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^{87}Rb NMR Relaxation and Vortex Dynamics in Rb_3C_{60}

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Abstract

^{87}Rb NMR relaxation measurements in superconducting fullerene Rb_3C_{60} for various values of the external field are reported. In the normal state the results are in agreement with T_1T= Constant. Below T_c, while no evidence of Hebel-Slichter peak is present in temperature dependence of T_1^{-1}, maxima are observed in T_2^{-1} at $T = 22K$ characteristic of vortex field fluctuation. T_1 shows an anomalous field dependence not yet explained.

1. Introduction

The discovery of superconductivity in alkali metal fullerides A_3C_{60} (A=K,Rb,Cs) has recently attracted considerable interest to the superconducting together with normal state properties of these materials. Their superconducting transition temperatures as high as 33K are the highest known, apart from those of the high T_c ceramics. Alkali metal fullerides are three dimensional, ionic, molecular solids therefore seem to be different also in structure from other known superconductors. The observed superconductivity is well explained in the framework of BCS theory in the weak coupling limit with a gap energy $2\Delta = 3.52\ kT_c$. Also the vortex state properties such as temperature and field dependences were between the subjects of recent studies.

Nuclear Magnetic Resonance (NMR) have been found to provide a wealth of information such as the structure, molecular dynamics, phase diagrams and electronic properties of alkali fullerides. On the other hand, NMR studies on vortex state and its dynamics were concentrated mostly on high T_c superconductors. Flux line dynamics is evidenced by the extra narrowing of the NMR line with respect to the rigid lattice value in $YBa_2Cu_3O_{7-8}$

[1] and $YBa_2Cu_4O_8$ [2]. Other NMR investigations demonstrate the effect of vortex dynamics on the spin-lattice and spin-spin relaxation rates together with characteristic peaks (maxima in T_1^{-1} and T_2^{-1}) as a function of temperature in $HgBa_2CuO_{4+\delta}$ and $YBa_2Cu_4O_8$ [2], [3], [4].

In this paper, we report ^{87}Rb NMR spin-lattice relaxation rate T_1^{-1}, and spin-spin relaxation rate T_2^{-1} measurements for various values of external magnetic field as a function of temperature. The measurements were performed with a homebuilt spectrometer at H_o=1.4 Tesla and with Bruker spectrometers, an MSL 200 for measurements at 2.3 T and 5.9 T and an AMX 400 for H_o=9.4 Tesla.

2. Experimental Results

The Rb_3C_{60} sample was prepared by direct reaction between pure C_{60} and pure Rb resulting from the thermal decomposition of rubidium azide (RbN_3). Superconducting transition temperature T_c measured by the detuning of a resonant circuit was found to be T_c=27.5K at H_o=5.9 T and T_c=28.5 K at H_o=2.3 T.

^{87}Rb NMR spectra is obtained by the Fourier Transformation of half of the echo signal. The spectrum at room temperature seen in Fig 1. shows the typical structure with two lines associated with the tetrahedral (T) and octahedric (O) Rb sites plus a further line attributed to a distorted tetrahedral T′ site.

Since the pulse length maximizing our ^{87}Rb signal is close to half of the $\pi/2$ pulse for a ^{87}Rb nucleus in reference RbCl solution, it is concluded that the observed lines correspond to the $(+1/2 \leftrightarrow -1/2)$ central transition broadened by second order quadrupolar interaction.

The ^{87}Rb nuclear spin-lattice relaxation rates were measured by monitoring the recovery of the magnetization from the echo intensity after a short rf sequence in order to saturate only the $(+1/2 \leftrightarrow -1/2)$ central transition and leave the spin population of other energy levels unchanged. The magnetization recovery was observed not to be described by an exponential law. In the case for a nuclear spin I = 3/2 and magnetic relaxation mechanism due to the fluctuations of the effective magnetic field at the nuclear sites, the recovery law for each site follows the equation [5]

$$y(t) = \frac{s(\alpha) - s(t)}{s(\alpha)} = 0.1e^{-2wt} + 0.9e^{-12wt}, \tag{1}$$

where w is the magnetic transition probability. It is found that in the first decade, where the magnetization grows up to 90 % of its initial value, the recovery is well fitted by the sum of two exponential functions with time constants corresponding to the relaxation times for the two different sites. Since the relative heights of the two components were 2:1 we assigned the long component obtained from the intercept at 0.33 of the normalized recovery law to the O site and the short component to the T site. Since T site has been found to have a relaxation rate close to the one for T′ site at room temperature, all data is analysed in terms of the two components T_1^O and T_1^T. In Fig.2 we report ^{87}Rb spin-lattice relaxation times so obtained as a function of temperature.

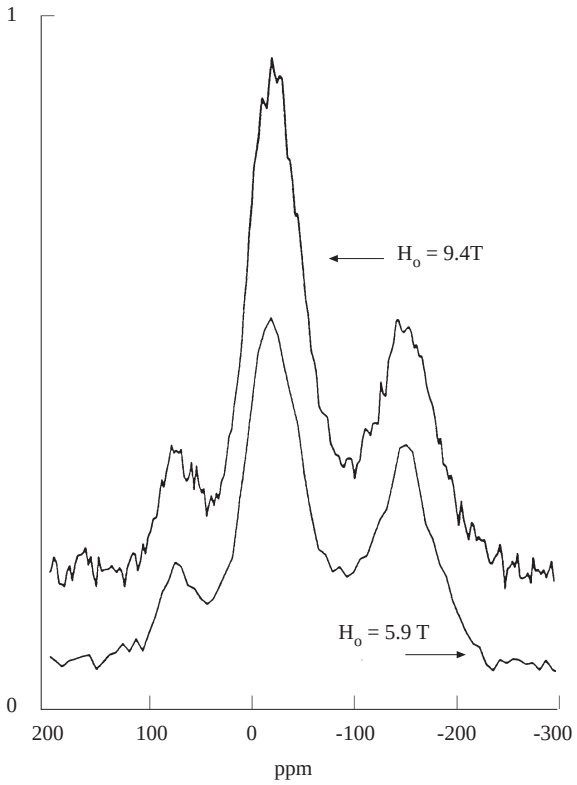

Figure 1. ^{87}Rb NMR spectrum obtained from the Fourier Transform of the echo signal at H_o=9.4 T and H_o=5.9 T.

The ^{87}Rb spin-spin relaxation times were measured by the standard two pulse Hahn echo sequence. In Fig.3 we report ^{87}Rb spin-spin relaxation rates as a function of temperature at H_o=9.4 T and H_o=1.5 T.

The error in spin-lattice and spin-spin relaxation time measurements was about 10%.

3. Analysis of the Data

In metals, the expected spin-lattice relaxation mechanism is the Korringa mechanism in which the nuclear spins are coupled to the noninteracting conduction electron spins through the Fermi contact interaction yielding a temperature independent relaxation [6]:

$$\frac{1}{T_1T} = \frac{\pi k_B}{h}\gamma_N^2 A^2 \; N(E_F)^2. \tag{2}$$

Here, A is the hyperfine coupling constant, γ_N is the nuclear gryomagnetic ratio and $N(E_F)$ is the density of states at the Fermi energy. In the normal state where $T > T_c$, from the ralaxation data in Fig.2, one observes that the product T_1T is nearly constant

as expected for a metal. However, this is contrary to what is found in mixed alkali-doped fulleride Rb_2CsC_{60} where $K^2(T)T_1T = \text{Const}$ holds [7].

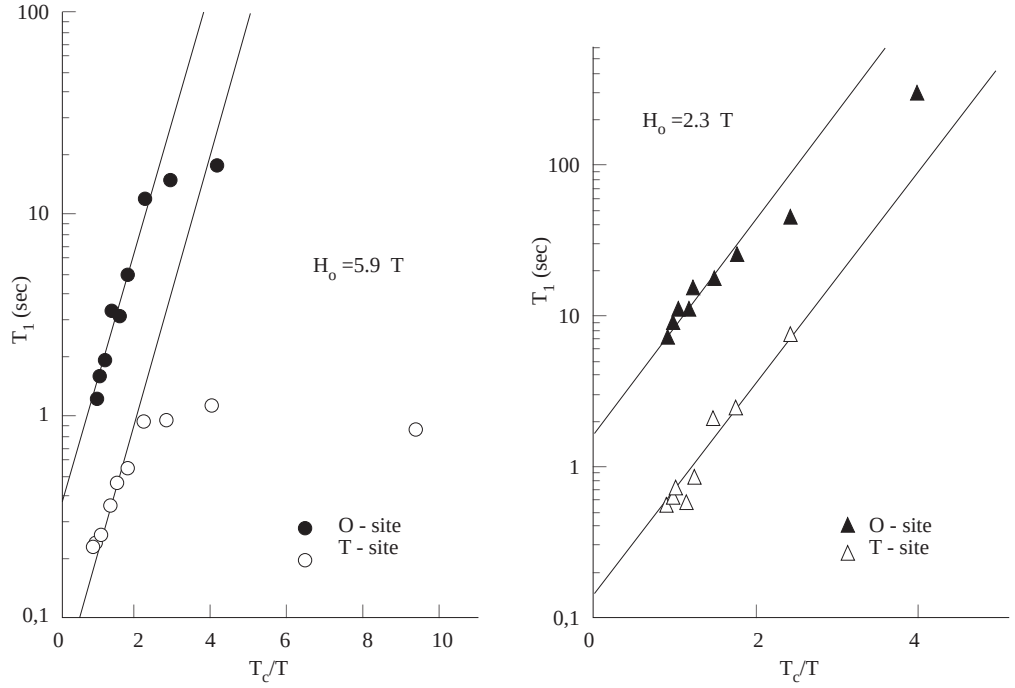

Figure 2. ^{87}Rb Relaxation times for the octahedral and the tetrahedral sites as a function of T_c/T. The solid lines represent the behaviours expected for a BCS superconductor in the limit of weak coupling with an energy gap $\Delta = 45$ K. 2a) $H_o = 5.9$ T, 2b) $H_o = 2.3$ T.

An unexpected result observed in the normal state is the field dependence of the relaxation rates seen in Fig.4, where $(T_1T)^{-1}$ is reported for different values of magnetic fields as a function temperature. The sizable decrease in the relaxation rate on going from $H_o = 9.4T$ to $H_o = 1.4T$ remains unclear since it is opposite to the effect expected in the case of the relaxation due to molecular motions or to relaxation due to magnetic interactions in low dimensional systems.

In the superconducting phase where $T < T_c$, from the temperature behaviour of relaxation, there is no evidence for the coherence peak or Hebel-Slichter (HS) peak [8] characteristic of conventional superconductors which would appear as an increase in $(T_1)^{-1}$ with decreasing temperature just below T_c. At this point one should remark that in Rb_3C_{60} the occurance of HS peak was reported only in low field (1.5 T) μ-SR experiments [9]. Several possibilities such as the quenching effect due to the field [10], pair-breaking interactions [11], [12-14], gap anisotropy [11], d-wave pairing [15] were suggested as the cause of the suppression of HS peak in various superconductors.

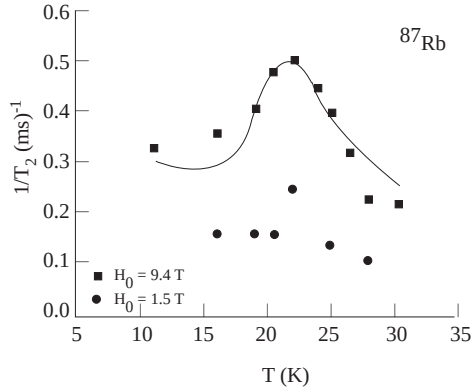

Figure 3. ^{87}Rb spin-spin relaxation rates T_2^{-1} at $H_o = 9.4$ T and $H_o = 1.5$ T. The solid line is the theoretical fit as explained in the text.

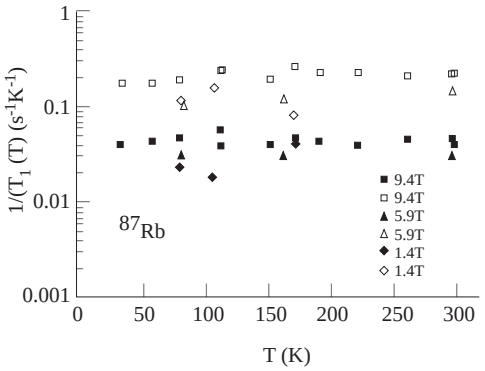

Figure 4. $(T_1T)^{-1}$ above T_c for different values of the external magnetic field as a function of temperature. Open and full symbols represent relaxation rates related to O and T sites respectively.

It is observed that T_1^{-1} decreases dramatically below T_c. The data is fitted quite well by the Arrhenius law $T_1^{-1} = \exp(\Delta/T)$ with $\Delta = 45$ K both for $H_o = 5.9$ T and $H_o = 2.3$ T. Such a temperature dependence is consistent with the opening of an energy gap 2Δ for electronic excitations in the superconducting state. The $2\Delta = 3.2T_c$ value obtained is in accordance with the BCS result $2\Delta = 3.5T_c$. For lower temperatures a marked departure of the relaxation rate from the Arrhenius law is present.

Regarding the vortex dynamics, our results are controversial. The nuclear spin-lattice relaxation can be affected by the flux lines via two mechanisms. The first one is related to the fast relaxation of the nuclei in the vortex core where the metal is in its normal state followed by spin diffusion to more distant nuclei in the superconducting region. In the rapid spin diffusion limit the core relaxation rate should behave as [16] $R_{1s} = R_{1n}S_n/(S_n+S_s)+R_{1s}S_s/(S_n+S_s) \approx R_{1n}H\xi^2/\Phi_o$ where one assumes that the relaxation in the normal metal R_{1n}, to be much faster than the one in the SC phase. The fact that for $T \leq 7$K the deviation of T_1 from the activated behaviour is much more pronounced in strong fields could be thought as an indication of vortex core relaxation.

The second contribution from the flux lines is a direct mechanism in which the nuclear spins are relaxed by the local magnetic fluctuations due to the thermally excited vortex motions. A peak is expected in T_1^{-1} when an effective correlation time τ_c is of the order of the inverse Larmor frequency, that is when the condition $\tau_c \approx \omega_L^{-1}$ is met [2], [3]. A similar effect can also be induced in the temperature bahaviour of spin-spin relaxation rate T_2^{-1} with a maximum [3].

As can be seen from Fig.3, spin-spin relaxation rate T_2^{-1} displays maxima around $T = 22$ K at $H_o = 9.4$ T and $H_o = 1.5$ T. We postulate that these peaks are due to the motion of the flux lines and try to analyze the data in terms of a simple model involving

pseudo-diffusive motion of flux lines used in references [2] and [3] to model characteristic peaks observed in the temperature dependence of T_1^{-1}. Since the maximum in T_2^{-1} should reflect the dynamical contribution related to the spectral density at zero frequency $[J(\omega) \to 0]$ we obtain the following equation:

$$\frac{1}{T_2} = \left(\frac{1}{T_2}\right)_{\max} \frac{\delta\omega\ \tau_c}{0.63} \ln \frac{\tau_c^{-2} + \delta\omega^2}{\delta\omega^2}. \tag{3}$$

Here, τ_c is a temperature and the field dependent correlation time for vortex fluctuations assumed to follow a thermally activated temperature bahaviour, $\tau_c = \tau_o e^{U/T}$ (U is the activation energy for thermal depinning). The fitting of Eq. (3) to our data taking ^{87}Rb linewidth $\Delta\nu = 10$kHz yielded the curve in Fig.3 with fitting parameters $\tau_o = 3.10^{-10}$ s and $U = 220$ K for $H_o = 9.4$ T. We did not consider fitting for $H_o = 1.5$ T since we have too few points around the maximum at this field. At this point one may wish to note that in an another work [16], for Rb_3C_{60} peaks observed in the temperature dependences of T_2^{-1} around $T = 17.5$ K at $H_o = 7.5$ T and were attributed to vortex daynamics. Their estimated pinning potential value $U = 25$ meV and attempt frequency $\tau_o^{-1} = 10^{10} s^{-1}$ are in good agreement with our value.

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