Extreme Fermi surface smearing in maximally disordered NiFeCoCr solid solution

Throughout history, advances in civilization have often been driven by advances in our mastery of materials. An alloy is a combination of at least two different elements. Alloy design has traditionally focused on there being one principal element (e.g., Cu) to which a small amount of something else is added, for example adding Sn to make bronze. In a class of materials known as "high entropy alloys", there are typically at least four different elements present at nearly equal concentrations [1,2]. This class of materials has shown unexpected and technologically important properties, such as their strength at low temperature. Further progress in exploiting their properties can be accelerated by understanding how the electrons behave in these complex alloys.

The Fermi surface of a metal is a surface of constant energy that separates the occupied and unoccupied electron momentum states, and its shape is crucial for determining the properties of the metal [3]. For most metals at low temperature, the Fermi surface is a sharp discontinuity between occupied and unoccupied states in momentum space, but the maximal disorder in these high entropy alloys smears this discontinuity across a significant fraction of the Brillouin zone. Using a simple argument based on Heisenberg's uncertainty principle, this smearing can be related to the distance the electrons can typically travel before being scattered (known as the mean-free-path). The electrons in most metals can travel unhindered over very long distances (even up to a cm), but it has been possible to show [4] that the electrons in the equiatomic NiFeCoCr alloy considered here would scarcely make it to the next atom before being scattered.

High-resolution X-ray Compton scattering experiments were performed at SPring-8 **BL08W**, allowing direct visualization of the impact of compositional disorder on the Fermi surface [4]. The nature of these alloys poses significant challenges to alternative techniques (such as quantum oscillatory methods or angle-resolved photoemission) since they are impossible to cleave and the electronic mean-free-path is extremely short. The Compton scattering technique is particularly powerful because the high-energy X-rays unambiguously probe the bulk electronic structure. Although the measured Compton profiles are actually projections of the electron momentum density in which two of the three momentum components have been integrated over, by measuring profiles down carefully chosen crystallographic directions (in this case it was 15 different directions) it is possible to tomographically reconstruct the full three-dimensional electron momentum density from this set of profiles. It is then straightforward to convert this electron momentum density into the occupation numbers within the first Brillouin zone, with the Fermi surface being associated with a change in the occupation number. First-principles electronic structure calculations were performed using the Korringa–Kohn–Rostoker (KKR) method within the Coherent Potential Approximation (CPA) and the Disordered Local Moment (DLM) state.

Figure 1 shows the calculated Bloch spectral function for the equiatomic NiFeCoCr alloy. For an ordered material, the spectral function would be an

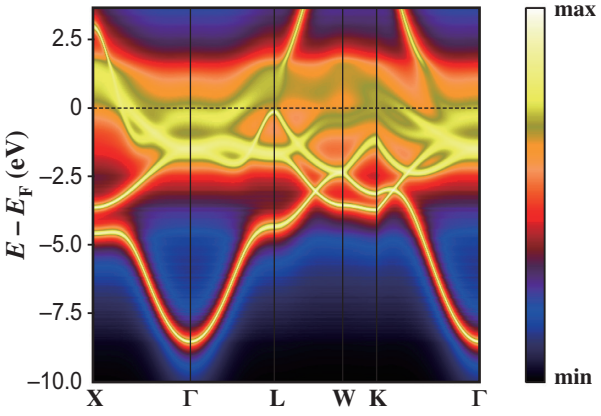

Fig. 1. The logarithm of the Bloch spectral function of NiFeCoCr from the KKR-CPA-DLM calculations.

image of the band structure of that material, showing sharply defined electronic energy bands. However, in a disordered alloy, the bands are smeared both in energy and crystal momentum, ***k***, due to the finite quasiparticle lifetime and mean-free-path, respectively. In Fig. 1 it can be seen that there is very significant smearing of the bands near the Fermi energy which implies that the Fermi surface is going to be substantially smeared.

Figure 2 shows a cut through the occupation number along the [110] direction, revealing a succession of three heavily smeared steps in occupation corresponding to three different Fermi surface sheets (the existence of which was also predicted by the first-principles calculations). By fitting a set of three smeared step functions to the ***k***-dependence of the occupation number, it was possible to extract the coherence length (which can be thought of as a ***k***-resolved mean-free-path).

This was the approach used to produce the colors on the Fermi surface sheet that is shown in Fig. 3. The colors indicate the effective coherence length for electrons located on that part of the Fermi surface (the average over all of the Fermi surfaces would be the mean-free-path of the alloy). It is important to note that the coherence length in this particular alloy is very close to the so-called Mott–Ioffe–Regel limit which describes the semi-classical upper bound for coherent electron transport.

Beyond their technological relevance, studies of these materials will also shed light on fundamental physics. Since the effects of chemical disorder are similar to those of strong electron-electron correlations on transport, these alloys may also be helpful in understanding the behavior of electrons in strongly correlated materials such as high-temperature superconductors. More recent work has probed the nature of the magnetism in these using magnetic Compton scattering at BL08W and XMCD at BL25SU [5].

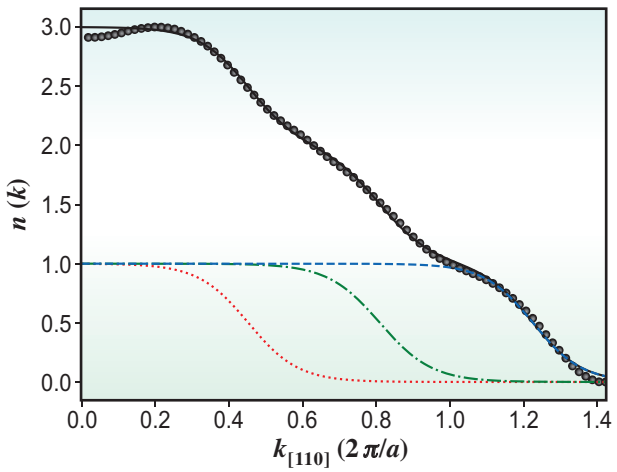

Fig. 2. Occupation number, $n(\boldsymbol{k})$, from the experiment along the [110] direction. $n(\boldsymbol{k})$ has been fitted with three *tanh* functions (one for each Fermi surface sheet) to represent smeared step functions from which the coherence length can be extracted.

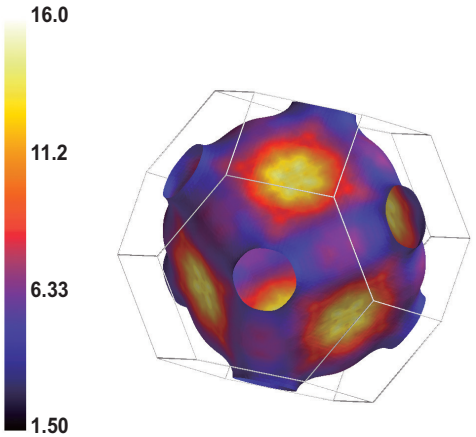

Fig. 3. One of the electron Fermi surface sheets obtained from the Compton experiment as an isodensity of the occupation number. The colors show how the coherence length (in angstrom) extracted from the experiment varies across the Fermi surface. The wireframe box shows the first Brillouin zone.

Stephen B. Dugdale

H.H. Wills Physics Laboratory, University of Bristol, United Kingdom

Email: S.B.Dugdale@bristol.ac.uk

References

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