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Cooper-pair formation by anharmonic rattling modes in the β-pyrochlore superconductor KOs_2O_6

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Abstract. We study the influence of anharmonic rattling phonons in the β-pyrochlore superconductor KOs_2O_6 using the strong-coupling Eliashberg approach. In particular, by analyzing the specific heat data, we find that the rattling phonon frequency changes discontinuously at the critical temperature of the first-order phase transition. Solving the strong-coupling Eliashberg equations with effective temperature-dependent $\alpha^2F(\omega)$, we investigate the consequence of this first-order phase transition for the anomalous temperature dependence of the superconducting gap. We discuss our results in the context of the recent experimental data.

The superconductivity in the family of β-pyrochlore oxides AT_2O_6 (A = alkali and T = transition metal atom), e.g. KOs_2O_6, $RbOs_2O_6$ and $CsOs_2O_6$, exhibits many exotic properties [1]–[6]. In these compounds the T atoms have octahedral oxygen coordination and are arranged on a corner-sharing pyrochlore lattice. This leaves large voids to incorporate alkali atoms, which are therefore confined in rather anharmonic (flat) potential wells.

Among them, KOs_2O_6 with the highest superconducting transition temperature $T_c = 9.6$ K shows the most anomalous behavior. For example, the electrical resistivity demonstrates strong concave T dependence down to T_c [1], in contrast to the normal T^2 behavior in Rb and Cs compounds [2]–[6]. Specific heat measurements have found the existence of low-frequency

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Einstein modes and that the T-linear coefficient of the specific heat $\gamma = 70\,\mathrm{mJ\,K^{-2}\,mol^{-1}}$ (see [7, 8]) is strongly enhanced over the value obtained from band structure calculations [9, 10]. The latter show that conduction states comprise nearly half-filled (5/12) t_{2g} bands with a bare value of $\gamma_b = 17\,\mathrm{mJ\,K^{-2}\,mol^{-1}}$. It is a matter of dispute how much of the enhancement is due to lattice vibrations or electronic correlations [7, 9].

The electronic structure calculations have indicated that these anomalies may be partly due to highly anharmonic low-frequency rattling motion of the alkali ions inside an oversized cage formed by the Os and O ions [9]. Furthermore, this is consistent with the x-ray observation of anomalously large atomic displacement for the K ions [11], and the low-frequency phonon structures seen in photoemission spectra [12]. Moreover, recent NMR data [13] have indicated that the relaxation at the K sites is entirely caused by fluctuations of the electric field gradient ascribed to the highly anharmonic low-frequency dispersionless oscillation (rattling) of K ions in the cage. Recently, Dahm and Ueda [14] have developed a phenomenological model to describe the influence of the anharmonic phonons on the NMR relaxation and the electrical resistivity.

Various experiments have been carried out in KOs_2O_6 to elucidate the mechanism of superconductivity and gap symmetry. Most importantly, thermal conductivity [15] and laser photoemission spectroscopy (PES) [12] measurements have revealed an almost isotropic s-wave superconducting gap. Furthermore, the absence of the coherence peak in the K-NMR relaxation rate expected for the isotropic gap can be explained as due to strongly overdamped phonons [13]. Simultaneously, penetration depth measurements by μSR [16] and scanning tunneling experiments [17] indicate a sizeable anisotropic component admixture, approximately 30% according to the latter. Alternatively, a two-gap model for the two Fermi surface sheets [9] has been proposed [16, 17], also because the pressure dependence was found to be opposite to that of a single band Bardeen–Cooper–Schrieffer (BCS) model. As a whole the experimental results point to a phonon-dominated mechanism for Cooper pairing in β-pyrochlores, even in KOs_2O_6.

Nevertheless, the large value of the electron–phonon coupling constant $\lambda_{ep} \approx 2.38$, the presence of the low-energy rattling phonon modes and their influence on the electronic properties, and finally, the first-order structural phase transition at $T_p < T_c$ [8] make an investigation of the superconductivity in KOs_2O_6 very interesting. In particular, the novel aspects of the strong-coupling Eliashberg theory in the presence of coupling between conduction electrons and anharmonic rattling phonons require a separate analysis. Previously, an attempt to consider the effect of the temperature-independent rattling phonons has been made in [18] in relation to $CsOs_2O_6$.

In this work, we analyze the superconductivity in KOs_2O_6 using the standard Eliashberg formalism. We neglect the sizeable anisotropic gap admixture for simplicity. The exotic character of superconductivity in KOs_2O_6 is due to a large contribution from a strongly anharmonic local rattling mode, usually not included in a quasi-harmonic treatment of lattice vibrations and their renormalization by the coupling with conduction electrons. We find that the phonon spectrum can be modeled by the two Lorentzians peaked at the energies of the two Einstein modes, ω_{E1} and ω_{E2}, representing the lowest energy rattling phonons. An additional contribution arises from the Debye frequency at ω_D. To explain the superconducting transition temperature in KOs_2O_6, we have employed the mean-field analysis of the temperature dependence of the lowest Einstein mode proposed previously [14]. We show that its energy should decrease with temperatures down to $T_p < T_c$ and then jump to a higher frequency

indicating the first-order structural phase transition. Solving the nonlinear Eliashberg equations below T_c with temperature-dependent electron–phonon coupling function, $\alpha^2F(\omega)$, we compare our results with those from various experiments.

On the real frequency axis the finite temperature Eliashberg equations for the superconducting gap $\Delta(\omega, T)$ and the renormalization function $Z(\omega, T)$ are given by the following equation [19]:

$$\Delta(\omega, T) = \frac{1}{Z(\omega, T)} \int_0^\infty d\omega' \mathrm{Re}\left\{\frac{\Delta(\omega', T)}{\sqrt{\omega'^2 - \Delta^2(\omega', T)}}\right\}\left[K_+(\omega, \omega', T) - \mu^* \tanh\left(\frac{\beta\omega'}{2}\right)\right], \quad (1)$$

$$\omega(1 - Z(\omega, T)) = \int_0^\infty d\omega' \mathrm{Re}\left\{\frac{\omega'}{\sqrt{\omega'^2 - \Delta^2(\omega', T)}}\right\} K_-(\omega, \omega', T), \quad (2)$$

where

$$K_\pm(\omega, \omega', T) = \int_0^\infty d\Omega\, \alpha^2F(\Omega)\left[\frac{f(-\omega') + n(\Omega)}{\omega' + \omega + \Omega} \pm \frac{f(-\omega') + n(\Omega)}{\omega' - \omega + \Omega}\right.$$
$$\left. \mp \frac{f(\omega') + n(\Omega)}{-\omega' + \omega + \Omega} - \frac{f(\omega') + n(\Omega)}{-\omega' - \omega + \Omega}\right] \quad (3)$$

and

$$\alpha^2F(\Omega) = \alpha^2_{E1}F_{E1}(\Omega) + \alpha^2_{E2}F_{E2}(\Omega) + \alpha^2_P F_P(\Omega) \quad (4)$$

is the generalized electron–phonon coupling function averaged over the Fermi surface. We assume that there are three contributions to the Eliashberg function. The last one arises from the usual dispersive acoustic phonons limited by the Debye energy $T_D = 325$ K. The first two contributions are due to local low-energy rattling phonons whose energies are designated by ω_{E1} and ω_{E2}. The latter requires special consideration.

It has been shown recently that the low-energy phonons ascribed to the heavy rattling of the K ions confined in an oversized cage made of OsO_6 octahedra is responsible for the unusual scattering processes in KOs_2O_6 [8]. In particular, it has been assumed that contrary to the $RbOs_2O_6$ and $CsOs_2O_6$ cases where a single Einstein mode due to rattling motion is enough, two modes exist at $\hbar\omega_{E1} = 22$ K and $\hbar\omega_{E2} = 61$ K in KOs_2O_6 corresponding to the first two excited energy levels of the corresponding anharmonic potential. Simultaneously, the mean-field description of the local alkali-ion anharmonic motion has been developed in [14]. Starting from the standard anharmonic Hamiltonian

$$H = \frac{p^2}{2M} + \frac{1}{2}ax^2 + \frac{1}{4}bx^4, \quad (5)$$

where x, p and M are the spatial coordinate, momentum and mass of the alkali ion, respectively, and a, b are constants with $b > 0$, one finds that according to *ab initio* calculations [9] for KOs_2O_6, the quadratic term becomes negative, i.e. $a < 0$, which results in a double well potential. We note that in reality the double wells are not in the plane [9]. Treating the Hamiltonian (5) in the self-consistent quasi-harmonic approximation, Dahm and Ueda [14] have found that the oscillation of potassium ions can be described by an effective harmonic oscillation with effective low-energy frequency that now depends on temperature. In particular, it decreases monotonically upon decreasing temperature[5]. At low enough temperatures ($T \ll \omega_0$), this

[5] It is important to remember that the effective frequency, ω_0, is a thermodynamical average frequency and should not be mixed with the discrete, temperature independent energy levels of the original Hamiltonian.

frequency becomes nearly temperature independent. Obviously this model requires a certain modification when applied to KOs_2O_6. In particular, although this effective model works well at high temperatures, it fails to explain the occurrence of the first-order phase transitions at temperature $T_p < T_c$ and the occurrence of the second Einstein frequency at higher energy.

Therefore, for modeling the contribution of the rattling K ions to the Eliashberg function, we adopt two Einstein-like modes centered at ω_{E1} and ω_{E2}. Furthermore, to take into account the upper-lying energy levels of the shallow potential we use the self-consistent quasi-harmonic approximation for the lowest mode ω_{E1} that results in its temperature dependence. Most importantly, before entering the temperature-independent regime, the effective rattling phonon frequency shows a discontinuous jump toward higher frequency that will result in a specific heat anomaly at T_p. The increase of the ω_{E1} at T_p is in accordance with recent experiments showing no structural changes below the phase transition [8, 13, 20]. Indeed, below T_p the K ions seem to be stabilized in their equilibrium positions, which corresponds to the increase of ω_{E1}.

To find the exact shift of the lowest frequency at T_p, and also the approximate positions of the rattling mode frequencies, we simulated the specific heat data around T_p following the model proposed by Hiroi *et al* [8] with temperature-dependent ω_{E1}. The contribution from the two frequencies representing the rattling modes is given by the following equation [8]:

$$C = aC_{E1} + (1-a)C_{E2}, \tag{6}$$

where $a = 0.24$ and ω_{Ei} with $i = 1, 2$ are the corresponding Einstein phonons. As mentioned above, for the sake of simplicity we assume that the higher mode is temperature independent and the lower one follows the mean-field temperature dependence that originates from integrating out the upper-lying energy levels with temperature. Generally, for the contribution of temperature-dependent Einstein frequency one finds

$$C_E = 3R\left(\frac{\hbar\omega_E}{k_B T}\right)^2 \frac{\exp\left(\hbar\omega_E/k_B T\right)}{\left(\exp\left(\hbar\omega_E/k_B T\right)-1\right)^2}\left[1 - \frac{\partial \ln \omega_E}{\partial \ln T}\right], \tag{7}$$

where R is the gas constant. The last term is absent for the upper temperature-independent frequency, ω_{E2}. The results of the fit are shown in figure 1. The change of the specific heat at $T_p = 7.5\,\mathrm{K}$ is reproduced by assuming a jump of the lower Einstein frequency from $\hbar\omega_{E1} = 27.4\,\mathrm{K}$ to $\hbar\omega_{E1} = 30\,\mathrm{K}$ at this temperature with ω_{E2} being constant. Above T_p the lower frequency mode is temperature dependent and the overall behavior of $\omega_{E1}(T)$ is shown in figure 2. Note that the strong temperature dependence of the lowest rattling phonon mode frequency has been recently observed by Raman scattering [20, 21] and inelastic neutron scattering experiments [22]. We further note that the increase of the lower frequency at T_p is consistent with the exact diagonalization studies performed recently [23].

In addition to the anharmonic temperature dependence of the rattling phonon mode that modifies the usual approximation for the Eliashberg function, the conduction electrons couple to the rattling phonons, further renormalizing their energy positions and introducing an extra damping. In particular, the spectral function of the low-energy rattling phonon frequency is given by

$$\alpha^2 F_{E1}(\omega) = -\frac{\alpha^2(T)}{\pi}\mathrm{Im}D(\omega) = \frac{\alpha^2(T)}{\pi}\frac{4\omega_{E1}(T)\Gamma_0\omega}{\left(\omega^2-\omega_r^2\right)^2 + 4\Gamma_0^2\omega^2}, \tag{8}$$

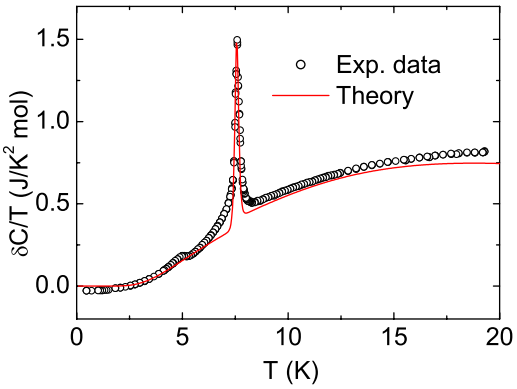

Figure 1. Theoretical fit to the experimental specific heat data from [8] using $\hbar\omega_{E2} = 59\,\mathrm{K}$ and $\hbar\omega_{E1}(T = T_c) = 27.4\,\mathrm{K}$. Assuming the jump of the characteristic rattling phonon frequency at T_p from $\hbar\omega_{E1} = 27.4\,\mathrm{K}$ to $\hbar\omega_{E1} = 30\,\mathrm{K}$, we find a corresponding increase in the specific heat curve in qualitative agreement with experimental data.

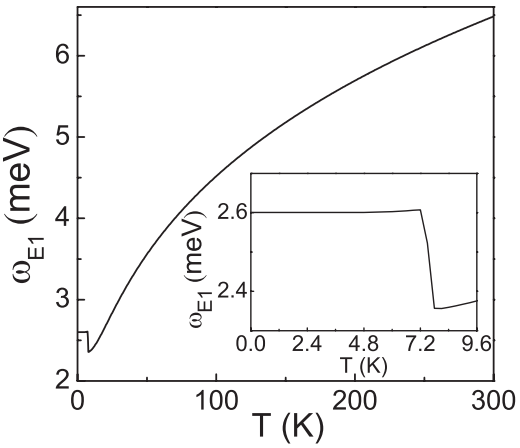

Figure 2. Calculated temperature dependence of the lower frequency ω_{E1} of the rattling phonon as obtained using the quasi-harmonic approximation [14]. The parameter of the model $b\hbar/M^2\omega_0^3(T = 300\,\mathrm{K}) = 0.25$ has been fixed by assuming the room temperature position of the rattling phonon frequency around 6 meV as found by neutron scattering experiments [24]. At T_p the effective frequency demonstrates a discontinuous jump to a higher energy (as shown in the inset) whose position is determined by the absolute value of the specific heat jump shown in figure 1.

where Γ_0 is an anharmonic phonon damping rate. The real part of the rattling phonon self-energy leads to a renormalized phonon frequency,

$$\omega_r^2(T) = \omega_{E1}^2(T) + 2\omega_{E1}(T)\mathrm{Re}\Pi(\omega). \tag{9}$$

Following a previous estimation [14], we have used $-\mathrm{Re}\Pi(\omega) = 1\,\mathrm{meV}$ and $\Gamma_0 = 0.25\,\mathrm{meV}$. It is important to remember that the electron–phonon coupling parameter $\alpha^2(T) \sim c/\omega_{E1}(T)$ with $c = 1.23\,\mathrm{meV}^2$ is also temperature dependent.

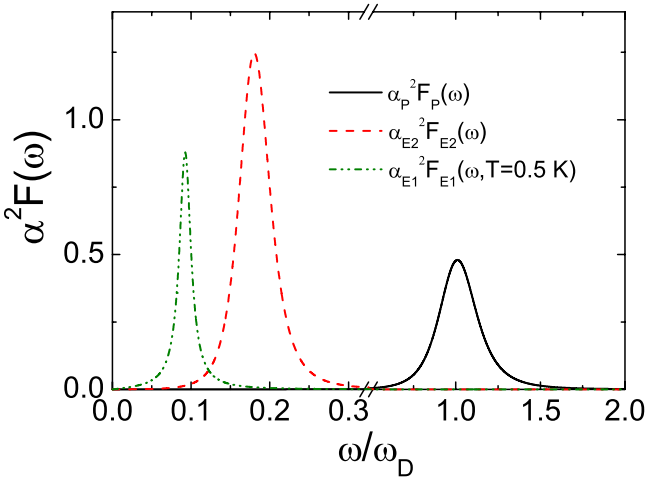

Figure 3. Calculated Eliashberg function, equation (4), for KOs_2O_6 for $T = 0.4$ K. The solid curve denotes the usual phononic contribution centered around Ω_D and the (red) dashed curve refers to the upper rattling phonons frequency centered around ω_{E2}. Here, we employ $\alpha^2 F_i(\omega) = \frac{c_i \omega_i \Gamma_i^3}{\pi((\omega-\omega_i)^2+\Gamma_i^2)^2}$, where $i = E2$, p. We further use $\Gamma_i = \omega_i/5$, $c_{E2} = 0.767$, and $c_p = 0.3$. The (green) dashed-dotted curve refers to the lower energy rattling mode contribution to $\alpha^2 F(\omega)$ with parameters as described in the text.

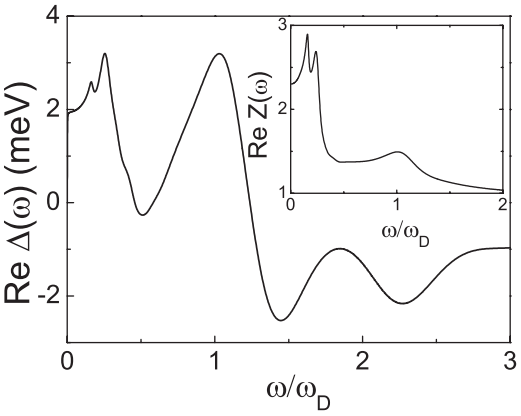

Figure 4. Calculated frequency dependence of the real part of the superconducting gap function $\Delta(\omega)$ for KOs_2O_6 compounds. The inset shows the renormalization function $Z(\omega)$ at $T = 0.5$ K. Note that we set the cutoff frequency to $8\omega_D$ and introduce a finite damping $\Gamma_0 = 0.25$ meV. The results are not sensitive to further increase of the cutoff frequency.

In figure 3, we show the Eliashberg function $\alpha_i^2 F_i(\Omega)$ including all three contributions for $T = 0.5$ K, i.e. well below T_p. In contrast to the usual Eliashberg theory, the $\alpha^2 F(\omega)$ spectrum and the coupling constant $\lambda = 2\int_{-\infty}^{+\infty} d\Omega\, \alpha^2 F(\Omega)/\Omega$ are now temperature dependent. Setting $\mu^* = 0.091$ and solving equations (1)–(3) in the linearized limit, we find the superconducting transition temperature $T_c = 9.6$ K. In figure 4, we show the results of the solution of the

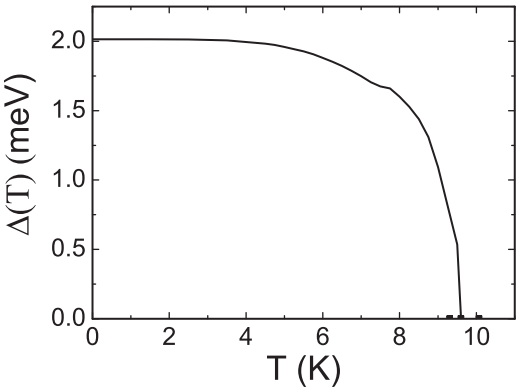

Figure 5. Calculated temperature dependence of the superconducting gap $\Delta_0(T)$ as determined from $\mathrm{Re}\Delta(T,\omega)=\omega$ for KOs_2O_6. The kink in the temperature dependence refers to the first-order phase transition that occurs at $T_p = 7.5\,\mathrm{K}$ and the corresponding change in the coupling constant λ.

Eliashberg equations for $T = 0.5\,\mathrm{K}$. One finds the typical behavior of a strong-coupling superconductor. In particular, $\mathrm{Re}\Delta(\omega)$ shows three peaks at the energy of the rattling phonons and at ω_D. For larger energies, it becomes negative, reflecting the effective repulsion for $\omega > \omega_D$.[6] Due to the presence of the low-energy rattling phonons one obtains a strong renormalization of the quasi-particle mass and we also find that $2\Delta_0/k_B T_c \approx 5.0$. The latter is in good agreement with the experimentally observed value [12].

One of the important consequences of the temperature-dependent phonon spectrum is the slightly anomalous behavior of the superconducting gap as a function of temperature. In figure 5, we show the temperature dependence of the superconducting gap. One finds a kink at $T = T_p$ due a discontinuous change of the phonon frequency and the corresponding coupling constant. In particular, due to the slight increase of ω_{E1} below T_p the electron–phonon coupling constant decreases, which results in a decrease of the superconducting gap with respect to its value without the structural phase transition. At the same time, overall we find relatively small modification of the superconducting properties due to the structural phase transition. This is consistent with experimental observations [8], which show a weak connection between the structural transition and superconductivity in KOs_2O_6. The most important result is that due to the temperature-dependent low-energy mode, the superconductivity is enhanced in KOs_2O_6. In particular, the dimensionless coupling constant $\lambda(T)$ increases with decreasing temperature due to a decrease in ω_{E1}. Further, it allows us to understand qualitatively why T_c in KOs_2O_6 is higher than that in the Rb and Cs counterparts since the characteristic energies of the rattling modes effectively lie at higher energies in these compounds. At the same time, using simply different values of ω_{E1} and ω_{E2} for all three compounds [21] one does not get the exact values of T_c without further modification of the μ^* value. We also find that the electron–phonon coupling strength changes from $\lambda(T > T_p) = 1.7$ toward $\lambda(T < T_p) = 1.6$ and is somewhat lower than estimated previously [8]. This is because a simple application of the McMillan–Dynes formula for estimating λ is probably questionable in KOs_2O_6 due to the temperature dependent rattling mode, which does not exist in the simple Eliashberg theory. Our obtained results, although

[6] The slightly negative value of $\mathrm{Re}\Delta(\omega)$ at energies of around $0.5\omega_D$ is an artefact of the small value of $\alpha^2F(\Omega)$ in this range. In particular, it becomes positive once the damping Γ is taken smaller.

showing an anomaly at T_p, cannot explain fully the photoemission data of [12] as is evident from figure 5. On the other hand, the tunneling data of [17] give almost no temperature dependence of $\Delta(T)$ for $T < 6$ K; this is in much better agreement with the theoretical curve in figure 5.

Our calculation shows that the superconducting properties of KOs_2O_6 should be sensitive to the external pressure. Due to the proximity to the first-order phase transition, the application of pressure may result in a modification of the electron–phonon coupling strength. Such an unusual behavior has indeed been recently found experimentally [25] although a detailed understanding both theoretically and experimentally is still required.

In summary, we have investigated the superconducting properties of the β-pyrochlore compound KOs_2O_6 based on the strong-coupling Eliashberg approach. Analyzing the specific heat data we find that the rattling phonon frequency changes discontinuously at the critical temperature of the first-order phase transition. Solving the strong-coupling Eliashberg equations with temperature-dependent $\alpha^2F(\omega)$, we discuss the consequence of this first-order phase transition for the anomalous temperature dependence of the superconducting gap. In particular, we have found that the superconducting gap as a function of temperature is anomalous, reflecting the temperature dependence of the Eliashberg function.

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