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BARIUM TIN OXIDE ORDERED PHOTOCATHODES: FIRST MEASUREMENTS AND FUTURE PERSPECTIVES*

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Abstract

Single crystalline photocathodes with small electron effective mass are supposed to enable ultra-low emittance beams, by taking advantage of the conservation of transverse (crystal) momentum. We present a preliminary study on photoemission from epitaxial films of La-doped $BaSnO_3$ with (100) orientation. We demonstrate here the possibility of generating and characterizing electron beams by exciting photoelectrons solely from the conduction band. We report quantum efficiency and mean transverse energy meaurements as a function of photon energy from the bare and Cs-activated La-doped $BaSnO_3$ surface.

INTRODUCTION

High brightness photoemission electron sources are required for several relevant state-of-art applications such as free electron laser (FEL) and ultrafast electron diffraction (UED). The performances of these instruments (i.e. shortest wavelength for FEL and coherence length for UED) are limited by the transverse brightness of the electron beam. A high transverse brightness electron beam is characterized by a small transverse emittance, that is a measure of the phase-space volume occupied by the beam, and it is a conserved quantity. At the photocathode the normalized 2D transverse rms emittance $\varepsilon_n = \sigma_x\sqrt{MTE/m_ec^2}$, where σ_x is the rms transverse beam size, and MTE= σ_{pT}^2/m_e is the mean transverse energy, related to the rms transverse momentum spread σ_{pT} [1–3]. In order to reduce the emittance without limiting the maximum beam charge ($\propto \sigma_x^2$), σ_{pT} must be small. The initial MTE of the beam is a property of the photocathode. In particular, in the photoemission process, the transverse crystal momentum of the excited electron is conserved if the surface of the emitter is ordered. For this reason, recent work on photocathode materials has focused on single-crystalline material, in order to exploit the conservation of transverse momentum and suitable band structure to reduce the transverse momentum spread below the thermal energy value [2, 4].

$BaSnO_3$ (BSO) is a perovskite structure oxide that can be electron doped by Ba-La substitution ($Ba_{1-x}La_xSnO_3$, BLSO). BLSO is a transparent conducting oxide characterized by high room-temperature mobility and high quality thin film samples show mobile carriers down to an electron concentration of the order of 10^{18} cm^{-3}; by increasing La doping up to a few percent, extremely high carrier concentration can be achieved, above 10^{20} cm^{-3}. [5] The effective mass of the carriers in the conduction band is reported to be close to 0.2 m_e. These properties make BLSO an interesting material for the study of the photoemission process near threshold, in view of implementing the use of ordered photocathodes as bright electron sources.

EXPERIMENTAL RESULTS

Sample Growth and Characterization

The BLSO thin films were grown on buffered $GdScO_3$ or $TbScO_3$ substrates by adsorption control molecular beam epitaxy, as described in ref. [5]. This technique allows excellent control on sample stoichiometry, as evidenced by the enhanced mobility, and produces samples with smooth, unreconstructed surfaces, as demonstrated by 2D reflection high energy electron diffraction (RHEED) patterns showing 1×1 periodicity. The effective carrier density n and mobility of the samples was measured by Hall effect and resistivity versus temperature measurements; the nearly constant n as a function of temperature demonstrates that BLSO behaves as a degenerate semiconductor [5].

We investigated the photoemission from two samples: a 30 nm BLSO on 200 nm BSO buffered $GdScO_3$ substrate with $n = 0.14\times10^{21}$ cm^{-3}, corresponding to $N = 0.009$ electrons per unit cell, and a 35 nm BLSO on 200 nm BSO/$SrSnO_3$ buffered $TbScO_3$ substrate with $n = 0.07\times10^{21}$ cm^{-3}, corresponding to $N = 0.005$. The properties of the samples are not significantly affected by the particular choice of substrate/buffer [5].

Measurement Setup

The quantum efficiency (QE) and MTE measurements were performed *ex-situ* on the samples. BSO is an extremely stable material [6] and its bulk properties are unaffected by exposition to atmosphere; however the surface can be affected by contamination.

The $N = 0.009$ sample was cleaned in an ultrasonic bath of isopropyl alcohol and acetone and loaded in the measurement system described in ref. [1]. The photon source employed for the measurements is a 3.0-5.3 eV tunable UV laser radiation source generating sub-picosecond pulses at a 30MHz repetition rate. The MTE as a function of photon energy has been measured via the solenoid scan technique. [1, 7]

The $N = 0.005$ sample, after solvent cleaning, was annealed in vacuum (in 10^{-11} Torr background pressure) at 400°C for 20 min. After cooling, it has been exposed to Cs

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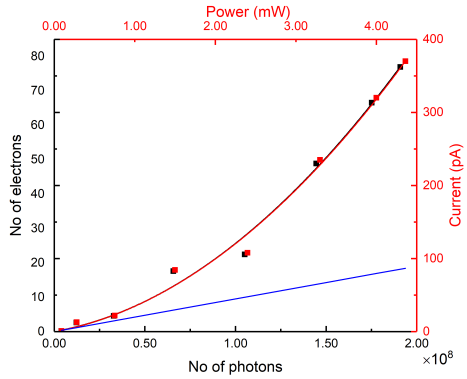

Figure 1: Black (red) symbols and axes: Electron yield (photocurrent) as a function of photon flux (light power) for the $N = 0.009$ sample with bare surface, for a photon energy of 4.75 eV. The black and red lines represent a quadratic fit to the data, the blue line is the linear (one photon) component.

flux while monitoring the photocurrent generated by illuminating the sample with a 405 nm laser diode and measured by a lock-in amplifier. The sample has been transfered, without breaking vacuum, from the preparation chamber to the transverse energy measurement system, described in ref. [7]. The MTE has been measured by voltage scans [7] ($V = 2.5 - 10$ kV). The photon source we used is a NKT SuperK laser (wavelength 405-800 nm, bandwidth 10 nm, pulse duration 6 ps at 78 MHz), and the laser spot size on the sample has been varied from 100 μm to 500 μm in order to obtain the MTE for each wavelength by a linear fit of ε_n as a function of σ_x. [3]

Results

The photoemission threshold E_{th} for the non-activated $N = 0.009$ sample is found to be at 3.95 ± 0.5 eV. This is in agreement with the measured values of the bandgap E_g, about 3.1 eV, and workfunction W, about 6.5-7.5 eV, of undoped BSO: we expect to emit electron from the conduction band, thus the required energy for escape is near to the electron affinity $E_{aff} = W - E_g$. In Fig. 1 we report the photocurrent as a function of light power at $\hbar\omega = 4.75$ eV. The quadratic behavior shows that there is a significant contribution of two photon processes, exceeding the linear one photon contribution, implying that there are significantly more occupied states involved in the two-photon case. This can be explained by considering that for $2\hbar\omega > 7$ eV, the valence states (with higher density of states than the conduction ones) can contribute to photoemission. Indeed, pure two-photon photoemission has been observed for intense laser pulses at 3.56 eV. The extracted linear component of the photocurrent QE at 4.75 eV (blue line in Fig. 1) is 10^{-7} electrons/photon.

In Fig. 2 we report the QE of the $N = 0.005$ sample as a function of photon wavelength after Cs-activation and after aging of 1 week and 12 days (in the background pressure of 6×10^{-11} Torr). The QE of the activated sample is found to be higher in comparison to the one of the non-activated

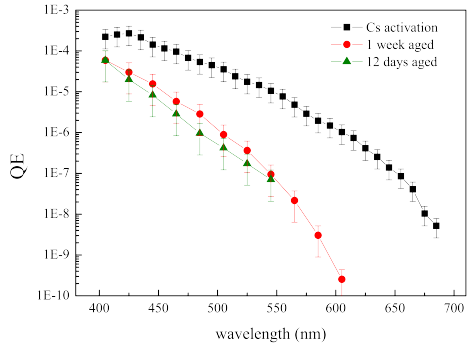

Figure 2: Quantum efficiency of the Cs-activated BLSO film ($N = 0.005$) as a function of laser wavelength. Black symbols represent the as activated sample, red and green symbols show the data corresponding to the same sample measured after 1 week and 12 days since activation respectively.

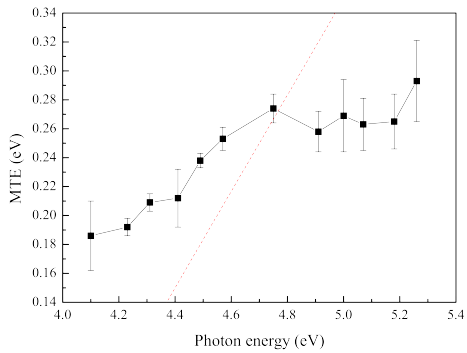

Figure 3: Black symbols: MTE as a function of photon energy for the $N = 0.009$ sample with bare surface, as obtained by solenoid scan measurements. Red line: $1/3 \cdot (\hbar\omega - E_{th})$, with $E_{th} = 3.95$ eV.

sample for comparable excess energy. For the non-aged sample $E_{th} \approx 1.8eV$, and it increases to 2.0 eV after 1 week. After 5 additional days the QE is only slightly changed. In Fig. 3 we report the MTE of the $N = 0.009$ sample with bare surface; for comparison we also report the behavior expected for disordered photocathodes, where above threshold MTE$\approx 1/3 \cdot (\hbar\omega - E_{th})$. [3] The power used to generate the electron beam has been tuned down in order to make negligible the two-photon contribution to photoemission. The MTE increases at first and then, for an excess energy larger than 0.75 eV, saturates to about 260 meV. In Fig. 4 the MTE of the Cs-activated $N = 0.005$ sample is reported for different aging. The MTE is weakly dependent on photon energy; the black and red lines show the $1/3 \cdot (\hbar\omega - E_{th})$ for $E_{th} = 1.8$ eV and 2.0 eV respectively. In this case the MTE is always lower than the disordered photocathode case.

DISCUSSION

The measured E_{th} for the $N = 0.001$ sample, close to E_{aff} of BLSO confirms that we are exciting electrons from the conduction band of the material, while electrons from the

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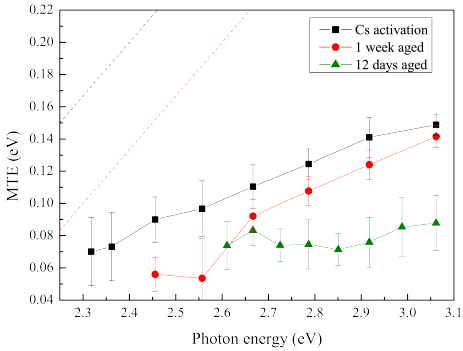

Figure 4: Symbols: MTE as a function of photon energy for the $N = 0.005$. Lines: one third of excess energy. Black: after Cs-activation; red: after 1 week since activation; green: after 12 days since activation.

valence band contribute via two-photon process at high laser power. The QE of the bare surface BLSO sample is found to be low ($\sim 10^{-7}$ for an excess energy of 0.8 eV), being 3 order of magnitude lower than the one of Cu for similar excess energy ($\sim 10^{-4}$). We note that the carrier density in the conduction band of BLSO is also 3 order of magnitude lower than the Cu one. The Cs activation, performed on the $N = 0.005$ sample, decreases the photoemission threshold of about 2 eV, and it greatly enhances the QE for comparable excess energy (about 3 order of magnitudes), despite the lower carrier density. The QE of ordered photocathodes has been modeled for single crystal metals [2, 8], and depends both on the initial and final states of photoemission, the latter being obviously different for the bare and Cs-activated samples. Furthermore the effective carrier density and band structure close to the surface of BLSO can dramatically differ from the bulk one (band bending, presence of surface states), as revealed by the ARPES measurements reported in ref. [9], and these surface properties are likely to be affected by the Cs layer.

In Fig. 5 we show ARPES measurements on a sample grown in the same conditions of the ones under investigation [9]. Near the Fermi level, a metallic state is visible, with a width that is compatible with an effective mass of 0.2 m_e. The valence band structure (shown in the right panel of Fig.5) can be described by *ab initio* calculations based on the generalized gradient approximation with spin-orbit coupling [9]. Based on the calculated band structure we estimated the MTE in the hypothesis of transverse momentum conservation, using the approach reported in ref. [1] to model the photoemission from crystalline metal photocathodes. The result is reported in Fig.6 as a function of excess energy, together with our experimental data on samples with disordered surface. The large difference in MTE for the N=0.009 and N=0.005 samples can be due both to different doping and different surface conditions, as discussed for the QE.

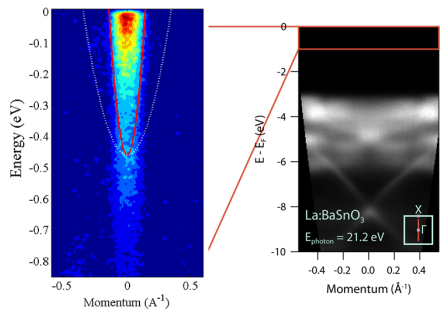

Figure 5: ARPES measurement on a BLSO sample with $N \approx 0.01$. The right panel shows the full band structures measured along the $\Gamma - X$ direction of the Brillouin zone, while the left panel shows the states close to the Fermi level. The red line represents a parabolic dispersion with $m_{eff} = 0.2m_e$, the white one for $m_{eff} = 1m_e$.

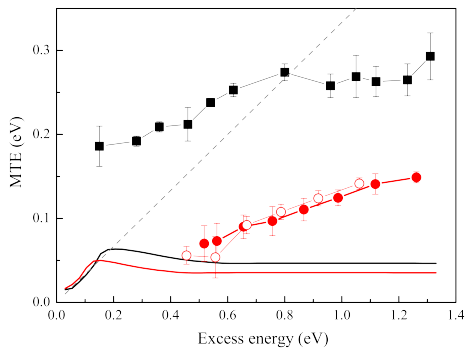

Figure 6: Solid lines: calculated MTE from band structure calculation for $N = 0.01$ (black) and $N = 0.005$ (red) as a function of excess energy. Dashed line: $1/3(\hbar\omega - E_{th}$ behavior. Symbols: MTE of the $N = 0.009$ sample (black) and of the $N = 0.005$ Cs-activated sample (red solid: after activation, red hollow: 1 week aged).

CONCLUSIONS

We performed tests on an innovative material for photocathode applications. BLSO is a promising materials thanks to its low effective mass in the conduction band and high carrier density. We tested the feasibility of experiments in which electrons are excited only from the conduction band, that is typically characterized by states with lower effective mass than the valence band. In particular, the Cs-activated photocathode outdoes metal photocathodes such as Cu, showing similar QE at lower photon energy and a slower increase of MTE with excess energy. Future experiments will aim to study the doping dependence of QE and MTE, and the conservation of transverse momentum in samples in which the ordered *as-grown* surface is preserved.

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