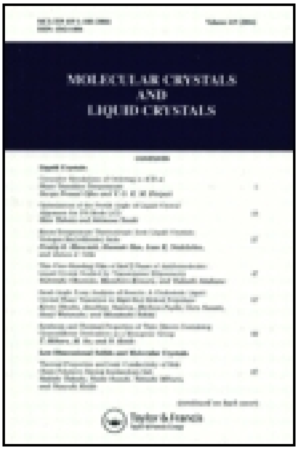

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Anomalous High Pressure Properties in Fullerene Superconductors

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Correlations among lattice parameter, external pressure, and critical temperature T_c has been surveyed on fullerene superconductors with a wide range of lattice parameters and various valence states. The observed value of dT_c/dP for $Na_2Rb_{0.5}Cs_{0.5}C_{60}$ and Li_3CsC_{60}, having small interfullerene separations, is about a few times lager than that of K_3C_{60}. In contrast, small dT_c/dP values are found in fullerides with expanded unit cells, such as $(NH_3)_xNaAA^{\dagger}C_{60}$ (A, $A^{\dagger}$ =K,Rb and Cs) and $A_3Ba_3C_{60}$. Interestingly, the chemical and physical pressure effects on T_c are considerably different in these fullerides with large interfullerene spacings. Our results suggest that the pressure dependence is scaled by an interfullerene distance when the interfullerene distances is small, but that this scaling fails when the unit cell is expanded.

Keywords: fullerene; superconductor; anomalous pressure effect

INTRODUCTION

Correlation between T_c and lattice parameters has provided useful information on the mechanism of fullerene superconductivity, particularly for A_3C_{60} (A=alkali metals) type materials. In this class of materials, T_c is scaled simply by the interfullerene distance. Moreover, the chemical substitution and application of external pressure yield the same relation between T_c and interfullerene distance within 10%[1-3]. This notable universal behavior strongly suggests that T_c of fullerene superconductors is controlled by the density of states at the Fermi Energy within the framework of the BCS weak coupling theory.

Recently, there appeared several superconducting fullerides showing considerably different T_c-lattice parameter relations. For an example, in the trivalent A_3C_{60} compounds, ammoniation yields antiferromagnetic insulator $NH_3K_3C_{60}$[4]. Also, the compounds of $(NH_3)_xNaAA'C_{60}$ ($0 \leq x \leq 1$, A'=K,Rb and Cs) exhibits a negative T_c-lattice parameter correlation which is totally different from the conventional universal relation[5]. While the smaller tetrahedral interstices are occupied by potassium ions, the octahedral site is occupied by a Na-NH_3 cluster. Due to the presence of the NH_3 molecule in the octahedral site, the Na ion is shifted from the center of the octahedral site. The authors suggest that the off-center Na ions in the octahedral site play a crucial role in the deviation from the universal trend. Another interesting T_c-lattice parameter correlation is found in the nonavalent series of fullerides with the chemical formula of $A_3Ba_3C_{60}$[6,7]. This group of compounds form a body-centered-cubic (bcc) cell that is isostructural with A_6C_{60} and Ba_6C_{60}. The interstitial site is randomly occupied by alkali metals and Ba. By changing the alkali species, one can tune the lattice parameter keeping the bcc structure. A common intriguing feature of these systems is that T_c decrease with increasing the lattice parameter under chemical control. Such anomalous behaviors recall a question whether these phenomena are controlled again simply by interfullerene distances, or by other hidden parameters. To investigate this issue, we have performed a measurement of T_c under physical pressure and compared the results with those derived from the chemical substitution.

EXPERIMENTAL

All samples were prepared by the reported method[5-9]. Phase purity was established by a high-resolution synchrotron x-ray powder diffraction and superconducting quantum interference device (SQUID) measurements. To derive a relation among T_c, lattice parameters, and pressure, we measured T_c for superconductivity and lattice parameter under pressure. The former was determined by an ac susceptibility measurement in a piston cylinder type Be-Cu high pressure apparatus. The pressure dependence of lattice parameter has been determined by powder x-ray diffraction measurements using the synchrotron radiations at BL-6C1, Photon Factory, KEK. For the application of hydrostatic pressure, the sample was loaded in a diamond anvil cell with mineral oil as a pressure medium. Typical compressibilities (*dlna/dP*) obtained were 0.0124(3) (GPa^{-1}) for the ammonia including superconductors and 0.0031(2) (GPa^{-1}) for the $A_3Ba_3C_{60}$ type materials, respectively.

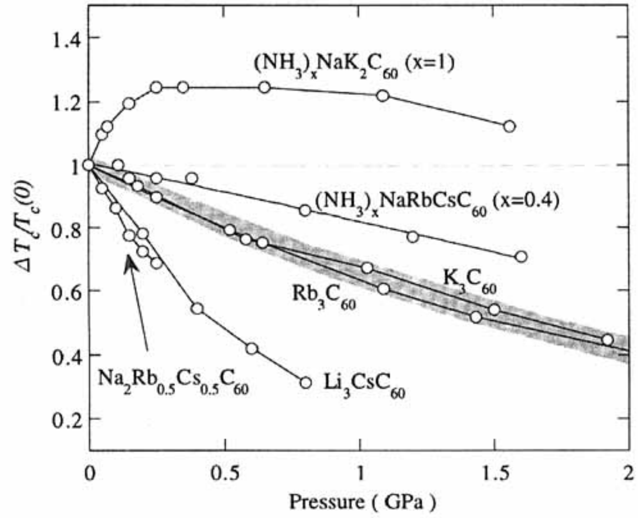


FIGURE 1. Relation between pressure and T_c that is normalized by the ambient value $T_c(0)$ for the trivalent fullerides.

RESULTS AND DISCUSSION

Figure 1 shows the relation between pressure and T_c that is normalized by the ambient pressure value $T_c(0)$ for the trivalent fullerides. Conventional compounds K_3C_{60} and Rb_3C_{60} display almost the same pressure dependence with the initial slope of $dT_c/dp/T_c(0)=-0.3$ (GPa^{-1}), which are in a fair agreement with the literature[2,3]. In the expanded ammoniated trivalent fullerides $(NH_3)_xNaAA^{\dagger}C_{60}$, on the other hand, $dT_c/dp/T_c(0)$ value varies depending on the chemical compositions. The most unique behavior is observed in $(NH_3)_xNaK_2C_{60}$ (x=1). T_c goes up at low pressure, eventually saturates and finally decreases slightly. When the ammonia concentration x is smaller, the $dT_c/dP(P<0.5GPa)/T_c(0)$ value is smaller and the whole pressure dependence approaches to the "normal" behavior as observed in

$(NH_3)_{0.4}NaRbCsC_{60}$ compounds. When we focus on the results above 0.5 Gpa, $dT_c/dp/T_c(0)$ is comparatively smaller in these large spacing fullerides than that of the normal ones. In contrast to this, that of the small lattice compound $Na_2Rb_{0.5}Cs_{0.5}C_{60}$ and Li_3CsC_{60}, is about a few times lager than K_3C_{60}[9]. We also measured the pressure dependence of T_c for the LUMO+1-derived compounds, $K_3Ba_3C_{60}$ and Ba_4C_{60} (Figure 2). In this system, the changing of $dT_c/dp/T_c(0)$ in Ba_4C_{60} is larger than that of the expanded $K_3Ba_3C_{60}$. These results suggest the possibility that the pressure dependencies are scaled by interfullerene distance.

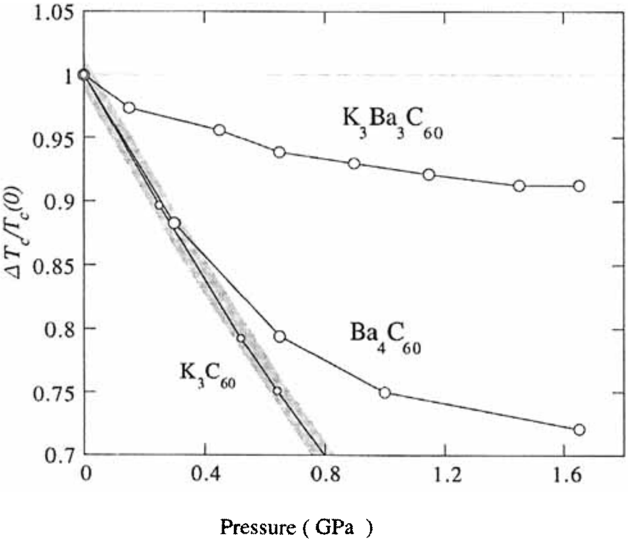
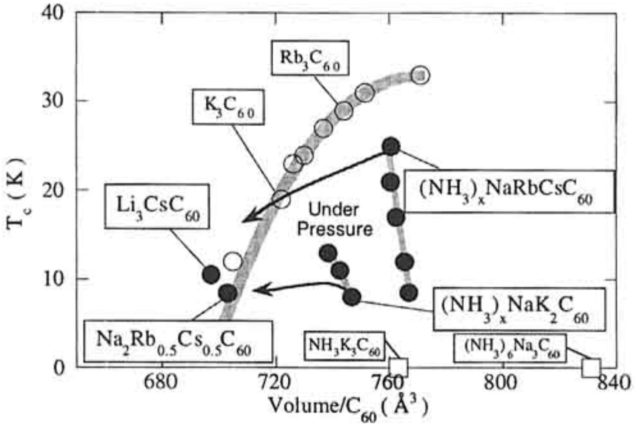
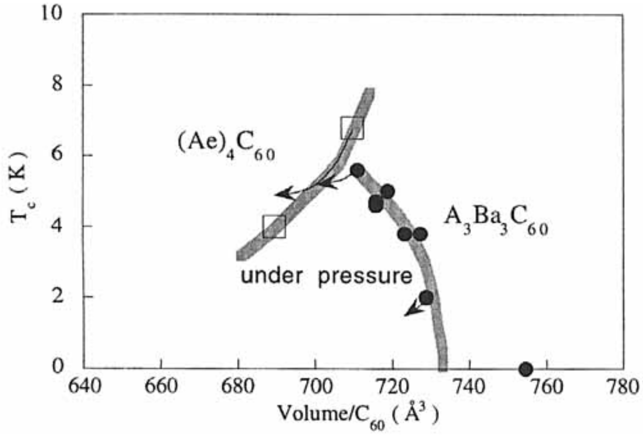


FIGURE 2. $\Delta T_c/T_c(0)$ vs external pressure for the LUMO+1-derived superconductors, $K_3Ba_3C_{60}$ and Ba_4C_{60}.

Figure 3 summarizes the relations between T_c and lattice parameters for trivalent superconductors (a) and $(Ae)_4C_{60}$ (Ae=alkaline earth metals) and $A_3Ba_3C_{60}$ superconductors (b), taking account of the pressure dependence of lattice parameters [10]. Plots are given both for the chemical and physical control of lattice parameters. Shaded lines display the relation between T_c and volume/C_{60} obtained by chemical control, while thin lines with arrows are those derived by a physical control (application of external pressure). It is to be noted that T_c for $K_3Ba_3C_{60}$ and $Rb_3Ba_3C_{60}$ decreases with the reduction of the unit volume. This trend is totally opposite to the chemically obtained relation. The similarity of both materials is that the anomalous pressure properties have been observed only in the large interfullerene distance regime.



(a) the ammoniated trivalent fullerides



(b) $(Ae)_4C_{60}$ and $A_3Ba_3C_{60}$ superconductors

FIGURE 3. The anomalous pressure effects in (a) the ammoniated trivalent fullerides and (b) $(Ae)_4C_{60}$ and $A_3Ba_3C_{60}$. Shaded lines display the relation between T_c and volume/C_{60} obtained by chemical control, while thin lines with arrows are those derived by physical control.

CONCLUSIONS

We have shown that T_c-lattice parameter correlation obtained by the chemical and physical means considerably differs from each other both for $(NH_3)_xNaAA'C_{60}$, and $A_3Ba_3C_{60}$ compounds. Our results indicates that the pressure dependencies are possibly scaled by an interfullerene distance only in the case of small interfullerene separations and that this scaling qualitatively fails when the unit cell is expanded. The present finding contrast in a striking way with the empirical rule, implying that, in the large interfullerene distance regime, T_c is not controlled simply by lattice parameters.

Acknowledgments

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