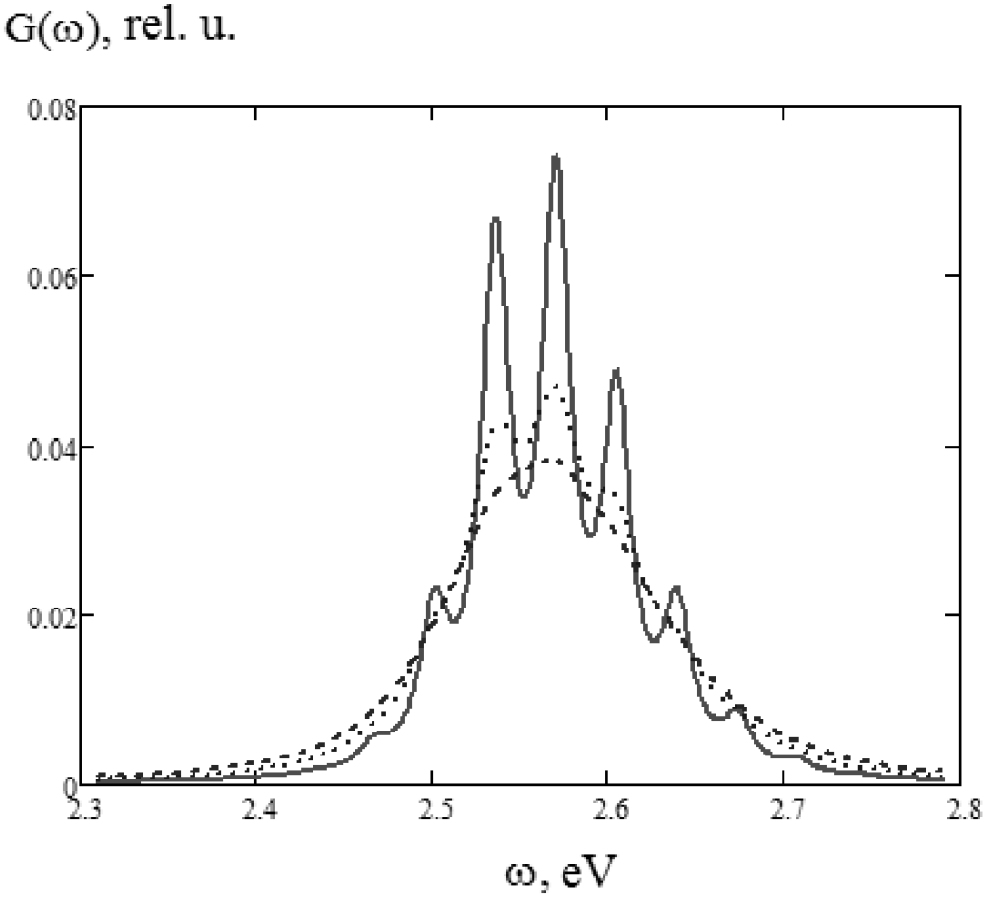

Figure 1: The absorption spectrum of the {$SrGa_2S_4$+$MgGa_2O_4$}: Eu^{2+} phosphor at room temperature T = 300 °K and different phonon relaxation constants: solid curve - γ = 10^{-2} eV, dotted curve - 2·10^{-2} eV, dashed curve - 3·10^{-2} eV.

From this figure it is seen that for small values of the parameter g maxima show themselves in the absorption spectrum that are caused by the contribution of absorbed/emitted phonons to the process. With increasing relaxation constant these maxima are broadened, so for large g the absorption spectrum represents a smooth bell-shaped curve with a maximum falling on a photon energy of 2.57 eV (λ = 482.5 nm).

The evolution of the absorption spectrum with decreasing temperature (for T = 60 °K) is shown in Fig. 2.

It is seen that in this case the absorption spectrum does not undergo significant changes (especially for large values of the parameter γ), increasing in amplitude approximately by an order of magnitude. For small phonon relaxation constants, phonon maxima in the low-frequency wing of the spectrum smooth out with decreasing temperature. The high-frequency wing in this case practically does not change.

Presented in Fig. 3 is the absorption spectrum in the limit of a small phonon relaxation constant γ = 10^{-3} eV for two values of temperature T = 300 °K and T = 60 °K. In this case, peaks corresponding to phonon absorption are clearly seen. The distance between two nearest peaks is 0.034 eV, which corresponds to the phonon energy. The solid curve in this figure is multiplied by 5. It is seen that with increasing temperature new phonon maxima appear in the low-frequency wing of the absorption spectrum.

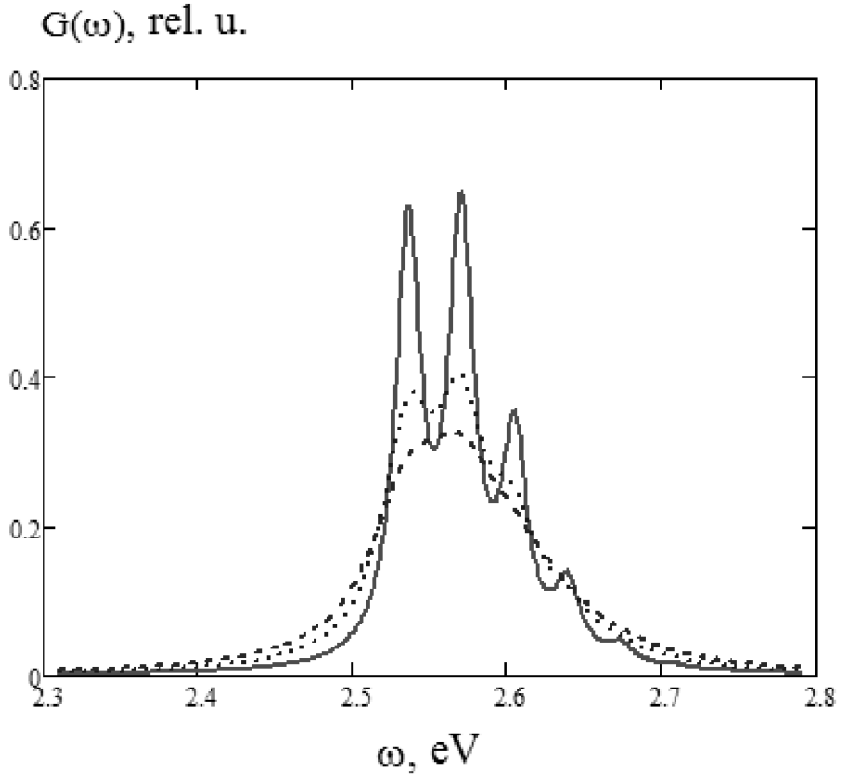

Figure 2: The same as in Fig. 1 for T = 60 °K.

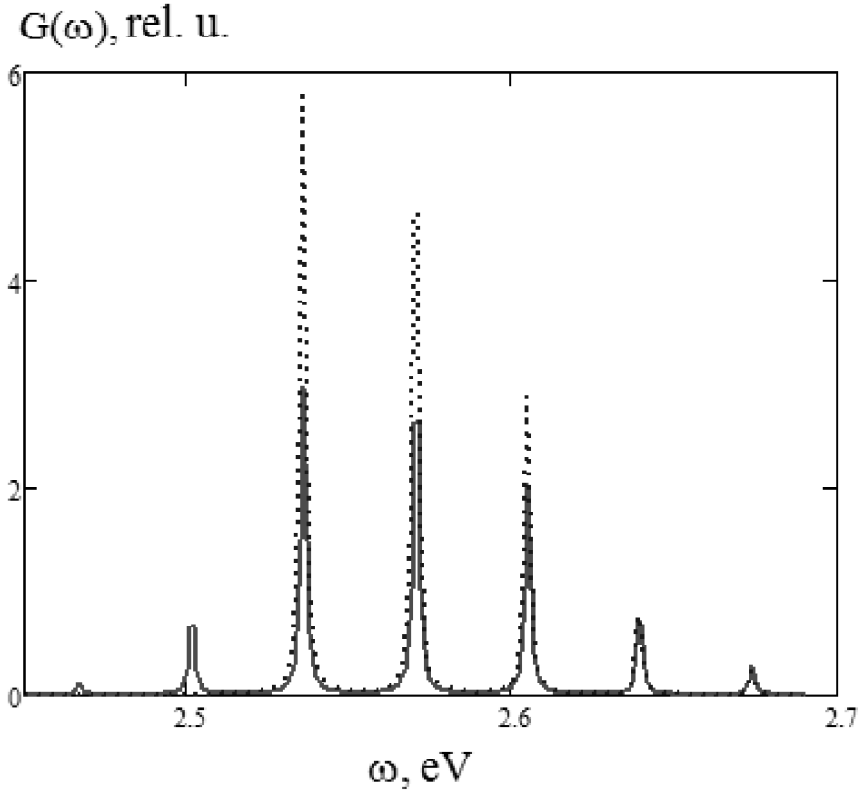

Figure 3: The absorption spectrum of the {$SrGa_2S_4$+$MgGa_2O_4$}:Eu^{2+} phosphor at $\gamma = 10^{-3}$ eV and different temperatures: solid curve - T = 300 °K, dotted curve - T = 60 °K.

Let us consider absorption by the $\{SrGa_2S_4+MgGa_2O_4\}:Eu^{2+}$ system of an USP of a corrected Gaussian shape, the Fourier transform of which looks like

$$E_{cor}\left(\omega',\omega,\tau,\varphi\right)=i\tau\sqrt{\frac{\pi}{2}}\frac{\omega'^2\tau^2}{1+\omega^2\tau^2}\left\{e^{-i\varphi-(\omega-\omega')^2\tau^2/2}-e^{i\varphi-(\omega+\omega')^2\tau^2/2}\right\}, \tag{3}$$

here ω is the carrier frequency, ω′ is the current frequency, τ is the pulse duration, φ is the initial phase. The pulse amplitude in the formula (3) is assumed to be equal to one.

Let us calculate the probability of absorption during the action of a pulse (3) as a function of its duration τ by the known formula [3]:

$$W(\tau,\omega)=\frac{c}{4\pi^2}\int_0^\infty\sigma(\omega')\frac{\left|E(\omega',\omega,\tau)\right|^2}{\hbar\omega'}d\omega', \tag{4}$$

where $\sigma(\omega')$ is the photoabsorption cross-section. Hereafter we will be interested in the relative value of the probability, so in the formula (4) it is possible to make the replacement $\sigma(\omega')\to G(\omega')$.

The results of calculations by the formulas (1)-(4) are presented in the figures given below for different problem parameters. The curves in Figs. 4-7 are normalized so that in case of long durations t the absorption probabilities are close to each other.

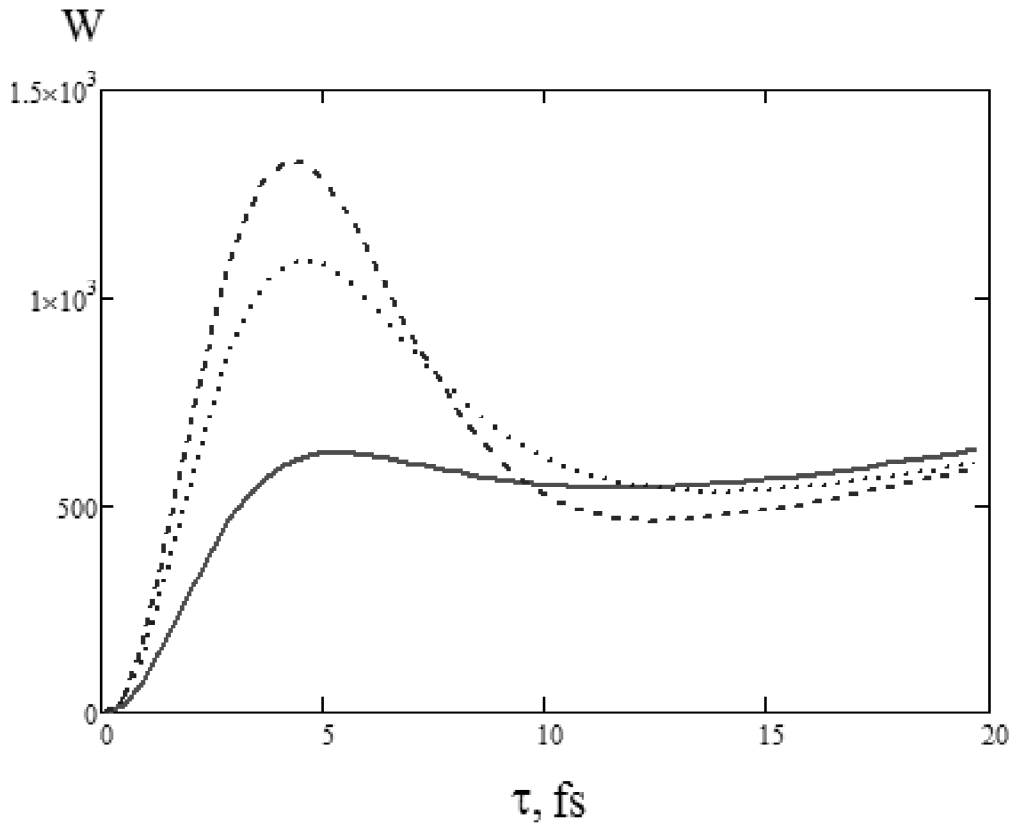

Figure 4: The relative probability of USP absorption in the phonon wing of the absorption line of the $\{SrGa_2S_4+MgGa_2O_4\}$: Eu^{2+} phosphor at the carrier frequency ω = 2.4 eV for γ = 0.01 eV and different temperatures: solid curve - T = 300 °K, dotted curve - T = 600 °K, dashed curve - T = 60 °K.

From Fig. 4 it follows in particular that the position of the maximum of the dependence $W(\tau)$ weakly depends on temperature. In this case, with decreasing temperature the maximum becomes more pronounced. With increasing pulse duration, the absorption probability becomes a linear function of τ as follows from traditional theoretical consideration.

In the case under consideration, the maximum falls on a pulse duration about 5 fs, which corresponds approximately to 3 cycles at the carrier frequency $\omega = 2.4$ eV.

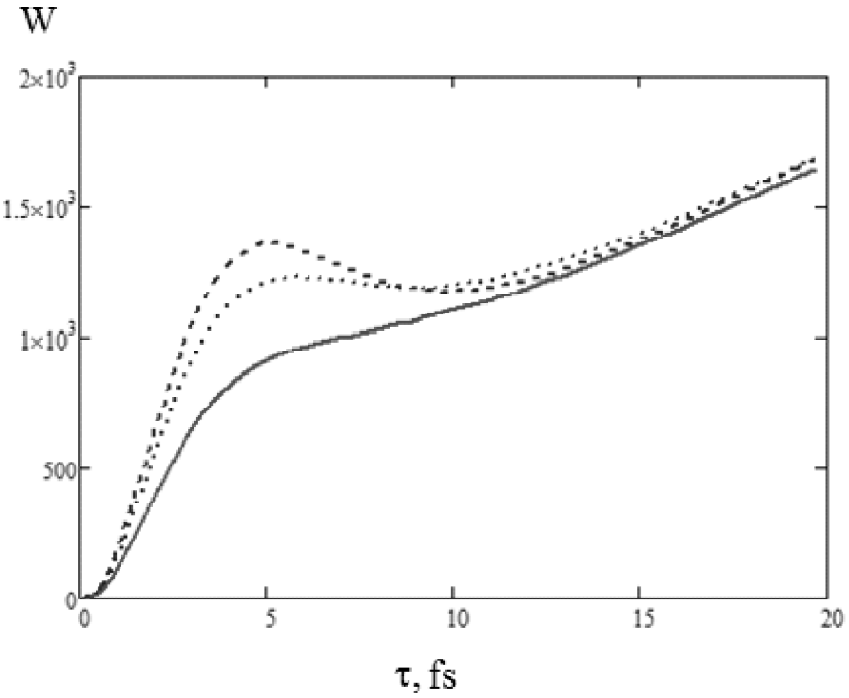

Figure 5: The same as in Fig. 4 for $\gamma = 0.03$.

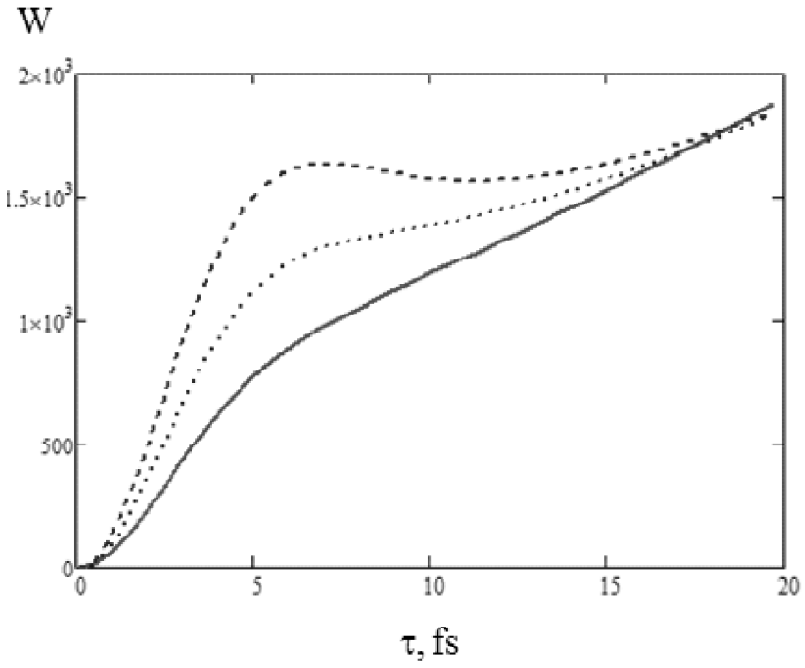

Figure 6: The same as in Fig. 4 for the carrier frequency $\omega = 2.7$ eV.

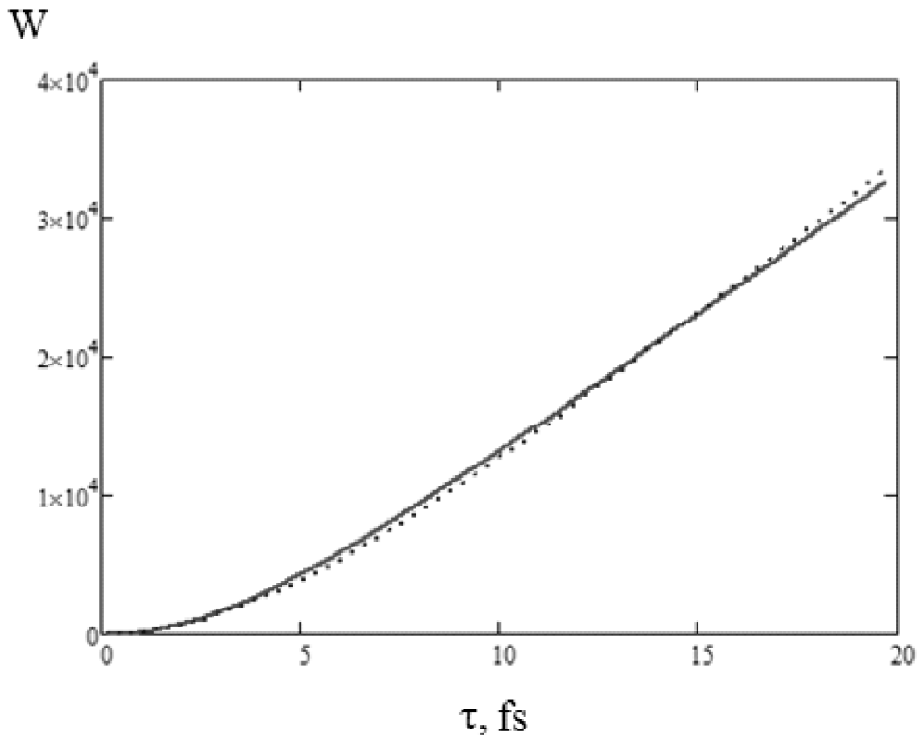

Figure 7: The same as in Fig. 4 for the carrier frequency ω = 2.55 eV.

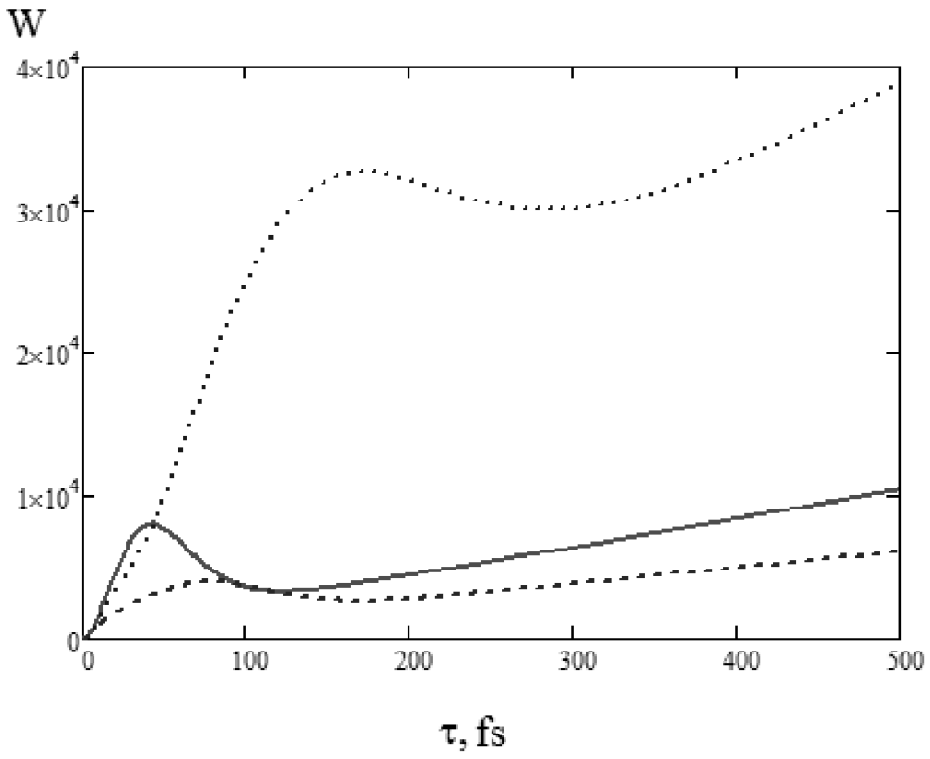

Figure 8: The relative probability of USP absorption in the phonon wing of the absorption line of the {$SrGa_2S_4$+$MgGa_2O_4$}: Eu^{2+} phosphor for γ = 10^{-3} eV, T = 300 °K at different carrier frequencies: solid curve - ω = 2.55 eV, dotted curve - ω = 2.6 eV, dashed curve - ω = 2.63 eV.

From Figs. 4-7 it follows that the nonlinear dependence of the function $W(\tau)$ for specified values of the parameter γ takes place at the edges of the phonon absorption band, that is, for ω = 2.4 eV and 2.7 eV. In this case, with decreasing temperature the nonlinearity increases, so a maximum and a minimum appear, and with increasing parameter γ the nonlinearity decreases. For the carrier frequency at the center of the absorption line ω = 2.55 eV the function $W(\tau)$ increases monotonically for all pulse durations, for short τ this increase being quadratic, and for long durations it is linear.

Presented in Fig. 8 is the dependence $W(\tau)$ for different USP carrier frequencies at T = 300 °K and for a small value of the parameter $\gamma = 10^{-3}$ eV. It is seen that in this case nonlinearity takes place also in the central region of frequencies of the absorption spectrum, which is caused by the quasi-linear behavior of the spectrum (Fig. 3).

References

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