Nature of the low temperature ordering of Pr in PrBa$_2$Cu$_3$O$_{6+x}$

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Theoretical model is presented to describe the anomalous ordered phase of Pr ions in PrBa$_2$Cu$_3$O$_{6+x}$ below $T_{Pr} \approx 12 - 17$ K. The model considers the Pr multipole degrees of freedom and coupling between the Cu and Pr subsystems. We identify the symmetry allowed coupling of Cu and Pr ions and conclude that only an ab-plane Pr dipole ordering can explain the Cu spin rotation observed at $T_{Pr}$ by neutron diffraction by Boothroyd et al. [A. T. Boothroyd et al., Phys. Rev. Lett. 78, 130 (1997)]. A substantial enhancement of the Pr ordering temperature is shown to arise from the Cu-Pr coupling which is the key for the anomalous magnetic behavior in PrBa$_2$Cu$_3$O$_{6+x}$.

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I. INTRODUCTION

Non-superconducting PrBa$_2$Cu$_3$O$_{6+x}$ compound attracted considerable attention in the last two decades because of its intriguing magnetic and electronic properties within the ReBa$_2$Cu$_3$O$_{6+x}$ family of compounds (Re=rare earth atom). In spite of the great efforts from both experimental and theoretical sides, numerous problems remain unresolved such as the suppression of superconductivity and the nature of the long-range ordered state of Pr sublattice with a unique, about an order of magnitude larger than for other rare earth atoms, ordering temperature. Although, there exists a discussion whether the ground state of PrBa$_2$Cu$_3$O$_{6+x}$ is really a non-superconducting, insulating material with a magnetically ordered Cu and Pr sublattices, herein we consider this modification of PrBa$_2$Cu$_3$O$_{6+x}$ as "canonical" and treat it solely herein. We note that superconductivity in PrBa$_2$Cu$_3$O$_{6+x}$ was reported and a non-magnetic Pr ground state was found by 141Pr NMR. It was suggested that these observations may result from a non-stoichiometric compound, i.e. when Pr occupies only about a half of the rare-earth sites, and the other half is occupied by the non-magnetic Ba.

The theoretical model of Fehrenbacher and Rice, i.e. the hybridization of Pr 4f and the nearest neighbor O 2p orbitals is the most accepted model to account for the absence of superconductivity in PrBa$_2$Cu$_3$O$_{6+x}$. It is suggested that the localization of holes in the hybridized 4f-2p orbitals renders the material non-superconducting. Although the non-superconducting nature of PrBa$_2$Cu$_3$O$_7$ compound is a challenging and interesting problem, we concentrate herein exclusively on the low-temperature Pr ordering.

PrBa$_2$Cu$_3$O$_{6+x}$ is an insulator for every $x$ and the Cu spins in the CuO$_2$ planes order antiferromagnetically at temperatures of $T_N \approx 350$ K and 250 K for $x = 0$ and $x = 1$, respectively. As the temperature is further decreased, the Pr sublattice undergoes a phase transition in the temperature range $T_{Pr} \approx 12 - 17$ K depending on $x$, which appears as an anomaly in the temperature dependence of thermodynamic quantities, Mössbauer spectroscopy, neutron diffraction, and NMR. It showed that the Pr magnetic moments order antiferromagnetically below $T_{Pr}$. However, there is no consensus among these experiments with respect to the magnitude of the ordered Pr moment, its direction, and the nature of this transition.

Neutron diffraction studies found $0.56 \mu_B$ ($x = 0.92$) and $1.15 \mu_B$ ($x = 0.35$) ordered moment with direction tilted out from the $ab$-plane. The rotation of Cu moments within the CuO$_2$ plane was found to accompany the Pr ordering which suggested that the Pr spin structure is non-collinear along the $a$-axis. As the temperature is further decreased, the Pr sublattice also undergoes an ordering at temperature $T_{Pr} = 12$ K. This ordering is accompanied by the rotation of Cu spins within the CuO$_2$ planes in such a way that the new spin structure is non-collinear along the $c$-direction (phase AFI).

II. GENERAL FORM OF THE CU-PR INTERACTION

In PrBa$_2$Cu$_3$O$_6$, the Cu spins are antiferromagnetically ordered along the [100] direction in the CuO$_2$ planes with the ordering vector $Q = [\pi/a, \pi/a, \pi/c]$ below the temperature $T_{Cu} \approx 350$ K (phase AFI). As the temperature is further decreased, the Pr sublattice also undergoes an ordering at temperature $T_{Pr} = 12$ K. This ordering is accompanied by the rotation of Cu spins within the CuO$_2$ planes in such a way that the new spin structure is non-collinear along the $c$-direction (phase AFI) and the Cu spin structure is characterized by the ordering vector $Q = [\pi/a, \pi/a, 0]$. The Cu spin structure
in the CuO$_2$ planes is shown in Fig. 1 in the temperature range $T \leq T_{Pr}$.

First, we construct the invariant form of the interaction between the multipole moments of a Pr ion at position $r_0 = (0, 0, 0)$ and the surrounding eight Cu spins, Cu[1-8], shown in Fig. 1. Although the local symmetry at Pr site is tetragonal, a Cu-Pr pair has lower symmetry. For example, the Cu[1]-Pr pair has reflection symmetry with respect to the $[1,1,0]$ mirror plane. The transformation of total angular momentum $J$ of Pr ion under this operation is: $J_x \rightarrow -J_y$, $J_y \rightarrow J_x$, and $J_z \rightarrow -J_z$. The same transformation is applied for the Cu spin $S$. Thus, spin operators $J_x - J_y$ ($S_x - S_y$) and $J_z$ ($S_z$) are odd, while $J_x + J_y$ ($S_x + S_y$) is even under this reflection. Not only the bilinear products of the Pr dipole moments and Cu spins appear in the interaction but also the bilinear products of rank-3 octupole and rank-5 triaxial dipole operators of Pr ion and Cu spins since they are allowed by the time reversal symmetry. The octupole operators $T_x^3$ and $T_y^3 - T_y^3$ are even, while $T_{xyz}$ and $T_y^3 + T_y^3$ are odd operators under the present transformation. In the case of tetragonal symmetry, there is one more independent magnetic operator, namely the rank-5 triaxial dipole operator $V_{1u} = J_x J_y J_z (J_z^2 - J_y^2)$ which is even under the present reflection. Thus, the invariant form of the interaction for the Cu[1]-Pr pair is constructed as

$$H_{Cu-Pr}^{[1]} = \{J_x + S_y\} [c_{11} (J_x + J_y) + c_{12} T_y^3 + c_{13} A_{1u}]$$

$$+ (S_x - S_y) [c_{21} (J_x - J_y) + c_{22} J_z + c_{23} T_{xyz}] + S_z [c_{31} (J_x - J_y) + c_{32} J_z + c_{33} T_{xyz}],$$

(1)

where $c_{ij}$ are constants which are not determined by the symmetry itself. The pair Cu[2]-Pr is connected to the pair Cu[1]-Pr by a $\pi/2$ rotation around the c-axis. Under this operation the transformation of the angular momentum components is given by $J_x \rightarrow -J_y$, $J_y \rightarrow J_x$, and $J_z \rightarrow J_z$. The same transformation holds also for the Cu spin components. Thus, the form of the interaction for the Cu[2]-Pr pair can be obtained from Eq. (1) by applying the $\pi/2$ rotation. Repeating the appropriate rotations and reflections, the interaction can be obtained for all the eight Cu-Pr pairs. The Cu spin at position $r$ is expressed by Fourier transformation as $S(r) = \sum_q S_q e^{-iqr}$. Using this expression, we obtain the total interaction as

$$H_{Cu-Pr} = \sum_{k=1}^8 H_{Cu-Pr}^{[k]} = 8 \sum_q \{(c_{11} + c_{21})(S_x^q J_x + S_y^q J_y)c_y c_z + (c_{11} - c_{21})(S_x^q J_y + S_y^q J_x)s_z s_y c_z$$

$$+ c_{22} J_z \{S_x^q s_x c_y s_z - S_y^q c_x s_y s_z\} + c_{31} [J_x^q s_x s_y c_z - J_y^q s_x s_y s_z] + c_{32} J_z^q c_x c_y c_z$$

$$+ c_{12} T_y^3 \{S_x^q s_x c_y s_z + S_y^q c_x s_y s_z\} + c_{31} A_{1u} [S_x^q c_x s_y s_z + S_y^q s_x c_y s_z] + c_{33} T_{xyz} \{S_x^q c_x s_y s_z - S_y^q s_x c_y s_z\},$$

(2)

where $c_k(s_k)$ denotes $\cos(q_k a/2) \sin(q_k c/2)$ for $k=x, y,$ and $\cos(q_k c/2) \sin(q_k c/2)$ for $k=z$.

Now we discuss the multipole order of Pr sublattice based on expression (2). In phase AFI, the Cu spin component $S_x^q$ with $q = Q$ is non-zero. In this phase there is no coupling between the Cu spins and Pr magnetic moments because the wave vector $Q$ leads to $c_x = c_y = c_z = 0$ and $s_x = s_y = s_z = 1$, which gives that none of the terms survives in expression (2). On the other hand, a coupling of Cu spins and Pr magnetic moments is present in phase AFIII. The non-collinear Cu spin structure in phase AFIII can be described by the appearance of the extra spin component $S_y^q$ in addition to the component $S_x^q$. The wave vector $Q$ gives $c_z = s_x = 0$, and $s_x = c_y = c_z = 1$. Together with the condition $S_y^q \neq 0$, we find that the only surviving term in the form of Cu-Pr interaction is the coupling $S_y^q J_y$. We note that the coupling $S_y^q J_y$ is also allowed by symmetry since the [100] and [010] domains in phase AFI are equivalent, which gives that the domains $S_y^q \neq 0$, $S_x^q = 0$ and $S_y^q \neq 0$, $S_x^q = 0$ are also equivalent.

Thus, we conclude that the neutron diffraction result for the Cu spin rotation below $T_{Pr}$ is compatible with Pr dipole
moments lying along the crystalline [100] or [010] direction, and no deviation from this direction is allowed by symmetry in contrast to the structure proposed in Ref. [8]. Since the $S^Q_y, S^Q_z$ Cu spin components alternate along the $a$ and $b$ directions, the Pr dipole moments also form an AFM structure in the $ab$-planes with the ordering vector $Q$. The magnetic structure for the Cu spins and Pr dipole moments below $T_{Pr}$ has the structure shown in Fig. [1].

III. CEF MODEL FOR THE COUPLED CU-PR SYSTEM

We now turn to model a CEF model of the Pr ion 4$f$ electrons to describe the Pr ordering in PrBa$_2$Cu$_3$O$_6$. The model consists of the coupled Pr and Cu subsystems and allows to calculate experimentally relevant quantities. The Pr ion has $4f^2$ electronic configuration in PrBa$_2$Cu$_3$O$_6$, which gives $J = 4$ as the total angular momentum based on the Hund’s rule for the ground state. In the Pr subsystem, we consider AFM interaction between the in-plane $J_x$ and $J_y$ dipole moments described by the Hamiltonian

$$H_{Pr} = \frac{1}{2z} \sum_{(i,j)} \Lambda [J_{x,i}J_{x,j} + J_{y,i}J_{y,j}],$$

where $z = 4$ is the number of nearest-neighbor Pr ions and $\Lambda$ is the coupling constant. The Hamiltonian (3) is taken in a quasi-triplet subspace, where the ground state is the doublet

$$|d\pm\rangle = a|\pm 3\rangle + \sqrt{1-a^2}| \mp 1\rangle,$$

with a near singlet excited state

$$|s\rangle = \frac{1}{\sqrt{2}}(|2\rangle + | -2\rangle),$$

where the states are expressed by the eigenstates of $J$. The doublet and singlet states are separated by the gap $\Delta_{11,17}$. The other CEF levels lie at much higher energy therefore these can be neglected when describing the low temperature behavior.

We treat the Cu subsystem phenomenologically and consider the free energy expansion:

$$F_{Cu} = -\frac{1}{\beta} \ln \text{Tr}_S \left( e^{-\beta H_{Cu}} \right)$$

where $F_{Cu}$ contains the part corresponding to the Cu spin components, $S^Q_x$ and $S^Q_y$, which are not relevant for our discussion. We assume that $\langle S^Q_y, S^Q_z \rangle$ is not critical around $T = T_{Pr}$, thus, $a_s > 0$. Parameters $b_s$ and $c_s$ determine the anisotropy as we discuss below.

The local coupling between the Cu and Pr subsystems is assumed as

$$H_c = z' \sum_i \lambda \left[ J_{x,i} \langle S^Q_y \rangle + J_{y,i} \langle S^Q_z \rangle \right],$$

which corresponds to the only non-vanishing term in expression (2) for $Q = [\pi/a, \pi/a, 0]$. Here, $z' = 8$ is the number of Pr nearest-neighbor Cu ions and $\lambda$ is the Cu-Pr coupling constant. The series expansion of the total free energy has the form

$$F = -\frac{1}{\beta} \ln \text{Tr}_S \left( e^{-\beta H_{Cu}} \right) - \frac{1}{\beta} \ln \text{Tr}_J \left( e^{-\beta(\delta H_{Pr} + H_c)} \right)$$

$$\equiv F_0 + F_{Cu},$$

where

$$F_0 = a_J (\langle J^1_{Pr} \rangle^2 + \langle J^2_{Pr} \rangle^2) + b_J (\langle J^3_{Pr} \rangle^2$$

$$+ \langle J^4_{Pr} \rangle^2 + \lambda' (\langle J^1_{Pr} \rangle \langle S^Q_y \rangle + \langle J^3_{Pr} \rangle \langle S^Q_z \rangle))$$

$$+ a_s (\langle S^Q_x \rangle^2 + \langle S^Q_y \rangle^2) + b_s (\langle S^Q_x \rangle^2 + \langle S^Q_z \rangle^2)^2$$

$$+ 4c_s (\langle S^Q_x \rangle^2 \langle S^Q_y \rangle^2)^2,$$

where $a_J = \frac{1}{\Lambda^2} \left( \frac{1}{\Lambda} + \frac{2}{\lambda} (2\gamma^2 (e^{-\Delta/T} - 1) - (e^{-\Delta/T} + 2)) \right)$.

The coefficient $a_J$ is expressed in Eq. (2) as

$$a_J = \frac{1}{\Lambda^2} \left( \frac{1}{\Lambda} + \frac{2}{\lambda} (2\gamma^2 (e^{-\Delta/T} - 1) - (e^{-\Delta/T} + 2)) \right).$$

From the condition $\partial F / \partial \langle S^Q_y \rangle = 0$ we obtain

$$\langle S^Q_y \rangle = -\frac{\lambda' \gamma}{2a_s} \langle J^1_{Pr} \rangle.$$

Substituting this expression into the free energy expansion $F$, the second order coefficient of $\langle J^1_{Pr} \rangle$ is obtained as

$$\tilde{a}_J = a_J - \frac{(\lambda' \gamma)^2}{4a_s}.$$

We define the temperature $T_0$ which corresponds to the non-coupled system with $\lambda = 0$ and the temperature $T_{Pr}$ for
the coupled system \( \lambda \neq 0 \). \( T_0 \) is obtained from the condition \( a_f = 0 \), where the second-order coefficient has the form \( a_f = \alpha_f(T - T_0) \) in the vicinity of the phase transition. However, this transition temperature is modified due to the coupling to the Cu subsystem (\( \lambda \neq 0 \)). \( T_{Pr} \) is obtained from the condition \( \alpha_f = 0 \), which gives

\[
T_{Pr} = T_0 + \frac{(\lambda a_f)^2}{4a_f^2 a_s}.
\]

Thus the Pr transition temperature is enhanced due to the Cu-Pr coupling irrespective of the sign of the coupling parameter \( \lambda \).

The rotation of the Cu spins in the CuO\(_2\) plane is expressed as \( \phi = \tan^{-1}(\langle S_{Cu}^y \rangle / \langle S_{Cu}^x \rangle) \), where \( \phi = 0 \) corresponds to the \( x \)-direction. The Cu magnetic moment is expressed as \( \mu_{Cu} = \gamma_{Cu} \mu_B (\langle S_{Cu}^y \rangle^2 + \langle S_{Cu}^x \rangle^2)^{1/2} \), where \( \gamma_{Cu} \approx 2 \) is the Cu g-factor. An ordered magnetic moment of \( 0.066 \mu_B/\text{Cu}[2] \) is found also for the AFI phase due to quantum fluctuations. Our model includes this effect phenomenologically as this ordered magnetic moment is retained above \( T_{Pr} \) in the AFI phase.

Figure 2 shows the enhancement of the Pr transition temperature due to the Cu-Pr coupling and the corresponding Cu spin rotation angle as a function of coupling parameter \( \lambda \), calculated within the above model. We used \( \Delta = 4 \) K and \( a = 0.96 \) for the CEF parameters, where the latter value is obtained from the high-temperature susceptibility data in \( \text{PrBa}_2\text{Cu}_3\text{O}_6 \). Neutron scattering found the interaction parameters as \( \Delta = 1.2 \) K and \( \lambda = 1.74 \) K and an ordered Cu moment of \( \mu_{Cu} = 0.64 \mu_B \) for \( x = 0.35 \), which were used in the calculation of Fig. 2 as fixed parameters. Clearly, the model accurately reproduces the \( \phi = 20^\circ \) rotation of the Cu spins which was observed experimentally in Ref. 8. In addition, the model accounts for about 70% enhancement of the Pr ordering temperature with this parameter set.

The ground state Pr magnetic moment and the enhancement of \( T_{Pr} \) strongly depends on the value of the CEF energy gap, \( \Delta \). In Fig. 3 we show the calculated \( T_{Pr} \) as a function of \( \Delta \), where we fix the interaction and CEF parameters as \( \Delta = 1.2 \) K, \( \lambda = 1.74 \) K, \( a = 0.96 \). The value of the phenomenological parameter \( a_s \) is chosen for each \( \Delta \) value so that it reproduces the experimental values \( \phi = 20^\circ \) and \( \mu_{Cu} = 0.64 \mu_B \). For small values of \( \Delta \) the phase transition is second-order. The transition temperatures \( T_0 \) and \( T_{Pr} \) are suppressed with increasing energy gap \( \Delta \), and the phase transition changes to first-order above a critical value of \( \Delta \). Increasing further \( \Delta \), the phase transition disappears. These behaviors are due to the fact that the \( J_\epsilon \) dipole order is interaction-induced within the quasi-triplet subspace since the dipole operator \( J_\epsilon \) has non-vanishing matrix element only between the doublet and singlet states. Around \( \Delta \sim 10 \) K the ratio \( T_{Pr}/T_0 \) is considerably enhanced due to the Cu-Pr coupling.

IV. MEASUREMENT OF THE LOCAL FIELDS

Finally, we discuss the field-angle dependence of the local magnetic field acting on a Pr ion. This local field could be measured by a local magnetic probe such as e.g. \(^{89}\text{Y}\) using NMR or \( \text{Gd}^{3+} \) using ESR which can be substituted into the Pr sublattice in a low concentration.

The magnetic field \( H_{\text{probe}} \) acting on the local magnetic probe has five different sources: exchange and dipole fields from the surrounding Cu spins, exchange and dipole fields from the surrounding Pr dipole moments, and the external magnetic field. Contributions except the external magnetic field are directed along the \( x \)-direction, and we express their effect by the field \( h_x \). We keep the field \( h_y \) fixed as the direction of the external magnetic field \( H \) is changed which is the situation for small values of the magnetic field. For a gen-
er-al direction of the external magnetic field \( \mathbf{H} \), we write the Hamiltonian of the local probe as

\[
\mathcal{H}_{\text{probe}} = \hbar \gamma \mathbf{S} \cdot \mathbf{H}_{\text{probe}},
\]

where \( \gamma \) is the gyromagnetic ratio of the local probe, and \( \mathbf{S} \) is its spin (either electron or nuclear). Thus, we obtain the experimentally detected magnetic resonance shift, \( h_{\text{probe}} \) (in magnetic field units):

\[
h_{\text{probe}} = S(h_{\text{res}} - h_0),
\]

where \( h_{\text{res}} \) is the resonance field and \( h_0 \) is the resonance position for the AFI phase. The external magnetic field is expressed by field angles as \( \mathbf{H}/|\mathbf{H}| = [H_x, H_y, H_z] = [\cos \phi \sin \theta, \sin \phi \sin \theta, \cos \theta] \), and we assume that \([S_x, S_y, S_z] \sim [H_x, H_y, H_z]\). This gives

\[
h_{\text{probe}} = (h_x + H \cos \phi \sin \theta) \cos \phi \sin \theta + H(\sin \phi \sin \theta)^2 + H(\cos \theta)^2.
\]

Figure 4 shows the field angle dependence of \( h_{\text{probe}} \) by changing the direction of the magnetic field in the planes \([001]\) and \([110]\). We estimate the change of \( h_{\text{probe}} \) as

\[
\frac{h_{\text{probe}}[100] - h_{\text{probe}}[110]}{h_{\text{probe}}[110] - h_{\text{probe}}[001]} = \sqrt{2} - 1,
\]

which ratio can be checked in the NMR or ESR experiments.

V. DISCUSSION AND SUMMARY

We studied the nature of the Pr ordered phase in \( \text{PrBa}_2\text{Cu}_3\text{O}_6 \) in a quasi-triplet CEF model of \( 4f^2 \) electrons, where we also included the symmetry allowed coupling between the Pr and Cu ions. The reason to include Cu-Pr coupling is the Cu spin rotation observed at the Pr ordering temperature in neutron diffraction, which has not been observed for other rare earth \( \text{ReBa}_2\text{Cu}_3\text{O}_6 \) compounds. In the \( 4f \) rare-earth series, the localized electrons of Pr ion tend to hybridize rather strongly with conduction electrons as in e.g. \( \text{PrFe}_1\text{P}_{12} \). This can be the reason why the Cu-Re coupling is much stronger in \( \text{ReBa}_2\text{Cu}_3\text{O}_6 \) for Re=Pr compared to the other rare-earth ions, Re.

We showed by a general symmetry analysis that there is no coupling between the Cu and Pr subsystems in phase AFI, but a coupling emerges in phase AFIII. We prove that only the AFM ordering of \( J_{z} \) \((J_{y})\) dipole moments of Pr ions is consistent with the Cu spin rotation observed at \( T = T_{Pr} \) by neutron diffraction. The interpretation of the neutron diffraction data in Ref. 8 includes a tilting of the ordered Pr magnetic moments out of the \( ab \)-plane. However, the presence of non-zero \( J_{z} \) dipole component does not follow from the general symmetry analysis as it was shown in Section 4.

Upon identifying of the Pr order parameter, we studied the Pr ordering temperature in the Cu-Pr coupled quasi-triplet CEF model by changing the interaction and CEF parameters. We found that the Cu-Pr coupling enhances the Pr ordering temperature, \( T_{Pr} \), compared to the uncoupled ordering temperature, \( T_0 \).

By fixing the Pr-Pr and Pr-Cu interaction parameters to the values obtained by neutron diffraction and the CEF parameter to that obtained by high-temperature susceptibility measurements, we found that the Pr ordering temperature is considerably enhanced due to the Cu-Pr coupling in the parameter range \( \Delta \sim 8 - 10 \) K. Namely, the enhancement of the ordering temperature is as large as \( T_{Pr}/T_0 \sim 8 - 12 \) in this interval. This observation explains the uniquely large ordering temperature for Re=Pr in the series \( \text{ReBa}_2\text{Cu}_3\text{O}_{6+x} \).

In addition, we predicted the magnetic field angle dependence of the local magnetic field acting at a Pr site in the AFM ordered phase of the Pr ions. This can be directly compared to measured NMR or ESR spectra which detect the local magnetic field by a local magnetic probe.

Our model contains the minimal number of parameters which is required to account quantitatively for the experimental data. Given that the parameter values are assumed from independent measurements and no fit is performed, the agreement is reasonable. First, the realistic estimate \( \Delta \sim 8 - 10 \) K gives a \( T_{Pr} \) of \( 5 - 3.6 \) K which is to be compared to the experimental \( T_{Pr} \) of 12 K. We note that this estimated range gives
the Pr transition temperature $T_0$ for the uncoupled system as $0.7 - 0.3$ K, which is close to the typical values of the ordering temperatures for other rare-earth ions such as Yb (0.35 K), Nd (0.5 K) or Dy (1 K). This fact may also indicate that the uniquely large ordering temperature for Pr arises from the Cu-Pr coupling. Second, the interval $\Delta \sim 8 - 10$ K is not very far from the estimate $\Delta \approx 17$ K based on the analysis of the high-temperature susceptibility data. Furthermore, the estimated interval for $\Delta$ and also the splitting pattern of the quasi-triplet subspace is consistent with the results of neutron scattering.

In summary, the model we propose herein describes the main features of PrBa$_2$Cu$_3$O$_y$ system, and demonstrates the importance of the Cu-Pr coupling which may be the key point to understand the anomalous behavior of this compound such as the enhancement of the Pr ordering temperature. Further studies which include also the itinerant character of $4f^2$ electrons are necessary to give an extensive description of this rather complicated system.

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