NQR and Low-Field NMR of $^{181}$Ta in the Low-Temperature CDW State of 2H-TaS$_2$

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NQR and low-field NMR spectra of $^{181}$Ta are reported to probe the electronic environment of inequivalent tantalum sites in the low-temperature CDW state of a layered compound 2H-TaS$_2$. The overall features are found to be consistent with the structure consisting of nine-atom snowflake clusters in a locally commensurate CDW state.

Key words: NQR, NMR, $^{181}$Ta, CDW, TaS$_2$.

Transition metal dichalcogenides have been the subject of intensive research because of their electronic instabilities which result in charge density waves (CDW) [1]. The 2H-polytype of TaS$_2$ undergoes a phase transition at 75 K from the high-temperature normal metallic state to the incommensurate CDW phase transition at 75 K. The quadrupole moment of $^{181}$Ta ($I = 7/2$) is very large ($Q = 3.28$ barn), and NQR of $^{181}$Ta was found to be useful for the study of the local electronic environments around Ta nuclei, as was demonstrated in a case of 1T-TaS$_2$ [3]. A pulsed NQR spectrum of $^{181}$Ta in 2H-TaS$_2$ at 4.2 K was taken using an incoherent high-power rf oscillator and a highly sensitive receiving system. A somewhat complex NQR spectrum of $^{181}$Ta has been obtained at 4.2 K as shown in Fig. 1, with clear peaks at frequencies of 95.5, 140, 190.2, 194.0 MHz and weak ones at 105.3 and 284.6 MHz. The peak positions of the spectrum are reliable but relative intensities should be considered to be only qualitative. A single Ta site creates three NQR transitions of $\pm 1/2 \leftrightarrow \pm 3/2$, $\pm 3/2 \leftrightarrow \pm 5/2$ and $\pm 5/2 \leftrightarrow \pm 7/2$, and the ratios of the three frequencies are determined by the asymmetry parameter $\eta$ as

$$\eta = (V_{xx} - V_{yy})/V_{zz},$$

where $V_{zz}$, $V_{yy}$, and $V_{xx}$ are the principal values of electric field gradient (EFG) tensor at the resonant nuclei, and $|V_{zz}| \geq |V_{yy}| \geq |V_{xx}|$.

If the assignment for the three transitions is done, the quadrupole frequency

$$\nu_0 = 3e^2qQ/[2I(2I-1)\hbar]$$

can be determined. The peak frequencies of 95.5, 190.2 and 284.6 are in the ratio of nearly 1:2:3, indicating the existence of a Ta site with nearly axial symmetry ($\eta = 0.03$). But the assignment of the other peaks in Fig. 1 to the NQR transitions is not unique due to possibilities that some transitions are overlapping and that some transitions (probably $\pm 5/2 \leftrightarrow \pm 7/2$) are missing. Therefore, we analyze our data with the help of low-field NMR and TDPAC experiments, as is described below.

The powder spectra of $^{181}$Ta NMR in a swept magnetic field for the transitions between degenerate NQR states $\pm m$ have shown to be a good method to give values of $\eta$ of the EFG independent of the magnitude of the EFG (or the magnitude of the quadrupole frequency) if the NMR frequency is much smaller than $\nu_0$ [4]. The signals from the $\pm 1/2$ transitions are ex-
Table 1. Assignment of the peak frequencies (MHz) in Fig. 1 to the NQR transitions and derived values of $\eta$ and $v_0$ for the three Ta sites in 2H-TaS$_2$ at 4.2 K.

<table>
<thead>
<tr>
<th>Ta site</th>
<th>$A$</th>
<th>$B$</th>
<th>$C$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$f (\pm 1/2 \leftrightarrow \pm 3/2)$</td>
<td>105.3</td>
<td>105.3</td>
<td>95.5</td>
</tr>
<tr>
<td>$f (\pm 3/2 \leftrightarrow \pm 5/2)$</td>
<td>140.0</td>
<td>194.0</td>
<td>190.2</td>
</tr>
<tr>
<td>$f (\pm 5/2 \leftrightarrow \pm 7/2)$</td>
<td>(221)*</td>
<td>(293)*</td>
<td>284.6</td>
</tr>
<tr>
<td>$\eta$</td>
<td>0.37±0.02</td>
<td>0.15±0.02</td>
<td>0.03±0.03</td>
</tr>
<tr>
<td>$v_0$ (MHz)</td>
<td>75.2±0.5</td>
<td>98.4±0.5</td>
<td>95.5±0.2</td>
</tr>
</tbody>
</table>

* Signals at frequencies with parentheses were not observed.

In summary, the overall features of the NQR and low-field NMR spectra are consistent with the nine-atom snowflake cluster model proposed from the TDPAC experiments. Three inequivalent Ta sites have definite values of the quadrupole parameters in spite of the incommensurate CDW state observed by differ-
fraction experiments. The signals must come from locally commensurate regions separated by regions of phase slip, confirming again the concept of discommensurations.

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