odd interactions in quantum magnets and liquids





Sasha Chernyshev













- I. interactions of magnons
- II. triangular AF, $S(q, \omega)$
- III. ⁴He, roton
- IV. XY magnets, lifetime







Mike Zhitomirsky

Sasha Chernyshev





magnons ≈ bosons, why?

- spins commute on different sites
- effective Weiss field for each spin
- local raising (lowering) operators







what would bosons do?



• exactly the same for a ferromagnet

• GS and excitations are e-states of $S_{
m tot}^z$

• **GS**
$$S_{\text{tot}}^{z} |GS\rangle = SN |GS\rangle$$

• magnon
$$\Rightarrow \Delta S^z = -1$$

magnon # preserved

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what would bosons do in an AF?



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formally ...

V. G. Bar'yakhtar, V. L. Sobolev, and A. 1 FEBRUARY 1971 VOLUME 3, NUMBER 3 PHYSICAL REVIEW B G. Kvirikadze, ZhETP 65, 790 (1973); Dynamics of an Antiferromagnet at Low Temperatures: Spin-Wave S. M. Rezende and R. M. White, PRB Damping and Hydrodynamics* **14**, 2939 (1976); **18**, 2346 (1978); A. B. Harris and D. Kumar Yu. A. Kosevich and A. V. Chubukov, Department of Physics, University of Pennsylvania, Philadelphia, Pennsylvania 19104 Sov. Phys. JETP 64, 654 (1986)]; and HKHH, 71 S. Tyc and B. I. Halperin, PRB 42, B. I. Halperin and P. C. Hohenberg 2096 (1990); P. Kopietz, PRB **41**, 9228 (1990). $\hat{\mathcal{H}}_0 = \sum_{\mathbf{k}} \varepsilon_{\mathbf{k}} b_{\mathbf{k}}^{\dagger} b_{\mathbf{k}} + \frac{1}{4} \sum_{\mathbf{k}_i} V_{\mathbf{k}_1, \mathbf{k}_2; \mathbf{k}_3, \mathbf{k}_4}^{(1)} b_{\mathbf{k}_1}^{\dagger} b_{\mathbf{k}_2}^{\dagger} b_{\mathbf{k}_3} b_{\mathbf{k}_4}$ $+\frac{1}{3!}\sum_{\mathbf{k}}V^{(2)}_{\mathbf{k}_{1},\mathbf{k}_{2};\mathbf{k}_{3},\mathbf{k}_{4}}\,b^{\dagger}_{\mathbf{k}_{1}}b^{\dagger}_{\mathbf{k}_{2}}b^{\dagger}_{\mathbf{k}_{3}}b_{\mathbf{k}_{4}}$ $+\frac{1}{4!}\sum V^{(3)}_{\mathbf{k}_{1},\mathbf{k}_{2};\mathbf{k}_{3},\mathbf{k}_{4}}b^{\dagger}_{\mathbf{k}_{1}}b^{\dagger}_{\mathbf{k}_{2}}b^{\dagger}_{\mathbf{k}_{3}}b^{\dagger}_{\mathbf{k}_{4}}$

- are they valid?
- is this all?

what else would bosons do?

• condense!
$$\langle b_{\mathbf{k}=0} \rangle \neq 0$$

- # of bosons **not** preserved
- Bogolyubov substitution

$$b_{\mathbf{k}_1}^{\dagger} b_{\mathbf{k}_2}^{\dagger} b_{\mathbf{k}_3} b_{\mathbf{k}_4} \Rightarrow b_{\mathbf{k}_1}^{\dagger} b_{\mathbf{k}_2}^{\dagger} b_{\mathbf{k}_3} \langle b_0 \rangle$$

• cubic anharmonicities

$$\mathcal{H} = \dots + \frac{1}{2!} \sum_{\mathbf{k}_{i}} V_{\mathbf{k}_{1},\mathbf{k}_{2},\mathbf{k}_{3}}^{(3,1)} b_{\mathbf{k}_{1}}^{\dagger} b_{\mathbf{k}_{2}}^{\dagger} b_{\mathbf{k}_{3}} + \text{H.c.} \rightarrow \mathbf{H.c.}$$
$$+ \frac{1}{3!} \sum_{\mathbf{k}_{i}} V_{\mathbf{k}_{1},\mathbf{k}_{2},\mathbf{k}_{3}}^{(3,2)} b_{\mathbf{k}_{1}}^{\dagger} b_{\mathbf{k}_{2}}^{\dagger} b_{\mathbf{k}_{3}}^{\dagger} + \text{H.c.} \rightarrow \mathbf{H.c.}$$

- corresponds to a non-collinear antiferromagnet
 - GS: spin rotational symmetry broken completely



BEC antiferromagnets

T. Matsubara and H. Matsuda, Prog. Theor. Phys. 16, 569 (1956);E. G. Batyev and L. S. Braginskii, Sov. Phys. JETP 60, 781 (1984).

BEC: U(1) symmetry breaking, order parameter

$$\mathbf{\Delta} = |\Delta| e^{i\varphi}$$





collinear vs non-collinear

frustration (competing interactions) \rightarrow **non-collinearity** \rightarrow transverse-longitudinal coupling \rightarrow 3-boson terms

> transverse (T) "one-magnon" $S^- \approx \sqrt{2S} a^{\dagger}$



longitudinal (L) "two-magnon" $S^z = S - a^{\dagger}a$

strong T-L



remaining U(1) symmetry forbids $S^z S^{\pm}$ terms

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two classes of (quantum) [AF]magnets

collinear (quartic anharmonicity)



no coupling with 2-magnon continuum

weakly-interacting excitations

 \downarrow

small renormalizations even in low D and S=1/2



non-collinear (qubic anharmonicity)



direct coupling 1-to-2-magnon states $\downarrow \downarrow$ strong(er)-interacting excitations $\downarrow \downarrow$

large spectrum renormalizations in low D and S=1/2





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most important qualitative difference

magnons in **non-collinear*** AFs:

• often decay even at T=0 (yield broad peaks)



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decays

frustration (competing interactions) \rightarrow **non-collinearity** \rightarrow transverse-longitudinal coupling \rightarrow 3-boson terms \rightarrow decays

$$\mathcal{H} = \dots + \frac{1}{2!} \sum_{\mathbf{k}_i} V^{(3,1)}_{\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3} b^{\dagger}_{\mathbf{k}_1} b^{\dagger}_{\mathbf{k}_2} b_{\mathbf{k}_3} + \dots$$

- three-boson terms are necessary for (1-in-2) decays
- "kinematic" conditions (*E* and *k* conservations) make it sufficient
- same conditions that favor cubic terms favor decay kinematics

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in contrast to ...

PHYSICAL REVIEW B

VOLUME 3, NUMBER 3

1 FEBRUARY 1971

Dynamics of an Antiferromagnet at Low Temperatures: Spin-Wave Damping and Hydrodynamics*

A. B. Harris and D. Kumar Department of Physics, University of Pennsylvania, Philadelphia, Pennsylvania 19104

and

B. I. Halperin and P. C. Hohenberg Bell Telephone Laboratories, Murray Hill, New Jersey 07974 (Received 14 July 1970)



Regime A: $\epsilon_{\vec{k}} \ll \tau^3 \ll 1$, $\Gamma_{\mathbf{i}} = \hbar^{-1} \Sigma''(\mathbf{k}, \omega_{\mathbf{r}} \epsilon_{\mathbf{i}})$ $= (2\omega_{E}/S^{2})\epsilon_{V}^{2}\tau^{3}(2\pi)^{-3}(a|\ln\tau| + a');$ (9.1a)Regime B: $\tau^3 \ll \epsilon_r \ll \tau \ll 1$. $\Gamma_{\vec{k}} = (8\omega_E/3S^2) \epsilon_{\vec{k}}^2 \tau^3 (2\pi)^{-3} [b \ln(\tau/k) + b'];$ (9.1b)Regime C: $\tau \ll \epsilon_{\vec{v}} \ll \tau^{1/3} \ll 1$, $\Gamma_{\vec{k}} = (\pi \omega_E / 108S^2) \epsilon_{\vec{k}} \tau^4 ;$ (9.1c)Regime D: $\tau^{1/3} \ll \epsilon_z \ll 1$. $\Gamma_{\vec{k}} = (\omega_E/2S^2\pi^3) \tau^5 \zeta(5) [g(\hat{k}) \epsilon_{\vec{k}}^2]^{-1}$. (9.1d)





- in **collinear** AFs 1-to-3 decay conditions are uncommon
- even if --- their effect is weak

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common?



 for a spiral-like state (more than one type of Goldstone mode) decay conditions are always fulfilled for a (large) range of k



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part II.









Martin Mourigal

Wesley Fuhrman



Mike Zhitomirsky



Sasha Chernyshev



$S(q, \omega)$, triangular lattice AF

- why? previous attempts* not intuitive ...
 ... and contain too many terms ...
- show effects of decays in S(q, ω)

$$\overline{\mathcal{S}^{\alpha_0\beta_0}(\mathbf{q},\omega)} = \int_{-\infty}^{\infty} \frac{dt}{2\pi} e^{i\omega t} \left\langle S^{\alpha_0}_{\mathbf{q}}(t) S^{\beta_0}_{-\mathbf{q}}(0) \right\rangle$$



 $\left[\mathcal{S}^{\text{tot}}(\mathbf{q},\omega) = \boldsymbol{\alpha} \, \mathcal{S}^{x_0 x_0}(\mathbf{q},\omega) + \boldsymbol{\beta} \, \mathcal{S}^{y_0 y_0}(\mathbf{q},\omega) + \gamma \, \mathcal{S}^{z_0 z_0}(\mathbf{q},\omega)\right]$



S(**q**,ω), II

• transverse (1-magnon) components

$$\begin{aligned} \mathcal{S}^{xx}(\mathbf{q},\omega) &= \frac{S}{2}\Lambda_{+}^{2}(u_{\mathbf{q}}+v_{\mathbf{q}})^{2}A_{11}(\mathbf{q},\omega) \\ \mathcal{S}^{yy}(\mathbf{q},\omega) &= \frac{S}{2}\Lambda_{-}^{2}(u_{\mathbf{q}}-v_{\mathbf{q}})^{2}A_{11}(\mathbf{q},\omega) \end{aligned}$$

• longitudinal (2-magnon) part (broad in ω)

$$\mathcal{S}^{zz}(\mathbf{q},\omega) = \frac{1}{2} \sum_{\mathbf{k}} (u_{\mathbf{k}} v_{\mathbf{k}-\mathbf{q}} + v_{\mathbf{k}} u_{\mathbf{k}-\mathbf{q}})^2 \delta(\omega - \varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}-\mathbf{q}})$$



 $A(q, \omega), S=1/2$







 $A(q, \omega), S=3/2$







S(q,ω), S=3/2







S(q,ω), S=1/2





complicated features persist



Μ $S(q, \omega), S=1/2, 3/2$ Y₁



• roles of transverse and longitudinal components

Nordíta, *NDFCM*, M. Mourigal, W. Fuhrman, **SC** and **MZ**, PRB **88**, 094407 (2013)



K'

Μ

$S(q, \omega)$, constant energy scans







$S(q, \omega)$, constant energy scans





experiments

Magnon breakdown in a two dimensional triangular lattice Heisenberg antiferromagnet of multiferroic LuMnO₃

Joosung Oh,^{1,2} Manh Duc Le,^{1,2} Jaehong Jeong,^{1,2} Jung-hyun Lee,^{3,4} Hyungje Woo,^{5,6} Wan-Young Song,^{3,4} T. G. Perring,⁵ W. J. L. Buyers,⁷ S-W. Cheong,⁸ and Je-Geun Park^{1,2,3,*}



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PRL **111**, 257202 (2013)



spin-gap systems, triplet decays



piperazinium hexachlorodicuprate (PHCC)

A. Kolezhuk and S. Sachdev, Phys. Rev. Lett. **96**, 087203 (2006), M. E. Zhitomirsky, Phys. Rev. B **73**, 100404 (2006)

order not necessary to have decays

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MZ and SC, RMP 85, 219 (2013)



T. A. Masuda, etal., PRL 96, 047210 (2006)

IPA-CuCl₃



other systems/experiments: BEC





conclusions

- ✓ decays in magnon spectra of non-collinear magnets are generic
- ☑ enhanced by lower D and small spin
- non-Lorenzian features in $S(\mathbf{k}, \omega)$ and decay regions can be used for fingerprinting

- \blacksquare finite-*T*?
- ✓ more experiments ?



parts III. and IV.



neutron-scattering spin-echo



90° 180° echo

The Neutron Spin-Echo Technique at Full Strength

Joël Mesot





Neutron Resonant Spin Echo



Coil displacement (mm)

ILL, IN22, and ZETA





Thermal neutron three-axis spectrometer with polarization analysis IN22

IN22 is a three-axis spectrometer (CRG) equipped for full polarization analysis. The option CRYOPAD and a 15 Tesla cryomagnet are optimised for inelastic scattering. The option ZETA provides neutron resonance spin echo (NRSE).



CRG three-axis spectrometer IN22 with the ZETA resonant neutron spin-echo setup



III. odd interactions in superfluid ⁴He



MAX.PLANCK.CESELISCHAP









experiments: Björn Fåk (CEA, ILL) Thomas Keller (Munich, Stuttgart)

Nordíta, NDFCM, 7-16-14

theory: Mike Zhitomirsky (CEA) Sasha Chernyshev (UC Irvine)

arXiv:1206.1498; PRL **109**, 155305 (2012)

spectrum of ⁴He



K. H. Andersen *et al.*, J. Phys. Cond. Mat. **6**, 821 (1994).

- *T*-dependence of roton's:
 - lifetime
 - energy

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(long) history ...

- L. D. Landau and I. M. Khalatnikov, *The theory of the viscosity* of helium II: I. Collisions of elementary excitations in helium II, Zh. Eksp. Teor. Fiz. **19**, 637- 650 (1949).
- roton lifetime (linewidth)

$$\Gamma(T) \propto N_r(T) \propto \sqrt{T} e^{-\frac{\Delta(T)}{T}}$$

- K. Bedell, D. Pines, and A. Zawadowskii, PRB 29, 102 (1984).
- prefactors + energy shift

$$\delta(T) = \Delta(T) - \Delta_0 \propto N_r(T) \propto \sqrt{T} \, e^{-\frac{\Delta(T)}{T}}$$

* **<u>no</u>** three-boson interaction needed/directly involved ** Hartree-term gives the same for $\delta(T)$







neutron-scattering spin-echo, (>1.2K)

VOLUME 44, NUMBER 24

PHYSICAL REVIEW LETTERS

16 June 1980



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is this the end of the story?



neutron-scattering spin-echo, (>0.88K)

VOLUME 77, NUMBER 19

PHYSICAL REVIEW LETTERS

4 NOVEMBER 1996

High-Resolution Measurements of Rotons in ⁴He





the $T \rightarrow 0$ limit.



phonons?



- where are the phonons ?
- roton-*phonon* scattering:





BF, TK, MZ, **SC**, PRL **109**, 155305 (2012).

* (= r-r at T=0.5K, where both $\sim 10^{-6}$ K)

neutron-scattering spin-echo, (>0.5K)







phonons to the rescue

• **more** roton-*phonon* scatterings:



• three-particle (decay, coalescence) are necessarily positive





theory vs experiment





- theory:
 - quantitatively (!) explains positive shift at low T
 - provides reasonable fit
 - three-particle interactions dominate the new effect



conclusions/outlook

- \square clear sign of the odd (3-particle) interactions in the roton energy shift
- ☑ LK ++ [LK-ZC (?)] theory is formulated
- $\ensuremath{\boxtimes}$ neutron spin-echo allows to reach new regimes

- even lower T?
- phonon-phonon scattering, pressure dependence, etc.



IV. lifetime of gapped excitations in (collinear) antiferromagnets









theory: Sasha Chernyshev (UC Irvine) Mike Zhitomirsky (CEA)

Nordíta, NDFCM, 7-16-14

EUTRON POR SCIENCE Institut Laue-La

experiments: Louis-Pierre Regnault, Nicolas Martin (CEA, ILL)

arXiv: 1206.4690; PRL **109**, 097201 (2012)

material

L. P. REGNAULT AND J. ROSSAT-MIGNOD PHASE TRANSITIONS IN QUASI TWO-DIMENSIONAL PLANAR MAGNETS

L. J. De Jongh (Ed.), Magnetic Properties of Layered Transition Metal Compounds 271–321. © 1990 Kluwer Academic Publishers. Printed in the Netherlands.





 $BaNi_2(PO_4)_2$

lifetime/linewidth/damping/decay rate



- lifetime⁻¹ = linewidth in "simple" AFs
- spin waves: scattering on?
 - impurities
 - themselves
 - combination of the two (impurity-assisted)





where from?

- by the nature of spin-to-bosons mapping:
- \Rightarrow magnon-magnon scattering $J\mathbf{S}\cdot\mathbf{S}, D(S^z)^2 \Rightarrow \varepsilon_{\mathbf{k}}b^{\dagger}_{\mathbf{k}}b_{\mathbf{k}} + V^{\mathrm{m-m}}_{\mathbf{k},\mathbf{p},\mathbf{k}',\mathbf{p}'}b^{\dagger}_{\mathbf{k}'}b^{\dagger}_{\mathbf{p}'}b_{\mathbf{p}}b_{\mathbf{k}}$ • \Rightarrow impurity [site/bond defect] scattering $\delta J[\delta D] \Rightarrow \delta J[\delta D]b^{\dagger}_{\mathbf{k}'}b_{\mathbf{k}}$
 - \Rightarrow and impurity-assisted magnon-magnon scattering $\delta J [\delta D] \Rightarrow V_{\mathbf{k},\mathbf{p},\mathbf{k}',\mathbf{p}'}^{\mathrm{imp}} b_{\mathbf{k}'}^{\dagger} b_{\mathbf{p}'}^{\dagger} b_{\mathbf{p}} b_{\mathbf{k}}$





damping, theory expectations, I



• local distortions $\rightarrow \delta D$, $\delta J \rightarrow$ conventional impurity scattering (2D):

• gapped on thermally excited gapless:

$$\Gamma_{\mathbf{k}\to0}^{\mathrm{m-m}} \approx \frac{\pi^3}{15} \, \frac{\tilde{g}^2}{c} \left(\frac{T}{c}\right)^5$$

 $\Gamma_{\mathbf{k}}^{\mathrm{imp}} \approx \Gamma_0 \propto n_{\mathrm{imp}} \overline{\delta D}^2 \, \frac{m \, \omega_{\mathrm{max}}^2}{\Lambda \, 2}$

• (and on gapped):
$$\Gamma^{\beta\beta\to\beta\beta}_{\mathbf{k}\to 0} \approx \frac{g_{\beta}^2 m^2 T}{4\pi} e^{-\Delta/T}$$

• numbers for m-m scattering are known/derivable!

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* [3D $\rightarrow T^7$]



standard lore: $\varrho = \varrho_0^{imp} + \varrho^{ee}(T)$



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damping, theory expectations, II



- reasons for "stronger" potential:
 - m-m interactions are singular but cancel out
 - impurities violate that cancellation
 - dynamically-induced "strong" disorder: optical spin-flip "sits" at impurity, scatters acoustic mode stronger ...

beyond the standard model ...





better picture ...





cross-checks, predictions

T=0 and *T>0* impurity terms must be related ($\Gamma_0 \sim \widetilde{A} \sim n_{imp} \overline{\delta D}^2 m$) \blacksquare true, in our fit: $\Gamma_0 \approx \widetilde{A} \approx 25 \ \mu eV$ does disorder strength make sense?

$$\blacksquare$$
 estimate: $n_{\rm imp}(\overline{\delta D}/D)^2 \approx \Gamma_0/\omega_{max} \approx 10^{-2}$

translates into a (very reasonable) statement that in $BaNi_2(PO4)_2$, strong modulation of magnetic couplings of order 1 is spread over 1 in 100 unit cells

predictions:

⇒ 3D:
$$\Gamma_{3D}^{imp} \propto n_{imp} T^{9/2}$$

⇒ AFs with non-collinear order → 3-magnon coupling BaCo₂(AsO₄)₂
new diagrams:
no change in m-m
lower power of T in impurity-induced: $\Gamma_{\mathbf{k}\to 0}^{imp,T} \approx \widetilde{A}_3 \left(\frac{T}{c}\right) \ln \frac{T}{\omega_0}$
 $\widetilde{A}_3 \propto n_{imp} |\delta g_3|^2$



conclusions

- \square general case: low-T lifetime of a magnetic excitation is completely dominated by the effects induced by a simple structural disorder
- \square support from experiments
- ☑ further predictions are made
- \square should be relevant to other systems

