Vortex dynamics in Rb_3C_{60} observed by ^{87}Rb and ^{13}C NMR

G. Zimmer and M. Mehring
2. Physikalisches Institut, Universität Stuttgart, Pfaffenwaldring 57, D-70550 Stuttgart, Federal Republic of Germany

F. Rachdi
Groupe de Dynamique des Phases Condensées, Université des Sciences et Technique du Languedoc, 34060 Montpellier, France

J. E. Fischer
Department of Materials Science and Engineering, University of Pennsylvania, Philadelphia, Pennsylvania 19104
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The vortex dynamics in Rb_3C_{60} is investigated by ^{87}Rb and ^{13}C NMR. It is shown that spin-spin relaxation as well as two-dimensional exchange experiments allow an estimation of the time scale of vortex fluctuations. The effective pinning potential is deduced from the temperature dependence of the NMR parameters. [S0163-1829(96)53030-4]

The superconducting state of A_xC_{60} is interesting in several respects (see, for example, Refs. 1–7). The pairing mechanism was the subject of intense research activity. Furthermore, investigations of magnetic properties, in particular the length scales and critical fields, were performed. Also the vortex state and its dynamics were subjects of recent publications. Different experimental techniques were used to explore its temperature and field dependence. It was noted early on, that vortex dynamics must be taken into account in B_{c2} extrapolations.[8] The nonlogarithmic time decay of the magnetization was a further hint of the vortex state.[9] The effective pinning potential was deduced from measurements of $H(T)$ on the transition boundary between short- and long-range order recently.[10] However, the time scale and amplitude of vortex motion remained unclear.

NMR investigations concerning the dynamic properties of the vortex state concentrated so far on high-T_c superconductors. In a recent two-dimensional (2D) NMR experiment on $YBa_2Cu_3O_{7-\delta}$,[11] the absence of exchange broadening led to the conclusion that long-range diffusion of vortices is not observable. Other NMR investigations demonstrated the influence of vortex state and its dynamics on relaxation rates in Tl and Hg cuprates (for example, Refs. 12 and 13) and organic superconductors.[14,15] Concerning the alkali fullerides, most NMR experiments concentrated on the electronic properties which reflect the opening of the gap and led to an estimation of $\Delta(0)$.[5,6]

In this article, we investigate the vortex dynamics from T_2 relaxation and demonstrate vortex motion by a 2D-NMR exchange experiment. If the vortex lattice has reached the ''rigid lattice'' situation, the field distribution leads to a characteristic asymmetric line shape[16,17] which has been demonstrated in classical[18,19] and high- T_c superconductors.[12] We have observed similar asymmetric ^{87}Rb line shapes in Rb_3C_{60}.[20] With decreasing temperature the penetration depth $\lambda(T)$ decreases, leading to an increased broadening. If the fluctuation of the local field due to vortex dynamics exceeds the broadening, dynamical averaging occurs resulting in line narrowing. More direct evidence for the vortex motion is, however, obtained from a 2D-NMR exchange experiment as will be discussed below.

The sample, a nominal Rb_3C_{60} compound, was prepared similar to the procedure outlined in Ref. 21. The onset T_c value of the superconducting transition was found to be 25–26 K in a 7.8 T field, which is in agreement with the reduction of T_c in high magnetic fields. Care was taken to prevent heating of the sample by strong rf pulses. The measurements were performed with home-built pulsed NMR spectrometers at different Larmor frequencies (7.8 and 4.3 T). T_2 relaxation times were measured by a Hahn echo sequence, incrementing the delay time between the two pulses.

The ^{87}Rb T_2^{-1} relaxation rate as a function of temperature is shown in Fig. 1 for the different lines of Rb_3C_{60}(O: octahedral, T: tetrahedral, T': special tetrahedral). In the superconducting state, where the different lines are indistinguishable, the labels indicate the left-hand side (O line), the maximum (T line) and the right-hand side (T' line) of the spectrum. Two main features are visible. There is a peak in the relaxation rate and a steady increase in T_2^{-1} with decreasing temperature. Since the difference in line shift of the O, T, and T' line, caused by the Knight shift, decreases with decreasing temperature in the superconducting state, flip-flop processes become allowed between neighboring Rb spins, leading to an increase in T_2^{-1}.

The narrow relaxation peak, observed at 17.5 K (7.8 T) will be discussed in the following. The equilibrium position of Rb^+ ions in octahedral sites should be off center with respect to regular positions as proposed by Drechsler *et al.*[22] and observed experimentally in extended x-ray-absorption fine structure (EXAFS) and neutron scattering experiments,[23–25] but not in x-ray diffraction.[26] A low activation energy was proposed, indicating a dynamic disorder at high temperatures. At low temperatures, the freezing could cause a relaxation peak. However, we discard this mechanism for being responsible for the observed relaxation peak. Instead we hold the vortex dynamics causing field fluctuations responsible for this peak. A relaxation peak in T_2^{-1}should occur, if the condition $\Delta\omega\tau_c \simeq 1$ is met. Here, $\Delta\omega$ is the ''amplitude'' of the fluctuating local field and τ_c the correlation time.

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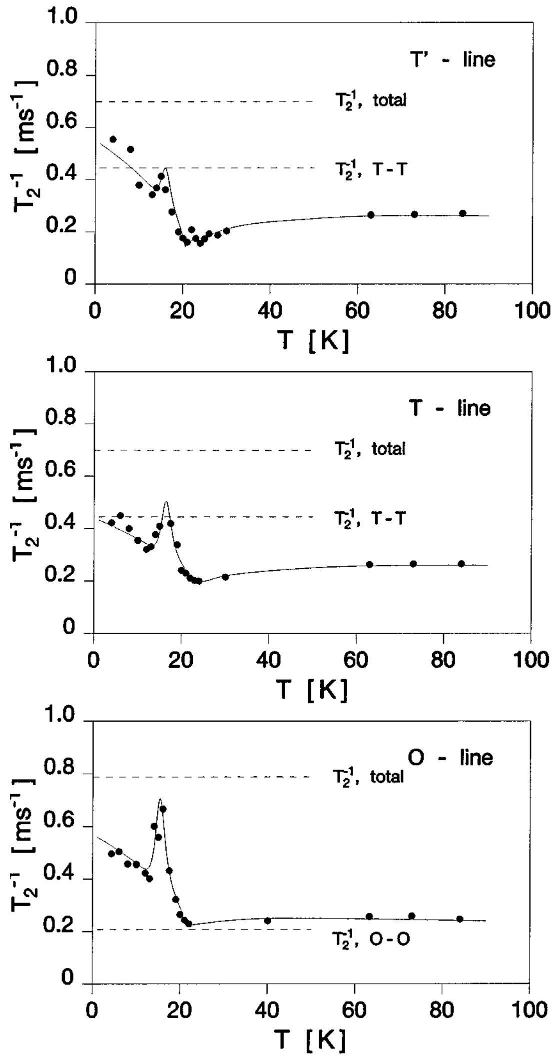

FIG. 1. Temperature dependence of the ^{87}Rb spin-spin relaxation rate for the different lines (O, T, T') observed in the normal state of Rb_3C_{60}(7.8 T). For comparison, the calculated values for T_2^{-1} caused solely by dipolar relaxation are also plotted.

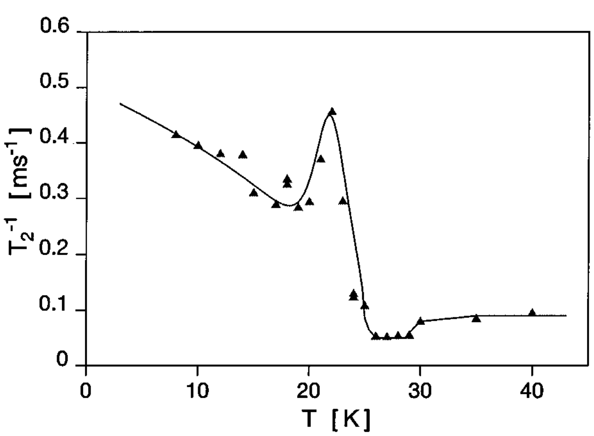

FIG. 2. ^{13}C T_2^{-1} relaxation rate at 4.3 T. Similar features as in the ^{87}Rb relaxation are observed.

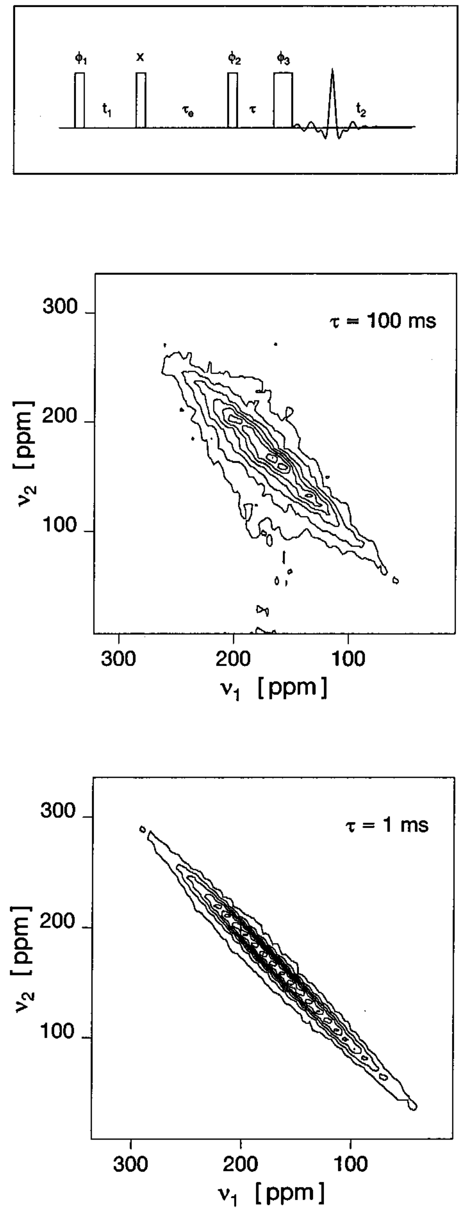

FIG. 3. Pulse sequence of the 2D exchange experiments. The two different time domains for correlation are $t_1$and t_2. Incrementing the exchange time τ_e allows the determination of the frequency of fluctuations. ϕ_i indicate the phases of the different pulses used for phase alternation. Spectra at 21.5 K in a 4.3 T field are shown for two different exchange times τ_e. The off diagonal intensity increases with exchange time, indicating low frequency fluctuations due to vortex dynamics.

In order to identify the relaxation mechanism, measurements on different isotopes and at different fields are necessary. In Fig. 2 the temperature dependence of the ^{13}C T_2^{-1} relaxation rate is presented (4.3 T). As for ^{87}Rb, a sharp relaxation peak is observed, which is shifted to higher temperatures. This temperature dependence is expected in the case of vortex dynamics: The effective pinning potential is strongly field dependent and increases at lower fields. Therefore, the relaxation peak should occur at higher temperatures as is observed. The peak is modeled by the following equation:

$$\frac{1}{T_2}=\Delta\omega(B_0)^2\,\frac{2\tau_c(B_0,T)}{1+\Delta\omega(B_0)^2\tau_c(B_0,T)^2},$$

where τ_c is the temperature and field dependent correlation time of vortex fluctuations and is assumed to follow a thermally activated temperature dependence:

$$\tau_c(B_0,T)=\tau_{c0}\exp\left(\frac{U_{\rm eff}(B_0)}{k_BT}\right).$$

Correlation effects between vortices are neglected within this model. Using an attempt frequency of $\tau_{c0}^{-1}=10^{10}\ \mathrm{s}^{-1}$, a simulation of the peak at 7.8 and 4.3 T leads to the following values for the effective pinning potential $U_{\rm eff}$: 25 meV (7.8 T) and 32 meV (4.3 T). Since the peak in the relaxation rate occurs at a temperature T_p, when the condition $\Delta\omega\tau_c=1$ is satisfied, and depends therefore on the time scale of the experiment, T_p must be distinguished from the irreversibility temperature observed in susceptibility measurements. $T_{\rm irr}$ indicates the cross-over from the low temperature vortex glass to a dynamic vortex state. We therefore expect $T_p>T_{\rm irr}$.

In order to prove this line of reasoning, we have performed a 2D-NMR exchange experiment which measures directly the fluctuation rate of the local magnetic field due to vortex dynamics. In Fig. 3 the pulse sequence of this experiment is shown. The first two pulses constitute a preparatory sequence of duration t_1 which labels the frequency of the nuclei in their different local fields and restores the magnetization back into z direction ($\parallel B_0$). The sequence is followed by an exchange time τ_e, where the spin magnetization evolves in the time dependent local magnetic field. The magnetization is finally detected by a Hahn echo which evolves for time t_2. A 2D Fourier transformation leads to a 2D spectrum which reveals frequency correlations before and after the exchange period: For small values of τ_e, the local magnetic field does not change appreciably, and the frequency ν_2 observed in the detection period t_2 is equal to ν_1. However, if the vortex fluctuation frequency is in the range of $1/\tau_e$, the vortex motion changes the local field during τ_e, leading to off diagonal intensity in the 2D-NMR spectrum. The experiment probes fluctuations in the frequency range $\Delta\omega<1/\tau_e<1/T_1$, where T_1 is the spin lattice relaxation time of nuclear magnetization. Therefore, only temperatures below the T_2^{-1} relaxation maximum can be used for this type of experiment. Spectra at 21.5 K for two different exchange times are shown in Fig. 3. Off diagonal intensity, which proofs a change in local field during τ_e, is observed at larger values of τ_e, indicating ‘‘long-range motion’’ of vortices in this temperature range. Zero point motion would not lead to such off diagonal intensity but instead to only a small broadening. The observation of large off diagonal intensity leads to the conclusion that the fluctuation amplitude of vortices amounts to about intervortex spacing within the exchange time. The fluctuation frequency can be deduced from the broadening of the 2D spectra as a function of exchange time. Alternatively, one can look at the decrease of the central part of the spectrum. Because the integral of the spectrum stays constant, the amplitude decreases, when the spectrum broadens. The time constant of this decay, which can be described by a single exponential, amounts to 0.084 s. Within the independent vortex model, an effective pinning potential can be estimated which amounts to 38 meV using an attempt frequency $10^{10}\ \mathrm{s}^{-1}$. The difference between $U_{\rm eff}$ deduced from T_2 and from the exchange spectra is within experimental error. The rather low attempt frequency of $10^{10}\ \mathrm{s}^{-1}$ points to motion of flux bundles rather than to single vortex motion. In this sense the determined pinning potential should be considered as an effective potential.

In summary we have demonstrated directly large-range vortex hopping by applying a 2D-NMR exchange experiment. The strongly temperature dependent hopping is rather low at 21.5 K and amounts to 12 s^{-1}. It was shown that the peak in the T_2 relaxation is intimately connected with the vortex dynamics and allows the determination of the hopping rate over a wider temperature range, which results in an effective pinning potential. We found the pinning potential to be field dependent as is expected.

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