Thomson scattering from the antiferroquadrupolar ordering phase in TmTe

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Abstract. On the basis of the recently established crystal electric field level scheme of $\Gamma_8 - \Gamma_6 - \Gamma_7$, we investigate the Thomson scattering intensity of the superstructure reflections $G = (\frac{2\pi}{a})(h, k, \ell)$ with $h, k, \ell = 2n + 1$ in the $\Gamma_3$-type ($O_{yz}, O_{zx}, O_{xy}$) antiferroquadrupolar (AFQ) ordering phase of TmTe. The intensities show strong dependence on $G$ and the direction of the applied magnetic field $H$. These dependences on $G$ and $H$ similar to those found in the AFQ phase of CeB$_6$, another AFQ material with $\Gamma_5$-type ($O_{yz}, O_{zx}, O_{xy}$) order parameter having the $\Gamma_8$ ground states, may provide us a useful tool to distinguish the component of the AFQ order of $\Gamma_3$-type and $\Gamma_5$-type.

1. Introduction
Thulium telluride (TmTe) is a magnetic semiconductor crystallizing in the rocksalt structure ($Fm\overline{3}m$) with the lattice constant being equal to 6.354 Å. This material is considered to show an antiferroquadrupolar (AFQ) order below $T_Q \approx 1.8$ K[1] and, then, an antiferromagnetic (AFM) order below $T_N \approx 0.43$ K. The thulium monochalcogenide family usually shows valence instability of Tm. The Tm ion in this material, however, is divalent in an ideal crystal.[2] Then, the ground multiplet becomes $^2F_2$ in the configuration $(4f)^{13}$. Under a cubic point symmetry ($Fm\overline{3}m$), the multiplet splits into two doublets $\Gamma_6, \Gamma_7$ and one quartet $\Gamma_8$. Since the overall level splitting does not exceed 15 K, we should include full multiplets into the analysis. Among the basic properties which have not achieve the consensus yet, the crystal electric field (CEF) level scheme is one of the most urgent issues to be addressed. Clementyev et al. performed inelastic neutron scattering measurements and proposed their data were well fitted by means of the CEF level schemes $\Gamma_8(0) - \Gamma_7(4.6K) - \Gamma_6(10.7K)$ or $\Gamma_8(0) - \Gamma_6(4.9K) - \Gamma_7(11.1K)$.[3] One the other hand, some claimed the ground multiplet might be $\Gamma_7$.[4]

Recently, through the investigation based on the magnetic phase diagram, the present author’s group has demonstrated that the level scheme $\Gamma_8 - \Gamma_6 - \Gamma_7$ is the most promising candidate of the CEF level scheme.[5] Their analysis has shown that the component of the AFQ order parameter is $O_2^0$ in zero field while in the presence of the applied field, their components depend on the direction of the field. The $O_2^0$ and $O_2^1$ states are stabilized by the field along [001] and [110] directions, respectively. These findings are consistent with the results predicted by the mean field analysis on a classical model[6, 7] and those confirmed by the neutron diffraction experiments.[8, 9, 10] As a possible direct observation of the AFQ order, we have shown that the azimuthal angle dependence of the resonant x-ray scattering intensity at the Tm $M_5$ edge can be employed to identify the order parameter and CEF.[5]
In this paper, we investigate the Thomson scattering intensity from the AFQ phase with the $\Gamma_3$-type ($O_{2h}^3, O_{2h}^5$) of TmTe assuming the $\Gamma_8 - \Gamma_6 - \Gamma_7$ level scheme and using the model Hamiltonian defined in Ref. [5]. We show that its dependence on the scattering vector and the applied magnetic field exhibits characteristic behavior and that the situation is similar to that found in another AFQ phase material with the $\Gamma_5$-type ($O_{yz}, O_{xz}, O_{xy}$) order of CeB$_6$.[11, 12, 13, 14, 15] Although both materials experience the AFQ phase with $\Gamma_8$ ground states under the cubic symmetry, we discuss that the difference of the component of the AFQ order may be distinguished by investigating the Thomson scattering data.

Sections 2 and 3 report the theoretical preparation in evaluating the Thomson scattering intensity and our findings for it from the AFQ phase in TmTe, respectively. A comparison between the results for TmTe and those for CeB$_6$ will be shown in section 3, too. A summary is given in section 4.

2. Theoretical framework
The Thomson scattering amplitude measured in units of the classical electron radius $r_0$ at the $j$-site $\mathbf{R}_j$ is written as

$$f_j = (\epsilon' \cdot \epsilon) e^{i \mathbf{G} \cdot \mathbf{R}_j} \langle \psi_j | e^{i \mathbf{G} \cdot \mathbf{r}} | \psi_j \rangle,$$

(1)

where $\mathbf{G}$ is the scattering vector. The polarizations of the incident and scattered photons are denoted as $\epsilon$ and $\epsilon'$, respectively. We restrict our interest in the $\sigma - \sigma'$ channel corresponding to $\epsilon' \cdot \epsilon = 1$. The eigenstate is represented by $| \psi_j \rangle$ which is expanded as $\sum |\ell_z, s_z, \frac{3}{2} \rangle \langle \ell_z, s_z, \frac{3}{2}| \psi_j \rangle$ within the space spanned by the total angular moment $J = \frac{7}{2}$. With the help of Rayleigh expansion, we can express the matrix element as

$$\langle \ell_z s_z | e^i \mathbf{G} \cdot \mathbf{r} | \ell'_z s'_z \rangle = \delta_{s_z, s'_z} \sqrt{4\pi} \sum_{\ell = 0}^{\infty} \ell' \sqrt{2\ell + 1} |j_{\ell'}\rangle Y_{\ell, \ell_z}^\ast (\theta_G, \varphi_G) \epsilon' (3\ell_z; 3\ell'_z),$$

(2)

$$|j_{\ell'}\rangle = \int_0^\infty r^2 R_{4f}^2 (r) j_{\ell'} (|\mathbf{G}|r)|dr,$$

(3)

where $j_{\ell'}$ is a spherical Bessel function of order $\ell$. Radial part of the $4f$ wave function is denoted as $R_{4f}(r)$, which is evaluated by the Hartree-Fock approximation in the following calculation.[16]

The Gaunt coefficient is represented as $\epsilon' (3\ell_z; 3\ell'_z)$. The arguments $\theta_G$ and $\varphi_G$ of the spherical harmonics $Y_{\ell, \ell_z}$ mean the spherical coordinate of $\mathbf{G}$.

The eigenstates are evaluated by solving the model Hamiltonian defined in Eq. (1) of Ref. [5] which includes the CEF term, the Zeeman term, and the terms describing the multipole interactions. The parameters needed to define the Hamiltonian are chosen as the same values as those used in Ref. [5]. As for the applied magnetic field, we consider they are along [001] and [110] directions with $|\mathbf{H}| = 4$ T.

3. Numerical results
In the fcc structure, four $K$-domains are given rise to with the magnetic modulation vectors $\mathbf{K}_1 = (1, \frac{1}{2}, \frac{1}{2}, \frac{1}{2})$, $\mathbf{K}_2 = (\frac{1}{2}, 1, \frac{1}{2}, \frac{1}{2})$, $\mathbf{K}_3 = (\frac{1}{2}, \frac{1}{2}, 1, \frac{1}{2})$, and $\mathbf{K}_4 = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2}, 1)$ where $K_i$ are measured in units of $\frac{2\pi}{a}$. Different $K$-domains are connected to the different magnetic Bragg reflections in the fcc structure. For instance, the intensities from $\mathbf{G} = (\frac{2n+1}{2}, \frac{2n+1}{2}, \frac{2n+1}{2})$ and $(\frac{2n+1}{2}, \frac{2n+3}{2}, \frac{2n+3}{2})$ are brought about by $\mathbf{K}_1$ and $\mathbf{K}_2$-domains, respectively. In the absence of the applied magnetic field, each $K$-domain has three $S$-domains where the choices of the quantization axis are different. The one is defined by the crystal $z$-axis as a quantization axis of $J$. We denote it as $S_1$-domain. The other two, named as $S_2$ and $S_3$, are obtained by rotating $S_1$ through angles $\pm 120^\circ$ about the modulation vector $\mathbf{K}_i$ in each $K$-domain. In zero field, all the three $S$-domains
contributes independently. On the other hand, in the presence of the applied field only \( K \)-domain consideration is needed. In the present treatment, we expect every domain is equally populated.

First, we summarize the results for a series of \( G = \left( \frac{2n+1}{2}, \frac{2n+1}{2}, \frac{2n+1}{2} \right) \). No intensities are detected in zero field and \( H || [110] \) due to the symmetrical reason. When the magnetic field is applied along [001] direction, however, relatively weak none-zero intensity is observed with a single-peak structure as a function of \( n \). Its maximum occurs around \( n = 9 \) and the dominant and secondary contributions are from the terms proportional to \( \langle j_4 \rangle \) and \( \langle j_0 \rangle \), respectively, in Eq. (2).

As for the detectability, it would be better to turn our attention to another series of \( G \) which gives larger intensities than that of \( \left( \frac{2n+1}{2}, \frac{2n+1}{2}, \frac{2n+1}{2} \right) \). Figure 1 shows the results of Thomson scattering intensities for \( G \) being a series of \( \left( \frac{2n+1}{2}, \frac{2n+3}{2}, \frac{2n+3}{2} \right) \). Intensities are measured in units of \( r_0^2 \). In this series of \( G \), the intensities in zero field and \( H || [110] \) exhibit similar behavior as a function of \( n \) as shown in Fig. 1 (a) and (c). After achieving the maximum around \( n = 2 \) or 3, the intensities decay rapidly when \( |G| \) exceeds \( 1 \ \text{Å}^{-1} \) since the dominant contribution is due to the term proportional to \( \langle j_2 \rangle \). It seems reasonable since the primary order parameter is \( O_2^2 \) in both cases. The qualitative difference may be attributed to the induced dipole and octupole moments emerged in the presence of the applied field. When the field is applied in the [001] direction, the intensity shows two-peak structure as a function of \( n \) as shown in Fig. 1 (b). Such a characteristic behavior may be easy to detect in an actual experiment. The intensity includes a considerable contribution from the term proportional to \( \langle j_2 \rangle \) which is absent for \( \left( \frac{2n+1}{2}, \frac{2n+1}{2}, \frac{2n+1}{2} \right) \).

![Figure 1. Thomson scattering intensities from TmTe with \( G = \left( \frac{2n+1}{2}, \frac{2n+1}{2}, \frac{2n+1}{2} \right) \) with \( n = 1 \sim 7 \). (a) zero field, (b) \( H || [001] \), and (c) \( H || [110] \). Intensities are measured in units of \( r_0^2 \). Symbols \( \circ \), \( \bullet \), and \( \times \) denote the full intensity, the terms proportional to \( \langle j_2 \rangle^2 \) and \( \langle j_4 \rangle^2 \), respectively.](image-url)

It would be worthwhile to compare the present results with those of CeB\(_6\) in the AFQ phase (phase II).[11, 17] In CeB\(_6\), the ground multiplet is also the \( \Gamma_8 \) quartet with \( J = \frac{5}{2} \) but is widely isolated (540 K). A notable difference between TmTe and CeB\(_6\) relevant to the behavior of the Thomson scattering intensity is that the AFQ order is believed to be of \( \Gamma_3 \)-type \( (O_2^2, O_0^2) \) and of \( \Gamma_5 \)-type \( (O_{yz}, O_{xx}, O_{yy}) \), respectively. Kono et al. obtained zero intensity for CeB\(_6\) in zero field with \( G = \left( \frac{2n+1}{2}, \frac{2n+1}{2}, \frac{2n+1}{2} \right) \).[15] This is the same as our present case of TmTe. On the other hand, a prominent difference is found from the work by Yakhou et al.[11] The relative intensities of Thomson scattering in the AFQ phase of CeB\(_6\) in the absence of the magnetic field at the superlattice spots \( G = \left( \frac{5}{7}, \frac{3}{7}, \frac{3}{7} \right), \left( \frac{5}{7}, \frac{1}{7}, \frac{1}{7} \right), \) and \( \left( \frac{7}{14}, \frac{7}{14}, \frac{7}{14} \right) \) were listed as about 1, 0.02, and 0.02, respectively. The corresponding values in the present case of TmTe are 1, 0.08, and 20.3, respectively. Note that these values are obtained by multiplying a geometrical factor \( \cos(2\theta_B) \) in the \( \tau - \pi' \) channel where \( \theta_B \) is the Bragg angle. There are several candidates for the origin of this huge difference of the relative intensity at \( \left( \frac{7}{14}, \frac{7}{14}, \frac{7}{14} \right) \). For instance, it may be due
to the difference of the values of $J$, the inclusion or exclusion of the excited states $\Gamma_{7,6}$, and the difference of the $\Gamma_3$-type and $\Gamma_5$-type components. Although we cannot specify a single reason at the present, it is safe to say that such a huge difference does not disappear unless the values of the parameters might be modified considerably.

4. Summary

On the basis of the CEF scheme $\Gamma_8 - \Gamma_6 - \Gamma_7$ spanned by $J = \frac{2}{3}$, we have investigated the Thomson scattering intensities of TmTe in the AFQ phase. We have demonstrated that the intensity exhibits characteristic dependence on the scattering vector and the direction of the applied magnetic field. We have considered three case; (i) zero field, (ii) $\mathbf{H} \parallel [001]$, and (iii) $\mathbf{H} \parallel [110]$. The primary order parameters of cases (i) and (iii) are $O_{\beta}^{2}$ while that of case (ii) is $O_{\alpha}^{2}$. Reflecting this difference, intensity of case (ii) shows different dependence on $\mathbf{G}$ compared to those of cases (i) and (iii), which are similar to each other.

The strong dependence of the Thomson scattering intensity on the direction of $\mathbf{G}$ similar to found in the present work are also known for CeB$_6$ in the AFQ phase.[12, 17] By comparing the present results and those of CeB$_6$, we can conclude that the experimental observation of Thomson scattering in TmTe has realistic possibility. If we pay attention to the difference of the relative intensities of Thomson scattering at superlattice reflections between TmTe and CeB$_6$ for the appropriate $\mathbf{G}$ spots, we may get a useful clue to distinguish the component of the AFQ order of $\Gamma_3$-type ($O_{\beta}^{2}, O_{\alpha}^{2}$) and $\Gamma_5$-type ($O_{yz}, O_{zx}, O_{xy}$) realized in TmTe and CeB$_6$, respectively.

The Thomson scattering experiment on TmTe in the AFQ phase may clarify the situation.

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