arXiv:cond-mat/0611709v1 [cond-mat.str-el] 28 Nov 2006

The model of the optical switching center and the polarization anomalies in absorption spectra of cupric oxide after fast particles irradiation

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We consider the model of the optical switching center – a system with the following properties: It has two (or more) metastable states $|1\rangle$, $|2\rangle$, separated by a potential barrier U; it can switch from one state to another by absorbing the photons with energy $\hbar\omega \sim U$; the transition $|1\rangle \to |2\rangle$ is allowed only for a certain light wave polarization p_1 and the transition $|2\rangle \to |1\rangle$ – for other polarization p_2; these polarizations p_1, p_2 are orthogonal. The optical properties of this system are studied and are found to exhibit unconventional polarization dependence. In particular, the absorption spectrum observed in natural (unpolarized) light can display new features, that are absent in the spectra, obtained in two independent polarizations. We discuss these results in connexion with the (yet unexplained) experimental findings[1], where the similar anomalous polarization dependence of the absorption spectra of cupric monoxide CuO after the fast particle bombardment is reported.

Keywords: Optical nonlinearity, switching, polarization, cuprates

I. INTRODUCTION

The copper oxides and related materials show a number of unconventional physical properties, that have placed them among the most studied systems in the past decade. Of especial interest is their liability to the formation of various metastable and nonuniform states. Examples include the phase separation phenomena in doped HTSC cuprates[2], the stripe structures in HTSC's and non-superconducting oxides, such as the cupric monoxide CuO[3], the catalytic properties of CuO in certain chemical processes[4]. A general conclusion to be drawn from these facts is that the equilibrium state of the systems may be readily disturbed by means of various perturbations, such as chemical doping.

An important information about the concomitant changes of the electronic structure of these systems can be obtained in optical studies. In a series of papers[1,5,6] the evolution of the optical absorption spectra of CuO single crystals under the bombardment with the fast particles have been studied in a systematic way. It was found, that the bombardment leads to the progressive growth of the spectral weight within the dielectric gap and the appearance of new peaks in absorption spectra, that have no counterparts in the spectra of the non-irradiated good single crystalline samples. Similar results have been reported recently for the absorption spectra of CuO powder with nanosize grains after the influence of the converging spherical shock waves[7].

Overall, these findings can be interpreted within the picture of the phase separation, stimulated with the fast particles bombardment or the shock waves. The new spectral features, emerging in these conditions can possibly be attributed to the surface plasmon (Mie) resonances, as it was shown in[7]. Although the effective medium model of[7] argue the novel phase to be presumably "metallic", the microscopic physics of cuprates in the phase separation regime remains an issue and any additional detailed experimental data concerning the optical properties of these systems is of especial interest.

The present work is motivated with the investigations of the absorption spectra of CuO single crystal, irradiated with the fast He^+ nuclei, in different polarizations[1]. Surprisingly, it was found, that the spectrum in unpolarized light looks quite differently from the spectra in two plane polarizations and shows a new feature, not observed in any of these polarizations. To the best of our knowledge, no similar results have been ever reported for other systems. We interpret these findings as a manifestation of a subtle kind of optical nonlinearity, which origin is related to the effect of the bobmardment and the concomitant nucleation of some novel phase. In general, the nonlinearity take place each time as the system modifies its properties (susceptibilities, etc.) under the influence of an external perturbation, so that the response is no longer proportional to the magnitude of the perturbation. Thus, it is plausible, that the metastability and phase separation may favour the enhancement nonlinear phenomena.

The article is organized as follows. The next section contains a short overview of the experiment and its discussion. In sections 3, 4 we formulate our model and examine its optical properties in different polarizations. In section 5 the microscopic grounds of the model and its application to cuprates are considered. In section 6 the summary of the work is given.

II. EXPERIMENT

As the experimental findings under consideration have been published elsewhere[1], we restrict ourself here to the most essential facts. The CuO single crystalline platelet, cut out in crystallographic *ac* plane, have been exposed to the bombardment with the He^+ nuclei and the absorption coefficient of the irradiated sample have been measured within the range of 0.2 – 1.4 eV, the light propagating

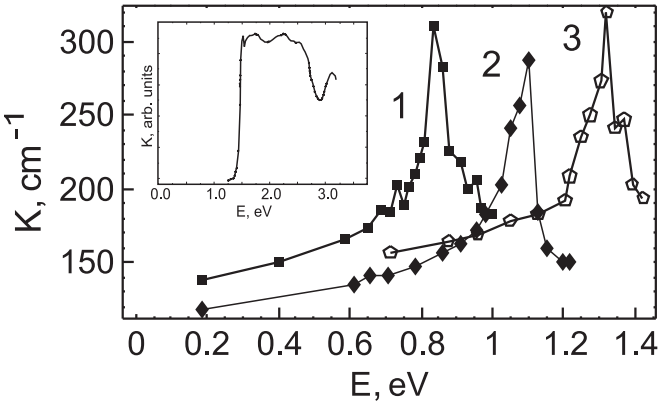

FIG. 1: The absorption spectra of CuO sample after the bombardment with He^+ nuclei[1]. Spectra 1 and 2 are obtained in plane polarizations $\mathbf{E} \parallel [\bar{1}01]$ and $\mathbf{E} \perp [\bar{1}01]$, respectively. Spectrum 3 is obtained in unpolarized light. Solid lines are guides to the eye. Inset: the absorption spectra of CuO high quality single crystal.

normally to the platelet. The bombardment was found to result in a significant destruction of the fundamental band edge and the appearance of some novel sharp peaks within the range 0.9 – 1.1 eV, that have no counterpart in the spectra of good CuO single crystals, shown at the inset of figure 1.

Main results are summarized at the figure 1. It is seen, that the change of the polarization of incident light from $\mathbf{E} \parallel [\bar{1}01]$ to $\mathbf{E} \perp [\bar{1}01]$ leads to the shift of the peak from ~ 0.9 eV to ~ 1.1 eV. Taking into account the anisotropy of the cupric monoxide, this rather strong dependence of the spectra on the polarization is not surprising. However, really striking is the spectrum, obtained in the unpolarized light, that shows again the similar peak shifted to still higher frequency (~ 1.3 eV).

The origin of these new spectral features and especially their relative positions in polarized and natural light pose a challenging problem. It is clear, that as far as the medium is linear, the spectrum, obtained in natural light, should always reduce to the superposition of the spectra, obtained in any two different polarizations, whatsoever are the mechanisms of the light extinction within the medium. Indeed, the unpolarized light may be regarded as a random mixture of two light waves with different polarizations. If the waves travel through the sample independently, the resulting optical spectrum will display the features, arising from both waves.

In contrast to this natural picture, the examination of the spectra at the figure 1 leads us to the conclusion, that such a superposition principle for the light waves in the system under consideration is violated. If we choose the polarizations $\mathbf{E} \parallel [\bar{1}01]$ and $\mathbf{E} \perp [\bar{1}01]$ as the basic ones, then the shape of the unpolarized light spectrum (curve 3 at figure 1) may only be understood, if we assume, that the propagation properties of one of these basic waves (or rather the corresponding system of two normal modes with elliptical polarization, see Section 5) is modified in presence of the second wave, so as to give rise to the peak at 1.3 eV and to hide the peaks at 0.9 eV and 1.1 eV.

In other words, the optical response of the system at a given frequency should depend on the parameters of the incident light wave. This implies the optical nonlinearity of the system. As a rule, the optical nonlinearity in non-magnetic media is described is terms of the dielectric permittivity ε, that depends on the intensity of the incident light wave, $\varepsilon = \varepsilon(\omega, |E|^2)$, where ω is its frequency and E – the electric vector. However, in the present case we have to deal with a somewhat subtler and less conventional type of nonlinearity, when the propagation of wave with a given polarization depends on the intensity of *other* wave, propagating simultaneously with this one.

We believe, that this unusual behaviour can be understood, assuming that the bombardment of CuO sample with the fast He^+ nuclei leads to the formation of the optical switching centers (OSC) – the specific structures, that provide the optical nonlinearity as discussed above. The basic ideas of the OSC model are discussed in the two next sections.

III. THE MODEL

Consider a system, that contains the structures (hereafter called the centers) with two equivalent metastable states $|1\rangle$, $|2\rangle$, separated by a potential barrier, high enough to suppress the spontaneous transitions between them via tunnelling and thermal activation. Thus, at low temperatures, the center is always frozen in one of the two states. However, it may surmount the barrier and hop to the other state when absorbing a portion of energy from an external energy reservoir.

The key points of the model are as follow: i) the center undergoes the transitions $|1\rangle \leftrightarrows |2\rangle$ when absorbing a photon of an appropriate frequency and ii) the transition $|1\rangle \to |2\rangle$ is allowed in a certain photon polarization p_1 while the reverse transition $|2\rangle \to |1\rangle$ is allowed in the polarization p_2, *orthogonal* to p_1. This is possible, if the transitions involve an intermediate excited state $|\psi'\rangle$ and the transitions $|i\rangle \to |\psi'\rangle$ are allowed in different polarizations for $i = 1, 2$. Then the height of the barrier is $U = E_{\psi'} - E_{1,2}$, E being the corresponding energy level (figure 2). Thus, the center acts as an optical switch, which is triggered when irradiated by a light wave of an appropriate polarization but does not feel the light of the complementary (orthogonal) polarization.

Now, we examine the effects of interaction of the centers with light. Let us first consider the case of the incident light wave with one of the special (p_1 or p_2) polarizations, say p_1. Those centers, that are in the state $|2\rangle$ do not feel this wave, while the centers in the state $|1\rangle$ absorb it and are excited to the state $|\psi'\rangle$. Then, these may either return to the state $|1\rangle$ or switch to the state $|2\rangle$. The similar considerations hold for the light with the polarization p_2. Given an arbitrary polarization, the

wave function of the photons may be written as

$$|p\rangle = \cos\varphi\,|p_1\rangle + \sin\varphi\,|p_2\rangle, \tag{1}$$

where by assumption, two basic polarizations are orthogonal, $\langle p_1|p_2\rangle = 0$. If the light is unpolarized, φ is a random variable. It follows from eq.(1), that

$$I_1/I_0 = \cos^2\varphi, \quad I_2/I_0 = \sin^2\varphi, \tag{2}$$

where I_0, $I_{1,2}$ stand for the intensities of the incident wave and those of its components with polarizations $p_{1,2}$, respectively. Since the photons of both p_1- and p_2- polarization are present in this mixed state in ratio $\cos^2\varphi : \sin^2\varphi$, the center will switch continuously between the metastable states $|1\rangle$, $|2\rangle$: the photons with different polarizations throw the center from one potential well into the other.

Restricting ourself to the three-level model as discussed above, we may write down the kinetic equations for the occupancy number of $|1\rangle$, $|2\rangle$, $|\psi'\rangle$ states, denoted n_1, n_2 and n_3, respectively:

$$\dot{n}_1 = -W_{31}n_1 + W_{13}n_3, \tag{3}$$
$$\dot{n}_2 = -W_{32}n_2 + W_{23}n_3, \tag{4}$$
$$\dot{n}_3 = W_{31}n_1 + W_{32}n_2 - (W_{13} + W_{23})n_3, \tag{5}$$
$$n_1 + n_2 + n_3 = 1, \tag{6}$$

where W_{ij} is the probability for a center to undergo the transition $|j\rangle \to |i\rangle$. The equilibrium ($\dot{n}_{1,2} = 0$) relative numbers of the centers in $|1\rangle$, $|2\rangle$ states are found to be:

$$n_1 = \frac{W_{13}W_{32}}{W_{23}W_{31} + (W_{13} + W_{31})W_{32}}, \tag{7}$$
$$n_2 = \frac{W_{23}W_{31}}{W_{23}W_{31} + (W_{13} + W_{31})W_{32}}. \tag{8}$$

The excitation of the center from i-th potential well is stimulated with light of an appropriate polarization. It follows from the symmetry arguments, that the transitions $|1,2\rangle \to |\psi'\rangle$ are governed by the same matrix element, multiplied by the relative number of photons of that polarization (eq.2), whence:

$$W_{31} = W\cos^2\varphi, \tag{9}$$
$$W_{32} = W\sin^2\varphi. \tag{10}$$

The transition amplitude W is found from the Fermi's golden rule:

$$W = \frac{4\pi^2}{\hbar^2}\rho(\omega_{12})\,|\langle\psi'|\mathbf{d}|1,2\rangle|^2, \tag{11}$$

where $\mathbf{d}$ is the dipole moment operator and $\rho(\omega_{12})$ is the spectral density of radiation at the frequency of the transition, the latter being proportional to the height of the potential barrier, separating the metastable states of the center (figure 2):

$$\hbar\omega_{12} \sim U. \tag{12}$$

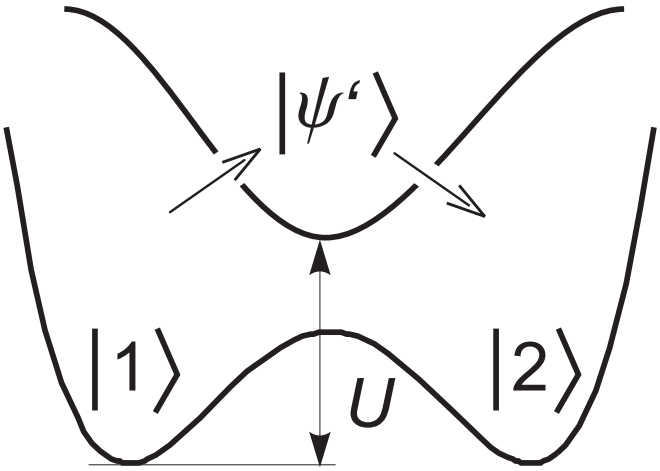

FIG. 2: Schematic picture of the optical switching transition. The center is excited from its ground state $|1\rangle$ to the intermediate state $|\psi'\rangle$, absorbing the photon with polarization p_1, and then may either relax back to its initial state or switch to another ground state $|2\rangle$.

As for the transitions $|\psi'\rangle \to |1,2\rangle$, these may be both spontaneous and stimulated, and their total probabilities W_{13}, W_{23} include the contributions of both channels. Thus, we write:

$$W_{13} = W'_{13} + W''_{13}\cos^2\varphi, \tag{13}$$

and similarly for W_{23}, where the probability W'_{13} of the spontaneous transition obviously does not depend on the polarization of incident photons and W''_{13} corresponds to the transition, induced with photons of p_1 polarization, their relative number being $\cos^2\varphi$.

Having in mind to account for the polarization anomalies in experimental spectra (figure 1), as discussed in previous section, we are mainly interested in the general properties of the polarization dependence of the stationary occupancy numbers $n_{1,2}(\varphi)$. Taking into account eqs.(9-13) and assuming $W'_{i3} = W'$, $W''_{i3} = W''$ for $i = 1, 2$ to make the general formulae (7,8) more tractable, one gets:

$$n_1 = \widetilde{n}(\varphi) + \frac{\sin^2\varphi}{1 + (\widetilde{W}/W')\cos^2\varphi\sin^2\varphi}, \tag{14a}$$
$$n_2 = \widetilde{n}(\varphi) + \frac{\cos^2\varphi}{1 + (\widetilde{W}/W')\cos^2\varphi\sin^2\varphi}, \tag{14b}$$

where $\widetilde{W} = W + 2W''$ and

$$\widetilde{n}(\varphi) = \frac{(W''/W')\,\cos^2\varphi\sin^2\varphi}{1 + (1 + 2W''/W')\cos^2\varphi\sin^2\varphi}. \tag{15}$$

The function $\widetilde{n}(\varphi)$ is symmetric in the number of the photons with p_1, p_2 polarization, while the second terms in rhs. of eqs.(14) show, that the equilibrium number n_1 of the centers, sensitive to the photons with p_1 polarization depends on the fraction of the photons with p_2 polarization in the beam (2), and vice versa.

Without the latter assumption of equal probabilities W'_{i3}'s, W''_{i3}'s, more complicated expressions are obtained instead of (14), but the qualitative results still remain the same: $n_1 = 0$, $n_2 = 1$ under the irradiation by light with p_1 polarization ($\varphi = 0$) and $n_1 = 1$, $n_2 = 0$ for the p_2 polarization ($\varphi = \pi/2$). The reason is that due to the switching described above, the center acquires a finite (albeit low) probability of *irreversible* escape from one state to the other under the irradiation with p_1 or p_2 polarization and thus all centers are switched for a long enough time. For other polarizations, both n_1 and n_2 are non-zero, because once switched, the center has a finite probability to switch back due to the presence of the photons with an appropriate polarization in the incident wave.

IV. THE OPTICAL RESPONSE

The switching transitions give rise to the resonant optical absorption at the frequency ω_{12}, eq.(12). We next put forward the general arguments to propose a plausible form of the dielectric tensor with regard to the switching transitions. To be specific, we consider the transmission of light through the film, containing the switching centers. In the case of normal incidence, the electric vector of the light wave lies in plane of the film. For the sake of simplicity, it will be assumed, that the in-plane part of the dielectric permittivity tensor $\hat{\varepsilon}$ is diagonal within the basis of the polarization states $|p_{1,2}\rangle$:

$$\hat{\varepsilon} = \begin{pmatrix} \varepsilon_1 & 0 \\ 0 & \varepsilon_2 \end{pmatrix}, \tag{16}$$

i.e. the corresponding plane waves are the normal modes for the system. In general, $\varepsilon_{1,2}$ contain the contributions from different optical transitions. E.g., as far as the optical transitions between the discrete levels are considered, it is common to express the dielectric permittivity matrix elements as a sum of lorentzian functions:

$$\mathcal{L}(\omega; \{\omega_i, I_i, \gamma_i\}) = 1 + \frac{1}{4\pi} \sum_i \frac{I_i}{\omega^2 - \omega_i^2 + i\gamma\,\omega_i}, \tag{17}$$

where ω_i, I_i and γ_i are the resonance frequency, intensity and damping factor of $i-$th optical transition, respectively. However, we emphasize, that the specific form of the spectral line is not important in what follows and the Lorentzian function is chosen rather arbitrarily.

Now, we focus on those contributions to the $\hat{\varepsilon}$ tensor, that are related to the switching transitions $|1\rangle \leftrightarrows |2\rangle$. By definition, their intensities are:

$$I_{1\to 2} = \hbar\omega_{12} P_1, \tag{18a}$$
$$I_{2\to 1} = \hbar\omega_{12} P_2, \tag{18b}$$

where P_i is the probability for the photon with polarization p_i to be absorbed by a center:

$$P_1 = W_{21} n_1, \tag{19a}$$
$$P_2 = W_{12} n_2. \tag{19b}$$

Here W_{ij} is the amplitude of the transition $|j\rangle \to |i\rangle$, $W_{ij} \propto W_{i\,3} W_{3\,j}$ (eqs.3-6) and $n_{1,2}$ are given by eqs.(14). Taking into account (19), the substitution of (18) into (17) makes it possible to write down the dielectric permittivity tensor (16) in the form:

$$\hat{\varepsilon} = \begin{pmatrix} \varepsilon_1 + \chi \cdot n_1(\varphi) & 0 \\ 0 & \varepsilon_2 + \chi \cdot n_2(\varphi) \end{pmatrix}, \tag{20}$$

where φ is the polarization angle of incident light wave relative to the basis $|p_{1,2}\rangle$ (1). All contributions to the conventional linear optical response of the system are absorbed in $\varepsilon_{1,2}$ terms, whereas the angle-dependent terms describe the contribution of the switching transitions. The angular dependence of $\hat{\varepsilon}$ matrix elements arises due to the feedback between the extinction of a certain normal mode and the intensity of other normal mode with complementary polarization. Such a feedback, provided by OSC's, is described by eqs.(14,18,19).

The dielectric permittivity tensor $\hat{\varepsilon}$, eq.(20), describes the optical response of a nonlinear system, which parameters vary with the polarization of incident light. Taking into account eqs.(2,14,20), we may write:

$$\hat{\varepsilon}_{11} = \hat{\varepsilon}_{11}(\omega, I_2/I_0), \tag{21a}$$
$$\hat{\varepsilon}_{22} = \hat{\varepsilon}_{22}(\omega, I_1/I_0). \tag{21b}$$

This type of nonlinearity differs from the conventional one,

$$\hat{\varepsilon} = \hat{\varepsilon}(\omega, I_0), \tag{22}$$

when the nonlinear effects are governed by the intensity I_0 of incident light.

All terms in eq.(20) are functions of frequency, which explicit form should be specified proceeding from a relevant microscopic theory. E.g., the function χ will depend on the profile of the potential barrier (figure 2) via the transition amplitudes W, W', W'' (14), etc. It is not our intention here to develop such a detailed theory, as it would require a number of additional model assumptions. Instead, we stress, that crucial is the angular dependence in eq.(20) itself, and not a special form of the functions $\varepsilon_{1,2}(\omega)$ and $\chi(\omega)$. However, taking into account, that $\chi(\omega)$ describes the hopping of the OSC over the potential barrier, it seems reasonable to assume, that it should exhibit a resonance at the frequency ω_{12}, eq.(12).

Given the dielectric permittivity tensor, it is straightforward to derive the absorption coefficient K of the film with thickness d for an arbitrary polarization of light (for simplicity, the effects of boundary reflections are neglected):

$$I_{out}/I_{in} = \exp(-Kd) = U_1 \cos^2\varphi + U_2 \sin^2\varphi, \tag{23}$$

where I_{in} and I_{out} are the intensities of incident and transmitted wave, respectively, $U_i = \exp(-2(\omega/c)n''_i d)$ and $n_i = n'_i + in''_i = \varepsilon_i^{1/2}$ are refractive indices, ε_i's being the eigenvalues of the dielectric permittivity tensor. The

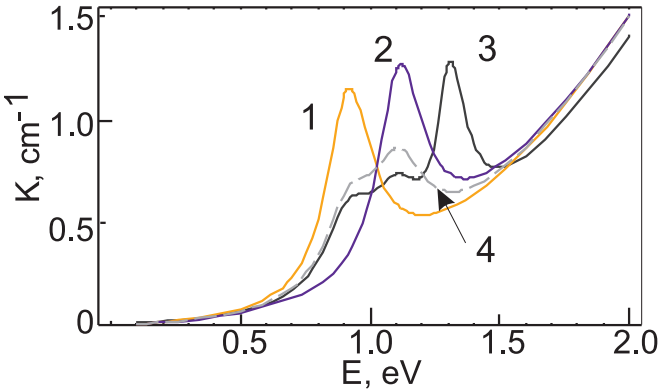

FIG. 3: The absorption coefficient, obtained with eqs.(23,24). Curves 1 and 2 depict the response in two plane polarizations, $\varphi = 0$ and $\varphi = \pi/2$, respectively. The curve 3 simulates the spectrum, obtained in unpolarized light. Noteworthy is the novel peak at 1.3 eV, not observed in two previous polarizations. The dashed curve 4 is the unpolarized light spectrum of a conventional medium and is obtained using the trivial counterpart of the dielectric tensor, eq.(20), where the angle-dependent terms are omitted.

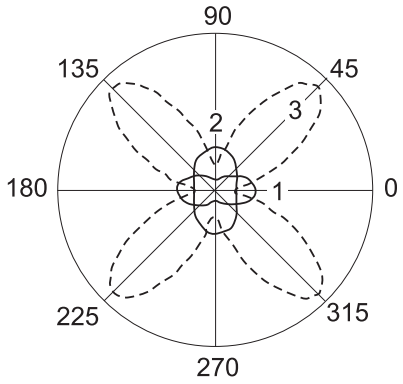

FIG. 4: The absorption coefficient $K(\omega, \varphi)$, eq.(23), at selected frequencies as a function of polarization angle φ. 1: $\omega = \omega_1$. 2: $\omega = \omega_2$. 3: $\omega = \omega_3$ (see text).

absorption coefficient for unpolarized light is obtained by averaging over the polarizations:

$$K_{unpol.} = \frac{1}{2\,\pi} \int_0^{2\,\pi} K(\varphi)\, d\varphi. \tag{24}$$

The key result of the present model is that the contribution of the $\chi(\omega)$ term in the dielectric tensor, eq.(20), to the absorption coefficient is effective only in presence of both normal modes with polarizations p_1 and p_2. This may be seen, noting, that the absorption coefficient and other functions of optical response are expressed as the functions of the "matrix elements":

$$\langle \mathbf{E}|\hat{\varepsilon}|\mathbf{E}\rangle = |E_0|^2 \left((\varepsilon_1 + \chi\, n_1(\varphi)) \cos^2\varphi + (\varepsilon_2 + \chi\, n_2(\varphi)) \sin^2\varphi \right) \tag{25}$$

where $\mathbf{E} = E_0(\cos\varphi, \sin\varphi)$ is the electric vector of incident light. With $n_{1,2}$ given by eqs.(14), it is obvious, that the contribution of $\chi(\omega)$ to this quantity vanishes for φ close to 0 and $\pi/2$.

For illustrative purposes, we consider a simple case, when $\varepsilon_{1,2}(\omega)$ and $\chi(\omega)$ show well resolved resonances at different frequencies. All contributions in the $\hat{\varepsilon}$ tensor (20) have been simulated with the Lorentzian functions (17). The "conventional" contributions due to some optical transitions in the medium are chosen to be $\varepsilon_{1,2}(\omega) = \mathcal{L}(\omega; \{\omega_{1,2}, 1.1, 0.2\}, \{2.5, 20.2, 2.0\})$ with $\omega_1 = 0.9$ eV and $\omega_2 = 1.1$ eV. The OSC contribution have been taken in the form: $\chi(\omega) = \mathcal{L}(\omega; \{\omega_3, 5.5, 0.1\})$ with a sharp resonance at $\omega_3 = 1.3$ eV. The ratios $\widetilde{W}/W'$ and W''/W' in eq.(14) are found to show a minor effect on the final results when varying in a broad range and are (arbitrarily) set equal to 1.0 and 0.2, respectively. As shown in figure 3, the absorption spectra are different in two basic polarizations (curves 1 and 2), that is typical of the anisotropic media. The sharp peaks, observed in two polarization, are also visible as the slight features at the curve 3, simulating the unpolarized light spectrum. In intermediate polarizations, the switching transitions come into play, and the curve 3, simulating the natural light spectrum, displays a novel strong resonance at 1.3 eV, that emerges due to $\chi(\omega)$ and is absent in both plane polarizations. This behaviour is clearly at variance to what is expected for any conventional system, where the unpolarized spectrum should be merely a superposition of the spectra, observed at two different polarizations. For comparison, such a "usual" unpolarized spectrum, obtained from eqs.(20,24) with $\chi \equiv 0$ is also shown at figure 3 (curve 4). Of course, the numerical quantities in this example do not pretend to be the fitting parameters, relevant to the experimental spectra at figure 1.

The polarization dependence of the model spectra is shown at figure 4. It is seen, that the indicatrix of the absorption coefficient at the resonance frequency ω_1 behaves as $K(\omega_1, \varphi) \sim \cos^2\varphi$ and is maximum at the polarization angle $\varphi = 0$, while at the resonance frequency ω_2, $K(\omega_2, \varphi) \sim \sin^2\varphi$ is maximum at $\varphi = \pi/2$. The absorption coefficient at the switching frequency ω_3 is minimum in both of these basic polarizations, but shows a pronounced maximum at $\varphi = \pi/4$. The reason is that the switching of OSC requires both normal modes to be excited, and this condition is optimally fulfilled in $\pi/4$-polarization.

V. DISCUSSION

The theoretical interpretation of the optical data on cupric oxide CuO is particularly difficult because of its low symmetry monoclinic tenorite structure. For the geometry of experiment, as described in Section 2, the tensor $\hat{\varepsilon}$ of an ideal single crystalline sample is decomposed

into the scalar ε_b and the in-plane part[8]:

$$\hat{\varepsilon}_{ac} = \begin{pmatrix} \varepsilon_{xx} & \varepsilon_{xz} \\ \varepsilon_{zx} & \varepsilon_{zz} \end{pmatrix}, \quad (26)$$

where the orthogonal coordinate axes $\mathbf{x}$, $\mathbf{z}$ lie in plane of the film (the crystallographic ac plane). The normal modes are the transverse waves with elliptic polarizations, propagating with (complex) refractive indices $n_{1,2}$:

$$2n_{1,2}^2 = (\varepsilon_{xx} + \varepsilon_{zz}) \pm ((\varepsilon_{xx} - \varepsilon_{zz})^2 + 4\varepsilon_{xz}\,\varepsilon_{zx})^{1/2}. \quad (27)$$

Within the disordered sample, the propagation direction of light may deviate from that of the $\mathbf{b}$ axis because of the scattering on the nonuniformities, induced with the fast particle bombardment. The excitation of both transverse and longitudinal modes can therefore be expected. Thus, more realistic theoretical approaches should deal with the non-diagonal tensors of a general form, as distinct from the model assumption, used in eq.(20), that have been made for the illustrative purposes.

However, we would like to emphasize, that the interpretation of the experimental spectra at figure 1 does not reduces to the accurate analysis of the normal modes of some disordered low-symmetry linear medium. In fact, two different problems should be distinguished here: those of the explanation of $K(E)$ dependence for a certain polarization, and those related to the relative positions of the three spectra. While the form of the $\hat{\varepsilon}(\omega)$ tensor determines the shape of the optical spectrum for any particular polarization, the problem of the abnormal polarization dependence of the spectra is beyond the scope of a linear response theory, as have been discussed in Section 2. We believe, that it can be resolved only assuming the explicit dependence of the dielectric tensor on the polarization of incident light.

The polarization-dependent switching effect, proposed in previous sections, provides the required general form of the $\hat{\varepsilon}$ tensor, eq.(20), and makes it possible to realize, how in principle the abnormal polarization dependence of the absorption spectra of CuO (figure 1) could be understood. As the theoretical spectra at figure 3 have been obtained with rather arbitrarily chosen numerical values of the model parameters, our present results are preliminary and allow but a qualitative comparison with experiment. However, the polarization dependence of the calculated spectra (figure 4) stems from general formulae (14,20) and is not related to some particular choice of the fitting parameters. Overall, the spectra at the figure 3 capture the most essential features of experiment, except that the curve 3, simulating the natural light spectrum, displays the traces of the peaks 1 and 2, in contrast to the corresponding spectrum 3 at figure 1. We believe, that rather large experimental errors, noted in[1], make it difficult to decide, whether some slight features can in fact be resolved at the broad tails of the experimental spectra.

The natural question arises about the microscopic physics of OSC. To our opinion, under certain conditions the Jahn-Teller clusters may be regarded as their possible prototypes. To be specific, we consider $E - b_1 - b_2$ - problem[9], that arises e.g. in the Jahn-Teller theory of the square cluster with the D_{4h} point symmetry when its electron ground state orbital $E_u\{x, y\}$ doublet is coupled to b_{1g} and b_{2g} local lattice modes. The vibronic interaction causes the spontaneous symmetry breaking of the cluster and the profile of its adiabatic potential along the active Jahn-Teller coordinate is shown schematically at figure 2. E.g., if the vibronic coupling with b_{1g} mode dominates, the cluster acquires one of two equivalent static b_{1g} (rhombic) distortions (the ground state average value of the corresponding normal coordinate being non-zero: $\langle Q_{b1g}\rangle = \pm Q^0_{b1g}$). The resulting vibronic ground state wave functions of the cluster frozen in one of the two potential wells have the symmetry properties: $\Psi_\pm \sim x, y$. If there is also some excited high-symmetry state $\psi' \sim A_{1g}$, then the photon-activated switching between the two Jahn-Teller distorted configurations, involving the intermediate ψ' state, can be considered (figure 2). Noteworthy is that the dipole transition matrix elements $\langle\psi'|(\mathbf{E}\cdot\mathbf{d})|\Psi_\pm\rangle$ are allowed in orthogonal polarizations, $\mathbf{E} \parallel x$ and $\mathbf{E} \parallel y$, respectively. Thus, the Jahn-Teller cluster may have all essential ingredients of the OSC as discussed in Section 3. In principle, the similar arguments may also be relevant for other cases of a spontaneous symmetry breaking and the role of the lattice subsystem is not crucial. In this connexion we note an interesting idea, put forward in[10], about the possibility of the "purely electronic" Jahn-Teller effect in strongly correlated systems, that consists in the correlational shift of the electron shells, similar to the conventional vibronic distortion of the lattice.

The Jahn-Teller effect may in fact play an important role in the physics of cuprates. In particular, it was shown in a series of papers[11,12,13,14], that many peculiarities of the optical and structural properties of CuO and other cuprates allow a consistent interpretation within the model of polar electron-hole pseudo-Jahn-Teller (PJT) centers, which underlying physics is dominated by the near-degeneracy of A_{1g} and E_u molecular orbitals. The authors note, that the favourable conditions for the nucleation of PJT centers is the chemical doping, fast particles bombardment or other perturbations, that may locally disturb the stability of the parent system. Thus, we believe the PJT centers in cuprates to be a plausible candidates to the OSC's, considered in this work.

VI. SUMMARY

In conclusion, the aim of the present work is to explain the polarization anomalies observed in the optical absorption spectra of cupric monoxide CuO after the bombardment with He^+ nuclei[1]: the spectra, obtained in unpolarized light were found to display new features, that are not observed in two different plane polarizations

(figure 1). Note, that for such an effect to be revealed, the comparative analysis of at least three spectra in different polarizations is required. To the best of our knowledge, no similar results have been ever observed in experiment or investigated theoretically. Leaving aside a number of involved problems, concerning the electronic structure of cupric monoxide and its changes under the bombardment, we addressed the question: how, at least qualitatively, these polarization anomalies could be explained. To this end we have proposed the model of a nonlinear system, which dielectric permittivity $\varepsilon = \varepsilon(\omega, \varphi)$, eqs.(20,21), depends explicitly on the polarization angle φ of light, propagating through the system. This type of nonlinearity differs from the conventional one, $\varepsilon = \varepsilon(\omega, |E|^2)$, where $|E|^2$ is the intensity of light. In our model, such a nonlinearity arises due to the special structures, the optical switching centers (OSC), which behaviour is as follows: i) The center has two metastable states, $|1\rangle$, $|2\rangle$; ii) it can hop from $|1\rangle$ to $|2\rangle$, absorbing the photons with polarization p_1 and iii) back from $|2\rangle$ to $|1\rangle$, absorbing the photons with polarization p_2, *orthogonal* to p_1. These switching transitions $|1\rangle \leftrightarrows |2\rangle$ give rise to the novel channel of optical absorption, that is only effective in presence of waves with both polarizations. This explains, how the new spectral features, not observed in two orthogonal polarizations p_1 and p_2, can emerge in unpolarized light.

We propose, that the systems with the properties, typical of OSC, can be found among the Jahn-Teller clusters. Taking into account, that the polar Jahn-Teller centers have been shown to play an important role in the optics of copper oxides[1,5,6,11,12,13,14], we believe, that the present model may indeed capture some interesting and yet little explored physics of cuprates. More detailed theory, that allows the quantitative comparison with experiment, will be deferred for other publication.

Acknowledgments

The author is indebted to Drs. Yu.P. Sukhorukov and Yu.D. Panov for many valuable comments. The discussions with Profs. N.G. Bebenin and V.Y. Shur are also acknowledged. The work is supported by Grant 04-02-96068 RFBR URAL 2004.

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