The interface between heavy fermions and normal electrons investigated by spatially-resolved nuclear magnetic resonance

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We have studied the superlattices with alternating block layers (BLs) of heavy-fermion superconductor CeCoIn5 and conventional-metal YbCoIn5 by site-selective nuclear magnetic resonance (NMR) spectroscopy, which uniquely offers spatially-resolved dynamical magnetic information. We find that the presence of antiferromagnetic fluctuations is confined to the Ce-BLs, indicating that magnetic degrees of freedom of f-electrons are quenched inside the Yb-BLs. Contrary to simple expectations that the two-dimensionalization enhances fluctuations, we observe that antiferromagnetic fluctuations are rapidly suppressed with decreasing Ce-BL thickness. Moreover, the suppression is more prominent near the interfaces between the BLs. These results imply significant effects of local inversion-symmetry breaking at the interfaces.

The physics of materials with strong electron correlations is remarkably rich, and in these materials the entanglement of charge, spin and orbital degrees of freedom often leads to the emergence of exotic quantum phases. It has been shown that in the presence of strong spin-orbit coupling, the introduction of broken spatial inversion symmetry can produce further notable effects on the electronic properties, even when the global inversion symmetry is preserved in the whole crystals. Moreover, recent advances in fabricating epitaxial superconductors, such as rare-earth and actinide compounds. Recent advances in fabricating epitaxial superlattices consisting of heavy-fermion block layers (BLs) and conventional-metal BLs provide unique opportunity to study the effect of locally broken inversion symmetry at the interfaces between BLs.

CeCoIn5/YbCoIn5 superlattices, several highly unusual superconducting properties have been observed, in particular when the thickness of the Ce-BLs is only a few unit cells. Although the importance of the interfaces between Ce- and Yb-BLs has been emphasized, the lack of spectroscopic information prevents us from understanding their physical properties at the microscopic level. Thus the electronic and magnetic structures at the interfaces remain largely unexplored.

Nuclear magnetic resonance (NMR) appears to be a particularly powerful probe, providing spatially-resolved microscopic information on the magnetic properties. We have performed NMR measurements on CeCoIn5(5)/YbCoIn5(5) superlattices grown by molecular beam epitaxial technique, where n = 5 and 5 layers of CeCoIn5 and 5 layers of YbCoIn5 were stacked alternately as shown in FIG. 1(b). Here the set of CeCoIn5(n) and YbCoIn5(5) repeats 40 times for n = 5 and 30 times for n = 9. Panels of FIG. 1(c) depict the NMR spectra of the n = 9 and n = 5 superlattices for H || c, along with the spectra of CeCoIn5 (Ce) [top] and YbCoIn5 (Yb) [bottom] thin films. The thickness of the Ce and Yb thin films is 500 nm and 350 nm, respectively.

From detailed analysis of the field dependence of the spectra we are able to obtain the site-selective NMR information, i.e. spectroscopic information resolved for Ce and Yb-BLs separately. In the thin films, NMR signals arising from two In sites, the In(1) located at the center of the Ce/Yb-In layer and the In(2) site located on the lateral faces, and the Co site can be clearly identified. The largest principal axis of the electric field gradient is parallel to the c axis at the In(1) and Co sites, while it is perpendicular at the In(2) site. The parameters for the NMR line fitting are listed in the Table 1, along with those of the bulk CeCoIn5.

Here we focus on the spectra arising from the central transition (+1/2 ↔ −1/2) of the In(1) site, which range...
Knight shift $K(T)$ at the fixed frequency $\omega_0$ is defined as

$$K(T) = \frac{(H_0 - H_{\text{res}})}{H_{\text{res}}} = A_{hf}\chi(T)$$

where $H_{\text{res}}$ and $H_0$ are resonant magnetic fields of a sample and a bare nucleus that has the relation of $\omega_0 = \gamma_n H_0$ with the nuclear gyromagnetic ratio $\gamma_n$. $A_{hf}$ and $\chi(T)$ are the hyperfine coupling constant and the local static susceptibility, respectively. The shift from $\mu_0 H_0 = 12.27$ T ($K = 0$) is proportional to the local static susceptibility at the In(1) site.

The nuclear spin-lattice relaxation rate $T_1^{-1}$ provides microscopic information on the dynamical magnetic properties; $(T_1 T)^{-1}$ is proportional to the momentum $q$-summed imaginary part of the dynamical susceptibility, i.e., $(T_1 T)^{-1} \propto \sum_\mathbf{q} A_{hf}\chi''(q, \omega)/\omega$. Figure 2 depicts $(T_1 T)^{-1}$ at the In(1) site of two thin films and superlattices with $n = 5$ and $9$ at $\mu_0 H \sim 12$ T. Although the In(1) site is located at the symmetric position in the unit cell, In(1) sees the AFM fluctuations from rare earth ions since the bond axes of the ordered moments are not coincident with the unit cell. In the Ce thin film, $(T_1 T)^{-1}$ is temperature independent (Korringa relation), which is typical behavior of the uncorrelated nonmagnetic metal. In the Yb thin film, $(T_1 T)^{-1}$ is strongly enhanced at low temperatures, indicating the presence of strong AFM fluctuations. In Fig. 2, dashed line represents $(T_1 T)^{-1}$ of the CeCoIn$_5$ single crystal measured at the same field, indicating that the AFM fluctuations in the Ce thin film are the same as those in bulk single crystals.

Figures 3(a) and (b) show the expanded NMR spectra at $3.2$ K near the In(1) central transition of the $n = 9$ and $n = 5$ superlattices. The NMR spectra of these superlattices contain signals from three different layers, i.e., Ce- and Yb-BLs and CeIn$_3$ buffer layers, each of which has different Knight shift. It should be stressed that the spectra of the Ce and Yb thin films, respectively. The maximum at around $12.22$ T observed in the $n = 5$ and $9$ superlattices and the Yb thin film (asterisk and dashed peaks) arises from the CeIn$_3$ buffer layer. Owing to the zero asymmetric parameter at the In(1) site, central-transition peak (+1/2 $\leftrightarrow$ −1/2) of the In(1) site is not shifted by the electric quadrupole interaction, but shifted by the hyperfine interaction related to the local spin susceptibility.
FIG. 2. (color online). The nuclear spin-lattice relaxation rate divided by temperature \((T_1T)^{-1}\) at the In(1) site in thin-film CeCoIn₅ (magenta squares), thin-film YbCoIn₅ (khaki squares) and CeCoIn₅\(n\)/YbCoIn₅\(5\) superlattices with \(n=9\) (triangles) and \(n=5\) (circles). In the superlattices, \((T_1T)^{-1}\)'s in the Ce-BLs (orange and red) and Yb-BLs (dark and light green) are shown separately. \((T_1T)^{-1}\) in bulk CeCoIn₅ measured at \(\mu_0 H \sim 12.1\) T is also shown with dashed line.

that the spectra from each layer are well separated in this field range. In FIGs. 3(a) and (b), the red, orange, and yellow shaded regions represent the spectra of Ce-BLs, the green region represents the spectra of Yb-BLs and the peak shown by asterisk represent the spectra of CeIn₃ buffer layers. These assignments are made by the straightforward comparison with the spectra of CeCoIn₅ and YbCoIn₅ thin films. A salient feature is that the shape of the spectra of Ce-BLs for \(n=9\) is different from that of \(n=5\). The spectrum in the lower-field region of \(n=9\) has a much larger weight compared with that of \(n=5\). This naturally implies that the spectra in higher field regimes shaded by yellow arise from the outer CeCoIn₅ layers close to the interfaces (FIG. 3(e)), because the fraction of the interface layers increases rapidly with the reduction of \(n\). In FIGs. 3(a) and (b), the red to yellow gradation is a schematic representation of the NMR signal arising from a inner and outer (interface) layers in Ce-BLs. The colors of the spectra correspond to the colors in the Ce-BL in FIG. 3(e). Thus the field dependence of \((T_1T)^{-1}\) enables us to resolve the layer dependence of the magnetic fluctuations even within a same Ce-BLs. The field dependence of \((T_1T)^{-1}\) shown in FIG. 3(c) indicates that even within the Ce-BLs AFM fluctuations have strong spatial dependence; AFM fluctuations near the interface are weaker than those at the inner Ce-layers. This indicates that the suppression of the AFM fluctuations is larger near the interface than in the inner layers. The field dependence of \(1/T_1\) in FIG. 3(c) is clearly recognized from the recovery of the nuclear magnetization \(m(t)\) at a time \(t\) after a saturation pulse(FIG.3 (d)). The recovery \((m(\infty) - m(t))/m(\infty)\) at 12.180 T is more slowly damped than the recovery at 12.115 T.

The temperature dependence of \((T_1T)^{-1}\) of the Ce- and Yb-BLs in each superlattice is plotted separately in FIG. 2. Here the relaxation rate of Ce-BLs is determined at the maximum of the broad peak, which represents an average of the AFM fluctuations in the whole Ce-BLs. It is clear that while relaxation rate in the Yb-BLs is essentially unchanged, but the magnitude of \((T_1T)^{-1}\) in the Ce-BLs is suppressed as the Ce-BLs thickness is reduced. We analyze \((T_1T)^{-1}\) in terms of the Curie-Weiss (CW) formula \((\mu_0 H)^{-1} \propto C/(T + \theta)\), assuming that dynamical susceptibility is dominated by the AFM fluctuations. Here \(\theta\) is the Weiss temperature and \(C\) is related to the Curie constant. The experimental data can be fitted by the CW formula as shown in FIG. 4. The reduction of \(n\) leads to a serious reduction of \(C\), indicating that the AFM fluctuations are suppressed as the Ce-BLs become thinner. We emphasize that the proximity of \(f\)-electrons with magnetic moment to nonmagnetic Yb-layers is unlikely to be the origin of this reduction, because of the following reasons. First, magnetic fluctuations in the Yb-BLs are essentially the same as those in the YbCoIn₅ thin
face into two sheets with different spin structures, where $\alpha$ two-dimensional plane, $\nabla\theta$ parallel to the Fermi level. The spatial dependence of the relaxation rate within the Ce-BLs [FIG. 3(c)] strongly suggests that the breaking of the local inversion symmetry at the interface reduces the AFM fluctuations through the helical anisotropy of the spin configuration shown in FIG. 3(e).\(^{22,23}\) In fact, similar situation has been reported in ferromagnetic metal MnSi, where helical spin structure is stabilized by the spin-orbit coupling.\(^{24}\) With the reduction of $n$, the fraction of the noncentrosymmetric interface layers increases rapidly, leading to the suppression of the AFM fluctuations. The present results suggest that the local inversion symmetry breaking plays a key role for the magnetic properties at the interface of strongly correlated electron systems, which is expected to host a fertile ground for observing exotic properties.\(^{25}\)

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FIG. 4. (color online). Plot of $T_\parallel$ against $T$ in the $n = 9$ and $n = 5$ superlattices compared with the result of pure CeCoIn$_5$ films. The data can be fitted by the Curie-Weiss law [$T_\parallel = (T + \theta)/C$].

The observed suppression of the magnetic fluctuations in the Ce-BLs is opposite to the enhancement of fluctuations expected from two-dimensionalization, which leads to the enhancement of the density of states at the Fermi level. The spatial dependence of the relaxation rate within the Ce-BLs [FIG. 3(c)] strongly suggests that the breaking of the local inversion symmetry at the interfaces between Ce- and Yb-BLs plays a decisive role for determining the magnetic properties of the Ce-BLs, in particular when their thickness is only a few unit-cell thick. In the absence of inversion symmetry, an asymmetric potential gradient $\nabla V$ yields a spin-orbit interaction. When $\nabla V$ is perpendicular to the two-dimensional plane, $\nabla V \parallel c$, Rashba spin-orbit interaction $\alpha_R g(k) \cdot \sigma \propto (k \times \nabla V) \cdot \sigma$ splits the Fermi surface into two sheets with different spin structures, where $g(k) = (-k_y, k_x, 0)/k_F$, $k_F$ is the Fermi wave number, and $\sigma$ is the Pauli matrix. The energy splitting is given by $\alpha_R$, and this interaction locks spin configurations within the $ab$ plane with clockwise rotation on one sheet and anticlockwise on the other, as shown in FIG. 3(e).\(^{21\text{st}}\)

Here, the inversion symmetry is locally broken at the top and the bottom layers of the Ce-BLs at the immediate proximity to the Yb-BLs. In the presence of the local inversion symmetry breaking together with the fact that the Ce atom has a large atomic number, the Rashba-type spin-orbit coupling is expected to be strong in the present superlattices. The importance of the local inversion symmetry breaking at the interface has been emphasized experimentally through the peculiar angular variation of upper critical field, which can be interpreted as a strong suppression of the Pauli pair-breaking effect.\(^{15}\) The Fermi surface splitting due to the Rashba coupling should modify seriously the nesting condition and hence is expected to reduce the commensurate AFM fluctuations with $Q = (\pi/a, \pi/a, \pi/c)$, which is dominant in bulk CeCoIn$_5$. In addition, it has been pointed out that the broken inversion symmetry at the interface reduces the AFM fluctuations by lifting the degeneracy of the fluctuation modes through the helical anisotropy of the spin configuration shown in FIG. 3(e).\(^{22,23}\) In fact, similar situation has been reported in ferromagnetic metal MnSi, where helical spin structure is stabilized by the spin-orbit coupling.\(^{24}\) With the reduction of $n$, the fraction of the noncentrosymmetric interface layers increases rapidly, leading to the suppression of the AFM fluctuations. The present results suggest that the local inversion symmetry breaking plays a key role for the magnetic properties at the interface of strongly correlated electron systems, which is expected to host a fertile ground for observing exotic properties.\(^{25}\)
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