Optical fiber system for the high resolution resonant Raman spectroscopy at ³He temperature in a high magnetic field

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Abstract. A high-resolution resonant Raman spectroscopy system has been developed by using optical fibers and applied to the spin-flip Raman scattering experiment in CdZnTe/CdZnMnTe quantum wells at ³He temperature in a high magnetic field up to 14 T. Excitation light from an external cavity laser diode or Ti:sapphire laser was introduced into the liquid ³He chamber by using polarization maintain fiber, and was incident on the sample at the angle of 45 degree through a GRIN collimation lens and a prism to deflect the reflected light from the luminescence/scattering detection pass. Luminescence and scattered light were collected efficiently into the bundled multi-mode fibers through a high NA objective lens and introduced into the slit of a 1.26 m single grating spectrometer equipped with a multi-channel CCD detector and a photon counter. The fiber system provides a convenient way to investigate the spin dynamics in an ideal environments; ultra-low temperature and radiation-free condition. Furthermore, the system is more sensitive than the conventional system utilizing a cryostat with the optical windows where multiple Fresnel loss and the small numerical aperture degrade the sensitivity.

1. Introduction
Growing demand for the coherent spintronic devices provides a strong motivation for understanding the coherent evolution of spin states and spin-spin interactions in semiconductor nano-structures. Spin-flip Raman scattering experiment have been widely used to investigate the precession and relaxation of the spin states. However, in order to fully clarify the interplay between the free carrier spins and the magnetic ion spins or the nuclear spins, a high-sensitive and high-resolution spin-flip Raman experiment in ultra-low temperature is strongly desired.

We have developed a home-made resonant spin-flip Raman spectroscopy system at ³He temperature by utilizing optical fibers. Optical fiber enabled to insert the sample holder into the small bore of super-conducting solenoid, so Raman experiments in an ultra-low temperature and a high magnetic field exceeding 10 T could be realized by a rather compact facility. Furthermore, the thermal radiation which may disturb spin states was completely excluded. The use of polarization maintaining fiber in enabled the polarization analysis of Raman scatterings. Also the use of bundled multi-mode fibers in combination with high NA objective lens enabled a high sensitive detection of Raman scattering which is essential for the study of spin dynamics with minimal optical disturbance.
2. System setup
Whole experimental setup is shown in Fig. 1. Excitation light from an etalon-tuned Ti:sapphire ring laser or an external cavity diode laser was coupled into the polarization maintaining fiber (HB-750: Fibercore Limited) and was introduced into a home-made \(^3\)He cryostat with 19 mmφ inner bore, equipped with a 14T superconducting magnet. Luminescence and scattered light were collected by a pair of miniature lens into the bundle of 7 or 19 multi-mode fibers (AFS105/125Y, 105μm-core step-index fiber: Fiberguide Industries), and were introduced to the slit of a 1.26 m single-grating spectrometer (Spex 1269) equipped with a multi-channel CCD detector and a photon counter. Although the ultimate resolution of the spectrometer was 0.006 nm with 6 μm slit width, this could be attained only in the use of photon counter. Usually, we used a 1024ch-CCD detector with 25 μm pixels (Jobin Yvon) for the efficient measurement, so the actual resolution was limited to 0.02 nm (36 μeV). Acquiring the spectra in the mini-step mode interpolating the CCD pixel size we were able to obtain a smoothed spectrum and could determine the spectral peak with the accuracy of several μeV.

The line-widths of etalon-tuned Ti:sapphire ring laser and the home-made external cavity laser were well below the spectrometer resolution. External cavity laser was constructed in Littman configuration [1] by utilizing commercially available laser diodes (635nm: Sanyo-DL5038-021, 650nm: Sanyo-DL3147-060, 690nm: Hitachi-HL6738MG etc.) in the visible and infrared region. Typical tuning range of the external cavity laser was about 10 nm around the specified wavelength and additional temperature tuning in the range of -10 ~ 80 °C enabled 30 nm overall tuning. External cavity diode lasers are versatile, i.e., they cover the spectral region which is not covered by Ti:sapphire laser and can be easily combined with a high magnetic facility because of the high portability.

The sample holder made with a copper block was inserted in to the liquid \(^3\)He pot of 19 mmφ inner bore at the center of superconducting solenoid. Detailed structures of sample holders for Faraday and Voigt configurations are shown in the Fig. 2. The excitation light was collimated with a graded-index lens (Linos: 39 9713, p=0.25) and was incident on the sample at the angle of ~45 ° through a prism. The spot size of the excitation beam was around 100~200μm. The axis of polarization maintaining fiber was aligned against the reflection plane. Polarization of the launched field was rotated by the use of \(\lambda/2\) plate in front of the fiber coupler. Since the index of semiconductor is pretty high, 3~4, the optical transition is approximated by the normal incidence. We preferred the p-polarization incidence because of less reflection of the excitation light on the sample surface.

Luminescence and scattered light was collected with a near IR achromatic lens (f=7.5 mm, 5 mmφ) and then refocused on the facet of the bundled multimode fibers with a lens (f=15 mm, 5 mmφ). In
Voigt configuration, a polarization beam-splitter cube was used between the lenses in reflection geometry to analyze the polarization of the luminescence and the Raman scattering. Placing a stack of \(\lambda/4\) plate and linear polarizer between the lenses we can analyze the polarization in Faraday configuration. 7 multi-mode bundled fibers seemed to be enough to collect the Raman scattering. However, 19 bundled fibers system had a higher allowance for the optical alignment and seemed to be less sensitive to the bubbling of liquid \(^3\)He. The signal intensity decreased to about one half of the original when liquid \(^3\)He was immersed in the sample chamber. The sample was glued on the Cu sample holder with Apiezon® N grease and the temperature was measured by a RuO resister.

Figure 2. Sample holders for Voigt and Faraday configurations.

3. Some results
Resonant spin-flip Raman scattering experiment was carried out on Cd\(_{0.93}\)Zn\(_{0.07}\)Te/Cd\(_{0.48}\)Mn\(_{0.04}\)Zn\(_{0.48}\)Te quantum wells where a significant interplay between two-dimensional carriers in the well and the manganese spins in the barriers were expected [2]. Figure 2 shows the luminescence and Raman spectra of a 4 nm quantum well observed at 0.4 K under the excitation of three different photon energies. The spectra were observed at 100 \(\mu\)W excitation intensity, using a multi-channel CCD. Data acquisition time was 1 sec. and the spectral resolution is 0.02 nm (36 \(\mu\)eV).

Under the off-resonant excitation, the spectra presented an intact exciton luminescence line-shape with a low energy tail ascribed to the bound exciton. On the other hand, the spectra presented rich spectral features under the resonant excitations. At zero-field a hump on the low energy side of excitation laser line was observed in addition to the normal luminescence spectra. The hump became significant with decreasing excitation photon energy, where the exciton localization was expected. Furthermore, the hump increased with decreasing temperature. These reflect the exciton magnetic polaron effects observed in CdMnTe epitaxy layers by A. Gornik et.al [3] and G. Mackh et.al. [4].

Figure 3. Spectra of 4 nm CdZnTe/CdZnMnTe quantum well at 0.4 K in Voigt configuration.
In a magnetic field the hump adjacent to excitation laser line became sharp peaks ascribed to spin-flip Raman scattering as seen in the Fig. 3. At the excitation of the absorption peak of the heavy-hole exciton we observed 6 multiple Mn$^{2+}$ spin-flip Raman scatterings even in the non-magnetic quantum well [5]. The line width of first Stokes-line was about 50 $\mu$eV and increased slightly with magnetic field. Second and third stokes lines had larger line-widths and the values reached 100 $\mu$eV at 14 T. So, a high resolution analysis of Mn$^{2+}$ spin-flip line may reveal the spin dynamics of Mn$^{2+}$ ion through the broadening mechanism. At the excitation of the exciton-luminescence peak we observed conventional electron spin-flip Raman scattering of which energy is well described by a modified Brillouine function. The spin-flip Raman intensity strongly depended on the temperature. These effects strongly suggest that the spin-flip Raman scatterings strongly correlate with exciton localization effects.

Figure 4 shows the Raman spectra of 9 nm CdZnTe/CdZnMnTe quantum well at 0.4 K in Voigt configuration. In contrast to the adjacent 4 nm quantum well, Mn$^{2+}$ spin-flip Raman scattering was very weak and an unusual low-energy spin-flip Raman scattering was observed. The spin-flip energy seemed to be zero up to 4 T and then increased linearly with a magnetic field as shown in Fig. 4. Furthermore, the spin-flip energy increased with temperature. These unusual behaviors resembles “softening mode” of spin resonance observed in p-doped ferromagnetic CdMnTe quantum well [6].

![Figure 4. Spectra of 9 nm CdZnTe/CdZnMnTe quantum well at 0.4 K in Voigt configuration.](image)

High-sensitive and high-resolution spin-flip Raman scattering spectroscopy at a $^3$He temperature in a high magnetic field was carried out on CdZnTe/CdZnMnTe quantum wells and revealed some new aspects. Fiber system provides a convenient and excellent way to study the correlated spin states in the dilute magnetic semiconductors and the Quantum Hall States at ultra-low temperature. Furthermore, the optical fiber system will play an important role in the study of the high-coherence spin state of quantum dots in ultra-low temperature and thermal radiation-free environment.

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References
[1] Liu K and Littman G M 1981 *Optics Letters* 6 117
[2] Koudinov V A, Kusrayev G Yu, Zakharchenya P B, Wolverson D, Davies J J, Wojtowics T, Karczewski G, Kossut J 2003 *Phys. Rev. B* 67 115304
[3] Mack G, Ossaw W, Yakovlev R D, Waag A, Landwehr G, 1994 *Phys. Rev. B* 49 10248
[4] Gornik A, Ginter J, Gaj A J 1983 *J. Phys. C* 16 6073
[5] Stuhler J, Schaael G, Dakl M, Waag A, Landwehr G, 1995 *Phys. Rev. Lett.* 74 2576
[6] Scalbert D, Teppe F, Vladimirova M, Tatarenko S, Cibert J, Nawrocki M 2004 *Phys.Rev. B* 70 245304