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Effects of Rattling Phonons on the Dynamics of Quasiparticle Excitation in the \( \beta \)-Pyrochlore KO\( \text{S}_2\)O\( \text{6} \) Superconductor


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Microwave penetration depth \( \lambda \) and surface resistance at 27 GHz are measured in high quality crystals of KO\( \text{S}_2\)O\( \text{6} \). Firm evidence for fully gapped superconductivity is provided from \( \lambda(T) \). Below the second transition at \( T_p \approx 8 \text{ K} \), the superfluid density shows a steplike change with a suppression of effective critical temperature \( T_c \). Concurrently, the extracted quasiparticle scattering time shows a steep enhancement, indicating a strong coupling between the anomalous rattling motion of K ions and quasiparticles. The results imply that the rattling phonons help to enhance superconductivity, and that K sites freeze to an ordered state with long quasiparticle mean free path below \( T_p \).

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Recently, the roles of phonons have become refocused on a plethora of novel physical properties in strongly correlated electron systems, such as their interplay with superconductivity in high-\( T_c \) cuprates [1], and the heavy-fermion behavior in filled-skutterudites [2] possibly due to unconventional motions of ions [3]. In the \( \beta \)-pyrochlore superconductor KO\( \text{S}_2\)O\( \text{6} \) with a relatively high \( T_c (= 9.5 \text{ K}) \) [4], the low-energy local vibration, or the anharmonic “rattling” motion of K ions with large excursions inside an oversized Os-O atomic cage, has been demonstrated both theoretically [5] and experimentally [6–11]. It is believed that the pronounced rattling of K ions is responsible for the unusual convex temperature dependence of resistivity \( \rho(T) \) in the normal state of KO\( \text{S}_2\)O\( \text{6} \) [6], indicating that the rattling strongly influences the electronic structure. It is also suggested [3] that such low-lying anharmonic phonons may give rise to an exotic superconducting state through the possible formation of heavy quasiparticles due to off-center degrees of freedom of ions. Very little is known, however, about the effects of the rattling on the superconducting properties.

What is intriguing in KO\( \text{S}_2\)O\( \text{6} \) is that inside the superconducting state below \( T_c \), a second transition occurs at \( T_p \approx 8 \text{ K} \), where specific heat shows an almost field-independent anomaly [6,7,10]. The high-field transport measurements have revealed that the concave \( \rho(T) \) at high temperatures changes to a typical Fermi-liquid dependence \( AT^2 \) below \( T_p \) [8,10]. This result naturally suggests that the K rattle responsible for the anomalous transport properties should be frozen below the transition \( T_p \). This provides a unique opportunity to study how this unusual rattling affects superconductivity and quasiparticle dynamics in the superconducting state.

In addition to these odd behaviors, strong electron correlations appear to have important contributions in transport and thermodynamic properties of KO\( \text{S}_2\)O\( \text{6} \). The Sommerfeld coefficient \( \gamma \) is estimated as high as 70 mJ/K\(^2\) mol [10], which is largely enhanced from the band calculation value [5]. At \( T_c \), the specific heat shows a large jump \( \Delta C/T_c = 200 \text{ mJ/K}^2 \text{ mol} \), and the upper critical field is found to be very high, up to 32 T (\( T \rightarrow 0 \text{ K} \)) with a steep slope of \( dH_{c2}(T)/dT = -3.4 \text{ T/K} \) [12]. These results indicate the pairing of electrons with enhanced effective mass \( m^* \), which in many cases invokes unconventional superconductivity. Moreover, the coefficient \( A \) in the \( AT^2 \) dependence of \( \rho \) below \( T_p \) follows the Kadowaki-Woods (KW) relation expected for strong correlation systems having large \( \gamma \) [8,10].

To clarify the effect of rattling phonons on superconductivity, microwave surface impedance \( Z_s(T) \) is a powerful low-energy electronic probe of quasiparticles, from which the magnetic penetration depth \( \lambda(T) \) and quasiparticle scattering time \( \tau \) can be extracted. The number of excited quasiparticles is most directly related to \( \lambda(T) \), since the superfluid density \( n_s \) is proportional to \( \lambda^{-2} \). Previous measurements of \( \lambda \) via \( \muSR \) [13] imply anisotropic gap with nodes, which contradicts the recent thermal conductivity results [8].

Here we report precise measurements of \( Z_s(T) \) in KO\( \text{S}_2\)O\( \text{6} \), from which fully gapped superconductivity with a large gap value \( \Delta = 25 \text{ K} \) is unambiguously demonstrated. The superfluid density shows a clear anomaly near \( T_p \), and the effective \( T_c \) is reduced by the freezing transition. This suggests that the rattling motion is helpful for superconductivity. Furthermore, the quasiparticle scattering rate rapidly decreases below \( T_p \), indicating enormous inelastic scattering in the normal state due to underlying strong correlations.

The surface impedance \( Z_s = R_s + iX_s \) is measured by a cavity perturbation method with the hot finger technique [14]. We used a 27 GHz TE\(_{011}\)-mode superconducting Pb cavity, whose temperature is maintained at 1.4 K, and the sample temperature is controlled up to 100 K. The inverse of quality factor \( 1/Q \) and the shift in the resonance fre-
unambiguously indicates that the quasiparticle excitation is of the activated type and the superconducting gap is nodeless. The obtained value of $\lambda(0) = 261$ nm is consistent with the $\mu$SR results [13], and by using the coherence length $\xi(0) = 3.2$ nm [12], the Ginzburg-Landau parameter is evaluated as $\lambda(0)/\xi(0) \sim 82$, indicating the London limit. The long London penetration depth $\lambda_L = (\frac{m^*}{\mu_0 n_p e^2})^{1/2}$ gives another support for the pairing of electrons with enhanced mass.

In Fig. 3, we plot the temperature dependence of superfluid density $n_s(T)/n_s(0) = \lambda^2(0)/\lambda^2(T)$. Again, it is clearly incompatible with a $d$-wave calculation with line nodes [19]. We also compare the low-temperature data with the expectation of weak-coupling BCS $s$-wave superconductors, and found that above 3 K the data deviates, which can be explained by the strong electron-phonon coupling. The conclusion of full gap superconductivity is reinforced by the observed coherence peak in $X_s(T)$ in Fig. 1. We note that such a coherence peak in $X_s(T)$ and the flat temperature dependence in low-temperature $\lambda(T)$ are observed in all the samples we measured. Our conclusion is also consistent with the observed weak field dependence of thermal conductivity $\kappa$ in the low-temperature limit [8]. In contrast, the $\mu$SR reports strong field dependence of the effective penetration depth $\lambda_{\text{eff}}$ [13], but it has been pointed out that the theoretical models employed for the data analysis may have insufficient accuracy [20], and the origin of $\lambda_{\text{eff}}(H)$ is still controversial [21].

Next, we discuss the effect of the second transition. The superfluid density in Fig. 3 exhibits a steplike change near $T_p$. This indicates that the transition clearly affects the superconducting condensates. The temperature dependence of superfluid density below 8 K extrapolates to
zero at a temperature $T_0 \sim 8.7$ K noticeably lower than the actual $T_c$. This immediately indicates that below $T_p$ where the K rattle responsible for the anomalous $\rho(T)$ is frozen, the effective $T_c$ is reduced considerably. We note that recent measurements of the lower critical field $H_{c1}(T)$ [22], which is also related to the superfluid density, show a similar reduction of the effective $T_c$ below $T_p$, consistent with our observation. These results lead us to infer that the rattling motion of K ions helps to enhance superconductivity in this system, although further theoretical investigations are necessary to clarify the microscopic origins of the observed behavior.

To see the effect on the quasiparticle dynamics, we extract microwave conductivity $\sigma = \sigma_1 - i\sigma_2$ from the surface impedance by $Z_s = (i\mu_0\omega/\sigma)^{1/2}$. The extracted real part $\sigma_1(T)$ is plotted in Fig. 4. Below $T_c$ the conductivity goes up with lowering temperature and below $T_p$ it increases more rapidly. This dependence is markedly different from the usual BCS expectation in strong-coupling $s$-wave superconductors [15], where $\sigma_1(T)$ shows a small coherence peak just below $T_c$ and it decreases rapidly below $\sim 0.9T_c$ and continues to decrease exponentially at lower temperatures. The observed enhancement of $\sigma_1(T)$ is consistent with the recent thermal conductivity data in a similar crystal [8], where $\kappa/T$ is enhanced in the superconducting state [inset of Fig. 4]. Such a big enhancement in $\sigma_1(T)$ has been also reported in high-$T_c$ cuprates [23,24] and heavy-fermion CeCoIn$_5$ [25,26], and it has been believed to be a characteristic feature of unconventional ($d$-wave) superconductors with large electron correlation effects. In contrast, KO$_2$Os$_6$ is an $s$-wave superconductor and such a behavior is nevertheless observed, which indicates the uniqueness of this system.

Since the effect of coherence factors appears only in the vicinity of $T_c$ and the observed enhancement of $\sigma_1(T)$ is much bigger, we can employ the simple two-fluid analysis which has been known to be useful to evaluate the quasiparticle scattering time $\tau$ in the superconducting state [23,24]. Here, the superfluid density $n_s$ and normal fluid (quasiparticle) density $n_n$ gives the total carrier density $n$, and the real part of conductivity can be written as $\sigma_1 = n_c\epsilon^2\tau/m^*\frac{1}{1+(\sigma_n\tau)^2}$. By using the conductivity and the superfluid density data, we get $\tau(T)$ as depicted in Fig. 5. It demonstrates that the quasiparticle scattering time is enhanced in the superconducting state and reaches an order of magnitude larger at $\sim 0.7T_c$ than at $T_p$. We note that such an enhancement of $\tau$ has been suggested by the thermal conductivity data [8], but one has to argue that the lattice contribution to $\kappa$ should be small compared to the electronic part. In contrast, the microwave conductivity is purely electronic.

The enhancement of $\tau$ (or the suppression of the scattering rate $1/\tau$) below $T_c$ is an indication that there is enormous inelastic scattering in the normal state which is reduced in the superconducting state by the opening gap in the electronic spectrum. To see this more clearly, we compare $1/\tau$ with the quantity $(n_s(T)/n)^2\rho(T)/\rho(T_c)$ in the inset of Fig. 5. Here $\rho(T)$ is the resistivity in the normal state obtained by applying strong magnetic fields (13 T) to destroy superconductivity [8], whose temperature dependence should mostly come from the normal-state scattering rate. The factor $(n_s(T)/n)^2$ represents that in the isotropic
by the two-fluid analysis. The inset compares \(1/\tau(T)\) with \((n_s(T)/n)^2 \rho(T)/\rho(T_c)\).

The quasiparticle scattering manifested by the steep enhancement of \(\tau\) at low temperatures. The quasiparticle mean free path \(l(T) = v_F \tau \sim \xi T_T/\hbar\) reaches a long value \(\sim 45\) nm at \(\sim 0.7 T_c\), and shows no saturation behavior, which may rule out disordered glasslike freezing below \(T_p\). Consistently, a theoretical suggestion that an ordered state of K sites appears below \(T_p\) has been made [5].

In summary, from the microwave surface impedance in high quality single crystals of \(\text{KO}_2\text{O}_6\), we clarify the following three points. (i) The superconducting ground state is fully gapped and electron-phonon coupling is strong. (ii) The superfluid density shows a steplike anomaly near the transition at \(T_p\), which suggests that the rattling motion is an important ingredient to the enhanced superconductivity. (iii) The quasiparticle scattering is rapidly decreased below \(T_p\), suggesting strong coupling between the rattling phonons and quasiparticles. Our results highlight that in spite of the conventional \(s^\pm\) wave ground state, this pyrochlore superconductor with unusual structural and electronic properties gives remarkable features in the superconducting state. An immediate question arises on how the transition affects vortex physics, which deserves further studies.

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