論 文 の 内 容 の 要 旨

論文題目　Terahertz spectroscopy of sublevel structures in single self-assembled InAs quantum dots
(単一自己組織化InAs量子ドットにおける量子準位構造のテラヘルツ分光に関する研究)

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In this thesis, we have investigated the intersublevel transition in single InAs quantum dots (QDs). Self-assembled InAs QDs are very attractive materials owing to their atom-like discrete energy levels. So far, terahertz (THz) intersublevel spectroscopy was mainly performed on ensembles of InAs QDs. For an introduction of this work, in Chapter 1, we first reviewed these important pioneering works. However, the THz spectra obtained by the conventional transmission measurements on ensembles of QDs are strongly affected by inhomogeneous broadening, hindering detailed discussions on the physics of intersublevel transitions. THz spectroscopy on single QDs is, therefore, highly desirable. The typical diameter of InAs QDs is about several tens nm and the energy separations between the zero-dimensional sublevels are typically 10-100 meV; the corresponding electromagnetic wavelength is about 12-120 μm. This large mismatch between the size of the QDs and the wavelength of the radiation gives extremely small scattering cross sections, making conventional transmission measurements on single QDs extremely difficult. As shown later, we used a single electron transistor (SET) geometry and successfully overcame this difficulty.

In Chapter 2, we describe the single electron tunneling process within the framework of the constant interaction model. How to obtain the information on the sublevel structures and the charging energies from the Coulomb stability diagram is presented. On the basis of this knowledge, we discuss the photoexcitation mechanism and the selection rule for the terahertz (THz) intersublevel transitions in the QD SET.

In Chapter 3, we first describe how to use the SET geometry to overcome the great mismatch between the size of the QDs and the wavelength of the THz electromagnetic wave. We used a solid immersion lens and the nanogaps electrodes integrated with a bowtie antenna to tightly focus the THz field on a single QD. By doing this, we can strongly enhance the effective THz field on a single QD. The other problem is that the THz absorption by a single QD is too small to be measured. We used the QD-SET itself as a THz detector. We detected the intersublevel transition as THz-induced photocurrent in the SET. Furthermore, we could vary the electron number in the QD by using the backgate.

Next, we describe the sample preparation. Particularly, we explain how to align the nanogap electrodes integrated with an antenna with a single QD. We used three different fabrication methods; i.e., random fabrication, selected fabrication, and QD mapping fabrication. The first method has an advantage of protecting the sample surfaces during the process. The second method has higher fabrication yield. The third allows us to select the individual dot and achieve a very high fabrication yield.

In Chapter 4, we presented the results of the THz photocurrent measurements. When the QD-SET sample was illuminated by a wideband THz radiation, an electron in the lower energy states absorbs a THz photon and makes a transition to the upper energy state and, then, tunnels out to the electrodes. When an electron in the electrodes tunnels into the lower empty state, the QD-SET returns to its initial state. We call this process "N↔N-1 excitation". However, the photocurrent distribution with respect to the Coulomb diamonds indicates that there is another mechanism for the photocurrent generation, which allows the photocurrent to flow even when the upper energy state is below the Fermi level of the electrodes. The total energy calculation shows that, after one electron is photoexcited to an upper sublevel, another electron in the electrode can subsequently tunnel into the lower empty state. Once the lower empty energy state is filled by another electron, all the energy levels are pushed up by E_C and the photoexcited electron goes above the Fermi levels and tunnels out, producing a photocurrent. During this photoexcitation process, the electron number changes between N and N+1. Thus, we call this process the "N↔N+1 excitation", which agrees with the observed photocurrent distribution. For deeper understanding of the photocurrent generation process, we performed numerical calculations with rate equations and found critical conditions for efficient photocurrent generation.

In Chapter 5, we presented the intersublevel transition spectra measured in QD-SETs within the few-electron regime. We observed sharp photocurrent peaks, whose linewidths are consistent with the tunnel coupling with the electrodes. When the p shell is fully occupied, we observed rather simple photocurrent spectra induced by the p→d-2s shell intersublevel transitions. The selection rule for the intersublevel transition predicts three intersublevel transitions; namely, $p_-\rightarrow d_-$, $p_-\rightarrow 2s$, and $p_+\rightarrow d_+$, recalling the fact that the THz electric field is polarized along the direction of the nanogap electrodes. Having this in mind, we decomposed the observed photocurrent peak into three peaks ($p_-\rightarrow d_-$, $p_-\rightarrow 2s$, and $p_+\rightarrow d_+$) by numerical fitting. The intensity change of the photocurrent peaks with V_G qualitatively agrees with the numerical calculation that takes into account the electron-electron Coulomb repulsion. However, when the p shell is half filled, the photocurrent spectra exhibited rather complicated behavior as a function the gate voltage, most likely due to the fluctuation in the electron configuration when the empty p state is filled back from the electrode.

High temperature operation of QD devices is of great interest for device applications. We measured the temperature dependence of the intersublevel transitions spectra. Surprisingly, even at 150 K, we observed a photocurrent peak at sublevel spacing of about 13 meV. At lower temperatures (<30 K), the linewidth shows a weak temperature dependence, while at higher temperatures (30-150 K), the linewidth is found to increase rapidly with increasing temperature.

In Chapter 6, we focused on manybody effects in the intersublevel transitions in QDs. We perfomed spectroscopy on intersublevel transitions in single InAs quantum dashes (QDHs). InAs QDHs are grown on (211)B-oriented GaAs substrates, and elongated along [011] direction. The typical length of the InAs QDHs is 100-500 nm. The QDH SET that we measured was in the many-electron regime, and the Coulomb stability diagram showed the sublevel spacing in the order of 3-5 meV. We observed a large photocurrent about 10 pA and sharp photocurrent peaks. The photocurrent spectra showed peaks at around 15 meV, which are much larger than the excited state energy obtained from transport measurements. This trend qualitatively agrees with our previous result obtained for the QD SETs and the depolarization shift is most likely the main contribution to this discrepancy.

In the last chapter of this dissertation, we summarized the whole work in this thesis and also gave a remark on future prospects. We systematically studied the intersublevel transition spectra in single self-assembled QDs. We think the knowledge we have obtained from this research will be useful for both developing devices and understanding fundamental physics. The technology we developed for the coupling between THz wave and single nm-scale structures may also be applied to other fields, such as nano-chemistry, pharmaceutical science, and even molecular biology.