Effects of Rattling Phonons on the Dynamics of Quasiparticle Excitation in the β-Pyrochlore KOs_2O_6 Superconductor

Y. Shimono,[1] T. Shibauchi,[1] Y. Kasahara,[1] T. Kato,[1] K. Hashimoto,[1] Y. Matsuda,[1,2] J. Yamaura,[2] Y. Nagao,[2] and Z. Hiroi[2]

[1]*Department of Physics, Kyoto University, Sakyo-ku, Kyoto 606-8502, Japan*

[2]*Institute for Solid State Physics, University of Tokyo, Kashiwa, Chiba 277-8581, Japan*

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Microwave penetration depth λ and surface resistance at 27 GHz are measured in high quality crystals of KOs_2O_6. Firm evidence for fully gapped superconductivity is provided from $\lambda(T)$. Below the second transition at $T_p \sim 8$ 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 β-pyrochlore superconductor KOs_2O_6 with a relatively high $T_c(\approx 9.5$ K) [4], the low-energy local vibration, or the anharmonic "rattling" motion of K ions with large excursion 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 KOs_2O_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 KOs_2O_6 is that inside the superconducting state below T_c, a second transition occurs at $T_p \sim 8$ 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 KOs_2O_6. The Sommerfeld coefficient γ is estimated as high as 70 $\mathrm{mJ/K^2\,mol}$ [10], which is largely enhanced from the band calculation value [5]. At T_c, the specific heat shows a large jump $\Delta C/T_c \approx 200$ $\mathrm{mJ/K^2\,mol}$, and the upper critical field is found to be very high, up to 32 T ($T \rightarrow 0$ K) with a steep slope of $dH_{c2}(T)/dT = -3.4$ 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 ρ below T_p follows the Kadowaki-Woods (KW) relation expected for strong correlation systems having large γ [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 τ can be extracted. The number of excited quasiparticles is most directly related to $\lambda(T)$, since the superfluid density n_s is proportional to λ^{-2}. Previous measurements of λ by μSR [13] imply anisotropic gap with nodes, which contradicts the recent thermal conductivity results [8].

Here we report precise measurements of $Z_s(T)$ in KOs_2O_6, from which fully gapped superconductivity with a large gap value $\Delta \approx 25$ 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-

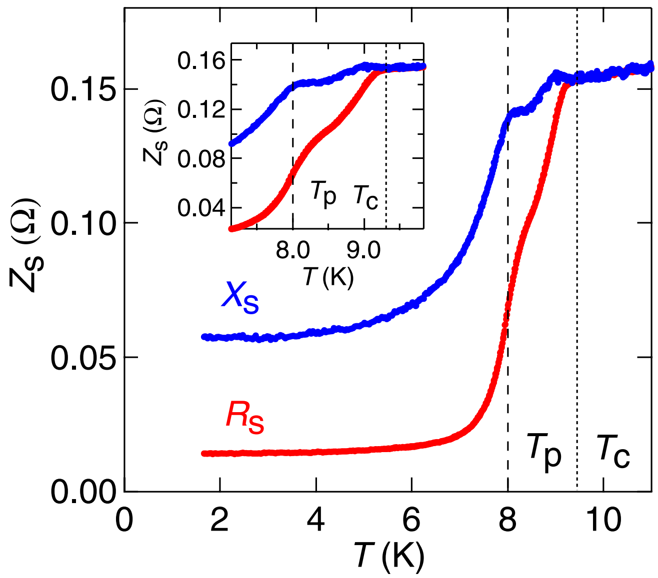

FIG. 1 (color online). Temperature dependence of the surface resistance R_s and reactance X_s at 27 GHz in a KOs_2O_6 single crystal. The inset shows an expanded view near the superconducting transition T_c and the second transition T_p.

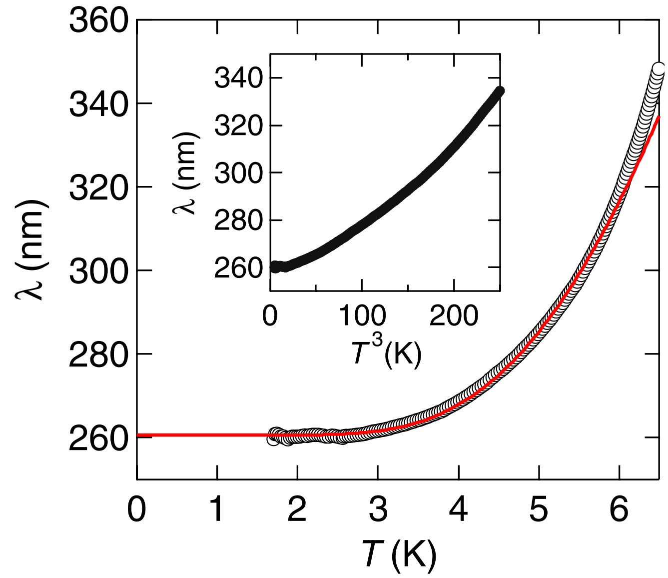

FIG. 2 (color online). Temperature dependence of the microwave penetration depth λ below 6.5 K in KOs_2O_6. The solid line is a fit to $\lambda(0) + C\exp(-\Delta/k_BT_c)$ with $\lambda(0) = 261$ nm and $\Delta = 24.5$ K. The inset shows λ vs T^3.

quency $\omega/2\pi$ are proportional to the real and imaginary parts of Z_s, respectively [15]. Details of the experimental setup are described elsewhere [16,17].

Single crystals of KOs_2O_6 were grown by the technique described in Ref. [6]. It has been known that once partial hydration takes place, the anomaly in specific heat at T_p tends to collapse. Before microwave measurements, we checked the specific heat anomaly and a special care was taken to keep the crystals in a dry atmosphere. We measured several crystals with shiny surfaces, and we here focus on the results of the crystal having most pronounced anomaly at T_p. The skin depth at 27 GHz is much smaller than the sample dimensions ($0.5 \times 0.5 \times 0.2$ mm^3), which ensures the skin depth regime.

Figure 1 shows the temperature dependence of R_s and X_s. In the normal state we can use the expected relation (in the Hagen-Rubens limit) $R_s = X_s = (\mu_0\omega\rho/2)^{1/2}$ to determine the absolute value of Z_s. Just below T_c, we observe a coherence peak in the reactance $X_s(T)$ near 9 K, which is a typical feature in s-wave superconductors [15]. A striking feature is that $Z_s(T)$ shows distinct anomalies near T_p, which we will discuss later.

The surface reactance is proportional to the penetration depth by $X_s = \mu_0\omega\lambda$. The temperature dependence of λ at low temperatures is demonstrated in Fig. 2. It is clear from the figure that $\lambda(T)$ has a flat temperature dependence at low temperatures, obviously different from T, T^2, or T^3 dependence [18] expected in the superconducting gap function with line or point nodes [see inset of Fig. 2]. The data below 6 K can be fitted to an exponential dependence $\lambda(T) - \lambda(0) \propto \exp(-\Delta/k_BT_c)$, with $\Delta \approx 24.5$ K, giving a strong-coupling value of $2\Delta/k_BT_c \approx 5.1$. This unambiguously indicates that the quasiparticle excitation is of the activated type and the superconducting gap is nodeless. The obtained value of $\lambda(0) \approx 260$ nm is consistent with the μSR results [13], and by using the coherence length $\xi(0) \approx 3.2$ nm [12], the Ginzburg-Landau parameter is evaluated as $\lambda(0)/\xi(0) \sim 82$, indicating the London limit. The long London penetration depth $\lambda_L = (\frac{m^*}{\mu_0 n_s e^2})^{1/2}$ 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 κ in the low-temperature limit [8]. In contrast, the μSR reports strong field dependence of the effective penetration depth $\lambda_{\rm eff}$ [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 $\lambda_{\rm eff}(H)$ 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

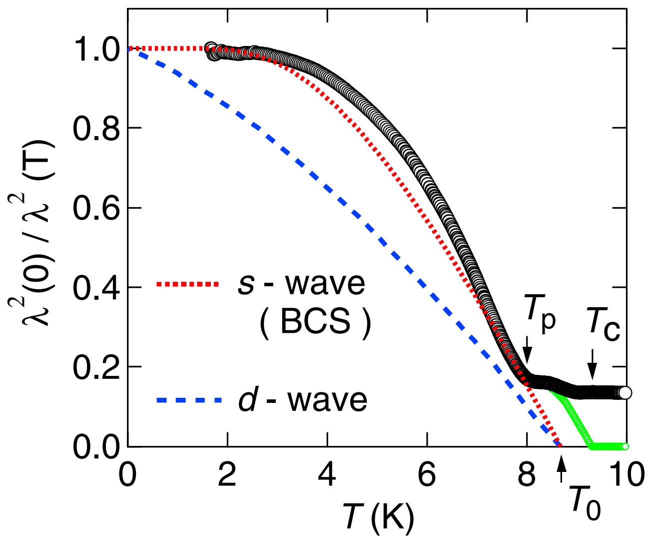

FIG. 3 (color online). $\lambda^2(0)/\lambda^2(T) = n_s/n$ as a function of temperature. The data below 8 K extrapolates to zero at $T_0(<T_c)$. Above T_c, λ is limited by the normal-state skin depth [16], but the superfluid density n_s should become zero at T_c as shown in the green solid line. The red dotted line is a weak-coupling BCS prediction for s-wave superconductors in the London limit. The blue dashed line is a calculation for d-wave superconductors with line nodes [19].

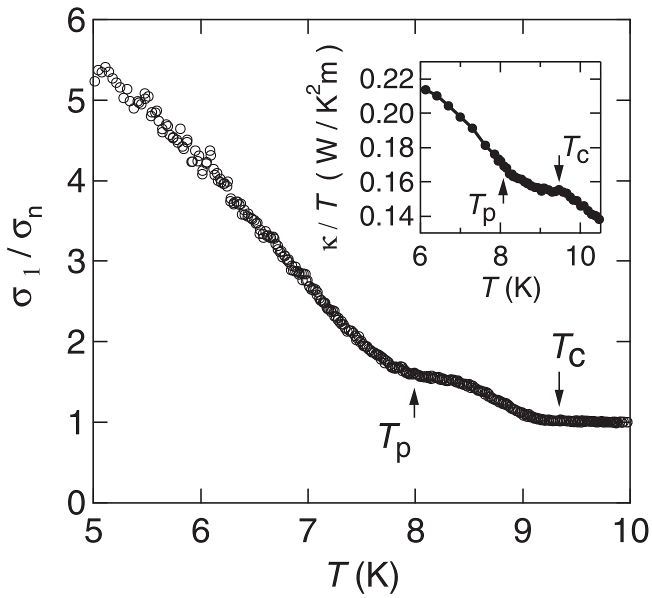

FIG. 4. Temperature dependence of microwave conductivity σ_1 of KOs_2O_6 at 27 GHz normalized to the normal-state value σ_n at T_c. The inset shows the $\kappa/T(T)$ data [8].

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 κ/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, KOs_2O_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 τ 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 $\sigma_1 = \frac{n_n e^2 \tau}{m^*} \frac{1}{1+(\omega\tau)^2}$. 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_c. We note that such an enhancement of τ has been suggested by the thermal conductivity data [8], but one has to argue that the lattice contribution to κ should be small compared to the electronic part. In contrast, the microwave conductivity is purely electronic.

The enhancement of τ (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 $(n_n(T)/n)^2\rho(T)/\rho(T_c)$ 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 $(n_n(T)/n)^2$ represents that in the isotropic

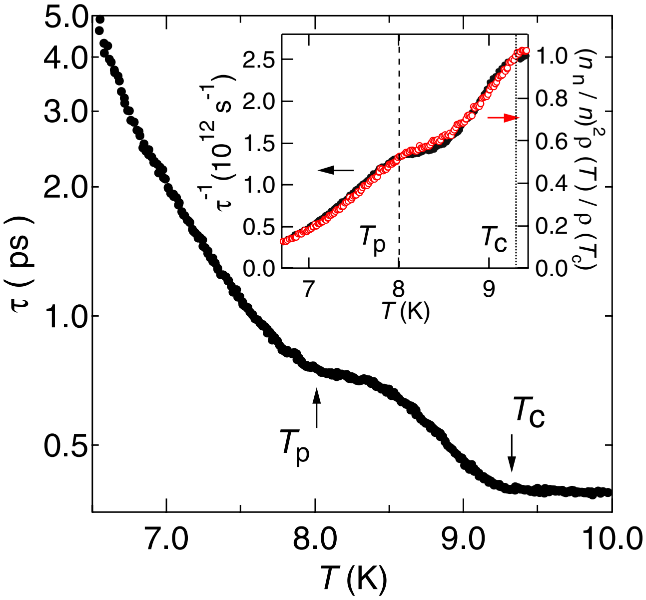

FIG. 5 (color online). Temperature dependence of quasiparticle scattering time $\tau(T)$ extracted from the surface impedance by the two-fluid analysis. The inset compares $1/\tau(T)$ with $(n_n(T)/n)^2\rho(T)/\rho(T_c)$.

gap case the scattering rate between quasiparticles can be simply proportional to the number of quasiparticles *and* the number of scatterers. The temperature dependence of both quantities below T_p is in very good correspondence. This simple analysis implies that the effect of the second transition on the quasiparticle dynamics is twofold: (1) The normal-state scattering mechanism changes from strong phonon-dominated scattering with concave temperature dependence of $\rho(T)$ above T_p to strong electron-electron scattering as revealed by AT^2 dependence below T_p [8,10]; (2) the number of quasiparticles in the superconducting state changes at T_p which also changes the interquasiparticle scattering manifested by the steep enhancement of τ. These effects are consistent with the views of the rattling freezing as the nature of the transition, and suggest strong electron-electron correlations inherent in this system, which is consistent with the observation of the KW relation at low temperatures. The quasiparticle mean free path $l(T) = v_F\tau \sim \xi\Delta\tau/\hbar$ reaches a long value $\sim$45 nm at $\sim 0.7T_c$, 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 KOs_2O_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-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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