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Tunneling magnetoresistance studies of $Sr_3Ru_2O_7$

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We have performed planar tunneling measurements on $Sr_3Ru_2O_7$, which has a field-tuned metamagnetic quantum phase transition. Our previously reported work revealed an unusual oscillation in the tunneling magnetoresistance of junctions made on $Sr_3Ru_2O_7$ single crystals [J. Hooper *et al.*, Phys. Rev. Lett. **92**, 257206 (2004)]. We here report further characterization of this intriguing phenomenon, revealing that the oscillation has a systematic dependence on the tunnel barrier, field orientation, and temperature. We discuss possible origins of this unusual oscillation phenomenon in light of these new experimental results.

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I. INTRODUCTION

Considerable study has been devoted to quantum phase transitions (QPTs), since proximity to a QPT often results in non-Fermi liquid behavior and the formation of novel ground states, including unconventional superconductivity.[1–4] QPTs in most studied materials result from depressing the characteristic temperature of a second-order phase transition to zero temperature by tuning external parameters such as pressure, chemical doping, and magnetic field.[5–9] $Sr_3Ru_2O_7$, the material studied in this paper, was recently found to show a new kind of quantum criticality,[10,11] which occurs when a critical end point terminating a line of first-order metamagnetic phase transitions in the temperature-field phase diagram is suppressed to zero temperature. This finding has generated a great deal of interest within the field of quantum criticality because of its unique features. Since the tuning parameter for this QPT is a magnetic field, which is much more convenient experimentally than using pressure or chemical doping, it enables many more experimental techniques for systematic investigation of this new phenomenon.

$Sr_3Ru_2O_7$ is the bilayer member of the Ruddlesden-Popper series of ruthenates $Sr_{n+1}Ru_nO_{3n+1}$. Its ground state is a Fermi liquid with strong electronic correlations and an exchange-enhanced paramagnetism.[12] It undergoes the metamagnetic transition under moderate magnetic fields with the transition field ranging from 4.9 T (for field parallel to the *ab* plane) to 7.9 T (for field parallel to *c*).[10,11] Strong evidence for critical fluctuations near this metamagnetic transition have been observed in experiments.[10,11,13,14] A new ordered nonsuperconducting phase has been observed very near to the quantum critical end point.[15,16] These fascinating features open new routes to explore the novel physics associated with quantum criticality.

To further study the physical properties in the vicinity of this metamagnetic QPT, we previously measured the magnetoresistance of planar tunneling junctions fabricated on single crystals of $Sr_3Ru_2O_7$.[17] We found that the tunneling magnetoresistance showed an unusual oscillation under applied magnetic fields $H \| c$, with the amplitude of the oscillation increasing significantly above a field of 5 T. Our analysis indicated that this oscillation is not due to the Shubnikov-de Haas (SdH) effect, and thus its origin must be something other than Landau levels crossing the Fermi surface. We proposed that this oscillation might be influenced by the metamagnetic quantum fluctuations.[17] Here we report further characterization of this new phenomenon, in particular its dependence on the tunnel barrier, field orientation, and the temperature. Our results indicate that this oscillation is a very robust and reproducible feature, with a systematic dependence on external parameters. We further discuss its possible origins in terms of these new results.

II. EXPERIMENT

Single crystals used for these tunneling measurements were grown in floating-zone furnaces at Kyoto and Tulane using the method described by Perry and Maeno.[18] The residual resistivities of these samples were typically on the order of ~3 $\mu\Omega$ cm. In the following, we present data from batches C6J10 and C6J06 grown at Kyoto, although the crystal grown at Tulane exhibited the same behavior. Junctions were fabricated by the method previously described.[17] A thin tunnel barrier (typically ~60 Å of Al_2O_3 or SiO) was evaporated on cleaved (001) faces or finely polished surfaces parallel to the *c* axis. A gold counterelectrode (~500 Å) was then evaporated on top of the tunnel barrier. The Al_2O_3 layer was typically prepared by three sequential evaporations of 20 Å of Al in high vacuum, each followed by oxidation in air. SiO junctions were prepared by a direct evaporation of 60 Å of SiO. Typical junction surface area was on the order of 1 mm × 1 mm. Tunneling measurements were made using a four-point method, with two wires attached to the crystal and two wires attached to the counterelectrode. All measurements were taken in a ^{3}He cryostat equipped with a 9-T superconducting magnet.

III. RESULTS AND DISCUSSION

We have made more than thirty Al_2O_3 junctions. Their resistances were found to be very different from junction to

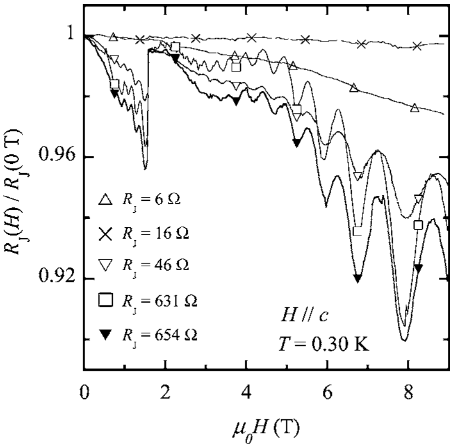

FIG. 1. Normalized tunneling magnetoresistance in the field range 0–9 T for several junctions with different tunnel barrier strengths. Junctions with R_J=6, 16, and 46 Ω were made on polished (100) surfaces; junctions with R_J=631 and 654 Ω were made on cleaved (001) surfaces. Measurements were taken in the configuration $H \| c$ at T=0.3 K. All data are normalized to the value of the junction resistance at 0 T.

junction. The junction resistance at 0.3 K under zero magnetic field ranged from ~6 Ω to 1200 Ω for the junctions measured in these experiments. This variation in junction resistance can be attributed to the surface roughness of the sample. Polished surfaces usually had a roughness of ~1–3 μm; for the cleaved surface, the roughness is mainly due to terraces, which are typically on the order of a few micrometers in height. This surface roughness would cause the tunnel barrier to be inhomogeneous at the junction interface, which could in turn result in the variation of effective tunneling areas and barrier strength (see below for more discussions).

Figure 1 shows the normalized junction resistance as a function of magnetic field applied along the *c* axis at 0.3 K for five different Al_2O_3 junctions with zero field resistance R_J=6, 16, 46, 631, and 654 Ω. Although the junctions with R_J=6, 16, and 46 Ω are all prepared on polished faces parallel to the *c* axis and have comparable nominal junction areas, they exhibit very different behavior under magnetic field. The junction with R_J=46 Ω displays remarkable oscillations, while the junction with R_J=16 Ω shows only a minimal oscillation pattern above 5 T and the junction with R_J =6 Ω shows no oscillations at all. The value of junction resistance normalized to the effective tunneling area reflects the tunnel barrier strength for a given tunnel junction. Since these three junctions are expected to have comparable effective tunneling areas (due to similar preparation), the systematic reduction of the oscillation with junction resistance suggests that the oscillation feature is dependent on tunnel barrier strength; specifically, the oscillation appears to occur only above a certain barrier strength.

The junctions with R_J=631 and 654 Ω [made on cleaved (001) surfaces] shown in Fig. 1 display similar oscillation patterns as the junction with R_J=46 Ω (made on the polished surface), but have a much greater amplitude. Given that $Sr_3Ru_2O_7$ has a layered quasi-two-dimensional crystal structure, it is reasonable to question why this oscillation does not appear to depend on crystal orientation. This might be interpreted in the following way. Although these junctions are nominally made on the (001) face, in-plane tunneling might be heavily involved due to the existence of terraces on the cleaved surface. If the tunneling is extremely anisotropic along different crystallographic directions and the in-plane direction has a much higher tunneling probability than the *c* axis, the in-plane tunneling through the terrace edges could dominate the transport properties of a junction made on a cleaved surface. This has been proven to be possible experimentally; for example, in the spin-triplet superconductor Sr_2RuO_4, a zero-bias conductance peak corresponding to Andreev surface bound states is expected only for in-plane tunneling, but is also observed in *c*-axis cleaved junctions.[19] Therefore it is reasonable to assume that our *c*-axis junctions made on cleaved (001) faces may be dominated by in-plane tunneling, and thus it is not surprising to observe similar oscillations in these two types of junctions.

Though the interface area of all junctions is nominally 1 mm × 1 mm, the actual effective tunneling area should be much smaller for both types of junctions, due to surface roughness. For junctions made on a cleaved (001) face, tunneling most likely takes place primarily via the terrace edges as discussed above, and the effective tunneling area of these terraces should be extremely small. For in-plane junctions on a polished face, the effective tunneling area could be extremely small as well since the polished surface may have roughness. This could result in a very inhomogeneous distribution of tunnel barrier with some small regions having a much lower barrier strength. Tunneling would primarily occur through such regions of the junction in this circumstance. If there is considerable surface roughness on the polished junctions, we can expect that both types of junctions might have a comparable effective tunneling area. Here, if we assume that the junctions made on a cleaved face shown in Fig. 1 have a comparable effective area as those in-plane junctions made on polished faces, the larger resistance of these junctions would suggest that their tunnel barrier strength per unit effective tunneling area should be much higher compared to the other three lower-resistance junctions on polished surfaces. Thus the observation of larger oscillation amplitudes in these (001) junctions, therefore, is consistent with our above argument that a higher tunnel barrier enhances the oscillations. Figure 1 also shows that increasing tunnel barrier strength causes the kink in the background at about 5 T to become much more remarkable. In addition, the smaller amplitude oscillations around 1 T, below the discontinuous jump at ~1.58 T, were also found to change systematically with the tunnel barrier; they become unobservable when the junction resistance is less than 16 Ω, similar to the behavior of the larger amplitude oscillations seen at high fields.

To investigate the field orientation dependence of the oscillation, we performed tunneling measurements in the configuration $H \| ab$ using junctions prepared on cleaved (001) surfaces. We observed different oscillation patterns for this configuration. Figure 2 shows the experimental data obtained on eight different Al_2O_3 junctions. The oscillations in this field configuration begin to develop at 4 T where the background shows a kink, and there are usually only four or five

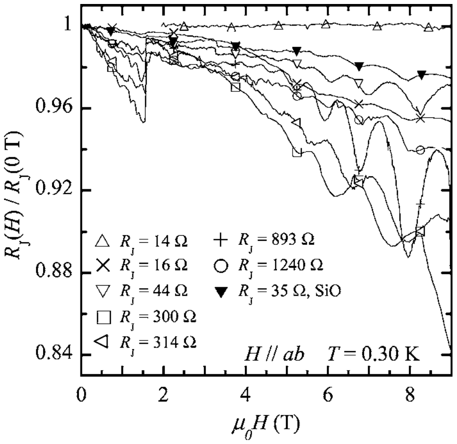

FIG. 2. Normalized tunneling magnetoresistance in the field range 0–9 T for several junctions [made on cleaved (001) surfaces] with different tunnel barrier strengths. For the junction with $R_J = 14\ \Omega$, one lead broke during measurement, so the data below 2 T for this sample is unavailable. Measurements were taken in the configuration $H \| ab$ at $T = 0.3$ K. All data are normalized to the value of the junction resistance at 2 T.

oscillation peaks observed below 9 T. This is in sharp contrast to the situation for $H \| c$, where the oscillation develops at 3 T, enhances significantly in amplitude above 5 T, and there are typically nine or ten oscillation peaks below 9 T. In the lower field region <2 T for $H \| ab$, we also observed the discontinuous jump at 1.58 T, but the small-amplitude oscillations around 1 T exhibit very different features than for $H \| c$.

For further comparison of the larger-amplitude oscillations at higher fields between these two field orientations, we plot the oscillation patterns with the background subtracted on the scale of $1/\mu_0 H$ in Fig. 3 for two typical junctions measured under $H \| ab$ and $H \| c$. We found that the oscillation frequency depends on the field range for both orientations. In our previous report,[17] we performed Fourier transform analysis of junction resistance versus $\mu_0 H$ with $H \| c$ for different field ranges, which yielded a frequency of 50 T^{-1} for the 3–5 T range and a frequency of 0.7 T^{-1} for the 5–9 T range.[27] However, since so few oscillations are included in the field range, it is unclear if any physical significance can be attributed to this analysis. Here, in order to allow comparison with the quantum oscillations reported for bulk samples, we roughly estimated the frequency of junction resistance versus $1/\mu_0 H$ in the field range 2–9 T by directly measuring and averaging the oscillation periods shown in Fig. 3. Although the oscillations are not precisely periodic in the inverse field, we estimate frequencies of ~44 T for $H \| c$ and ~32 T for $H \| ab$, an order of magnitude smaller than the lowest frequencies seen in de Haas-van Alphen and Shubnikov-de Haas measurements.[13,20]

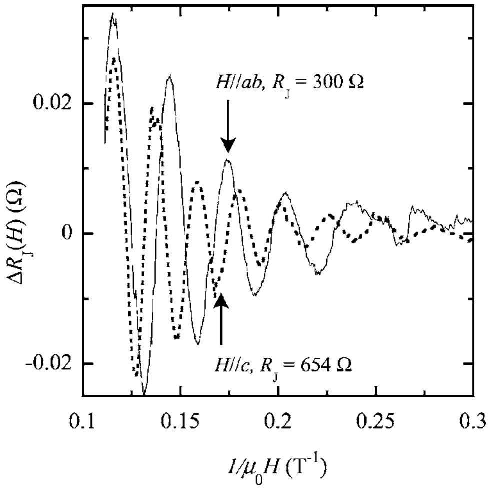

FIG. 3. Tunneling magnetoresistance versus reciprocal field. The background has been subtracted, leaving only the oscillation pattern. Data is shown for two junctions with $R_J = 300$ and 654 Ω made on cleaved (001) surfaces, one with $H \| ab$ (solid line) and the other with $H \| c$ (dashed line).

As seen in Fig. 2, the oscillation maxima and minima for $H \| ab$ are junction dependent, suggesting a variation of oscillation frequency. Such a variation presumably reflects the in-plane anisotropy of the Fermi surface (FS) of $Sr_3Ru_2O_7$. As discussed above, the in-plane tunneling through the edge of terraces likely dominates the transport properties on these junctions. The random crystalline orientation on the edge of terraces could result in different orientations of the magnetic field relative to the tunneling direction among different junctions, which would possibly allow the tunneling to probe different portions of the Fermi surface. Band structure calculations[21] revealed that the FS of $Sr_3Ru_2O_7$ is much more complicated than Sr_2RuO_4. The yz and xz bands split pairwise into even and odd combinations due to interlayer coupling; the zone folding due to orthorhombicity yields small cylindrical lens-shaped FSs. Such complexity in band structure makes it difficult for us to make direct comparisons between our observations and theoretical results.

From Fig. 2, we also observed a similar dependence of the oscillation on tunnel barrier as seen in Fig. 1 for junctions with resistance less than 1000 Ω; specifically, the oscillation amplitude enhances with an increase of tunnel barrier. However, for the junction with $R_J = 1240\ \Omega$, the oscillation is dramatically suppressed compared to the junction with $R_J = 893\ \Omega$. This implies that an upper limit in barrier strength may exist, above which the oscillation is depressed with a further increase in tunnel barrier strength.

In addition, it is worthwhile to note that the junctions with $R_J = 300$ and 314 Ω in Fig. 2 and the junctions with $R_J = 631$ and 654 Ω in Fig. 1 are actually two identical junctions measured in two separate coolings. In the first experiment with $H \| ab$, the resistances of these two junctions at zero field were 300 and 314 Ω, respectively, while in the second cooling with $H \| c$, their junction resistances increased up to 631 and 654 Ω. Although this dramatic increase in junction resistance suggests that the junctions had changed relative to the first experiment, the similarity in oscillation patterns between these two junctions and the junction with $R_J = 46\ \Omega$ for $H \| c$ (see Fig. 1) provides further support for our argument that the oscillation does depend on field orientation. The most direct way to probe the field orientation dependence of the oscillation is to perform measurements on the same junction during a single cooling using a rotator. However, such measurements cannot be made currently with our cryostat since it is not equipped with a rotator.

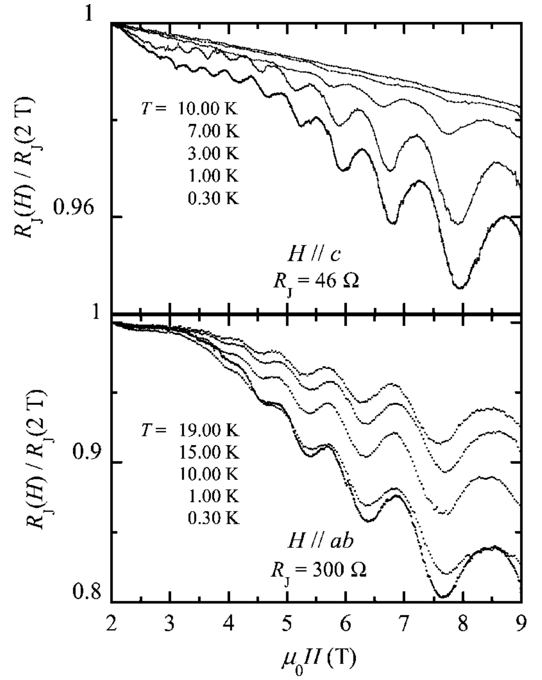

FIG. 4. Temperature dependence of normalized tunneling magnetoresistance for two typical junctions, (a) made on a polished face parallel to the c axis with field applied $H \| c$ and (b) made on a cleaved (001) face with field applied $H \| ab$. All data are normalized to 2 T.

Also in Fig. 2, we have included the tunneling magnetoresistance data for a typical junction whose tunnel barrier is composed of SiO. A similar oscillation phenomenon is seen in this junction. This observation excludes the possibility that the oscillation behavior we observed in the tunneling is an artifact of the specific material used in creating the tunneling barrier. Experimentally, however, Al_2O_3 is a better choice for fabricating junctions with higher tunnel barriers.

Figure 4 shows the temperature dependence of the oscillations measured on two typical junctions. For $H \| c$, the oscillation is suppressed as temperature is increased up to 7 K, and the oscillation maxima and minima are temperature dependent. However, for $H \| ab$, the oscillation amplitude diminishes more slowly with increasing temperature and is still quite prominent up to 19 K (the highest available temperature in our ^{3}He cryostat); the positions of oscillation maxima and minima are less sensitive to temperature compared to the situation for $H \| c$. These distinct differences again suggest that the observed oscillation in tunneling depends on the field orientation.

Overall, based on the experimental data presented above, the oscillation of tunneling magnetoresistance in $Sr_3Ru_2O_7$ is a very robust and reproducible phenomenon, with a systematic dependence on external parameters such as junction resistance, field orientation, and temperature. Next, we consider the possible origin of this unusual oscillation behavior. The analysis in our previous report has indicated that this oscillation is unrelated to traditional quantum oscillations caused by orbit quantization (the SdH effect).[17] Our current experimental observations, especially the systematic barrier strength dependence of the oscillation, further confirms that our oscillation is truly from the tunneling process. The fact that we did not observe any oscillation features in junctions whose zero-field resistance is below ~16 Ω for both $H \| ab$ and $H \| c$ is probably because the barrier between $Sr_3Ru_2O_7$ and the counterelectrode is not in the tunneling regime. The entire junction in this case is likely shorted by a small area where the tunnel barrier is extremely low; the electronic transport through this area may behave more like the transport in a dirty metallic region rather than tunneling. The observation of constant or slightly decreasing tunneling magnetoresistance in these junctions, in sharp contrast with high resistance junctions which show a large negative magnetoresistance, suggests that this is a very possible scenario.

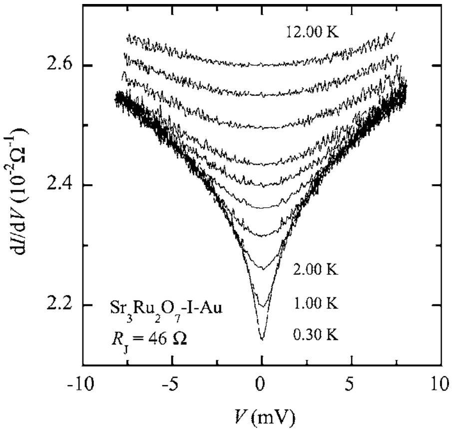

FIG. 5. Tunneling conductance versus bias voltage for a typical $Sr_3Ru_2O_7$ junction made on a polished (100) surface. Measurements are shown for T=0.30, 1.00, 2.00, 3.00, 4.00, 5.00, 6.00, 8.00, 10.00, and 12.00 K.

Ordinarily, for tunneling between a nonsuperconducting metal such as $Sr_3Ru_2O_7$ and a counterelectrode, the tunneling resistance is inversely proportional to the density of states (DOS) at the FS, as discussed in our earlier report.[17] The decreasing background trend in R_J with increasing applied field as shown in Figs. 1 and 2 would thus indicate an increasing DOS as the field is ramped up, which is consistent with the current theoretical picture of $Sr_3Ru_2O_7$.[11,15,22] However, since no corresponding oscillation feature has been observed in any bulk measurement of $Sr_3Ru_2O_7$, it is difficult to understand the oscillatory behavior in R_J as being directly due to an oscillation of DOS.

Since tunneling is a surface probe technique, is it possible that this oscillation phenomenon originates from an unusual surface electronic state? We think this possibility cannot be excluded, for the following reasons. First, a surface electronic state which differs from that of the bulk has already been observed in the related compound Sr_2RuO_4. The tunneling spectrum dI/dV of Sr_2RuO_4 exhibits a sharp cusp feature around zero bias, suggesting a suppression of DOS at the FS.[23] This cusp feature was found to be related to a surface normal layer caused by surface structure reconstruction.[23,24] For $Sr_3Ru_2O_7$, we observed a similar feature in tunneling spectra for all the junctions we measured; shown in Fig. 5 is the spectrum obtained on a typical junction. In our previous discussion, we suggested that this cusp feature might be related to antiferromagnetic correlations in the bulk. However, considering the similarity to Sr_2RuO_4 in terms of the tunneling spectra, we certainly cannot exclude the existence of an unusual surface electronic state in $Sr_3Ru_2O_7$. Secondly, as shown in Fig. 1, we observed a very remarkable barrier-strength dependent discontinuous jump at about 1.58 T in the tunneling magnetoresistance, implying a sharp change in the FS. However, no anomaly was observed around this field in bulk resistivity, magnetization, or specific heat measurements.[10,14] This appears to be another indication of a surface state. All other results in our current experiments are not incompatible with the idea of an unusual surface electronic state. The differences in the oscillation patterns and temperature dependences between $H\|ab$ and $H\|c$ mirrors the anisotropy of the FS of $Sr_3Ru_2O_7$, but any surface state might also have an anisotropy similar to that of the bulk material.

Since the physical properties of $Sr_3Ru_2O_7$ are strongly affected by the metamagnetic quantum criticality, a very natural question is whether this oscillation is associated with critical fluctuations in the quantum critical regime. Our previous measurements on tunneling spectra as a function of magnetic field for $H\|c$ revealed that the FS changes prominently starting from 5 T and then sharply around the metamagnetic transition field ~8 T.[17] The fact that the significant enhancement in oscillation amplitude also takes place above 5 T appears to suggest that the larger-amplitude oscillation near the critical field is related to critical fluctuations. Our current data obtained on junctions with higher tunnel barriers for $H\|c$ (see Fig. 1) seems to provide further support for this view; in these junctions, the oscillation amplitude enhances significantly above 5 T, the point at which the kink of the background becomes more remarkable as well.

For $H\|ab$, the oscillation develops near the metamagnetic transition field of ~5 T and enhances gradually with increasing field; in contrast to $H\|c$, here the most prominent oscillation is seen at fields well above the transition point. This does not seem to be consistent with the above argument. Furthermore, we observe oscillations in the low-field range 0–2 T for both field configurations, a region which should not be strongly affected by the quantum critical point based on the phase diagram derived from bulk measurements.[11,16] However, if we assume that the surface electronic state differs from that of bulk as discussed above, we do not necessarily expect properties probed by tunneling to be fully analogous to bulk properties. The metamagnetic transition may survive on the surface with modified features. To clarify this issue, further investigations on the surface electronic state of $Sr_3Ru_2O_7$ are highly desirable.

Finally, we will discuss other possible scenarios which might be associated with the oscillations observed in our experiment. Since $Sr_3Ru_2O_7$ is a strongly correlated system,[12] tunneling should reveal the DOS renormalized by many-body effects, not the single-particle DOS. As the system approaches the metamagnetic transition, quantum critical fluctuations may result in multiple electron-correlation mechanisms, which would vary the degrees of quasiparticle renormalization. This would result in a change in the renormalized DOS with the magnetic field. Additionally, we know that the quasiparticle DOS is related to the tunneling conductance via the tunneling matrix element H_T^2. In general, H_T^2 is assumed to be constant and can be removed from the tunneling integral, as shown in Eq. (1) in Ref. 17. Nevertheless, in the vicinity of the metamagnetic transition of $Sr_3Ru_2O_7$, H_T^2 might become energy dependent due to quantum criticality and cannot be factored out of the integral. It can also change with the magnetic field. Under this circumstance, the oscillation in tunneling resistance does not merely indicate an oscillation of the quasiparticle DOS. These issues have been discussed theoretically in the literature.[25,26]

IV. CONCLUSION

We have investigated the tunneling magnetoresistance of $Sr_3Ru_2O_7$ using various junctions prepared on cleaved (001) surfaces and polished surfaces parallel to the c axis of $Sr_3Ru_2O_7$ single crystals. Our experimental results reveal that the unusual oscillation phenomenon observed in tunneling magnetoresistance in our previous work is a robust and highly reproducible feature, with a systematic dependence on external parameters. The dependence of these oscillations on tunnel barrier strength further confirms our previous argument that this oscillation is truly from tunneling; increasing the barrier strength (up to a certain limit) enhances the amplitude of the oscillation feature. The differences in the oscillation patterns and temperature dependences between $H\|ab$ and $H\|c$ mirrors the anisotropy of the FS of $Sr_3Ru_2O_7$. The variation of oscillation frequency observed on junctions prepared on various cleaved (001) surfaces further suggests that the FS of $Sr_3Ru_2O_7$ should have an in-plane anisotropy.

Although the current experimental data are not sufficient for us to make a definitive conclusion on the origin of this oscillation, we propose that it may originate from an unusual surface electronic state. Further work is certainly desirable to resolve this issue. Regardless of the precise mechanism that may be responsible for this new oscillation phenomenon, our experimental observations suggest that magnetotunneling might be a sensitive probe of the Fermi surface, especially for materials with magnetic phase transitions.

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[27] In our previous report (Ref. 17), the unit of oscillation frequency obtained by Fourier transform analysis versus $\mu_0 H$ was incorrectly listed as T. However, the discussions related to this frequency given there are still approximately valid since the oscillation frequency obtained from a Fourier analysis of R_J versus $1/\mu_0 H$ is also very small, below 50 T.