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AC SUSCEPTIBILITY INVESTIGATIONS OF SUPERCONDUCTING DOPED FULLERENES A_xC_{60}

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Abstract We present investigations of the real and the imaginary parts of the complex ac magnetic susceptibility $\chi = \chi' - i\chi''$ of granular fulleride superconductors K_3C_{60} and Rb_3C_{60} . The magnetic response $\chi(T)$ has been studied as a function of the alternating magnetic field amplitude (0.01 G - 20 G), the driving field frequency (10 Hz - 1 kHz) and external magnetic dc fields (up to 14 T). From this we determined the upper and lower critical fields and the critical current densities.

INTRODUCTION

The discovery of superconductivity in doped fullerenes A_xC_{60} ($A_x = Ba_6^1$, Ca_5^2 , K_3^3 , Rb_3^4 , $RbCs_2^5$) with transition temperatures T_c up to 33 K for $RbCs_2C_{60}$ revived the study of vortex dynamics using the ac magnetic susceptibility ⁶⁻⁸. In the case of these granular systems this contact-free method is a powerful tool. Here we apply this technique to K_3C_{60} and Rb_3C_{60} .

Sample Preparation

K_3C_{60} and Rb_3C_{60} were prepared from phase pure C_{60} (99.99 %) ⁶ according to the method of Mc Cauley et al. ⁹. In order to enhance the homogeneity of the samples the doped C_{60} powder was pressed (20 MPa) and sintered during the preparation. Grain sizes larger than 500 Å can be estimated from X-ray diffraction investigations. The average particle size of these granular material determined by scanning electron microscopy was (10 ± 5) μm. Another Rb_3C_{60} sample was prepared without using external pressure from a RbTl alloy and a mixture of C_{60} and 10% C_{70} ¹⁰.

RESULTS

The microstructure of the investigated samples can be described as arrays of superconducting grains weakly coupled by Josephson junctions acting like a second phase ($H_{c1}^j, H_{c2}^j, j_c^j$). Passing through the Shubnikov phase of the single grains and the Josephson matrix, respectively, strongly influences the normal-to-superconducting state transition. This is reflected in a steplike change in the real part $\chi'(T)$ and dissipation peaks in the imaginary part $\chi''(T)$. Similar effects are frequently observed in other granular materials like high- T_C ceramics or A15 compounds¹¹. The ac susceptibility results for K_3C_{60} ($T_C=17.8$ K) and Rb_3C_{60} ($T_C=29.5$ K, #1) shown in Fig.1 are typical and compare well to dc susceptibility measurements (SQUID). Moreover $\chi_{ac}'(T)$ lies on top of the zero field cooled curve $\chi_{dc}(T)$.

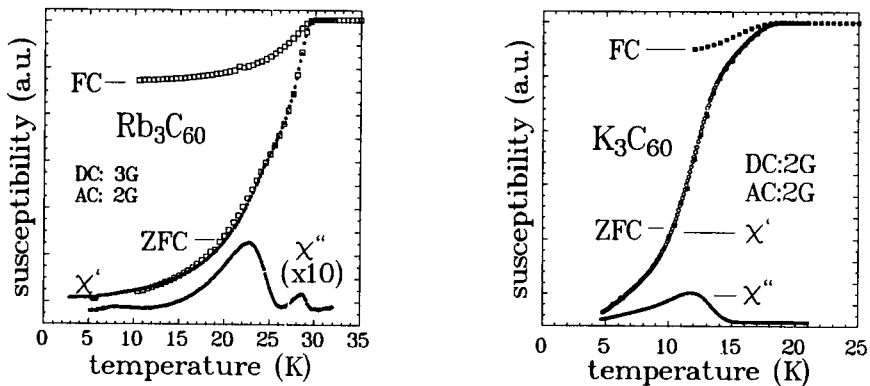


FIGURE 1 Ac susceptibilities of Rb_3C_{60} (left) and K_3C_{60} (right) compared to dc susceptibility results ($B_{ac} \approx B_{dc} \approx 2$ G, $\nu_{ac}=107$ Hz).

Variation of the ac field amplitude

The susceptibility results clearly demonstrate the decoupling of the superconducting grains with increasing ac field strengths (Fig.2). According to Bean's model¹¹ the ac loss peaks correspond to different lower and upper critical fields for the single grain and Josephson matrix. Fig.3 shows the peak fields $B_p(T_p)$ obtained from Fig.2. The critical current density j_c is related to $B_p(T_p)$, by

$$j_c = B_p(T) / (\mu_0 r). \quad (r: \text{radius of particle or grain}) \quad (1)$$

Both samples show nearly the same temperature dependences for the intergrain peak field $B_p \propto (1-T/T_c)^\alpha$ with $\alpha=4.1 \pm 0.2$ for K_3C_{60} and $\alpha=4.7 \pm 0.3$ for Rb_3C_{60} (Fig.4).

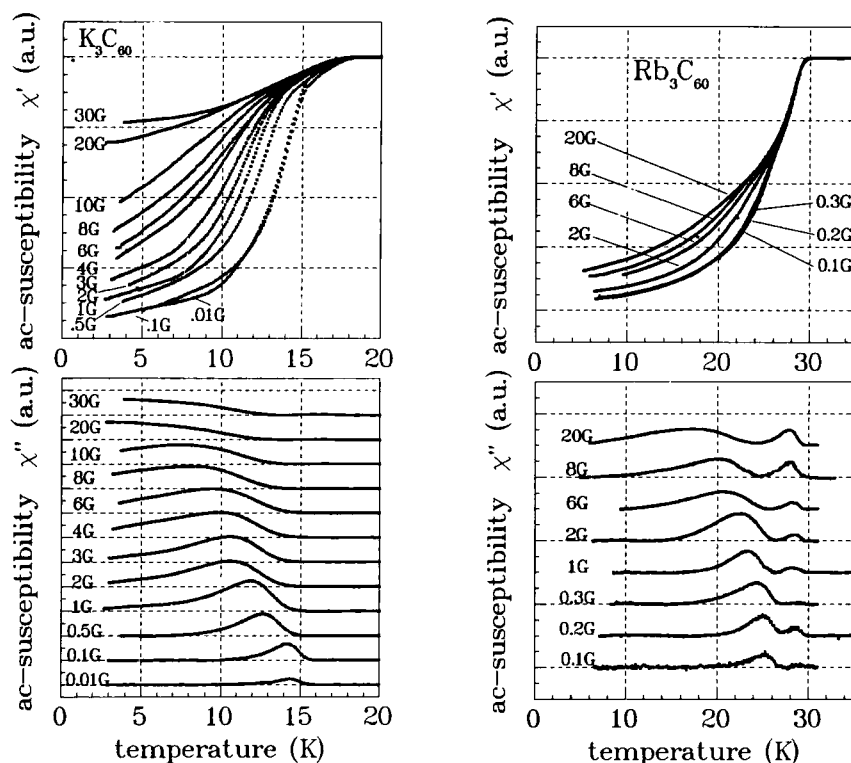


FIGURE 2 Real and imaginary parts of the ac susceptibility as a function of temperature and applied ac field for K_3C_{60} and Rb_3C_{60} , #1, ($\nu=107$ Hz).

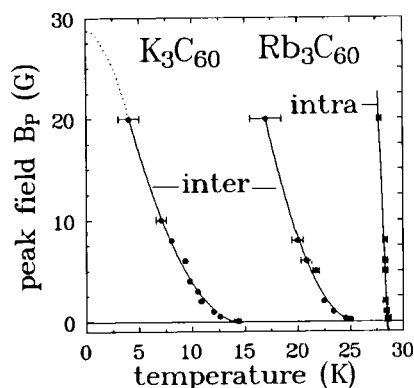


FIGURE 3 Peak fields for A_3C_{60} .

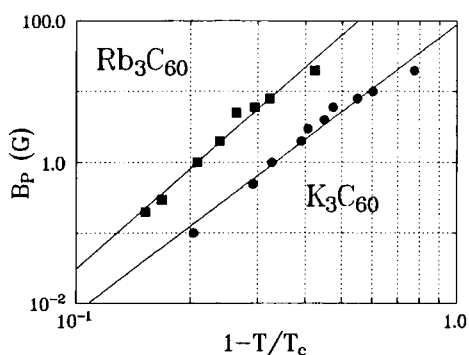


FIGURE 4 Intergrain peak fields.

Frequency dependence

$\chi'(T)$ shows a broadening of the transition width and a shift of the intergrain dissipation peak to lower temperatures with decreasing frequency. T_c and the intra-grain loss peak are not affected. This behaviour could be understood in terms of inter-grain flux creep (with pinning potentials U_0 in the meV range)¹³.

Variation with the external magnetic dc field

A sharp superconducting transition of the single grains can be observed in Fig.5. Furthermore a shoulder in the $\chi'(T)$ curves indicates the transition to the superconducting state of the matrix.

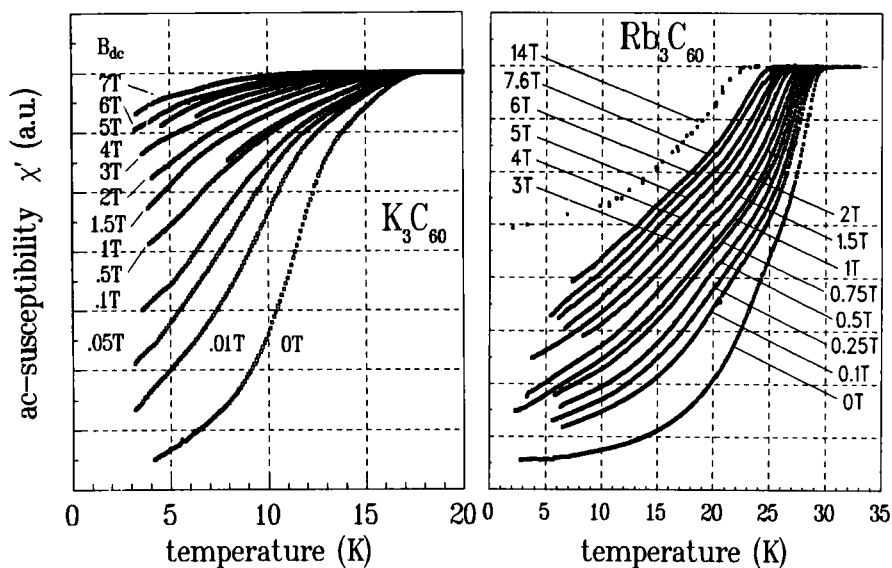


FIGURE 5 Real parts $\chi'(T)$ of the ac susceptibility of K_3C_{60} and Rb_3C_{60} , #1 under various dc magnetic fields ($B_{ac}^{rms}=2$ G, $\nu=107$ Hz).

We determined the upper critical field $B_{c2}(0)$ of the single grains from the temperature dependence of $B_{c2}(T)$ (Fig.6) by the Werthamer-Helfand-Hohenberg relationship¹⁴

$$B_{c2}(0) = -0.69 T_c (\partial B_{c2} / \partial T)_{T=T_c}. \quad (2)$$

$B_{c2}(0)$ is connected to the Ginzburg-Landau coherence length ξ_{GL} through¹⁵

$$B_{c2}(0) = \Phi_0 / (2\pi \xi_{GL}^2). \quad (3)$$

Table I lists these critical parameters.

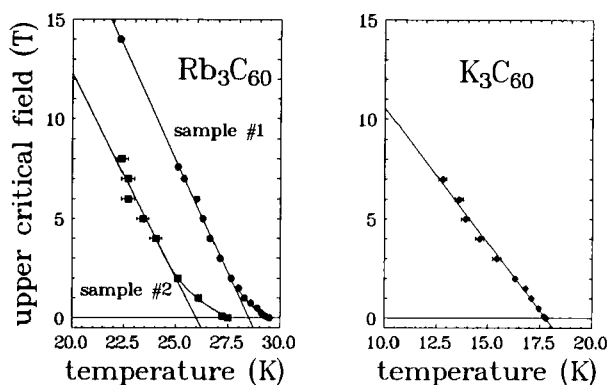


FIGURE 6 Upper critical fields for Rb_3C_{60} and K_3C_{60} .

TABLE I Superconducting quantities of A_3C_{60} compounds.

	T_c K	$\partial B_{c2}/\partial T$ T/K	$B_{c2}(0)$ T	ξ_{GL} Å	$H_{c1}^j(0)$ G	$H_{c1}(0)$ G	$j_c^j(T)$ A/cm ²	$j_c(T)$ A/cm ²
Rb_3C_{60} #1	29,5	$2,3 \pm 0,2$	44 ± 3	27 ± 1	60 ± 10	200 ± 50	$10^4(17K)$	$10^5-10^6(28K)$
Rb_3C_{60} #2	27.5	$2,1 \pm 0,3$	37 ± 6	30 ± 3	no ac loss peaks observed			
K_3C_{60}	17.8	$1,4 \pm 0,3$	17 ± 4	44 ± 1	6 ± 1	-	$10^4(4K)$	

DISCUSSION

The ac susceptibility measurements were strongly influenced by the underlying microstructure of the sample. Both samples, prepared under the same conditions, show nearly identical temperature dependences of j_c^j . The estimated j_c values (Tab.I) are comparable to those of polycrystalline high- T_c bulk material.

ξ_{GL} (Tab.I) exceeds the unit cell dimensions a_0 (≈ 14 Å) and displays the three dimensional nature of the superconductivity in doped fullerenes. An enhancement of $B_{c2}(0)$ and a reduction of the calculated ξ_{GL} value caused by *dirty limit effects* ($\ell_0 \ll \xi_0 \Rightarrow B_{c2}(0) \propto \Phi_0/\xi_0 \ell_0^{15}$, ξ_0 : Pippard coherence length) cannot be ruled out, since small distance (1-10 a_0) intragrain defects, like stacking faults for example, limit the electron mean free path ℓ_0 . This has to be kept in mind, especially in the case of K_3C_{60} where $\xi_0 \approx 150$ Å¹⁶ (this is in contrast to Rb_3C_{60} where ξ_0 is much smaller).

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