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THE ROLE OF ELECTRON-PHONON SCATTERING IN TRANSVERSE MOMENTUM CONSERVATION IN PbTe(111) PHOTOCATHODES

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Abstract

The state of the art in creating high quality electron beams for particle accelerator applications and next generation ultrafast electron diffraction and microscopy involves laser-generated photoemission. A high quality beam requires that electrons emerge from the surface with low mean transverse energy (MTE). Recent density-functional theory calculations by T. Li and W. A. S. [arXiv:1704.00194v1 [physics.acc-ph] (2017)] suggest that PbTe(111) will produce low-MTE photoelectrons due to the low effective electron mass associated with its electronic band structure. Based on this, we measured the distribution of photoelectrons from PbTe(111) and found the MTE to be about 20x larger than expected. To explain the apparent lack of transverse momentum conservation, we carried out many-body photoemission calculations including electron-phonon scattering. Our results are in far better agreement with the experiment, underscoring the importance of electron-phonon scattering in photoemission from PbTe(111), and suggest that cooling could mitigate the phonon effects on the MTE for this material.

INTRODUCTION

Current developments in high-brightness electron beams utilize laser-generated photoemission and photocathode materials with high quantum efficiency and low mean transverse energy (MTE) [1], where MTE is proportional to the square of rms transverse momentum of the emitted photoelectrons. Recently PbTe(111) was predicted to produce very low MTE of ≤ 15 meV due to the small effective mass associated with the valence band maximum of the electronic band structure, as calculated using density-functional theory [2] (Fig. 1, gray curve). However, this assumes the electrons instantly transition to vacuum from the valence band maximum without accounting for their propagation through the photocathode material. Moreover, this predicted MTE is 10-20x smaller than the observed MTE in our recent experiments at room temperature (Fig. 1, blue curve). To better explain this observation, we next perform *ab initio*, first-principles MTE calculations including the propagation of the photoelectrons through the material at room temperature.

The red curve in Fig. 1 shows the *ab initio* result of accounting for the propagation of photoelectrons through the material. With the propagation accounted, the magnitude of the calculated MTE is now in far better agreement with the experiment. However, the overall shape of the calculated MTE is still somewhat different: there is a sharp threshold around 4.8 eV without a similar feature in the experiment. On the other hand, the prediction does show a dip around 5.0-5.5 eV, which is similar to a dip seen in the experiment centered about 0.5 eV lower in photon energy.

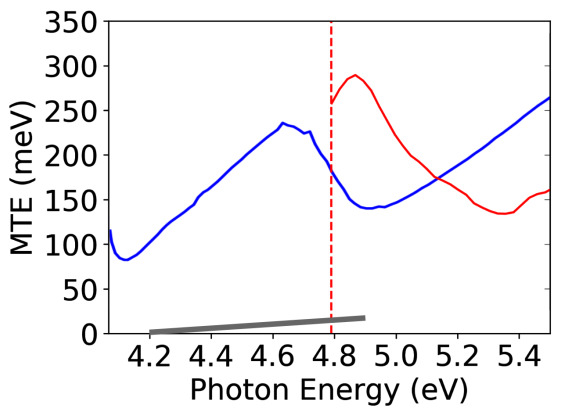

Figure 1: Electron mean transverse energy (MTE) versus photon energy. Initial prediction neglecting propagation through the material [2] (gray curve), experimental result at room temperature (blue curve), *ab initio* result at room temperature including propagation through the material (red curve), calculated emission threshold (vertical dashed red line), below which there are no excited electrons.

Because of the discrepancies between our calculated and experimental MTEs, we next include electron-phonon scattering (scattering between electrons and crystal vibrations) in our *ab initio* calculations. There are two reasons to suspect this may be an important effect. First, the experiment was done at room temperature, where we would expect non-negligible phonon activity [3]. Second, unlike photons, whose momenta are ~1000x smaller than typical electron crystal momenta (and so the effect of photon excitations on the electrons' momenta is negligible), phonons carry significant momenta, and hence can significantly affect the MTE of the photoelectrons.

MANY-BODY APPROACH

In this work, we consider only photoelectrons that originate from the bulk of the photocathode material, as opposed to those from the surface, because of the greater number of the bulk electrons. Figure 2 shows the processes that such bulk electrons undergo. During emission, an entering photon excites an electron from the valence band into a bulk conduction band, either directly (Fig. 2.1a) or mediated by a phonon (Fig. 2.1b). During the phonon-mediated excita-

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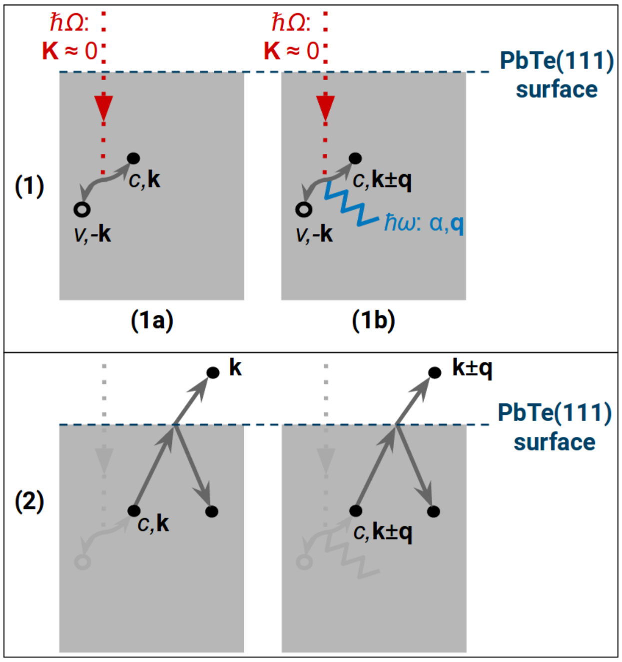

Figure 2: Many-body photoemission model. Direct excitation into a crystal state (1a), phonon-mediated excitation into a crystal state (1b), emission from excited crystal state into vacuum (2). Incoming photon of energy $\hbar\Omega$ and momentum $\mathbf{K}$ (dotted red arrow), absorbed/emitted phonon of energy $\hbar\omega$ and momentum $\mathbf{q}$ from phonon branch α (jagged blue line), resulting hole in valence band v with momentum $-\mathbf{k}$ (open circle), resulting electron in conduction band c with momentum $\mathbf{k}$ or $\mathbf{k} \pm \mathbf{q}$ (filled circle), coherent outgoing scattering state including reflection and transmission at crystal surface (straight gray arrows).

tion, where the phonon can be absorbed or emitted by the electron, the photon and phonon scatter coherently with the electron. The resulting state of the electron is a coherent outgoing scattering state, which includes incoming, reflected, and transmitted components (Fig. 2.2). The incoming part is inside the material going towards the PbTe(111) surface, the transmitted part is in vacuum going away from the surface, and the reflected part in the material going away from the surface. In this work, for simplicity the transmission coefficient is assumed to be 100%; future refinements of our model would be to employ surface calculations to calculate transmission and reflection coefficients.

We calculate the transition rates of both direct and phonon-mediated excitations using JDFTx, a plane-wave density-functional theory software with a capability to calculate such transition rates [4]. Then, we calculate the MTE of the transmitted electrons as a function of photon energy.

RESULTS AND DISCUSSION

Figure 3 shows the calculated MTEs at room temperature both with and without electron-phonon scattering. The blue curve labeled "total" has the contributions from the direct excitations and the phonon-mediated excitations, while the red curve labeled "no phonon" only has the contributions from the direct excitations. (This red curve is the same as the red curve in Fig. 1.) We see that our calculated MTE including the electron-phonon scattering (Fig. 3, blue curve) has much better agreement with our experimental result (Fig. 1, blue curve) than our calculated MTE without the electron-phonon scattering (Fig. 1 or 3, red curve). In addition to having approximately the same values as the experimental MTE, the calculated MTE with the electron-phonon scattering also has features similar to the ones observed in the experimental MTE: for the photon energy ranges shown in Figs. 1 and 3, both MTE curves start with a tiny dip, then increase, and then have a large dip. Finally, we note that there is a small, constant horizontal shift of ~0.4 eV between the features seen in the experimental MTE and the calculated MTE due to issues related to the band gap from density-functional theory and the actual work function of our sample.

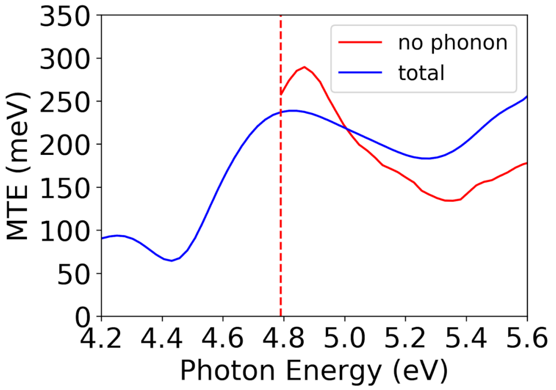

Figure 3: Calculated MTE at room temperature. Total MTE (blue), direct-only MTE (red, identical to the red curve in Fig. 1), direct threshold (vertical dashed red line), below which there are no electrons from the direct excitations. The total MTE now agrees much better with the experiment (Fig. 1, blue curve) than the direct-only MTE, underscoring the importance of electron-phonon scattering effects on the MTE for PbTe(111).

Because the electron-phonon scattering significantly affects the MTE at room temperature for PbTe(111), we now investigate whether we can control the phonon effects on the MTE in this material by lowering the temperature. Accordingly, we next recalculate the MTEs at cryogenic temperature, where there are almost no phonon absorptions (because there are almost no phonons in the environment), but there are still phonon emissions [3]. Figure 4 shows the calculated MTE at ~30 K. Again, the blue curve has the contributions from the direct excitations and the phonon-mediated excitations, while the red curve only has the contributions from the direct excitations. At this temperature, the direct-only ("no phonon") MTE in red is practically identical to the total MTE in blue above the direct threshold, which suggests that the number of photoelectrons from the phonon-mediated excitations only is very small. This is further verified by cal-

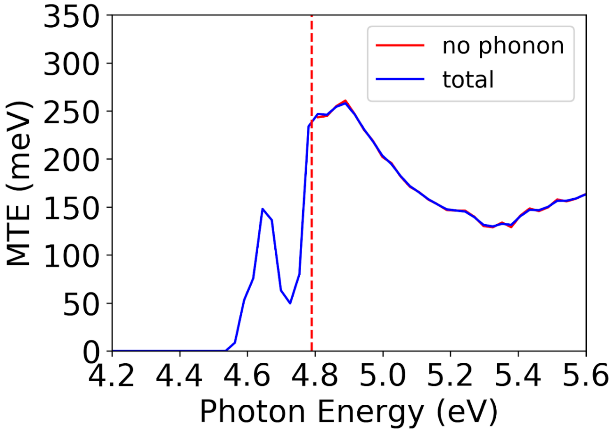

Figure 4: Calculated MTE at ∼30 K. Total MTE (blue), direct-only MTE (red), direct threshold (vertical dashed red line), below which there are no electrons from the direct excitations. Above the direct threshold, the total MTE is identical to the direct-only MTE.

culating the photocurrent ratio, the number of these electrons divided by the total number of photoelectrons just above the direct threshold at 4.8 eV. The photocurrent ratio at ∼30 K is only $\sim 10^{-6}$ over the entire range of the photon energy in the figure; in contrast, the photocurrent ratio at room temperature is ∼0.1 over the same range of photon energy. Below the direct threshold, the features in the total MTE in Fig. 4 are caused by a tiny number of phonon-mediated photoelectrons, without any contributions from direct excitations. Because of the low numbers, these features may not be detected experimentally. Hence, for this material, the MTE at ∼30 K is practically identical to the direct-only MTE, with a magnitude similar to the experimental MTE at room temperature.

Since the number of phonon-mediated photoelectrons at ∼30 K is very small below the direct threshold at 4.8 eV, the contribution to the MTE from these phonon-mediated photoelectrons may actually become smaller than the surface contribution that we ignore in this work. If this is the case, then we would expect to get a total MTE curve at ∼30 K that behaves similarly to the MTE from the previous work in ref. [2] (Fig. 1, gray curve) below 4.8 eV, and to the direct-only MTE in Fig. 4 above 4.8 eV. This means that it is possible to acquire very low MTE experimentally in PbTe(111) by cooling it down to ∼30 K and using appropriate photon energies. To explore this possibility, future *ab initio* calculations would include photoelectrons that instantly transition to vacuum.

FUTURE WORK

To improve the photoemission model, future work would calculate the transmission and reflection coefficients of the outgoing scattering states of the photoelectrons, which this work assumes to be 100% and 0, respectively. Future theoretical work would also include the surface contributions to verify whether the very low MTE predicted in ref. [2] should be obtainable at ∼30 K. Finally, we recommend low-temperature experiments on PbTe(111) which may finally yield the originally predicted low MTE.

CONCLUSION

Prior work had initially predicted PbTe(111) to produce a very small MTE of ≤ 15 meV, due to the small effective mass associated with the valence band maximum. However, the MTE observed in our experiments at room temperature is 10-20x larger. By accounting for the propagation of the photoelectrons through the photocathode material and including electron-phonon scattering, *ab initio* MTE calculations can reproduce the experimental MTE well at room temperature. Our *ab initio* calculations also show that the electron-phonon scattering effects on the MTE become negligible at ∼30 K, with photoemission dominated by direct excitations and the MTE still having a similar magnitude to the experimental MTE at room temperature.

This work gives deeper understanding into important processes underlying photoemission and will improve future computational searches for better photocathode materials for high-brightness beam applications.

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