Polarization-sensitive photocurrent in the resistive Ag/Pd films

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Abstract. We report on the observation of helicity dependent photocurrent of the 20 μm thick silver–palladium (Ag/Pd) films manufactured by the thick-film technology. The transverse photocurrent is observed at oblique incidence of laser radiation with different wavelengths in the spectral range of 266 – 2100 nm. At the wavelength range of 532 - 2100 nm the polarity of the transverse photocurrent is positive (negative) for the left- (right-) circular polarized beam. We show that action of high temperature on the films in vacuum results in the decrease of longitudinal photocurrent due to the reduction of PdO content. The photon drag effect is suggested to be the origin of the polarization-sensitive photocurrent in the Ag/Pd films. The obtained results show that the Ag/Pd resistive films may be of interest for polarization-sensitive measurements.

1. Introduction

Interaction of pulsed laser radiation with matter can lead to the interesting phenomena. One of such phenomena is helicity dependent transverse photocurrent induced by the circular polarized laser beam. The transverse photocurrent is perpendicular to the plane of the incidence. The origin of helicity dependent photocurrent can be the circular photogalvanic effect (CPGE) [1] and the photon drag effect [2].

The CPGE arises in gyrotropic media where the symmetry with respect to mirror transformation is broken, and is conditioned by the peculiarities in the band structure of gyrotropic crystals [3]. It should be noted that depending on the type of symmetry, the CPGE can be observed both at oblique and normal incidence of elliptically polarized light onto the sample surface [4].

The photon drag effect (PDE), first discovered in [5, 6], leads to the generation of the photo-electromotive force (photocurrent) by transferring the momentum of photons of incident radiation to the charge carriers in the intraband or interband energy transitions. Unlike the CPGE, the PDE can be observed in centrosymmetric media [2]. In the presence of circular polarization of exciting radiation, the PDE can be accompanied by the helicity dependent photocurrent.

The present work is aimed at studying the spectral and temperature dependences of a polarization-sensitive photocurrent in the silver-palladium (Ag/Pd) films at the wavelength range of 266 - 2100 nm.
2. Experimental

2.1. Ag/Pd films
Ag/Pd resistive films have stable electrical characteristics and have long been used in electronics. They are widely applied for obtaining hybrid microcircuits, multocrystal modules, integrated circuits assemblies and passive electronic components such as resistors, inductive elements, multilayer capacitors. The Ag/Pd films are produced using technologies [7] based on burning of a special paste, which contains silver oxide (Ag₂O) and palladium (Pd), at high temperatures on the insulating substrate surface. The optical and electronic properties of the Ag/Pd films are varied in a wide range depending on a number of factors including thermodynamics and kinetics of Pd oxidation during burnout and firing, Ag diffusion and migration, properties and concentration of inorganic and organic additives. The film studied in our experiments was fabricated at temperature of 878 K using a well-known technology of the thick-film resistors on ceramic substrates. The size of the film obtained was 20 × 20 mm (figure 1a). The thickness of the film was about 20 μm. To measure the photocurrent the film was provided with two parallel silver film electrodes which were arranged along the opposite sides of the film between the dielectric substrate and the Ag/Pd film. Scanning electron microscopy showed that the Ag/Pd film is porous with pore size ranging from 25 to 500 nm (figure 1b).

Figure 1. (a) Photograph of the Ag/Pd film, where (1) substrate; (2) Ag/Pd film; (3) silver measuring electrodes located between the substrate and the film, and (b) the scanning electron microscope image of a film surface section.

2.2. Experimental setup
The transverse photocurrent in the resistive Ag/Pd film was induced by laser radiation at different wavelengths in the range of 266 – 2100 nm. In the range of 266 – 1064 nm we used a passive Q-switched single-mode pulsed YAG:Nd³⁺ laser (λ = 1064 nm) with frequency converters to second (λ = 532 nm), third (λ = 355 nm), and fourth (λ = 266 nm) harmonics. To carry out the experiments in the range of 1350 – 2100 nm we have used an automated frequency tunable infrared laser source (Laser Vision). The linearly polarized radiation of laser was converted into circularly polarized radiation by zero order quartz quarter-wave plates (figure 2a). These quarter-wave plates were orientated with their planes perpendicular to the direction of laser radiation propagation. The ellipticity of the laser beam was determined by the angle φ between the vertical axis x’ and the slow axis nₑ (figure 2a). The radiation fell onto the Ag/Pd film at an angle α = 45°. The measuring electrodes A and B were orientated in such way that they were parallel to the incidence plane σ (the transverse configuration). The laser radiation did not fall on these electrodes during the experiments. The pulsed voltage induced by polarized laser radiation was measured using a TDS7704B digital oscilloscope (Tektronix) with an impedance of r = 50 Ω connected directly to the sample electrodes.
To study the temperature influence on the photocurrent in Ag/Pd films we used $p$-polarized laser pulses of 1064 nm and the longitudinal configuration, where the electrodes were oriented perpendicular to the incidence plane and the photocurrent along $x$ axis was measured (figure 2b). The Ag/Pd film was cyclically heated in vacuum. The film was placed in a vacuum chamber, where atmosphere pressure was reduced upon $1.3 \times 10^{-3}$ Pa. Heating was performed with the special 15 Ω ceramic isolated wolfram heater. The same oscilloscope was used for measuring photocurrent. Temperature $T$ was changed cyclically: first the Ag/Pd film was heated to $T = 270^\circ$ C and this temperature was carried awhile, then the film cooled down. During the process of heating and cooling the film was irradiated by the laser and the photocurrent was registered. The film resistance was also registered by a digital ohmmeter. After cooling film upon the room temperature the heating and cooling process was repeated.

To estimate the changing composition of the Ag/Pd film the X-ray diffraction (XRD) analysis was performed using a D2 PHASER diffractometer with monochromatized CuKα radiation.

3. Experimental results

A nanosecond optical pulse produces an electric current directed perpendicular to the $\sigma$ in the film that manifests itself as a unipolar nanosecond electric pulse of voltage $U_y$. The photocurrent can be determined as $i_y = U_y/r$.

Experimental investigation of the value $i_y$ as function of the incidence angle $\alpha$ show that the relation $i_y(\varphi, \alpha) = -i_y(\varphi, -\alpha)$, where $-90^\circ < \alpha < 90^\circ$, holds true in the entire range of wavelengths (266 – 2100 nm) and for any fixed angle of $\varphi$. That means that the signal polarity depends on the direction of the wave vector $k$ of incident radiation. According to [2, 8], this allows us to consider the observed effect as PDE.

The dependences of the photocurrent on the angle $\varphi$ have been obtained for the wavelengths of 266, 355, 532, 1064, 1350, 1450, 1650, 1750, 1850, 1950 and 2100 nm. For example insets in figure 3 shows the normalized photocurrent dependences of the peak amplitude of the measured signal on $\varphi$ for $\lambda = 355$ nm and 1750 nm. The normalized photocurrent is determined as $\overline{i_y}(\varphi) = i_y(\varphi)/i_{y, \text{max}}$, where $i_{y, \text{max}}$ is the maximum absolute value of $i_y$ in the range $0 < \varphi < 180^\circ$. Similar experimental dependences of $\overline{i_y}(\varphi)$ have been obtained for other wavelengths. For all wavelengths the signal is absent at $\varphi = 0, 90^\circ, 180^\circ$ ($p$-polarized light). The photocurrent is also absent in the case of $s$-polarization, which has been proven by additional experiments. For $\lambda = 1750$ nm (figure 3, right inset) the signal is positive for...
circular polarization with the negative sign ($0 < \phi < 90^\circ$) and negative for circular polarization with the positive sign ($90^\circ < \phi < 180^\circ$), the photocurrent depending considerably on the degree of ellipticity of light polarization, i.e. on the angle $\phi$. However for $\lambda = 355$ nm (figure 3, left inset) on the experimental dependence obtained with both left- and right-hand polarized light and the phase angle varied within $0 < \phi < 90^\circ$ and $90^\circ < \phi < 180^\circ$, respectively, the signal polarity takes both negative and positive values.

![Figure 3](image.png)

**Figure 3.** Ratio $\xi$ of the amplitudes of photocurrent circular and linear contributions vs. the incident radiation wavelength $\lambda$. Insets show the normalised photocurrent as a function of the angle $\phi$ (dots), obtained for the excitation wavelengths of 355 (left inset) and 1750 nm (right inset). Curves (1) show approximating dependences, curves (2) and (3) – dependences of circular and linear contributions, respectively. Polarization ellipses of radiation for different angles $\phi$ are shown at the top of insets.

For all wavelengths the data obtained is well approximated with relation:

$$\tilde{i}_y = i_1(\lambda)\sin(2\phi) - i_2(\lambda)\sin(4\phi),$$  \hspace{1cm} (1)

where $i_1(\lambda)$ and $i_2(\lambda)$ represent the magnitudes of the helicity-sensitive (circular) and insensitive (linear) contributions of normalized photocurrent, respectively. The magnitudes of both contributions change with changing the wavelength independently. For $\lambda = 355$ nm $i_1 = 0.527$, $i_2 = 0.602$ and for $\lambda = 1750$ nm $i_1 = 0.986$, $i_2 = 0.083$. The dependence $\xi = i_1(\lambda)/i_2(\lambda)$ has been plotted in figure 3. It can be seen that, in the ultraviolet spectral region, the magnitude $\xi$ is comparable with unity, while in the infrared region for the wavelengths of 1350 – 2100 nm the amplitude $i_1(\lambda)$ of the circular contribution is many times greater than the amplitude $i_2(\lambda)$ of the linear contribution. Thus, it may be assumed that virtually “pure” circular PDE is observed in the wavelength range of 1350 – 2100 nm. Expression (1) indicates that, for the counterclockwise-polarized light $0 < \phi < 90^\circ$ the photocurrent $i_y(\phi)$ is strictly positive for $\xi > 2$. It follows from figure 4 that for $\lambda = 532$ nm the ratio $\xi \approx 2$. This means that in the spectral range of 532 $< \lambda < 2100$ nm, for transverse experimental geometry, the photovoltaic signal has positive polarity in the case of the negative sign of circular polarization and negative polarity in the case of the positive sign. Thereby the pronounced polarization dependence of the photocurrent makes the Ag/Pd films an interesting material for fabrication of helicity sensitive photon drag photodetectors.
The results of experimental investigations of the temperature influence on the longitudinal photocurrent $i_x=U_x/r$ are presented in the figure 4, where $U_x$ is longitudinal photo-induced voltage. Here, inset demonstrates the example of the changing longitudinal photocurrent during a cycle of heating and cooling. One can see that during heating $i_x$ increases. When the film cools down, $i_x$ reduces and becomes lower than initial value. Figure 4 shows the plots of $i_x$ vs. quantity of heating and cooling cycles. It can be seen that every cycle of heating and cooling leads to decrease of longitudinal photocurrent. The behaviour of the interelectrode resistance of the film is similar. All these results obtained can be explained by the varying of the content of Ag/Pd film during of it heating. It is well known that PdO is unstable at high temperature [9, 10]. In our experiments decomposition of PdO occurs in vacuum. For this reason oxygen is removed from the films and, therefore, Pd is not oxidized backward.

XRD patterns of the film is shown in figure 5. We can see that XRD pattern of the film before heating contains the Ag-Pd, PdO, and Al$_2$O$_3$ reflections (figure 5, curve 1). The Ag-Pd and PdO reflections are in agreement with composition of initial resistive paste, therefore corresponding solid solutions are present in the film. Al$_2$O$_3$ reflection belongs to substrate on which the film was produced. After cyclical heating and cooling the intensity of the PdO reflections have been reduced with respect to Ag-Pd reflections (figure 5, curve 2). Therefore the content of PdO in the film after heating is lower than before. Moreover new little reflections of Pd appear in the XRD pattern. Thus PdO content is reduced, that results in the photocurrent decrease. These results indicate that PdO content influence on polarization-sensitive photocurrent in Ag/Pd films.
Figure 5. X-ray diffraction patterns of the film before (1) and after (2) heating and bar diffraction patterns of detected phases (CuKα).

4. Conclusion

Thus, the present work demonstrates that helicity dependent photocurrent is generated in Ag/Pd resistive films at broad wavelength range from 266 to 2100 nm. In the wavelengths range of 532 - 2100 nm, the polarity of the photocurrent is unambiguously determined by the sign of circular polarization. These results demonstrate the possibility of employing resistive Ag/Pd films for the creation of high-performance sensors of the circular polarization sign of the incident light in a wide spectral range.

The action of high temperature on the films in vacuum results in the photocurrent decrease due to the reduction of PdO content.

The PDE is suggested to be the origin of the helicity dependent photocurrent.

Acknowledgments

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