Journal of Physics: Conference Series **663** (2015) 012010 doi:10.1088/1742-6596/663/1/012010

Low–energy spin–wave excitations in amplitude–modulated magnetic structure of $PrNi_2Si_2$

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Abstract. Inelastic neutron scattering (INS) experiments and random phase approximation calculations have been used to investigate the low–energy spin–wave excitations in $PrNi_2Si_2$. The modulated magnitude of the ordered magnetic moments of Pr^{3+} ions implies that the associate, longitudinally polarized magnetic excitations are more intense and dispersive than the usual transverse spin waves. Within the random phase approximation the results are in good overall agreement with the predictions made by the model determined previously from the paramagnetic excitations. The most unusual observation is the well–defined amplitude mode detected close to the magnetic Bragg point existing simultaneously with the phason mode. At low energies, an extra mode is observed to hybridize with the magnetic phasons in the neighborhood of the magnetic Brillouin zone center. A magnetoelastic interaction between the magnetic excitations and the longitudinal phonons is able to explain part of the disturbances, but it is concluded that the extra mode must be of some other, unknown origin.

1. Introduction

In incommensurate magnetic structures, the only low–energy mode that is expected to be is the pseudo–Goldstone mode of broken translations, known as a phason [1]. This excitation is a purely longitudinal mode associated with the phase of the complex order parameter and represent a modulation of the magnetic moment only. This phason mode is quite unusual, where typically the low–energy collective modes are transverse spin–waves, which correspond to small rotations of the magnetic moments away from their ordered axes. In the spin–wave theory, longitudinal modes are often expected to be highly damped, and then hard to be observed [2]. For a recent notable exception to this rule is the case of amplitude–modulated (AM) magnetic structure of $PrNi_2Si_2$ [3]. It presents a peculiar magnetic character: it is one of the few examples [4, 5, 6, 7, 8, 9, 10] in nature where the magnetic ions, Pr^{3+}, have a longitudinal AM magnetic structure below $T_N = 20$ K, which is stable down to 0 K. In this longitudinally polarized phase, the length of the magnetic moments is modulated and the magnitude of the exchange field changes from site to site. In this way, the material is characterized by an Ising–type magnetic structure, where the magnetocrystalline anisotropy confines the magnetic moments along the c–axis. In most "equal-moment" systems the moments saturate at low temperatures, in which case the longitudinal excitations are quenched, allowing only the presence of transversely polarized spin waves. The AM magnetic structure of $PrNi_2Si_2$ at low temperature (4 K) is well described by a single wave vector $\vec{k} = (0, 0, k_z)$ with $k_z = 0.87$ (in r.l.u.) such that the magnetic moment at the site l is given by the expression $< M_l^z >= \cos(2\pi z k_z)$, where z is the coordinate of the Pr^{3+} ions [11]. The observation of a third harmonic at even lower temperatures reflects a weak tendency to squaring up of the AM moment, but the observed moment ratio between the third and first harmonics is far from the limit of perfect squaring [11, 12]. The dispersion and intensities of both longitudinal and transverse excitations in $PrNi_2Si_2$ were measured along the high–symmetry directions below T_N in several INS experiments [3, 13, 14, 15, 16, 17]. Information about both the experimental details and all the longitudinal and transverse excitations observed in $PrNi_2Si_2$ can be found elsewhere [3]. In this contribution, we now present the results of the dispersive lowest–energy excitations and a discussion of the possible interpretation of this dispersion considering a coupling to the longitudinal phonons.

2. Results and discussion

The excitations observed in the ordered phase relate in a direct way to the excitations in the paramagnetic phase via the level diagram shown in Figure 1 [18]. The *cc* polarized paramagnetic excitations are the cooperative excitations deriving from the transition between the $\Gamma_1^{(1)}$ and Γ_2 states at zero exchange field, and the longitudinal magnetic excitations in the ordered phase derive from the same transition at non-zero exchange fields. Similarly, the $\Gamma_1^{(1)}$–$\Gamma_5^{(1)}$ excitations in the paramagnetic phase become the transverse spin waves in the ordered phase. In a comparison with the paramagnetic phase, the behavior of the excitations in the ordered phase is more complicated because the translational symmetry is reduced, but, on the other hand, they are more well defined since the temperature is lower.

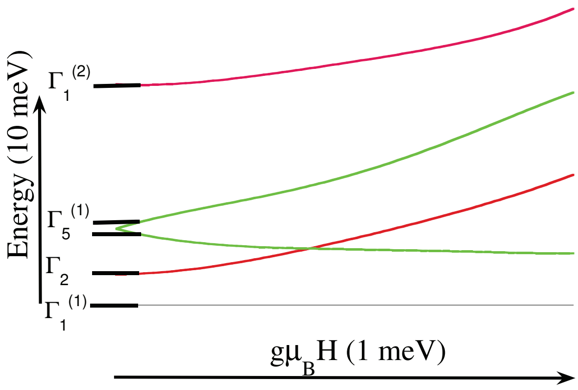

Figure 1. (Color online) The mean field low–lying excitation energies for the Pr^{3+} ions as functions of an exchange field ($g\mu_B H$) along the z axis, calculated with the use of the crystal field parameters given by reference [16]. The classification of the levels is the one applying at zero field.

The INS cross sections measured in the ordered phase of $PrNi_2Si_2$ have been analyzed in terms of the CF and exchange parameters determined from the paramagnetic properties of the system. The previous analysis and the present one are both based on the random phase approximation (see Figure 2). By involving interactions between neighbors 8 layers apart along the c axis, the RKKY exchange interaction in $PrNi_2Si_2$ is long ranged [13]. This indicates that the RPA should be trustworthy, in particular in the zero temperature limit where line–width effects are unimportant. The agreement between experiments and theory is quite good at energies above the range of the phason mode above 2 meV (not shown, see reference [3]). The analysis of the paramagnetic excitations leaves some room for modifications of the exchange parameters and utilizing the remaining degrees of freedom, it might be possible to improve the comparison

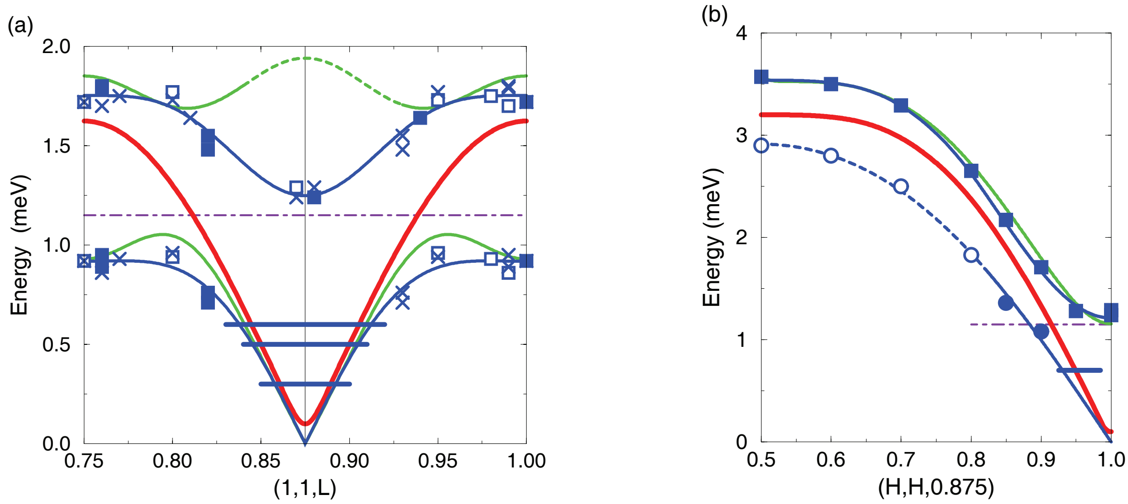

Figure 2. (Color online) The dispersion at low energies of the magnetic excitations in $PrNi_2Si_2$ at $T = 1.6$ K for the wave vectors (a) $(1, 1, L)$ and (b) $(H, H, 0.875)$. The different blue symbols denote the experimental results. The solid blue squares are the constant Q–scan results obtained at the wave vectors shown, the open blue squares in (a) denote results obtained in the neighboring magnetic Brillouin zones shifted to the present one, and the blue crosses show all the experimental results when being reflected with respect to the zone center at $(1, 1, 0.875)$. The circles in (b) denote low intensity peaks, and the open ones indicate peaks that are so weak that their presence may be questioned. In both figures the horizontal line segments indicate results obtained in constant–E scans, and the solid blue lines are least squares Fourier fits to (lines connecting) the experimental points. The solid red lines show the calculated energies of the phason mode. The green lines are the final excitation energies when the longitudinal phonons and the longitudinal magnetic excitations interact according to the magnetoelastic model. The horizontal dot–dashed violet lines indicate the position of a possible flat level interacting with the phasons (see text).

in some of the cases. The most noticeable discrepancy is that the RPA model predicts the energy dispersion of the phason mode along $(H, H, 0.875)$ at 1.6 K to be about 10% smaller than observed [see Figure 2(b)]. This defiance may possibly be removed by a modification of the exchange parameters, but we have not tried to do that, because it might as well be related to the interaction occurring between the phasons and the extra mode at about 1.2 meV.

The major problem in the analysis of the experimental excitation spectrum in $PrNi_2Si_2$ is the clear observation that the phason mode is interacting with an extra level lying at an energy of about 1.2 meV (see Figure 2). It can be excluded that this level should originate from a $4f$ orbital of the Pr^{3+} ions. One possible candidate is the longitudinal acoustic phonons transferred by the modulation of the ordered moments to the relevant range in wave vectors and energies. Using a magnetoelastic model we are able to explain some of the hybridization phenomena, [3])but it failed in an essential way to predict the right behavior in the close neighborhood of the magnetic Brillouin zone center. The possibility that low–symmetry interactions should have increased the effective period for the longitudinal excitations by a factor of two, and hence introduced an extra low lying amplitude mode, can also be dismissed. The RPA might be wrong in placing the principal amplitude mode at energies about 3 meV instead of 2 meV. The RPA also predicts that the intensity of the amplitude mode should be small relatively to the phason mode at all wave vectors. Since we expect that, at least, this more qualitative prediction should be correct, the large intensity of the upper mode compared to the lower one at the zone boundaries in Fig. 2(a) disqualifies this possibility. The well–defined systematic hybridization behavior observed in the experiments seems to exclude that the extra level should be an experimental artefact. Is it possible that this extra level is established by the electrons on the Ni ions or by the conduction electrons? We do not think so, since, at the low energies in question, these outer electrons are expected just to constitute a passive medium for establishing the RKKY interaction. The only remaining possibility is, as far as we can figure out, that the level is caused by "impurities". The most likely origin of a well–defined impurity level would be that a minor percentage of the Pr ions are displaced from their right position in the crystal. More experimental investigation of the properties of the phonons and the significance of the crystal quality have to be done in order to clarify the cause for the extra level.

3. Conclusions

In summary, we have shown that $PrNi_2Si_2$ is an ideal system for studying the behavior of the magnetic excitations in an amplitude modulated magnetic structure. The most unusual observation being the well–defined amplitude mode observed close to the magnetic Bragg point existing simultaneously with the phason mode. All of them are well accounted for in an RPA model, except for the mysterious hybridization of the phasons with an extra level of unknown origin. This conclusion compares in many ways with the one made in the case of another system with Pr^{3+} ions placed in metallic surroundings, namely that of elemental Pr metal [19, 20].

Acknowledgments

Financial support has been received from Spanish MEC and FEDER Grant No. MAT2011-27573-C04-02 and FC-15-GRUPIN14-037 (Asturias Government, Spain).

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