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Multinuclear NMR/NQR study of $HgBa_2CuO_{4+x}F_y$ superconductors with different oxygen and fluorine content

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Abstract

We have measured $^{63,65}Cu$ NMR/NQR spectra, ^{199}Hg and ^{19}F NMR spectra and spin-lattice relaxation rate on a series of powder $HgBa_2CuO_{4+x}F_y$ samples with different oxygen and fluorine content. Comparison of ^{63}Cu and ^{199}Hg relaxation data to numerical calculations for several types of the order parameter symmetry gives the best coincidence with the d-wave symmetry. ^{19}F NMR line width and relaxation rate are strongly influenced by the magnetic flux-line motion. The ^{19}F magnetization recovery curve is described by a stretched exponential function. The ^{19}F spin-lattice relaxation strongly depends on the fluorine content evidencing that at higher doping level part of F atoms occupy the apical O2 position.

Keywords: NMR; Spin-lattice relaxation

We present ^{63}Cu, ^{19}F and ^{199}Hg NMR results performed on a series of powder $HgBa_2CuO_{4+x}F_y$ samples with different oxygen and fluorine content. The details of the synthesis and fluorination are described in [1]. The temperature dependences of the ^{63}Cu and ^{199}Hg spin-lattice relaxation rate have been numerically calculated using the effective random phase approximation (see Ref. [2], for instance) for different symmetries of the order parameter and $2\Delta/k_BT$ values. The calculations have been performed for the optimally oxygen doped sample $HgBa_2CuO_{4.13}$ below $T_c = 97$ K and compared with the experimental data [3]. We have found that for ^{199}Hg nuclei in the orientation $\boldsymbol{B}_0 \| (a,b)$ plane as well as for ^{63}Cu nuclei in both orientations the experimental results are best described by the $d_{x^2-y^2}$-wave symmetry assuming the gap parameter $2\Delta/k_BT \cong 7$.

For ^{19}F NMR study we used two powder $HgBa_2CuO_4F_x$ samples prepared under different fluorination conditions. The sample no.1F has $T_c = 97$ K and the sample no.2F has $T_c = 96$ K [1]. Sample no.2F has higher fluorine content and is slightly overdoped. ^{19}F NMR experiments have been performed on nonoriented powder samples in a magnetic field of 7 T. As it has been discussed in Ref. [1], the most probable lattice site for fluorine is randomly occupied non-stoichiometric O3 position in the middle of the Hg–Hg mesh. We found that for the sample no.1F in the whole temperature range 10–300 K the echo intensity I is perfectly described by the stretched exponential dependence:

$$I = A - B \cdot \exp(-\tau/T_1)^{0.5}, \tag{1}$$

where T_1 is the spin-lattice relaxation time; τ is the time delay between the saturation sequence and the spin-echo sequence. The stretched exponential function (Eq. (1)) is widely used in the description of relaxation processes in disordered systems like glasses (see Ref. [4], for instance).

In contrast to sample no.1F in sample no.2F with higher fluorine content the recovery of the nuclear magnetization is best described by a double exponential function. The resulting temperature dependences of the spin-lattice relaxation rate R_1(T) for both $HgBa_2CuO_4F_x$ samples are shown in Fig. 1.

We found one order of magnitude difference between rates of the fast and slow relaxation components for the

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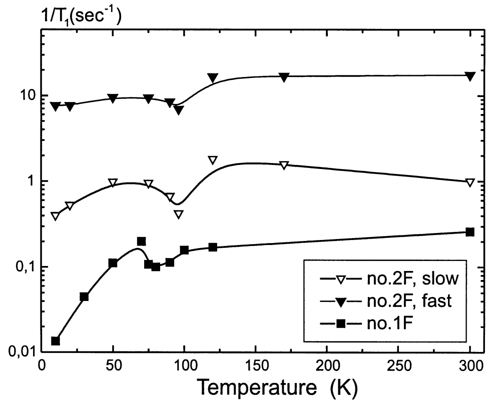

Fig. 1. ^{19}F NMR spin-lattice relaxation rate for two $HgBa_2CuO_4F_x$ samples: (a) sample no.1F, stretched exponential fit; (b) sample no.2F, double exponential fit-slow component; (c) sample no.2F, double exponential fit-fast component.

sample no.2F. This yields an evidence that at higher doping level a part of fluorine atoms occupies an apical O2 position and exhibits a fast nuclear spin-lattice relaxation due to proximity to the copper site. This coincides with the observation of the second pair of copper lines in the NQR spectrum of the sample no.2F.

The existence of the maximum at around $T_{max} \simeq 65$ K has been observed on the $1/T_1$ temperature dependence for the sample no.1F and on both $1/T_1$ components for the sample no.2F. Earlier the similar behavior of the $R_1(T)$ dependence was observed in Y124 and $HgBa_2CuO_{4+\delta}$ oriented powder samples for ^{89}Y, ^{19}F [5] and ^{199}Hg nuclei [6]. As it was shown in Ref. [5] that below T_{max} nuclear spin-lattice relaxation is affected by the thermal fluctuations of the flux lines with the correlation time $\tau_e = \tau_0 \exp(U/T)$, where U is the activation energy in assumption of FL pseudo diffusive motion under a restoring force. It is well known that for orientation $H \perp c$ and with coherence length $\xi_\perp$ of an order of the inter-plane spacing the FLs are self-trapped between the superconducting planes and thus give a negligible contribution to R_1. But this happens only for a very narrow angle range $\Delta\theta$ around the orientation $H \perp c$ of the applied magnetic field: $\Delta\theta < 1^\circ$ in $YBa_2Cu_3O_{7-\delta}$ [7]. Therefore, the FL contribute essentially to nuclear spin-lattice relaxation even for nonoriented powder samples used in our experiments and even for unfavorable partial self-orientation of powder grains towards $H \perp c$ which occurs below T_c due to magnetic anisotropy.

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