The title compound (dineopentylbiferrocene)(fluorotetracyanoquinodimethane)$_3$ shows a very interesting ionic(I)-ionic(II) phase transition around 120 K. At high temperature the compound is in the monovalent state, while at low temperature it is in the divalent state. The magnetic property also changes drastically accompanied by this phase transition. At the high temperature phase, the donor molecule dineopentylbiferrocene has one spin ($S = 1/2$) coming from the low spin Fe$^{3+}$ ion and the acceptor which consists of fluorotetracyanoquinodimethane trimer also carries one spin ($S = 1/2$) as $\pi$ electron. On the other hand, at the low temperature phase the donor has two spins because two iron atoms of the donor molecule become Fe$^{3+}$ ion. Moreover, the acceptor become non magnetic because of the formation of the singlet pair of two $\pi$ electrons.

In order to investigate this phase transition from the microscopic point of view in detail, we performed X-band ESR measurements, and high-field and multi-frequency ESR measurements. In the X-band ESR measurements on the aligned single crystals, a broad single absorption line was observed at room temperature. We cannot observe the signals originated from the Fe$^{3+}$ ion and the $\pi$ electron separately, presumably due to the exchange interaction between them. The line width of this absorption line broadens as the temperature was decreased and it seems to disappear around the transition temperature. It suggests that the ESR signal becomes very broad or out of the range of this measurement condition (9.6 GHz, 0.6 T) at the low temperature phase. In the high-field and multi-frequency ESR measurements of powder sample at 1.9 K in the frequency region from 70 to 500 GHz, we have succeeded in observing the absorption lines due to Fe$^{3+}$ ions and found that the small energy gap of about 60 GHz (~ 3 K) opens at the zero field. This is consistent with the fact that the ESR signal cannot be observed in the X-band ESR measurements. The detailed analysis and discussion will be presented.