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Decay of quasiparticles in quantum spin liquids

M. E. Zhitomirsky
Commissariat à l'Energie Atomique, DSM/DRFMC/SPSMS, 38054 Grenoble, France

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Magnetic excitations are studied in gapped quantum spin systems, for which spontaneous two-magnon decays are allowed by symmetry. Interaction between one- and two-particle states acquires nonanalytic frequency and momentum dependence near the boundary of two-magnon continuum. This leads to a termination point of the single-particle branch for one-dimensional systems and to strong suppression of quasiparticle weight in two dimensions. The momentum dependence of the decay rate is calculated in arbitrary dimensions and the effect of external magnetic field is discussed.

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A number of magnetic materials with quantum disordered, or spin-liquid, ground states have been discovered in the past two decades. The well-known examples include the spin-Peierls compound $CuGeO_3$ (Ref. 1), dimer systems $Cs_3Cr_2Br_9$ (Ref. 2) and $TlCuCl_3$ (Ref. 3), integer-spin antiferromagnetic chains,[4] and many others. A common property of all these systems is the presence of a spin gap in the excitation spectrum, which separates a singlet, $S=0$ ground state from $S=1$ quasiparticles. The low-energy triplet of magnons can be split due to intrinsic anisotropies or under applied magnetic field. Recent neutron experiments[5,6] on two organic spin-gap materials, piperazinium hexachlorodicuprate (PHCC) and IPA-$CuCl_3$, have revealed drastic transformation of triplet quasiparticles undergoing at high energies. Upon entering two-magnon continuum (see Fig. 1), spontaneous ($T=0$) decay of a magnon into a pair of quasiparticles becomes possible, which leads to a rapid decrease in the quasiparticle lifetime in the former case[5] and complete disappearance of the single-particle branch in the latter system.[6]

Quasiparticle instability is well documented for another type of quantum liquid—superfluid ^{4}He. Predicted by Pitaevskii nearly 50 years ago,[7–9] this instability was later confirmed by neutron scattering measurements.[10,11] In liquid helium, interaction between one- and two-particle states is enhanced in the vicinity of a decay threshold by a large density of roton states and leads to avoided crossing: the single-particle branch flattens at energies below twice the roton energy and ceases to exist completely above that energy scale. The aim of the present work is to investigate the role of spontaneous two-magnon decays in quantum spin liquids. Note, that an analogy with ^{4}He has been previously invoked in Ref. 5 for an interpretation of the neutron scattering results for PHCC.

We consider an isotropic spin system with a quantum disordered ground state, which is separated by a finite gap Δ from low-energy spin-1 excitations. A bare dispersion of propagating triplets, sometimes also called triplons, is given by $\varepsilon(\mathbf{p})$. Bosonic operators $t_{\mathbf{p}\alpha}$ and $t^\dagger_{\mathbf{p}\alpha}$ destroy and create a quasiparticle with momentum $\mathbf{p}$ in one of the three polarizations $\alpha=x,y,z$. Interaction between one- and two-particle states is, generally, described by two types of cubic vertices shown in Figs. 2(a) and 2(b). In superfluid ^{4}He the presence of such particle nonconserving processes is determined by a Bose condensate, which absorbs or emits an extra particle.[8,12] In quantum spin systems with singlet ground states one finds a more diverse situation. Symmetry of a majority of low-dimensional dimer systems, such as, for example, a dimerized chain,[13] an asymmetric ladder, and a planar array of dimers,[14] allows for the presence of cubic vertices. These should be contrasted with a symmetric ladder,[15] where one- and two-magnon states belong to sectors with different parity under permutation of two legs and, consequently, do not interact. There is also a suggestion that cubic vertices exist in a Heisenberg antiferromagnetic spin-1 chain beyond the nonlinear sigma-model description.[16] In the following, we shall assume the presence of interaction between one- and two-particle states and consider its consequences for the dynamic properties of a spin liquid.

In Heisenberg magnets, rotational symmetry in the spin space fixes uniquely the tensor structure of cubic vertices. Specifically, the decay vertex, Fig. 2(a), has the following form:

$$\hat{V}_3 = \frac{1}{2}\sum_{\mathbf{k},\mathbf{q}} \Gamma(\mathbf{k},\mathbf{q})\epsilon^{\alpha\beta\gamma} t^\dagger_{\mathbf{k}\alpha} t^\dagger_{\mathbf{q}\beta} t_{\mathbf{k}+\mathbf{q}\gamma}. \tag{1}$$

Conservation of the total spin during the decay process requires that two created spin-1 quasiparticles must form an $S=1$ state, which is imposed by the antisymmetric tensor $\epsilon^{\alpha\beta\gamma}$. The vertex is, consequently, antisymmetric under permutation of the two momenta $\Gamma(\mathbf{k},\mathbf{q})=-\Gamma(\mathbf{q},\mathbf{k})$. Similar consideration applies to the source-type vertex, Fig. 2(b), which is antisymmetric under permutation of any two of three outgoing lines.

An important kinematic property of the energy spectrum $\varepsilon(\mathbf{p})$ is a type of instability at a decay threshold momentum $\mathbf{p}_c$, beyond which the energy conservation

$$\varepsilon(\mathbf{p}) = \varepsilon(\mathbf{q}) + \varepsilon(\mathbf{p}-\mathbf{q}) \tag{2}$$

is satisfied and two-magnon decays become possible. The extremum condition imposed on the right-hand side of Eq. (2) yields that two quasiparticles at the bottom of continuum always have equal velocities $\mathbf{v}_\mathbf{Q}=\mathbf{v}_{\mathbf{p}-\mathbf{Q}}$, where $\mathbf{v}_\mathbf{k}=\nabla_\mathbf{k}\varepsilon(\mathbf{k})$. Then, the simplest possibility is that both momenta are also equal with $\mathbf{Q}=\frac{1}{2}(\mathbf{p}+\mathbf{G})$, where $\mathbf{G}$ is a reciprocal lattice vector. We have verified the above assertion for several model

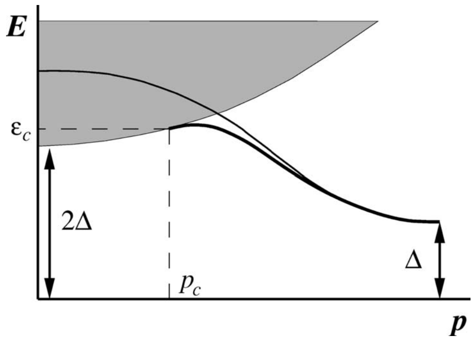

FIG. 1. Schematic structure of the energy spectrum of a quantum magnet with spin-liquid ground state. The thin solid line is a bare spectrum of triplet quasiparticles $\varepsilon(\mathbf{p})$, the full solid line is a renormalized single-particle branch, and the shaded region is two-particle continuum. The decay instability threshold is denoted by p_c.

dispersion curves[17,18] as well as for a few experimental fits.[5,6,19] The common feature of all analyzed cases is the presence of two regimes: for a small total momentum $\mathbf{p}$ of a magnon pair the minimum energy corresponds to equal momenta of two quasiparticles, whereas for $\mathbf{p}$ near the Brillouin-zone boundary two particles of the lowest-energy pair have different momenta. Such a bifurcation is closely related to the appearance of bound pairs of magnons at large momenta for one-dimensional spin systems.[13,20] Kinematic instability at the decay threshold in all considered cases corresponds, however, to a decay into a pair of quasiparticles with equal momenta. In the following we shall focus on such type of a model-independent instability.

Let us consider the second-order contribution to the normal self-energy from the decay processes shown in Fig. 2(c). Standard calculation yields

$$\Sigma_{11}(\omega,\mathbf{p}) = \int \frac{d^Dq}{(2\pi)^D}\frac{\Gamma(\mathbf{q},\mathbf{p}-\mathbf{q})^2}{\omega-\varepsilon(\mathbf{q})-\varepsilon(\mathbf{p}-\mathbf{q})+i0}, \quad (3)$$

where D is the number of dimensions. We shall be interested in the behavior of the energy spectrum at small $\Delta\mathbf{p}=\mathbf{p}-\mathbf{p}_c$ and $\Delta\omega=\omega-\varepsilon_c$ and expand accordingly all functions under integral in $\Delta\mathbf{q}=\mathbf{q}-\mathbf{Q}$, where $\mathbf{Q}=\frac{1}{2}(\mathbf{p}+\mathbf{G})$. The main difference with the Pitaevskii's analysis[8] is that the antisymmetric vertex vanishes at the decay threshold such that

$$\Gamma(\mathbf{Q}+\Delta\mathbf{q},\mathbf{Q}-\Delta\mathbf{q}) \approx 2\Delta\mathbf{q}\cdot\nabla_{\mathbf{k}}\Gamma(\mathbf{k},\mathbf{q})|_{\mathbf{k},\mathbf{q}=\mathbf{Q}}. \quad (4)$$

Including all dimensional constants in the vertex $g_3 \sim|\nabla_{\mathbf{k}}\Gamma(\mathbf{k},\mathbf{q})|$ and performing angular integration we obtain in $D\geqslant 1$

$$\Sigma_{11}(\omega,\mathbf{p}) \simeq -g_3^2\int_0^\Lambda \frac{q^{D+1}dq}{q^2+\mathbf{v}_2\Delta\mathbf{p}-\Delta\omega-i0}, \quad (5)$$

where $\mathbf{v}_2=\nabla_{\mathbf{k}}\varepsilon(\mathbf{k})|_{\mathbf{k}=\mathbf{Q}}$ and Λ is a lattice cutoff. The self-energy remains finite as $\Delta\omega,|\Delta\mathbf{p}|\to 0$, though it contains an important nonanalytic contribution $\tilde{\Sigma}(\omega,\mathbf{p})$. We shall absorb a regular part of $\Sigma_{11}(\omega,\mathbf{p})$ into $\varepsilon(\mathbf{p})$ and determine a new corrected spectrum from the Dyson's equation: $G^{-1}(\omega,\mathbf{p})=\omega-\varepsilon(\mathbf{p})-\tilde{\Sigma}(\omega,\mathbf{p})=0$.

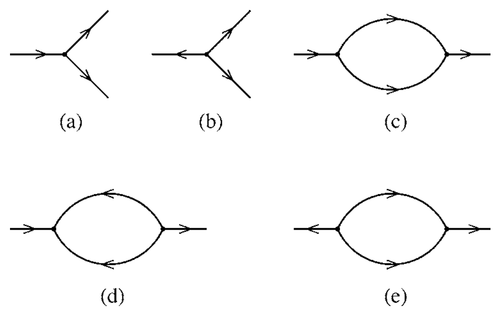

FIG. 2. Two cubic vertices of the (a) decay and (b) source types and their contributions to the (c) and (d) normal and (e) anomalous self-energies.

Two other second-order diagrams constructed from cubic vertices are presented in Figs. 2(d) and 2(e). Correction to the normal self-energy shown in Fig. 2(d) is an analytic function of frequency and momentum and can be absorbed into $\varepsilon(\mathbf{p})$. The anomalous self-energy $\Sigma_{12}(\omega,\mathbf{p})$, Fig. 2(e), has the same dependence on ω and $\mathbf{p}$ as the one-loop diagram (5). Using the Belyaev's expression for the normal Green's function of a Bose gas,[8,12]

$$G^{-1}(\omega,\mathbf{p}) = \omega-\varepsilon(\mathbf{p})-\Sigma_{11}(\omega,\mathbf{p}) + \frac{\Sigma_{12}(\omega,\mathbf{p})\Sigma_{21}(\omega,\mathbf{p})}{\omega+\varepsilon(-\mathbf{p})+\Sigma_{11}(-\omega,-\mathbf{p})}, \quad (6)$$

one can conclude that inclusion of the anomalous self-energy simply modifies a coefficient in front of the nonanalytic contribution (5) without changing its functional dependence. A similar result applies to the vertex correction from multiple two-particle scattering processes. Since a one-loop diagram is finite for $\omega=\varepsilon_c$, $\mathbf{p}=\mathbf{p}_c$, the renormalized vertex $\tilde{g}_3$ also remains finite and the vertex correction amounts to replacement $g_3^2\to g_3\tilde{g}_3$ in front of the integral in Eq. (5). Thus, the following analysis based on analytic properties of the one-particle Green's function is not restricted to the lowest-order perturbation theory used in the derivation of Eq. (3) and is quite general.

One-dimensional gapped spin systems. Let us begin with a stable region, where spontaneous decays are forbidden: $\Delta\omega<0$, $\Delta p>0$, such that $(v_2\Delta p-\Delta\omega)>0$. Separating the nonanalytic part in the integral (5) and expanding $\varepsilon(p)\approx\varepsilon_c+v_1\Delta p$ we obtain for the inverse Green's function

$$G^{-1}(\omega,p) = \Delta\omega - v_1\Delta p - \lambda\sqrt{v_2\Delta p-\Delta\omega}, \quad (7)$$

with $\lambda=\pi g_3^2/2$. The one-particle Green's function with such a dependence on ω and p has been briefly discussed in Ref. 8. In the following we give more details relevant for the case of Eq. (7).

The condition for a pole $G^{-1}(\omega,p)=0$ is transformed to a quadratic equation, which yields

$$\bar{\varepsilon}(p) = \varepsilon_c + v_1\Delta p - \frac{\lambda^2}{2} + \sqrt{\frac{\lambda^4}{4}+\lambda^2(v_2-v_1)\Delta p}. \quad (8)$$

Away from the crossing point for $\Delta p\gg\lambda^2/(v_2-v_1)$, the slope of the single-particle branch coincides with the bare magnon velocity $d\bar{\varepsilon}(p)/dp \approx v_1 < 0$, whereas for $\Delta p \to 0$ the slope changes sign to $d\bar{\varepsilon}(p)/dp = v_2 > 0$. In the crossover region $\Delta p \sim \lambda^2/(v_2 - v_1)$ the quasiparticle weight Z,

$$Z^{-1} = \left. \frac{\partial G^{-1}}{\partial \omega} \right|_{\omega=\bar{\varepsilon}} = 1 + \frac{\lambda^2 + \sqrt{\lambda^4 + 4\lambda^2(v_2 - v_1)\Delta p}}{4(v_2 - v_1)\Delta p}, \quad (9)$$

is continuously suppressed and vanishes, once the single-particle branch touches the two-magnon continuum.

On the opposite side of the crossing point, $\Delta p < 0$, $\Delta\omega > 0$, and $(\Delta\omega - v_2\Delta p) > 0$, the nonanalytic part of the self-energy becomes purely imaginary $\tilde{\Sigma} = -i\lambda\sqrt{\Delta\omega - v_2\Delta p}$. Formally, condition $G^{-1}(\omega, p) = 0$ yields after transformation to a quadratic equation the same solution as in the stable region. For small negative Δp the root (8) is real and satisfies $\Delta\omega - v_1\Delta p < 0$ and $\Delta\omega - v_2\Delta p < 0$. The latter of the above two relations contradicts the assumption made in the beginning of the paragraph for the decay region, whereas the former is inconsistent with zero of $G^{-1}(\omega, p)$ in the region, where $v_2\Delta p - \Delta\omega > 0$, see Eq. (7). Thus, inside the continuum, a physical pole of the one-particle Green's function reappears only at a finite distance from the crossing point, where $\mathrm{Re}\{\Delta\omega\} > v_2\Delta p$ or

$$\Delta p < -\frac{\lambda^2}{2(v_2 - v_1)}. \quad (10)$$

Disappearance of single-magnon excitations inside the continuum is quite similar to the termination point in the energy spectrum of superfluid ^{4}He (Refs. 7–11). In both cases the single-particle branch approaches tangentially the boundary of the two-particle continuum.

Further away from the lower edge of the continuum in the region, where $\bar{\varepsilon}(p) \approx \varepsilon_c + v_1\Delta p$, one can find for the inverse quasiparticle lifetime

$$\gamma_p \simeq 2\lambda\sqrt{(v_2 - v_1)|\Delta p|}. \quad (11)$$

Such a square-root dependence of the decay rate follows also from a simple golden rule consideration.[16] We have seen, however, that the full dependence of the self-energy on frequency and momentum must be kept in the vicinity of the decay threshold. Besides, if the cubic vertices have no special smallness, i.e., λ is of the order of the bandwidth of $\varepsilon(p)$, the perturbative regime, where Eq. (11) applies, is never realized. Such a situation apparently occurs in the quasi-one-dimensional gapped spin system IPA-$CuCl_3$, where no trace of single-magnon excitations has been observed inside the continuum.[6]

Two-dimensional gapped spin systems. Below decay threshold in the stable region, where $(\mathbf{v}_2\Delta\mathbf{p} - \Delta\omega) > 0$, the nonanalytic part of the self-energy becomes

$$\tilde{\Sigma}(\omega, \mathbf{p}) = \lambda(\mathbf{v}_2\Delta\mathbf{p} - \Delta\omega)\ln\frac{R}{\mathbf{v}_2\Delta\mathbf{p} - \Delta\omega}, \quad (12)$$

where $\lambda = g_3^2/2$ and R is an energy cutoff. Although a pole of the Green's function cannot be found analytically, it is easy to verify that the solution of $G^{-1}(\omega, \mathbf{p}) = 0$ exists all the way down to the crossing point. The quasiparticle branch touches tangentially the boundary of the continuum $\nabla_{\mathbf{p}}\bar{\varepsilon}(\mathbf{p}) = \mathbf{v}_2$ for $\mathbf{p} = \mathbf{p}_c$, whereas the quasiparticle weight is gradually diminished and vanishes at the boundary.

Above the decay threshold $[(\mathbf{v}_2\Delta\mathbf{p} - \Delta\omega) < 0]$ the self-energy acquires the imaginary part

$$\tilde{\Sigma}(\omega, \mathbf{p}) = -\lambda(\Delta\omega - \mathbf{v}_2\Delta\mathbf{p})\left(\ln\frac{R}{\Delta\omega - \mathbf{v}_2\Delta\mathbf{p}} + i\pi\right). \quad (13)$$

In the perturbative regime, where $\bar{\varepsilon}(\mathbf{p}) \approx \varepsilon_c + \mathbf{v}_1\Delta\mathbf{p}$, the decay rate of magnons grows linearly inside the continuum

$$\gamma_p \simeq 2\pi\lambda(\mathbf{v}_1 - \mathbf{v}_2)\Delta\mathbf{p}. \quad (14)$$

In the close vicinity of the crossing point we write instead $\bar{\varepsilon}(\mathbf{p}) \approx \mathbf{v}_2\Delta\mathbf{p} + a$ with $\mathrm{Re}\, a > 0$ and transform the equation on the pole of the Green's function to

$$a\left[\ln\frac{R}{a} + i\pi\right] = \frac{(\mathbf{v}_1 - \mathbf{v}_2)\Delta\mathbf{p}}{\lambda} = b > 0. \quad (15)$$

With logarithmic accuracy the solution of the above equation is

$$\mathrm{Re}\, a = \frac{b}{\sqrt{\ln^2 R/b + \pi^2}}, \quad \mathrm{Im}\, a = \frac{-\pi b}{\ln^2 R/b + \pi^2}. \quad (16)$$

A single-particle branch of a two-dimensional gapped spin system can be, therefore, continued inside the decay region. However, the quasiparticle weight is suppressed in a range of momenta near the crossing point,

$$|(\mathbf{v}_1 - \mathbf{v}_2)\Delta\mathbf{p}| \simeq Re^{-1/\lambda}. \quad (17)$$

Suppression of the quasiparticle peak has been experimentally observed in two-dimensional quantum-disordered antiferromagnet PHCC (Ref. 5).

Three-dimensional gapped spin systems. The nonanalytic part of the self-energy is given in this case by

$$\tilde{\Sigma}(\omega, \mathbf{p}) = -\lambda(\mathbf{v}_2\Delta\mathbf{p} - \Delta\omega)^{3/2}, \quad \Delta\omega < \mathbf{v}_2\Delta\mathbf{p},$$

$$\tilde{\Sigma}(\omega, \mathbf{p}) = -i\lambda(\Delta\omega - \mathbf{v}_2\Delta\mathbf{p})^{3/2}, \quad \Delta\omega > \mathbf{v}_2\Delta\mathbf{p},$$

where $\lambda = \pi g_3^2/2$. Such a weakly nonanalytic term can be treated perturbatively near the crossing point. In particular, the quasiparticle weight is not suppressed as $\mathbf{p} \to \mathbf{p}_c$ and $\bar{\varepsilon}(\mathbf{p})$ does not change its slope at the crossing. The magnon decay rate in the three-dimensional case is

$$\gamma_p \simeq 2\lambda[(\mathbf{v}_1 - \mathbf{v}_2)\Delta\mathbf{p}]^{3/2}. \quad (18)$$

An experimentally relevant question is how an external magnetic field affects the decay processes. If the Zeeman energy is smaller than the spin gap $g\mu_B H < \Delta$, the role of an applied field reduces to splitting a triplet of low-energy excitations. This can, in principle, lead to breaking the resonance condition (2). To study such a possibility one should transform from the vector basis $t_{\mathbf{p}\alpha}$ to states with a definite spin projection on the field direction

$$t_{\mathbf{p}0} = t_{\mathbf{p}z}, \quad t_{\mathbf{p}\pm} = \mp\frac{1}{\sqrt{2}}(t_{\mathbf{p}x} \mp i t_{\mathbf{p}y}). \quad (19)$$

The decay vertex (1) taken in the new basis has the following form:

$$i\epsilon^{\alpha\beta\gamma}t^{\dagger}_{\mathbf{p}\alpha}t^{\dagger}_{\mathbf{q}\beta}t_{\mathbf{k}\gamma}=(t^{\dagger}_{\mathbf{p}+}t^{\dagger}_{\mathbf{q}-}-t^{\dagger}_{\mathbf{p}+}t^{\dagger}_{\mathbf{q}-})t_{\mathbf{k}z}+(t^{\dagger}_{\mathbf{p}+}t^{\dagger}_{\mathbf{q}z}-t^{\dagger}_{\mathbf{p}z}t^{\dagger}_{\mathbf{q}+})t_{\mathbf{k}+}+(t^{\dagger}_{\mathbf{p}z}t^{\dagger}_{\mathbf{q}-}-t^{\dagger}_{\mathbf{p}-}t^{\dagger}_{\mathbf{q}z})t_{\mathbf{k}-}. \quad (20)$$

In all decay channels a destroyed one-particle and a created two-particle states experience the same energy shifts. Therefore, the decay threshold momentum $\mathbf{p}_c$ does not change for all three split branches of the spin-1 excitations.

Once the Zeeman energy exceeds the triplet gap, $H>H_c=\Delta/g\mu_B$, a Bose condensation of magnons takes place in $D>1$ (Ref. 21). The ground state acquires a nonzero magnetization and a long-range order of transverse spin components. Such a canted antiferromagnetic structure opens an additional channel for spontaneous decays of low-energy excitations in the gapless branch, similar to the prediction made for ordered antiferromagnets in the vicinity of the saturation field.[22]

Intrinsic magnetic anisotropies, which become important in materials with spins $S\geqslant 1$, can also affect the decay processes. Anisotropy changes differently the dispersion of triplet excitations and modifies the resonance condition (2). The decay rate of spin-1 excitations depends, then, on a polarization, as was experimentally observed in bond-alternating quasi-one-dimensional antiferromagnet NTENP (Ref. 23). The tensor structure of the decay vertex (1) can be also modified due to the absence of spin-rotational symmetry. Dynamic properties of spin liquids with different types of anisotropies deserve further theoretical investigations.

In conclusion, by studying analytic properties of the one-particle Green's function near the decay threshold we have found that crossing of a single-particle branch into two-magnon continuum is described solely by a growing linewidth of magnons only for three-dimensional quantum spin liquids. In two dimensions there is, in addition, strong suppression of a one-magnon peak near the crossing point, whereas in one dimension a single-magnon branch terminates at the continuum boundary.

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