This paper, together with a recent paper by Bengs and Levitt, discusses and revisits the problem of the relaxational dynamics of a collection of interacting spins coupled to the vibrations of the lattice. It is common to assume that the spins are weakly coupled to the lattice and to use a perturbative approach (Born-Markov approximation) that allows to write a master equation of the Redfield kind for the density matrix reduced to the spin degrees of freedom [equation 21 of the main text]. Equation 21 does not have a Lindblad form. A Lindblad form is required to respect the physical properties of a density matrix (positive, trace conserving…).

In practice, in the literature further approximations are proposed for the evolution equation of the spin density matrix. The first two (i.e. the ones discussed in this paper) are:

To introduce a secular approximation that reduces Eq. 21 to Eq 40 or the identical Eq. 43. The equation has a Lindblad form and, by construction, relaxes the interacting spins to their Boltzmann equilibrium. Namely to \sigma_{Boltzmann) = exp(-\beta H_S)

In NMR (however see the historical paper by Tom Barbara) one is used instead to a semi-classical approximation of the Redfield equation which leads to Equation 65. Unfortunately Eq.65 does not have a Lindblad form. In general, for more than 1 spin this equation leads to misleading results that are discussed in this paper as well as in the previous paper by Bengs and Levitt. In my opinion this equation is very clumsy: on one side it does not reproduce a physical evolution and one the other side it is phenomenological. It works when the physics of the problem is well described by a single spin, but it will fail to capture many body effects, among others. Still within the Markovian assumption, instead of designing further approximations to turn Redfield's equation into a physically sound form (Lindblad), one can go one step back and examine the many possible ways of making the Markovian approximation. This leads to another proposal I would like to mention:

Recently a more general Lindblad evolution has been proposed (PERLind approaches, see e.g. Nathan & Rudner PRB 2020). It holds at the same level of approximation as Redfield's (Eq.21), i.e. second-order perturbation with respect to the coupling to the lattice, but it is of Lindblad form. In the limit of very weak coupling with the lattice one recovers the secular approximation of Eq. 40. For moderate coupling it captures the competition between the interactions among the spins and the coupling with the lattice. As a result the stationary state of this equation is not exactly exp(-\beta H_S) even if, in a strong magnetic field, the total magnetisation will be indistinguishable from the one predicted by Boltzmann. Indeed Boltzmann equilibrium is not expected to hold outside weak system-bath coupling. Recently we used this arguably more general equation to show that the spin temperature (generated by dipolar interactions) can be suppressed at high temperature due to the effect of the lattice vibrations (Maimbourg, Basko, Holtzmann, Rosso, PRL 2021).

In conclusion I think it is important (within the Markovian assumption) to stick with well-defined Lindblad forms and I think that this discussion is important. I also wish to advertise that a lot of physics can be found beyond the secular approximation.

We would like to thank you for the insightful comments and for pointing out recent investigations of Linblad operators. We added the suggested references to the papers of Nathan & Rudner, and of Maimbourg et al. on generalizations of the Linblad approach in the revised manuscript.