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Cavityless optical bistability in quasi-one-dimensional Peierls semiconductors

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***Abstract*. The theoretical possibility of obtaining cavityless optical bistability in quasi-one-dimensional Peierls semiconductors is demonstrated. The nonlinear interaction of the system with an electromagnetic field is caused by the red shift of the lower edge of an optical transition in the semiconductor with increasing concentration of nonequilibrium electron – hole pairs.**

Keywords: *optical bistability, quasi-one-dimensional Peierls semiconductor*.

1. Introduction

The prospects for using optical bistability in optical memory elements and other devices in quantum electronics attract considerable interest in its study and in the search for new materials possessing this nonlinear optical property [1 – 10]. One of the most well studied materials, whose parameters exhibit jump-like hysteresis behaviour upon a continuous variation of the parameters of the incident radiation, is cadmium sulfide [11 – 16]. An increase in the concentration of nonequilibrium photoexcited electron – hole pairs in this material leads to a substantial transformation of the electronic spectrum due to a strong electron – phonon interaction. As a result, the cavityless optical bistability appears with increasing absorption when the parameters of the incident radiation are adequately chosen [1, 11 – 16].

The red shift of the lower edge of the absorption band taking place in cadmium sulfide with increasing concentration of electron–hole pairs has been also observed in a number of quasi-one-dimensional Peierls semiconductors: mixed-valence platinum complexes [17], charge-transfer salts TTF – TCNQ, NMP – TSNQ, TTT(TCNQ)$_2$ [17], blue bronze $K_{0.3}MoO_3$ [18, 19], vanadium dioxide [20, 21], TaS_3 [19, 22, 23], $TaSe_3$, $(NbSe_4)_{10/3}I$, $(NbSe_4)_2I$, NbS_3, $NbSe_3$, etc. [19]. In particular, a short, high-power laser pulse can induce in vanadium dioxide the semiconductor – metal phase transition (when the band gap of the electronic spectrum of the system vanishes), which occurs via the electronic mechanism [21, 24, 25]. Note that in quasi-one-dimensional Peierls semiconductors, unlike cadmium sulfide, the transformation of the electronic spectrum upon variation of the concentration of electron – hole pairs is caused by a strong electron – phonon interaction [17, 19, 20, 26 – 32].

Thus, there are good premises for observing cavityless optical bistability with increasing absorption in quasi-one-dimensional Peierls semiconductors. The theory of this phenomenon was developed in a series of papers [33 – 35] where the mechanism of the appearance and conditions for the observation of the cavityless optical bistability and photo-induced phase transition in Peierls semiconductors were discussed.

In this paper, we study, based on the theory developed in papers [33 – 35], the dependences of the band gap of the electronic spectrum of a quasi-one-dimensional Peierls semiconductor and the concentration of nonequilibrium photo-excited electron–hole pairs on the intensity and frequency of the incident radiation.

2. Basic equations

The Peierls system considered in this paper represents a one-dimensional chain of atoms with one external electron in each atom. In the high-temperature metal phase, the atoms in the chain are arranged equidistantly and the electron conduction band is half-filled. At sufficiently low temperatures a one-dimensional metal becomes unstable, and a metal – semiconductor phase transition occurs at some critical temperature. In this case, the atoms in the chain come close together in pairs [17, 28].

The equations describing the behaviour of a spatially homogeneous quasi-one-dimensional Peierls semiconductor with one external electron per atom in a light field have, in the dipole approximation, the form [23, 33, 35]

$$\frac{\mathrm{d}n}{\mathrm{d}t} = \frac{8\pi I}{cn_1\hbar^2}\sum_{|k|\leqslant\pi/2} d_k^2 g\left(\frac{2\varepsilon_k}{\hbar}-\omega_0\right)\tanh\frac{\varepsilon_k-\mu}{2k_\mathrm{B}T} - \frac{n-n_0}{\tau}, \quad (1)$$

$$\frac{\mathrm{d}^2\xi}{\mathrm{d}t^2}+\frac{1}{\tau_\mathrm{p}}\frac{\mathrm{d}\xi}{\mathrm{d}t} = \frac{4}{NMR^2}\left(\sum_{|k|\leqslant\pi/2}\frac{\partial\varepsilon_k}{\partial\xi}\tanh\frac{\varepsilon_k-\mu}{2k_\mathrm{B}T} - A\xi\right), \quad (2)$$

where $k = 0, \pm 2\pi/N, ..., \pm\pi/2$ are dimensionless quasi-wave electron numbers in the one-dimensional electron band from the first Brillouin zone whose width equals 2π; n_1 is the refractive index of the medium; n and n_0 are the concentrations of electrons in the conduction band upon irradiation of the semiconductor and in the absence of

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irradiation, respectively; ξ is the order parameter of the metal–semiconductor phase transition (the reduced amplitude of the Frölich phonon mode); τ and τ_p are the interband electron and phonon relaxation times, respectively; I is the incident-radiation intensity; μ is the Fermi quasi-level of electrons in the conduction band; k_B is the Boltzmann constant; T is the absolute temperature; ω_0 is the central frequency of the incident-radiation spectrum; N and M are the concentration and mass of atoms involved in the formation of the one-dimensional electron band, respectively; R is the effective radius of the atomic wave function of an electron; $g(\omega)$ is the incident-radiation spectrum normalised to unity;

$$\varepsilon_k = 2b\,\mathrm{sign}(\cos k)(\cos^2 k + \sinh^2 \xi)^{1/2} \tag{3}$$

is the electronic spectrum of a Peierls semiconductor in the strong coupling approximation; $4b$ is the width of the electron conduction band in the metal phase (for $\xi = 0$);

$$d_k = \frac{2d}{1+\varphi_k^2}\left|(1-\varphi_k^2)\sinh\xi\cos k - 2\varphi_k\cosh\xi\sin k\right| \tag{4}$$

is the matrix element of the dipole moment operator for electronic transitions with the quasi-wave number k from the valence band to the conduction band;

$$\varphi_k = \frac{\cosh\xi\cos k - \mathrm{sign}(\cos k)(\cos^2 k + \sinh^2\xi)^{1/2}}{\sinh\xi\sin k}; \tag{5}$$

$2d\sinh\xi = d_0$ is the matrix element of the dipole moment operator at the centre of the Brillouin zone (for $k = 0$); and

$$A = 2\sum_{|k|\leqslant\pi/2}\left(\frac{\partial^2\varepsilon_k}{\partial\xi^2}\tanh\frac{\varepsilon_k}{2k_BT_0}\right)\Bigg|_{\xi=0} \tag{6}$$

is the generalised coefficient of rigidity of a crystal lattice, which can be expressed in terms of the critical temperature T_0 of the metal–semiconductor phase transition [25].

The dynamic equations (1) and (2) for a quasi-one-dimensional Peierls semiconductor should be supplemented with the electric neutrality equation relating the Fermi quasi-level μ and the electron concentration n in the conduction band:

$$n = \frac{N}{2} - \sum_{|k|\leqslant\pi/2}\tanh\frac{\varepsilon_k - \mu}{2k_BT}. \tag{7}$$

Equations (1) and (2), along with equation (7) and auxiliary relations (3)–(6), completely describe the dynamics of parameters $n(t)$ and $\xi(t)$ of a Peierls semiconductor at the given temperature T and parameters of incident radiation such as the intensity I, the central frequency ω_0 of the emission spectrum and its shape $g(\omega)$.

3. Stationary solution for a nondegenerate semiconductor

We will analyse equations (1)–(7) for a nondegenerate semiconductor, when Fermi quasi-levels of electrons, $\mu_e = \mu$, and holes, $\mu_p = -\mu$, lie within the band gap of the electronic spectrum ε_k (3):

$$0 \leqslant \mu < \frac{\varepsilon}{2} - 2k_BT, \tag{8}$$

where

$$\varepsilon = 4b\sinh\xi \tag{9}$$

is the band gap of the electronic spectrum ε_k (3) of a quasi-one-dimensional Peierls semiconductor.

By excluding the Fermi quasi-level μ from equations (2) and (7), we find in the stationary case ($d\xi/dt \equiv 0$) the approximate relation between the band gap ε (9) and the electron concentration n in the conduction band for $T < T_0/2$:

$$n = \frac{N\varepsilon}{4\pi b}\ln\frac{\varepsilon_0}{\varepsilon}, \tag{10}$$

where ε_0 is the band gap of the electronic spectrum (3) in the absence of irradiation.

We will assume below that the envelope of the optical spectrum of radiation incident on the semiconductor has the Lorentzian shape

$$g(\omega) = \frac{\tau_0}{\pi[1+(\omega\tau_0)^2]}, \tag{11}$$

where τ_0 is the inverse half-width of the spectrum. In this case, we obtain in the stationary regime ($I = \mathrm{const}$, $\xi = \mathrm{const}$, $n = \mathrm{const}$) from equations (1), (2), and (7), taking into account (9), the approximate dependence $I(\varepsilon, \omega_0)$:

$$I = \frac{cn_1\hbar^2}{32\pi^2d^2\tau(\tau_0^3\omega_0)^{1/2}} \times(x-x_0)\left\{(1+x^2)\left[x+(1+x^2)^{1/2}\right]\right\}^{1/2}, \tag{12}$$

where

$$x = \tau_0\left(\frac{\varepsilon}{\hbar} - \omega_0\right) \tag{13}$$

is a dimensionless parameter that characterises the detuning of the lower edge of the optical transition $\varepsilon/\hbar$ from the carrier radiation frequency ω_0; and $x_0 = \tau_0(\varepsilon_0/\hbar - \omega_0)$ is the detuning in the absence of irradiation when the band gap is ε_0. Equation (12) was derived assuming that the central frequency ω_0 of the emission spectrum is close to the frequency of the lower edge of the optical transition:

$$|\varepsilon - \hbar\omega_0| \ll \hbar\omega_0. \tag{14}$$

Taking into account equations (7) and (10), the nondegeneracy condition (8) can be written in a more convenient form

$$\varepsilon_0 - \varepsilon < \frac{(\varepsilon_0 k_BT)^{1/2}}{2}; \tag{15}$$

where it is assumed, as in (10), that $T < T_0/2$.

4. Results of numerical calculations and discussion

The numerical calculations according to equations (10) and (12) were performed for the following values of parameters typical for vanadium dioxide [20]: the concentration of vanadium atoms was $N \simeq 7\times10^{22}$ cm^{-3}, the width of the conduction band of the electronic spectrum in the metal

phase was $4b \simeq 1.1$ eV, the band gap of the electronic spectrum in the low-temperature semiconductor phase was $\varepsilon_0 = 4b \sinh \xi_0 \simeq 0.6$ eV, the characteristic time of the interband electronic relaxation was $\tau \approx 3 \times 10^{-11}$ s [36], the matrix element of the dipole moment operator of the system for electronic transitions from the valence band to the conduction band at the centre of the Brillouin zone was $d_0 \approx 10^{-18}$ g cm s^{-1} [37, 38], the inverse half-width of the incident radiation spectrum was $\tau_0 \approx 10^{-13}$ s, and the refractive index of the medium at the incident-radiation frequency was $n_1 \approx \sqrt{10} \approx 3.16$.

The results of numerical calculations are presented in Figs 1–3. Figs 1 and 2 show the dependences of the band gap ε of the electronic spectrum of a Peierls semiconductor and the electron concentration n in the conduction band on the light-field intensity I for different central frequencies ω_0 of the light-field spectrum. Our analysis showed that, when the condition

$$\varepsilon_0 - \hbar\omega_0 > 1.62\hbar/\tau_0 = 0.011 \text{ eV}, \tag{16}$$

which bounds below the light-field carrier frequency ω_0, is fulfilled, the dependences $\varepsilon(I)$ and $n(I)$ are ambiguous within some region I ($I_2 < I < I_1$). At points I_1 and I_2, where $\partial\varepsilon/\partial I = 0$ and $\partial n/\partial I = 0$, ε and n change jump-wise (as shown by the arrows in Figs 1 and 2). The difference $\Delta I = I_1 - I_2$ determines the width of the region of optical bistability upon increasing absorption.

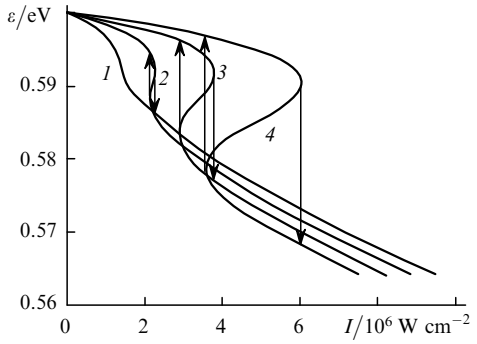

Figure 1. Dependences of the band gap ε of the electronic spectrum of a Peierls semiconductor on the light-field intensity I for the central frequency of the spectrum $\hbar\omega_0 = 0.592$ (*1*), 0.588 (*2*), 0.584 (*3*), and 0.580 eV (*4*).

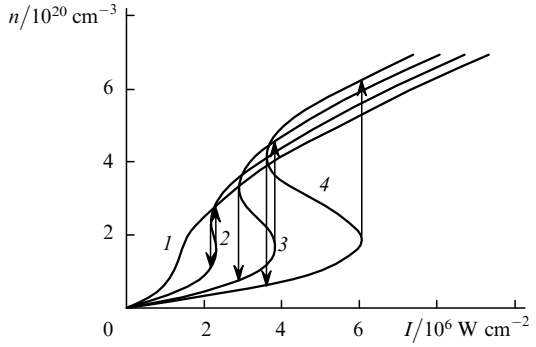

Figure 2. Dependences of the electron concentration n in the conduction band of a semiconductor on the light-field intensity I for the central frequency of the spectrum $\hbar\omega_0 = 0.592$ (*1*), 0.588 (*2*), 0.584 (*3*), and 0.580 eV (*4*).

Note that the condition (15) at the temperature 100 K and the band gap $\varepsilon_0 = 0.6$ eV in the absence of irradiation imposes the following restriction on band gap ε in the presence of irradiation:

$$0 < \varepsilon_0 - \varepsilon < 0.036 \text{ eV}. \tag{17}$$

One can see from Figs 1 and 3 that the inequality (17) is valid for the solutions obtained by us, including the bistability regions of the system.

Fig. 3 shows the tree-dimensional surface $I(\hbar\omega_0, \varepsilon)$, which illustrates a complicated nonmonotonic and ambiguous dependence of the inner parameter ε of the system on the external parameters I and ω_0.

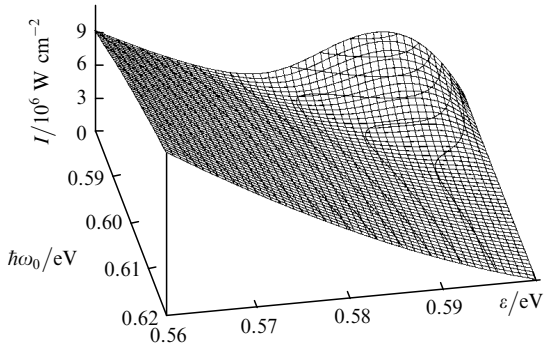

Figure 3. Dependence of the band gap ε of the electronic spectrum of a quasi-one-dimensional Peierls semiconductor on the intensity I and the central frequency ω_0 of the light-field spectrum.

The bistability of absorption of electromagnetic waves in VO_2 films near the critical temperature $T_0 = 340$ K of the semiconductor–metal phase transition was observed experimentally [39–41]. The values of the radiation wavelength ($\lambda = 2.2$ mm) and intensity ($I_{1,2} \sim 1$ W cm^{-2}), the time of switching between the semiconductor and metal states ($\tau_s \sim 0.1$ s), as well as the temperature dependences of the intensities $I_{1,2}$ indicate to the thermal mechanism [40] of optical bistability observed in papers [39–41].

By using the expression

$$\gamma = \frac{16\pi^2}{cn_1\hbar^2} \sum_{|k| \leqslant \pi/2} \varepsilon_k d_k^2 g\left(\frac{2\varepsilon_k}{\hbar} - \omega_0\right) \tanh\frac{\varepsilon_k - \mu}{2k_B T}, \tag{18}$$

for the electronic mechanism of switching between two semiconductor states with a small absorption coefficient γ_1 for $\hbar\omega_0 < \varepsilon$ and a large absorption coefficient γ_2 for $\hbar\omega_0 > \varepsilon$, we obtain the numerical estimates

$$\gamma_1 = \gamma(\hbar\omega_0 < \varepsilon) \approx 3 \times 10^4 \text{ cm}^{-1}, \tag{19}$$

$$\gamma_2 = \gamma(\hbar\omega_0 > \varepsilon) \approx 1.1 \times 10^5 \text{ cm}^{-1}. \tag{20}$$

The experimental absorption coefficient of the semiconductor phase of VO_2 measured at frequencies $\hbar\omega_0 > \varepsilon$ lies within $(0.15 - 1.8) \times 10^5$ cm^{-1} [20], in accordance with the numerical estimate (20).

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