Summary of thesis:
Nuclear Magnetic Resonance Studies on Iron Chalcogenide FeSe

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In the thesis, I report the nuclear magnetic resonance (NMR) results on the iron chalcogenide FeSe. I have observed a suppression of antiferromagnetic (AFM) spin fluctuations and pseudogap (PG) behavior on the nuclear spin-lattice relaxation rate $1/T_1$, which are ascribed to the superconducting (SC) fluctuation effect around the critical temperature T_c. The results suggest the presence of preformed Cooper pairs associated with FeSe being in the BCS-BEC crossover regime, which was theoretically proposed[1, 3]. Besides, I have found that the SC diamagnetic shielding effect becomes weak in the field-induced SC phase (*B* phase), and suggest that the SC character would be changed at the *A*-*B* transition, although superconductivity persists beyond the *A*-*B* transition[2].

FeSe has the simplest structure and relatively low T_c ~ 9 K among the family of iron-based superconductors (FeSCs), but exhibits several distinct properties compared to other members. Especially, the extremely large ratio of SC gap to Fermi energy (Δ/E_F ~ 0.1) suggests that FeSe is in the Bardeen-Cooper-Schrieffer (BCS) – Bose-Einstein condensate (BEC) crossover regime. In this regime, preformed Cooper pairs were theoretically-predicted and can lead to an enhancement of SC fluctuations in the vicinity of T_c. Another interesting property in FeSe is the existence of magnetic field (H)-induced SC state (*B* phase) above a low-H phase (*A* phase), whose origin is still unknown. Since the above properties were only studied by macroscopic probes such as transport and thermodynamics measurements, we consider that microscopic probe of NMR measurement can give an important clue to the above properties in FeSe.

To achieve this goal, I have performed ^{77}Se-NMR measurements on single crystal sample from μ_0H = 0.5 to 19 T with temperature (T) down to 90 mK. From the results of $1/T_1T$ in various H, I found that the AFM spin fluctuations develop rapidly below the structural transition temperature T_s ~ 90 K. The enhancement of AFM spin fluctuations is H-independent in a wide T range down to T^*, but they start to be suppressed by H below T^*. In the range of $T_c(H) < T < T^*(H)$, the $1/T_1T$ curve exhibits a broad maximum at $T_p(H)$. This behavior is similar to the PG behavior in high-T_c cuprate superconductors, indicating that the AFM spin fluctuations are suppressed above T_c[1].

From the H-dependence of $1/T_1T$, I found that at T = 10 K, which is slightly above $T_c(0)$, $1/T_1T$ is strongly suppressed below 3 T, in contrast to the H-independent behavior of $1/T_1T$ above 20 K[1]. This behavior is consistent with the results of the magnetic torque measurement on FeSe, and can be explained in the framework of SC fluctuations [3]. Therefore, the SC fluctuation effect, which is related to the preformed pairs, is confirmed from the T- and H-dependence of $1/T_1T$. Based on these results, I suggest that the preformed pairs would exist in FeSe, and suppress AFM fluctuations below T^*.

Another anomaly of H-dependence of $1/T_1T$ is in the high-H region. At $T < 10$ K, $1/T_1T$ increases throughout $H_{c2}(T_c)$ with the increasing H, and exhibits a kink structure at H_{anom}, above which the increase of $1/T_1T$ is modest[1]. From the phase diagram of FeSe, the H_{anom} line seems to connect with the A-B transition line at $\mu_0H^* = 13.5$ T in the low-T region, thus it is interesting to investigate the SC properties in the B phase.

In order to investigate the B phase above H^*, I have performed ac susceptibility and ^{77}Se-NMR measurements in the field range from 12.5 T to 14.75 T below $T = 1.6$ K. I have found that superconductivity persists beyond the A-B transition line at H^* from the ac susceptibility measurement. From the decrease of broadening of spectrum linewidth arising from the SC diamagnetic effect, I suggest that the SC character is changed at H^*, and H^* is linked to H_{anom} above $T_c(H)$. Thus, I consider that the SC pairing state might be modified and the superconductivity in the B phase becomes weaker due to the reduction of Fermi surfaces.

I believe that these results provide deep insights into the unconventional properties in normal and SC states of FeSe.

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