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Carbon Isotope Effect in Single-Crystal Rb_3C_{60}

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We have synthesized single crystals of isotopically enriched Rb_3C_{60}. The very sharp superconducting transitions in single crystal superconducting fullerides allow us to determine the isotope effect on the superconducting transition temperature, T_c, with unprecedented accuracy. We find that the carbon isotope shift exponent α_{carbon} for 99% ^{13}C substitution is 0.21±0.012, significantly smaller than other values reported in the literature[1-6]. This, coupled with the near-zero rubidium isotope effect reported by B. Burk et. al.[7], should place considerable constraints on any theoretical model of superconductivity in the alkali fullerides.

1. EXPERIMENTAL

Commercially available 99% enriched ^{13}C powder was used as a starting material in the preparation of $^{13}C_{60}$. Rods of $^{13}C_{60}$ were formed using a method similar to that reported by C.-C. Chen et.al.[5] The rods were arc-burned in a helium atmosphere to produce fullerene soot. C_{60} was extracted from the soot using HPLC chromatography. A similar batch of natural abundance C_{60} was prepared as a control from graphite rods. It should be noted that as natural abundance carbon is approximately 1.1% ^{13}C, both samples have similar isotopic purity (99%).

Crystals were grown from the C_{60} powder using a vapor transport method under flowing argon. Crystals were intercalated with rubidium following a previously reported method[8].

2. RESULTS

Figure 1 shows the resistive transitions of two samples each of natural abundance carbon and 99% ^{13}C enriched Rb_3C_{60}. The resistively measured transitions are much narrower in temperature than the isotope shift. The transitions are also nearly parallel, reducing the dependence of the measured isotope shift value on the choice of definition of T_c. We chose to define T_c as the maximum in the first derivative of resistance with temperature,

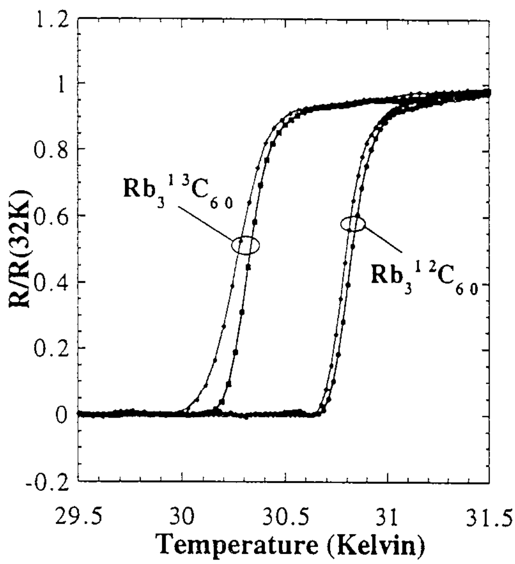

Figure 1. Resistive transitions in $Rb_3{}^{12}C_{60}$ and $Rb_3{}^{13}C_{60}$.

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which gave a value which was most consistent from sample to sample of the same composition. The measured shift in T_c is then 505±30mK. Assuming that the transition temperature depends on the isotope mass to the negative power of α, this gives a value of $\alpha_{carbon}=0.21\pm0.012$.

3. DISCUSSION

We find a value of α_{carbon} lower than, and outside the error margins of, any reported in the literature[1-6]. Using the frequency distribution of the electron-phonon coupling function due to Schluter, et. al.[9], we calculate $\lambda=1.05$ and $\mu^*=0.21$. These values are 20-25% larger than those obtained assuming $\alpha_{carbon}=0.3$, and should place serious constraints on theories of superconductivity in A_3C_{60}.

The need for large values of λ and μ^* to explain the high T_c and small α_{carbon} of Rb_3C_{60} hints that the alkali metal phonons may be playing a larger role in the superconductivity than is indicated by their small isotope effect. The possibility of a large alkali metal mode contribution to λ masked by an anharmonic potential is intriguing. In fact, experiments by our group[7] indicate that α_{Rb} may be negative, as is the hydrogen isotope effect in palladium hydride, where the hydrogen ions see a strongly anharmonic potential. No such experiments to determine the potassium isotope effect in K_3C_{60} have been published; such an experiment could shed light on this possibility.

The sharp transitions in single crystals of A_3C_{60} should also help to elucidate the previously reported[6] possibility of an anomalous isotope shift in isotopically disordered samples.

4. CONCLUSION

We have measured the carbon isotope using high-quality single crystals of 99% isotopic purity. We find $\alpha_{carbon}=0.21\pm0.012$, a value lower than any reported in the literature. This low value of α_{carbon} should have a significant impact on theories of superconductivity in C_{60} compounds, and also raises the interesting possibility of a significant contribution to superconductivity in A_3C_{60} by the alkali metal modes, whose isotope effect may be masked by the effects of an anharmonic potential.

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REFERENCES

1. T. W. Ebbesen, et. al. Nature **365**, 620 (1992)
2. A. A. Zakhidov, et. al. Phys. Lett. A **164**, 355 (1992)
3. A. P. Ramirez, et. al. Phys. Rev. Lett. **68**, 1058 (1992)
4. P. Auban-Senzier, et. al., Synth. Met. **55-57**, 3027 (1993)
5. C.-C. Chen and C. M. Lieber, J. Amer. Chem. Soc. **114**, 3141 (1992)
6. C.-C. Chen and C. M. Lieber, Science **259**, 655 (1993)
7. B. Burk, et. al., Phys. Rev. Lett. **72**, 3706 (1994)
8. X.-D. Xiang, et. al. Science **256**, 1190 (1992)
9. Schluter, et. al. Phys. Rev. Lett. **68**, 526 (1992)