

# The effect of quasiparticle-self-energy on $\text{Cd}_2\text{Re}_2\text{O}_7$ superconductor

F. S. Razavi, Y. Rohanizadegan, M. Hajialamdari, M. Reedyk, R. K. Kremer, and B. Mitrović

**Abstract:** The magnitude and the temperature dependence of the superconducting order parameter  $\Delta(T)$  of single-crystals of  $\text{Cd}_2\text{Re}_2\text{O}_7$  ( $T_c = 1.02$  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 at a value of 0.22(1) meV,  $\approx 40\%$  larger than the BCS value. 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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In a recent review on the surface properties of superconductors for microresonator and its application J. Zmuidzinas [1] stated several issues concerning surface properties of superconductors for application purposes. One of these issues is the broadening of the density of states (DOS) compared to that predicted by the BCS theory. Dynes et al. [2] interpreted correctly this broadening in their experiments on Pb-Bi alloy as a result of the lifetime broadening at finite temperatures. They tried to account for the lifetime broadening by replacing the energy  $E$  in the BCS formula for the DOS by  $E - i\Gamma$ , where  $\Gamma$  is a phenomenological broadening parameter. Subsequently this approach has been applied in many experimental studies where a broadened DOS was obtained (see [3] where some of these works are cited.) However, Mitrović and Rozema [3] pointed out that such treatment of lifetime effects in superconductors cannot be justified microscopically, at least not for conventional strong-coupling superconductors such as Pb or Nb. They proposed [3] that the broadening of the DOS can be accounted for by substituting the real gap  $\Delta$  by a complex gap function  $\Delta(E)$ . We measured the point-contact spectra of  $\text{Cd}_2\text{Re}_2\text{O}_7$  by soft point-contact spectroscopy and modified Blonder-Tinkham-Klapwijk (BTK) theory [4] to fit the experimental data based on complex gap function.

A large number of transition metal (TM) oxides crystallize with the  $\alpha$ -pyrochlore structure with the general composition  $\text{A}_2\text{B}_2\text{O}_7$ , where A is a larger and B is a smaller TM cation. Amongst the  $\alpha$ -pyrochlores,  $\text{Cd}_2\text{Re}_2\text{O}_7$  is the only one which shows superconductivity at  $\sim 1$  K [5, 6]. At room temperature,  $\text{Cd}_2\text{Re}_2\text{O}_7$  exhibits a cubic structure (space group  $Fd\bar{3}m$ ). At  $T_{S1} \sim 200$  K,  $\text{Cd}_2\text{Re}_2\text{O}_7$  undergoes a metal-to-metal second order structural phase transition (PT) to a non-centrosymmetric

tetragonal structure (space group  $I\bar{4}m2$ ) followed by a first order PT at  $T_{S2} \sim 120$  K to another tetragonal structure (space group  $I4_122$ ) [7, 8, 9]. These two PTs have a profound effect on the electronic and the magnetic properties of  $\text{Cd}_2\text{Re}_2\text{O}_7$ . The electrical resistivity and the magnetic susceptibility drop sharply below  $T_{S1}$  [7, 8]. Heat capacity measurements below  $T_{S2}$  showed an exceptionally large electronic Sommerfeld coefficient of  $\gamma = 15$  mJ/(K<sup>2</sup> mol Re) [5, 10]. 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-5d character with electron or hole pockets at various points of the Brillouin zone [11]. However, the band structure of  $\text{Cd}_2\text{Re}_2\text{O}_7$  in the low-temperature structure ( $T < T_{S2}$ ) indicated localized Cd 4d and itinerant Re-5d electrons and a quasi two dimensional Fermi surface [12]. The results of the Re nuclear quadrupole resonance and the Cd nuclear magnetic resonance at low temperature, and in the superconducting state, revealed no magnetic or charge ordering [13]. Just below  $T_c$ , the <sup>187</sup>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  $\sim 0.8$  K, an exponential decrease consistent with weak-coupling BCS theory and an isotropic gap [13]. Vyaselev *et al.* calculated the Wilson ratio and obtained a value of 0.34 indicating a strong electron-phonon coupling incompatible with weak-coupling theory [13]. The strong electron-phonon coupling in  $\text{Cd}_2\text{Re}_2\text{O}_7$  is also implied by a large value of the mass enhancement parameter  $1 + \lambda = \gamma_{\text{exp}}/\gamma_{\text{band}} = 2.63$  extracted from the specific heat measurements and the band structure calculations [14]. The resulting  $\lambda = 1.63$  is comparable to what is found for mercury [15]. The far-infrared spectroscopy measurements on  $\text{Cd}_2\text{Re}_2\text{O}_7$  crystals in the superconducting state at  $\sim 0.5$  K revealed two strong absorption peaks near 9.6 and 19.3 cm<sup>-1</sup> which completely vanish above  $T_c$  possibly indicating strong electron-phonon coupling and the concomitant lifetime broadening in the superconducting state [16].

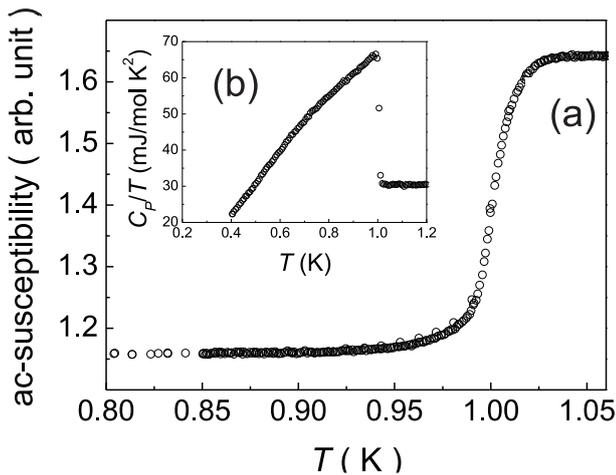
Using point-contact spectroscopy on a single crystals of  $\text{Cd}_2\text{Re}_2\text{O}_7$ , we measured the temperature dependence of the

F. S. Razavi, Y. Rohanizadegan, M. Hajialamdari, M. Reedyk, and B. Mitrović. Department of Physics, Brock University, St. Catharines, Ontario L2S 3A1, Canada

R. K. Kremer. Max-Planck-Institut für Festkörperforschung, Heisenbergstraße 1, 70569 Stuttgart, Germany

superconducting order parameter of  $\text{Cd}_2\text{Re}_2\text{O}_7$  which differs markedly from the BCS temperature dependence of the energy gap. For  $T \rightarrow 0$  K the order parameter approaches a value between 0.22(1) to 0.26(1) meV, i.e.  $2\Delta(0)/k_{\text{B}}T_c > 5.0(1)$  indicating that  $\text{Cd}_2\text{Re}_2\text{O}_7$  is a strong-coupling superconductor.

Single crystals of  $\text{Cd}_2\text{Re}_2\text{O}_7$  were grown from high purity elements, Cd, Re and oxygen, following published procedures. The ac-susceptibility and the heat-capacity measurements proved a superconducting transition at 1.02 K and a width of the transitions of  $\pm 30$  mK (see Fig. 1), consistent with data reported previously [10].

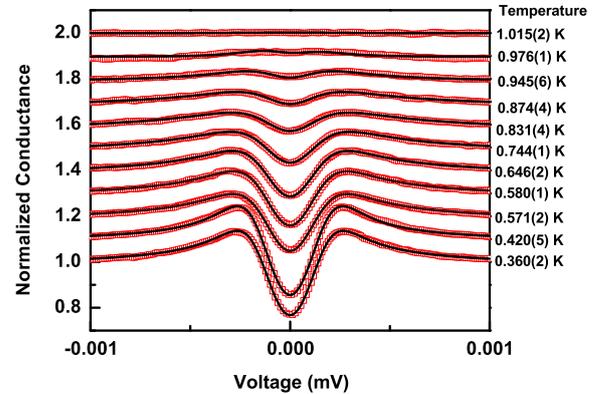


**Fig. 1.** (a) Temperature dependence of the ac-susceptibility of a single crystal of  $\text{Cd}_2\text{Re}_2\text{O}_7$ . (b) Heat capacity of  $\text{Cd}_2\text{Re}_2\text{O}_7$ .

The point-contact spectra were measured by employing the soft point-contact spectroscopy (PCS) method reviewed in detail by Daghero and Gonnelli [17]. The sample was cooled in a home-built single-shot  $^3\text{He}$  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  $^3\text{He}$  and it was measured with a custom calibrated Cernox CX1030 temperature sensor. The stability of temperature measurements were less than  $\pm 3$  mK below and  $\pm 0.7$  mK above 0.7 K, respectively. All data were collected between  $\pm 1$  mV in order to prevent the Joule heating of the sample below 1 K, as the sample temperature increased by about 20 to 50 mK at 2 mV. The accuracy in current measurements is of the order of  $10^{-9}$  A and for a voltage of the order of  $\pm 2 \times 10^{-8}$  V. The measurement error on  $dI/dV$  is of the order of 0.5% (maximum error in  $dI/dV \pm 0.01$ ). The fitting program stops iteration when the total standard deviation of all points is less than  $10^{-8}$ . To reduce the error in  $dI/dV$  calculation we used the 5 points adjacent averaging. However, we observed only a negligible change in the energy gap parameter if no averaging is used.

As the cubic crystal of  $\text{Cd}_2\text{Re}_2\text{O}_7$  can grow along [001], [100], [110], and [111] directions then the associated plane to these directions can be easily identified using the Laue X-ray back scattering. Several measurements were done by injecting a current in the (001), (100), (110), or (111) planes and voltage leads in the same plane or in two different planes. As an example, a set of characteristic normalized conductance spectra collected by injecting the current in the (001) plane with

one of the voltage leads in the the same plane and the other voltage lead and the current lead connected in (100) plane, is displayed in Fig. 2. All spectra were normalized to the value of the conductance  $dI/dV$  measured at a voltages between  $\simeq \pm 2.0$  mV in the normal state at 1.2 K.



**Fig. 2.** (a) Voltage dependence of the normalized conductance ( $\square$ ) for  $\text{Cd}_2\text{Re}_2\text{O}_7$ . For clarity the normalized conductance is shifted by 0.1 relative to the previous temperature indicated on the right. The solid black lines represent the fits obtained using our generalized BTK theory with a complex gap.

An attempt to fit the normalized conductance spectra of  $\text{Cd}_2\text{Re}_2\text{O}_7$  using the Blonder–Tinkham–Klapwijk (BTK) theory [4] without considering the broadening of DOS did not provide satisfactory results, especially for the spectra at temperatures close to  $T_c$  where finite lifetime effects can play an important role [18]. The BTK theory is based on the Bogoliubov equations for the two-component wave function

$$[1] \quad \psi(\mathbf{r}, t) = \begin{pmatrix} u(\mathbf{r}, t) \\ v(\mathbf{r}, t) \end{pmatrix}$$

for particle ( $u$ ) and hole ( $v$ ), 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 the 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 [19, 20, 21]. The resulting theory has the form identical to the BTK theory but with the normalized superconducting quasiparticle DOS given by the so-called Dynes equation [2]  $N_s(E) = \text{Re} \{ (E - i\Gamma) / \sqrt{(E - i\Gamma)^2 - \Delta^2} \}$ , where  $\Delta$  is the superconducting energy gap. However, it was pointed out that the Dynes equation cannot be justified microscopically, at least not for conventional strong-coupling superconductors such as Pb or Nb [3]. In the strong-coupling (Eliashberg) theory of superconductivity the normalized superconducting quasiparticle density of states is given by [22, 23]

$$[2] \quad N_s(E) = \text{Re} \frac{E}{\sqrt{E^2 - \Delta^2(E)}},$$

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

By using the Bogoliubov equations which include the self-energy effects [24], we have generalized the BTK theory. Their time Fourier transform has the form

$$[3] \quad \left\{ \left[ -\frac{\hbar^2}{2m} \nabla^2 - \mu(\mathbf{r}) \right] \tau_3 + \Sigma(\mathbf{r}, E) \right\} \psi(\mathbf{r}, E) = E \psi(\mathbf{r}, E),$$

where

$$[4] \quad \Sigma(\mathbf{r}, E) = (1 - Z(\mathbf{r}, E)) \tau_0 + \phi(\mathbf{r}, E) \tau_1$$

is the  $2 \times 2$  electron self-energy matrix.  $\tau_0$  is a unit matrix and  $\tau_1$  and  $\tau_3$  are Pauli matrices [22, 23]. 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  $\Delta(\mathbf{r}, E)$  is related to the pairing self-energy  $\phi(\mathbf{r}, E)$  and the renormalization function  $Z(\mathbf{r}, E)$  by  $\Delta = \phi/Z$ . 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 which are parallel to the interface, and a  $\delta$ -function potential at the normal metal (N)–superconductor (S) interface) [4], we arrive at a theory which is identical in form with the BTK theory [4] but with the real gap  $\Delta$  replaced by the complex gap function  $\Delta(E)$ . Specifically, the conductance of a N–S interface at a voltage  $V$  is given by

$$[5] \quad \frac{dI_{NS}}{dV} = S \int_{-\infty}^{+\infty} dE \frac{df(E - eV)}{dV} [1 + A(E) - B(E)],$$

$S = 2N(0)ev_F\mathcal{A}$ .  $N(0)$  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  $\mathcal{A}$  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  $v_F$ )

$$[6] \quad A(E) = \frac{|u|^2|v|^2}{|\gamma|^2}$$

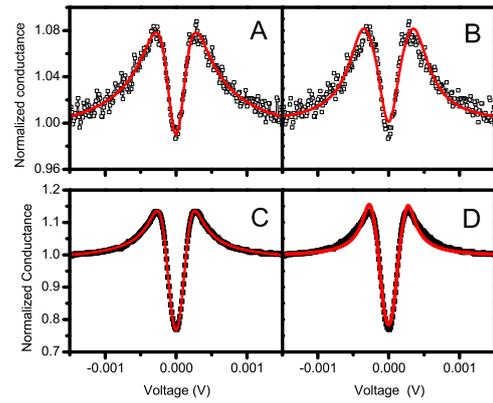
$$[7] \quad B(E) = \frac{[|u|^4 + |v|^4 - 2\text{Re}(u^2v^2)]z^2(z^2 + 1)}{|\gamma|^2}$$

$$[8] \quad \gamma = u^2 + (u^2 - v^2)z^2$$

$$[9] \quad u, v = \frac{1}{\sqrt{2}} \sqrt{1 \pm \sqrt{E^2 - \Delta^2(E)}/E}.$$

The branch of  $\sqrt{E^2 - \Delta^2(E)}$  is taken to have positive real part. 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 as was demonstrated in [18, 3]. 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  $z$ .

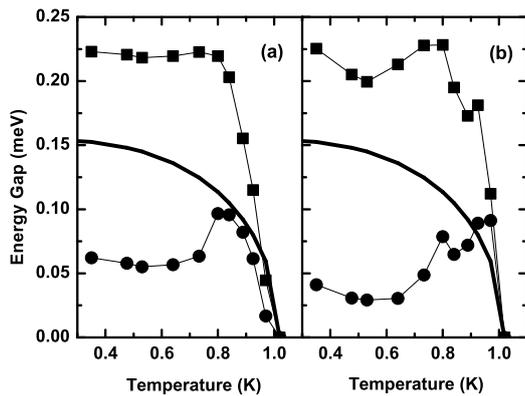
Two characteristic fits to the conductance spectra ( $dI/dV$  vs  $T$ ) at different temperatures are displayed in Fig. 3, panels A and C. For comparison we also show the fits using a



**Fig. 3.** Experimental data ( $\square$ ) and theoretical fits for the normalized conductance spectra of  $\text{Cd}_2\text{Re}_2\text{O}_7$  at 0.831 K (A and B) and 0.360 K (C and D). Solid lines in Figs. 3A and 3C represent the fits obtained using our generalized BTK theory with a complex gap. The  $z$  parameter varies slightly with temperature ( $z = 0.70 \pm 0.03\%$ ). Solid lines in Figs. 3B and 3D show the fits using the phenomenological approach including the Dynes broadening parameter  $\Gamma$  [19, 20, 21]. In 3A and 3B no 5 points adjacent averaging on  $dI/dV$  were carried out.

phenomenological approach [19, 20, 21] based on the Dynes broadening parameter (panels B and D in Fig. 3). We found that theoretical fit using the BTK model modified by including a complex  $\Delta$  provides improved agreement with the experimental data at the higher temperature, in particular at low voltages (see panels A and B in Fig. 3). As pointed out in [3] the Dynes formula for DOS at low energies gives an unphysical constant value  $\Gamma/\Delta$  which can be significant if  $\Gamma$  is not a very small fraction of the gap  $\Delta$ , which is the case near  $T_c$  for a strongly coupled superconductor. On the other hand, the strong coupling formula for DOS at low energies  $E$  gives  $(\Delta_2/\Delta)^2 E$ , where  $\Delta_2$  is the imaginary part of the gap, and it vanishes at  $E=0$ . When the damping rate is low, which might be expected at low temperatures, the two approaches coincidentally give numerically nearly identical results [3].

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. 4a and the caption for measurement procedure). For comparison, we also show in Fig. 4b the energy gap versus temperature obtained by fitting the BTK theory modified by including a phenomenological broadening parameter  $\Gamma$ . The magnitude and 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 of the superconducting order parameter found in this study give a strong indication that  $\text{Cd}_2\text{Re}_2\text{O}_7$  is a strong coupled superconductor with a remarkably large gap resulting in a ratio  $2\Delta(0)/k_B T_c \sim 5$ , which is comparable to values found e.g. in Pb–Bi alloys with very strong electron-phonon coupling [25]. Such a large value of the gap to transition temperature ratio seems inconsistent with a low value of the normal-

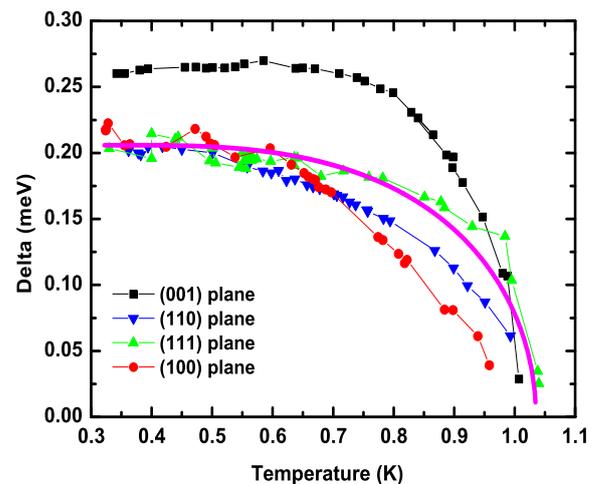


**Fig. 4.** Superconducting energy gap  $\Delta(T)$  of  $\text{Cd}_2\text{Re}_2\text{O}_7$  (square symbols) obtained by fitting the experimental data of Fig. 2 for the normalized conductance at different temperatures using (a) the generalized BTK theory presented in this work and (b) the phenomenological approach [19, 20, 21]. The circles in (a) give the values of the imaginary part of the gap at the gap edge obtained in the fits while the circles in (b) give the Dynes broadening parameter  $\Gamma(T)$ . The thick lines give the BCS temperature dependence of the gap. Here, the current was injected between (001) and (111) planes and voltage was measured between these two planes.

ized jump in the electronic specific heat at transition temperature  $\Delta C_e/\gamma T_c = 1.15$  reported in [10], which is below the BCS value of 1.43. Theoretical studies [26] have shown that for a crystalline superconductor with  $2\Delta(0)/k_B T_c \sim 5$  the normalized jump in the specific heat  $\Delta C_e/\gamma T_c$  should be about 3 (see Fig. 57 in [26]). While in principle it is possible to obtain  $\Delta C_e/\gamma T_c$  below the BCS value in the extreme strong-coupling limit when  $k_B T_c/\omega_{\text{ln}}$  [27] exceeds about 0.7 (see Fig. 59 in [26]), such a large value of  $k_B T_c/\omega_{\text{ln}}$  would imply  $\lambda \gtrsim 10$  (see Fig. 64 in [26]) which is far greater than  $\lambda = \gamma_{\text{exp}}/\gamma_{\text{band}} - 1 = 1.63$  reported in [14]. On the other hand, the gap anisotropy [28] and/or multigap superconductivity [29] could lead to  $\Delta C_e/\gamma T_c$  which is less than the BCS value 1.43. In the case of anisotropic gap  $C_e$  does not approach zero exponentially with decreasing temperature but as a power law. Although the specific heat data on  $\text{Cd}_2\text{Re}_2\text{O}_7$  presented in [10] was limited to the temperatures above 0.37 K, Hiroi and Hanawa used the entropy balance to argue that the specific heat must decrease exponentially with decreasing temperature rather than as a power law.

We also checked for anisotropy in  $\Delta(T)$  for different crystallographic planes as shown in Fig. 5. Both injected and measured voltage are in the same plane. We observed the  $\Delta(T)$  values for all planes saturate at about  $\Delta(T) = 0.22$  meV except for the (001) plane, where the value is slightly larger and saturates at about 0.26 meV. None of  $\Delta(T)$ -curves follows the BCS temperature dependence given by the solid line in Fig. 5, except perhaps the gap for (110) plane. The observed anisotropy in the superconducting order parameter  $\Delta(T)$  in  $\text{Cd}_2\text{Re}_2\text{O}_7$  may be related to the anisotropy of the Fermi surface as reported by band structure calculations [12].

The non-BCS temperature dependence of the gap shown in Figs. 4a and (squares) 5 and a large damping rate as given



**Fig. 5.**  $\Delta(T)$  of  $\text{Cd}_2\text{Re}_2\text{O}_7$  measured for different crystallographic planes. The solid line gives the BCS temperature dependence of the gap.

by the imaginary part of the gap function in Fig. 4a (circles) are quite unusual even for strongly coupled electron-phonon superconductors such as Pb where  $\Delta(T)$  closely follows the BCS curve (see Fig. 44 in [23]). A very similar behavior was observed in the related ternary  $\beta$ -pyrochlore  $\text{KOs}_2\text{O}_6$  ( $T_c = 9.6$  K) using photoemission spectroscopy [30] (see Fig. 3 in [30]), and  $2\Delta(0)/k_B T_c$  for this compound was estimated to be  $\geq 4.56$ . An additional similarity between  $\text{Cd}_2\text{Re}_2\text{O}_7$  and  $\text{KOs}_2\text{O}_6$  is an enhanced value of  $\gamma_{\text{exp}}/\gamma_{\text{band}}$  which for the latter compound is 6–7 [31]. Superconductivity in the  $\beta$ -pyrochlore osmium oxides is usually attributed to a low-energy rattling vibrational mode of the alkali metal atoms. The observed first order structural phase transition occurring below  $T_c$  in  $\text{KOs}_2\text{O}_6$  is attributed to a freezing of the rattling motion of the K atoms [30, 14]. It is tempting to associate the superconductivity in  $\text{Cd}_2\text{Re}_2\text{O}_7$  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 [16]. A detailed examination of our (see Fig. 1) and published heat capacity and upper critical field data of  $\text{Cd}_2\text{Re}_2\text{O}_7$  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 [7]. We rule out Cd or combined Cd–O related vibrations since preliminary results on  $^{116}\text{Cd}$  and  $^{18}\text{O}$  isotope enriched samples,  $^{116}\text{Cd}_2\text{Re}_2^{18}\text{O}_7$  and  $\text{Cd}_2\text{Re}_2^{18}\text{O}_7$ , showed no isotope effect on  $T_c$  [32]. 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.* [7] in context with the freezing at the high-temperature structural PT. Interestingly,  $\text{ReO}_3$  also exhibits unusually large anisotropic thermal vibrations of the oxygen atoms and a proximity to a low-pressure structural phase transition [33].

In summary, we have measured N–S conductance spectra below the superconducting transition temperature of the  $\alpha$ -pyrochlore superconductor  $\text{Cd}_2\text{Re}_2\text{O}_7$  by the 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 to derive the temperature dependence and the magnitude of the superconducting order parameter. The magnitude of the gap at  $T = 0$  indicates that  $\text{Cd}_2\text{Re}_2\text{O}_7$  is a strong-coupling superconductor. The temperature dependence of the order parameter is markedly different from that of the weak-coupling BCS gap.

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