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Diffuse Neutron Scattering from GaAs

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Diffuse neutron scattering from powder sample of GaAs was measured using a neutron diffractometer. Numerical calculations of the diffuse background have been made based on the crystal structure of GaAs including the correlation between the thermal displacements of atoms. The main contribution to the observed oscillatory diffuse scattering is from the correlation between the thermal displacements of gallium and arsenic atoms.

KEYWORDS: diffuse scattering, thermal vibration, GaAs, neutron diffraction

§1. Introduction

Anomalously strong and oscillatory diffuse scattering from α-AgI type solid electrolytes has been studied in X-ray and neutron scattering experiments.[1)] The theoretical treatment including the disordered arrangements of atoms and the correlations among the thermal displacements of atoms has been succeeded in explaining the observed diffuse scattering of the solid electrolytes. In the case of ordered solid electrolyte crystals, CuI and AgBr, the temperature dependence of the oscillatory diffuse scattering was mostly explained in termes of the correlations among the thermal displacements of atoms.[2)]

The correlation effect between the thermal displacements of atoms would not be specific in solid electrolytes. We estimate the oscillatory diffuse scattering even from normal crystals due to the thermal correlation. The intensity of the oscillatory diffuse scattering would show clear temperature dependence; the lower the temperature is, the weaker becomes the intensity. In order to measure the oscillatory diffuse scattering in semiconductor, neutron diffraction measurement of ordered crystal GaAs is performed.

In this paper, the diffuse neutron background intensity of GaAs has been measured at 8 K and 290 K. Numerical calculations of the diffuse background have been made based on the crystal structure of GaAs including the correlation between the thermal displacements of atoms.

§2. Experimental

Neutron scattering measurements were carried out for a powder GaAs (99.9999%, Furuuchi Chemical) in a cryostat at 8 K and 290 K with the use of a triple-axis type spectrometer TAS-2 installed at JRR-3M in Japan Atomic Energy Research Institute. The GaAs powder sample was set in a vanadium container with 8 mm in diameter. The incident neutron wave length of 1.436 Å was used. Neutron diffraction data were collected for 60 sec. per step at 0.1° intervals over the 2θ range of 20° to 118° by a step-scan mode. Figure 1 shows the powder diffraction intensities of GaAs at 8 K and 290 K. Several sharp Bragg lines and a large diffuse scattering were observed at 290 K. The first and second peak of the oscillatory diffuse scattering appear at $2\theta \sim 45°$ and 85°, respectively. The intensity of the first peak of the oscillatory diffuse scattering is weaker than that of the second peak in the measurement. The oscillatory characteristic in the diffuse scattering of GaAs at 8 K is not clear. The clear temperature dependence of the oscillatory diffuse scattering from GaAs crystals, which was predicted with the correlation effect, was confirmed by the measurement.

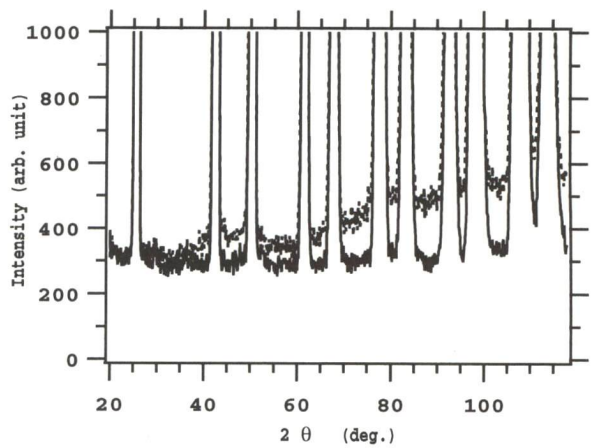

Fig. 1. Observed neutron powder diffraction intensity for GaAs. The solid lines are observed intensities at 8 K and the broken lines observed intensities at 290 K.

§3. Analysis and Discussion

Rietveld refinements of the neutron scattering data of GaAs with RIETAN-94[3,4)] by the background function including the correlation between the thermal displacements of atoms in place of Legendre polynomials have been carried out. The background function[1,5)] is shown

as follows;

$$I_B = k[b_{Ga}b^*_{Ga}\{1-\exp(-2M_{Ga})\} + b_{As}b^*_{As}\{1-\exp(-2M_{As})\}] + k(b_{Ga}b^*_{As} + b_{As}b^*_{Ga}) \times[\exp\{-(M_{Ga}+M_{As})(1-\lambda_{Ga-As})\} - \exp\{-(M_{Ga}+M_{As})\}]Z\sin(Qr)/Qr + k(\sigma^{Inc}_{Ga} + \sigma^{Inc}_{As}) + c_B \quad (1)$$

where $\exp(-M_i)$ $(= \exp(-\frac{1}{2} < (\vec{Q}\cdot\Delta\vec{r}_i)^2 >) = \exp\{-B_i(\sin\theta/\lambda)^2\})$ is the Debye-Waller factor of the i atom. $\Delta\vec{r}_i$ is the displacement caused by thermal vibration. b is the neutron scattering length, k a scale factor depending on the experimental conditions, Z the number of nearest neighboring sites. σ^{Inc} is the incoherent scattering cross section. The constant c_B was added for the corrections of electrical noise, the background by cosmic rays, etc. λ_{Ga-As} and r correspond to the value of the correlation between the thermal displacements of nearest-neighboring gallium and arsenic atoms and the atomic distance between these atoms, respectively. The correlation among the thermal displacements of atoms λ_{Ga-As} is written as follows;

$$\lambda_{Ga-As} = 2 < \Delta\vec{r}_{Ga}\cdot\Delta\vec{r}_{As} > \div\{< (\Delta\vec{r}_{Ga})^2 > + < (\Delta\vec{r}_{As})^2 >\} \quad (2)$$

The values of the correlation among the displacements of Ga and As atoms are 0 and $2\sqrt{B_{Ga}B_{As}}/(B_{Ga}+B_{As})$ in the case of no correlations among thermal displacements and the perfect correlation, respectively. The values of the correlation between the thermal displacements of atoms are assumed to be 0 except the nearest-neighboring sites. Crystals of GaAs are assumed to belong to zinc blende type structure with the space group $F\bar{4}3m$. Gallium and arsenic atoms occupy 4 (b) and 4 (a) sites, respectively. The number of nearest neighboring sites Z is 4 in the zinc blende type structure. Six parameters $(k, M_{Ga}, M_{As}, \lambda_{Ga-As}, r, c_B)$ in background functions are used in eq. (1), where thermal parameters M_{Ga} and M_{As} are same as those for the treatment of Bragg reflections.

Table I. Final refined parameters and the obtained R factors at 8 K and 290 K.

Parameters	8K	290K
a (Å)	5.6472(8)	5.6536(7)
λ_{Ga-As}	0.65(2)	0.62(1)
B_{Ga} (Å^2)	0.01(35)	0.45(64)
B_{As} (Å^2)	0.24(25)	0.67(69)
R_{wp} (%)	5.6	5.2
R_p (%)	4.1	3.7
R_e (%)	2.7	2.8
R_I (%)	1.1	1.0
R_F (%)	0.6	0.7

The observed and calculated neutron diffraction intensities at 290 K and 8 K are shown in Figs. 2 and 3, respectively. The thick solid lines in Figs. 2 and 3 show the calculated diffuse background intensity. It is found that the observed scattering intensities are well explained by the model including the thermal diffuse scattering in the eq. (1). The derived thermal parameters and lattice constants at 8 K and 290 K in Rietveld refinements are shown in Table I. The value of thermal parameter B_{As} is extremely greater than that of B_{Ga} at 8 K. However, the value of B_{As} is almost equal to that of B_{Ga} at 290 K.[6] The standard deviations of the thermal parameters are very large in the calculation. This might be occurred by a relatively small number of independent Bragg reflections. The distance between nearest-neighboring gallium and arsenic atoms r is almost equal to $\sqrt{3}a/4$, where a is the lattice constant.

The values for 2θ of the first and second peak in diffuse scattering of GaAs in Fig. 1 are greater than those of super ionic conductor CuI which belongs to zinc blende type structure.[2] They are related to the reciprocal of the distance between nearest-neighboring atoms; the distances in CuI and GaAs are 2.60 and 2.45 Å at 290 K, respectively. The main contribution to the oscillatory

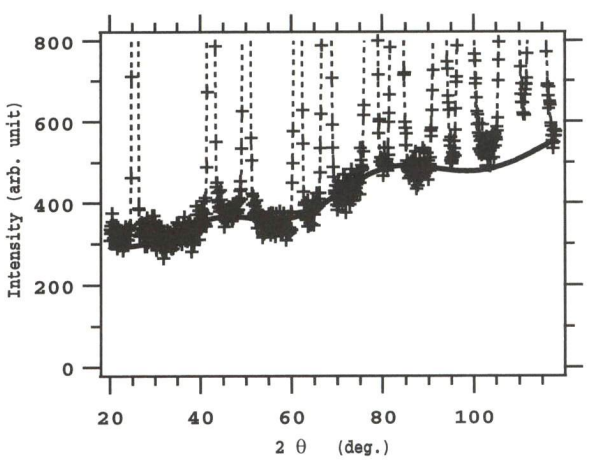

Fig. 2. Rietveld refinements patterns for GaAs at 290 K by the neutron diffraction measurement. The solid lines are calculated diffuse scattering intensity, the crosses (+) observed intensities and the broken lines calculated intensity.

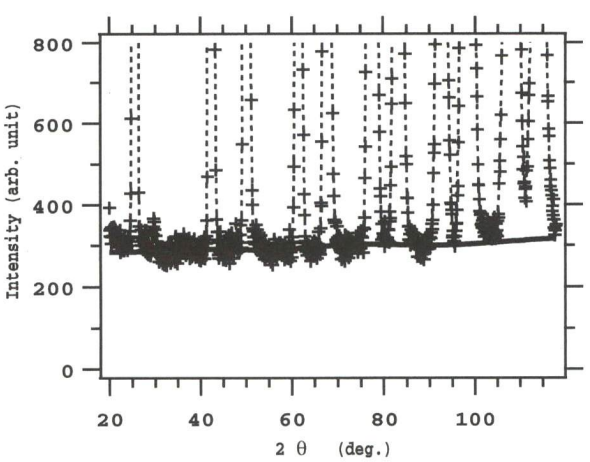

Fig. 3. Rietveld refinements patterns for GaAs at 8 K by the neutron diffraction measurement. The solid lines are calculated diffuse scattering intensity, the crosses (+) observed intensities and the broken lines calculated intensity.

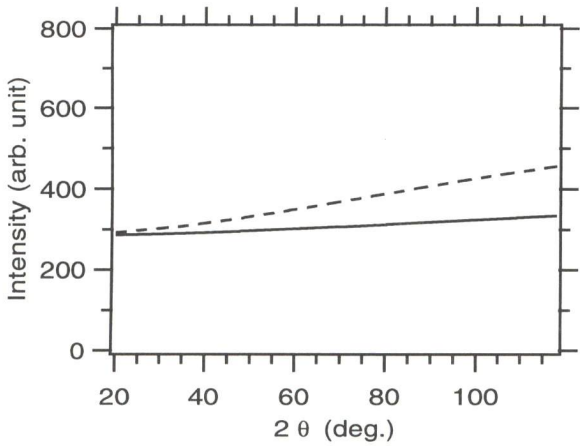

Fig. 4. Calculated diffuse scattering intensity at 8 K (solid lines) and 290 K (broken lines) based on the approximation that the atoms vibrate independently.

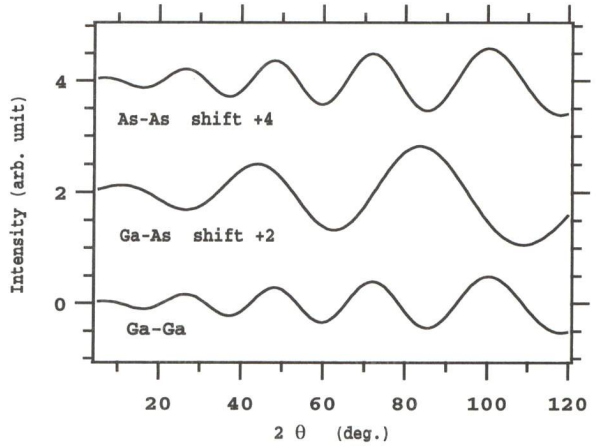

Fig. 5. Expected intensities from the contributions of Ga-Ga, Ga-As and As-As pairs to the diffuse scattering. The value of the correlation λ_{A-B}(A, B=Ga, As) was assumed to be 0.65.

diffuse scattering of super ionic conductors was from the correlation among the thermal displacements of cation and anion. It is suggested that the main contribution of GaAs would be from the correlation among the displacements of gallium and arsenic atoms.

When we assume λ=0 in eq. (1) where the atoms vibrate independently, the calculated intensity of the diffuse scattering is shown in Fig. 4. It is found that the major part of the observed diffuse scattering intensity in Fig. 1 could be explained by the assumption. However, the remaining weak oscillatory part in the observed diffuse scattering intensity can not be reproduced in Fig. 4. The second term of the right hand side in eq. (1) shows the oscillating scheme in the diffuse scattering. Expected contributions from Ga-Ga, Ga-As and As-As pairs to the diffuse scattering were calculated as follows;

$$I^{\mathrm{OSC}}_{\mathrm{A-B}} = (b_{\mathrm{A}} b^*_{\mathrm{B}} + b_{\mathrm{B}} b^*_{\mathrm{A}})[\exp\{-(M_{\mathrm{A}} + M_{\mathrm{B}})(1 - \lambda_{\mathrm{A-B}})\} - \exp\{-(M_{\mathrm{A}} + M_{\mathrm{B}})\}] \sin(Qr)/Qr \quad (3)$$

where the values of the correlation λ_{A-B} (A, B=Ga, As) were assumed to be 0.65 at 290 K. The distances between Ga-Ga, Ga-As and As-As are $\sqrt{2}a/2$, $\sqrt{3}a/4$ and $\sqrt{2}a/2$, respectively. The obtained results are shown in Fig. 5. The maximum and minimum positions in the observed diffuse scattering of GaAs in Fig. 1 correspond to those calculated from Ga-As pairs in Fig. 5. It is found that the main contribution to the observed oscillatory diffuse scattering is from the correlation between the thermal displacements of gallium and arsenic atoms. The contribution from the correlation among Ga-Ga and As-As atoms should be very weak to produce the observed oscillating scheme in the diffuse scattering of GaAs at 290 K in Fig. 1. In the case of a strong correlation among atoms even to a long distance, we need to apply the well-known theory of the correction for the thermal diffuse scattering (TDS).[7] We have to estimate the contribution of TDS around the Bragg reflections in the correction.

The characteristics of the oscillatory diffuse scattering intensities from semiconductor GaAs are similar to those of solid electrolytes measured by means of neutron diffraction. The background function including the correlation between the thermal displacements of atoms would be very effective to analyze the scattering data from other crystals with the measurement by X-ray and neutron diffraction.

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