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## <sup>19</sup>F NMR probe of structural features and flux-line motion in fluorinated Hg-1201

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## Abstract

 $^{19}\mathrm{F}$  NMR spectroscopy has been applied to study local copper environment and flux-line motion in the oxy-fluorine superconductors  $\mathrm{HgBa_2CuO_4F_x}$  with different fluorine content. The temperature dependence of the  $^{19}\mathrm{F}$  NMR linewidth reveals the crossover at 35 K between the low-temperature thermally activated regime with correlation time  $\tau = \tau_0 \exp(U/T)$  and high-temperature "melted" regime with  $\tau = \mathrm{const.}$  © 2000 Published by Elsevier Science B.V. All rights reserved.

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<sup>19</sup>F nuclei provide an excellent microscopic NMR probe of the magnetic flux line (FL) behavior as well as structural peculiarities of the fluorinated Hg-1201 superconductors. We present here for the first time an unambiguous evidence from <sup>19</sup>F NMR that fluorine atoms are successfully incorporated in the superconducting matrix of Hg-1201 with different fluorine content.

For our measurements we used two powder  ${\rm HgBa_2CuO_4F_x}$  samples prepared under different fluorination conditions. The sample no. 1F has  $T_{\rm c}=97\,{\rm K}$  and the sample no. 2F has  $T_{\rm c}=96\,{\rm K}$  [1]. Sample no. 2F has higher fluorine content and is slightly overdoped. The <sup>19</sup>F NMR experiments were performed at a magnetic field of 7.01 T in the temperature range of 4.2–300 K using the Fourier transform of the echo signal after a  $(\pi/2-\pi)$  pulse sequence. The fluorine nuclear spin-lattice relaxation rate was measured using the saturation recovery method.

Typical <sup>19</sup>F NMR spectra of the sample no. 1F measured at different temperatures are presented in Fig. 1. No measurable shift has been found with respect to the reference frequency  $F_0$  reflecting the quasi-2D character

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of superconductivity in the layered cuprates with an extremely low density of charge carriers outside the  $Cu-O_2$  layers. The spectral linewidths  $\Delta v_{1/2}$  at half maximum vs temperature are plotted in Fig. 2. Below  $T_c$  the spectra exhibit rapid broadening with decreasing temperature caused by an inhomogeneous distribution of the magnetic field in the flux-line lattice. This evidences that fluorine atoms are successfully incorporated in the superconducting matrix of Hg-1201.

The remarkable feature of the observed  $^{19}$ F linewidth temperature dependence is the existence of a good pronounced crossover for both samples at around  $T_{\rm cr} \simeq 35\,\rm K$ . A similar crossover has been observed for  $^{89}$ Y and  $^{19}$ F in YBa<sub>2</sub>Cu<sub>4</sub>O<sub>8</sub>: F,  $^{89}$ Y in YBa<sub>2</sub>Cu<sub>4</sub>O<sub>8</sub> [2,3]; and  $^{89}$ Y in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> [4], and was attributed to FL motion. Following this approach the free induction decay in the presence of a random Brownian motion is given by [5]

$$s(t) = e^{-\gamma^2 \langle \Delta B^2 \rangle \tau_c^2 \cdot [\exp(-t/\tau_c) - 1 + t/\tau_c]}.$$

Here  $\tau_c$  is the characteristic correlation time of FL motion;  $\langle \Delta B^2 \rangle$  is the second moment of the magnetic field distribution in the rigid FL lattice. To extract the  $\tau_c$  values the NMR spectra were fitted in Refs. [2-4] by the Fourier transform of Eq. (1). In order to minimize the data manipulation errors we use Eq. (1) directly to fit the half echo intensity decay in the time domain. The

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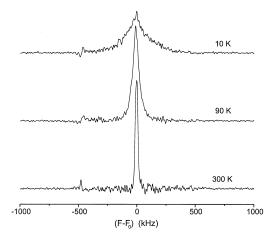


Fig. 1.  $^{19}$ F NMR spectra of the sample no. 1F measured at different temperatures in the magnetic field of 7.01 T. Reference frequency  $F_0$  corresponds to the bare  $^{19}$ F nuclei ( $^{19}\gamma = 40.0538$  MHz/T).

resulting  $\tau_{\rm c}(T)$  dependence is plotted in the insert in Fig. 2. It exhibits a crossover at  $T_{\rm cr} \simeq 35\,{\rm K}$  between the thermally activated FL movement  $(T < T_{\rm cr})$  and the melted regime with  $\tau = {\rm const}~(T > T_{\rm cr})$ .

In conclusion, the <sup>19</sup>F NMR spectroscopy applied to HgBa<sub>2</sub>CuO<sub>4</sub>F<sub>v</sub> samples yields several interesting results:

- (i) Below  $T_{\rm c}$  the spectra exhibit rapid broadening with decreasing temperature which provides for the first time an unambiguous evidence that fluorine atoms are successfully incorporated in the superconducting matrix of Hg-1201.
- (ii) The temperature dependence of the <sup>19</sup>F NMR linewidth reveals the crossover at 35 K between the low-

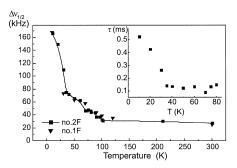


Fig. 2. Temperature dependences of  $^{19}$ F NMR linewidth for the samples  $HgBa_2CuO_4F_x$ . Insert: correlation time  $\tau$  of magnetic FL motion extracted from half echo decay by fitting to Eq. (1) in the time domain.

temperature thermally activated regime and high-temperature melted regime.

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