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Spin dynamics in $La_{2-x}Sr_xCuO_{4+\delta}$ probed by electron paramagnetic resonance

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Abstract

We report on EPR experiments on single crystals of $La_{2-x}Sr_xCuO_{4+\delta}$ for Sr-concentrations $0.07 \leqslant x \leqslant 0.20$. Intrinsic EPR signals have been observed for all concentrations. The anomalous temperature dependence of the g-shift and of the line width are explained in terms of a strong coupling of the EPR probe to the spin fluctuations in the CuO_2 planes.

The physics of the cuprate superconductors is dominated by antiferromagnetic spin fluctuations and in modern theories even the pairing interaction is thought to be mediated via magnetic excitations [1]. Recently, electron paramagnetic resonance (EPR) measurements of $YBa_2Cu_3O_{6+\delta}$ single crystals [2] and powder samples of $La_{2-x}Sr_xCuO_{4+\delta}$ [3,4] provided strong evidence for the possibility to characterize the spin fluctuations via EPR-techniques. In this article we present the first intrinsic EPR spectra of undoped single crystals of $La_{2-x}Sr_xCuO_{4+\delta}$ for Sr-concentrations $0.07 \leqslant x \leqslant 0.20$.

The EPR-measurements were performed at X-band frequency (9.1 GHz) on high quality single-crystalline samples [5] for temperatures between 4.2 and 300 K. Well-defined EPR-signals were observed in $La_{2-x}Sr_xCuO_{4+\delta}$ with a Sr-concentration between 0.07 and 0.2. Typical EPR spectra are shown in Fig. 1(a) for a crystal with $x = 0.16$ at 45 K. Upon rotation of the c-axis from parallel ($\Theta = 0°$) to perpendicular ($\Theta = 90°$) with respect to the magnetic field H, the resonance field is shifted by ≈ 0.1 kG. At all angles the spectra could be described with an asymmetric Lorentzian line shape with a dispersion to absorption ratio of about 0.9. The angular dependence of the resonance field for $c \| H$ to $c \perp H$ (Fig. 1(b)) provides strong evidence for the intrinsic origin of the EPR-line and can be described with anisotropic g factors in an $s = \frac{1}{2}$ system with uniaxial symmetry, namely, $g = (g_{\|}^2 \cos^2 \Theta + g_{\perp}^2 \sin^2 \Theta)^{1/2}$ (solid line in Fig. 1(b)) with $g_{\|} = 2.089$ and $g_{\perp} = 2.022$.

The temperature dependencies of the EPR intensities for three different samples as determined by the area under the absorption curve and of the g factors are shown in Figs. 2(a) and (b). The EPR intensity is proportional to the static susceptibility of the EPR probe. As can be seen from Fig. 2(a) a Curie-like behavior describes the low-temperature results for all concentrations (see e.g. the solid line for $x = 0.075$) which points towards localized EPR active centers. Especially the intensities of the samples with higher Sr-concentrations seem to be composed of a constant susceptibility contribution above $T = 50$ K and a Curie contribution below 50 K. We want to mention that a nearly temperature-independent behavior of the EPR intensity was also found on single crystals of $YBa_2Cu_3O_{6+\delta}$ [2].

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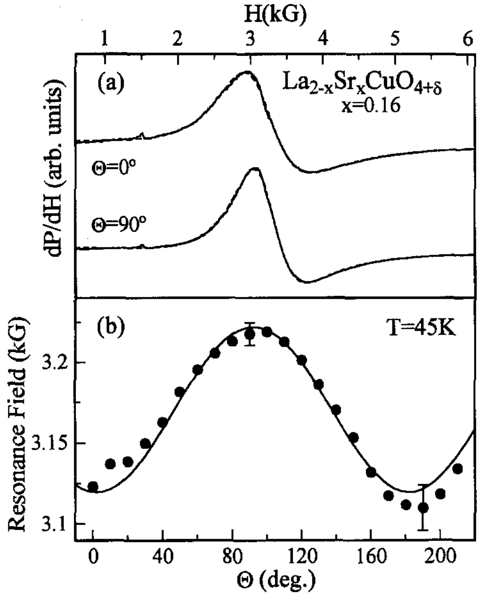

Fig. 1. (a) Absorption derivative $\mathrm{d}P/\mathrm{d}H$ versus magnetic field H in $La_{2-x}Sr_xCuO_{4+\delta}$ at two different crystal orientations at 45 K. The fits with Lorentzian line shapes are indicated by dashed lines. (b) Angular dependence of the resonance field by rotating the c-axis with respect to the magnetic field. The results of the fits as described in the text are indicated by solid lines.

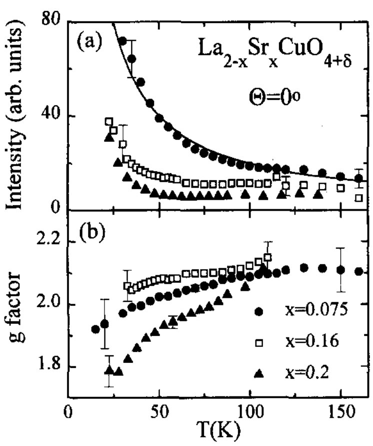

Fig. 2. (a) Temperature dependence of the intensity and (b) of the g factor of the absorption line in $La_{2-x}Sr_xCuO_{4+\delta}$ at a parallel alignment of the c-axis to the magnetic field ($\Theta = 0°$). A Curie-type behavior of the intensity is fitted to the $x = 0.075$ data (solid line in (a)).

The Sr-concentration x clearly influences the temperature dependence of the g factor which determines the resonance field of the EPR spectra. As can be seen in Fig. 2(b), for $T < 120\,\mathrm{K}$ the g factors increase with increasing temperature, an effect that seems to diminish by approaching optimal doping ($x = 0.15$). For $T > 120\,\mathrm{K}$ an almost constant temperature dependence is observed. The g factor mainly is determined by the static susceptibility of the host material of the EPR probe. The temperature dependence of the g factors resembles measurements of the static bulk susceptibility which is determined by the spin fluctuations in the CuO_2-planes [6]. The mechanism of relaxation of the EPR probe is reflected by the width ΔH of the EPR absorption line. Its temperature dependence at a fixed angle of $\Theta = 90°$ is shown in Fig. 3 for three Sr-concentrations. Distinct minima of the line width are observed for all Sr-concentrations at temperatures $T_{\min}$ which depends linearly on the superconducting transition temperature T_c as demonstrated in the inset of Fig. 3. Above $T_{\min}$ the line width increases linearly with 27 G/K without significant dependence on the Sr concentration. This increase reflects the spin-lattice relaxation via delocalized charge carriers (Korringa relaxation).

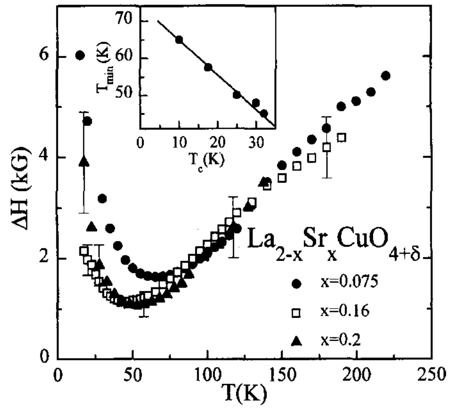

Fig. 3. Temperature dependence of the line width ΔH in $La_{2-x}Sr_xCuO_{4+\delta}$. The inset shows the temperatures $T_{\min}$ of the line width minima vs. T_c.

The steep increase of the line width below $T_{\min}$ seems to be a common feature of all high-T_c superconductors we investigated by EPR measurements so far [2–4]. In Fe^{3+}-doped powder samples, Cieplak et al. [7] and Shengelaya et al. [8] described the line broadening at low temperatures with spin-glass-like freezing which is connected to a charge carrier localization. A different approach successfully described the EPR line width data of $YBa_2Cu_3O_7$ single crystals [2]. There an exchange narrowed EPR line is assumed for EPR probes which are strongly coupled to the spin fluctuations in the plane. This makes it possible to attribute the steep increase of the line width to a strong depression of the spin susceptibility of the CuO_2 plane which is consistent with INS and NMR results.

A proper description of the EPR results in $La_{2-x}Sr_xCuO_{4+\delta}$ has to consider both concepts, carrier localization and spin dynamics. We believe that the EPR probes in the $La_{2-x}Sr_xCuO_{4+\delta}$ samples are holes which are strongly coupled to the Cu spins (magnetic polarons). The steep increase of the line width towards low temperatures can be ascribed to the opening of a spin gap as described in Ref. [2]. Compared with $YBa_2Cu_3O_{6+\delta}$ the much smoother low-temperature line width broadening in $La_{2-x}Sr_xCuO_{4+\delta}$ refers to different spin dynamics in both systems, which is also theoretically proposed by Millis and Monien [9].

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