

Answer to the response of Prof. D. Louca

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Data: 21 April 2006, 20:04:46 Temat:[Web]resub LQK1020Radwanski

RECVD: Fri Apr 21 15:04:45 2006

Resubmission to: Physical Review Letters

New files with this resubmission: RadwanskiComLoucaHS2.tex

Deleted files from previous submission: RadwanskiComLoucaHSx.tex

Details of changes:

Dear Editor,

As You have noticed, 4.04.2006, that our Comment is too long we removed 1 sentence in the last paragraph before conclusion. Thus, we resubmit our Comment.

Our answer to Prof. D. Louca, her the Response to the Comment LQK1020/Radwanski, is attached. We gave answer to all critics of Prof. D. Louca. So, we expect our Comment to be published.

Dear Editor, (Dr. Millev)

Dear Prof. D. Louca,

We appreciate that the Authors of the commented paper, namely Prof. D. Louca, maintain their claim that the excited state in LaCoO_3 is the IS state in contrary to our view of the HS state presented in Phys. Rev. B 67 (03) 172401. Such clear controversy can be solved in the future. Thus, we insist on publication of our Comment. Moreover, we do not have anything against the response to be published in a form as it is.

Coming to details, we would like to note that the recalled by Prof. Louca calculations with hybridization of the ligand oxygen by Potze *et al.* [Phys. Rev. B 51, 11501 (1995)] are, according to our judgment, very qualitative. The idea of the excited IS state was later developed by Korotin *et al.* [Phys. Rev. B 54, 5309 (1996)] from the same laboratory yielding 10 eV wide energy spectrum for d states (Fig. 2). We take this result to be contradicted by experiment of Louca *et al.*, who have found, after Noguchi *et al.* (2002) and Podlesnyak *et al.* (2005), a distinct low-energy excitation of 0.6 meV only. Thus, we would like to point out that this excitation has been predicted by our ionic QUASST picture, but NOT by the recalled Potze and Korotin calculations. We will not comment Prof. Louca remark about our QUASST approach. We note only, that usefulness or useless of any theoretical approach in Physics is easily verified. We have applied QUASST successfully to a great number of $3d$ oxides, let mention FeBr_2 [Phys. Rev. B 63 (01) 172404], NiO , CoO , YTiO_3 , $\text{Na}_2\text{V}_3\text{O}_7$. We repeat that a distinct low-energy excitation of 0.6 meV revealed in the commented paper has been predicted by our ionic QUASST approach pointing out the existence of atomic-like low-energy structure in $3d$ solids.

In the situation of the revealed so strong opposition to our interpretation of LaCoO_3 presented in Phys. Rev. B 67 (03) 172401, proving that the excited state is the HS state, authors of the commented paper are strongly encouraged to write a Comment to our paper in Phys. Rev. B 67 (2003) 172401. Already now, in advance, we declare that we will support their efforts to publish their Comment. They are strongly welcome to formulate their objections against our a simple crystal field calculation. Such a Comment seems to be needed, indeed, as they knew our paper but they simply ignored it in the commented paper.

An argument of Prof. Louca that our Comment should not be published as it does not present any new insight is unfair. In contrary to the Louca's view of the IS state we claim that the excited state (quasi-triplet) originate from the HS state. So, it is new.

We do not discuss experimental results about FM and AFM correlations but we question an inferred conclusion about the nature of the excited state to be the IS state. We are convinced that the results revealed by Prof. Louca can be equally well described by our HS state, but it goes beyond the present Comment. For a fruitful discussion the Authors, Prof. Louca, are encouraged to clarify their meaning of the IS state and to draw their low-energy electronic structure, even schematically, with relevant energies and degeneracies.

Finally, apart of the above-mentioned arguments, we have a scientific right to publish Comment in order to express our scientific disagreement with the published view with the relevant critics. It allows for formulation of the scientific controversy, in this case about the origin of the excited state, IS or HS state, that will be solved in the due time.

Thus, to the Editor of Phys. Rev. Lett., we insist on publication of our Comment. We do not have anything against the Louca's response to be published as it is.

Sincerely Yours, R. J. Radwanski and Z. Ropka, Krakow, 21-04-2006.

Remarks intended solely for the editor:

Dear Editor, (Dr. Millev)

We appreciate that the Authors of the commented paper, namely Prof. D. Louca, maintain their claim that the excited state in LaCoO_3 is the IS state in contrary to our view of the HS state presented in Phys. Rev. B 67 (03) 172401. Such clear controversy can be solved in the future. Thus, we insist on publication of our Comment. Moreover, we do not have anything against the Louca's response to be published in a form as it is.

Sincerely Yours, R. J. Radwanski and Z. Ropka

2nd Rejection with two referee reports

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From: Physical Review Letters <prl@ridge.aps.org>
To: zofiaropka@fizyk.instytut.serwery.pl
Data: 7 czerwca 2006, 03:48:54
Temat: Your manuscript LQK1020 Radwanski
Pliki: (none)
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Re: LQK1020
Comment on "Nanomagnetic droplets and implications to orbital ordering in $\text{La}_{1-x}\text{Sr}_x\text{CoO}_3$ ": the origin of the excited state in LaCoO_3 by R.J. Radwanski and Z. Ropka

Dr. Z Ropka
Center of Solid State Physics
Snt Filip 5 31-150 Krakow, POLAND

Dear Dr. Ropka,

The above manuscript has been reviewed by our referees.

The resulting reports include a critique which is sufficiently adverse that we cannot accept your paper on the basis of material now at hand. We enclose pertinent comments.

If you feel that you can overcome or refute the criticism, you may resubmit to Physical Review Letters. With any resubmittal, please include a summary of changes made and a brief response to all recommendations and criticisms.

Yours sincerely,
Yonko Millev
Assistant Editor Physical Review Letters
Attached: Referee reports A and B

Report of Referee A – LQK1020/Radwanski

This comment focuses on two related items. The first is a claim that the authors of the original paper, Phelan *et al.*, should have referenced an earlier paper by Ropka and Radwanski that calculates the first excited state in LaCoO_3 to be the high-spin state and accounts for some experimental data published by Noguchi *et al.*. The second issue is a claim that Phelan *et al.* misidentify the excited state as the intermediate spin state. I tend to agree with Radwanski and Ropka on the first point. Their previous paper was a significant piece of work on the same compound investigated by Phelan *et al.* and contained demonstrated agreement with experimental data. The result should have been mentioned, most probably during the section where the basic controversy over the nature of the first excited state is described. This mention would be proper even if only to point out why Phelan *et al.* believe it is wrong. I have less sympathy for the second argument. It is grossly overstated to claim that the nature of the first excited state has been proven to be the high spin state. It is entirely reasonable for Phelan *et al.* to argue otherwise and clearly many others do so as well.

I do have some concerns with the response of Dr. Louca. Firstly, it is true as stated by Dr. Louca that, "the comment does not invalidate any of our experimental observations". However, if correct it could invalidate, or at least complicate, conclusions drawn from those observations. Secondly, the response details what physics Dr. Louca believes is missing from the model used by Radwanski and Ropka. While this discussion may explain why the calculation by Radwanski and Ropka is wrong, it does not, by itself, establish that the calculation is wrong. Establishing the correctness of the theory can only be done by comparison to experimental data. Radwanski and Ropka have made such a comparison, at least to some extent. Phelan *et al.*'s own data is contrary evidence and should be discussed as such. Finally, I urge Dr. Louca to be careful that the tone of his response does not become too personal.

The appropriate action in the initial writing of the paper would have been to cite the work of Radwanski and Ropka. However, I do not believe that a missing reference is adequate reason to publish a separate comment. In their follow-up correspondence, Radwanski and Ropka state that, "We are convinced that the results revealed by Prof. Louca can be equally well described by our HS state, but it goes beyond the present Comment." Such an assessment is precisely what could make this comment interesting enough to be published. But since it is not present, I do not believe there is enough scientific content to merit the publication of this comment.

Report of Referee B – LQK1020/Radwanski

I have read the manuscript of the Comment and letters to the Editor from the authors of the Comment and the commented paper. The authors of the Comment concluded in a strong tone that the excited state of Co ion in LaCoO_3 should take a high spin ($S=2$) state, in sharp contrast to the conclusion of an intermediate spin ($S=1$) state in the commented paper.

It is my impression that the discussion in the Comment seems to be off the mark, and I cannot recommend the publication of the present manuscript in the Comment section of Physical Review Letters, even though I understand its theoretical viewpoint.

In the commented paper, it has been claimed that the tendency of the coexistence of ferromagnetic and anti-ferromagnetic correlations in LaCoO_3 revealed by neutron scattering experiment is consistent with the $S=1$ excited state with active e_g -orbital degree of freedom and dynamical orbital correlations. It should be noted that the experimental results first exist, and then, the results are interpreted by the spin and orbital degree of freedoms of the excited state. The authors of the commented paper have concluded that their results can be understood by the $S=1$ intermediate spin state. If the authors of the Comment insist on the high-spin $S=2$ state, it is necessary to show their interpretation of the experimental results on the basis of the $S=2$ high-spin state. If such an interpretation looks reasonable, then the authors of the commented paper should seriously reconsider their interpretation. In that situation, I am willing to recommend the publication of the manuscript as a Comment paper.

I think that the commented paper did not treat the theoretical aspects of the excited spin state. Here the point is how to interpret the experimental data. I understand that the authors of the Comment strongly believe the $S=2$ high-spin state as the excited state of LaCoO_3 and their belief is based on reliable calculations. Thus, I suggest the authors of the Comment to show their interpretation of the experimental results of the commented paper, on the basis of the $S=2$ high-spin state. Without that, it is difficult to recommend the publication of the manuscript as a Comment paper.

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Comments in: Physical Review Letters

Resubmission after two referees reports

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Data: 9 czerwca 2006, 14:31:12
Temat: [Web] resub LQK1020 Radwanski

Subject: LQK1020

Manuscript code: LQK1020

Comment on "Nanomagnetic droplets and implications to orbital ordering in $\text{La}_{1-x}\text{Sr}_x\text{CoO}_3$ ": the origin of the excited state in LaCoO_3 by R. J. Radwanski and Z. Ropka

RECV D: Fri Jun 9 09:31:12 2006

Resubmission to: Physical Review Letters

Resubmission type: resubmit

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Deleted files from previous submission:

RadwanskiComLoucaHS2.tex 06-09-2006

RadwanskiComLoucaHS2.pdf 06-09-2006

Details of changes:

Dear Editor,

In the answer of Your email of 7 June, 2006 with two referee reports we resubmit our Comment.

Thank You very much for long referee reports as well as for their quite positive reports. Both referee suggest us to explain within our approach the experimental results of Phelan/Louca.

We did it by putting two sentences to the last acapit before conclusions. Namely that the HS state in contrary to Phelan/Louca claim is Jahn-Teller active. As is clearly written on p. 4 (left, top) of the commented paper the rejection of the HS state and favouring the IS state was related to the fact that Phelan/Louca need the Jahn-Teller active state, which according to them is only IS state. It is not true.

We appreciate publishing of our Comment.

Sincerely Yours,

Ryszard Radwanski

Rejection with two 2nd referee reports

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From: Physical Review Letters <prl@ridge.aps.org>
To: zofiaropka@fizyk.instytut.serwery.pl
Data: 27 July 2006, 18:36:56
Temat: Your_manuscript LQK1020 Radwanski
=====

Re: LQK1020

Comment on "Nanomagnetic droplets and implications to orbital ordering in $\text{La}_{1-x}\text{Sr}_x\text{CoO}_3$ ": the origin of the excited state in LaCoO_3 by R. J. Radwanski and Z. Ropka

Dr. Z Ropka
Center of Solid State Physics
Snt Filip 5
31-150 Krakow, POLAND

Dear Dr. Ropka,

The above manuscript has been reviewed by our referees.

On the basis of the resulting reports, we conclude that the paper is unsuitable for publication in Physical Review Letters. We enclose comments from the criticism that led to this editorial decision.

In accordance with our standard practice (see enclosed memo), this concludes our review of your manuscript.

Yours sincerely,
Yonko Millev
Assistant Editor
Physical Review Letters

Second Report of Referee A – LQK1020/Radwanski

The key element of my first review of this comment was that the only relevant scientific point that would make this comment publishable would be a discussion of why the authors believe that the results by Phelan *et al.* could be equally well explained by having the high spin state as the first excited state rather than the intermediate spin state as argued by Phelan *et al.* The other referee had a similar comment. In their cover letter, Ropka and Radwanski note that "Both referee suggest us to explain within our approach the experimental results of

Phelan/Louca. We did it by putting two sentences to the last paragraph before conclusions.”

In my estimation these two sentences are not nearly enough to accomplish the task. The requirement was to explain how their result could explain Phelan’s data rather than merely including a statement asserting that it could. Assertion does not equal explanation. Along with a general explanation, the onus for the comment is to explain what is wrong with the arguments by both Phelan *et al.* and their reference 21 which argue that the high spin state is not consistent with the data. I understand that Ropka and Radwanski believe those are incorrect, but they spend no time explaining why.

As it stands, most of this comment consists of a restating of a previous paper and recounting some background information. Neither of those two subjects merit a publication as a comment in Physical Review Letters.

Second Report of Referee B – LQK1020/Radwanski

I read the revised manuscript and a reply. To respond to my points raised in the first report, the authors added a couple of sentences to mention that ”the HS state in contrary to Phelan/Louca claim is Jahn-Teller active”. However, the experimental results in the commented paper cannot be understood only from such a naive point whether the state is Jahn-Teller active or not. I feel some sympathy with the claim of the authors, but just a mention is not enough, if the authors want to publish the manuscript as an independent paper in the Comment section. Even if both cases (HS and IS states) are Jahn-Teller active, the type of the relevant orbital (e_g and/or t_{2g}) should be different, leading to the different spin and orbital correlations. In conclusion, I cannot recommend the publication of the present manuscript in the Comment section of Physical Review Letters.

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APPEAL to DAE

From: zofiaropka@fizyk.institut.serwery.pl
To: Physical Review Letters (prl@ridge.aps.org)
Data: 23 listopada 2006, 11:59:23
Temat: from-author LQK1020 Radwanski - APPEAL

From: R. J. Radwanski and Z. Ropka, 23 Nov 2006
To Editor of Phys. Rev. Lett.

LQK1020

Comment on "Nanomagnetic droplets and implications to orbital ordering in $\text{La}_{1-x}\text{Sr}_x\text{CoO}_3$ ": the origin of the excited state in LaCoO_3 by R. J. Radwanski and Z. Ropka

Dear Editor,

Despite of Your subsequent negative decision of 27 July 2006 we insist on publication of our Comment LQK1020, in which we have argued that a claim of Phelan *et al.* in PRL 97 (2006) 027201, that the first excited state is the IS state, is incorrect - we correct that the excited state is the HS state.

Recently, October 27, 2006 Phys. Rev. Lett. has been published in a paper of Haverkort, PRL 97 (2006) 176405, with **the same claim as ours**. They wrote: "To summarize LaCoO_3 can be described by a LS ground state and a triply degenerated HS excited state ...".

In these circumstances our answering to two referee reports seems not to be necessary any more. Conclusion is clear - our Comment with correction of the excited state is scientifically important and Comment should be published. Finally we can add a remark of the second report of A: "the onus of the Comment is to explain what is wrong with the arguments by both Phelan *et al.* and their reference 21", i. e. Potze PRB 51 (1995) 11501, - they did not take into account strong intra-atomic correlations, importance of which we argue by last 10 years, at least.

In conclusion, in such circumstances publication of our Comment is fully justified and an appeal to DAE is not necessary. If not, please treat this letter as our APPEAL TO DAE.

We hope for the scientific cooperation.

Sincerely Yours,
R. J. Radwanski and Z. Ropka