Zeeman splitting of the ^{63,65}Cu NQR line in optimally doped $Hg_1Ba_2Ca_2Cu_3O_{8+\delta}$ below T_c

H. Breitzke [a,1], K. Lüders [a], K. A. Lokshin [b] E. V. Antipov [b]

[a] *Physics Department, Free University Berlin, Arnimallee 14, D-14195 Berlin, Germany*

[b] *Dept. of Chemistry, Moscow State University, 119899 Moscow, Russia*

Abstract

We report pure zero field nuclear magnetic resonance (NQR) measurements on the optimally doped three layer compound $Hg_1Ba_2Ca_2Cu_3O_{8+\delta}$ (Hg-1223) with T_c = 134 K. Above T_c two Cu line pairs are observed in the spectrum corresponding to the two inequivalent Cu lattice sites. Below T_c the Cu NQR spectrum shows a splitting of these two line pairs into six line pairs. From a multi exponential spin lattice relaxation with a dependency on the initial conditions, we find a formation of magnetic moments in the CuO layers as the origin of the line splitting. Furthermore, the magnetic moments are *oriented parallel* to the symmetry axis of the electrical field gradient tensor with magnitudes of the order of 1000 G.

Key words: High-T_c superconductivity; Hg-cuprates; NQR; Zeeman splitting

1. Introduction

Most cuprate high-T_c compounds exhibit anomalous broadened Cu NQR spectra at low temperatures [1–3]. Recently, Chakravarty et al. [4] added a further phase to the electronic phase diagram of high-T_c cuprates, by the proposal of the so called d-density wave (DDW) state. One of the observable features of a DDW state is a staggered magnetization, due to the formation of macroscopic currents in the CuO layers may result in a broadening of the Cu NQR spectrum due to a Zeeman splitting of the NQR resonance lines. The HgBaCaCuO family possess a highly ordered crystal lattice and the flatest CuO layers of all cuprate superconductors [6]. Together with the anomalous broadened Cu NQR spectra of this high-T_c family, this compound is an interesting candidate to study wether a stripe phase ordering and/or a magnetic ordering is present.

2. Experimental

The Hg-1223 sample was prepared at the Moscow State University by K. A. Lokshin and E. V. Antipov [5]. The T_c value (134 K) of the sample was determined by ac susceptibility measurements with a Quantum Design susceptometer (PPMS). An impurity content of less than 5 % within the sample was found from neutron powder diffraction and x-ray powder diffraction [5]. The NQR measurements were carried out on a phase-coherent pulsed NQR spectrometer using a standard Hahn spin echo sequence with pulse duration of 4 μs and 8 μs and a pulse separation time of 23 μs. In order to distinguish between the two possible origins of a NQR line splitting (charge segregation/removing of the Kramers degeneracy) we recorded the spin-lattice relaxation curves by employing the method of saturation recovery with different numbers of saturating pulses. The spin-spin relaxation time was recorded by varying the pulse separation of the spin echo sequence.

[1] E-mail:breitzke@physik.fu-berlin.de

3. Results and discussion

Fig. 1 shows the evolution of the ^{63,65}Cu spectra with temperature. Above T_c two pairs of lines occur corresponding to the inner and outer layers of the unit cell. Below T_c, the spectra can not be fitted by using two pairs of resonance lines. Instead a reasonable agreement can be achieved by using six pairs of lines, strongly indicating a splitting of the Cu line of each CuO layer. This assumption is proved by the multi exponential spin-lattice relaxation with a clear dependence on the number of saturating pulses, Fig. 2. Fitting the relaxation curves with double exponential functions results in reasonable good fits but with different time constants for various numbers of saturating pulses. However, the spin-spin relaxation time is purely mono-exponential. This can only be understood by a complete removal of the Kramers degeneracy of the quadrupole levels resulting in an unequally spaced four level system. A stripe formation is excluded from the number of the resonance line pairs and their relative position to each other. We deduced the value of the magnetic field from the frequency spacing of the resonance lines from the expression $B = \delta\nu/(2 \cdot \gamma)$, with γ as the gyromagnetic ratio of the ^{63}Cu nucleus. The results for each CuO layer are shown in Fig. 3. With decreasing temperature the magnitude increases and reaches a maximum value of the order of 800 G at 4.2 K. From the spin-lattice relaxation and the number of resonance line pairs we deduced an orientation of the magnetic moments parallel to the EFGs symmetry axis. The orientation is in agreement with the DDW theorie, whereas the magnitude exeeds the predicted order [4] by a factor of 8. A consistent interpretation of the observed magnetic fields, however, requires further detailed investigatigations especially of the doping dependency of the temperature onset of the magnetic field formation.

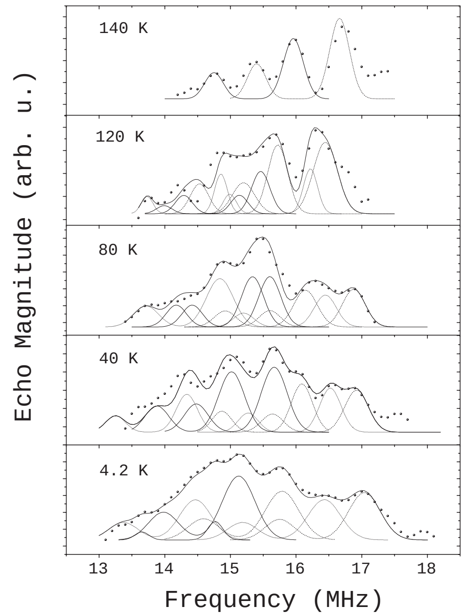

Fig. 1. Measured ^{63,65}Cu NQR spectra for various temperatures (dots). Below T_c the spectra are described by six pairs of lines indicating the splitting of the resonance frequencies.

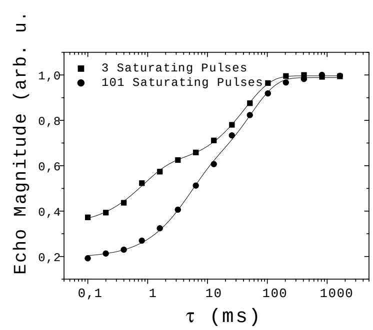

Fig. 2. Longitudinal relaxation curve at $\nu = 16.6$ MHz for three (squares) and 101 (circles) saturating pulses. The solid curve denotes a fit using a two exponential function.

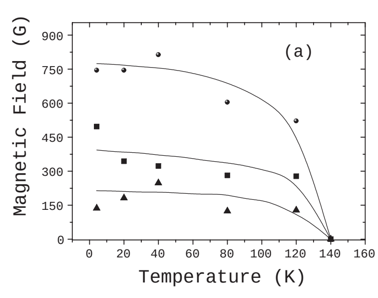

Fig. 3. Temperature dependence of the extracted magnetic field for the CuO layers. Both outer layers are denoted by circles and squares, the inner layer is denoted by triangels. Solid lines are guides to the eyes.

References

[1] Yo Kohori, Ko-ichi Ueda, and Takao Kohara, Physica C **185 189** (1991) 1187

[2] Kenji Fujiwara, Yoshio Kitaoka, Kunisuke Asayama, Hiroyi Sasakura, Sinnosuke Minamigawa, Kiyotaka Nakahigashi, Shigemitsu Nakanishi, Mineo Kogachi, Nobuo Fukuoka, Akira Yanase, J. Phys. Soc. Jpn. **58** (1989) 380

[3] A. A. Gippius, E. V. Antipov, W. Hoffmann, K. Lueders, Physica C **276** (1997) 57

[4] Sudip Chakravarty, R. B. Laughlin, Dirk K. Morr, Chetan Nayak, Phys. Rev. B **63** (2001) 94503

[5] K. A. Lokshin, D. A. Pavlov, S. N. Putilin, E. V. Antipov, D. V. Sheptyakov, A. M. Balagurov, Phys. Rev. B **63** (64511) 2001

[6] J. L. Wagner, B. A. Hunter, D. G. Hinks, J. D. Jorgensen, Phys. Rev. B **51** (15407) 1995