Ultraviolet Sensitive Ultrafast Photovoltaic Effect in Tilted $KTaO_3$ Single Crystals *

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An ultraviolet sensitive ultrafast photovoltaic effect is observed in tilted 10° $KTaO_3$ (KT) single crystals. The rise time of the transient photovoltaic pulse is 497.4 ps and the full width at half maximum is 974.6 ps under irradiation of a 266 nm laser pulse with 25 ps duration. An open-circuit photovoltage sensitivity of 328 mV/mJ and a photocurrent sensitivity of 460 mA/mJ are obtained. The experimental results demonstrate the potential applications of KT single crystals in ultraviolet detection.

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Much attention has been focused on the development of high quality and reliable ultraviolet (UV) photodetectors due to their wide and important applications in civil living and military activities.[1] Especially, UV detectors with high discrimination against visible light are more favorable for occasions requiring detection of UV radiation in a background of visible radiation. Therefore, in the past decades, much research has been focused on wide bandgap materials, such as GaN, ZnO, SiC, diamond, and so on, which appear to be attractive choices for selective UV detection.[2−5] However, these devices require a complicated fabrication process and costly manufacturing.

Potassium tantalite ($KTaO_3$, KT) appears to be excellent in the field of electronic industry. KT and its compounds $KTa_{1-x}Nb_xO_3$ (KTN), as an important family of perovskite oxides, have a variety of prolific physical characteristics, such as ferroelectric, electro-optic, photorefractive, piezoelectric, acousto-optic, and nonlinear optical properties, which are highly relevant for applications in lead-free and biocompatible transducers, optical waveguides, holographic gratings, digital deflectors, and optical modulators etc.[6−13] Thus, interest in exploring this KT family in multifunctional devices is growing. However, as we know, the photoelectric effect in KT crystals has not been widely studied. Our previous work has reported UV photovoltaic effects in tilted $SrTiO_3$, $LaAlO_3$, ZrO_2 single crystals.[14−16] In this Letter, we report the ultrafast and ultraviolet sensitive photovoltaic effect in KT single crystals. The KT crystal presents a highly sensitive and picosecond-order photovoltaic response to UV irradiation.

The wafers used in the present study are commercial KT (001) single crystal with a purity of 99.99% and mirror single polished. The (001) plane is tilted to the wafer surface with an angle of 10°, as shown in the inset of Fig. 2. The tilting of the c-axis was further confirmed by x-ray diffraction measurement with the usual $\theta - 2\theta$ scan. The geometry of the sample is 5 mm × 10 mm with a thickness of 0.5 mm. The absorption spectrum of the KT single crystal was measured, as shown in Fig. 1. The absorption peak and the absorption edge were located at 340 nm and 360 nm, respectively. The bandgap of KT single crystal, determined by the absorption spectrum, is about 3.5 eV as shown in the inset of Fig. 1.

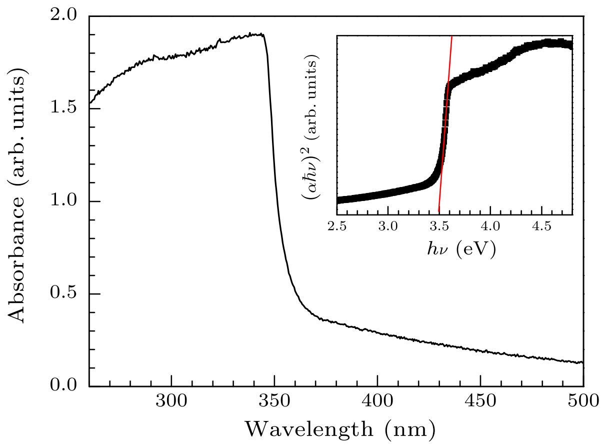

Fig. 1. Absorption spectrum of $KTaO_3$ single crystal with a thickness of 0.5 mm. Inset: a plot of the relationship of $(\alpha h\nu)^2$ with single photon energy $h\nu$.

To measure the photovoltage, two indium electrodes, painted on the surface of KT single crystal, are separated by 1 mm. The electrodes were always kept in the dark to prevent the influence of the electrical contacts. A fourth harmonic of an actively-

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passively mode-locked Nd:YAG laser was used as the excitation source, operating at a wavelength of 266 nm with 25 ps duration at a 1 Hz repetition rate and with an energy density of 12.7 µJ/mm^2. Thus, the on-sample energy was 63.5 µJ. The photovoltaic signals were measured by a sampling oscilloscope (Tektronix TDS7254B, 2.5 GHz bandwidth) with an input impedance of 1 MΩ. The schematic measurement circuit is shown in the inset of Fig. 2.

Figure 2 shows a transient open-circuit photovoltaic signal when the tilted 10° KT single crystal was irradiated directly by a YAG laser pulse. The peak photovoltage V_p was 20.8 mV and the corresponding photovoltage sensitivity was 328 mV/mJ. The rise time (10–90%) was 3.2 µs and the full width at half maximum (FWHM) was 142 µs. It should be noted that the transient signal had a sharp rise at the beginning, but with a slow fall at the end due to the RC effect in the measurement circuit.

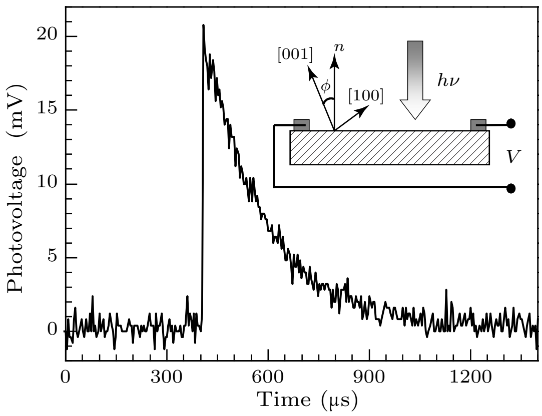

Fig. 2. Transient open-circuit photovoltaic pulse for the tilted $KTaO_3$ single crystal under irradiation of a 266 nm YAG laser pulse. Inset: the schematic circuit of the measurement system.

To reduce the influence of the measurement system, we put a 0.5 Ω resistance connected in parallel with the tilted KT single crystal as shown in the inset of Fig. 3. The photovoltage was measured under the same experimental conditions as Fig. 2. As shown in Fig. 3, the rise time and the FWHM were dramatically reduced to about 497.4 ps and 974.6 ps, respectively. The peak photovoltage V_P was 14.6 mV. The photocurrent, calculated by $I_p = V_p/0.5\,\Omega$, was 29.2 mA and the corresponding photocurrent sensitivity was 460 mA/mJ. The dip of the signal and slight oscillation behavior may come from impedance mismatching in the circuit. The experimental results show that the photovoltaic response of tilted KT single crystal to UV irradiation is not only a picosecond ultrafast process but also a highly sensitive process.

In order to understand the mechanism of the photovoltaic effect, we measured the photovoltages in tilted KT single crystal with a 1.064 µm Nd:YAG laser and its second harmonic laser. No photovoltaic signals were observed for the reason that the photon energies of 1.064 µm and 532 nm wavelengths are smaller than the bandgap of KT single crystals. The experimental results indicate that the photoelectric effect plays an important role in the photovoltaic process. In addition, we also measured the photovoltages when the tilted KT wafer was illuminated through the bottom side. The photovoltaic signal polarity was reversed.

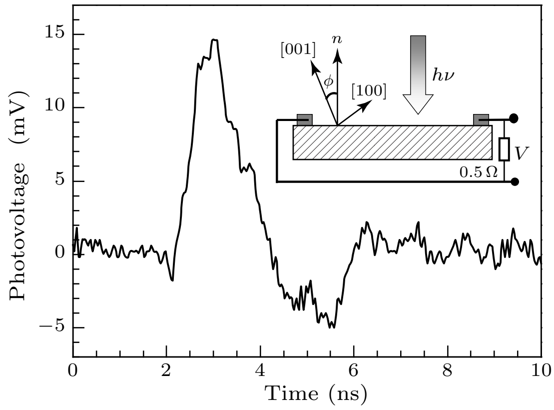

Fig. 3. Transient photovoltaic pulse of the tilted $KTaO_3$ single crystal under irradiation with a 0.5 Ω resistance connected in parallel with the single crystal. Inset: the schematic circuit of the measurement system.

The photo-induced voltage in the tilted KT wafer can be understood as follows: incident photons with energy larger than the bandgap of KT crystal were absorbed to generate electrons and holes, at the same time, a thermal gradient field was formed under the laser irradiation in c-axis orientation of KT crystal. As said in thermoelectric theory, a lateral thermoelectric field $E(x)$ was built according to the formula[17]

$$E(x) = (S_{ab} - S_c) \sin 2\phi (dT/dz)/2, \qquad (1)$$

where S_{ab} and S_c are the Seebeck coefficients of the KT crystal along the ab plane and c-axis, ϕ is the vicinal cut angle of the KT single crystal, and dT/dz denotes the temperature gradient in the direction of irradiation (perpendicular to surface of the KT single crystal). The thermoelectric field provoked the separation of electrons and holes in the lateral direction and resulted in the photovoltage between the two electrodes. From the thermoelectric formula (1), we can see that the lateral photovoltage has a sinusoidal relationship with the tilted angle ϕ. When ϕ is zero, no lateral field can be built and no photovoltage can be observed in the lateral direction, which is in agreement with our experimental result.

In conclusion, large open-circuit photovoltage sensitivity of 328 mV/mJ and the photocurrent sensitivity of 460 mA/mJ have been observed in tilted KT single crystal to UV irradiation at room temperature. Ultrafast photoelectric responses with 497.4 ps rise time and 974.6 ps FWHM have been achieved. The experimental results can be well explained by the combination of the photoelectric effect and the Seebeck effect. It is worth noting that this device is based on commercial as-supplied KT single crystal and no external bias is required, which means the advantage of a simple fabricating process and low cost. Our present work manifests that KT single crystal has attractive potential applications in visible-blind photodetectors.

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