Magneto-Optical Spectroscopy of Excitonic Fine-Structure in Diamond

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Introduction

Diamond is one of the group-IV semiconductors attracting increasing attention for the potential usage of the localized spin as a quantum qubit with an extremely long coherence time. However, the large energy gap (5.5 eV), deep impurity states, and difficulty in fabricating high-purity diamond crystals has hindered detailed studies on its intrinsic properties. The fundamental material parameters such as the values of g-factors and the magnitude of spin-orbit interaction in the valence bands are still controversial. In our previous investigation of photoluminescence under a magnetic field up to 10 T in NHMFL, the g-factor of the light-hole (LH) exciton was found to be 2.6 [1]. This leads to the Zeeman splitting of only 3 meV at 10 T, which is of the same order as the fine-structure splitting. Therefore, we have applied higher magnetic fields to reveal the effect of external perturbation on the excitonic fine-structure levels originating from other species of holes.

Experimental

We used the 25 T Split-Florida Helix to measure absorption spectra of ultrapure diamond at 10 K. The light source was a deuterium lamp (StellarNet Inc., SL3). The light transmitted through the sample mounted on the probe was spectrally resolved by a high-dispersion monochromator (f=750 mm, grating: 2400 grooves/mm) and detected by a charge-coupled device camera (Princeton Instruments, PyLoN). The absorption peaks due to excitonic fine-structure were analyzed by taking the derivatives of the spectra, and measured as a function of the field strength up to 23 T, applied along the 001 or 110 axis of the diamond crystal in Voigt geometry. Long exposure times such as 80 minutes were chosen to acquire sufficient signal to noise ratios.

Results and Discussion

Figure 1 shows the peak positions in the absorption spectra as a function of magnetic field. At zero field, we confirmed four-level splitting of the excitonic fine-structure [2]. We have observed clear splitting of the sublevels with applied magnetic fields. The g-factors of the heavy-hole (HH) exciton and the split-off (SO) hole exciton were found to be 0.75 and 1.4, respectively. The degeneracy of the sublevels agreed with the prediction as described in Ref. [2], verifying that the origin of the fine-structure splitting comes from the competition among electron-hole exchange, spin-orbit, and mass-anisotropy interactions. We also succeeded in detecting absorption due to the dark state of spin-triplet nature, which becomes weakly allowed under a high magnetic field. These observations made it possible to quantify the amplitudes of the above-mentioned three interactions.

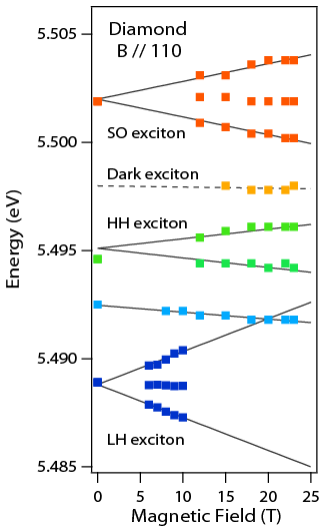

Fig.1 Field dependence of the excitonic fine-structure levels in ultrapure diamond at 10 K.

Conclusions

Our measurements of the excitonic fine-structure in diamond under high magnetic fields provided precise values of g-factors. Also, we clarified the spin singlet-triplet nature of the excitonic fine-structure levels. These results give a clue to elucidate the spin relaxation mechanism of delocalized carriers in intrinsic diamond and deepen the scientific aspect of diamond such as the few-body physics of recently found excitonic N-body bound states [3].

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