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Supplementary materials:



Fig. S1. θ-dependent Raman intensity of all modes in a polar coordinate.

Based on group theory analysis, the form of the Raman tensor (R) is determined by the point group of the material and the symmetry of the Raman mode. Therefore, the Raman scattering intensity is closely related to the crystal structure, in addition to the polarization directions of the incident and scattering lights. the Raman intensity (I) can be expressed as,

$$I \propto |\boldsymbol{e}_s \cdot \boldsymbol{R} \cdot \boldsymbol{e}_i|^2 \tag{1}$$

Where e_s and e_i are the polarization vectors of scattering and incident lights, respectively. *R* is the second-order polarizability tensor of the Raman mode. For the C2/m GeAs,

$$R(A_g) = \begin{pmatrix} b & 0 & d \\ 0 & c & 0 \\ d & 0 & e \end{pmatrix}; R(B_g) = \begin{pmatrix} 0 & f & 0 \\ f & 0 & g \\ 0 & g & 0 \end{pmatrix}$$
(2)

Where the alphabets b - g are Raman tensor elements. In our experiment, the layer stacking direction is along the a-axis, and the sample is placed in the b-c plane. This means the polarization vectors e_s and e_i are fixed on the b-c plane. Assuming $e_i = (0, \cos \theta, \sin \theta)^T$, then $e_s = (0, \cos \theta, \sin \theta)$ in parallel configuration and $e_s = (0, -\sin \theta, \cos \theta)$ in perpendicular configuration. Table SI lists the derived Raman intensity for the two modes of A_g and B_g. Note that due to the randomness of the sample placement, θ is actually (θ + φ), where φ is the angle between the crystal axis and the polarization direction of the incident light. For this work, φ =30°.

Mode	Configuration	Intensity expression
A_g	// (XX)	$I \propto (c \cos^2 \theta + e \sin^2 \theta)^2$
	⊥(XY)	$\frac{e-c}{1}\sin 2\theta$
B _g	// (XX)	$I \propto (g \sin 2\theta)^2$
	$\perp(XY)$	$I \propto (g \cos 2\theta)^2$

Table SI. Raman intensity for $A_{g}\,\text{and}\,B_{g}$ modes.