Electron-hole drops in vanadium dioxide

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The detection of anisotropy in the coefficient of reflection, induced by picosecond excitation of a vanadium dioxide film with quantum energy 1.17 eV, is reported. The phenomenon is related to the formation of electron-hole drops.

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In an previous paper,[1] we presented results from a study of the fast kinetics of a metal-semiconductor phase transition (MSPT) in a vanadium dioxide film when the MSPT is excited by radiation pulses with duration $\sim 6\times 10^{-12}$ s. The time τ_i obtained in Ref. 1, during which the MSPT is initiated, is $\tau_i \sim 10^{-12}$ s and is less than the phonon transit time $\tau_i \sim d/v_{3s} \sim 10^{-10}$ s (d is the film thickness, and v_{3s} is the velocity of sound). For this reason, we investigated in detail the leading edge of the MSPT kinetics with time resolution 2×10^{-11} s using the method of temporal probing with pre-excitation.[2] Single picosecond pulses were obtained with the help of a pulsed YAG:Nd^{3+} laser, operating in the self-locking regime. The parameters of a single amplified pulse are radiation wavelength, 1.06 μm; energy ~ 3 mJ; duration $(18-20)\times 10^{-12}$ s; spectral width $\Delta\lambda \sim 0.7$ Å; contrast $>10^4$. The ratio of the probing and exciting beam intensities is 1:10.[3] The angle of incidence of the beams on the film did not exceed 10°.

As in Ref. 1, we studied a vanadium dioxide film placed on an aluminum reflecting layer deposited on a polychore substrate. In the experiments we monitored the reflection coefficient of the film, whose variation we attributed to the development of MSPT. The experiments were performed at film temperatures $T=300$ K and $T=77$ K. With the help of a $\lambda/2$ plate and a Glans prism placed in the path of the exciting beam, we determined the MSPT kinetics for cases in which the exciting and probing beams were linearly polarized in mutually orthogonal and parallel planes (see Fig. 1). The results, presented in Fig. 1, show that when a vanadium dioxide film is excited by a picosecond pulse with power density $P \sim 3\times 10^8$ W/cm^2 and quantum energy $\hbar\omega = 1.17$ eV, the change in the reflection coefficient in a time $\sim 25\times 10^{-12}$ s is distinctly anisotropic. At the same time, the long axis of the polarization ellipse is oriented along the vector **E** of the exciting field. The time variation of the induced anisotropy $\gamma = (R_{\parallel} - R_{\perp})/(R_{\parallel} + R_{\perp})$ is shown in Fig. 2. It is evident that the induced anisotropy has a pulsed nature, coinciding along the leading edge with the pumping pulse and lagging behind it insignificantly (by $\sim 5\times 10^{-12}$ s) along the trailing edge. For a more complete description of this phenomenon, we add the fact that it is observed both at $T=77$ K and at $T=300$ K with the difference that at $T=300$ K the quantity γ decreases ($\gamma \sim 0.1$).

We performed a series of special experiments in order to verify that the observed phenomenon is not a result of the interference interaction of the excited and probing

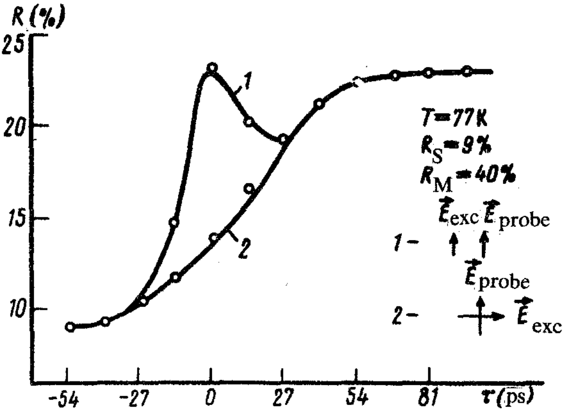

FIG. 1. Leading edge of MSPT in a vanadium dioxide film. R_s and R_m are the coefficients of reflection in the semiconducting and metallic phases.

beams. As a result, the energy in the exciting beam is pumped in the direction of the probing beam. For this purpose, we varied the ratio of the exciting and probing beam intensities from 1:10 to 1:10 (Ref. 3) for fixed geometry of the experiment. However, the results of the measurements were the same as in Fig. 1.

In our opinion, the reason for the observed phenomenon is the formation of electron-hole drops (EHD) in the semiconducting phase of the vanadium dioxide film, induced by the powerful pumping pulse. During the formation process, the liquid drop with a metallic conductivity is located in the electric field of the light wave. In this case we should expect the appearance of the electrostriction effect, leading to the deformation of the drop. As a result, the drop assumes an ellipsoidal shape.[3] The magnitude of the deformation of a spherical conducting drop with radius r and surface tension σ, located in an external field, can be determined in analogy with Ref. 3. We write the electrostatic energy

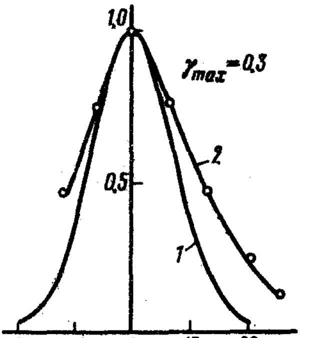

FIG. 2. The induced anisotropy γ in the reflection coefficient (curve 1). The pumping pulse is Gaussian (curve 2). Both curves are normalized to their maximum value.

$$U_{\mathrm{el}} \cong -\frac{3VE^2}{8\pi} - \frac{3V}{10\pi}\frac{a-b}{r}E^2$$

and the surface energy

$$U_{\mathrm{sur}} \cong \frac{7V\sigma}{2r} + \frac{V\sigma}{8r^3}(a-b)^2$$

for an elongated ellipsoid with semiaxes a and b ($a > b = r$, and V is the volume), located in an electric field with intensity E. Minimizing the sum $U_{\mathrm{el}} + U_{\mathrm{sur}}$ with respect to the parameter $a - b$, we obtain

$$\frac{a-b}{r} = \frac{6}{5}\frac{rE^2}{\pi\sigma}.$$

This expression is valid for an elongated, nearly spherical ellipsoid, i.e., for $\sqrt{1 - b^2/a^2} \ll 1$. It permits estimating the magnitude of the energy in the field E^2, for which the electrostriction effect will stretch the EHD along the electric field vector **E** into an ellipsoid with semiaxes $(a-b)/r \sim 0.1$. We assume r and σ to be equal to the well-known values for EHD in Ge, i.e., $r \sim 10^{-4}$ cm, $\sigma \sim 10^{-4}$ erg/cm^2.[4] Then $E^2 \sim 2.5 \times 10^{-8}$ J/cm^3. This estimate shows that the energy density of the field obtained experimentally ($\sim 5 \times 10^2$ J/cm^3) is much greater than the quantity presented and is sufficient to deform a spherical drop into an ellipsoid even if σ of the drop is $\sim 5 \times 10^2$ erg/cm (Al,Sn). Furthermore, it is clear that the volume of the film filled with EHD shaped like an elongated ellipsoid and oriented with the long semiaxes along the vector **E** of the pumping light field, has distinct anisotropic properties.

Thus, a *d-d* – electron-hole plasma, excited in the semiconducting state of vanadium dioxide by quantum energy 1.17 eV at pumping power densities $P \sim 3 \times 10^8$ W/cm^2 condenses into EHD within a time $< 10^{-11}$ s.

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