

Out-of-equilibrium electron dynamics of the quasi-two dimensional correlated metal BaNiS₂

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Abstract

We present a time-resolved Angle Resolved Photoelectron Spectroscopy (tr-ARPES) study on BaNiS₂, which is the metallic precursor of the metal to insulator transition (MIT) in the quasi-2D compound BaCo_{1-x}Ni_xS₂ with $x_{cr} \sim 0.22$. This MIT is of particular interest because it is merely driven by electron-electron correlation [1] and is associated with a competition between an insulating antiferromagnetic phase and an unconventional paramagnetic metallic one [2] similar to High-Tc oxide superconductors [3], such as cuprates.

A recent theoretical and experimental work has confirmed the multi-orbital band structure of BaNiS₂, identified several Dirac-like band dispersions in the vicinity of the Fermi level and a very large Rashba splitting of the bands at X resulting from the spin-orbit interaction coupled to a strong crystal field [4]. This is particularly surprising, because it reveals a strong spin-orbit interaction even in the absence of heavy elements. We have successfully measured the full band structure of BaNiS₂ by means of ARPES, fig 1(b). In addition, a series of tr-ARPES experiments taken on the FemtoARPES setup [5] with 1.5 eV as pump energy and 6.3 eV as probe pulse allowed us to observe the unoccupied electronic states, fig 1 (c,b), and to study the out-of-equilibrium dynamics of the photoexcited carriers close to the Fermi level in a k-resolved fashion. The excess electrons then relax back to the valence band with a decay time of about 600fs thanks to their strong interband and intraband scattering processes with the electron-phonon coupling playing the key role, fig 1(e,f). It should also be pointed out that the time evolution of the excess populations is very well balanced for electrons and holes at all time delays.

However, due to some geometric restrictions on the FemtoARPES setup, we cannot study the electron dynamics through the entire k-space by 6.3 eV photon energy. To overcome this issue, we will plan to perform a detailed time-resolved study of BaNiS₂ at Attolab using XUV source.

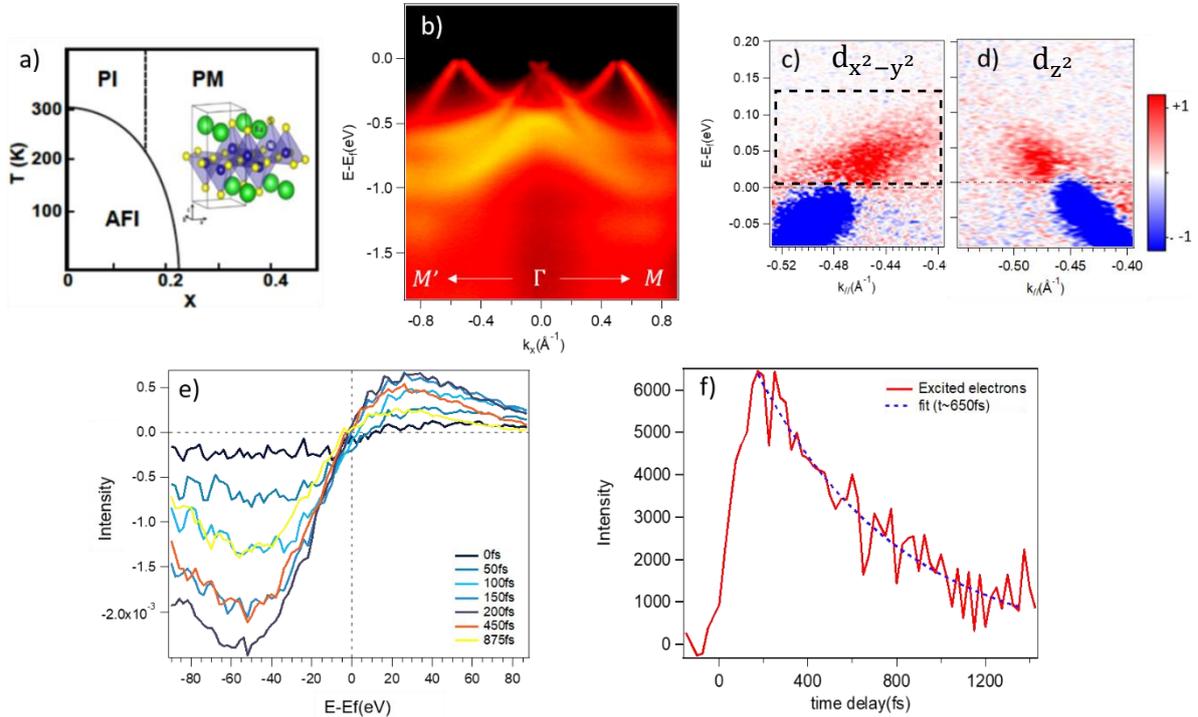


Fig 1. a) The phase diagram of BaCo_{1-x}Ni_xS₂. b) The band dispersion of BaNiS₂ by 70 eV photon energy. c,d) Excess electron (red) and hole (blue) populations are detected from the difference tr-ARPES images obtained before and after photoexcitation at 200fs for the bands along ΓM direction. e) The integral of intensity vs. electron binding energy for the d_{z^2} band for different time-delays. f) Ultrafast time evolution of the excited states in the k-E integration window depicted in dashed black contour in (c).

References

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