CEMS Topical Meeting Online

Emergent Oxide Interface Phenomena meets Novel Growth Techniques

Friday, July 16th 2021

10:00–17:30

Abstracts

Transport properties of the correlated metal $SrVO_3$ in the ultraclean limit

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Progress in thin film synthesis resulting in ever cleaner materials enables experimental access to the intrinsic properties of materials. While this approach has been perfected in the case of established semiconductor material systems using molecular beam epitaxy and resulted in the discovery of new physical phenomena, it is still in its infancy for more complex material system with a more unfavourable growth kinetics, such as perovskite oxides, in which carriers experience sizeable electron-electron coupling.

In this talk I will present efforts to grow ever cleaner perovskite oxide systems exemplifying this on the correlated metal $SrVO_3$. Changes in the magnetotransport properties as a function of film orientation and degree of disorder will be discussed. Transport peculiarities observed at high magnetic fields in the ultraclean limit points towards a pronounced scattering time anisotropy that has remained elusive in systems with pronounced defect density.
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Superconductivity at $LaAlO_3/KTaO_3$ interfaces

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Abstract:

Very recently superconductivity with a transition temperature (T_c) up to 2.2 K was discovered at $KTaO_3$-based interfaces [1-3]. In this talk we will present our progresses in this newly discovered superconducting oxide interface. We confirmed that the superconductivity depends strongly on the orientation of the interface and discovered a T_c ~0.9 K superconductivity at (110)-orientated interface [2]. We controlled the superconductivity of (111)-orientated interface with electric field, achieving a superconductor-to-insulator quantum phase transition [3]. Strikingly, we found that the tuning effect was achieved by change in the spatial profile of the carriers in the interface and hence, effective disorder, rather than carrier density. A few interesting quantum phenomena (i.e. quantum metal) that are related to the interplay between disorders and two-dimensional superconductivity were observed. We also observed a critical thickness of $LaAlO_3$, ~3 nm [4]. The interface is conducting (insulating) when $LaAlO_3$ is thicker (thinner) than this thickness. Our further study on this critical thickness suggests that the determinant mechanism of the interface conduction is the electron transfer from oxygen vacancies in the $LaAlO_3$ film to the $KTaO_3$ substrate.

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From Molecular Beam Epitaxy to high magnetic field quantum oscillations and charge ordering

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ABSTRACT: Cuprate superconductors present a major challenge in condensed matter physics not only due to their electron correlations but also due to their complex crystal structure. Complex crystal structures, i.e. various cations at various lattice positions, demand for the utmost caretaking when being synthesized. In particular, Molecular Beam Epitaxy is well known to be the foremost versatile tool and technique that allows for the synthesis of such materials without the necessity to compromise on impurity phases. First, I introduce our custom designed Molecular Beam Epitaxy equipment which is empowered by e-guns, not effusion cells, and controlled by electron impact emission spectroscopy. After presenting several material systems I present high magnetic field quantum oscillation data on films synthesized by our Molecular Beam Epitaxy systems. Furthermore, I show results on the charge-ordering mechanism using resonant elastic x-ray scattering. These data support the notion that the appearance of superconductivity in cuprate superconductors is tied to the coordination of the copper in these systems. Using molecular beam epitaxy also infinite-layer cuprates, i.e., a thermodynamically unstable phase, can be synthesized. Moreover, superconducting thin films of the low-temperature superconductor Sr_2RuO_4 were grown by molecular beam epitaxy. The highest superconducting transition temperature was 1.4 K and those films showed a residual-resistivity ratio (RRR) of 114. High magnetic field (up to 24 T) magnetoresistance measurements above 0.5 K show Shubnikov-de Haas oscillations thus allowing us to study the intrinsic electronic structure of this material along with the influence of in-plane epitaxial strain in comparison to bulk samples. I will conclude with an outlook that the top of the superconducting transition temperature in cuprate superconductors has not been reached, yet.

Thermal Laser Epitaxy: New Opportunities for Oxide Epitaxy

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ABSTRACT. In thermal laser epitaxy, both the substrate and the individual evaporation sources are heated by high-power CW lasers. This method combines the advantages of MBE and PLD, allowing the efficient thermal evaporation and epitaxial deposition of any combination of elements from the periodic table.
We demonstrate and discuss the deposition of elemental layers from the majority of the elements in the periodic table, including W, Ta, C, Ir and Si, at 1 Å/s with laser powers less than 500 W, the growth of binary oxides from the elements in ozone/oxygen background pressures as high as 10^{-2} hPa, as well as the epitaxial growth of refractory metals on sapphire at substrate temperatures well above 1000 °C with chamber pressures during growth below 2×10^{-10} hPa.

Keywords: thermal laser epitaxy, laser evaporation, oxide epitaxy, adsorption-limited growth

In oxide epitaxy, MBE is generally regarded as the method producing the highest quality heterostructures. To address new opportunities in adsorption-limited deposition, however, higher substrate and source temperatures are required, together with higher active oxygen background pressures. Following pioneering work in the 1960s and 1980s[1], we explore continuous wave (CW) laser heating for both the substrate and individual elemental sources to overcome these limitations[2]. In such a system, shown in Fig. 1, a long wavelength infrared laser is used to directly heat oxide substrates that are transparent to the radiation of standard heaters, allowing ultrahigh substrate temperatures above 2000 °C. Most elements in the periodic table are metals with melting points at which the vapor pressure is in or even above the range of fluxes required for deposition. Under these conditions, heating the central region of a source target by a laser produces temperature gradients large enough so that the target does not melt as a whole, as demonstrated for a 12 mm diameter Si source in Fig. 2. All these materials can therefore be evaporated without crucibles, by merely suspending them between support points. Using a working distance of 60 mm, we achieve growth rates of more than 1 Å/s. We have experimentally verified such crucible-free evaporation already for a majority of the non-radioactive elements in the periodic table (Fig. 3)[3].

We obtain chamber pressures of 6×10^{-11} hPa at substrate temperatures of 1600 °C and 2×10^{-10} hPa during the deposition of Ru at a rate of 0.2 Å/s (Figs. 4/5). The source targets are transferable in the same way as the substrates, allowing a flexible operation with different source materials in subsequent runs. The laser heating is energy efficient, with less than 500 W sufficient for all elements in the entire periodic table for deposition on 2” substrates (Fig. 6). We have achieved reliable deposition of oxides by evaporation in up to 10^{-2} hPa ozone/oxygen, such as the oxide layer shown in Fig. 7. As there are no hot filaments in the vacuum, both highly corrosive background gases and ultra-pure operation are possible. The small source targets enable short working distances, allowing an efficient use of the source material, e.g. when working with isotope pure materials. The same scaling arguments as in MBE apply, permitting a straightforward upscaling of the epitaxy process to large substrate sizes. The process is agile, the substrate and the sources can be ramped at extreme rates due to their small heat capacity with direct heating and the absence of crucibles and the associated thermal expansion mismatch problems, thereby allowing high throughput volumes. And finally, the process chamber can be very small and simple, allowing both long uptimes and fast and easy rebuild or replacement in case of contamination or damage.

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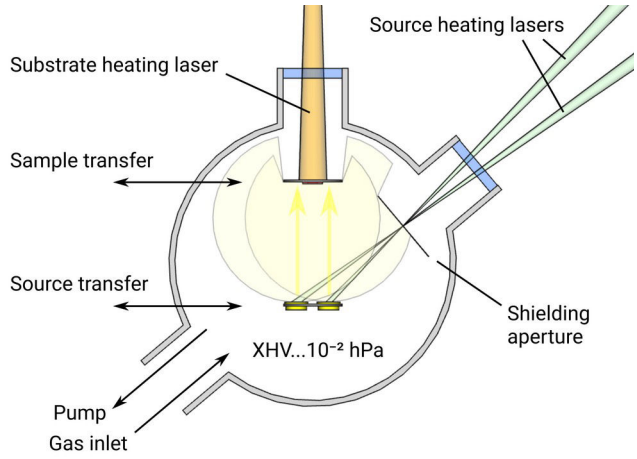

Fig. 1: Schematic representation of a TLE system. Both the substrate and the sources are heated by CW lasers, both can be transferred in and out of the growth chamber.

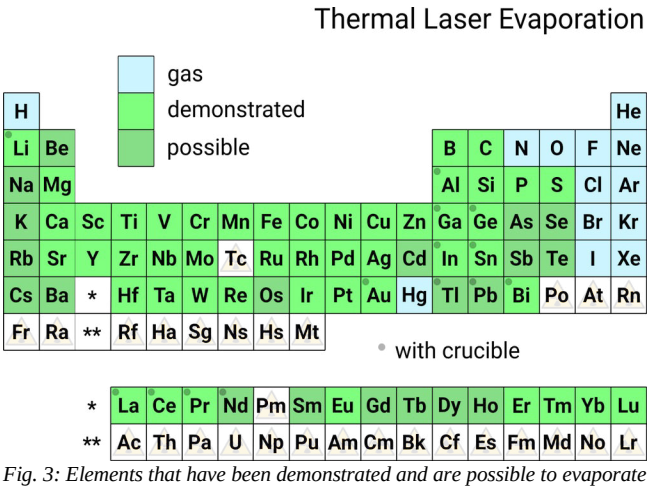

Fig. 3: Elements that have been demonstrated and are possible to evaporate or sublimate by thermal laser evaporation.

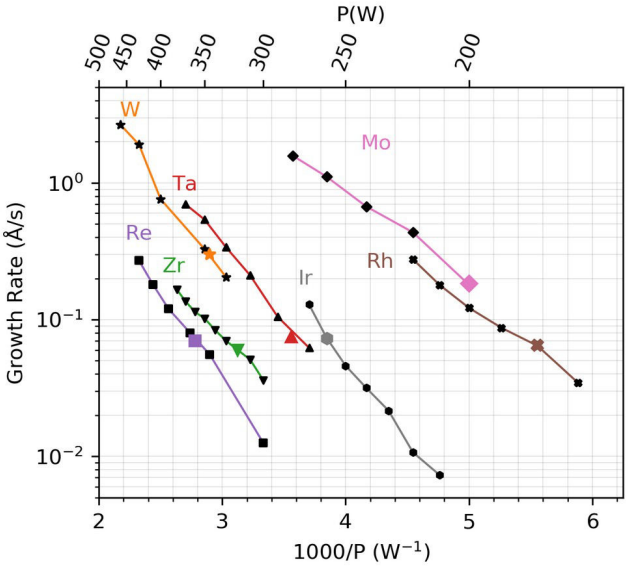

Fig. 6: Power vs. growth rate for selected elemental metals. The melting points are marked by colored symbols.

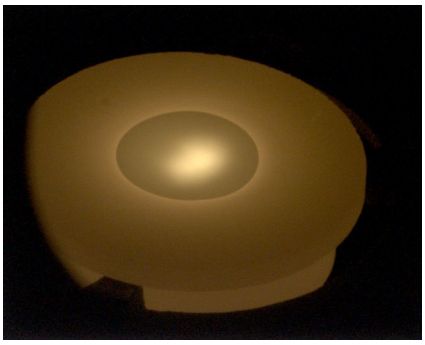

Fig. 2: Melt pool on a 12 mm diameter Si source

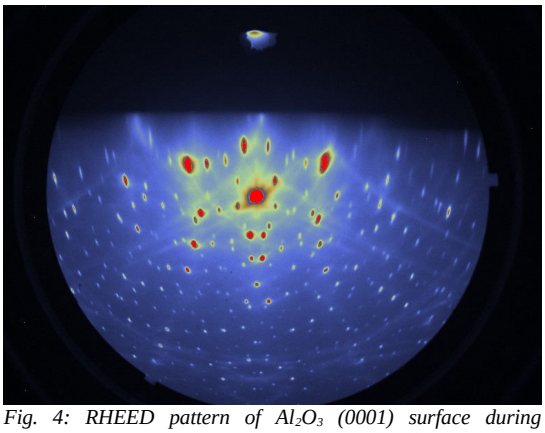

Fig. 4: RHEED pattern of Al_2O_3 (0001) surface during substrate preparation at 1600 °C with 7.5x10^{-11} hPa chamber pressure

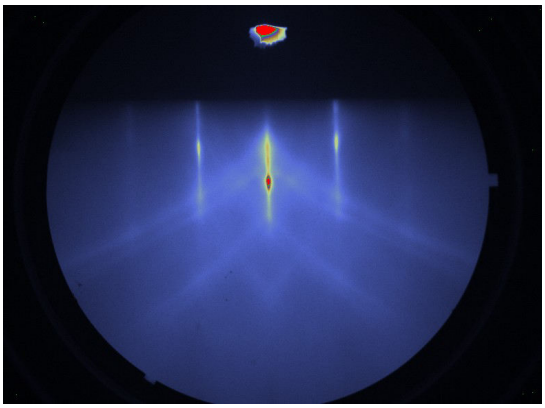

Fig. 5: Epitaxial Ru layer grown on the sapphire surface shown in Fig. 5 at 1000 °C with 2x10^{-10} hPa chamber pressure.

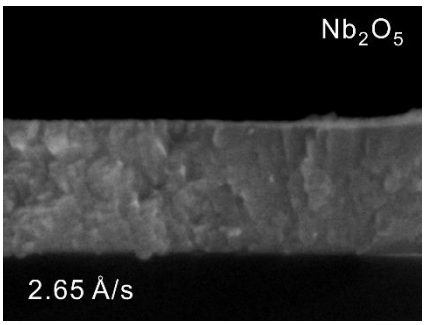

Fig. 7: Oxide layer grown by laser evaporating Nb metal in an oxygen/ozone atmosphere of 10^{-3} hPa on an unheated Si substrate.

Electronic structure of buried oxide interfaces explored by soft-X-ray ARPES

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Pushing ARPES to soft-X-ray photon energies enhances its **k**-resolving capabilities with larger probing depth and resonant photoexcitation allowing chemical specificity. These spectroscopic abilities make soft-X-ray ARPES ideal for 3D materials, buried interfaces and impurity systems [1]. I will focus on its applications to oxides and their interfaces, in particular in the context of *e-ph* interaction.

Ce-doped CMO. – The applications to 3D systems are based on sharp definition of out-of-plane momentum resulting from the enhanced photoelectron delocalization. We show the development of a Fermi surface in CMO upon Ce doping, where 3D mobile electrons coexist with much heavier quasi-2D strongly coupled polarons. This dichotomy results from different filling of the energy bands compared to the phonon energy going beyond the Migdal approximation [2].

LAO/STO. – Resonant soft-X-ray ARPES at the Ti *L*-edge exposes electron dispersions of the d_{xy}-, d_{yz}- and d_{xz}-derived subbands of the mobile electrons in the interface quantum well buried below the LAO layer. The peak-dip-hump spectral function of these electrons manifests polaronic nature of the interface charge carriers, which fundamentally limits their temperature-dependent mobility [3].

γ-Al_2O_3/*STO*. – Owing to the symmetry breaking between the spinel γ-Al_2O_3 and perovskite STO, the d_{xy}-states in STO shift above the d_{xz}/d_{yz} ones and depopulate. The resulting electron-density shift away from the defect-rich interface and weakening of the *e-ph* interaction contribute to the giant mobility boost compared to LAO/STO observed in transport experiments [4].

Finally, I mention the capabilities of soft-X-ray ARPES to other classes of materials such as 3D topological materials, buried metal/semiconductor interfaces, magnetic impurities, etc. In the instrumental perspective, I introduce iMott the multichannel spin detector, whose giant efficiency boost will allow access to spin textures of buried heterostructure and impurities.

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Towards spin-orbit coupling effects in strong electron correlation systems

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Oxide compounds constitute a wide material class featuring a wealth of quantum phenomena and functionalities governed by the interplay of various degrees of freedom and electron correlation effects [1]. Particularly, materials with a relativistic spin-orbit coupling interaction (SOI) are nowadays in the spotlight. Large SOI affects both electronic and magnetic structure and is anticipated to lead to the emergence of novel electronic quantum phases laying the ground to future technologies. Thereby, the control of materials and their interfaces is inevitable to unravel the material fundamentals.

In this talk, I will discuss a surprising observation of the SOI in a high mobility two-dimensional electron system of ZnO heterostructures [2]. It constitutes perhaps one of the tantalizing aspects of reach spin physics found in ZnO [3, 4]. Striking here is that the SOI appears in the regime of strong Coulomb interaction. In fact, the SOI can profoundly compete against Coulomb interaction, which could lead to the emergence of unconventional electronic and spin phases [5]. Thus, the SOI observation unravels another facet of emerging phenomena in ZnO and marks ZnO as an appealing platform enabling to explore the interplay between correlation effects and the spin-orbit coupling, one of the outstanding problems in modern solid state physics. Furthermore, I will present the recent activities on developing heterostructures based on oxide tantalate material platform, in which both SOI and electron correlation effects are stronger pronounced than in ZnO.

References:

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