Magnetism and superconductivity in $Pr_xY_{1-x}Ba_2Cu_3O_7$

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Using a model of trivalent Pr ions in $Pr_xY_{1-x}Ba_2Cu_3O_7$ with a crystal-electric-field level structure where the three lowest singlets are separated from the others by a large gap of about 500 K, we show that it is possible to account both for the low-temperature staggered magnetization of the antiferromagnetic ordered Pr ions and for the high-temperature magnetic susceptibility. We discuss the suppression of superconductivity by the magnetic moment of Pr^{3+} and show the importance of the antiferromagnetic exchange interactions for pair breaking by ions with singlet-level structure. Exchange-interaction parameters are evaluated.

I. INTRODUCTION

The phase diagram of $Pr_xY_{1-x}Ba_2Cu_3O_7$ shows that Pr ions destroy superconductivity of the 1:2:3 compound [$T_c(x \approx 0.5)=0$] and that the antiferromagnetic (AF) order of the Pr subsystem starts to build up for $x > 0.5$ reaching at $x = 1$ the maximum Néel temperature of 17 K.[1,2] As shown by neutron scattering the zero-temperature magnetic moment of the Pr ion at $x = 1$ is $0.74\mu_B$,[3] while its high-temperature moment is about $2.9\mu_B$ (less than the Pr^{3+} free-ion value of $3.58\mu_B$ and more than the Pr^{4+} value of $2.54\mu_B$).[1,4–6] In the region of $x \approx 0.5$ the metallic character of the conductivity becomes more like that of a semiconductor.[5] This unique behavior of the Pr ions in the 1:2:3 compound raises many questions both on the microscopic and the phenomenological levels. Suggestions of trivalent, tetravalent, and mixed valency of the Pr ions have been made on the basis of various experiments (see, e.g., Refs. 6 and 7). Band-structure calculations[8] as well as high-energy-spectroscopy experiments[7] favor the Pr^{3+} description. It is obvious that because of the singlet character of Pr^{3+} states in the orthorhombic crystal electric field (CEF) its effects are significant both for magnetic properties and pair breaking. Recently Goodman *et al.*[9] demonstrated by inelastic-neutron-scattering measurements together with extensive calculations of the CEF states that Pr is trivalent in $Pr_xY_{1-x}Ba_2Cu_3O_7$. The main feature of the CEF spectrum of Pr^{3+} is a gap of about 500 K which separates the three lowest singlets from the rest of the singlets. The energy differences between these three singlets are an order of magnitude smaller than the gap energy. Goodman *et al.*[9] showed that the CEF naturally explains the high-temperature magnetic moment, which determines the Curie-Weiss susceptibility. In their analysis, however, the exchange interaction between the Pr ions is not taken into account, so the AF order of the Pr ions needs explanation. Besides, the inelastic-scattering peaks for the transitions between the lowest states are too broad and of too low intensity to serve as a reliable source of information about the two lowest-energy splittings. These small splittings control the onset of the AF ordering and the value of the staggered field for a given exchange interaction. These splittings are also of concern for pair breaking because Pr^{3+} ions may destroy the superconductivity via the Abrikosov-Gor'kov mechanism.[10] As shown by Keller and Fulde,[11] when $T_c(x)$ is less than the separation between singlet states the nonmagnetic character of the latter makes pair breaking noneffective, causing long tails in the $T_c(x)$ curve while the experiments show an abrupt decrease in the $T_c(x)$ curve. We suggest here that the AF interaction between the Pr ions, which causes the AF ordering at low temperatures, is also important for the explanation of the pair-breaking $T_c(x)$ curve for $x > 0.4$.

We use the two energy separations between the lowest singlets as parameters that, together with the exchange interaction, give a fair fit to the temperature dependence of the AF ordered magnetic moment and to the high-temperature magnetic moment. The same parameters are used for calculations, in accordance with experiment, of $T_c(x)$. The magnetic susceptibility in the whole range of temperatures is also calculated. The last cannot be fitted to experimental data for $T < T_N$.[1,3,4] The reason for this discrepancy is not understood yet but it is plausible that this part of the susceptibility is caused by some kind of impurities that dominate over the AF susceptibility of the Pr^{3+} ions.

II. ANTIFERROMAGNETISM

In order to treat the antiferromagnetic order at low temperatures we consider the following Hamiltonian for the subsystem of the Pr ions:

$$\mathcal{H}_{\mathrm{Pr}} = \mathcal{H}_0 - \frac{1}{2}\sum_{i,j} \mathcal{J}_{ij}\mathbf{S}_i \cdot \mathbf{S}_j, \tag{1}$$

where $\mathcal{H}_0$ is the single-ion Hamiltonian that contains also the crystal electric field and the spin-orbit interaction, $\mathbf{S}_i$ is the total spin of the Pr ion at site i, and $\mathcal{J}_{ij}$ is an effective exchange interaction between the Pr ions.

In the random phase approximation (RPA) we assume an effective field $\mathbf{H}_{\mathrm{eff}}(\mathbf{q}) = \mathcal{J}(\mathbf{q})\langle \mathbf{S}(\mathbf{q})\rangle/\mu_B$ acting on ev-

ery Fourier component of the spin. $\mathcal{J}(\mathbf{q})$ and $\mathbf{S}(\mathbf{q})$ are the Fourier transforms of the interaction and the spin, respectively.

The level structure of $\mathcal{H}_0$ consists of three lowest levels separated from the rest of the levels by a gap of about 500 K. At low temperatures, $T < T_N$, we neglect all but the three lowest levels. As proposed by Goodman *et al.*,[9] the states are $|\Gamma_1\rangle$, $|\Gamma_4\rangle$, and $|\Gamma_2\rangle$ with energies 0, 1.4, and 3.8 meV, respectively. The nonvanishing matrix elements of $\mathbf{S}$, as calculated with the CEF parameters of Ref. 9 using the 33 lowest levels of Pr^{3+} ($L = 5, S = 1$) are $M_x \equiv \langle\Gamma_1|S_x|\Gamma_2\rangle = -0.51$, $M_y \equiv \langle\Gamma_1|S_y|\Gamma_4\rangle = -0.49i$, and $M_z \equiv \langle\Gamma_2|S_z|\Gamma_4\rangle = 0.50$. As may be shown these are not sensitive to small changes of the CEF parameters that preserve the gap of about 500 K. The measurements indicate that the c axis (z direction) is the AF staggered field direction.[3] With $|\Gamma_1\rangle$ as the ground state of the Pr^{3+} it is impossible to fit the experimental values both of T_N and the staggered ordered moment $\langle\mu\rangle_{T=0}$. To meet the experimental demands the ground state must be connected by the matrix element M_z to one of the two upper singlets, so the ground state may be $|\Gamma_2\rangle$ or $|\Gamma_4\rangle$. Only in the case when the staggered field is in the a-b plane the assignment of $|\Gamma_1\rangle$ to the ground state would be in agreement with the AF data.[3] We define the energy splitting $\Delta E_\parallel = E(\Gamma_2) - E(\Gamma_4)$ and $\Delta E_\perp = E(\Gamma_1) - [E(\Gamma_2) + E(\Gamma_4)]/2$. Using the RPA in the usual way and taking into account the disorder in the simplest possible way we obtain for $x \leq 1$ the equation for the susceptibility $\chi_{\mu\mu}$ ($\mu = x, y, z$) for $T > T_N$:

$$\chi_{\mu\mu}(\mathbf{q}, T) = \frac{\chi^0_{\mu\mu}(T)}{1 - x\mathcal{J}(\mathbf{q})\chi^0_{\mu\mu}(T)/G^2_\mu\mu^2_B}, \tag{2}$$

where $\chi^0_{\mu\mu}(T)$ is the single-ion susceptibility and G_μ is the proportionality coefficient between the nonvanishing matrix elements of $L_\mu + 2S_\mu$ and S_μ for the three lowest $|\Gamma_n\rangle$ states ($n = 1, 2, 4$). In the present case $G_\mu \approx g_J/|g_J - 1| = 4$ is almost μ independent, g_J is the Landé factor for the $J = 4$ multiplet of Pr^{3+}. $\chi^0_{zz}(T)$ is given by

$$\chi^0_{zz}(T) = \frac{2\sinh\beta\Delta E_\parallel/2}{e^{-\beta\Delta E_\perp} + 2\cosh\beta\Delta E_\parallel/2} \frac{G^2_z\mu^2_B|M_z|^2}{\Delta E_\parallel/2} \tag{3}$$

with $\beta = 1/T$.

To obtain the equation for T_N we require that the denominator in Eq. (2) will vanish for $\mathbf{q} = \mathbf{Q} = (\pi/a, \pi/b, \pi/c)$ which is the wave vector corresponding to an antiferromagnetic order:[3]

$$x\mathcal{J}(\mathbf{Q})\chi^0_{zz}(T_N)/G^2_z\mu^2_B = 1. \tag{4}$$

$\langle\mu_z\rangle_T$ is equal to $\langle S_z(\mathbf{Q})\rangle_T$ multiplied by the factor G_z. For $T < T_N$ it is determined self-consistently by solving Eq. (4), replacing T_N by T and $\Delta E_\parallel$ in Eq. (3) by $\Delta E'_\parallel = \sqrt{\Delta E^2_\parallel + [2\mathcal{J}(\mathbf{Q})|M_z|\langle S_z(\mathbf{Q})\rangle_T]^2}$. At $x = 1$ and $T = 0$ we obtain an equation for $\langle S_z(\mathbf{Q})\rangle_T$:

$$\Delta E_\parallel/2\mathcal{J}(\mathbf{Q}) = |M_z|^2\sqrt{1 - \langle S_z\rangle^2_{T=0}/|M_z|^2}. \tag{5}$$

Figure 1 shows $\langle\mu_z\rangle^2_T$ versus T at $x = 1$ for $\Delta E_\parallel = 5.5$ meV, $\Delta E_\perp = 3$ meV and $\mathcal{J}(\mathbf{Q})$=10.7 meV. These

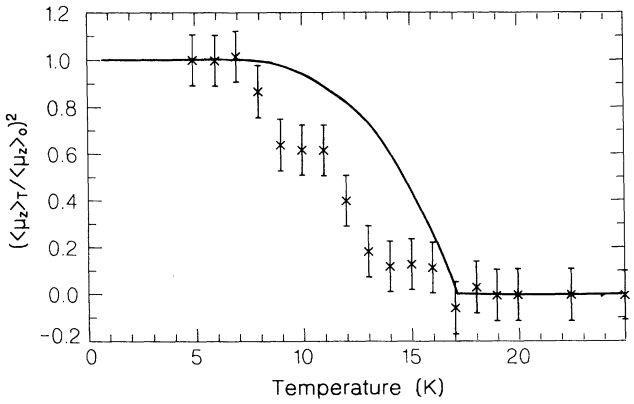

FIG. 1. The square of the staggered moment of a Pr^{3+} ion as a function of temperature at $x = 1$. The solid line is the RPA calculation as explained in the text and the points are from Ref. 3.

parameters were chosen so that by preserving the gap of about 500 K the experimental values of $\langle\mu_z\rangle_{T=0} = 0.74\mu_B$ and T_N=17 K are obtained with Eqs. (4) and (5). The calculated $\langle\mu_z\rangle_T$ curve in Fig. 1 resembles a typical mean-field (MF) behavior. Of course fluctuations have to be taken into account in a more accurate calculation.

From the phase diagram,[1,2] the Pr-ion Néel temperature falls down to zero at $x \approx 0.5$. To explain $T_N(x)$ we solve Eq. (4) for $x < 1$. We observe that to obtain in our model $T_N(x = 0.5) = 0$ the interaction $\mathcal{J}(\mathbf{Q})$ for $x = 0.5$ has to be twice larger than for $x = 1$. This may be explained by the increase of $\mathcal{J}$ because of the appearance of additional Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction at $x \leq 0.5$.

The high-temperature susceptibility for the powder at $\Delta E_\parallel \ll T < \Delta_{\text{gap}}$ may be written as

$$\chi(T) \approx \chi_0 + \frac{\mu^2_{\text{eff}}/3}{T + \theta}, \tag{6}$$

where χ_0 is a van-Vleck susceptibility due to the high-energy levels (we calculated $\chi_0 \approx 10^{-3}$ emu/mol). The Curie-Weiss part in Eq. (6) comes from the lower three levels. $\mu_{\text{eff}} = 2.93\mu_B$ is in accordance with experiment,[9] and θ is a sum of the contributions of the CEF splitting and the AF interaction through $\mathcal{J}(\mathbf{q} = \mathbf{0})$: $\theta = \theta_{\text{CEF}} + \theta_{\text{AF}}$, where $\theta_{\text{CEF}} \approx 10$ K and $\theta_{\text{AF}} \approx 2|\mathcal{J}(\mathbf{q} = \mathbf{0})|\sum|M_i|^4/3\sum|M_i|^2$ for $i = x, y, z$. To obtain the experimental value of the susceptibility θ_{AF} must be of the order of θ_{CEF}.

Measurements of the susceptibility at $T < T_N$ show a Curie-Weiss-type increase.[1,3,4] This is contradictory to the calculated AF directionally averaged susceptibility, which shows a small cusp and a saturation toward zero temperature. We believe that the low-temperature increase of the susceptibility comes from uncontrolled impurities beyond the Pr^{3+} system.

III. PAIR BREAKING

To explain the suppression of superconductivity we use the Abrikosov-Gor'kov mechanism of pair breaking[10] as

formulated by Keller and Fulde[11] to include the CEF. The Hamiltonian is now written

$$\mathcal{H} = \mathcal{H}_{Pr} + \mathcal{H}_e - \frac{1}{2}\sum_i \sum_{\mathbf{k},\mathbf{k}'} J\, e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}_i} \mathbf{S}_i \cdot \boldsymbol{\sigma}_{\alpha\beta} c^\dagger_{\mathbf{k},\alpha} c_{\mathbf{k}',\beta}, \tag{7}$$

where $\mathcal{H}_{Pr}$ and $\mathcal{H}_e$ are the Hamiltonians of the subsystems of the Pr^{3+} ions and the conduction electrons, respectively. $\mathcal{H}_e$ contains also an effective attractive pairing interaction between the electrons. The last term contains an exchange interaction J between a Pr^{3+} spin $\mathbf{S}_i$ at site i and the spin of the conduction electrons. $c^\dagger_{\mathbf{k},\sigma}$ is a conduction-electron creation operator and $\boldsymbol{\sigma}_{\alpha\beta}$ are the Pauli matrices.

Following Ref. 11, but using a wave-vector-dependent Stoner-type susceptibility [Eq. (3)] we arrive at the following equations for $T \approx T_c(x)$:

$$\bar{\omega}_n = \omega_n + \frac{T}{\tau} \sum_{\mu=x,y,z} \frac{1}{G^2_\mu} \sum_m \bar{\chi}_{\mu\mu}(\omega_n - \omega_m, x)\,\mathrm{sgn}(\bar{\omega}_m), \tag{8}$$

$$\bar{\Delta}_n = \Delta - \frac{T}{\tau} \sum_{\mu=x,y,z} \frac{1}{G^2_\mu} \sum_m \bar{\chi}_{\mu\mu}(\omega_n - \omega_m, x) \frac{\bar{\Delta}_m}{|\bar{\omega}_m|}. \tag{9}$$

Here the gap Δ is determined self-consistently as in Ref. 11 and $\tau^{-1} = \pi N(\varepsilon_F) x\, J^2/2\mu_B^2$, where x is the concentration of the Pr ions and $N(\varepsilon_F)$ is the density of states at the Fermi surface. In Eqs. (8) and (9) $\omega_n = 2\pi T(n+\frac{1}{2})$ are Matsubara frequencies. $\bar{\omega}_n$ and $\bar{\Delta}_n$ are averages of $\bar{\omega}_n(\mathbf{p})$ and $\bar{\Delta}_n(\mathbf{p})$ over $\mathbf{p}$. The last are related to the Nambu representation of the Green's function $g(\mathbf{p},\omega_n)$:

$$g^{-1}(\mathbf{p},\omega_n) = \begin{pmatrix} i\bar{\omega}_n(\mathbf{p}) - \bar{\varepsilon}_\mathbf{p} & \bar{\Delta}_n(\mathbf{p}) \\ \bar{\Delta}_n(\mathbf{p}) & i\bar{\omega}_n(\mathbf{p}) + \bar{\varepsilon}_\mathbf{p} \end{pmatrix}. \tag{10}$$

$\bar{\chi}_{\mu\mu}(\omega, x)$ in Eqs. (8) and (9) is an average of $\chi_{\mu\mu}(\mathbf{p}-\mathbf{q},\omega)$ over the Fermi surface:

$$\bar{\chi}_{\mu\mu}(\omega, x) = \frac{1}{N(\varepsilon_F)^2} \frac{1}{(2\pi)^6} \times \int_{S(\varepsilon_F)} \frac{d^2p}{|\mathbf{v}_\mathbf{p}|} \int_{S(\varepsilon_F)} \frac{d^2q}{|\mathbf{v}_\mathbf{q}|} \chi_{\mu\mu}(\mathbf{p}-\mathbf{q},\omega,x). \tag{11}$$

$\chi_{\mu\mu}(\mathbf{q},\omega)$ is the dynamic susceptibility which is obtained from the static one [Eq. (3)] by the RPA-MF as in Ref. 11.

Equations (8) and (9) are similar to the equations obtained by Ref. 11, except that the equations of Ref. 11 contain a paramagnetic susceptibility which is independent of x, while in our case there is an additional x dependence through $\bar{\chi}(\omega, x)$ which is caused by the antiferromagnetic interactions between the Pr ions. The susceptibility $\chi(\mathbf{q}, \omega = 0)$ diverges with the onset of antiferromagnetic order at $x \approx 0.5$. The integral in Eq. (11) will be enhanced appreciably for $x > 0.4$ if there is a nesting of the vector $\mathbf{Q} = (\pi/a, \pi/b, \pi/c)$ on the Fermi surface. When the subsystem of Pr ions tends toward antiferromagnetic order its susceptibility for wave vectors close to $\mathbf{Q}$ increases significantly and electrons will be magnetically scattered from one part of the Fermi surface to the other.

We have done the numerical calculations of the function $\bar{\chi}(\omega, x)$ using Eq. (11) and setting $\mathcal{J}(\mathbf{Q}, x < 0.5)$=20 meV, in accordance with Sec. II. The significant contribution to $\bar{\chi}$ comes from $\mathcal{J}(\mathbf{q} = \mathbf{Q})$, and we have used $\mathcal{J}(\mathbf{q}) = 2\mathcal{J}_{\mathrm{NN}}[\cos(q_x a) + \cos(q_y b)]$ where $\mathcal{J}_{\mathrm{NN}}$ stands for the nearest neighbor interaction; $-4\mathcal{J}_{\mathrm{NN}} = \mathcal{J}(\mathbf{Q})$ and the much smaller interaction in the z direction was neglected here. The $T_c(x)$ curve is then calculated with the help of Eqs. (8) and (9) in the same way as in Ref. 11. For the calculation of $\bar{\chi}(\omega, x)$ we have used the Fermi surface calculated by Pickett[12] for the $YBa_2Cu_3O_7$ compound. Only the parts of the Fermi surface arising primarily from the relevant CuO_2 layers were taken into account and we used $N(\varepsilon_F)$=2.5 states/eV cell calculated for the layers' density of states.[8] Note that there is no appreciable difference in the Fermi surface of the Y and Pr compounds.[8]

Figure 2 shows the results of the numerical calculations of $T_c(x)$ for the Abrikosov-Gor'kov model of paramagnetic impurities with a degenerate ground level $J = 4$, for the model of Keller and Fulde for paramagnetic impurities with CEF split levels as proposed in Sec. II, and for our model with the same parameters. A quite good fit to the experimental curve of $T_c(x)$ is obtained in our model for $J = 80$ meV. Note that $N(\varepsilon_F)J^2 = 16$ meV is of the order of RKKY contribution to $\mathcal{J}(\mathbf{Q})$ for x<0.5. As can be seen in Fig. 2, the Abrikosov-Gor'kov curve can also explain the behavior of $T_c(x)$ but the degenerate ground state is not consistent both with the explanation of the magnetic properties and the neutron-scattering experiments.

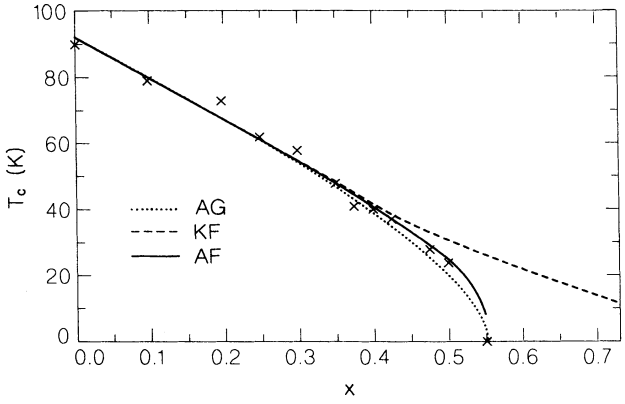

FIG. 2. $T_c(x)$ from experiment (Ref. 2) and from calculation for three models: Abrikosov-Gor'kov (AG) — without CEF splitting, for the multiplet $J = 4$, using $N(\varepsilon_F)J^2$=10 meV ($J = 63$ meV), Keller and Fulde (KF) and our antiferromagnetic model (AF). The two latter were calculated for the CEF levels given in Sec. II, and the exchange parameters $J = 80$ meV and $\mathcal{J}$=20 meV.

IV. CONCLUSIONS

The behavior of Pr^{3+} in $Pr_xY_{1-x}Ba_2Cu_3O_7$ is characterized by two main features: a small magnetic moment of the single ion due to a CEF and relatively strong exchange interactions which lead to antiferromagnetism and pair breaking. The CEF effects are common for many $RBa_2Cu_3O_7$ compounds (R=Ho,Er,Dy,Nd),[9,13] and they are well established by inelastic neutron scattering. In this sense Pr^{3+} is not an exception (apart from its weak neutron-scattering lines). The key question in the $Pr_xY_{1-x}Ba_2Cu_3O_7$ problem is about the reasons for the large exchange interaction compared to other rare-earth ions and for the metal-semiconductor transition at $x = 0.5$. The present band[8] and semiempirical-molecular-orbital calculations[14] show that the Pr^{3+} f electrons are highly hybridized with the oxygen electrons in the CuO_2 planes. But what the mechanism is for the exchange interaction and its change with x when $x > 0.5$ is not clear yet. The problem of the large exchange AF interaction cannot be avoided if one uses a model of Pr^{4+}. Furthermore, we examined a Pr^{4+} model and found that because of the Kramers character of Pr^{4+} its zero-temperature staggered magnetic moment (for T_N=17 K and the appropriate scaled CEF) will be twice larger than the experimental value. We have attributed the suppression of superconductivity to a magnetic pair-breaking mechanism. It is possible that change in Pr-ion concentration causes relocation of the holes in the 1:2:3 compound between the planes and the chains[15] which will also lead to the suppression of superconductivity. What we have shown here is that due to the antiferromagnetic exchange interaction between Pr^{3+} ions the singlet level system is effective in the suppression of superconductivity in the whole range of $T_c(x) \geq 0$.

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