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Absorption of ultrashort electromagnetic pulses on color centers in solids: dependence on current time

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ABSTRACT: In the framework of previously derived approach we investigate theoretically ultrashort electromagnetic pulse absorption on color centers in solids as a function of current time with account for strong electron-phonon coupling. Time dependence of absorption probability and temporal evolution of absorption spectra are calculated and analyzed. Particularly, it is shown that spectral maximum of the probability absorption shifts to lower frequencies with lowering of temperature.

Due to fast development of ultrashort electromagnetic pulses (USP) generation technologies [1] (Nobel prize 2023) it is of interest to consider USP-matter interaction in real time. This paper is devoted to the investigation of USP absorption by color centers in solids in the presence of strong electron-phonon coupling in picosecond time range.

We use here our approach for description of photoprocesses in terms of dimensionless probability at current time. This approach was suggested and validated in the paper [2].

The probability of excitation of an electronic-vibrational transition at a given time t by a pulse with duration τ is given by the formula [3]

$$W(t,\tau)=\omega_0\,\Omega_0^2\int_0^\infty G(\omega)\frac{\widetilde{D}(t,\tau,\omega)}{\omega}d\omega \tag{1}$$

here ω_0 is eigenfrequency of quantum transition,

$$\widetilde{D}(t,\tau,\omega)=E_0^{-2}\left|\int_{-\infty}^{t}dt'\exp(i\omega t')E(t',\tau)\right|^2 \tag{2}$$

$E(t',\tau)$ is electric field strength in the pulse, Ω_0 is Rabi frequency which is proportional to the electric field amplitude E_0, $G(\omega)$ is spectral profile of the absorption cross section which in the presence of a strong electron-phonon coupling (without taking into account phonon dispersion) is described by the following formula (in a system of units in which the Boltzmann constant and Planck constant are set equal to unity) [4]:

$$G(\omega)=\exp\left[-\frac{a}{2}\coth\left(\frac{\omega_1}{2T}\right)\right]\sum_{p=-\infty}^{\infty}\exp\left(\frac{p\omega_1}{2T}\right)I_p\left(\frac{a}{2\sinh(\omega_1/2T)}\right)L(\omega-\omega_0-p\omega_1). \tag{3}$$

here p is number of phonons emitted/absorbed during light absorption, ω_1 is phonon energy, a is heat release constant, T is temperature, ω_0 is zero phonon line frequency, I_p is modified Bessel function, $L(\omega)$ is Lorenzian:

$$L(\omega)=\frac{\gamma/\pi}{\omega^2+\gamma^2} \tag{4}$$

here γ is the spectral width of electron-phonon transition.

Let us consider, as an example, the absorption of radiation at the 4f-5d transition in the Eu^{2+} ion placed in multiphase phosphorus $SrGa_2S_4$+$MgGa_2O_4$, which is promising for the creation of highly efficient LEDs [5]. For this case we have the following numerical values of the parameters: $\omega_0 = 2.536$ eV (zero phonon line), a = 8, $\omega_1 = 34.5$ meV. We suppose in calculations that γ=0.272 meV.

Let us calculate absorption probability according to the formulas (1)-(4) for excitation of color center in solid by exponential pulse

$$E_{EP}(t,\tau)=\theta(t)E_0\exp(-t/\tau)\cos(\omega_c t) \tag{5}$$

here $\theta(t)$ is Heaviside step-function, τ, ω_c are pulse duration and carrier frequency. We suppose that exciting pulse is multi-cycle, i.e. $\omega_c\cdot\tau>>1$.

Absorption probability as a function of time is presented in Fig.1 for various numbers of absorbed phonons and in Fig.2 for various pulse durations.

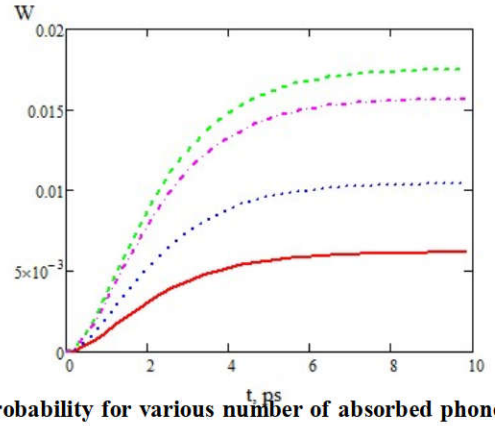

Fig.1. Time dependence of absorption probability for various number of absorbed phonons: solid curve – p=0, dotted line – p=1, dashed curve – p=3, dotted-dashed line – p=5; T=300 K, τ=2.4 ps, $\Omega_0=10^{-5}$ at. u.

As one can see from these figures absorption probability is monotonically increasing function of time for all considered values of problem parameters. Fig.1 demonstrates that absorption probability grows with increase of absorbed phonon numbers till p=3 after that probability decreases.

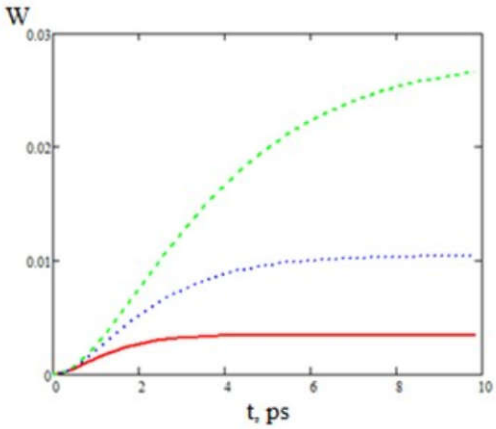

Fig.2. Time dependence of absorption probability for various pulse durations: solid curve – τ=1.2 ps, dotted line – τ=2.4 ps, dashed curve – τ=4.8 ps, one absorbed phonon p=1, T=300 K, $\Omega_0=10^{-5}$ at. u.

Fig. 2 shows that the probability of absorption increases with increasing pulse duration, which is intuitive.

Absorption spectra, i.e. dependence on carrier frequency of USP, are shown in Fig. 3, 4 for various times and temperatures.

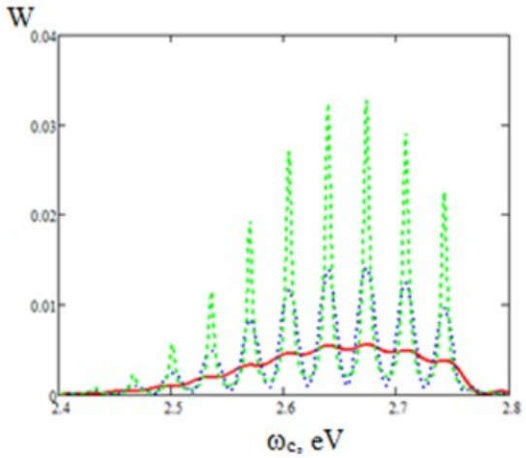

Fig.3. Temporal dynamics of photoabsorption probability: solid curve – t=0.12 ps, dotted line – t=0.24 ps, dashed curve – t=2.4 ps, T=300 K, τ=0.24 ps, $\Omega_0=10^{-4}$ at. u.

It can be seen that with increasing time, the spectral maxima become sharper, and with decreasing temperature, the spectral maximum of absorbed phonons.

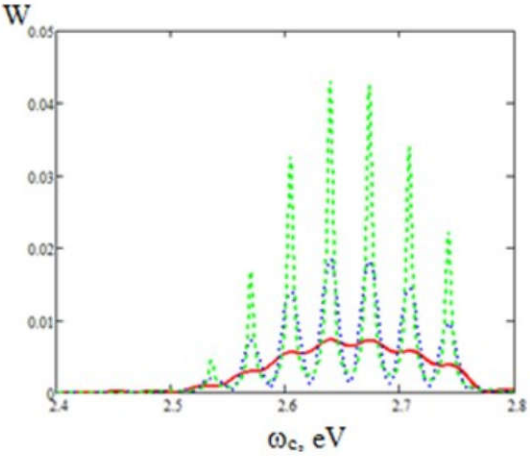

Fig.4. Temporal dynamics of photoabsorption probability: solid curve – t=0.12 ps, dotted line – t=0.24 ps, dashed curve – t=2.4 ps, T=77 K, τ=0.24 ps, $\Omega_0=10^{-4}$ at. u.

It also follows from the figures presented that at the temperatures considered, the contribution of the zero-phonon line is small compared to the phonon wing.

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