Observation of a Critical Charge Mode in a Strange Metal

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Quantum electronic matter has long been understood in terms of two limiting behaviors of electrons: one of delocalized metallic states, and the other of localized magnetic states. Understanding the strange metallic behavior which develops at the brink of localization demands new probes of the underlying electronic charge dynamics. Using a state-of-the-art technique, synchrotron-radiation-based Mössbauer spectroscopy, we have studied the longitudinal charge fluctuations of the strange metal phase of β-YbAlB$_4$ as a function of temperature and pressure. We find that the usual single absorption peak in the Fermi-liquid regime splits into two peaks upon entering the critical regime. This spectrum is naturally interpreted as a single nuclear transition, modulated by nearby electronic valence fluctuations whose long time-scales are further enhanced, due to the formation of charged polarons. Our results represent a direct observation of critical charge fluctuations as a new signature of strange metals.

The strange metal (SM) is a ubiquitous state of matter found to develop in quantum materials with strong correlations, often appearing as a fan-shaped region of the phase diagram centered around an unstable quantum critical (QC) point. SMs share many commonalities, most-notably a logarithmic temperature (T) dependence of specific heat $C/T \sim -\log T$, a linear-in-T resistivity $\rho(T) \sim T$ (*1*) and a strong violation of Kohlers law in the magnetotransport (*2*, *3*, *4*). These properties and their universality defy the standard concept of quasiparticle excitations and the conventional wisdom of momentum-relaxation-origin of the conductivity, central to the Fermi liquid (FL) theory of metals. This enigma has prompted a wide range of possible origins, including spin-density instability (*1*), Fermi surface instablity (*5*, *6*, *7*), valence quantum criticality (*8*), charge stripes (*9*), and nematicity (*10*, *11*, *12*) and motivated novel approaches, including the holographic duality (*13*, *14*, *15*) and simulation using cold atoms (*16*).

While the spin dynamics at quantum criticality has been extensively studied, little is known experimentally about the charge dynamics as appropriate laboratory probes are scarce. Conventionally, charge dynamics are studied using optical spectroscopy (*17*), but these methods only probe the low-momenta, divergence-free *transverse* components of the current $\boldsymbol{J} = \sigma \boldsymbol{E} \perp \boldsymbol{k}$ that, by the continuity equation, do not couple to fluctuations in the charge density. Longitudinal charge fluctuations can be probed by electron energy loss spectroscopy (EELS) but are limited to energies above the Debye energy due to a phonon background (*18*, *19*, *20*). A classic method to detect low frequency longitudinal charge dynamics is Mössbauer spectroscopy, successfully used in the past to detect the slowing of the charge dynamics at charge ordering transitions of Eu and Fe based compounds (*21*, *22*).

However, the widespread adoption of Mössbauer methods has been long hindered by the lack of suitable radioisotope sources. To overcome these difficulties, a new generation of Mössbauer spectroscopy has recently been developed using synchrotron radiation (SR) (*23*). SR-based Mössbauer spectroscopy (see Fig. 1 A) can be used for a wide range of Mössbauer

isotopes, providing improved energy resolution for these isotopes with shorter-lifetimes; it offers an unprecedented capability to select a particular nuclear transition, taking advantage of the perfectly polarized SR. This new approach presents an ideal probe to resolve *longitudinal* charge dynamics in materials for which conventional Mössbauer techniques are inapplicable.

Here we report the first direct observation of critical charge dynamics in a SM regime using SR-based ^{174}Yb Mössbauer spectroscopy. The heavy fermion metal β-$YbAlB_4$ provides an ideal platform to study a SM regime at ambient pressure in a stoichiometric crystal (*3*, *24*). In β-$YbAlB_4$, core level X-ray studies have established the presence of an intermediate valence state caused by valence fluctuations between two ionic configurations (*25*) $Yb^{2+} \rightleftharpoons Yb^{3+} + e^-$. Usually, in heavy fermion compounds, such valence fluctuations are too fast to be observed by Mössbauer spectroscopy (*26*, *27*, *28*, *29*), but here we show that this is not the case in the SM.

Mössbauer spectroscopy measures the shift in a nuclear absorption line due to changes in the local (q-integrated) charge density. The characteristic time-scale of the measurement is the lifetime of the nuclear excited state, $\tau_0 \sim 2.5$ns in ^{174}Yb. Charge fluctuations that are much shorter in time than τ_0 produce a single motionally narrowed absorption line, whereas charge fluctuations that are much longer in time than τ_0 produce a double peak absorption line, corresponding to the two different valence states of the Yb ion (see Figure 1 C). By fitting the Mössbauer absorption line-shape, one can detect charge fluctuations with time-scales in the range $\sim 0.1\tau_0$ to $\sim 10\tau_0$ (*30*).

β-$YbAlB_4$ exhibits quantum criticality without tuning in an intermediate valence state (*25*), and the application of an infinitesimal magnetic field B tunes the SM into a FL with $k_{\rm B}T_{\rm FL} \sim \mu_{\rm B}B$. The slope of the linear-in-T resistivity $\rho(T) \sim T$ over T between 0.5 and 25 K at ambient pressure, corresponds to a nearly quantum-saturated scattering rate $\tau_{tr}^{-1} = 0.4 \times k_B T/\hbar$ (*30*), thus establishing β-$YbAlB_4$ as a system with Planckian dissipation (*31*). This anomalous $\rho(T)$ and its extension over a broad pressure (p) range from ambient pressure to $p^* \sim 0.5$GPa

(*3*, *24*, *32*) (see Fig. 1 B) provides an excellent setting for high precision measurements of the critical charge fluctuations, likely of relevance to the broader family of SMs.

We have investigated how the QC behavior in the SM regime affects the charge dynamics, following their evolution as the SM regime at ambient pressure transforms into a FL regime under pressure. Above 9K at ambient pressure (Fig. 2 A) the Mössbauer spectra exhibit a single line feature. However, below $T^* \sim 10$K, as one enters the QC region, this peak broadens into a two-peak structure, with 5σ significance (*30*). Fig. 2 B shows how this two-peak structure observed for $p < 0.7$GPa at 2K coalesces into a single peak around $p \sim 1.2$GPa, ultimately sharpening into an almost resolution-limited peak at $p = 2.3$GPa characteristic of a Fermi liquid (*30*).

The local symmetry at the Yb site of β-$YbAlB_4$ with the orthorhombic structure allows us to rule out a nuclear origin of the double-peak structure. For $c \parallel \boldsymbol{k}_0$ (the propagation vector of the incident X-ray), the symmetry selects two degenerate nuclear transitions $I_g = 0 \rightarrow I_e^z = \pm 1$ from the five $E2$ nuclear transitions ($\Delta I^z = 0, \pm 1$, and ± 2) of the ^{174}Yb Mössbauer resonance (*33*) (see Figs. 1 A and C). The absence of magnetic order in β-$YbAlB_4$ (*24*, *32*) also eliminates magnetic and non-axially symmetric quadrupolar hyperfine interactions as explanations (*30*). This leaves a combination of the electric monopole and axially symmetric quadrupolar interactions, linking the hyperfine energy to the valence state of the rare-earth ion, as the only candidate for the observed splitting. The presence of a Mössbauer line splitting then implies a distribution of Yb valences within the crystal. We now argue that these result from slow dynamic charge fluctuations.

All Yb sites are crystallographically equivalent in β-$YbAlB_4$ and SR X-ray diffraction measurements (*34*) show that the lattice structure does not change up to 3.5GPa at 7K; furthermore the absence of any low-temperature phase transitions rules out the possibility of a charge density wave (*30*). Moreover, the residual resistivity ratio (RRR) exceeds 100, indicating the low

levels of quenched disorder in this material. Since disorder broadens the Mössbauer absorption peak, our ability to resolve the double-peak structure is consistent with this conclusion. An attempt to fit the Mössbauer spectrum with two nuclear transitions (i.e. a static hyperfine interaction), using a width corresponding to the experimental energy resolution, fails to reconstruct the feature at 2K and $\sim$ 0 mm/s (blue broken line). Thus the two-peak structure and line broadening observed for $T < 5$K and $p < 0.7$GPa must derive from a single nuclear transition that is dynamically modulated by fluctuations between two different Yb charge states (i.e. a time-dependent hyperfine interactions) (Fig. 1 C) (*30*).

We have analyzed our Mössbauer spectra at ambient pressure using a stochastic theory (*35*, *36*, *37*) with a single nuclear transition modulated by two different charge states (*30*). Fig. 2 A shows that the predicted spectra (red lines) well reproduce the two-peak structure in the spectra at low Ts and its subsequent collapse into a single line with increasing T.

At ambient pressure, the extracted fluctuation time τ_f between two different Yb charge states is unusually long compared to the electronic timescales, exhibiting a slow power-law growth $T^{-\eta}$ ($\eta \sim 0.2$) on cooling below T^* (Fig. 2 C). The energy difference between two selected nuclear transitions is almost independent of T up to 20K (*30*), so that the development of the two-peak structure in the observed spectra must derive from the marked low-T growth in τ_f. On the other hand, as shown in Fig. 2 B, the gradual collapse of the two-peak structure in the observed ^{174}Yb Mössbauer spectra at 2K with increasing p indicates that fluctuation timescale τ_f becomes shorter as a function of p. The spectra at p <1.2GPa can only be analyzed and reconstructed by the same stochastic model used at the ambient pressure, while the spectrum observed at 2.3GPa was simply fit using the static model. The line-width of this single absorption component was found to be $\Gamma = 1.11$ mm/s, slightly broader than the resolution limit $\Gamma_0 = \hbar/\tau_0 = 1.00$ mm/s (3mK), for ^{174}Yb Mössbauer spectroscopy (τ_0 =2.58ns).

As seen in Fig. 2 D, τ_f gradually decreases with increasing p, exhibiting a kink across $\sim p^*$

in between 0.5 and 1GPa, approaching the resolution limit at 2.3GPa. This is roughly consistent with previous $\rho(T)$ measurements in β-YbAlB$_4$ (*32*); at T <0.5K and under p, $\rho(T)$ displays $\rho \sim T^{\alpha}$ with α = 3/2 below p^* and further application of pressure increases the exponent to α = 2, stabilizing a FL state at about 1GPa (*32*). However, the Fermi liquid temperature $T_{\rm FL}$ depends on p, and only for $p \sim 2.3$GPa is the system in the FL regime at $T = 2$K (*32*).

The above consistency leads us to interpret the split line-shape observed in the Mössbauer spectra of the SM as unusually slow valence fluctuations between the Yb^{2+} and Yb^{3+} ionic-like states in β-YbAlB$_4$, on a timescale $\tau_f > 1$ns that follows an approximate power-law growth $\tau_f \sim T^{-0.2}$ with decreasing temperature below T^*. The Yb^{3+} ground state is a $J_z = \pm 5/2$ moment as deduced by varying incident angle of the X-ray (*30*). The slow charge fluctuations extend up to p^*, beyond which a conventional valence fluctuation state with rapid charge fluctuation takes over in the pressured regime corresponding to the FL regime.

The unusual aspect of the observed charge dynamics is that not only are they slower than the Planckian time $\tau_f \gg \tau_{tr} \sim 10^{-2}$ ns at 2K, but they are also slower than the characteristic time-scale of the lattice vibrations as we will show shortly. In this situation, the lattice is expected to adiabatically respond to the associated charge redistribution. Each valence fluctuation of Yb atoms is then dressed by N_p phonons, leading to the formation of a polaron (*38*, *39*) and renormalizing the matrix element for the charge fluctuations and providing a mechanism for enhancing their time-scale ($\tau_f \rightarrow \tau_f e^{N_p}$) (*30*). Analysis of the Mössbauer spectra allows us to directly check this scenario. We have used the T-dependence of the absorption components in the spectra to determine the Lamb-Mössbauer (recoil-free) factor $f_{\rm LM}$ in β-YbAlB$_4$, the equivalent of the Debye-Waller factor in a usual scattering experiment. Generally, $-\ln(f_{\rm LM}) = k_0^2\langle \Delta z^2 \rangle$, where Δz is an atomic displacement from a regular position in a crystal along the

direction of $\boldsymbol{k}_0$ (*40*). The expression for the variance in atomic position is

$$\langle \Delta z^2 \rangle \propto \int_0^\infty d\omega \frac{F(\omega)}{\omega} \overbrace{\left[\frac{1}{2} + \frac{1}{e^{\omega/T}-1}\right]}^{\frac{1}{2}\coth(\beta\omega/2)} \quad (1)$$

where $F(\omega)$ is the (partial) phonon density of states. In a Debye model, $F(\omega) \propto \omega^2$ which leads to $\langle \Delta z^2 \rangle \propto \left[3/2 + (\pi/\Theta_D)^2 T^2\right]$ at $T \ll \Theta_{\rm D}$, where $\Theta_{\rm D}$ is the Debye temperature (*30*). As seen in Fig. 3 A, this Deybe relation holds above T^* at ambient pressure, where τ_f ($\sim$ 1.15ns) is independent of T; from this we estimate $\Theta_{\rm D}$ = 95K, corresponding to the lattice response time $\tau_L \sim h/k_{\rm B}\Theta_{\rm D} \sim 0.5$ps, so that $\tau_f \gg \tau_L$. The estimated $\Theta_{\rm D}$ (= 95K) value is unusually smaller than that (195K) of a conventional valence fluctuation metal $YbAl_2$ (*41*). This indicates that the lattice vibrations are soft in β-$YbAlB_4$, suggesting an enhanced effective coupling between slow charge fluctuation modes and lattice vibrations.

Additionally, we see from Fig. 3A that in the QC regime below T^*, where τ_f develops temperature-dependence, $\langle \Delta z^2 \rangle$ departs from this Debye behavior, indicating an enhancement in the quantum fluctuations, $\langle \Delta z^2 \rangle = \langle \Delta z^2 \rangle_{\rm Debye} + \delta\langle \Delta z^2 \rangle$, of the Yb ions. Notably, the $\sqrt{\delta\langle \Delta z^2 \rangle} \sim 0.014$Å rms fluctuation observed here is comparable to the quantum fluctuations of the phonon mode, around 0.05Å estimated from $\frac{1}{4}k_{\rm B}\Theta_{\rm D} \sim \frac{1}{2}m_{\rm Yb}(k_{\rm B}\Theta_{\rm D}/\hbar)^2\langle \Delta z^2 \rangle$. Fig. 3 B shows that $\langle \Delta z^2 \rangle$ is approximately constant at 2K for $p < p^*$ and then drops when $p > p^*$, indicating that the anomalous vibrations of the lattice, $\delta\langle \Delta z^2 \rangle$, disappear in the FL regime at low temperatures.

The saturation of $\langle \Delta z^2 \rangle$ for $T < T^*$ and $p < p^*$ implies that the phonon spectrum $F(\omega)$ has changed its form to compensate the $\coth(\beta\omega/2)$ term in the integral (1). This then suggests that at energies and temperatures below T^*, $F(\omega)$ acquires a temperature-dependence $F(\omega, T) = \phi(\omega)\tanh(\omega/2T)$ that cancels the $\coth(\omega/2T)$ term in integral (1). The function $\tanh(\omega/2T) \sim \omega/2T$ for $\omega \ll T$ and $\tanh(\omega/2T) \sim 1$ for $\omega \gg T$, and thus has the Marginal Fermi Liquid (MFL) form. This enhancement in phonon density of states should be observable

in inelastic neutron scattering measurements. Since the phonons are linearly coupled to the charge density of the electrons, the appearance of a MFL component in the phonon spectrum is an indication of MFL behavior in the charge fluctuations. The enhancement of τ_f by polaron formation has been crucial for slowing the charge fluctuations down to time-scales accessible to Mössbauer spectroscopy.

A possible interpretation of our results is the quantum critical tuning of a critical end point of a classical valence transition (*42*) between the Yb^{2+} and Yb^{3+} ionic states. Such first order valence transition lines, with second order end-points, are well established in rare earth compounds. It has been suggested (*42*) that the tuning of such an endpoint to zero temperature may provide an explanation of the observed Mössbauer spectra.

An alternative interpretation is that the observed valence fluctuation modes are an intrinsic property of the SM regime connected with a spin charge separation that develops with the collapse of the f-electron Fermi surface (*43*, *44*, *45*, *46*). This scenario suggests that similar slow charge fluctuations will be manifested in the Mössbauer spectra of any partial Mott localization critical point, e.g. in other heavy-fermions and Iron-based superconductors.

In summary we have provided direct evidence for unusually slow charge fluctuations in the SM regime of β-$YbAlB_4$ using a state-of-the-art technique, SR-based Mössbauer spectroscopy. Because their time-scales are longer than that of the lattice response, we have inferred polaronic formation in the mixed valence regime (*38*, *39*). Both the slow charge fluctuation modes and the anomalous vibrations of the lattice disappear in the pressure-induced FL regime. An interesting possibility is that these observed slow charge modes are the origin of the linear resistivity often observed in SMs. Various theoretical approaches (*13*, *14*) have suggested that the novel transport properties of SMs are linked to the universal quantum hydrodynamics of a Planckian metal. Since the local equilibrium is established at the scale of Planckian time, it is natural to regard the slow charge fluctuations detected here as a possible signature of a new hydrodynamic mode.

This would lead us to expect that nano-second charge fluctuations and anomalous vibrations are not unique to β-$YbAlB_4$, but rather, are universal properties of SM regimes in quantum materials.

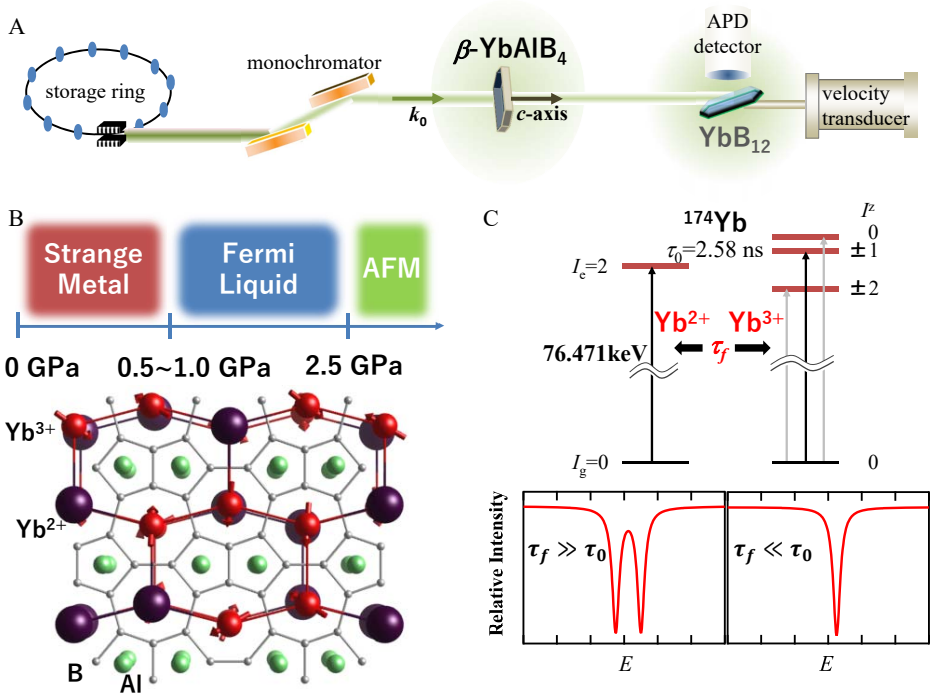

Figure 1: (A) Schematic of our experimental setup for the synchrotron-radiation-based ^{174}Yb Mössbauer spectroscopy (*47*). The ^{174}Yb nuclear resonance (E_γ =76.471keV) was obtained by synchrotron radiation using a monochromator. The c-axis of the single crystalline β-YbAlB$_4$ samples was aligned along the propagation vector $\boldsymbol{k}_0$ of the incident X-ray under both ambient and external pressure. The single-crystalline YbB$_{12}$ were cooled at 26K. A Si avalanche photodiode (APD) detector was used to measure delayed incoherent emission from ^{174}Yb nuclei in the YbB$_{12}$. (B) Schematic phase diagram of β-YbAlB$_4$ as a function of pressure at low temperatures (Top) and cartoon of the crystal structure of β-YbAlB$_4$ with a snapshot of the Yb valences, i.e., Yb^{2+} (large dark red sphere) and Yb^{3+} (small red sphere with arrow indicating magnetic moment) (Bottom). (C) (Top) Energy level diagrams of the excited ^{174}Yb (I_e=2) nuclear state with the lifetime of $\tau_0 = 2.58$ns surrounded by different charge configurations. The allowed Mössbauer transitions are indicated by arrows, where the black arrows represent two selected transitions for $c \parallel \boldsymbol{k}_0$. (Bottom) Two typical Mössbauer absorption spectra at limiting cases with $\tau_f \gg \tau_0$ and $\tau_f \ll \tau_0$ where τ_f is a characteristic timescale of fluctuation between two different charge configurations.

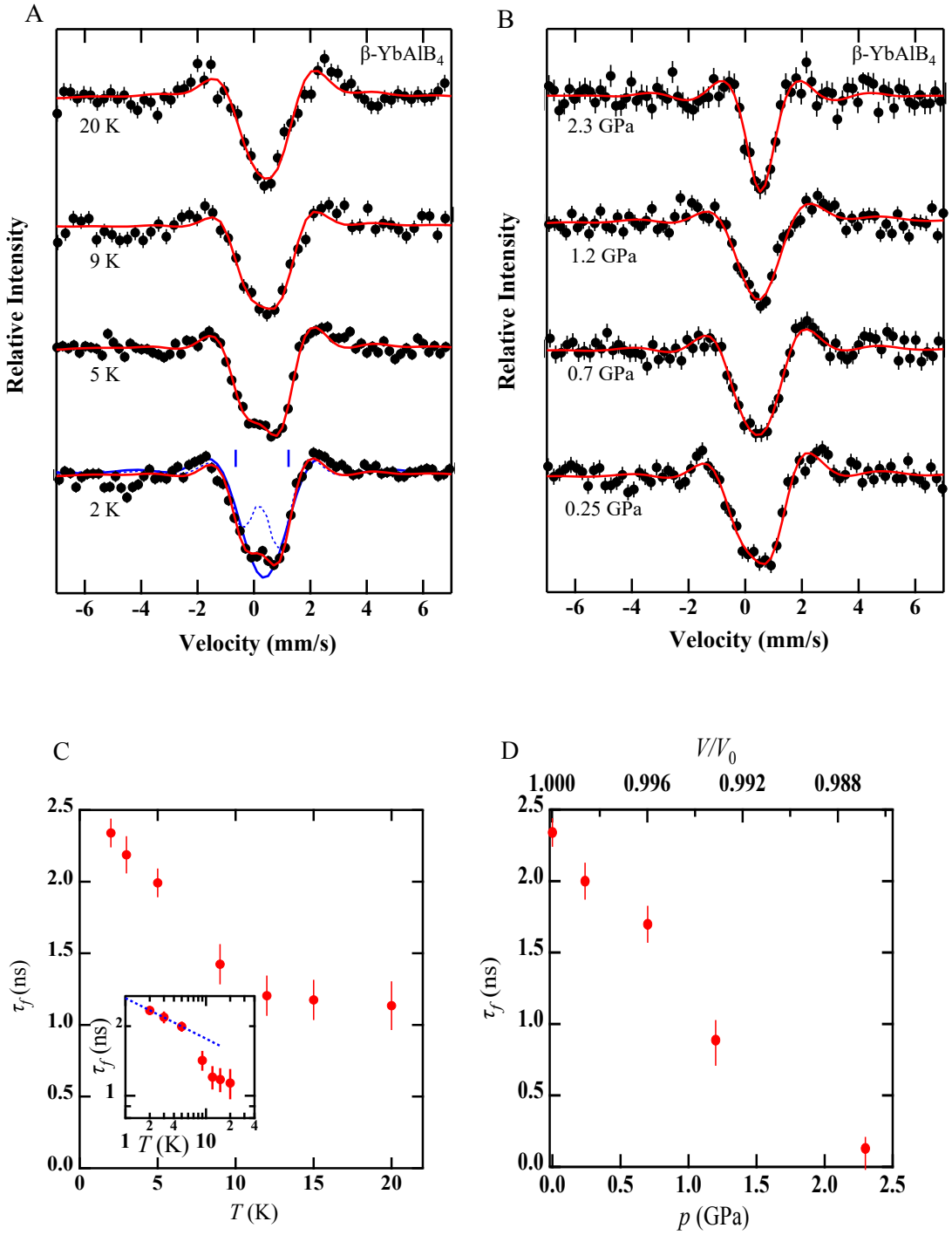

Figure 2: Selected synchrotron-radiation-based ^{174}Yb Mössbauer spectra of β-$YbAlB_4$ as a function of temperature (T) at ambient pressure (A) and under external pressure (p) at 2K (B). The c-axis of the single crystalline β-$YbAlB_4$ samples was aligned along the propagation vector $\boldsymbol{k}_0$ of the incident X-ray. The solid circles with error bar and the red solid lines present the observed and the analytical spectra, respectively. In A, the broken blue line in the spectrum at 2K represents the spectrum with two static nuclear transitions expected with our experimental energy resolution, whereas the solid blue line shows a fit to the wings of the lineshape, discarding the double-peak structure in the center. The deviation at the center corresponds to 5σ statistical significance (*30*). Temperature T (C) and pressure p (D) dependences of the refined fluctuation time τ_f between two different Yb charge states in β-$YbAlB_4$. (Inset in (C)) Log-log plots of τ_f versus T in β-$YbAlB_4$. The broken line represents $\tau_f \sim T^{-0.2}$.

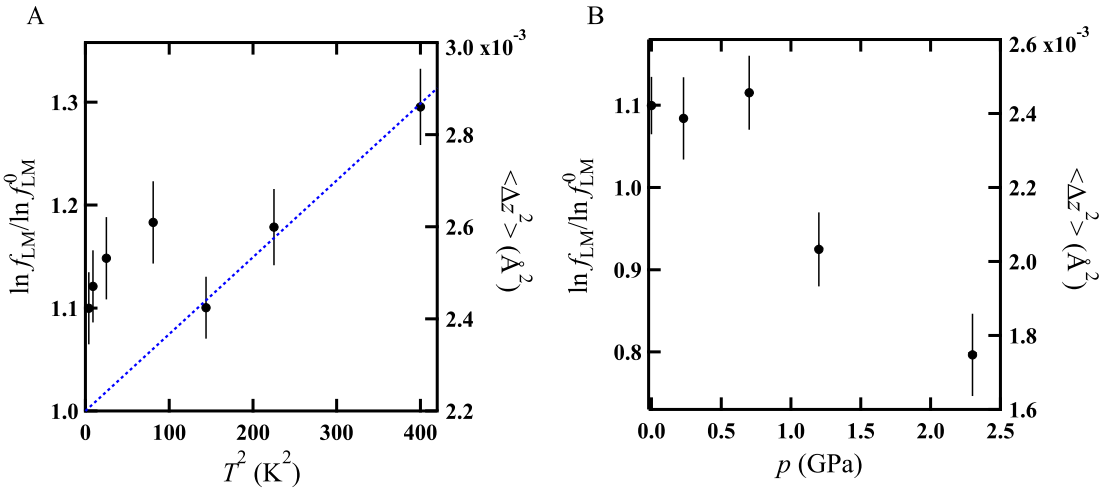

Figure 3: Lamb-Mössbauer factor $f_{\rm LM}$ ($\ln f_{\rm LM}/\ln f^0_{\rm LM}$) as a function of T^2 at ambient pressure (A) and under external pressure p at 2K (B) for β-$YbAlB_4$. In (A), the broken line represents a linear relation between $\ln f_{\rm LM}$ and T^2. In (A) and (B), $\ln f^0_{\rm LM}(\propto -\frac{3}{2}\frac{E_{\rm R}}{k_{\rm B}\Theta_{\rm D}^{\rm Yb}})$ was estimated above 12K at ambient pressure. For ^{174}Yb Mössbauer resonance of k_0=38.75Å^{-1}, $\langle\Delta z^2\rangle$ for the Yb ions was evaluated in β-$YbAlB_4$ from the T and p dependences of $\ln f_{\rm LM}/\ln f^0_{\rm LM}$ using $\Theta_{\rm D}^{\rm Yb}$= 95K. In (A) and (B), the $\langle\Delta z^2\rangle$ values (right axis) are $\sim 2.6\times10^{-3}$Å^2 in the SM regime and decrease to 1.7×10^{-3}Å^2 in the pressured regime corresponding to the FL regime, which is comparable with that for $YbAl_2$ (*41*).

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