EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Letter of clarification to the ISOLDE and Neutron Time-of-Flight Committee

P-502-Local Probing Of Ferroic And Multiferroic Compounds

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Requested protons: protons on the ISOLDE target, split into 2 runs over 2 years (2017, 2018)
Experimental Area: [ISOLDE HALL – GLM SSP collection chamber & building 508 r-008]

First we would like to thank the committee for the proposal analysis and the given opportunity to explain ambiguous aspects of our proposal. We fully understand the committee's concerns and, with this letter, we hope to give to the INTC a straight to the point answer that definitively clarifies all the issues raised by the committee.

We appreciate that the committee recognizes that the topic we propose to investigate is of very high scientific and technological interest and as mentioned by the committee "PAC studies are ideally suited to investigate structural, charge, and orbital correlations and their impact on the multiferroic behaviour".

PAC is, in fact, a very valuable technique for studying multiferroic materials as it provides local selective electrical and magnetic information. This information is critical in many multiferroics once clear-cut evidences of the existence of ferroic orders and their intrinsic cross-coupling effects are typically revealed only through local information which is not accessible by a crystallographic/magnetic approach based on long-range average models. Particularly, the lack of evidence on certain ferroic claims can only be mitigated by local methods.

In the past, we have already proven that PAC can be a key technique to ascertain for the existence of multiferroism and followed the first-order polar phase transition (10.1103/PhysRevLett.100.155702) or determined which local distortions are responsible for the polar order (10.1103/PhysRevB.86.224418; 10.1103/PhysRevB.84.014434). Additionally, we have a set of complementary techniques at our disposal, at our home institutions, and the necessary research collaboration with high-motivated young PhD students and Post-docs ensuring a timely conclusive interpretation of the research done at ISOLDE. With such complementary means, including the singular Perturbed Angular Correlation (PAC) technique we have a real opportunity to decisively contribute to a comprehensive knowledge of multiferroic systems.

Having said that, we would like to state that, if the proposal is approved and beam-time is attributed to this proposal we will proceed, focusing on fewer compounds, as recommended by the committee. This means that we will focus only on the so called "priority cases" during the course of this proposal, namely in bismuth ferrite [$BiFeO_3$] (pg. 3 in the proposal); Multiferroic [Lu-(Fe/Mn)-O] (pg. 5 of in proposal) and [$Ca_3(Mn/Ti)_2O_7$] Ruddlesden-Popper compounds (pg. 6 in the proposal).

Notice that, we deliberately selected these three, different but complementary, systems as they represent the paradigm of the three possible types of ferroelectricity in multiferroics which are known as the i) proper ferroelectricity [$BiFeO_3$]; ii) improper ferroelectricity [Lu-(Mn/Fe)-O]; and iii) hybrid improper ferroelectricity [$Ca_3(Mn/Ti)_2O_7$]. With this 3-branch study we aim, on the one hand, to study the particularities of each system, its order parameter temperature evolution and the coupling between the relevant degrees of freedom (e.g. magnetoelectric coupling). On the other hand, we have, at the same time, the opportunity to offer a unified view of the common grounds for apparently different mechanisms for ferroelectricity in multiferroics. Thus, although the challenge of understanding how to simultaneously break space-inversion and time reversal symmetries is by itself a compelling reason for our research, one is also committed to contribute to the search of a common strategy for tuning the coupling of magnetism and ferroelectricity.

Here we should stress that the proposed systems passed already all tests during the PAC proof of concept experiments. All of them presented very clear and wieldy spectra, warranting a faultless analysis and exciting results. In fact, first experimental data are very promising, already with significant information that can be seen in the proposal figures 1, 4, and 7. Thus, at this point it is clear that we have already a fully developed methodology, that allied with previous relevant experience in the field warrants the feasibility of our approach, providing clear results to address the questions of the proposal.

Concerning the number of measurements necessary to accomplish a comprehensive knowledge of the effects under study we need c.a. 12 points for a proper characterization of each sample. If the time required to run PAC experiments is around 4-5 hours (e.g. for ^{111m}Cd and ^{204m}Pb) and having at our disposal 4 fully operational PAC machines we would be able to characterize realistically around 6 different samples in a typical 4 day-run period. This is the necessary number of measurements to accomplish our goals allowing characterizing temperature effects across the phase transitions. Nevertheless, we have to admit that our reasoning for asking such huge amount of 50 shifts for all the systems included was not clear. In fact, it is desirable to have a continued period of 4 days of beam time (e.g. for the ^{111m}Cd isotope) to build systematics on the same type, same batch of samples. However, the number of shifts can be cut significantly if there are other users asking for the same beam while not using the six-detector PAC machines. Other users have already approved beam time for the next two years using the same targets we need to extract our isotopes. Thus, with proper coordination among users, we will manage to obtain the same proposal outputs with much less requested shifts.

The number of shifts required in the proposal for the test cases are no longer considered and for the experiments using the long-lived isotopes (149Gd, 172Lu and 204Bi) one shift per year is sufficient to perform all the necessary measurements.

Summarizing: As recommended, we will focus our study in three complementary systems. These systems are chosen as they are prototypes of the three types of ferroelectricity in multiferroics. With this study we expect to contribute to the search of an integrated view on the control magnetoelectric coupling. We will be able to perform our focused research with ca. half of the beam time previously requested (i.e. now 26 shifts).

Summary of beam time request:

ISOLDE Beam	Approximate intensity (ion/μC of p-beam)	Target	Ion source	SHIFTS 1 + 1 years	Machine days per year
^{111m}Cd (49 m)	$1 . 10^8$	Molten Sn	Vadis or MK5	7+6	4
^{149}Gd (9.3d)	$3. 10^9$	Ta foil	Surface ioniser	1 + 1	1/3
^{172}Lu (6.7d)	$2 . 10^7$	Ta foil	Surface ioniser	1 + 1	2/3
^{204m}Pb (67m)	$5 . 10^7$	UCx	Pb RILIS	3 + 4	2
^{204}Bi (11.2h)	$1 . 10^7$	UCx	RILIS	1 + 1	2/3
			TOTAL (two years)=	**13 + 13**	