# Multi-frequency and high magnetic field electron spin resonance in the quantum spin system $Ni(C_5H_{14}N_2)_2N_3(PF_6)$ and the diluted magnetic semiconductor GaN:Fe.

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(1) Quantum spin systems defined as low dimensional (one-dimensional(1D) or two-dimensional(2D)) antiferromagnets with a small spin value have attracted a number of condensed matter scientists in the last few decades, because they exhibit interesting phenomena, *e.g.* an energy gap in a one-dimensional antiferromagnet originated from a many body quantum effect, namely the Haldane gap, magnetization plateau due to a quantum origin, and high $T_c$ superconductivity. Recently, a considerable attention has been paid to the field-induced phenomena of the quantum spin-gap systems such as the $S$ = 1/2 weakly-coupled antiferromagnetic dimer, the $S$ = 1/2 or 1 bond-alternating 1D antiferromagnet and the $S$ = 1 1D Heisenberg antiferromagnet (HAF). The ground state of these spin-gap systems is a singlet and the first excited state is a triplet at zero field. The energy of one of the triplet states decreases with increasing magnetic field, and the singlet-triplet gap closes at a critical magnetic field $H_c$ which corresponds to a quantum phase transition point. In $TlCuCl_3$, one of the $S$ = 1/2 weakly-coupled antiferromagnetic dimer systems, the long range order appeared above $H_c$ is interpreted as a Bose Einstein condensation of magnons. For the $S$ = 1 1D HAF, it is expected that the system shows the Tomonaga-Luttinger liquid behavior above $H_c$, when the magnetic field is applied along the symmetric axis. The low energy physics around the energy gap of the spin gap system has been studied extensively. But the low energy nature between $H_c$ and $H_s$ where $H_s$ is the saturation field has not been clearly understood. Therefore, in order to understand the physics above $H_c$, we performed multi-frequency and high-field ESR measurements on single crystals of the $S$ = 1 quasi 1D HAF compound $Ni(C_5H_{14}N_2)_2N_3(PF_6)$, abbreviated as NDMAP, in magnetic fields up to about 55 T and at frequencies up to about 2 THz. We observed several ESR signals above $H_c$ for each crystallographic axis and one or two of them survive in the high-field region above about 15 T. One mode approaches a paramagnetic resonance line at high-fields and the other mode broadly changes with magnetic fields. These modes fit well with the conventional antiferromagnetic resonance (AFMR) modes with easy-plane anisotropy. This result suggests that the spin-excitation modes above $H_c$ change from modes affected by quantum fluctuations to those expected for a conventional antiferromagnet because of a suppression of the quantum fluctuations by magnetic field. In addition, we studied angular dependence of spin excitations below 14 T. We found two set of resonance modes caused by two types of $Ni^{2+}$ chains in NDMAP. We analyzed these data by comparing with a phenomenological field theory (PFT). The experimental results blow about 12 T are well fitted by the PFT, but the fitting by the PFT above 12 T is not good. This must be caused by a gradual change from the modes affected by the quantum fluctuations to those explained by the conventional antiferromagnetic resonance theory.

(2) Recently, semiconductors doped with transition metals have been studied extensively because some of them are predicted to be room temperature ferromagnetic semiconductors. Gallium arsenide (GaAs) semiconductors doped with Mn ions show the ferromagnetism at low temperatures. They have been studied not only for spintronics devices but also for high power and high frequency devices such as high-mobility transistors. Gallium nitride (GaN) doped with transition ions is another candidate for this kind of new device. Recently, it was reported that a GaN this film doped with Mn ions shows the ferromagnetism above room temperature. Therefore, the study on GaN doped with transition ions becomes one of hot topics. In addition, substrates with high resistivity and high crystal quality are needed to compose semiconductor devices. It is known that GaN substrates doped with Fe ions have high resistivity. Recent developments of sample preparation techniques enable us to obtain the very high quality GaN substrate. Therefore, to obtain fundamental information of Fe ions in the GaN substrate, we studied a high quality and high resistive GaN thin film dope with Fe ions (GaN:Fe) by electron spin resonance (ESR) measurements with a conventional X-band apparatus and a home made Q-band equipment. Observed ESR signals were analyzed with a spin Hamiltonian given by considering the local symmetry of the Ga site ($C_{3v}$) and assuming that the $Fe^{3+}$ ions ($S$ = 5/2) are substituted for the $Ga^{3+}$ ions. As a result, the angular dependence of the resonance fields and temperature dependence of the signal intensities are reproduced very well by the calculations. In addition, we compared the obtained parameters with the reported ones which were obtained from unintentional doping samples. Consequently, we confirmed that the $Fe^{3+}$ ions occupy some of the Ga sites in the GaN thin film, and the obtained $g$-value is close to that of free $Fe^{3+}$ ion than that of unintentional samples. The difference of the $g$-value between the intentional doped sample and the unintentional one would be caused by the quality of the GaN crystals.