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Do the *intra*molecular C=C stretching vibrational modes in ET mediate electron-pairing in κ-(ET)$_2$X superconductors?

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Isotopic labelling (^{13}C) of all six double-bonded carbon atoms in bis(ethylenedithio)tetrathiafulvalene (BEDT-TTF or ET), which results in large shifts (56–67 cm^{-1}) in the frequencies of *intra*molecular a_g modes at ~1500 cm^{-1}, is found to lead to *no measurable isotope effect on* T_c in the high-T_c ambient pressure organic superconductors κ-(ET)$_2$Cu(NCS)$_2$ (T_c=10.4 K) and κ-(ET)$_2$Cu[N(CN)$_2$]Br (T_c=11.6 K). Thus, the *intra*molecular C=C stretching vibrational modes in ET at energies near 1500 cm^{-1} are not dominant mediators of superconducting pairing in these molecular superconductors.

1. Introduction

The electron-pairing mechanism leading to superconductivity in molecular superconductors, such as the cation-radical salts of bis(ethylenedithio)tetrathiafulvalene (BEDT-TTF or ET) and the anionic alkali-doped buckminsterfullerenes (C$_{60}$), is an important unresolved issue [1–4]. In elemental and binary-alloy BCS-superconductors, the lattice vibrational modes (lattice phonons) mediate electron-pairing. A unique feature of molecular superconductors is the existence of several *intra*molecular vibrational modes, in addition to the lattice phonons. This, combined with the well-known fact that conduction electrons in organic molecular conductors strongly couple to the symmetrical (Raman-active)vibrational modes [5–8], has led several workers to propose that superconductivity in molecular systems may be mediated by the high-energy (500–1500 cm^{-1}) *intra*molecular vibrational modes [4,9–11]. In the ET-based organic superconductors, the relevant a_g modes are ν_3 and ν_4, which appear as the strongest bands in the Raman spectrum of ET at energies of ~1500 cm^{-1}, and ν_{12} and ν_{13}, at energies of ~500 cm^{-1}. The major contributors to ν_3 and ν_4 modes are the central C=C and the ring C=C stretching motions in about equal weight, and the ν_{12} and ν_{13} modes involve the collective C–S stretching motion [4,12].

The involvement of the *intra*molecular vibrational modes in mediating superconductivity can be directly probed by isotopic labelling at selected sites in the ET molecule. The isotopic substitutions result in shift in the frequencies of appropriate *intra*molecular vibrational modes, and thus, the possible involvement of specific vibrational modes in mediating electron-pairing can be determined. We have recently demonstrated [13], in contrast to an earlier report by Merzhanov et al. [14], that ^{13}C-labelling of the central C=C bond in ET does *not* result in any significant isotope effect on T_c in β*-(ET)$_2$I$_3$ (T_c~8 K, 0.5 kbar) within an intrinsic experimental uncertainty of ±0.2 K. Additionally, we have also reported on the absence of any measurable isotope effect, for the same isotopic labelling, in two ambient pressure κ-phase superconductors, κ-(ET)$_2$Cu(NCS)$_2$ (T_c=10.4 K) and κ-(ET)$_2$Cu[N(CN)$_2$]Br (T_c=11.6 K), within an uncertainty of ±0.1 K [15,16].

Substitution of the two central C=C carbon atoms

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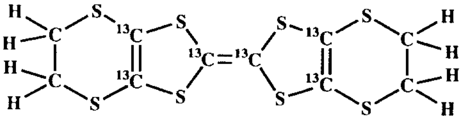
in ET with the ^{13}C isotope results in sizeable shifts in the frequencies (ω) of the ν_3 ($\Delta\omega = -34$ cm^{-1}) and ν_4 ($\Delta\omega = -26$ cm^{-1}) modes. If one assumes that the conventional BCS isotope shift is applicable to coupling involving intramolecular phonon modes, then $T_c \propto \omega$, so that $\Delta T_c/T_c = \Delta\omega/\omega \approx -2\%$, which is close to both the precision of our measurements and to the crystal-to-crystal variations in T_c among samples of the same isotopic content in our previous isotope effect studies. In order to test unambiguously whether the intramolecular modes ν_3 and ν_4 are involved in electron-pairing, we have now extended our isotope effect study to ET in which all six C=C atoms are ^{13}C-labelled, designated herein as ^{13}C(6)-ET. As expected, this substitution results in even larger shifts in the frequencies of ν_3 ($\Delta\omega = -67$ cm^{-1}) and ν_4 ($\Delta\omega = -56$ cm^{-1}) modes, and should give rise to an easily discernible isotope shift of $\Delta T_c/T_c = \Delta\omega/\omega \approx -4\%$, since it is well above our experimental uncertainty as well as the crystal-to-crystal variations in T_c.

We find, in this new study, that there is *no* measurable isotope effect on T_c in the high-T_c ambient-pressure ET-based superconductors, κ-$(ET)_2Cu(NCS)_2$ ($T_c = 10.4$ K) and κ-$(ET)_2Cu[N(CN)_2]Br$ ($T_c = 11.6$ K) within an uncertainty of ± 0.1 K, even when all six double-bonded carbon atoms in ET are ^{13}C-labelled. Our results, therefore, now establish that intramolecular vibrational modes involving the stretching motions of C=C bonds in ET, at frequencies near 1500 cm^{-1}, are not dominant mediators of electron-pairing in these superconductors.

2. Results and discussion

The ^{13}C-labelling of ET used in the present study is shown in the structure below. Both labelled ^{13}C(6)-ET and unlabelled ET were synthesized in identical, parallel experiments starting with $^{13}CS_2$ (nominally 99% enriched, Cambridge Isotope Laboratories, Woburn, Mass.) and unlabelled CS_2 (GFS Chemicals, Columbus, Ohio) respectively, following literature procedures [17–20].



^{13}C(6)-ET

Fourier transform laser desorption mass spectra of the ^{13}C(6)-ET and natural abundance ET are shown in fig. 1. The molecular ions in both these cases appear as clusters of peaks, with the highest intensity peaks at m/z 390 (fig. 1(a)) and 384 (fig. 1(b)) for ^{13}C(6)-ET and natural abundance ET, respectively, as expected. The peaks at higher m/z in these spectra arise due to the isotopes of higher mass (e.g., ^{34}S, ^{13}C) in natural abundance sulfur and carbon atoms. The peaks at m/z lower than 390 in the case of ^{13}C(6)-ET (in fig. 1(a)) arise due to less-than 100% enrichment in the starting $^{13}CS_2$. The intensities of the peaks at m/z 390, 389 and 388 in the ratio 100:13.5:1.25 is indicative of 88% of the ^{13}C(6)-ET product being ET with all six C=C carbons having been labelled with ^{13}C, 12% of the product being ET with five of the six C=C carbons having been labelled with ^{13}C, and a trace (< 1%) of the product being ET with four of the six C=C carbons having been labelled with ^{13}C. This is consistent with ~98% ^{13}C enrichment in the starting $^{13}CS_2$.

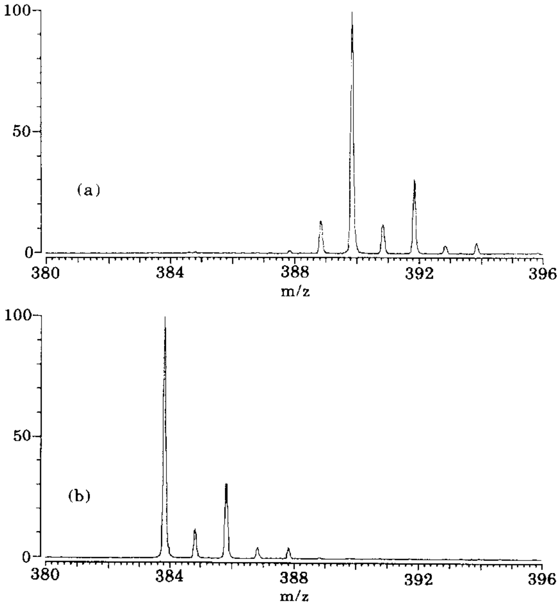


Fig. 1. Fourier transform laser desorption mass spectra of (a) ^{13}C(6)-ET and (b) unlabelled ET.

Fourier transform Raman spectra (λ_{exc} = 1064 nm) in the 1400–1600 cm^{-1} region, taken at liquid nitrogen temperature, of unlabelled ET, ^{13}C(2)-ET and ^{13}C(6)-ET are shown in fig. 2. The ν_3 and ν_4 modes at 1554 and 1492 cm^{-1} in the spectrum of unlabelled ET (fig. 2(a)), according to the normal coordinate analysis of Meneghetti et al., have nearly equal contributions from the central C=C and the ring C=C stretching motions in ET [12]. These frequencies are shifted to 1520 and 1466 cm^{-1} in ^{13}C(2)-ET (fig. 2(b)), and to 1487 and 1436 cm^{-1} in ^{13}C(6)-ET (fig. 2(c)). Therefore, ^{13}C-labelling of both the central C=C and the ring C=C bonds in ET results in substantially larger shifts (56–67 cm^{-1}) in the frequencies of the ν_3 and ν_4 modes than when only the central C=C bond is ^{13}C-labelled (26–34 cm^{-1}) [14,15].

The superconducting salts κ-(ET)$_2$Cu(NCS)$_2$ and κ-(ET)$_2$Cu[N(CN)$_2$]Br were synthesized starting from the unlabelled ET and ^{13}C(6)-ET, in strictly identical and parallel experiments by electrocrystallization, as previously described [21,22]. Superconducting transitions were determined with the use of a commercial AC susceptometer (Lake Shore Cryotronics, Inc.) operated with a modulation field and frequency of 1 Oe and 111.1 Hz, respectively. In these experiments, a single-crystal specimen was supported in a Delrin container and slowly cooled from room temperature to 4 K in a period of about 25 min to avoid the disorder-induced suppression of T_c, as previously reported for κ-(ET)$_2$Cu[N(CN)$_2$Br [23]. The AC susceptibilities, χ, were then recorded at temperature intervals of approximately 0.1 K on warming from 5 K to 20 K. Temperatures were determined with the use of a diode sensor calibrated to an accuracy of ±15 mK or better at temperatures below 20 K. In total, the superconducting transitions of four single-crystal specimens of the two κ-phase salts were determined for each of the two different

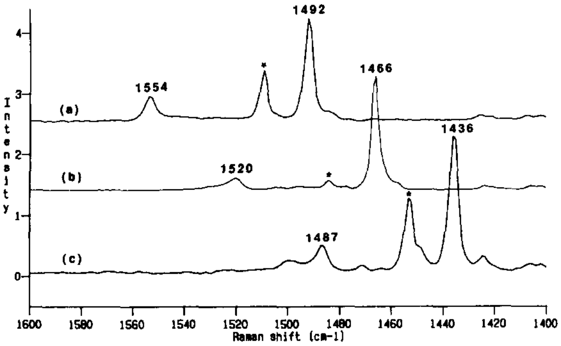


Fig. 2. Fourier transform Raman spectrum (λ_{exc} = 1064 nm) at liquid nitrogen temperature of (a) unlabelled ET, (b) ^{13}C(2)-ET, and (c) ^{13}C(6)-ET. The fundamental modes, ν_3 and ν_4, are marked with appropriate frequencies. The combination modes are marked with asterisks (⋆).

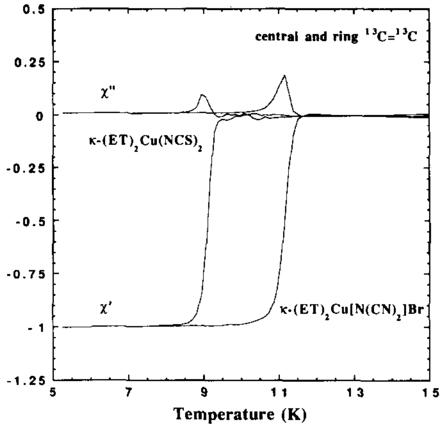


Fig. 3. The superconducting transitions, as determined from AC susceptibility measurements, of κ-(ET)$_2$Cu[N(CN)$_2$]Br and κ-(ET)$_2$Cu(NCS)$_2$ crystals with ^{13}C-labelling in the central and ring double-bonded carbon atoms of ET. The symbols χ' and χ'' refer to the real (in-phase) and imaginary components, respectively, of the volume susceptibility, corrected for demagnetization.

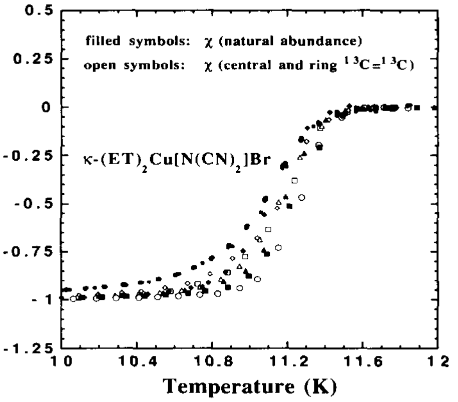


Fig. 4. Point-by-point volume susceptibilities χ' (corrected for demagnetization) for all eight crystals of κ-(ET)$_2$Cu[N(CN)$_2$]Br: four with isotopes of natural abundance (filled symbols) and four with ^{13}C-labelling (open symbols) in the central and ring double-bonded carbon atoms of ET.

isotopic compositions. The results of these experiments are summarized graphically by the superconducting transition curves shown in figs. 3 and 4 and by the list of T_cs reported in table 1.

In order to obtain consistent superconducting transition curves for an accurate comparison of isotope effects, we recorded the AC susceptibilities, both the in-phase or real (χ') and imaginary (χ'') components, as volume susceptibilities (dimensionless) and subsequently corrected these values for demagnetization. The demagnetization corrections are important because the crystals were thin platelets of variable thickness and the in-phase volume susceptibilities, χ', without demagnetization corrections were very large ($\gg 1$) at low temperatures [24]. The volume factors for each crystal were obtained from

Table 1

Superconducting transition temperatures [a] (T_c) at ambient pressure for κ-(ET)$_2$Cu[N(CN)$_2$]Br and κ-(ET)$_2$CU(NCS)$_2$ crystals with compositions of natural isotopic abundance (^{12}C crystals) and with ^{13}C-labeling (^{13}C crystals) in the central and ring C=C atoms in the TTF moiety of ET

κ-(ET)$_2$Cu[N(CN)$_2$]Br

^{13}C crystals	No. 1	No. 2	No. 3	No. 4	Average
T_{co} (K)	11.61	11.61	11.70	11.70	11.66±0.03
T_{cl} (K)	11.41	11.48	11.36	11.43	11.42±0.02
T_{cm} (K)	11.16	11.26	11.14	11.17	11.18±0.03
$\chi''_{\max}$ (K)	11.15	11.28	11.04	11.10	11.14±0.05
^{12}C crystals					
T_{co} (K)	11.89	11.72	11.67	11.63	11.73±0.06
T_{cl} (K)	11.42	11.36	11.38	11.40	11.39±0.01
T_{cm} (K)	11.22	11.10	11.09	11.17	11.14±0.03
$\chi''_{\max}$ (K)	11.21	10.97	11.08	11.19	11.11±0.06
			$\Delta T_c = T_c(^{13}\mathrm{C}) - T_c(^{12}\mathrm{C})$		
		T_{co} (K)	−0.07±0.07		
		T_{cl} (K)	+0.03±0.02		
		T_{cm} (K)	+0.04±0.04		
		$\chi''_{\max}$ (K)	+0.03±0.08		

κ-(ET)$_2$Cu(NCS)$_2$

^{13}C crystals	No. 1	No. 2	No. 3	No. 4	Average
T_{co} (k)	9.50	9.62	9.77	9.77	9.66±0.07
T_{cl} (K)	9.30	9.32	9.35	9.44	9.35±0.03
T_{cm} (K)	9.11	9.15	9.15	9.20	9.15±0.02
$\chi''_{\max}$ (K)	8.96	9.15	9.14	9.21	9.12±0.05
^{12}C crystals					
T_{co} (K)	9.88	9.93	9.44	9.72	9.74±0.11
T_{cl} (K)	9.30	9.47	9.27	9.25	9.32±0.05
T_{cm} (K)	9.06	9.11	9.13	9.08	9.10±0.02
$\chi''_{\max}$ (K)	9.03	9.00	9.10	9.14	9.07±0.03
			$\Delta T_c = T_c(^{13}\mathrm{C}) - T_c(^{12}\mathrm{C})$		
		T_{co} (K)	−0.08±0.13		
		T_{cl} (K)	+0.03±0.06		
		T_{cm} (K)	+0.05±0.03		
		$\chi''_{\max}$ (K)	+0.05±0.06		

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[a] T_{co} is the diamagnetic onset, T_{cl} the linearly extrapolated mean-field T_c (see text), T_{cm} the transition midpoint, and $\chi''_{\max}$ the maximum in the imaginary component of the AC susceptibility.

the mass of the crystal and the density of the crystal calculated from the X-ray unit-cell data [21]. Corrections for demagnetization were obtained in the following manner. First, demagnetization corrections [25] based on cylindrical shapes as a function of length-to-diameter ratios were applied to examine the screening fractions. The length-to-diameter ratios were estimated from the crystal dimensions for which the cross-sectional area perpendicular to the modulation field was expressed as an effective circular area. Although these corrections are approximate, they yielded shielding fractions (constant χ' at low temperatures) within better than 10% of unity in all cases. We then assumed, because of the high purity of our materials, that the screening fraction for each crystal is, in fact, unity, and we adjusted the demagnetization correction accordingly. This is clearly warranted at least in those cases where the approximately corrected values of χ' exceeded unity at low temperatures. The transition curves shown in figs. 3 and 4 and the T_cs listed in table 1 were obtained from such data normalized by a demagnetization factor giving unit screening fraction. Because the demagnetization correction changes the shape of the superconducting transition curve in regions where the susceptibility is large [24], this introduces possible systematic errors in derived values of T_c. We estimate, from an examination of our data for different values of the demagnetization correction, that such systematic errors would not exceed an uncertainty ±0.05 K.

Figure 3 illustrates the complete superconducting transition curves for one ^{13}C-labelled crystal each of κ-(ET)$_2$Cu[N(CN)$_2$]Br and κ-(ET)$_2$Cu(NCS)$_2$. These curves are typical of those determined here for all of the natural-abundance and ^{13}C-labelled crystal specimens. Several "superconducting transition temperatures" (T_cs) can be derived to characterize these curves, and these quantities are summarized in table 1. These T_cs represent the diamagnetic onset temperature (T_{co}), the conventional mean-field definition (T_{cl}), which is the extrapolation to zero susceptibility of the linear portion of the superconducting transition below onset, the transition midpoint (T_{cm}), and the maximum in the imaginary component of the AC susceptibility (χ''_{max}), which is observed to be close to the transition midpoint, T_{cm}. In addition, this table lists, for each definition, the average T_c for the groups of four crystals, the standard deviation in the mean, the isotopic shift, $\Delta T_c = T_c(^{13}C) - T_c(^{12}C)$, and the propagated standard deviation in this temperature difference.

Table 1 shows a variation in T_c from sample-to-sample of the order of 0.2 K or less for T_{cl}, T_{cm}, and χ''_{max}, and considerably more in some cases for the onset temperature, T_{co}, which is difficult to determine with precision. These variations reflect both limitations to the precision in the determinations of T_c and differences in crystal properties. Thus, the propagated statistical errors in the average T_cs represent a combined measure of precision and sample-to-sample variations. The isotope shifts, ΔT_c, derived from the average T_cs show *no* isotope effect significantly larger than the errors, of the order of ±0.05 K for the statistical errors or of the order of ±0.1 K including the possible systematic errors arising from the approximated demagnetization corrections.

Figure 4 shows the point-by-point susceptibilities (χ') in the 10–12 K temperature range for all eight crystal specimens of the κ-(ET)$_2$Cu[N(CN)$_2$]Br salt. This figure illustrates the variability of sample properties within the superconducting state as well as the *absence* of any discernible average isotope effect throughout the complete superconducting transition region. One observes in this figure that the superconducting transitions for seven of the eight crystals are reasonably sharp, having a 10%–90% completion width of approximately 0.5 K. For one crystal specimen with natural isotopic abundance, however, the transition is much broader, having a width of approximately 0.9 K. This tends to weight the average T_cs toward an apparent slight positive (inverse) isotope effect, although one not significantly larger than the statistical errors.

3. Concluding remarks

Our present results, taken together with those of our previous isotope effect studies [13,15,16], rule out the ν_3 and ν_4 modes (at ~1500 cm^{-1}) as being a dominant source of exchange mechanism for electron-pairing. However, the ν_{12} and ν_{13} modes (the other a_g species at ~500 cm^{-1}), involving the collective C–S stretching motions in the ET molecule, may still provide a phonon exchange pathway for electron-pairing. Further isotope effect studies with isotopic labelling at the sulfur atoms of ET are currently underway in our laboratory to investigate that possibility.

Additionally, our results are also consistent either with pairing mediated by *inter*molecular phonons, or with some mechanism of purely electronic origin. Note that for pairing mediated by intermolecular phonons in the conventional BCS mechanism [26], the entire ET molecules vibrate, with both acoustical and optical vibration modes. In this case, the relevant mass for the calculation of the isotope effect is that of the ET molecule, which is increased by 1.56% upon ^{13}C-labelling at the six C=C sites. Hence, with a conventional shift ($\alpha = 1/2$), the expected shift in T_c, $[1-(390/384)^{-1/2}]$, is on the order of 0.77%, or −0.08 to −0.09 K. Such a shift would be indiscernible in our studies, since it is in the same range as our experimental uncertainty and crystal-to-crystal variations in T_c. Further experiments with larger mass changes of the ET molecules would be necessary to investigate the possible role of intermolecular phonons in pairing. Finally, we note that, in the ET-based organic superconductors, the proximity of superconductivity to electronic and/or structural instabilities [1-4] may also account for the absence of any appreciable isotope effect in our observations [27].

Acknowledgements

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References

[1] J.M. Williams, J.R. Ferraro, R.J. Thorn, K.D. Carlson, U. Geiser, H.H. Wang, A.M. Kini and M.-H. Whangbo, Organic Superconductors (Including Fullerenes)-Synthesis, Structure, Properties and Theory (Prentice Hall, Englewood Cliffs, NJ, 1992).

[2] J.M. Williams, A.J. Schultz, U. Geiser, K.D. Carlson, A.M. Kini, H.H. Wang, W.-K. Kwok, M.-H. Whangbo and J.E. Schirber, Science 252 (1991) 1501.

[3] D. Jérome, Science 252 (1991) 1510.

[4] T. Ishiguro and K. Yamaji, Organic Superconductors (Springer, Berlin, Heidelberg, 1990).

[5] C.B. Duke, N.O. Lipari and L. Pietronero, Chem. Phys. Lett. 30 (1975) 415.

[6] N.O. Lipari, M.J. Rice, C.B. Duke, R. Bozio, A. Girlando and C. Pecile, Int. J. Quantum Chem. Symp. 11 (1977) 583.

[7] M.J. Rice and N.O. Lipari, Phys. Rev. Lett. 38 (1977) 437.

[8] C.B. Duke, Ann. New York Acad. Sci. 313 (1978) 166.

[9] K. Yamaji, Solid State Commun. 61 (1987) 413.

[10] A.P. Ramirez, A.R. Kortan, M.J. Rosseinsky, S.J. Duclos, A.M. Mujsce, R.C. Haddon, D.W. Murphy, A.V. Makhija, S.M. Zahurak and K.B. Lyons, Phys. Rev. Lett. 68 (1992) 1058.

[11] A.P. Ramirez, M.J. Rosseinsky, D.W. Murphy and R.C. Haddon, Phys. Rev. Lett. 69 (1992) 1687.

[12] M. Meneghetti, R. Bozio and C. Pecile, J. Phys. (Paris) 47 (1986) 1377.

[13] K.D. Carlson, J.M. Williams, U. Geiser, A.M. Kini, H.H. Wang, R.A. Klemm, S.K. Kumar, J.A. Schlueter, J.R. Ferraro, K.R. Lykke, P. Wurz, D.H. Parker, J.D.B. Sutin, J.E. Schirber, E.L. Venturini and P. Stout, J. Amer. Chem. Soc., in press.

[14] V. Merzhanov, P. Auban-Senzier, C. Bourbonnais, D. Jérome, C. Lenoir, P. Batail, J.-P. Buisson and S. Lefrant, C.R. Acad. Sci. Paris 314 (1992) 563.

[15] K.D. Carlson, A.M. Kini, R.A. Klemm, H.H. Wang, J.M. Williams, U. Geiser, S.K. Kumar, J.R. Ferraro, K.R. Lykke, P. Wurz, S. Fleshler, J.D. Dudek, N.L. Eastman, P.R. Mobley, J.M. Seaman, J.D.B. Sutin, G.A. Yaconi, D.H. Parker and P. Stout, Inorg. Chem. 32 (1992) 3356.

[16] U. Geiser, J.M. Williams, K.D. Carlson, A.M. Kini, H.H. Wang, R.A. Klemm, J.R. Ferraro, S.K. Kumar, K.R. Lykke, P. Wurz. D.H. Parker, S. Fleshler, J.D. Dudek, N.L. Eastman, P.R. Mobley, J.M. Seaman, J.D.B. Sutin, G.A. Yaconi and P. Stout, in: Proc. Int. Conf. on Synthetic Metals, Göteborg, Sweden, August 1992, Synth. Met., to be published.

[17] M. Mizuno, A.F. Garito and M.P. Cava, J. Chem. Soc. Chem. Commun. (1978) 18.

[18] G. Steimecke, H.-J. Sieler, R. Kirmse and E. Hoyer, Phosphorus and Sulfur 7 (1979) 49.

[19] K.S. Varma, A. Bury, N.J. Harris and A.E. Underhill, Synthesis (1987) 837.

[20] P.E. Reed, J.M. Braam, L.M. Sowa, R.A. Barkhau, G.S. Blackman, D.D. Cox, G.A. Ball, H.H. Wang and J.M. Williams, Inorg. Synth. 26 (1989) 386.

[21] A.M. Kini, U. Geiser, H.H. Wang, K.D. Carlson, J.M. Williams, W.K. Kwok, K.G. Vandervoort, J.E. Thompson, D.L. Stupka, D. Jung and M.-H. Whangbo, Inorg. Chem. 29 (1990) 2555.

[22] H.H. Wang, A.M. Kini, L.K. Montgomery, U. Geiser, K.D. Carlson, J.M. Williams, J.E. Thompson, D.M. Watkins, W.K. Kwok and K.G. Vandervoort, Chem. Mater. 2 (1990) 482.

[23] W.K. Kwok, U. Welp, K.D. Carlson, G.W. Crabtree, K.G. Vandervoort, H.H. Wang, A.M. Kini, J.M. Williams, D.L. Stupka, L.K. Montgomery and J.E. Thompson, Phys. Rev. B 42 (199) 8686.

[24] W.R. Goldfarb and J.V. Minervini, Rev. Sci. Instrum. 55 (1984) 761.

[25] G.W. Crabtree, Phys. Rev. B 16 (1977) 1117.

[26] J. Bardeen, L.N. Cooper and J.R. Schrieffer, Phys. Rev. 106 (1957) 162; ibid., 108 (1957) 1175.

[27] R.S. Markiewicz, Int. J. Mod. Phys. B 5 (1991) 2037.