Muons and Frustrated Magnetism in NiGa$_2$S$_4$ and Pr$_2$Ir$_2$O$_7$

D E MacLaughlin$^1$, Y Nambu$^{2,4}$, Y Ohta$^2$, Y Machida$^2$, S Nakatsuji$^2$ and O O Bernal$^3$

$^1$Dept. of Physics & Astronomy, Univ. of California, Riverside, California 92521, USA
$^2$Institute for Solid State Physics, Univ. of Tokyo, Kashiwa 277-8581, Chiba, Japan
$^3$Dept. of Physics & Astronomy, California State Univ., Los Angeles, California 90032-4221, USA

E-mail: macl@physics.ucr.edu

Abstract. Geometrically frustrated magnets are of interest because of the novel phenomena that arise from their exotic ground states and low-lying excitations. Muon spin rotation and relaxation ($\mu$SR) is a sensitive probe of magnetism on the local (atomic) distance scale, and is an attractive tool for the study of frustrated magnets. The muon carries a unit electric charge, however, which can have an appreciable effect on local properties. We discuss two cases where such an effect might be involved. In the 2D triangular antiferromagnet NiGa$_2$S$_4$ the ‘magnetic’ specific heat is field-independent up to 7 T, suggesting nonmagnetic excitations, but $\mu$SR experiments reveal Ni spin freezing below $\sim 9$ K and strong magnetic fluctuations down to 25 mK. Comparison with Ga nuclear quadrupole resonance data suggests, however, that the muon charge does not cause this discrepancy. In the pyrochlore iridate Pr$_2$Ir$_2$O$_7$ muon spin relaxation due to a distribution of quasistatic fields is observed over a wide temperature range. The data strongly suggest hyperfine-enhanced $^{141}$Pr nuclear magnetism, which requires a nonmagnetic Pr$^{3+}$ ground state. This may be due to lifting of the Pr$^{3+}$-non-Kramers degeneracy by the muon electric field or, at least in part, a property of a spin-liquid-like many-body ground state.

1. Introduction

Geometrical frustration in solids refers to situations where the geometry of a crystal lattice prohibits simultaneous satisfaction of all interactions between atoms [1]. In general frustration can lead to macroscopic degeneracy of the (classical or quantum) ground state and a very large density of low-lying excited states, due to the combinatorics involved in distributing the frustrated interactions. This in turn leads to novel phenomena not found in unfrustrated systems such as suppression of the ordering temperature, sometimes to $T = 0$. Magnets with geometrically frustrated interactions provide a testing ground for understanding the interplay between frustration, magnetic order, and a wide variety of exotic states.

Magnetic resonance techniques use spin probes (nuclei, electrons, muons) to determine magnetic properties on the atomic scale [2]. Electronic and/or nuclear spins in the environment of a spin probe produce a local field $B^{\text{loc}}$ at the spin-probe site. This local field fluctuates randomly.

---

4 Present address: Dept. of Physics & Astronomy, Johns Hopkins Univ., Baltimore, Maryland 21218, USA.
in time due to thermal excitations, and may also have a quasistatic component \( \langle B^{\text{loc}} \rangle \). The local nature of the spin probe renders magnetic resonance studies complementary to experiments that perform bulk averages, such as measurements of thermodynamic and transport quantities, and to diffraction and scattering techniques (neutron, x-ray, ...), which are reciprocal-space probes.

In time-differential muon spin rotation (td-\( \mu \)SR) \([3, 4]\) the spin probes are spin-polarized muons implanted in the sample. Usually positive muons (\( \mu^+ \)) are used, which can be considered short-lived impurities after they come to rest at interstitial sites in the sample. The \( \mu^+ \) spins precess in the resultant of \( B^{\text{loc}}(t) \) and any applied field, and eventually decay: \( \mu^+ \rightarrow e^+ + \nu_e + \bar{\nu}_\mu \) (mean lifetime 2.2 \( \mu s \)). The direction of the decay positron emission is correlated with the \( \mu^+ \) spin orientation at the time of decay. Thus the \( \mu^+ \) spin-polarization component in a given direction is proportional to the difference between parallel and antiparallel count rates for that direction. The time evolution of this asymmetry is the primary measurement in td-\( \mu \)SR.

In transverse-field \( \mu \)SR (TF-\( \mu \)SR) muon spin precession in a magnetic field perpendicular to the \( \mu^+ \) polarization results in oscillations of the asymmetry at the \( \mu^+ \) precession frequency. In zero applied field (ZF-\( \mu \)SR) or longitudinal field (LF-\( \mu \)SR) the only precession arises from components \( B^{\text{loc}}_\perp \) of the local field perpendicular to the \( \mu^+ \) spin direction. Thermal spin excitations give rise to fluctuations of \( B^{\text{loc}}_\perp \), which dephase or ‘relax’ the \( \mu^+ \) spin polarization.

\( \mu \)SR experiments are useful for studies of geometrically frustrated magnets (see e.g. \([5]\)) for several reasons. Muon spins are local probes in real space; unlike neutron diffraction, they do not depend on spatial correlation or long-range order to detect quasistatic magnetism or spin fluctuations. They are therefore ideally suited to studies of disordered spin freezing, which is often found in geometrically frustrated magnets. ZF-\( \mu \)SR is an extremely sensitive probe of weak quasistatic magnetism, being able to detect quasistatic moments as weak as \( 10^{-3} \mu_B \). The dynamic relaxation is strong and easy to measure.

The electric field from the \( \mu^+ \) charge can, however, perturb the phenomenon under study. In metals such effects are minimized by screening of the charge; superconducting and magnetic transition temperatures observed in \( \mu \)SR are typically the same as the bulk values, indicating that the muon charge does not have a major effect on the ordered state. \( \mu^+ \)-induced effects have been observed \([6, 7]\), however, and should as a rule be searched for carefully in all \( \mu \)SR data.

In the present paper we discuss \( \mu \)SR experiments in two prototypical geometrically frustrated antiferromagnets, the two-dimensional triangular insulator NiGa\(_2\)S\(_4\) \([8, 9]\) and the 3D pyrochlore Kondo lattice Pr\(_2\)Ir\(_2\)O\(_7\) \([10, 11]\), where there is reason to suspect \( \mu^+ \)-induced effects. We shall argue that in NiGa\(_2\)S\(_4\) such effects probably do not affect the results qualitatively, whereas in Pr\(_2\)Ir\(_2\)O\(_7\) the \( \mu^+ \) charge is more important. Even so, \( \mu \)SR experiments seem to yield reliable information for both materials.

2. NiGa\(_2\)S\(_4\)

2.1. Structure, specific heat

In the quasi-2D antiferromagnet NiGa\(_2\)S\(_4\) the Ni\(^{2+}\) ions (\( S = 1 \)) form a geometrically frustrated two-dimensional triangular sublattice \([8]\). Neutron scattering experiments reveal the short-range spin-correlated state at low temperatures shown in figure 1. The correlation length is short (~25 Å), and there is no long-range order. The ‘magnetic’ specific heat \( C_M \) (i.e. after correction for the phonon contribution) is shown in figure 2. It is unusual in a number of respects. There are two maxima of \( C_M(T) \), one centered around the Curie-Weiss temperature \( \theta_W \approx 80 \text{ K} \) and the other at \( T_{\text{peak}} \approx 10 \text{ K} \), where the magnetic susceptibility also exhibits a weak maximum. This

\( ^5 \) We note that \( \langle B^{\text{loc}} \rangle \) is sampled only for times shorter than the lifetime of the probe spin state, and thus appears stationary if its fluctuations are sufficiently slow, or ‘quasistatic’. Quasistatic and strictly static fields cannot be distinguished by magnetic resonance measurements, which merely put a lower limit (viz. the probe-spin lifetime) on the correlation time for fluctuations of \( \langle B^{\text{loc}} \rangle \).
seems to indicate freezing of single-spin excitations below $\sim |\theta_W|$, whereas more complicated highly degenerate low-energy states due to magnetic frustration in the triangular lattice are involved below 10 K. At low temperatures $C_M \propto T^2$, and at all temperatures $C_M$ is independent of applied field up to 7 T to within the experimental resolution of a few percent. There was no evidence for bulk spin freezing in the early data, and it was suggested that at low-temperatures NiGa$_2$S$_4$ is a ‘viscous’ (i.e. slowly-fluctuating) spin liquid.

2.2. $\mu$SR; comparison with NQR
In contrast, $\mu$SR experiments [12, 13] reveal the onset of a quasistatic $\mu^+$ field $\langle B_{\text{loc}} \rangle$ in NiGa$_2$S$_4$ below a freezing temperature $T_f \approx 9$ K (figure 3). The oscillations are strongly damped, indicative of a broad distribution of $\langle B_{\text{loc}} \rangle$ consistent with a short correlation length; the average field is $\langle B_{\text{loc}} \rangle_{\text{av}} \approx 2.5$ kG. The oscillation frequency $\omega_\mu$, which in conventional magnets is proportional to the order parameter in the magnetically ordered phase, is well described by the mean-field Brillouin function for $S = 1$ [figure 3(a)].

The dynamic relaxation in NiGa$_2$S$_4$ is also widely distributed, as evidenced by the sub-exponential polarization decay (upward curvature in a semilog plot of decay curve versus time; data not shown) observed in both $\mu$SR and $^{69}$Ga nuclear quadrupole resonance (NQR) [14] experiments. A relaxation function of the so-called ‘stretched exponential’ form $\exp[-(\Lambda t)^\beta]$ can be fit to the data; here $\Lambda$ is a typical rate associated with the distribution and $\beta < 1$ qualitatively parametrizes the distribution width. The temperature dependence of $\Lambda$ in NiGa$_2$S$_4$ is given in figure 3(b). Data were taken in a dilution refrigerator for temperatures below $\sim 3$ K, and in a gas-flow cryostat above $\sim 2$ K. The low-temperature data are less accurate because in the dilution refrigerator a significant fraction of muons stop in the silver cold finger.

In figure 3(b) it can be seen that $\Lambda$ increases strongly as $T_f$ is approached from above, as expected from critical slowing down at a phase transition. Below $T_f$ $\Lambda$ decreases to a minimum at $\sim 2$ K, and remains $\approx 5$ $\mu$s$^{-1}$ down to the lowest temperatures of measurement. A lower bound for the Ni-spin correlation time at 25 mK is $\Lambda^{-1} \approx 2 \times 10^{-7}$ s $\gg \hbar/k_B T_f \approx 10^{-12}$ s. Thus
in NiGa$_2$S$_4$ the Ni spins are either completely frozen or else form a very viscous spin liquid.

The behaviour of $\Lambda$ Below $\sim$1 K is in contrast to the zero-field $^{69}$Ga spin-lattice relaxation rate $1/T_1$ measured using NQR [14], shown in figure 4. NQR and $\mu$SR are both local spin-probe techniques, and in general would be expected to yield similar results. But below $\sim$1 K $1/T_1$(NQR) follows a $T^3$ law, compared to the temperature-independent $\mu^+$ relaxation rate $\Lambda$.

2.3. Effect of muon charge?

The field independence of $C_M$ in NiGa$_2$S$_4$ (figure 2) suggests a low-temperature state without spin freezing. This seems to conflict with the $\mu$SR evidence for at least quasistatic spin freezing and the strong magnetic field dependence of the $\mu$SR relaxation at low temperatures [13, 15]. It has been suggested [16] that the low-temperature state is in fact nonmagnetic, but the muon itself “...unveils a frozen magnetic moment in its vicinity.” However, there is also evidence for spin freezing from $^{69}$Ga NQR spectra below $\sim$2 K [14], which are broadened by about the same field distribution width as obtained from the $\mu$SR quasistatic relaxation rate. Unlike muons, $^{69}$Ga nuclei are native to the material and do not perturb the Ni$^{2+}$ magnetism.

The difference between $\mu$SR and NQR relaxation rates at low temperatures (figures 3 and 4) also suggests an effect of the muon on its magnetic environment. A similar discrepancy is found in the kagomé lattice-like compound volborthite [Cu$_3$V$_2$O$_7$(OH)$_2$·2H$_2$O], where it has been argued [5, 17] that, on the contrary, the effect is intrinsic: the symmetric position of $^{51}$V NMR nuclei with respect to the kagomé lattice filters out dominant spin fluctuations with a particular spatial configuration. Similarly, in NiGa$_2$S$_4$ the $^{69}$Ga nuclei are centered on triangles in the Ni planes, raising the possibility of a similar picture in which no effect of the $\mu^+$ charge is involved. This is of course speculation, and more work is needed to identify the fluctuation modes in NiGa$_2$S$_4$. But the case for $\mu^+$-induced magnetism is at least open to question.

---

6 Along with the spin-echo relaxation rate $1/T_2$, which we do not consider here.

7 Between $\sim$2 K and $T_\mu$ the $^{69}$Ga NQR spin-echo signal is ‘wiped out’, i.e. decays too quickly to be observable [14].
It should be noted that $T_N = 0$ for the ideal 2D Heisenberg antiferromagnet [18], with or without frustration, but perturbations (anisotropy, interlayer interactions, defects, ...) can produce a nonzero transition temperature. In this light the NQR and $\mu$SR evidence for a transition in NiGa$_2$S$_4$ is perhaps not surprising, although the transition may be more exotic [19]. But the extreme field independence of the specific heat remains difficult to understand.

3. Pr$_2$Ir$_2$O$_7$

Pr$_2$Ir$_2$O$_7$ is a metallic compound in which inelastic neutron scattering experiments [10] indicate a well-isolated Pr$^{3+}$ $\Gamma_3$ non-Kramers doublet crystalline electric field (CEF) ground state. Transport properties suggest a Kondo effect even below a RKKY interaction temperature \(\sim 20 \text{ K} \) [11]; geometrical frustration has been suggested as a mechanism for suppression of magnetic order and a spin-liquid state.

LF-$\mu$SR experiments in weak applied fields have been performed to characterize local quasistatic and dynamic magnetism in this compound [20]. Kubo-Toyabe (K-T) relaxation functions [21] are observed between 25 mK and 200 K, indicating a Gaussian distribution of quasistatic $\langle B_{\text{loc}} \rangle$ over this temperature range. The K-T functional form is expected from nuclear dipolar fields, but the $T \to 0$ quasistatic $\mu^+$ spin relaxation rate $\Delta(0) \approx 8 \mu s^{-1}$ is enhanced by a factor of $\sim 50$ over the expected value for $^{141}$Pr nuclei [20] and indicates a randomly-oriented quasistatic moment of order $0.1 \mu_B$. The temperature dependence of $\Delta$, shown in figure 5, shows no evidence of a magnetic phase transition. Instead, $\Delta(T)$ decreases smoothly with increasing temperature but remains nonzero up to 125 K. The data rule out ordering of the full Pr$^{3+}$ CEF ground-state moment (3.0 $\mu_B$) down to 25 mK.

3.1. Hyperfine-enhanced nuclear magnetism

We argue [20] that this quasistatic field distribution is due to hyperfine enhancement of the $^{141}$Pr nuclear magnetism [22, 23], in which $^{141}$Pr nuclear moments are enhanced by a Van Vleck-like mechanism when the Pr$^{3+}$ ions are in a nonmagnetic (e.g. singlet) ground state. Evidence that the origin of this relaxation is nuclear rather than electronic magnetism comes from a plot of $\Delta$ versus the molar bulk susceptibility $\chi_{\text{mol}}$, shown in figure 6. For small $\chi_{\text{mol}}$ (high temperatures) $\Delta \propto \chi_{\text{mol}}$, as expected from the hyperfine enhancement mechanism: the enhancement factor is $a_4 F$, where $a_4$ is the $^{141}$Pr atomic hyperfine coupling constant [22, 23]. The slope in figure 6 yields a reasonable estimate of the unenhanced $^{141}$Pr dipolar field [20].

**Figure 5.** Temperature dependence of weak LF-$\mu$SR quasistatic relaxation rate $\Delta$ in Pr$_2$Ir$_2$O$_7$, 25 mK $\leq T \leq 125$ K.

**Figure 6.** Relaxation rate $\Delta$ versus molar bulk susceptibility $\chi_{\text{mol}}$, with temperature an implicit parameter, in Pr$_2$Ir$_2$O$_7$, 2 K $\leq T \leq 125$ K.

$\Delta \propto \chi_{\text{mol}}$, as expected from the hyperfine enhancement mechanism: the enhancement factor is $a_4 F$, where $a_4$ is the $^{141}$Pr atomic hyperfine coupling constant [22, 23]. The slope in figure 6 yields a reasonable estimate of the unenhanced $^{141}$Pr dipolar field [20].

Ir and O nuclear magnetic moments are too small to contribute appreciably.
3.2. Effect of $\mu^+$ charge

Assuming this picture, the downward curvature in $\Delta$ versus $\chi_{\text{mol}}$ at low temperatures (figure 6) suggests that the local susceptibility $\chi_{\text{loc}}$ of Pr$^{3+}$ ions is reduced in the neighborhood of $\mu^+$ sites. This would happen if the Pr$^{3+}$ non-Kramers degeneracy were lifted by the $\mu^+$ electric field. Such an effect would also contribute to, and could conceivably explain completely, the nonmagnetic (singlet) Pr$^{3+}$ ground state required for hyperfine-enhanced nuclear magnetism [20].

To test for muon-induced reduction of $\chi_{\text{loc}}$ we have measured the $\mu^+$ frequency shift $K = \Delta \omega_{\mu}/\omega_{\mu}$ using TF-$\mu$SR [3]. In paramagnets $K$ is a measure of the local susceptibility: $K = a_{\mu} \chi_{\text{loc}}$, where $a_{\mu}$ describes the coupling between electronic spins and the $\mu^+$ spin [2]. In figure 7 $K$ is plotted against $\chi_{\text{mol}}$, with temperature an implicit parameter, before and after correction for the Lorentz and demagnetization fields. These are quite significant and the correction is not accurate, due to uncertainties in sample dimensions and the powder packing fraction. The error bars on the corrected shift $K_{\text{corr}}$ are mainly due to these uncertainties in the correction, and are not random between points. The curvature in $K_{\text{corr}}$ versus $\chi_{\text{mol}}$ (figure 7), like that in $\Delta$ versus $\chi_{\text{mol}}$ (figure 6), is evidence for a deviation of $\chi_{\text{loc}}$ from the bulk $\chi_{\text{mol}}$, i.e. for muon-induced modification of $\chi_{\text{loc}}$ [6, 7]. Other mechanisms, such as contributions from CEF excited states or the Kondo effect [24], would not lead to the observed hyperfine enhancement.

Figure 8 gives $\Delta$ versus $K_{\text{corr}}$. It can be seen that the relation is much more linear than in figure 6 over the entire temperature range, even if the correction is not accurate. We conclude that $\Delta$ tracks the local susceptibility accurately, yielding further evidence that the K-T relaxation in Pr$_2$Ir$_2$O$_7$ is due to hyperfine-induced $^{141}$Pr nuclear magnetism.

### Figure 7

Raw and corrected $\mu^+$ frequency shift versus molar bulk susceptibility $\chi_{\text{mol}}$, with temperature an implicit parameter, in Pr$_2$Ir$_2$O$_7$.

### Figure 8

Relaxation rate $\Delta$ versus corrected $\mu^+$ frequency shift $K_{\text{corr}}$, with temperature an implicit parameter, in Pr$_2$Ir$_2$O$_7$.

3.3. Phase transition

Recent specific heat measurements indicate a phase transition in some Pr$_2$Ir$_2$O$_7$ samples at $T_c \sim 0.8$ K, apparently correlated with Pr-Ir stoichiometry [25]. Examples are shown in figure 9. As shown in figure 10(a), however, ZF-$\mu$SR experiments on similar samples do not exhibit the large increase of quasistatic $\mu^+$ relaxation rate below $T_c$ expected for a magnetic transition. For the full non-Kramers doublet ground-state moment of 3.0 $\mu_B$ the quasistatic relaxation would not be directly observable, but would cause a decrease of the initial muon asymmetry (signal amplitude) to $\sim 1/3$ of its value above $T_c$. No such decrease is observed [figure 10(b)]. There is also no feature in the dynamic $\mu^+$ relaxation rate at $T_c$ [figure 10(a)].

The splitting of the ground-state Pr$^{3+}$ doublets noted above [20] could suppress the quasistatic magnetism locally. In metals this effect should be quite local because of screening of the $\mu^+$
Figure 9. Low-temperature specific heat of three samples of Pr$_2$Ir$_2$O$_7$. Data from [25].

Figure 10. (a) Quasistatic (open symbols) and dynamic (filled symbols) $\mu^+$ spin relaxation rates, and (b) $\mu$SR asymmetry (signal amplitude) in Pr$_2$Ir$_2$O$_7$, for samples with (PI53C) and without (PI8E) a specific-heat anomaly at $T_c$.

charge; in the intermetallics PrNi$_5$ [6] and PrIn$_3$ [7] significant CEF modification is confined to nearest-neighbour Pr$^{3+}$ ions. Lattice-sum calculations of the dipolar field at representative $\mu^+$ sites in Pr$_2$Ir$_2$O$_7$ show, however, that even for complete suppression out to 25 Å, including $\sim$900 Pr ions, the expected contribution to $\langle B_{\text{loc}} \rangle$ from more distant Pr ions would be $\sim$40 G $\approx 5 \mu s^{-1/\gamma_{\mu}}$. Onset of such a contribution at $T_c$ would be a $\sim$20% effect, i.e. easily visible; its absence [figure 10(a)] is evidence that the phase transition in Pr$_2$Ir$_2$O$_7$ is nonmagnetic.

4. Conclusions

4.1. NiGa$_2$S$_4$

In this compound NQR and $\mu$SR experiments appear at first sight to both confirm and contradict each other. Quasistatic spin freezing is seen by both techniques, but it is not clear whether or not the muon charge has a significant influence on the low-temperature spin dynamics since the temperature dependence of dynamic spin relaxation is different for the two techniques (cf. section 2.2). This may be due to differences in local surroundings and consequent filtering of spin fluctuations as in volborthite [5], but the question needs to be studied in more detail. The most important issue in NiGa$_2$S$_4$ remains how to reconcile the field-independent specific heat with the evidence from NQR and $\mu$SR for disordered quasistatic magnetism.

4.2. Pr$_2$Ir$_2$O$_7$

Here the evidence for $\mu^+$-induced modification of the $\mu$SR data (nonlinear $K$ versus $\chi_{\text{mol}}$) is fairly clear. It remains uncertain, however, whether $\mu^+$-induced CEF level splitting is the only mechanism for a Pr$^{3+}$ singlet ground state or, alternatively, whether a many-body singlet ground state associated with the frustrated Pr$^{3+}$ spin system also contributes.

Even though the $\mu^+$ electric field lifts the non-Kramers degeneracies of nearest-neighbor and possibly several near-neighbor Pr$^{3+}$ ions, a significant local field from more distant Pr$^{3+}$ spins would remain if these were frozen, with or without long-range order (section 3.3). The absence of $\mu$SR evidence for Pr$^{3+}$ magnetism (other than the $4f$ polarization involved in the
nuclear hyperfine enhancement mechanism) strongly suggests that in Pr$_2$Ir$_2$O$_7$ the ground state, low-lying excitations, and phase transition at 0.8 K are all nonmagnetic in character.

Acknowledgments
We are grateful for technical assistance from the TRIUMF Centre for Molecular and Materials Science, where these experiments were carried out. We wish to thank E J Ansalido and K Onuma for assistance with the experiments, and P Dalmas de Réotier, H Kawamura, R F Kiefl, R R P Singh, A Yaouanc, and S Zhao for useful discussions. This work was partially supported by U.S. NSF Grant nos. 0422671 and 0604015 (Riverside) and 00604015 (Los Angeles), by a Grant-in-Aid (No. 21684019) from the Japanese Society for the Promotion of Science (JSPS), and also by Grants-in-Aid for Scientific Research on Priority Areas (Nos. 17071003 and 19052003) from the Ministry of Education, Culture, Sports, Science and Technology (MEXT), Japan.

References
[1] Ramirez A P 1994 *Ann. Rev. Mater. Sci* 24 453–480
[2] Slichter C P 1990 *Principles of Magnetic Resonance* 3rd ed (New York: Springer-Verlag)
[3] Schenck A 1985 *Muon Spin Rotation Spectroscopy: Principles and Applications in Solid State Physics* (Bristol & Boston: A. Hilger)
[4] Blundell S J 1999 *Contemp. Phys.* 40 175–192
[5] Mendels P, Oleari A, Bert F, Bonod L, Collin G, Ueland B, Schiffer P, Cava R J, Blanchard N, Duc F and Trombe J C 2007 *J. Phys.: Condens. Matter* 19 145224–1–9
[6] Feyerherm R, Amato A, Grayevsky A, Gygax F N, Kaplan N and Schenck A 1995 *Z. Phys. B* 99 3–13
[7] Tashma T, Amato A, Grayevsky A, Gygax F N, Pinkpank M, Schenck A and Kaplan N 1997 *Phys. Rev. B* 56 9397–9405
[8] Nakatsuji S, Nambu Y, Tononura H, Sakai O, Jonas S, Broholm C, Tsunetsugu H, Qiu Y and Maeno Y 2005 *Science* 309 1697–1700
[9] Nakatsuji S, Nambu Y, Onuma K, Jonas S, Broholm C and Maeno Y 2007 *J. Phys.: Condens. Matter* 19 145232–1–7
[10] Machida Y, Nakatsuji S, Tononura H, Tayama T, Sakakibara T, van Duijn J, Broholm C and Maeno Y 2005 *J. Phys. Chem. Solids* 66 1435–1457
[11] Nakatsuji S, Machida Y, Maeno Y, Tayama T, Sakakibara T, van Duijn J, Balicas L, Millican J N, Macaluso R T and Chan J Y 2006 *Phys. Rev. Lett.* 96 087204–1–4
[12] Yasuanc A, Dalmas de Réotier P, Chapuis Y, Marin C, Lapertot G, Cervellino A and Amato A 2008 *Phys. Rev. B* 77 092403–1–4
[13] MacLaughlin D E, Nambu Y, Nakatsuji S, Heffner R H, Shu L, Bernal O O and Ishida K 2008 *Phys. Rev. B* 78 220403(R)–1–4
[14] Takeya H, Ishida K, Kitagawa K, Ibara Y, Onuma K, Maeno Y, Nambu Y, Nakatsuji S, MacLaughlin D E, Koda A and Kadono R 2008 *Phys. Rev. B* 77 054429–1–13
[15] MacLaughlin D E, Nambu Y, Nakatsuji S, Ishida K, Heffner R H, Shu L and Bernal O O 2009 *J. Phys.: Conf. Ser.* 145 012040–1–5
[16] Varma C 2008 Mysterious order for spins on a triangular lattice. *J. Club Condens. Matter Phys.* November 2008
[17] Bert F, Mendels P, Bonod L, Oleari A, Ladieu F, Trombe J C, Duc F, Baines C, Amato A and Hillier A 2006 *Physica B* 374–375 134–137
[18] Chakravarty S, Halperin B I and Nelson D R 1989 *Phys. Rev. B* 39 2344–2371
[19] Kawamura H, Yamamoto A and Okubo T 2009 *Z_2 vortex ordering of the triangular-lattice Heisenberg antiferromagnet* arXiv:0909.0121
[20] MacLaughlin D E, Ohta Y, Machida Y, Nakatsuji S, Luke G M, Ishida K, Shu L and Bernal O O 2009 *Physica B* 404 667–670
[21] Hayano R S, Uemura Y J, Imazato J, Nishida N, Yamazaki T and Kubo R 1979 *Phys. Rev. B* 20 850–859
[22] Murao T 1971 *J. Phys. Soc. Jpn.* 31 683–690
[23] Bleaney B 1973 *Physica (Utrecht)* 69 317–329
[24] Ohishi K, Heffner R H, Ito T U, Higemoto w, Morris G D, Hur N, Bauer E D, Sarrao J L, Thompson J D, MacLaughlin D E and Shu L 2009 *Phys. Rev. B* 80 125104–1–7
[25] Ohta Y, Machida Y and Nakatsuji S Low-temperature phase transition in Pr$_2$Ir$_2$O$_7$ unpublished