

Direct Observation of Itinerant Magnetism in the 5*f*-Electron System UTe

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Our electron photoemission experiments demonstrate that the magnetization of the ferromagnetic state of UTe is proportional to the binding energy of the hybridized band centered around 50 meV below E_F . This proportionality is direct evidence that the ferromagnetism of UTe is itinerant; i.e., the 5*f* electrons are not fully localized close to the atomic core. This mechanism of itinerant ferromagnetism differs from the traditional picture for 5*f*-electron magnetism in an essential and a novel way. We propose a simple model for the observed proportionality between the temperature dependence of the magnetization and the binding energy of the hybridized band near E_F . This model allows us to estimate the effective magnetic interaction and to identify signatures of itinerant ferromagnetism in other materials.

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For years, the mixed-valence regime of *f*-electron materials has been associated with a paramagnetic state in which the *f*-magnetic moments are compensated by a strong antiferromagnetic coupling to the conduction electrons. Although this so-called collective Kondo state may hold for dilute systems with a low concentration of *f*-magnetic moments, it vanishes in the concentrated limit where one *f*-magnetic moment per site is available. In the concentrated limit the number of conduction electrons contributing to the screening is smaller than the number of magnetic impurities, and therefore screening breaks down. As an alternative to the collective Kondo state, it was recently suggested that itinerant ferromagnetism may occur for the strong hybridization and strong coupling (mixed-valence) regime of the periodic Anderson model (PAM) [1]. For higher dimensions, different theoretical approaches [2–5] have shown that the PAM also has an itinerant ferromagnetic phase in the mixed-valent regime, that is, when the *f*-orbital energy level coincides with the Fermi level E_F . Numerical calculations have emphasized the role hybridization plays in the appearance of this state [5]. In contrast to the traditional Ruderman-Kittel-Kasuya-Yosida (RKKY) mechanism for localized *f*-electron magnetism, the magnetic moments in the mixed-valent regime of the PAM are itinerant.

To prove or disprove the theoretical predictions, one has to investigate a mixed-valent ferromagnetic *f*-electron system experimentally. The semimetallic monochalcogenide UTe is a ferromagnet with a relatively high Curie temperature $T_C = 104$ K [6,7], a large anisotropy, and an ordered moment of $2.25(5)\mu_B$ at 0 K [6]. The relatively

high Curie temperature requires a large value of the exchange interaction—too large to be successfully explained by RKKY interaction. Existing experimental and theoretical evidence seems to be mutually exclusive. Mixed valence was confirmed by the negative c_{12} elastic constant below T_C found by Brillouin scattering [8,9]. The relatively high Curie temperature was believed to result from hybridization and the high degree of delocalization of the uranium 5*f* states [10]. Failure to reproduce the negative c_{12} of UTe within the local-spin-density approximation was interpreted as a signature of possible mixed valence [11]. Some amount of hybridization of 5*f* and conduction states was suggested by photoemission [12] and early measurements of transport properties [13]. UTe was then described as a dense “Kondo system” with a strong interaction between the localized 5*f* and itinerant conduction electrons. A quasilocalized nature was inferred from magneto-optical Kerr spectroscopy coupled with local density approximation with a Hubbard U (LDA + U) calculations [14]. Finally, support for UTe as a partially localized 5*f*-electron system on the border of instability towards itinerancy, with well-defined excitations propagating in only one direction, comes from neutron inelastic scattering experiments [15,16]. Overall, information coming from experiments is hitherto contradictory in determining the extent of hybridization or localization in UTe.

Mixed valence has been previously suggested as coexisting with ferromagnetism [17,18], but a firm signature of such coexisting states has been elusive [19]. As previously noted, recent theoretical studies indicate that this coexistence is possible. We will show that features of the energy

spectrum near E_F , revealed by high resolution photoemission measurements, affirm the itinerancy of the ferromagnetic state.

Single crystals of UTe were grown by the mineralization technique [20] at Eidgenössische Technische Hochschule in Zürich. Magnetization measurements were performed at LANL as a function of temperature and applied magnetic field. When the sample was cooled below T_C in 0.1 T, the maximum moment at low temperatures was $0.8\mu_B$; but when cooled in 5 T, the magnetization at 5 K was substantially larger, $1.8\mu_B$, and much closer to the neutron-diffraction value of $2.25\mu_B$. We interpret this discrepancy as arising primarily from incomplete domain alignment under low-field conditions. The effective moment extracted from susceptibility measurements well above T_C was $2.84\mu_B$, similar to $2.81\mu_B$ reported earlier [21]. Photoemission measurements were performed at the Synchrotron Radiation Center, Stoughton, Wisconsin, using the Plane Grating Monochromator (PGM) beam line. Spin-polarized photoemission does not currently provide sufficient energy resolution to distinguish the subtle changes in the binding energy shown here, and therefore was not applicable. In our photoemission study, we utilized a PGM beam line with an electron energy analyzer system capable of both a higher energy resolution of 25 meV at 34 eV, and a higher angular resolution of 1° , than previously reported [22]. The single crystals of UTe were oriented and cleaved in a vacuum of better than 5×10^{-11} Torr prior to measurement. There were no signs of surface degradation over the duration of the experiment. We focused on the near- E_F region. The normal emission data collected in the range 21.2 to 45 eV exhibited structure composed of two bands B and F within the first 2 eV from E_F , with band F being located near E_F . Angle-resolved photoemission results show the F peak to have a dispersive nature, consistent with narrow band character. From the lack of photon energy dependence in normal emission data and from the photoionization cross sections [23], we infer a hybridized nature for the conduction band.

In Fig. 1 we show the photoemission data and line shape analysis for UTe at selected temperatures between 32 K and T_C . Line shape analysis of the data was performed following procedures established in Refs. [24,25]. Analysis shows the peak F position at 53 meV for a measurement temperature of 32 K and a smooth, monotonic convergence to E_F as T increases to T_C . With the peak F location moving toward E_F and the influence of the Fermi function increasing with temperature, we have deconvoluted the Fermi function from the fitted line shape to elucidate the movement of peak F with temperature more clearly. In Fig. 1(b) we show details for the fitting of peak F at 32 K, with the blue line being the best fit at 32 K and the red line with the same parameters but without the Fermi function cutoff at E_F . In the top frame of Fig. 1, we show the analysis for 32, 89, and 103 K UTe data. These results

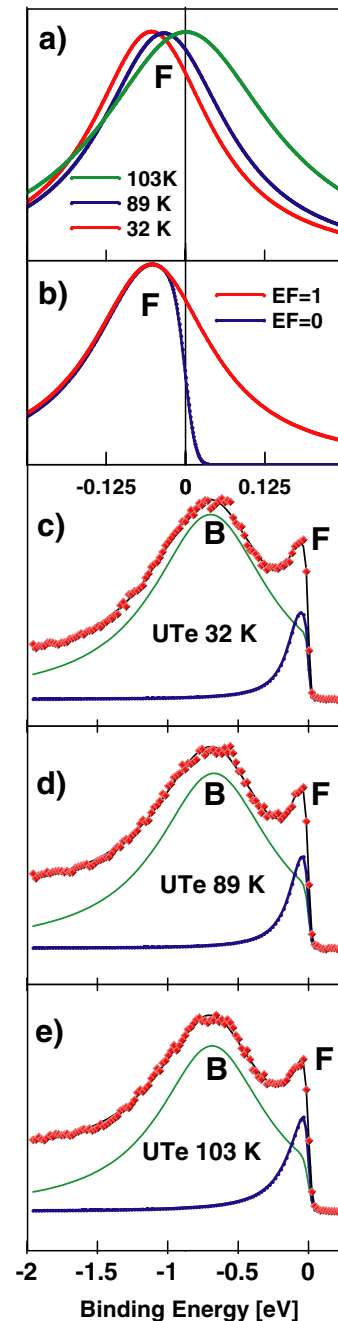


FIG. 1 (color online). Movement of the first peak (peak F) in the temperature-dependent photoemission spectrum of UTe. Panel (a) shows the fitted Lorentzian line shape at 32, 89, and 103 K, indicating the band movement towards E_F and increase in band width with increasing temperature. Panel (b) demonstrates the best fit at 32 K with and without the Fermi function cutoff. Panels (c)–(e) illustrate the original data (diamonds) and fitting results for both peaks B and F .

are directly obtained from the individual fits to the data in Fig. 1(c)–1(e) with the procedure defined above. The transition temperature of 104 K in UTe facilitated the examination of the temperature-related changes in the

bands below the Curie temperature without dominant broadening effects from the Fermi function. In particular, we see that band F in Fig. 1 approaches E_F with increasing temperature, and its center position crosses E_F at the transition temperature. The full width at half maximum (FWHM) of peak F is approximately 200 meV, and its binding energy (BE) at low temperatures is -53 meV. The FWHM and BE changes for peak B are insignificant compared to peak F .

The qualitative behavior of the bands can be understood within a simple ‘‘Stoner-like’’ mean-field treatment of the f - d electron system. Monte Carlo calculations for the PAM [5] show that there is instability towards itinerant ferromagnetism when the concentration of electrons is close to quarter filling and the Fermi energy of the non-interacting (paramagnetic) ground state coincides with the peak of the f -character density of states (F peak) [5]. The

Coulomb interaction induces a ferromagnetic ground state by polarizing most of the f electrons. As a consequence, the position of the F peak moves down relative to the Fermi level in order to satisfy the Pauli exclusion principle (see Fig. 2). In the mean-field description, the relative shift of the F peak is roughly half of the exchange splitting Δ_{ex} between the spin-up and spin-down bands. It is exactly $\Delta_{\text{ex}}/2$ if the f band is symmetric. The exchange splitting is $\Delta_{\text{ex}} = 2J_{\text{eff}}Jm/g\mu_B$, where J is the total angular momentum of each f electron, m is the magnetization per site, J_{eff} is the effective exchange interaction, g is the gyromagnetic factor, and μ_B is the Bohr magneton. From this expression, we conclude that the binding energy E_B is proportional to m with proportionality constant $\alpha = J_{\text{eff}}J/g\mu_B$; that is, $E_B(T) = J_{\text{eff}}Jm(T)/g\mu_B$. In Fig. 3 we compare the magnetization $m(T)$ multiplied by a constant $\alpha = 65$ meV/ μ_B with $E_B(T)$. The comparison strongly supports a linear relation between m and E_B , and it predicts an effective exchange interaction $J_{\text{eff}} = 11$ meV, assuming the $5f^3$ configuration and intermediate coupling with $g = 0.759$ and $J = 9/2$.

In contrast to the nonmagnetic collective Kondo state, which is expected for the intermediate-valence regime of a dilute system [26], our results for UTe show itinerant ferromagnetism coexisting with a mixed valency. From the observed linear relation between the F photoemission peak and the magnetization, we extracted an effective exchange interaction. It is important to note that the shift of the peak in the density of states relative to the Fermi level on a transition into the ferromagnetic state is a

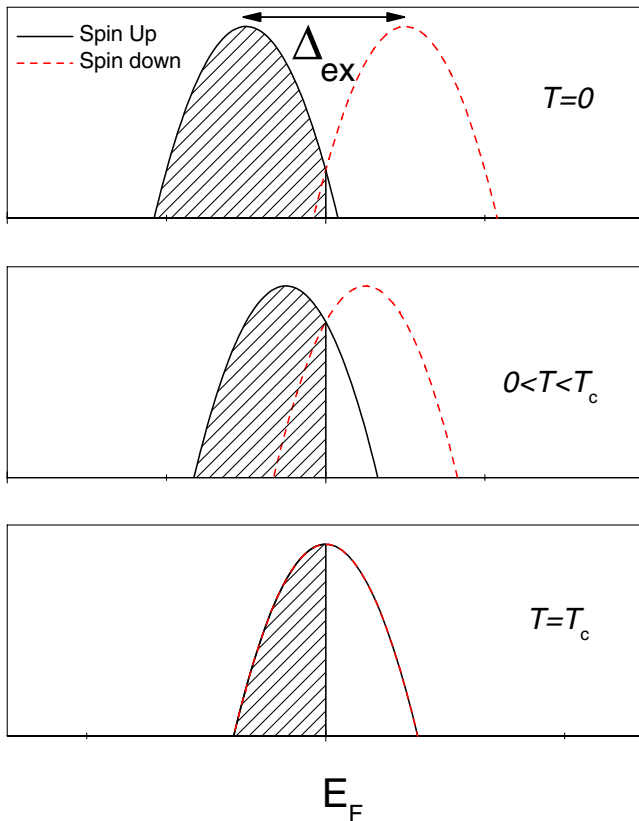


FIG. 2 (color online). At low temperatures ($T = 0$) the spin-up and spin-down bands are separated by the exchange splitting Δ_{ex} . Shading represents occupancy of the spin-up band located below the Fermi level, whereas the higher intensity (spin-down) band is empty. At temperature T , where $0 < T < T_c$, the bands are merging and occupancy of the spin-down band increases. In both cases, nonzero magnetization comes from moments not fully compensated in the lower band. At the Curie temperature (lowest panel), the bands merge and are equally occupied; hence, no effective moment is observed and the system becomes paramagnetic.

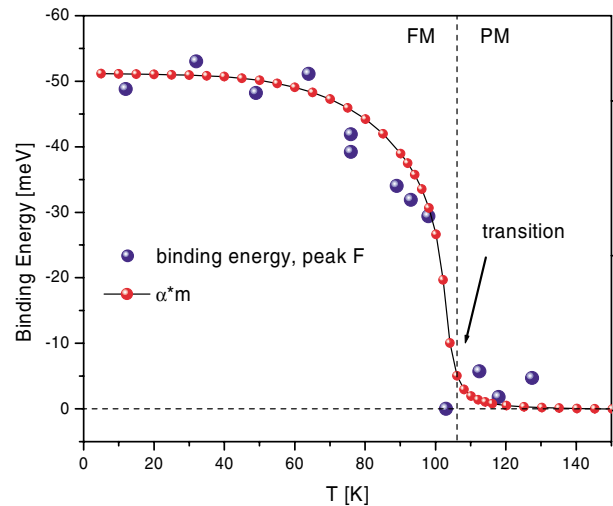


FIG. 3 (color online). The movement of band F towards Fermi level (binding energy = 0) is correlated with magnetization (m). Each blue point represents the band F position determined by fitting the photoemission spectrum at a given temperature, whereas the red points are the magnetization data scaled by a factor $\alpha = 65$ meV/ μ_B . The Curie temperature was determined by fitting a straight line to the high temperature part of the inverse magnetic susceptibility.

universal feature of itinerant ferromagnets. This behavior is a consequence of the Pauli exclusion principle and should be observed in photoemission experiments for other f -electron itinerant ferromagnets. We do acknowledge, however, that success in explaining the correlation between the F photoemission peak and the magnetization does not imply that all the general properties of UTe can be addressed within the mean-field (Stoner-like) theory. As it is explained in [5], the itinerant ferromagnetism of f -electron compounds requires more sophisticated approaches to explain properties such as the nonmonotonic dependence of T_c as a function of pressure [27].

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