Intrinsic Electron Paramagnetic Resonance in $La_{2-\xi}Sr_{\xi}CuO_4$: Manifestation of Three Spin Polarons

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Electron-Paramagnetic-Resonance (EPR) measurements on $La_{2-x}Sr_xCuO_4$ single crystals provide experimental evidence of a three-spin polaron of two Cu^{2+} ions and one p hole. The symmetry properties and the peculiar temperature dependence of the g-value of the EPR line indicate the presence of a dynamical Jahn–Teller distortion (Q_2-mode) and formation of a collective mode of polarons and surrounding strongly correlated Cu ions (bottleneck regime).

$La_{2-x}Sr_xCuO_4$ single crystals ($0.05 \le x \le 0.2$) without any paramagnetic impurities show in the temperature range $20\,K \le T \le 300\,K$ a broad but well defined single EPR line. The line shape indicates the typical behavior for metals, i.e., Lorentzian–Dyson form. It does not show any fine or hyperfine structure [1].

The angular dependence of the resonance fields for c-axis parallel to the external magnetic field $c \| H(\Theta = 0°)$ to $c \perp H(\Theta = 90°)$ (Fig. 1a) can be well described with anisotropic g-factors and $S = 1/2$.

$$g_{eff}^2 = g_{\|}^2 \cos^2 \Theta + g_{\perp}^2 \sin^2 \Theta \qquad (1)$$

with $g_{\|} = 2.08\,(9)$ and $g_{\perp} = 2.02\,(0)$.

For a center with $S > 1/2$ and an unresolved fine structure one expects a minimum in the angular dependence of the linewidth near $\Theta \approx 60°$ which clearly is not observed and strictly can be excluded (Fig. 1b). Considering the EPR intensity for the sample with $x = 0.075$, we have estimated that around 1% of the doped holes are included in the formation of the paramagnetic (pm) centers.

The simplest and natural choice for a model of the pm centers is a three-spin polaron (TSP) build up by one O-hole spin and two adjacent Cu spins (Fig. 2). This type of a quasiparticle has been proposed earlier by Emery and Reiter [2] and was considered in detail by Frenkel *et al.* [3].

The most important experimental evidence for this model can be found by an inspection of the temperature dependence of the g-factors (Fig. 3). $g_{\|}$ decreases with decreasing temperature to a rather unusual value $g_{\|} < 2$, showing a crossover with $g_{\perp}$.

In the case of antiferromagnetic coupling between surrounding Cu spins and TSP the value $g_{\|} < 2$ would be obtained with the usual value of Cu-spin susceptibility. However, the linewidth would be much too broad considering only the isotropic exchange. The experimental results can be explained with the assumption that the magnetic relaxation rates $\Gamma_{p\sigma} \gg \Gamma_{\sigma L}$ (p = polaron, σ = surrounding Cu ions, L = lattice), i.e., strong bottleneck limit [4]. An anisotropic part of the interaction contributes both to the linewidth and the effective g-factors even in the very deep bottleneck regime.

By resolving the coupled equations for the transverse magnetizations of the polarons and Cu spins one obtains the following expressions for the effective g-values for the case that the alternating magnetic microwave field is perpendicular to the c axis:

$$g_{\|}(\Theta = 0°) = \frac{\chi_{\perp}^{p} g_{\|}^{p}(1 + \delta\lambda_{\|}\chi_{\|}^{\sigma}) + \chi_{\perp}^{\sigma} g_{\|}^{\sigma}(1 + \delta\lambda_{\|}\chi_{\|}^{P})}{\chi_{\perp}^{p} + \chi_{\perp}^{\sigma}}$$

$$g_{\perp}(\Theta = 90°) = \frac{\chi_{\perp}^{p} g_{\perp}^{p}(1 - \frac{1}{2}\delta\lambda_{\perp}\chi_{\perp}^{\sigma}) + \chi_{\perp}^{\sigma} g_{\perp}^{\sigma}(1 - \frac{1}{2}\delta\lambda_{\perp}\chi_{\perp}^{P})}{\chi_{\perp}^{p} + \chi_{\perp}^{\sigma}} \qquad (2)$$

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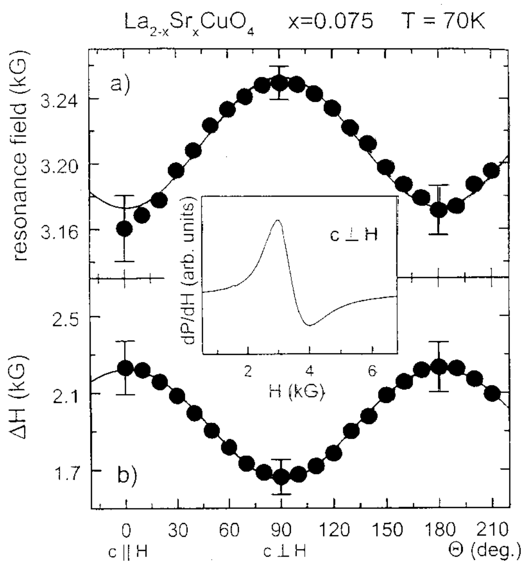

Fig. 1. (a) Angular dependence of the resonance field, and (b) of the linewidth ΔH by rotating the c axis with respect to the external magnetic field. An axial symmetric behavior is indicated by the solid line. The inset shows the EPR spectrum with a Lorentzian line shape for the crystal orientation $c \perp H$.

with

$$\delta\lambda_{\|} = [I_{\|}^{p} - I_{\perp}^{p}] \cdot \frac{1}{g_{\|}^{p} g_{\|}^{\sigma} \mu_B^2}$$

and

$$\frac{\delta\lambda_{\perp}}{\delta\lambda_{\|}} = \frac{g_{\|}^{p} g_{\|}^{\sigma}}{g_{\perp}^{p} g_{\perp}^{\sigma}}$$

Equation (2) demonstrates that in addition to the unusual terms for the bottleneck regime there are contributions $\delta\lambda$ due to the anisotropy of the coupling constants and g-factors. It is noticed that the terms with $\delta\lambda_{\|}$ and $\delta\lambda_{\perp}$ enter with an opposite sign.

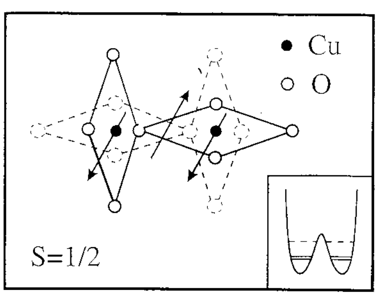

Fig. 2. Three-spin polaron which is regarded as the EPR active center in the CuO_2 plane. The Jahn–Teller distorted polaron has two degenerated configurations as indicated by the dashed line (Q_2-mode). The inset shows the corresponding double-well potential with the excited vibronic state (dashed line) and the ground state split by tunneling (solid lines).

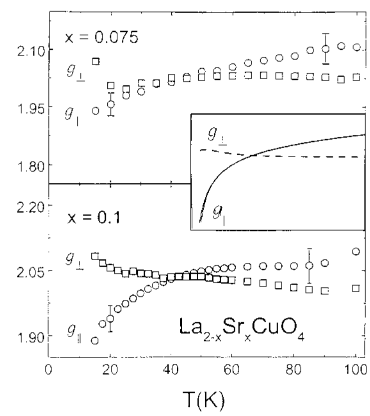

Fig. 3. Temperature dependence of the g values of $La_{2-x}Sr_xCuO_4$ as determined from the resonance field for two different crystal orientations $c \| H$ and $c \perp H$, respectively. The inset demonstrates that the model in connection with formula (2) describes the unusual crossover of the g values satisfactorily.

Equations (2) are consistent with the observed temperature dependence of g-factors, if for the polaron spin susceptibility a Curie law $\chi_{\|,\perp}^{p} = C_{\|,\perp}/T$ and for the Cu-spin susceptibility $\chi_{\|,\perp}^{\sigma}$ the values determined by NMR Knight shift [5]. The quality of the fits is significantly increased, allowing for $g_{\|,\perp}^{p} \neq g_{\|,\perp}^{\sigma}$.

Obviously the polaron is distorted due to a pseudo Jahn–Teller (JT) effect. As a result of the JT effect the ground state of the polaron is a mixture of the functions belonging to the Γ_3 representation. The principal g-values of the polaron for this state are [6]:

$$\begin{aligned} g_1^p &= 2 - 2u \cdot (2 - \cos\alpha - \sqrt{3}\sin\alpha) \\ g_2^p &= 2 - 2u \cdot (2 - \cos\alpha + \sqrt{3}\sin\alpha) \qquad (3) \\ g_3^p &= 2 - 4u \cdot (1 + \cos\alpha) \end{aligned}$$

Hereby, $u = \lambda/\Delta$, λ = spin-orbit parameter, and Δ is mainly determined by the cubic field splitting.

In most cases λ/Δ is of the order -0.1 or less. The normal coordinate Q_2 defines the new variable α by $Q_2 \sim \sin^2\alpha$ [6]. Because of the tunneling between two equivalent configurations of the polaron (Fig. 2) the first two g-values (g_1 and g_2) are interchanging, giving an averaged value $g_{\perp}^{p} = \frac{1}{2}(g_1^p + g_2^p)$, while the third one remains constant $g_{\|}^{p} = g_3^p$. As a result the uniaxial symmetry is restored. The fit parameter for $g_{\|}^{p}$ and $g_{\perp}^{p}$ for the sample with $x = 0.16$ are the following:

$$[I_{\perp} - I_{\|}] = 100\,\text{K}, \; I_{\perp} = -825\,\text{K}, \qquad C = 0.03\,\text{K},$$

$$\Theta_{\|} = 7.5\,\text{K}, \qquad \Theta_{\perp} = 7\,\text{K}$$

The other parameter, i.e., $g_{\|}^{\sigma} = 2.24$, $g_{\perp}^{\sigma} = 2.06$, $u = -0.03$, and $\alpha = 3\pi/4$ are independent on x.

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