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Coherent dipolar correlations in the low-temperature phase of geometrically frustrated $SrCr_{8-x}Ga_{4+x}O_{19}$

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Abstract

We propose a model for the nature of the low-temperature phase of a geometrically frustrated antiferromagnet with a Kagomé lattice, $SrCr_{8-x}Ga_{4+x}O_{19}$. We propose that the long-range dipolar interaction between the magnetic Cr^{3+} ions introduces correlations in their zero-point dynamics. The dipolar ground state has the spins performing correlated oscillations in a coherent state with a well defined global phase. We calculate the magnon excitations of such a dipolar array and find good agreement with the spin-wave velocities inferred from measurements of the specific heat. We can further explain the unusual muon spin-relaxation signal of this phase.

1. Introduction

The problem of the low-temperature magnetic phase of materials with strong geometric frustration against antiferromagnetic (AFM) order is a long-standing one [1]. Especially interesting are systems where there appears below a critical temperature a phase which has some properties of a spin-glass but also of an ordered spin phase, such as $SrCr_{8-x}Ga_{4+x}O_{19}$ (SCGO), on which we shall concentrate in this paper. Above the transition temperature ($T_c \simeq 3$ K) SCGO shows AFM correlations with a Curie–Weiss temperature [2] of $\theta_{CW} \simeq -500$ K, indicating strong exchange interactions ($J \sim 100$ K) between nearest-neighbour magnetic ions. The transition is to a low-temperature phase ($T_c/J \ll 1$) without static long-range spin ordering, but the transition does show a characteristic of spin-glass, i.e. large difference between the zero-field-cooled (ZFC) and field-cooled (FC) static magnetic susceptibility [2, 3]. On the other hand, specific-heat measurements [2, 4–10] point to a low-temperature phase which has spin-waves, i.e. a long-range order [8] on scales >650 Å. The experimental data also indicate almost no frozen (static) magnetic moment [6, 22], down to the lowest temperatures.

Classical [1, 11–13] and quantum [14–16] theoretical studies of the Heisenberg AFM on the two-dimensional Kagomé lattice have concluded that there is no finite-temperature

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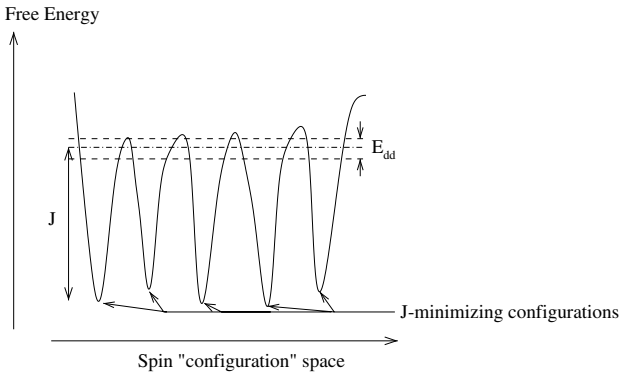

Figure 1. Schematic description of the free-energy surface of the spin configuration on the Kagomé lattice [1]. The ground state of the exchange interactions (J, dash–dotted line) is not localized in any particular minima due to the geometric frustration. The dipole-induced splitting of the ground state (equation (1)) is indicated by the pair of dashed lines.

transition to a phase with long-range spin ordering. There are indications that, although the ground state may be disordered, fluctuations will tend to select coplanar configurations, and maybe even a $\sqrt{3} \times \sqrt{3}$ state [14, 16], at $T \to 0$. Though these coplanar configurations are separated from non-coplanar configurations by zero-point energy barriers of height [1, 12–15] $\sim J$ (figure 1), the system appears not be localized at finite T in a particular domain of the ground-state manifold [16] (i.e. a 'spin-liquid'). For the system to be spread over the different coplanar states [15] it must retain a zero-point energy (i.e. quantum fluctuations) of the order of the barrier height, i.e. $\sim J$ (depending on the spin [15] S). In a semi-classical picture the ground state of the nearest-neighbour Heisenberg model for the frustrated Kagomé AFM is spread out over a multi-well spin-space potential [1, 12], where the minima of the potential are at the coplanar spin arrangements (figure 1), and the ground state is not localized in any minima due to the high ($\sim J$) zero-point energy.

We do not wish to attempt a detailed description of this ground state, which is a very complicated problem, but rather assume that all the correlations due to the nearest-neighbour exchange interactions are included in the large (of order J) zero-point energy of the spins in the ground state, and in the constraint of zero total spin moment on each triangle.

We here propose a new model to describe the low-temperature phase of SCGO. We first note that, in addition to the nearest-neighbour exchange interactions, there is a direct magnetic dipole–dipole interaction between the Cr^{3+} ions. In SCGO this dipolar interaction energy is of order (taking the Cr^{3+} magnetic moment as [4] 3.8 μ_B) $E_{dd} = (3.8\,\mu_B)^2/a^3 \sim 4$ K, where the nearest-neighbour distance is [8] $a = 2.93$ Å. This (three-dimensional) dipolar interaction is therefore of the order of the transition temperature and is important in determining the properties of the low-temperature phase [11, 17]. Other interactions beyond the exchange interactions are not considered in our treatment for the following reasons. The single-ion anisotropy [18] is weaker than the dipolar interactions (<1 K). Next-nearest-neighbour interactions, if important, will limit the ground state to such an extent as to freeze the spins [19], which is not observed[2]. Since we are dealing with a macroscopically degenerate ground state of the exchange interactions, we are looking for another interaction which will break this degeneracy strongly enough to produce some long-range order, while maintaining the dynamic

[2] The coherence of the oscillating spins can be destroyed if an additional static local magnetic moment (Fe ion) is added [5]. The system now has a spin-glass (SG) transition temperature $T_{SG} \sim 25$ K, i.e. an order of magnitude larger than the dipole-induced transition temperature T_c discussed in this paper.

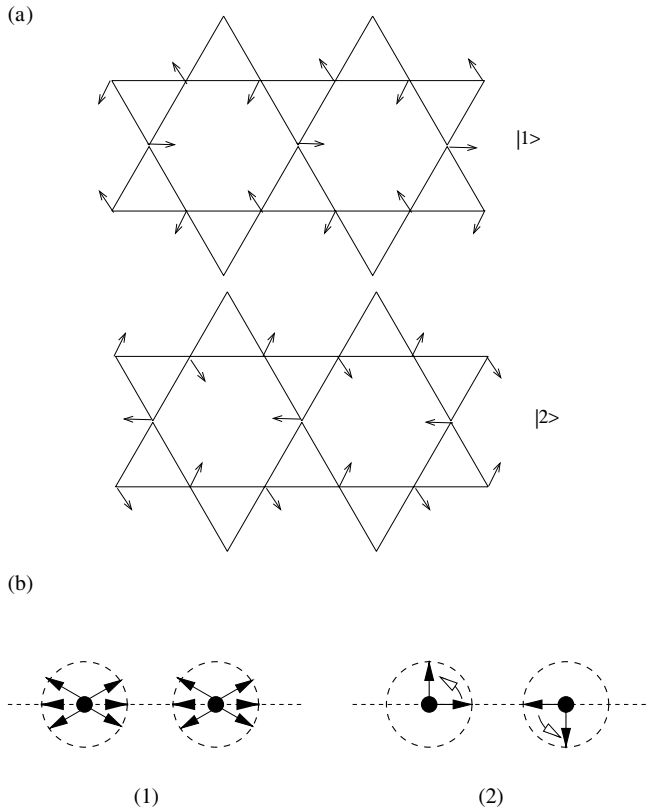

Figure 2. (a) The local minimum configuration for the spins with respect to the dipolar interactions in the Kagomé planes (the plane of the figure). The spins are in the Kagomé planes. The system makes zero-point coherent oscillations between these two configuration. (b) Schematic illustration of the spins (1) in the non-coherent ground state ('spin-liquid') due to the exchange interactions alone, and (2) making coherent zero-point oscillations between the two configurations of (a) in the dipole-induced coherent phase. The straight dashed lines represent the Kagomé planes, viewed sideways.

nature of the spins. In this work we propose that dipolar interactions produce a state with off-diagonal long-range order (ODLRO) which has no static diagonal long-range order (DLRO) of the spins in the Kagomé planes. This is the main proposal of this paper.

2. Coherent zero-point dipoles

Introducing the effect of dipolar interactions, the energies of the different spin configurations over which the ground state is spread are now split by the dipolar energy (figure 1). We find that due to the Kagomé geometry and the constraint of coplanar and zero total spin moment on each triangle (an angle of 120° between nearest neighbours), a model spin configuration that minimizes the dipolar interaction is as shown in figure 2(a). We do not consider the intervening triangular planes as participating in the dipolar ordering, since experiments [20] indicate they have a different behaviour from the Kagomé planes.

The configurations of figure 2 are in the class [21] $Q = 0$ or $Q_1 = Q_2$. These have slightly higher energy at $T = 0$ than the $\sqrt{3} \times \sqrt{3}$ configurations, as far as the exchange interactions

are concerned ('order from disorder' effect). We have to remember that these considerations pertain to an ordered state with a static moment at $T = 0$, since the exchange interactions alone do not lead to a finite-temperature transition. Our proposed state is not a state with a static moment, or static long-range order in the configurations of figure 2. Rather, we propose a state where the zero-point fluctuations of the exchange interaction are made to pass coherently through the states that minimize the dipolar interactions. Where the dipolar interactions are truly the only ones present, we could find a static $Q_1 = Q_2$ configuration, unlike the case of SCGO.

A final comment about the configurations of figure 2 relates to the work of Chandra *et al* [1]. These configurations have infinitely long 'fold lines', which exist in a typical coplanar configuration. Indeed, fold lines on all scales are needed for the system to explore all the coplanar configurations in the 'spin-liquid' phase. The non-static nature of the magnetic moment in the low-temperature phase is an indicator that the zero-point energy (per site) is indeed of order J [15]. The system therefore 'explores' the configurations that minimize the dipolar interactions ($|1\rangle, |2\rangle$ in figure 2). Even if the system can explore only a limited range of configurations, that do not include $|1\rangle$ or $|2\rangle$, there could still be configurations with a lower dipolar interaction energy that introduce correlations and coherence. The smaller dipolar energy will then give a lower transition temperature T_c.

The overall dipolar interaction energy is given by

$$E_{dd} = \sum_{i \neq j} \frac{\mu_i \cdot \mu_j - 3(\mu_i \cdot \hat{\boldsymbol{r}}_{ij})(\mu_j \cdot \hat{\boldsymbol{r}}_{ij})}{|\boldsymbol{r}_{ij}|^3} \tag{1}$$

where for Cr^{3+} ions in SCGO, $|\mu| = 3.8\ \mu_B$. The sum is performed for each spin i, summing for all $i \neq j$ in the real three-dimensional SCGO lattice. The experimental data indicate almost no frozen (static) magnetic moment [6, 22] at low temperatures, justifying taking the full magnetic moment of the Cr^{3+} ion as taking part in the coherent zero-point oscillations. Our assumption is that the spins have a high zero-point energy of order J, and this high zero-point energy keeps the spins dynamic even at $T = 0$. This means that the expectation value of the static moment is taken to be zero: $\langle \mu_i \rangle = 0$, while the spin–spin correlations are non-zero, given as the dipolar interaction energy $E_{dd} \propto \langle \mu_i \cdot \mu_j \rangle \neq 0$. Let us emphasize that we are using a semi-classical language when we describe the zero-point fluctuations as 'dynamic'.

The dipolar interactions are not strong enough to freeze this zero-point motion ($E_{dd} \ll J$), but cause this motion to be coherent. By coherent oscillations we mean that the spins in the Kagomé planes make synchronized zero-point oscillations between the different (and random) degenerate J-minimizing configurations (figure 1) through the configurations which locally minimize the direct magnetic dipolar interaction energy ($|1\rangle, |2\rangle$ in figure 2). The energy difference between the symmetric and antisymmetric combinations of these configurations

$$|S\rangle = (1/\sqrt{2})(|1\rangle + |2\rangle) \qquad |A\rangle = (1/\sqrt{2})(|1\rangle - |2\rangle) \tag{2}$$

is estimated as $E_0 = 2|E_{dd}| \simeq 8.0$ K, and is just the resonance energy split shown in figure 1, describing the coherent oscillations of the spins between the two equivalent configurations of figure 2. The dipolar energy in the configurations $|1\rangle, |2\rangle$ is $E_{dd} \simeq -4.0$ K. There is a long-range global phase which describes these spin oscillations, i.e. a state of macroscopic quantum resonance at the frequency given by the dipolar interactions (1). Since the intermediate J-minimizing configurations are random, this is a peculiar state of spin-liquid and ordered phase coexistence. The system moves between the random ('spin-liquid') J-minimizing configurations with frequency $\sim J/\hbar$, while oscillating between the dipole minimizing configurations of figure 2 with the much lower frequency $\sim E_0/\hbar$.

Our coherent ground state $|\psi\rangle$, describing the low-temperature ordered phase, is different from the spin-liquid ground state [16] ($|SL\rangle$) of the pure Heisenberg exchange Hamiltonian

(which we do not know explicitly). The difference is that its projection on the two states $|S\rangle, |A\rangle$ evolves as a coherent combination. This is similar to the double-well description of quantum resonance, e.g. of the ammonia molecule [23]. The symmetric and antisymmetric combinations are used as basis and the system performs oscillations between the two equivalent configurations $|S\rangle, |A\rangle$ at frequency $\sim\hbar/E_0$

$$P_{a,s}|\psi(t=0)\rangle = (|S\rangle + |A\rangle) \Rightarrow P_{a,s}|\psi(t)\rangle = (|S\rangle + |A\rangle e^{-iE_0 t/\hbar}) \tag{3}$$

where $P_{a,s}$ is the projection operator on the two states $|S\rangle, |A\rangle$. The time dependence here is in the semi-classical sense, describing the broken gauge symmetry of choosing a particular realization of the relative phase between the spins. This proposed state (3) has a lower average energy compared with the spin-liquid state, since it minimizes the dipolar energy more than random coplanar configurations do, where $P_{a,s}|SL\rangle = 0$. This 'spin-liquid' state is assumed to have no long-range correlations, temporal or spatial, so its projection on any particular state with long-range correlations (such as $|S\rangle$ or $|A\rangle$) between the spins is small.

Experimental evidence for this proposed arrangement of the zero-point oscillating spins may be indicated by a diffuse elastic-Bragg peak detected in neutron scattering [6]. The measurement indicates a lack of well defined long-range spatial order. In our model the spins zero-point oscillate in phase over the entire lattice, but the intermediate J-minimizing states through which the spins oscillate are disordered. The peak position at $\sim$1.4 Å^{-1} corresponds to a periodicity of $\sim$4.5 Å, which is the size of approximately two triangles in the Kagomé plane, and agrees with the periodicity of the dipolar arrangement we propose in figure 2(a).

An immediate result of our model is that the energy scale of the low-temperature phase is determined by the long-range (dipolar) interactions (1). This means that the absence of any measured critical behaviour at the Kagomé percolation concentration [24] $p_{percol} = 0.6527$ of the Cr atoms follows naturally. A phase transition driven by the nearest-neighbour exchange interactions would have been sensitive to the percolation transition. The linear dependence of the transition temperature [4] on the Cr concentration p follows naturally from the $1/r^3$ summation in (1). It was previously noted [25] that the independence of the qualitative properties of the low-temperature phase from the dilution p may indicate the occurrence of long-range interactions.

3. Spin-waves

The state we described above is of a quantum resonance state between the dipolar configurations of figure 2. We now want to introduce an excitation whereby the relative phase between the dipoles is spatially modulated. This means that the projection onto the configurations (2) of figure 2 is modulated by a factor $\exp -i2\pi \boldsymbol{k}\cdot\boldsymbol{r}$. Such a modulation is just an extra phase $2\pi\boldsymbol{k}\cdot\boldsymbol{r}$ in the relative phase between the components of $|\psi(t)\rangle$ in (3,1)

$$E(\boldsymbol{k}) = -\sum_{i\neq 0} \mu_0 \cdot \mu_i \left[\frac{3\cos^2(\hat{\mu}_0 \cdot (\boldsymbol{r}_0 - \boldsymbol{r}_i)) - 1}{|\boldsymbol{r}_0 - \boldsymbol{r}_i|^3}\right] \exp[2\pi i \boldsymbol{k}\cdot(\boldsymbol{r}_0 - \boldsymbol{r}_i)] \tag{4}$$

where $\boldsymbol{k}$ is the wavevector of the modulation. These collective modes are restricted to the Kagomé planes by the restriction of the nearest-neighbour exchange interaction to remain in the domain of coplanar configurations with 120° between spins on each triangle.

The resulting excitation spectrum (4) $E(k) - E(0)$ is shown in figure 3, compared with the inferred spin-wave velocity from specific-heat measurements [2, 26]. We find that the calculated dispersion is indeed linear at long wavelengths, and agrees reasonably well with the experimentally inferred spin-wave velocity, $\sim$80 m s^{-1}.

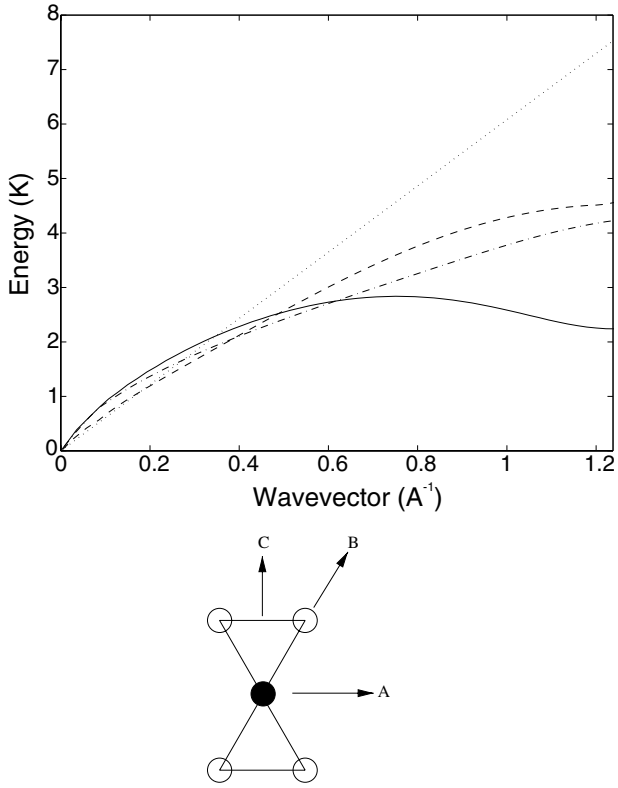

Figure 3. The calculated spectrum (equation (4)) of the spin waves in the Kagomé planes, for dilution $p = 0.89$, where $E_0 \simeq 7$ K. The linear spin-wave spectrum inferred from the $C_v \propto T^2$ dependence is shown by the dotted line. The calculated spin-waves are for modulations along the vectors shown below: A—solid curve, B—long-dashed curve, C—dash–dotted curve.

We also note that in the experiments there is a peak in C_v corresponding to a localized excitation of energy $\sim$10 K. This energy is quite close to the dipole resonance energy $E_0 \sim 7$ K (at $p = 0.89$). Our simple model should be viewed as quantitative only for the long-wavelength limit of the excitations. At higher energies a theory that describes the interplay of dipolar and exchange interactions [11], should apply. A full treatment of the problem of coupled exchange and dipolar interactions should take into account magnetic excitations of relatively low energy [16] ($\sim J/20$) that hybridize with the dipolar excitations.

4. Spin correlations and experimental probes

We shall now describe some of the responses of the low-temperature phase of SCGO to external probes, as follows from our proposed model.

In SCGO there is a marked difference between ZFC and FC static magnetic susceptibility [2, 3], similar to a static spin-glass. The measured cusp in the ZFC magnetic susceptibility defines the transition temperature T_c.

In the FC case we have a magnetic field (along the z-direction) which tends to align a component of the spins with the field, against the dipolar interactions that tend to keep the spins in the Kagomé planes. The states $|1\rangle$, $|2\rangle$ are therefore modified so that the spins are slightly out of the plane, in the direction of the external magnetic field. We therefore expect

to find a magnetic response which is similar to that of a normal AFM in a perpendicular external magnetic field, which is non-zero and almost constant with temperature [27]: $\chi_{FC} = \chi_{\perp} \sim 1/E_0 \neq 0$. The smallness of the dipolar interactions (E_0) accounts for the relatively large low-temperature FC susceptibility.

In the ZFC state the coherent order is established and in the limit of small applied fields, i.e. weak compared with the internal magnetic fields, the coherent oscillations in the planes (3) are much faster than the Larmor frequency μH. In this limit the individual oscillating spins are on average in the symmetric state $|S\rangle$ (2) in the xy-planes, which correspond to $S_z = \pm 1$. Such a state will respond as an AFM in parallel to the external field [27]: $\chi_{ZFC}(T = 0) = \chi_{\parallel} \to 0$ for $T \to 0$. This explains the vanishing susceptibility of the ZFC in small applied fields as $T \to 0$. The magnitude of the internal magnetic fields can be estimated from (1) as $H_0 \simeq E_{dd}/3.8\ \mu_B \simeq 10$ kG. Only for external fields approaching the size of these internal fields does the observed ZFC response approach the FC response [24, 26, 28]. Experiments on single crystals [18] have found that the susceptibility parallel to the Kagomé planes has a weaker ZFC versus FC difference. This is in accordance with our model where the dipolar coherence introduces an anisotropy with respect to the Kagomé planes.

Finally, muon spin-relaxation (μSR) experiments [24, 28] probe the spatial and temporal correlations of the spins. These experiments show that the low-temperature relaxation rate of polarized muons is finite and temperature independent in the low-temperature phase, which means that the magnetic spins are fluctuating even at $T \to 0$.

The wavefunction (3) describes a coherent state with ODLRO of the oscillating spins. This is a system with broken global gauge symmetry, in the form of a global phase of the zero-point oscillating spins (a complex order parameter). These long-range temporal correlations appear in the time dependence of the μSR which changes from exponential above the transition temperature to Gaussian [24] as $T \to 0$. Gaussian decay of the polarization arises in cases of long-range (time-independent) temporal correlations between the spins [29], as occurs here. The coherent zero-point oscillations of the spins will produce a zero average static magnetic moment, so there will therefore be no oscillating signal in the muon polarization decay as arises in cases of static magnetic order [29]. The muon spins that will be excited to oscillate in phase with the coherent oscillations of the Kagomé spins (with frequency $E_0/\hbar$) will see a constant magnetic field, with a resulting Gaussian time decay of the muon polarization [30]. The rate at which muons will follow the coherently oscillating Kagomé spins is given by second-order time-dependent perturbation theory [23]. It is equal to the probability per unit time of exciting the muon by the resonant oscillations of the lattice spins

$$P(t) \propto \frac{|W|^2}{\hbar^2 \omega} \tag{5}$$

where the matrix element coupling the muons to the local magnetic fields is $|W| \simeq \gamma_\mu \Delta B_\mu$, where ΔB_μ is the rms deviation of the internal magnetic field at the muon site and γ_μ is the muon magnetic moment. Since this rms field is due to the coherent zero-point oscillations of the Kagomé spins, it is of order H_0. The frequency $\omega = E_0/\hbar$ is that of the zero-point oscillations of the lattice spins, so the resulting rate of muon depolarization is given by

$$\lambda = \frac{|\gamma_\mu \Delta B_\mu|^2}{E_0} \simeq 20\ \text{MHz} \tag{6}$$

in excellent agreement with experimental results [24]. This rate of muon depolarization should apply as long as the probability is much less than unity, i.e. for $t \ll 1/\lambda \simeq 0.1\ \mu$s. Indeed at longer times there is a deviation from the Gaussian depolarization curve [24].

The coherent zero-point magnetic fields are linear with the dipolar energy $H_0 \propto E_0$, therefore linearly proportional to the dilution p. The decay rate is therefore linear in the

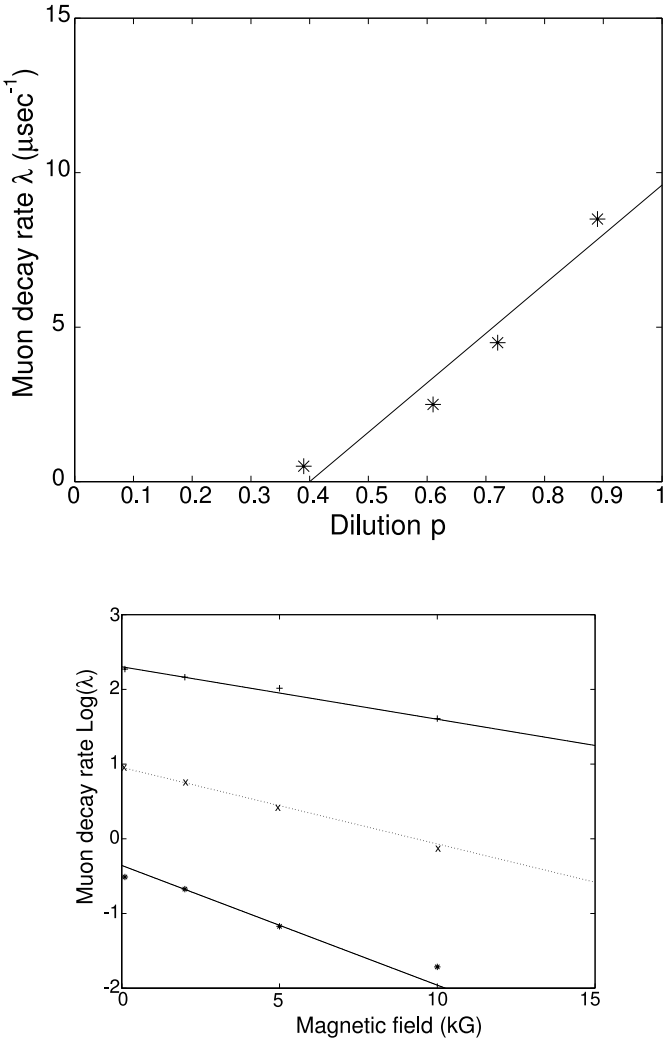

Figure 4. (a) The rate of muon depolarization in the Gaussian regime $\lambda \propto (p - p_c)$ (solid line), as a function of the dilution p for $H = 100$ G. (b) The rate of muon depolarization as a function of the magnetic field H for different dilutions (equation (7) straight lines). Experimental data (symbols) from [24].

dilution p (figure 4(a)). The linear dependence of the resulting decay rate λ on the dilution p is roughly supported by the experimental results [24], if we note that the zero of the coherent oscillations is shifted to a finite critical dilution [4], i.e. $\lambda \propto (p-p_c)$ with $p_c \simeq 0.4$ (figure 4(a)). For the dilution dependence of the transition temperature this critical dilution is $p_c \simeq 0.2$, which means that below this critical dilution the dipolar interaction between the Cr^{3+} ions is weaker than other types of interaction that destroy the coherent dipolar phase. For the muon depolarization p_c is larger, due to the weaker gyromagnetic moment of the muon which means that below this dilution the dipolar interactions of the Cr^{3+} ions and the muon are masked by other weak interactions.

As a function of the magnetic field there is a measured reduction [24] in λ (figure 4(b)). The decrease fits an activated behaviour with the external magnetic field

$$\lambda(H, p) \propto \exp(-H/\bar{H}_0(p)) \tag{7}$$

where from the experiment $\bar{H}_0 \simeq 16\,\mathrm{kG} \sim 2H_0$, and from the discussion above we expect a linear dependence on the dilution: $\bar{H}_0(p) \simeq \bar{H}_0 \cdot p$ (figure 4(b)). This is indeed observed.

Above the transition temperature the spin fluctuations are uncorrelated, resulting in an exponential decay of the muon polarization.

5. Conclusion

We conclude that a model where the zero-point oscillations of the Kagomé spins are correlated by the magnetic dipolar interactions describes some of the important features of the low-temperature phase of SCGO. The main property which is described by our proposed model is the occurrence of a coherent state of the oscillating spins which therefore remain dynamic (quantum resonance) while having a long-range order of the global phase. The other properties follow from the behaviour of the dipolar interaction energy, which scales linearly with the Cr concentration and is therefore insensitive to the percolation density.

We wish to comment that recent work [31] on $Ba_2Sn_2Ga_3ZnCr_7O_{22}$ (BSGZCO) shows similar behaviour to SCGO. The lower temperature of the susceptibility cusp and specific-heat peak in BSGZCO compared with SCGO (the ratio is $\sim 2/3$) can be partially attributed to the slightly larger dimensions of the unit cell. The dipolar energy ratio is given by $(a_{SCGO}/a_{BSGZCO})^3 \sim 0.87$, and correspondingly the transition temperature and the spin-wave velocity.

Our model does not present a complete solution to the low-temperature phase of this material. In particular, there should be a coupling of the dipolar excitations with the low-energy magnetic and non-magnetic excitations of the exchange interactions [16]. We nevertheless hope to have shown that the dipolar interactions can naturally explain some essential properties of this phase, and should therefore play an important role in a any future treatment [11].

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