Quantum Oscillations and Overcritical Torque Interaction in Sr$_2$RuO$_4$

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Sr$_2$RuO$_4$ is the only known layered perovskite oxide superconductor without copper; there is strong evidence for an unconventional (“p-wave”) pairing mechanism, and it has recently been shown to possess a cylindrical, “quasi-two-dimensional” Fermi surface. Using Sr$_2$RuO$_4$ as a test case for the detection of quantum oscillations with piezoactive microcantilevers, the piezovector torque magnetometry technique was successfully implemented on a dilution refrigerator. It was possible to reproduce the quantum-oscillatory magnetization data on all three Fermi surface sheets, in a crystal of microgram mass. Moreover, an absolute estimate of the amplitude of the oscillation is provided. We also investigated the phenomenon of torque interaction which distorts the magnetization signal and introduces harmonics and line-sources to the dHvA spectrum. As the torque interaction effect grows in strength, overcriticality is shown to lead to discrete magnetization jumps and to a near-asymptotically damped “sproing” effect.

I. INTRODUCTION

Recent discoveries of unconventional electronic behavior — such as high-$T_c$ superconductivity in the cuprates or colossal magneto-resistance in the manganates — have focused scientific interest on the transition metal oxides. To understand the properties of metallic oxides, it is important to probe the validity of the Fermi liquid picture and, if possible, to infer the shape of the Fermi surface and other quasiparticle properties. Measurement of the quantum-oscillatory part of the magnetization — the de Haas-van Alphen (dHvA) effect — is the most direct way to obtain such information. However, these investigations are difficult as the materials in question are frequently non-stoichiometric and are often only available as very small crystallites.

In particular, Sr$_2$RuO$_4$ has aroused great interest as the only known superconductor with the same (layered perovskite) crystal structure as the high-$T_c$ cuprates but without copper.

Moreover, it represents a possible candidate for p-wave superconductivity. Sr$_2$RuO$_4$ is one of the very few complex materials for which there is a good prospect that a deeper physical understanding will soon be achieved. High-purity stoichiometric crystals can be prepared, and its normal state Fermi surface is known to be “quasi-2D”, consisting of three slightly warped cylindrical sheets.

Here, we reexamine the dHvA effect in Sr$_2$RuO$_4$ using piezovector torque magnetometry: a new and ultra-sensitive technique which is especially suited for small microcrystals of anisotropic materials. It was possible to obtain absolute estimates for the quantum-oscillatory magnetization on all three Fermi surface sheets. The torque interaction effect and its remarkable manifestation in the “overcritical” case will also be discussed.

II. PIEZOLEVER TORQUE MAGNETOMETRY

The fundamental frequency of the quantum-oscillatory torque density (magnetic torque per unit volume) corresponding to a Fermi surface cylinder in a quasi-2D metal

\[ \tilde{\tau} = \frac{e^2 h_B Z F \sin \theta}{2 \pi^3 m^*} R \sin \left( \frac{2 \pi F}{B \cos \theta} \right) \]

(1)

where $B$ is the magnetic field which is applied at an angle $\theta$ to the low conductivity axis, $h_B$ is the Brillouin zone height (i.e. the length of the cylinder), $m^*$ the on-axis quasiparticle mass, and $F$ the dHvA frequency corresponding to the on-axis Fermi cylinder cross-section $A_F$ via $F = h A_F / 2 \pi e$. The number $R$ represents the damping factors related to finite temperature, sample inhomogeneity, spin-splitting, and the warping of the cylinder.

To record this small torque, we have employed piezoactive microcantilever torque magnetometry as shown in Fig. 1. This technique was pioneered only recently and we successfully implemented it at low temperatures and achieved quantum-oscillatory moment sensitivities of $10^{-12}$ Am$^2$ at 150 mK in a 17 T field.
FIG. 1. Piezolever torque magnetometry: an applied field $B$ induces a magnetic moment $m$, and the magnetic torque $T = m \wedge B$ in an anisotropic crystal flexes the cantilever on which the sample is mounted. This deflection is sensed via the piezoresistance of a resistive path implanted on the lever.

A small high-quality Sr$_2$RuO$_4$ crystal (200 × 150 × 20 µm$^3$, residual resistivity about 0.6 µΩ cm), prepared in a floating zone image furnace, was mounted on a piezolever of dimensions 170 × 50 × 5 µm$^3$. A second (empty) piezolever was used for background compensation in a Wheatstone bridge circuit, driven by a 50 nA amplitude AC current through each lever. Temperatures of 150 mK were achieved on a dilution cryostat during field sweeps from 18 T to 15 T.

The output signal and its dHvA spectrum for a sweep close ($\theta \simeq 2^\circ$) to the $c$-axis is shown in Fig. 2. All dHvA frequencies observed in the original study (which employed the standard field modulation technique) were reproduced with piezolever torque magnetometry.

One can extract the absolute quantum-oscillatory magnetization, and we estimate 460 A/m, 200 A/m, and 30 A/m, respectively, as the dHvA amplitudes (at $T \simeq 0$, $B \simeq 17$ T, and $\theta \simeq 0^\circ$) for the $\alpha$, $\beta$, and $\gamma$ sheets — to within a factor of two. These values are in agreement with Eq. (1).

III. TORQUE INTERACTION

For off-axis fields, the magnetization signal is affected by the torque interaction effect which is due to the feedback of the oscillating magnetic moment on the position of the lever in the field. As a result, the apparent magnetization profile gets sheared, and harmonics and sidebands are introduced to the dHvA spectrum. Fortunately, this effect can be compensated through numerical treatment of the experimental data. For large $\theta$ and high fields, however, the effect becomes strong enough (“overcritical”) to produce discrete jumps in the magnetization and hence in the position of the lever, see Fig. 3.

Although observed previously in arsenic, this remarkably macroscopic manifestation of an inherently quantum phenomenon was visible in our experiments at unprecedented strength. The “sproing” effect at the magnetization jumps — when the lever snaps into its new equilibrium position — was also recorded with a high-speed voltmeter: the lever movement turned out to be near-asymptotically damped by eddy currents. This casts some doubt on the practicability of proposed “dynamic mode” piezolever measurements at least on metallic samples.

IV. CONCLUSION

In conclusion, piezolever torque magnetometry measurements have been able to reproduce the quantum-oscillatory magnetization data on all three Fermi surface sheets of the layered perovskite oxide superconductor Sr$_2$RuO$_4$. Moreover, they provide an absolute estimate of the amplitude of the oscillation. This introduces the piezolever technique as an interesting alternative to conventional methods for dHvA experiments on anisotropic compounds if these are only available as microcrystals—a common situation for complex modern materials.

We also investigated the phenomenon of torque interaction which distorts the magnetization signal and introduces harmonics and sidebands to the dHvA spectrum. As torque interaction grows in strength, it leads to irreversibility effects and discrete magnetization jumps: a near-asymptotically damped “sproing” effect.

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FIG. 3. Experimental magnetization curve recorded on Sr$_2$RuO$_4$, with the magnetic field pointing 48° from the c-axis of the crystal. Torque interaction shears the sinusoidal profile, and this effect gets progressively more severe as the field is swept higher. Above about 12.5 T, the effect becomes “overcritical”, with different values of the magnetization during field upsweeps (grey) and downsweeps (black), and discrete magnetization jumps can be observed. The arrows refer to the sweep direction.

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