Supplementary Material to
"The origin of exciton mass in a frustrated Mott insulator Na_2IrO_3"

I. Pump fluence dependence. In an ideal setting the only effect of the $\hbar\omega = 1.5eV$ pump pulse is to generate non-equilibrium doublons and holons. However there is always some amount of inevitable parasitic sample heating, which potentially might have comparable effect on the overall signal. Therefore if the aim is to study the dynamics of electronic excitations on some order in the background, one should be careful to work in the regime where the pumping fluence is low enough to not to melt the order. The figure below (Fig.1) shows the fluence dependence of the signal dynamics. As can be seen, in the low fluence limit the relaxation dynamics is independent of fluence. In our experiment we work in this regime.

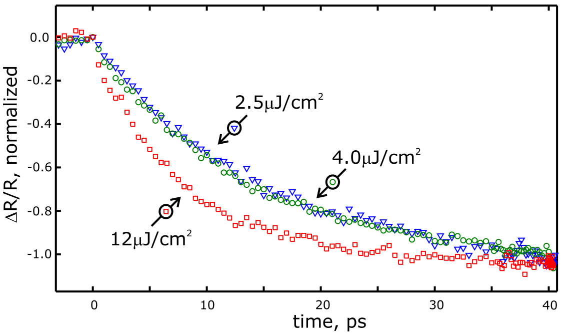

FIG. 1: **Pump-probe spectroscopy at 5K** Pump pulse ($1.5\,eV$) fluence dependence of the transient response of Na_2IrO_3. The decay rate of normalized curves is fluence independent for low fluence values indicating that in this regime the magnetic order is not melted by the pump pulse.

II. Push pulse effects. In this work we use the push pulse as a means to generate instant heating of the system already excited by the pump pulse and expect the push to be minimally coupled to the electronic degrees of freedom. In Fig.2a we show how the effect of the push pulse on the equilibrium electron configuration compares with that of the pump. As can be seen from the rightmost trace where the push pulse arrives before the pump, the response to the push pulse is very weak compared to the signal produced by pump pulse although the push pulse is much more intense ($100\mu J/cm^2$ for push and $4\mu J/cm^2$ for pump). This is compatible with the notion that push induced electronic transitions are prohibited due to selection rules. However as was mentioned in the main text there are still some electronic excitations generated by the push (otherwise the probe would not pick up any signal before pump pulse) even though in the first approximation there should be none. This is not surprising as the local moments picture is not exact and there is some inevitable admixture of $J_{\rm eff} = 3/2$ character to the $J_{\rm eff} = 1/2$ bands which is responsible for the generation of some "parasitic" electronic transitions between the lower and upper Hubbard bands.

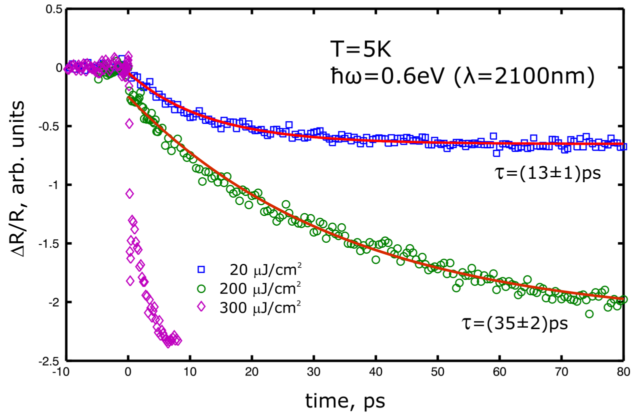

FIG. 2: **Push pulse effects at 5K**. Dependence of the relaxation rate on push fluence. Solid line is the fit to a single exponential curve with $\tau = (13 \pm 1)ps$. Note that the $20\mu J/cm^2$ fluence response is *faster* than the $200\mu J/cm^2$ one. The $300\mu J/cm^2$ is already melting the magnetic order and features the characteristic initial spike at $t = 0ps$

Fig.2b shows the fluence dependence of the response to the push pulse. In the lowest fluence regime the relaxation rate has the same (within error) time constant as in the $1.5\,eV$ pump case. This is expected, since a weak push pulse is not sufficient to melt the magnetic order and the "parasitic" doublons and holons relax much in the same way as those created by a $1.5\,eV$ pump pulse. However when the push fluence is increased to higher values

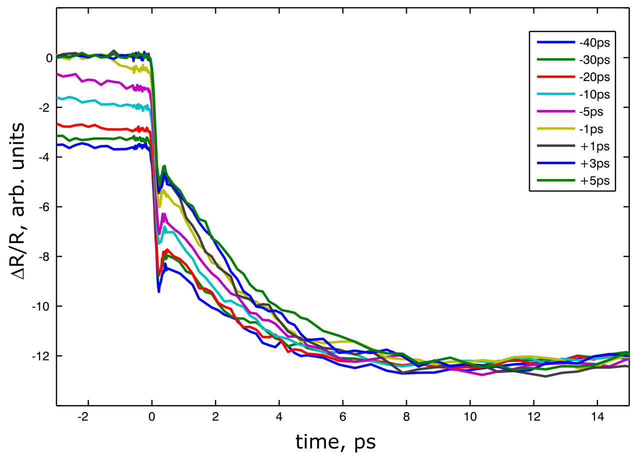

FIG. 3: **Effects of the push pulse arriving before the pump**. A closeup of the the Fig.3 from the main text in region around $t = 0ps$ with additional traces for which the pump pulse arrives after the $500\mu J/cm^2$ push pulse.

the behavior changes: the relaxation becomes *slower* at higher push fluence. This is opposite to the case of $1.5eV$ pumping, where the signal is getting *faster* with increasing pumping fluence (compare with Fig.1). This observation strongly suggests that the effect of the push pulse is qualitatively different from that of the pump pulse. In the latter case the laser pulse primarily causes (allowed) electronic transitions which subsequently relax by giving their energy away to the other degrees of freedom (spins, lattice). On the other hand the push pulse is only minimally coupled to the electronic degrees of freedom, therefore at high fluence values, alternative absorption mechanism begin to play a role. We already mentioned multi-phonon absorption and impulsive stimulated Raman scattering as possible candidates (there are Raman active modes in Na_2IrO_3 which are accessible with our push pulse bandwidth (Ref.[36] in the main text)). In general this is an interesting open problem which we plan to address in our future work. However we would like to emphasize that since we are only interested in the push as a means to instantaneously melt the magnetic order, the particular absorption mechanism does not affect the conclusions of this work.

Finally for the comparison purposes we also show a closeup of the Fig.3 from the main text where we add a few additional traces corresponding to the cases where the pump pulse arrives after an intense push pulse that is sufficiently strong to melt the magnetic order. The majority of the signal therefore is coming from the push pulse, however the pump pulse is also producing a noticeable contribution. This is particularly visible for the $+1ps$ trace in Fig. 3.