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Magnetic resonance in low-temperature paramagnets

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Contents

Abstract. Low-temperature paramagnets are a class of spin systems that remain in a disordered paramagnetic state down to the lowest temperatures despite the presence of noticeable spin-spin exchange coupling. The magnetic properties of these systems at low temperatures are due to collective spin excitations, and, in a magnetic field, such spin systems can demonstrate field-induced antiferromagnetic ordering. Magnetic resonance allows studying the details of the excitation spectrum in the paramagnetic low-field and ordered high-field phases of low-temperature paramagnets, revealing the fine structure of the spectrum of collective excitations, details of the interaction between excitations, changes in the excitation spectrum induced by the magnetic field, and the emergence of non-Goldstone modes of spin dynamics in high-field ordered phases.

Keywords: spin liquid, magnetic resonance, low-temperature paramagnets, quantum magnets

1. Introduction

The study of the properties of magnetic dielectrics, systems of exchange-coupled atomic spins on a regular crystal lattice, has a long history, including the prediction of antiferromagnetism by Landau [1] and Néel [2], confirmed by neutron diffraction experiments by Shull [3], and the discovery of spin waves (collective excitations of such spin systems) predicted by Bloch [4] in Brockhouse's experiments [5, 6] on inelastic neutron scattering. A large number of compounds are known in which ferro- or antiferromagnetic ordering occurs with decreasing temperature.

The 'traditional' scenario for the formation of magnetic order is as follows. A system of exchange-coupled spins with a Heisenberg Hamiltonian

$$\hat{\mathcal{H}} = \sum_{i,j} J_{ij}\hat{\mathbf{S}}_i\hat{\mathbf{S}}_j$$

at temperatures $k_\mathrm{B}T \gg |J|$ (J being the characteristic value of the exchange integral) is in a paramagnetic state in which the magnetic entropy is close to $S_\mathrm{max} = R\ln(2S+1)$, and at temperature $k_\mathrm{B}T_\mathrm{c} \sim |J|$ a transition to a magnetically ordered state occurs. At the moment of transition, the magnetic entropy is close to S_max, and at $T \to 0$, the magnetic entropy, now associated with collective excitations of the magnet, turns to zero.

However, in recent decades, systems that do not follow this line of behavior have been actively studied. It turns out that, in some cases (often associated with the one- or two-dimensionality of the spin subsystem, frustration of interactions, or another type of special geometry of exchange bonds), spin systems remain in a paramagnetic state, without demonstrating the traditional type of ordering, down to temperatures $k_\mathrm{B}T \ll |J|$, even if the magnetic entropy $S_\mathrm{m} \ll S_\mathrm{max}$. Systems of this type are an example of *a low-temperature paramagnet* — a state of the spin system analogous to the state of a quantum liquid in helium. The properties of such systems at low temperatures are due to collective excitations. The spectrum of these excitations, their interaction with each other, and the effect of a magnetic field on the excitation of a low-temperature paramagnet are of interest for theoretical and experimental studies.

In this brief review, we will consider a narrower class of low-temperature paramagnets: spin systems in which the spectrum of collective excitations is separated by an energy gap from the singlet ground state. The simplest example of such a spin system is one of dimers coupled by an antiferromagnetic exchange interaction: in an isolated pair of spins $s = 1/2$, there is a ground state with $S=0$ and triplet excitations with $S=1$. In the presence of a weak interdimer exchange interaction, the ground state of the collective system remains a singlet $S = 0$, and the triplet excitations become delocalized and acquire a dispersion $\varepsilon(\mathbf{q})$ with some gap Δ. Less trivial examples are one-dimensional chains of integer spins (Haldane magnets [7]) and 'spin ladder' systems [8, 9].

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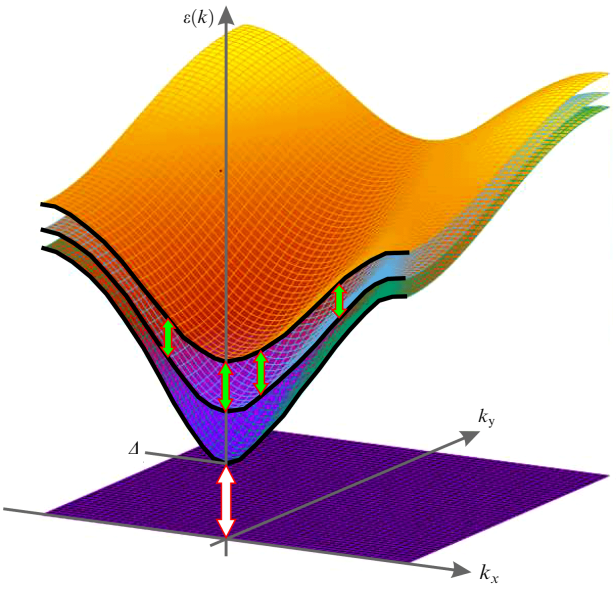

Figure 1. Schematic representation of spectrum of triplet excitations of a low-temperature paramagnet in an applied magnetic field. Arrows indicate possible magnetic resonance transitions between triplet sublevels and one of the singlet-triplet transitions.

The application of a magnetic field removes the degeneracy in the spin projection, splitting the sublevels of the triplet state with $S = 1$ (Fig. 1). Transitions between sublevels can be studied using magnetic resonance spectroscopy. The high energy resolution of this method makes it possible to study the properties of collective excitations and detect the fine structure of the triplet spectrum, while the observation of singlet-triplet transitions (which may be weakly allowed due to the low symmetry of the crystal) allows tracing the dependence of the gap on the magnetic field and temperature.

Of particular interest is the behavior of low-temperature paramagnets in strong magnetic fields. The dispersion of triplet excitations $\varepsilon(\mathbf{q})$ gives rise to two characteristic fields: the spin gap closing field $H_{c1} = \min(\varepsilon(\mathbf{q}))/(g\mu_B)$, in which a nonzero magnetization arises at $T = 0$, and the saturation field $H_{c2} \simeq \max(\varepsilon(\mathbf{q}))/(g\mu_B)$, in which complete polarization of the spin system occurs (see, e.g., [10]). In many cases, at a sufficiently low temperature and $H_{c1} < H < H_{c2}$, a phase transition to an unexpected field-induced antiferromagnetically ordered state is observed, which in the purely Heisenberg case can be described as Bose condensation of quasiparticles, where the magnetic field plays the role of a tunable chemical potential [11–15]. Magnetic resonance in the ordered phase measures the energy of antiferromagnetic magnons with $q = 0$, being a very sensitive marker of the appearance of non-Goldstone modes.

The experiments described in this paper were mainly carried out at the P.L. Kapitza Institute for Physical Problems.

2. Results and discussion

2.1 Magnetic resonance of collective triplet excitations

In fields lower than the spin gap closing field $H < H_{c1}$, the magnetic resonance response of a low-temperature paramagnet with a gapped excitation spectrum is associated mainly with allowed transitions between triplet sublevels.

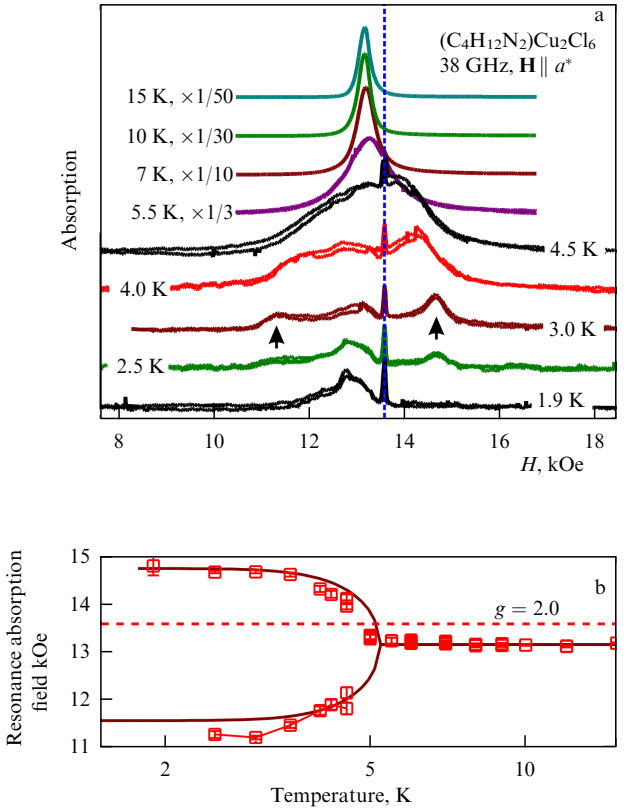

Figure 2. (a) Resonance absorption spectra in quasi-two-dimensional low-temperature paramagnet $(C_4H_{12}N_2)Cu_2Cl_6$ (in the literature referred to as PHCC) at different temperatures. High-temperature data are multiplied by factors indicated in figure for better presentation of data. Narrow absorption peaks near vertical dotted line are DPPH marker ($g = 2.00$). Vertical arrows show positions of split components of fine structure of EPR spectrum at low temperatures. (b) Dependence of resonance absorption field on temperature (symbols) and comparison with exchange narrowing model (curves).

The presence of a gap in the excitation spectrum leads to an exponential freezing of the population of the triplet sublevels and, accordingly, to the freezing of the intensity of the observed resonance absorption spectrum. Observation of such behavior of the magnetic resonance signal intensity, qualitatively different from that expected for a conventional Curie paramagnet, is one of the characteristic features of a low-temperature paramagnet with a gapped excitation spectrum. This exponential freezing of the population of triplet states at low temperatures makes the regime of an ideal gas of triplet excitations achievable, when quasiparticle excitations can be considered independent of each other and noninteracting.

An example of such behavior, observed in the quasi-two-dimensional dimeric low-temperature paramagnet $(C_4H_{12}N_2)Cu_2Cl_6$ (PHCC in the literature), is shown in Fig. 2: as the temperature decreases, the magnetic resonance signal loses intensity, splits into several components, and practically disappears at the lowest temperature of 1.7 K (the magnetic resonance signal of irregular shape remaining at this temperature is associated with a small number (less than 1%) of magnetic defects) [16].

The observed splitting of the magnetic resonance spectrum is associated with the influence of the effective crystal field on the collective excitations of the low-temperature paramagnet. In the case of axial symmetry of the crystal, the

effective Hamiltonian of the triplet excitation can be written as

$$\hat{\mathcal{H}}(\mathbf{q}) = \Delta_{\mathbf{q}} - g\mu_{\mathrm{B}}\mathbf{H}\hat{\mathbf{S}} + D_{\mathbf{q}}\hat{S}_Z^2 . \qquad (1)$$

Here, $D_{\mathbf{q}}$ is the wavevector-dependent value of the effective anisotropy constant, which is microscopically related to various anisotropic spin-spin interactions (some examples of such a relationship are considered in [17–19]), and the direction of the main anisotropy axis Z is determined by the symmetry of the crystal. As a result, for noninteracting triplet excitations, a fine structure of the spectrum characteristic of spins $S = 1$ arises: the magnetic resonance fields for the transitions with a change in the projection of the excitation's spin on the direction of the magnetic field $S_{\mathrm{H}}|+1\rangle \leftrightarrow |0\rangle$ and $|0\rangle \leftrightarrow |-1\rangle$ differ. Such a fine structure of the magnetic resonance spectrum (and, accordingly, the fine structure of the energy levels of triplet excitations) was observed in low-temperature paramagnets of various types [16, 20–24]. The high resolution of magnetic resonance spectroscopy makes it possible to reliably detect and characterize this effect, as well as to determine the direction of the anisotropy axes.

An interesting feature of such spectra, qualitatively distinguishing the magnetic resonance of delocalized triplet excitations from the 'classical' magnetic resonance of localized spins $S = 1$, is the averaging of the observed spectrum over the populated states in the entire first Brillouin zone: at a wavelength of $\lambda \sim 1$ cm used in magnetic resonance spectroscopy, the photon wave vector is practically zero, $k \approx 0$. Therefore, the law of quasi-momentum conservation in photon absorption processes allows 'vertical' transitions between triplet sublevels throughout the Brillouin zone. The dependence of the effective anisotropy constant on the wave vector $D_{\mathbf{q}}$ [17] can lead to the fact that, even in the low-temperature regime of an ideal gas of noninteracting triplet excitations, a dependence of the observed splitting of the magnetic resonance spectrum fine structure on temperature will arise, requiring special attention when analyzing the results.

This type of averaging the absorption signal over the populated region of k-space was successfully observed in the quasi-one-dimensional Haldane magnet $PbNi_2V_2O_8$ [20, 23]. In this compound, the Ni^{2+} ions form weakly coupled chains of spins with $S = 1$, and the effective anisotropy acting on triplet excitations is not due to spin-spin interactions but to a fairly noticeable single-ion anisotropy of the nickel ions. This allows determining the characteristic effective anisotropy constant from the anisotropy of the critical field for closing the spin gap H_{c1} measured at low temperatures. Figure 3 illustrates a comparison of the magnetic resonance spectrum measured at 15 K with the simulation results. The EPR absorption spectrum calculated using the effective anisotropy constant determined from the anisotropy of the field H_{c1} shows a clearly overestimated splitting of the absorption maxima compared to the experiment. At the same time, taking into account the three-dimensional dispersion of spin sublevels [25], which leads to the dependence of the effective anisotropy constant on the position in the Brillouin zone, gives significantly better agreement with the experiment. Modeling the position of the absorption maximum at different temperatures with the change in the characteristic volume of populated states considered (see Fig. 3) shows that, even in the absence of interaction of excitations with each other, averaging the absorption spectrum over all states can lead to the emergence of a noticeable temperature dependence of the observed fine structure of the EPR spectrum of collective triplet excitations [20].

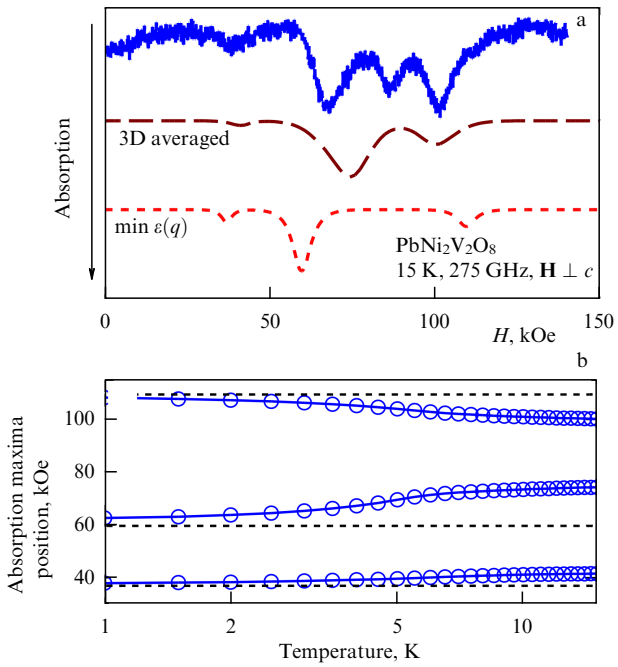

Figure 3. (a) Comparison of experimentally measured resonance absorption spectrum in an oriented sample of quasi-one-dimensional Haldane magnet $PbNi_2V_2O_8$ with results of modeling taking into account dependence of anisotropy parameters on wave vector and averaging over populated states in first Brillouin zone (dashed curve marked '3D averaged') and with anisotropy parameters for minimum of the spectrum (dotted curve marked 'min $\varepsilon(q)$'). (b) Model dependence of EPR absorption maxima position for noninteracting triplet excitations with averaging over populated states in first Brillouin zone (symbols). Dotted lines shows expected positions of resonance absorption fields for transitions to minimum of excitation spectrum.

As the temperature increases, the concentration of triplet excitations increases, and the excitations' gas becomes nonideal. The most obvious manifestation of this interaction is the effect of exchange narrowing [26]: the exchange interaction between triplet excitations leads to the merging of the fine structure components of the magnetic resonance spectrum into one common resonant absorption line (see Fig. 2). Such behavior was also observed in other low-temperature paramagnets [19, 22]. The observed temperature dependence of the resonance absorption field and the width of the magnetic resonance line can be described within the Anderson exchange narrowing model [26], assuming that the characteristic exchange frequency exponentially depends on temperature [27] (which actually reflects the exponential dependence of the concentration of triplet excitations on temperature). An example of the results of such modeling [20] is shown in Fig 2.

The interaction of triplet excitations is one of the channels of spin precession relaxation reflected in the width of the magnetic resonance line. In particular, if the relaxation process with the participation of N quasiparticles dominates, then, in the low-temperature limit, a relationship is expected between the width of the magnetic resonance line ΔH, the equilibrium concentration of excitations n, and the integrated intensity of resonance absorption I (or magnetiza-

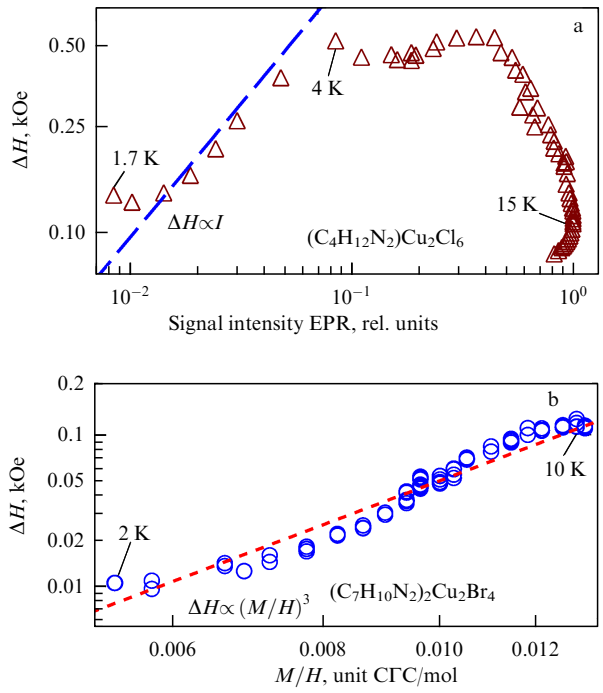

Figure 4. Relationship between EPR linewidth and intensity of resonance absorption or static magnetization for various low-temperature paramagnets. Figures also show some characteristic temperatures at which corresponding points were obtained. (a) Quasi-two-dimensional low-temperature paramagnet $(C_4H_{12}N_2)Cu_2Cl_6$ (PHCC). (b) Quasi-one-dimensional 'spin ladder' compound $(C_7H_{10}N_2)_2CuBr_4$ (DIMPY).

tion M) $I \propto M \propto n$:

$$\Delta H \propto n^{(N-1)} \propto I^{(N-1)} \propto M^{(N-1)}. \qquad (2)$$

An example of this type of dependence is shown in Fig. 4. In the quasi-two-dimensional dimeric magnet $(C_4H_{12}N_2)Cu_2Cl_6$ in the low-temperature limit, a direct proportionality is observed between the width of the magnetic resonance line and the absorption intensity, corresponding to the action of the exchange narrowing mechanism described above, for which pair interactions of quasiparticles with each other are of primary importance. For another low-temperature paramagnet, a 'spin ladder' system $(C_7H_{10}N_2)_2CuBr_4$, the dependence $\Delta H \propto (M/H)^3$ is observed, which indicates a dominant relaxation channel involving four quasiparticles. The microscopic nature of such a multiparticle interaction remains unclear.

2.2 Singlet-triplet transitions and magnetic resonance in field-induced antiferromagnetic phase

As the temperature decreases, the population of triplet excited states becomes zero, and at $T \to 0$ the magnetic resonance response due to them disappears. However, at low temperatures, other types of magnetic resonance arise that are specific to low-temperature paramagnets with a gapped excitation spectrum.

In some cases, the anisotropic spin-spin interactions present in the crystal mix the singlet ($S = 0$) ground state and triplet ($S = 1$) collective excitations, which allows transitions between these spin multiplets (see Fig. 1) [21, 22, 24, 29]. The law of quasi-momentum conservation during photon absorption requires that such transitions occur only for wave vectors present in the ground state: this is primarily $k = 0$ (the center of the Brillouin zone), corresponding to a spatially homogeneous state, and in some cases, e.g., in the presence of staggered components of spin-spin interactions, some additional (discrete) wave vectors may be present [24]. The minimum of the excitation spectrum $\varepsilon(\mathbf{q})$ may not be located at the center of the zone, so the singlet-triplet transition does not necessarily occur at the minimum of the excitation spectrum.

Magnetic resonance spectroscopy allows tracing the change in the frequency of the singlet-triplet transition depending on a magnetic field with high accuracy. In the most interesting case, when the singlet-triplet transition to the minimum of the spectrum is allowed, it is possible to trace the closing of the gap in the excitation spectrum in a field H_{c1} with high accuracy (significantly exceeding the accuracy of the inelastic neutron scattering technique).

Most low-temperature paramagnets with a gapped excitation spectrum demonstrate the occurrence of field-induced antiferromagnetic ordering above the field H_{c1}. The low symmetry of the crystals leads to the fact that, in this field-induced antiferromagnetic phase, the energy of antiferromagnetic magnons is nonzero, which allows observing antiferromagnetic resonance at $H > H_{c1}$. Within the framework of the classical theory of antiferromagnetic resonance [30, 31], the observed resonance mode at the frequency $\omega \ll \Omega_L = \gamma H$ corresponds to the antiferromagnetic resonance frequency $\omega = \sqrt{2H_A H_E}$, which is independent of the magnetic field in a traditional antiferromagnet (here, H_A and H_E are the anisotropy field and the exchange field). Within the framework of the mean field theory, this frequency is proportional to the value of the antiferromagnetic order parameter. In the case of a low-temperature paramagnet, even at $T = 0$, the antiferromagnetic order parameter arises from zero in the field H_{c1}, which leads to a change in this resonance frequency with the field. In this case, in the vicinity of the spin gap closing field, a situation is realized that is not typical for ordinary antiferromagnets: even at $T \to 0$, the antiferromagnetic order parameter in the vicinity of the field H_{c1} is far from saturation; therefore, longitudinal spin waves are noticeably present in the dynamics of the antiferromagnetic phase [32].

An example of observing such magnetic resonance signals is shown in Fig. 5 using the compound $TlCuCl_3$, in which a three-dimensional system of coupled antiferromagnetic dimers is formed. In this compound, the minimum of the excitation spectrum coincides with the center of the first Brillouin zone, so the field in which the singlet-triplet transition occurs increases with decreasing frequency (mode 'A' in the figure), turning to zero at the point of transition to the field-induced antiferromagnetic phase. Above the phase transition field H_{c1}, the antiferromagnetic resonance mode ('B') is observed, the frequency of which increases with the field, reflecting the growth of the antiferromagnetic order parameter. For comparison, note that the Larmor frequency $\Omega_L = \gamma H$ in a field of 70 kOe is about 200 GHz, i.e., much higher than the frequency of the observed antiferromagnetic resonance mode.

The anisotropic interactions present in the crystal affect all types of magnetic resonance response observed in low-temperature paramagnets. They lead to the appearance of a fine structure of the spectrum of triplet excitations, affect the behavior of the gap in the spectrum near the spin gap closing field H_{c1}, and lead to violation of XY symmetry and the appearance of a spin-wave mode with a nonzero frequency in

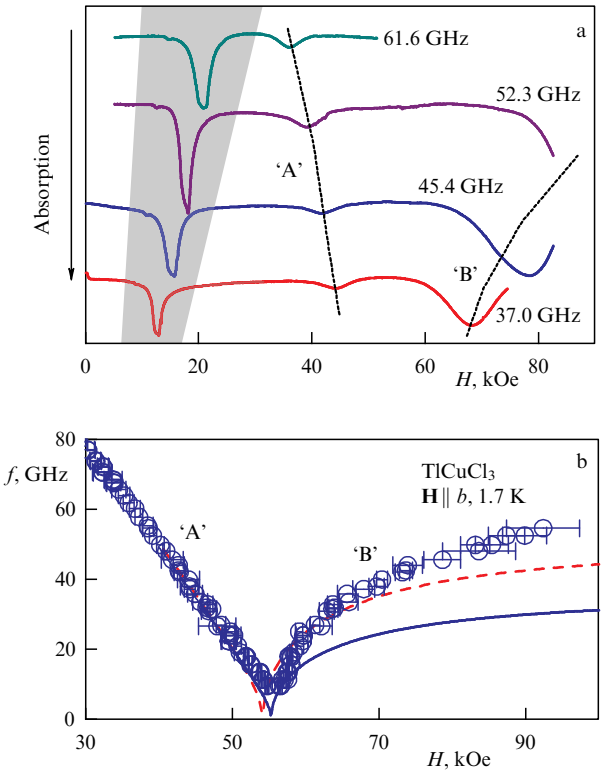

Figure 5. (a) Magnetic resonance spectra in a three-dimensional dimer magnet $TlCuCl_3$ at various microwave radiation frequencies, $\mathbf{H}||b$, $T = 1.7$ K. 'A' — absorption associated with singlet-triplet transition, 'B' — antiferromagnetic resonance in field-induced ordered phase. Dotted lines connect magnetic resonance signals of the same type for clarity, gray fill marks region of paramagnetic absorption associated with defects formed on crystal surface upon contact with atmosphere. (b) Frequency-field diagram of magnetic resonance in $TlCuCl_3$ for singlet-triplet transition ('A') and antiferromagnetic resonance ('B'). Curves are plotted within framework of Farutin–Marchenko hydrodynamic model [28] with anisotropy parameters determined from fine structure of spectrum of triplet excitations (solid curves) and for doubled values of anisotropy constants (dashed curve).

the field-induced antiferromagnetic phase. All these effects can be considered within the framework of the hydrodynamic model proposed by Farutin and Marchenko for describing the low-frequency spin dynamics of low-temperature paramagnets [28]. This model assumes a small gap in the spectrum of a low-temperature paramagnet and the proximity of such a system to traditional ordering. This allows describing the low-frequency spin dynamics both in the disordered low-field phase and in the antiferromagnetically ordered high-field phase as oscillations of a certain vector field, which has the meaning of an order parameter in the ordered phase. An example of calculating the frequencies of natural oscillations of the spin system in $TlCuCl_3$ within the framework of this model is shown in Fig. 5. The model demonstrates good semiquantitative agreement with experiment for the values of the anisotropy parameters describing the observed fine structure of the spectrum of triplet excitations (solid lines in Fig. 5). This discrepancy can be partly associated with the above-described effect of reducing the apparent splitting of the fine structure of the spectrum of triplet excitations when averaging over the populated states (see Fig. 3): doubling the values of the anisotropy constants improves agreement between the experiment and the model calculation (dashed lines in Fig. 5) [20].

We also note the unusual effect of 'inversion of anisotropy axes' upon transition to the ordered phase, which distinguishes low-temperature paramagnets from traditional spin systems. It turns out that the 'easy' anisotropy axis for triplet excitations in the low-field paramagnetic phase upon transition to the field-induced antiferromagnetic phase becomes a 'hard' anisotropy axis for the antiferromagnetic order parameter. Magnetic resonance spectroscopy has made it possible to systematically trace this effect in various low-temperature paramagnets [20]. This effect was first noted by Date and Kindo [18] for Haldane magnets; the hydrodynamic approach of Farutin–Marchenko [28] describes the observed inversion of anisotropy axes.

3. Conclusions

Low-temperature paramagnets are a class of spin systems whose unusual properties are essentially determined by the laws of quantum physics. Fundamental interest in such systems is associated with the formation of a correlated ground state not ordered in the traditional sense, the existence of well-defined quasiparticle excitations, and the existence of unusual phases in an applied magnetic field.

The singlet ground state and the gap in the spectrum of magnetic excitations determine the common physical properties of the studied compounds: freezing of magnetic susceptibility at low temperatures and the onset of field-induced antiferromagnetic ordering above the critical field of closing the spin gap. The high energy resolution of the magnetic resonance spectroscopy technique allows studying fine details of the excitation spectrum of a low-temperature paramagnet both in the quantum-disordered low-field phase and in the field-induced antiferromagnetically ordered phase above the critical field of closing the spin gap.

When studying low-temperature paramagnets with a gapped excitation spectrum, it is possible to observe and numerically characterize with high accuracy the fine structure of triplet levels in low fields, characterize spin relaxation processes, and observe and numerically characterize the emergence of non-Goldstone modes in the field-induced antiferromagnetic phase. In some spin-gap systems, it is possible to observe direct transitions between the singlet ground state and excited triplet states of the quantum system.

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