Critical slowing down in charge fluctuation in a strange metal probed by synchrotron radiation-based Mössbauer spectroscopy

A strange metal (SM) is a ubiquitous state of matter found to develop in quantum materials with strong correlations. It is often linked to quantum criticality (QC) at the border of magnetism. SMs share many commonalities, e.g., anomalous temperature dependences of the specific heat and resistivity. These observed properties are inconsistent with the quasiparticle excitation concept central to the Fermi liquid theory. This universality challenges the conventional wisdom of conductivity based on a momentum ($\boldsymbol{k}$) relaxation of quasiparticles.

Although spin dynamics have been extensively studied, little is known experimentally about charge dynamics. Charge dynamics are studied by optical spectroscopy, but the method only probes the *divergence-free* transverse components of the current $\boldsymbol{J} = \sigma\boldsymbol{E} \perp \boldsymbol{k}$. Mössbauer spectroscopy is a method used to detect low-frequency longitudinal charge dynamics. However, the widespread adoption of Mössbauer methods has been hindered by difficulties in preparing radioisotope sources. To overcome these difficulties, a new generation of Mössbauer spectroscopy has been developed using synchrotron radiation (SR) [1]. The new SR-based Mössbauer spectroscopy approach provides an ideal probe to resolve longitudinal charge dynamics in materials.

Figure 1(a) shows the experimental setup for SR-based ^{174}Yb Mössbauer spectroscopy at SPring-8 **BL09XU** and **BL19LXU**. The monochromatized SR pulse passed through a sample including ^{174}Yb nuclei and then encountered YbB_{12} known as a scatterer. This scatterer was moved using a velocity transducer to create a relative Doppler velocity between the sample and the scatterer. We independently observed weak nuclear resonant scattering delayed by the finite lifetime (τ_0) of the excited nuclear state. Figure 1(c) shows a delay time spectrum after each SR pulse from ^{174}Yb nuclei in the YbB_{12} scatterer. The delayed scattering signals in nuclear recoil-free absorptions were accumulated within the time window from $\tau_s = t_s/\tau_0$ to $\tau_e = t_e/\tau_0$ to measure a SR-based Mössbauer spectrum. As seen in Fig. 1(c), a typical time window in our experiments was in the range from $\tau_s \sim 3.1$ to $\tau_e \sim 6.2$.

Figure 2 shows the SR-based ^{174}Yb Mössbauer spectrum of YbB_{12} at 20 K. The full-width at half-maximum of one absorption component at 0 mm/s was evaluated to be ~1.2 mm/s which is much narrower than that ($2G_0$ = 2.00 mm/s) expected from τ_0 = 2.58 ns. This narrowing phenomenon is related to the time-window effect in the accumulation of the delay time spectrum. A Lorentzian shape with the expected width should be obtained in a spectrum within the time window from $\tau_s \sim 0$ to $\tau_e \sim \infty$. The SR-based ^{174}Yb Mössbauer spectrum is well analyzed using one nuclear transition with this time-window effect.

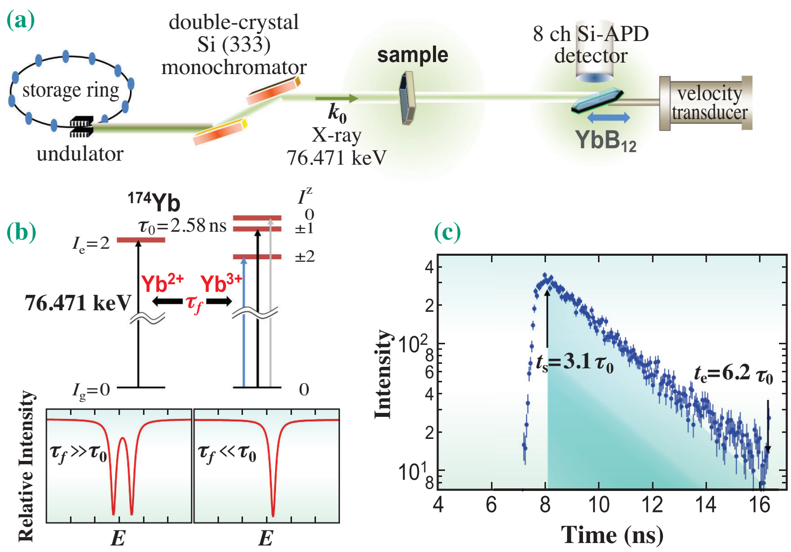

Fig. 1. **(a)** Schematic experimental setup for synchrotron-radiation-based ^{174}Yb Mössbauer spectroscopy. The c-axis of the single-crystalline β-$YbAlB_4$ was aligned along $\boldsymbol{k}_0$ of the incident X-ray and YbB_{12} was cooled at 26 K. **(b)** (Top) Energy diagrams of the excited ^{174}Yb (I_e = 2) nuclear state surrounded by different charge configurations. The allowed transitions are indicated by arrows, where the black ones represent the selected transitions for $c//\boldsymbol{k}_0$. (Bottom) Typical Mössbauer spectra at two limiting cases where τ_f is a timescale of fluctuation between two different charge configurations. **(c)** Typical delay time spectrum from YbB_{12}.

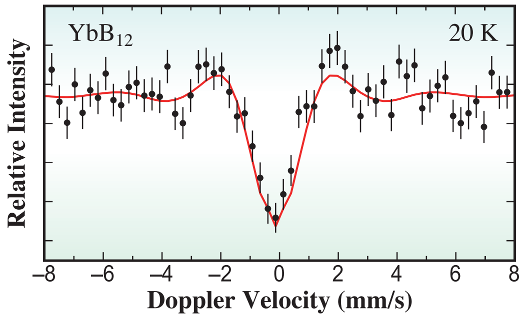

Fig. 2. Synchrotron-radiation-based ^{174}Yb Mössbauer spectrum of YbB_{12} at 20 K. The delayed scattering signals were accumulated within the time window (see Fig. 1(c)). The closed circles and red solid line present the observed and analytical spectra, respectively.

Since τ_s~3.1 is much larger than τ_s~0, an energy resolution higher than that of the conventional ^{174}Yb Mössbauer spectroscopy was achieved.

We report the direct observation of charge dynamics in an SM phase by SR-based Mössbauer spectroscopy. The heavy fermion metal β-$YbAlB_4$ provides an ideal platform to study a SM phase [2]. Accordingly, we have investigated how the QC behavior in the SM phase of β-$YbAlB_4$ affects the charge dynamics. Below T^*~8 K, as one enters the QC region, a two-peak structure is observed, as shown in Fig. 3(a). However, above T^*, the Mössbauer spectra exhibit a broad-single line feature.

The local symmetry at the Yb site of β-$YbAlB_4$ allows us to rule out a nuclear origin of the double-peak structure. For $c // \boldsymbol{k}_0$, the symmetry selects two degenerate nuclear $I_g = 0 \rightarrow I_e^z = \pm 1$ transitions from the five $E2$ nuclear transitions of the ^{174}Yb Mössbauer resonance (see Fig. 1(b)). The absence of magnetic order also eliminates magnetic hyperfine interactions. This leaves the electrical hyperfine interactions, linking to the valence state of Yb ions, as the only candidate for the observed splitting. The presence of Mössbauer line splitting then implies a distribution of Yb valences within the crystal. We argue that these result from slow dynamic charge fluctuations.

We have analyzed our Mössbauer spectra applying a stochastic theory with one nuclear transition modulated by two different charge states [3]. Figure 3(a) shows that the predicted spectrum well reproduces the two-peak structure in the spectrum at 5 K and its subsequent collapse into a broad single line with increasing temperature. The extracted fluctuation time τ_f between two different Yb charge states is unusually long compared with the electronic timescales (see Fig. 3(b)). The energy difference between two selected nuclear transitions is almost independent of temperature up to 20 K (see Fig. 3(c)), so that the development of the two-peak structure in the observed spectra must derive from the marked low-temperature growth in τ_f [4]. This consistency leads us to interpret the split line-shape observed in the ^{174}Yb Mössbauer spectra of the SM as unusually slow valence fluctuations between the Yb^{2+} and Yb^{3+} ionic states, on a timescale of τ_f >1 ns that follows a power-law growth with decreasing temperature below T^*.

In summary, we have provided direct evidence for unusually slow charge fluctuations in the SM phase of β-$YbAlB_4$ using SR-based ^{174}Yb Mössbauer spectroscopy. An interesting possibility is that these slow charge modes are the origin of the linear resistivity often observed in SMs. Various theoretical approaches [5] have suggested that the novel transport properties of SMs are linked to the universal quantum hydrodynamics of a Planckian metal.

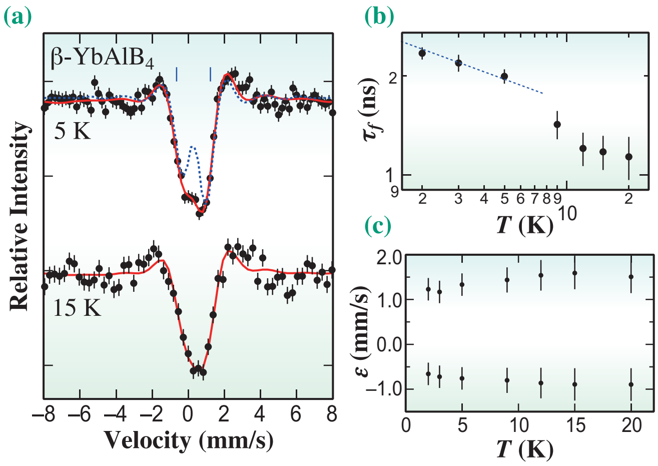

Fig. 3. **(a)** Selected synchrotron-radiation-based ^{174}Yb Mössbauer spectra of β-$YbAlB_4$ and **(b** and **c)** temperature dependences of the refined fluctuation time τ_f between two different Yb charge states and the refined energy ε values for two nuclear transitions. In (a), the closed circles and red solid line present the observed and the analytical spectra, respectively. The broken blue line in the spectrum at 5 K represents the spectrum with two nuclear transitions expected with our energy resolution. In (b), the broken line represents $\tau_f \propto T^{-0.2}$.

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References

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