Quasiparticle-Self-Energy and its Effect on the Superconducting Order Parameter of the Pyrochlore Superconductor Cd$_2$Re$_2$O$_7$

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Abstract

The magnitude and the temperature dependence of the superconducting order parameter $\Delta(T)$ of single-crystals of Cd$_2$Re$_2$O$_7$ ($T_c = 1.02$ K) was measured using point contact spectroscopy. In order to fit the conductance spectra and to extract the order parameter at different temperatures we generalized the Blonder-Tinkham-Klapwijk theory by including the self-energy of the quasiparticles into the Bogoliubov equations. This modification enabled excellent fits of the conductance spectra. $\Delta(T)$ increases steeply below the superconducting transition temperature of 1.02 K and levels off below $\sim$0.8 K depending on measurement directions where its value varied from 0.22(1) meV to 0.26(1) meV. Our results indicate the presence of a strong electron-phonon interaction and an enhanced quasiparticle damping and may be related to a possible phase transition within the superconducting region at $\sim$0.8 K.

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A large number of transition metal (TM) oxides crystallize with the α-pyrochlore structure with the general composition $A_2B_2O_7$, where $A$ is a larger and $B$ is a smaller TM cation. Amongst the α-pyrochlores, $Cd_2Re_2O_7$ is the only one which shows superconductivity at $\sim 1\text{ K}$\textsuperscript{2}. At room temperature, $Cd_2Re_2O_7$ exhibits a cubic structure (space group $Fd\overline{3}m$). At $T_{S1} \sim 200 \text{ K}$, $Cd_2Re_2O_7$ undergoes a metal-to-metal second order structural phase transition (PT) to a non-centrosymmetric tetragonal structure (space group $I\overline{4}m2$) followed by a first order PT at $T_{S2} \sim 120 \text{ K}$ to another tetragonal structure (space group $I4_122$)\textsuperscript{3–5}. These two PTs have a profound effect on the electronic and the magnetic properties of $Cd_2Re_2O_7$. The electrical resistivity and the magnetic susceptibility drop sharply below $T_{S1}$\textsuperscript{3,4}. Heat capacity measurements below $T_{S2}$ showed an exceptionally large electronic Sommerfeld coefficient of $\gamma =15 \text{ mJ/K}^2 \text{ mol Re}$. Band structure calculations for the room-temperature cubic structure revealed that the electronic density of states (DOS) at the Fermi level arises mainly from bands with Re-5$d$ character with electron or hole pockets at various points of the Brillouin zone\textsuperscript{7}. However, the band structure of $Cd_2Re_2O_7$ in the low-temperature structure ($T < T_{S2}$) indicated localized Cd 4$d$ and itinerant Re-5$d$ electrons and a quasi two dimensional Fermi surface\textsuperscript{8}. The results of the Re nuclear quadrupole resonance (NQR) and the Cd nuclear magnetic resonance (NMR) at low temperature, and in the superconducting state, revealed no magnetic or charge ordering\textsuperscript{9}. Just below $T_c$, the $^{187}$Re spin lattice relaxation rate exhibits a pronounced coherence peak with an increase of the relaxation rate by a factor of two and subsequently, below $\sim 0.8 \text{ K}$, an exponential decrease consistent with weak-coupling BCS theory and an isotropic gap\textsuperscript{9}. Vyaselev et al. calculated the Wilson ratio and obtained a value of 0.34 indicating strong electron-phonon coupling incompatible with weak-coupling theory\textsuperscript{9}. The far-infrared spectroscopy measurements on $Cd_2Re_2O_7$ crystals in the superconducting state at $\sim 0.5 \text{ K}$ revealed two strong absorption peaks near 9.6 and 19.3 cm$^{-1}$ which completely vanish above $T_c$ possibly indicating strong electron-phonon coupling in the superconducting state\textsuperscript{10}. 

In this article, we report point contact spectroscopy measurements on single crystals of $Cd_2Re_2O_7$ carried out in order to better understand the superconducting state of $Cd_2Re_2O_7$. Our measurements provide the temperature dependence of the superconducting order parameter which differs markedly from the BCS temperature dependence of the energy gap. For $T \to 0 \text{ K}$ the order parameter approaches a value between 0.22(1) to 0.26(1) meV, i.e $2\Delta(0)/k_B T_c > 5.0(1)$ indicating that $Cd_2Re_2O_7$ is a strong-coupling superconductor.
Single crystals of Cd$_2$Re$_2$O$_7$ were grown from Re$_2$O$_7$ and Cd (purity 99.99%) by a gas phase chemical transport technique$^{13}$. Re$_2$O$_7$ powder was prepared from Re metal (purity 99.99%) and O$_2$ (purity 99.9999%) by employing the Noddack method$^{12}$. To check the purity of the samples, we crushed several small crystals and performed X-ray powder diffraction measurements (see Fig. 1) where we found no impurity phases. The lattice parameter was refined to 1.028(5) nm, very close to the published data$^{1}$. Electron-microprobe analysis of selected Cd$_2$Re$_2$O$_7$ crystal proved an ideal Cd:Re ratio of one, within error bars of the method.

![Powder X-ray diffraction result of crushed single crystals of Cd$_2$Re$_2$O$_7$.](image1)

FIG. 1: Powder X-ray diffraction result of crushed single crystals of Cd$_2$Re$_2$O$_7$.

The ac-susceptibility and the heat-capacity measurements showed a superconducting transition at 1.02 K and a width of the transition of ±30 mK (see Fig. 2), consistent with data reported previously$^{6}$.

![Temperature dependence of the ac-susceptibility of a single crystal of Cd$_2$Re$_2$O$_7$.](image2)

FIG. 2: (a) Temperature dependence of the ac-susceptibility of a single crystal of Cd$_2$Re$_2$O$_7$. (b) Heat capacity of Cd$_2$Re$_2$O$_7$.

Andreev reflection spectra were measured in a specific crystallographic plane by employ-
ing the soft point contact spectroscopy (PCS) method reviewed in detail by Daghero and Gonnelli. The resistance of the contact to the sample was adjusted to be within the range (of \(\sim 10 \text{ Ohms} \)) of the resistance of the sample by repeatedly applying short high voltage pulses through the contact. The sample was cooled in a home-built single-shot \(^3\)He cryostat which enables the samples to be fully immersed in the cryogenic fluid reducing ohmic heating of the point contacts. The temperature was stabilized by controlling vapor pressure of liquid \(^3\)He and it was measured with a custom calibrated Cernox CX1030 temperature sensor.

![Graph](image)

**FIG. 3:** (a) A sample of I vs. V and (b) the calculated \(\frac{dI}{dV}\); no averaging was used in the process. The maximum error on the derivative is of the order of \(\pm 0.002\) and can not be represented on the graph.

The stability of temperature measurements was less than \(\pm 3 \text{ mK} \) below and \(\pm 7 \text{ mK} \) above 0.7 K, respectively. A graph of raw data I vs V is shown in Fig. 3a and its derivative is shown in Fig. 3b. No data averaging was done post measurement. The accuracy in current measurements is of the order of \(10^{-9} \text{ A} \) and for voltage of the order of \(\pm 2 \times 10^{-8} \text{ V} \). The measurement error on \(\frac{dI}{dV}\) is of the order of 0.2\% of its value. The fitting program stops iteration when the total standard deviation of all points is less than \(10^{-8}\).

Several measurements were done by injecting current in the (001), (100), (110), and (111) planes. As an example, a set of characteristic normalized conductance spectra collected by injecting the current in the (111) plane is displayed in Fig. 4. All spectra were normalized to the value of the conductance \(dI/dV\) measured at a voltage \(\simeq \pm 2.0 \text{ mV} \) in the normal state.
An attempt to fit the normalized conductance spectra of Cd$_2$Re$_2$O$_7$ using the Blonder–Tinkham–Klapwijk (BTK) theory\textsuperscript{16} did not provide satisfactory results, especially for the spectra at temperatures close to $T_c$ where finite lifetime effects can play an important role\textsuperscript{17}. The BTK theory is based on the Bogoliubov equations for the two-component wave function
\begin{equation}
\psi(r, t) = \begin{pmatrix} u(r, t) \\ v(r, t) \end{pmatrix}
\end{equation}
for particles and holes, respectively, but does not take into consideration any self-energy effects, i.e. finite quasiparticle lifetimes. Previous attempts to extend the BTK theory to include lifetime effects were purely phenomenological and assumed that the time-dependence of the particle and hole amplitudes $u$ and $v$ in the Bogoliubov equations are of the form $\exp[-i(E - i\Gamma)t/\hbar]$, where $E$ is the quasiparticle energy, and $\Gamma$ is its scattering rate\textsuperscript{18–20}. The resulting theory has a form identical to the BTK theory but with the normalized superconducting quasiparticle density of states given by the so-called Dynes equation\textsuperscript{21}
$$N_s(E) = \text{Re} \{(E - i\Gamma)/\sqrt{(E - i\Gamma)^2 - \Delta^2}\},$$
where $\Delta$ is the superconducting energy gap. However, it was pointed out that the above equation cannot be justified microscopically, at least not for conventional strong-coupling superconductors such as Pb or Nb\textsuperscript{22}.
strong-coupling (Eliashberg) theory of superconductivity the normalized superconducting quasiparticle density of states is given by \( N_s(E) = \text{Re} \frac{E}{\sqrt{E^2 - \Delta^2(E)}} \),

where \( \Delta(E) \) is the complex gap function, i.e. the renormalized pairing self-energy. All damping and retardation effects are contained in \( \Delta(E) \).

We have generalized the BTK theory by using the Bogoliubov equations which include the self-energy effects. Their time Fourier transform has the form

\[
\{[-\frac{\hbar^2}{2m} \nabla^2 - \mu(r)]\tau_3 + \Sigma(r, E)\} \psi(r, E) = E \psi(r, E),
\]

where

\[
\Sigma(r, E) = (1 - Z(r, E))\tau_0 + \phi(r, E)\tau_1
\]

is the 2×2 electron self-energy matrix. \( \tau_0 \) is a unit matrix and \( \tau_1 \) and \( \tau_3 \) are Pauli matrices. We have assumed that the self-energy is local in space which is justified if it arises from the electron-phonon interaction. The gap function \( \Delta(r, E) \) is related to the pairing self-energy \( \phi(r, E) \) and the renormalization function \( Z(r, E) \) by \( \Delta(r, E) = \phi(r, E)/Z(r, E) \). In the weak-coupling limit \( Z = 1, \Delta = \phi \) and Eq. (3) reduces to the familiar Bogoliubov equation.

Subsequently, by making the same assumptions as in the derivation of the BTK theory (spatially independent \( \mu, \phi \) and \( Z \), translational invariance along \( y \)- and \( z \)-directions and a \( \delta \)-function potential at the normal metal (N)–superconductor (S) interface), we arrive at a theory which is identical in form with the BTK theory but with the real gap \( \Delta \) replaced by the complex gap function \( \Delta(E) \). Details of the derivation are published elsewhere.

Specifically, the conductance of an N–S interface at a voltage \( V \) is given by

\[
\frac{dI_{NS}}{dV} = S \int_{-\infty}^{+\infty} dE \frac{df(E) - eV}{dV} [1 + A(E) - B(E)],
\]

with \( S = 2N(0)e v_F A \). \( N(0) \) is the single-spin Fermi level density of states in the normal state, \( e \) is the electron charge, \( v_F \) is the Fermi velocity and \( A \) the effective area of the N–S interface. The probability current densities for the Andreev reflection \( A(E) \) and for the
normal reflection $B(E)$ are given by (in units of the Fermi velocity $v_F$)

$$A(E) = \frac{|u|^2 v^2}{|\gamma|^2}$$

(6)

$$B(E) = \frac{[|u|^4 + |v|^4 - 2 \text{Re}(u^2 v^2)] z^2 (z^2 + 1)}{|\gamma|^2}$$

(7)

$$\gamma = u^2 + (u^2 - v^2)z^2$$

(8)

$$u, v = \frac{1}{\sqrt{2}} \sqrt{1 \pm \sqrt{E^2 - \Delta^2(E)/E}}.$$ 

(9)

The parameter $z$ in Eqs. (7-8) is a dimensionless barrier strength parameter related to the strength of the $\delta$-function potential $V(x) = H \delta(x)$ at the interface by $z = H/(\hbar v_F)$.

For $E$ not too far from the gap edge the real and the imaginary part of $\Delta(E)$ can be taken as constant$^{17,22}$. Thus, in applying Eqs. (5-9) to the experimental results there are three fit parameters at each temperature: the real and the imaginary part of the gap at the gap edge, and the barrier strength parameter $z$. Two characteristic fits to the conductance spectra ($dI/dV$ vs $T$) at different temperatures are displayed in Fig. 5a, and 5c. We found that the theoretical fit using the BTK model modified by including a complex $\Delta$ provides a significantly improved agreement with the experimental data. The variation of $z$ parameter

FIG. 5: Experimental data (□) and theoretical fits for the normalized conductance spectra of Cd$_2$Re$_2$O$_7$ at 0.831 K (a and b) and 0.360 K (c and d). Solid lines in Figs. 5a and 5b: represent the fits obtained using our generalized BTK theory with a complex gap. Solid lines in Figs. 5b and 5d show the fits using the phenomenological approach including the Dynes broadening parameter $\Gamma^{18-20}$. 

7
as a function of temperature did not show any systematic variation with temperature (see Fig. 6) and it shows a random fluctuation with temperature of the order of ± 3%. In

contrast, the fits with the BTK model extended by including a phenomenological broadening parameter Γ (Fig. 5b, and 5d) describe the experimental data less well, especially for temperatures near $T_c$. This finding renders strong support that the described theoretical amendments are essential for a description of conductance spectra of superconductors in the strong coupling limit.

By fitting the set of temperature dependent conductance spectra, we obtained the superconducting energy gap of $\text{Cd}_2\text{Re}_2\text{O}_7$ as a function of temperature (see Fig. 7). We also checked for anisotropy in $\Delta_0(T)$ for different crystallographical planes as shown in Fig. 8. Both injected and measured voltage are in the same plane. We observed the $\Delta_0(T)$ values for all planes saturate at about $\Delta_0(T) = 0.22$ meV, except however, for the (001) plane where the value is slightly larger and saturated at about 0.26 meV. The deduced values for $2\Delta(T \to 0)/k_BT_c$ which vary between 5.0(0.1)to 6.2(0.1) for different crystallographic planes indicate a magnitude characteristic for strong-coupling superconductors. The temperature dependence of the gap is markedly different from that expected from BCS theory. $\Delta(T)$ rises rapidly below $T_c$ and levels off below $\sim 0.8$ K whereas the imaginary part exhibits a maximum at $\sim 0.8$ K and subsequently decreases again but remains constant to the lowest temperatures measured.

The magnitude and the temperature dependence of the superconducting order parameter found in this study give an indication that $\text{Cd}_2\text{Re}_2\text{O}_7$ is a strong coupled superconductor.
FIG. 7: Superconducting energy gap $\Delta_0(T)$ of $\text{Cd}_2\text{Re}_2\text{O}_7$ (square symbols) for (001) plane obtained by fitting the experimental data for the normalized conductance at different temperatures using the generalized BTK theory presented in this work. The circles give the values of the imaginary part of the gap at the gap edge obtained in the fits (the errors in fitted values are less than the size of the symbols).

FIG. 8: $\Delta_0(T)$ of $\text{Cd}_2\text{Re}_2\text{O}_7$ measured for different crystallographic planes with a remarkably large gap resulting in a ratio $2\Delta(0)/k_BT_c > 5$, which is comparable to values found e.g. in Pb–Bi alloys with extremely strong electron-phonon coupling. The related ternary $\beta$-pyrochlore osmium oxides, $\text{AO}_2\text{O}_6$, where A is an alkali metal cation, also exhibit such large $2\Delta(0)/k_BT_c$ ratios, as observed for $\text{KO}_2\text{O}_6$. However, $T_c$ of $\text{KO}_2\text{O}_6$
is by an order of magnitude higher than in Cd$_2$Re$_2$O$_7$. In the superconducting region the density of states of KOs$_2$O$_6$ was measured using photoemission spectroscopy and the data were fitted using the Dynes formula. The resulting broadening parameter $\Gamma$ had temperature dependence quite similar to what we found for the imaginary part of the gap shown in Fig. 7. An additional similarity with the $\beta$-pyrochlore osmium superconductors is the substantial enhancement of the experimental Sommerfeld $\gamma$-term with respect to the band $\gamma$-term. Superconductivity in the $\beta$-pyrochlore osmium oxides is usually attributed to a low-energy rattling vibrational mode of the alkali metal atoms which may also account for the observed first order structural phase transition occurring below $T_c$ in KOs$_2$O$_6$. It is tempting to associate the superconductivity in Cd$_2$Re$_2$O$_7$ to similar low-energy vibrational excitations, e.g. to the low-energy IR modes recently observed by low-temperature optical spectroscopy in the superconducting state.

The temperature dependence of the gap edge is quite unusual even for strongly coupled electron-phonon superconductors such as Pb where $\Delta_0(T)$ closely follows the BCS curve (see Fig. 44 in Ref. 23). In Cd$_2$Re$_2$O$_7$ $\Delta_0(T)$ remains flat up to about 80% of the $T_c$ and then it drops precipitously to 0 at $T_c$. The imaginary part of the gap at the gap edge has a sharp peak near the temperature where the gap edge starts its rapid decrease. Such temperature dependence can result in systems where the superconductivity is caused by electrons coupling to a low frequency phonon mode as illustrated in Fig. 9.

The results shown in Fig. 9 were obtained for a model for the $\beta$-pyrochlore KOs$_2$O$_6$ where the superconductivity is assumed to result from the coupling to a low frequency anharmonic (rattling) mode. Using a very sharp cutoff Lorentzian model for the electron-phonon coupling function of a “rattler” $\alpha^2 F_R(\Omega) = (g\epsilon/\pi) \{1/[(\Omega - \Omega_R)^2 + \epsilon^2] - 1/(\Omega_c^2 + \epsilon^2)\}$, for $\Omega_R - \Omega_c \leq \Omega \leq \Omega_R + \Omega_c$, and $\alpha^2 F_R(\Omega) = 0$, for $|\Omega - \Omega_R| > \Omega_c$, with the rattling frequency $\Omega_R = 2.2$ meV, the cutoff frequency $\Omega_c = 2.1$ meV, the peak-width parameter $\epsilon = 0.01$ meV, and the the coupling strength parameter $g = 3.312$ (chosen such that $\lambda_R = \int_0^{+\infty} d\Omega \alpha^2 F_R(\Omega)/\Omega = 3$), the Eliashberg equations were solved on the real axis at various temperatures. The real and the imaginary part of the gap, at the gap edge, defined by $\text{Re} \Delta(E = \Delta_0(T), T) = \Delta_0(T)$, are shown in Fig. 9. The transition temperature $T_c$ was obtained by solving the Eliashberg equations on the imaginary axis with the cutoff in the Matsubara sums $\omega_c = 30$ meV and the Coulomb repulsion parameter $\mu^*(\omega_c) = 0.1$. The same $\omega_c$ and $\mu^*(\omega_c)$ were used in the real axis calculations. The resulting
FIG. 9: The real and imaginary part of the gap at the gap edge as a function of temperature obtained for a model electron-phonon spectrum in the form of a very sharp cutoff Lorentzian at 2.2 meV such that the electron-phonon mass renormalization parameter is $\lambda_R=3$ and $k_B T_c/\omega_0=0.24$. The calculated value of $2\Delta_0(0)/(k_B T_c)$ is 5.73.

$T_c$ was 6 K and $2\Delta_0(0)/(k_B T_c) = 5.73$.

Strong electron-phonon coupling may lead to structural PTs as it has been observed for KOs$_2$O$_6$ below $T_c$ and attributed to a freezing of the rattling motion of the K atom\(^{29}\). A detailed examination of our (see Fig. 2) and published heat capacity and upper critical field data of Cd$_2$Re$_2$O$_7$ reveals a change in the slope of these data at $\sim 0.8$ K which could in fact be due to a freezing of low-energy lattice degrees of freedom\(^3\). We rule out Cd or combined Cd–O related vibrations since preliminary results on $^{116}$Cd and $^{18}$O isotope enriched samples, $^{116}$Cd$_2$Re$_2^{18}$O$_7$ and Cd$_2$Re$_2^{18}$O$_7$, showed no isotope effect on $T_c$\(^{33}\). This finding rather suggests the importance of Re related lattice vibrations, possibly of a low-energy Re-lattice breathing mode as discussed by Hanawa et al. in context with the freezing at the high-temperature structural PT\(^3\). Interestingly, ReO$_3$ also exhibits unusually large anisotropic thermal vibrations of the oxygen atoms and a proximity to a low-pressure structural phase
transition\textsuperscript{34}. The observation of anisotropy in the superconducting order parameter $\Delta_0(T)$ in Cd$_2$Re$_2$O$_7$ might be as a result of the anisotropy of the Fermi surface as reported by band structure calculations\textsuperscript{35}.

In summary, we have measured N–S conductance spectra below the superconducting transition temperature of the $\alpha$-pyrochlore superconductor Cd$_2$Re$_2$O$_7$ by soft point contact spectroscopy. We developed and employed an extension of the BTK theory by including the quasiparticle self-energy into the Bogoliubov equations, and thus we were able to fit the conductance spectra as well as derive the temperature dependence and the magnitude of the superconducting order parameter. The magnitude of the gap at $T = 0$ indicates that Cd$_2$Re$_2$O$_7$ is a strong-coupling superconductor. The temperature dependence of the order parameter is markedly different from that of the weak-coupling BCS gap.

Acknowledgments

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