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Bosonic Collective Modes in Quantum Materials Studied with meV-resolved, Momentum-resolved EELS

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A crowning achievement of twentieth-century condensed matter physics is the Fermi Liquid paradigm, which reduces the incredibly complicated problem of $>10^{23}$ interacting electrons to a simple single-electron picture. However, a growing class of materials has been found which dramatically violate Fermi Liquid theory, requiring quantum interactions to be carefully accounted for and thus earning the name "quantum materials" [1]. Quantum materials exhibit powerful and exotic phases, including superconductivity, multiferroicity, metal-insulator transitions, topological phases, etc., and understanding their driving mechanisms is a holy grail of modern physics [1,2].

Currently, robust tools have been developed to understand quantum materials through the band-structure of their fermions (e.g. ARPES, SI-STM). However, more important than the band-structure itself is understanding how the fermions interact with one another in quantum materials (e.g. via bosonic collective modes), which is exactly encoded in the electron energy loss function $\varepsilon^{-1}(\mathbf{q},\omega)$. EELS is the most natural way to measure the loss function, provided it has the energy and momentum resolutions relevant to quantum materials, namely ≤10 meV and ~0.03 Å^{-1}. However, it is critical to measure the loss function in momentum-space rather than real-space, as the interactions are non-local and wave-like at these low-energies, just like the wave-like valence electrons themselves. At these scales, a wide variety of excitations are predicted to occur, such as electromagnons in multiferroics, topological plasmons in Weyl semimetals, and Cooper-pair Josephson modes in layered superconductors [2,3].

Here we summarize two of the first Momentum-resolved EELS (M-EELS) studies on quantum materials done with the first truly momentum-resolved reflection HR-EELS instrument [4]. Our instrument achieves 4 meV and 0.03 Å^{-1} resolutions at a 50eV primary beam energy, and functions within a double mu-metal shielded ultra-high vacuum system. The design includes a 5-axis cryostat, allowing for measurements from 20K to 400K, giving the entire home-built M-EELS system a cost of ~$450K.

First, we discuss M-EELS studies performed on $TiSe_2$ [5], a van der Waals 2D material which undergoes a charge density wave (CDW) transition at 220K. The origin of this transition has long been debated either to be a lattice instability, or a more exotic instability towards exciton superfluidity (i.e. "excitonium"). The former is driven solely by a soft phonon, while the latter requires a soft plasmon. Figure 2a-b shows M-EELS data on $TiSe_2$ as a function of momentum and temperature. At 300K, $TiSe_2$ behaves as an ordinary metal, with a plasmon dispersing to higher energy and damping away; however, at the CDW transition the plasmon dispersion dramatically reverses direction and goes to zero energy at the CDW wavevector, thus confirming $TiSe_2$ as the first example of excitonium in nature.

Second, we discuss M-EELS results on the "strange metal" phase of $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi-2212) [6], a high-temperature superconductor. The strange metal is a phase found in numerous material families which violates nearly all the expected behaviours of metals, and, among other properties, exhibits dissipation at the quantum limit of $\hbar/k_bT$ [7]. Figure 2c-d show the M-EELS spectra of the strange metal

Bi-2212, which exhibits a flat continuum from 0.1 to 1 eV that is momentum-independent, in stark contrast to the dispersing plasmons of ordinary metals. The flat energy-dependence indicates that electrons are never fully screened in strange metals, because the Coulomb interaction is as strong at 0.1 eV as at the nominal plasma frequency of 1 eV. The momentum-independence of this continuum means that charge disturbances are unable to propagate in space and that strange metals exhibit a new type of charge dynamics that are local to such a degree that the space and time axes are decoupled [6].

While our reflection M-EELS system is best suited for the study of surfaces and 2D quantum materials, vast opportunities remain for the study of bulk quantum materials using meV-resolution (S)TEM-EELS. To enter into the field of quantum materials, the grand challenge is to adapt a TEM-based EELS system to achieve (1) meV-resolution, (2) high momentum/angular resolutions of ~0.03 $Å^{-1}$ ($2\pi/100$ $Å^{-1}$ or ~0.2 mrad at 100 keV), and (3) low-temperature sample stages at LHe or LN_2 temperatures. At this meeting we will discuss some routes towards achieving these milestones and will show that they are well within modern capabilities provided momentum-resolution is made a priority [9].

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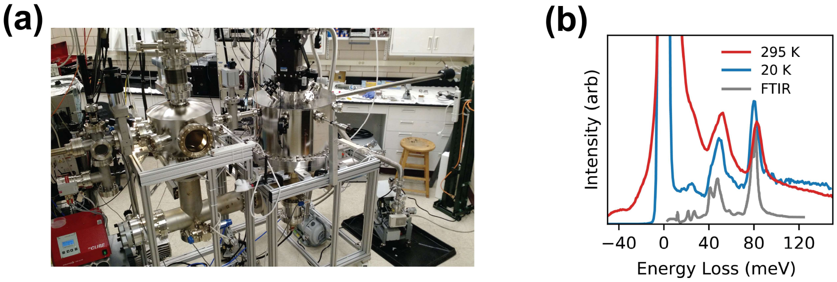

Figure 1. (a) Complete, functioning M-EELS instrument currently housed at UIUC. (b) Sample M-EELS spectra at q=0 showing phonons in Bi-2212, matching well with the FTIR loss function from [8].

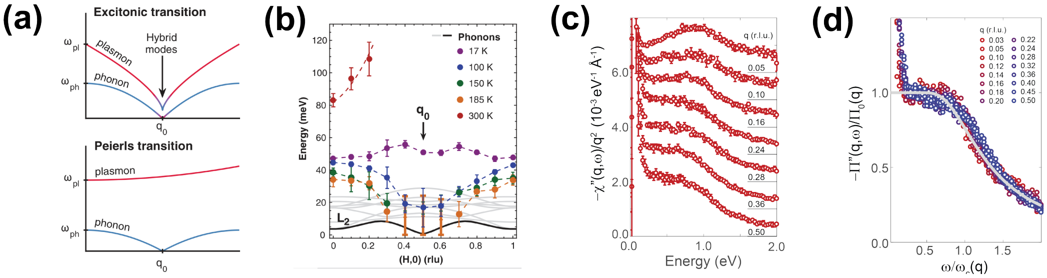

Figure 2. (a) Comparison of an excitonic and Peierls CDW transition, with a soft plasmon only occurring in the excitonic case. (b) M-EELS spectra of $TiSe_2$ showing the plasmon dispersion for a series of temperatures, clearly showing a soft plasmon [5]. (c) M-EELS spectra of Bi-2212, showing a momentum-independent continuum from 0.1 to 1 eV, at odds with ordinary plasmons [6]. (d) Collapse of the spectra in panel (c) emphasizing the universal and momentum-independent behavior.

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