Laser tuning in AlN single crystals

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In the fields of trace gas detection, electric field detection, visible optical communication, and beyond, laser tuning technology plays an increasingly important role, as shown in Fig. S1. Following the miniaturization of optoelectronic devices, it is desirable to integrate a large number of lasers with different wavelengths into one chip [1]. However, the dynamic wavelength allocation efficiency of fixed-wavelength laser is quite low, which greatly limits the monolithic integration of optoelectronics systems. Therefore, the development of laser wavelength tuning technology is urgent and crucial to the efficiency of optoelectronics [2–7]. Raman scattering is a physical phenomenon that can be explained by mature normal form [8,9]. However, most studies only pay attention to the fact that Raman spectrum is a non-destructive material characterization method with fingerprint identification characteristics, while its essential feature of inelastic scattering is always ignored [10]. The natural frequency shift characteristics indicate the possibility of Raman scattering as a feasible method to realize laser wavelength tuning [11–13].

An important prerequisite for achieving laser wavelength tuning by the Raman effect is the scattering light with polarization characteristics dependent on laser medium [14]. For isotropic optical crystals, the Raman scattering signal is often accompanied by depolarization phenomenon. Thus, optical crystals with strong anisotropy may be an ideal carrier for laser wavelength tuning. Here, we adopted wurtzite aluminum nitride (AlN) as the laser medium. As a mature binary semiconductor, AlN has a relatively simple phonon dispersion relationship compared with other multicomponent systems, which directly determines a “clean” Raman spectrum [15–18]. In addition, high-quality and large-size AlN single crystals (SCs) with different crystal planes have been fabricated *via* physical vapor transport (PVT) technology [19,20], which avoids the interference of Raman signals from other crystal planes.

In this letter, we discovered that when the polarization direction of incident light is along the *c*-axis of AlN SC, an “extinction phenomenon” will appear in $E_1(\text{TO})$ and E_2^2 modes, and only $A_1(\text{TO})$ mode can be observed. In this situation, the scattering light corresponding to $A_1(\text{TO})$ mode shows perfect linear polarization characteristics with a degree of polarization $((I_{\parallel} - I_{\perp}) / (I_{\parallel} + I_{\perp}))$ approaching 1. This phenomenon indicates that AlN can realize laser frequency shift with the help of Raman scattering. In addition, based on the theory that phonon frequency is limited by temperature, an ultra-high precision laser wavelength tuning (0.0006 nm K^{-1}) has been realized by controlling the temperature [21,22]. Our experimental results indicate that wurtzite AlN with high thermal conductivity and high stability may be an ideal carrier for laser wavelength tuning [23]. Theoretically speaking, changing laser medium can achieve arbitrary wavelength tuning, which may help with the development of integrated sensing field.

Here, we adopted backscattering geometry to collect the angle-resolved polarization Raman signal of *m*-plane AlN SC with Renishaw Raman spectrometer (inVia Reflex). The measurement geometry is shown in Fig. 1a [24–34], where a polarizer with fixed polarization direction was placed in the collecting optical path to ensure that the polarization direction of scattering light was strictly consistent with that of pump light. In order to avoid the influences of pump light absorption and resonance Raman scattering on the intensity of scattering light, 488-nm Ar⁺ gas laser was used as pump light and focused on the AlN surface through a 50× quartz lens [35–39]. Meanwhile, in order to smooth the Raman signal and prevent the sample from being damaged, exposure time and integration time were set to 1 s and 10 times, respectively.

Previously, we have reported an angle-resolved polarization Raman spectra of AlN microwires with a diameter of 200 μm under the excitation of a 532-nm laser, in

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Figure 1 Angle-resolved polarization Raman spectra of m-plane AlN. (a) Backscattering polarization Raman measurement geometry. The illustration is the optical photograph of m-plane AlN SC. (b) Angle-resolved polarization Raman spectrum of m-plane AlN SC. (c–e) Raman scattering intensities of $A_1(\text{TO})$, $E_1(\text{TO})$ and E_2^2 modes under different rotation angles.

which the information about its Raman tensor with a strong Raman scattering anisotropy was obtained [20,40]. However, laser wavelength tuning achieved by Raman scattering has a higher and stricter requirement on both the quality and flatness of crystal surface. Thus, an AlN SC with larger area and higher quality was used as the measured sample in our research. The only XRD diffraction peak [100] found in Fig. S2 indicates its perfect crystal orientation. The illustration in Fig. 1a is the optical photograph of a m-plane AlN SC with a diameter of 15 mm [41].

Fig. 1b is the angle-resolved polarization Raman spectrum of m-plane AlN SC, which directly presents the Raman scattering intensities of three vibration modes ($A_1(\text{TO})$ (607.87 cm^{-1}), $E_1(\text{TO})$ (665.87 cm^{-1}) and E_2^2 (653.40 cm^{-1})) under different rotation angles, of which the result is in accordance with that obtained by a 532-nm laser [20,40]. From the angle-resolved polarization Raman spectrum, it can be clearly observed that when the polarization direction of pump light is along the c -axis of AlN, $E_1(\text{TO})$ and E_2^2 modes will disappear, and only A_1

(TO) mode can be found with its intensity up to the maximum. The vibration laws of these modes follow the Raman selection rule, that is $I \sim |e_s \cdot R \cdot e_i|^2$, where e_s and e_i denote the polarization direction vectors of pump and scattering light, R is the Raman tensor of certain phonon mode [9]. Based on the results of previous studies on the Raman polarization of AlN SC [20,40], the Raman scattering intensities of different vibration modes under different rotation angles can be obtained as follows

$$\begin{aligned}
 I_{A_1(\text{TO})}(\theta) &\sim |a|^2 \cos^4 \theta + |b|^2 \sin^4 \theta \\
 &+ \frac{1}{2} |a||b| \sin^2(2\theta) \cos(\varphi_a - \varphi_b), \\
 I_{E_1(\text{TO})}(\theta) &\sim \frac{|c|^2}{2} \sin^2(2\theta), \\
 I_{E_2^2}(\theta) &\sim |d|^2 \cos^4 \theta,
 \end{aligned} \quad (1)$$

where a , b , c and d are Raman tensor elements, φ_a and φ_b denote the complex phases of corresponding Raman tensor elements a and b , and θ is marked as the angle between the polarization direction vector of pump light

and the b -axis of AlN. Fig. 1c–e present the Raman scattering intensities of $A_1(\text{TO})$, $E_1(\text{TO})$, and E_2^2 modes under different rotation angles and the fitting results based on Equation (1). For bulk materials, it is difficult to determine the crystal orientation through the angle-resolved polarization Raman spectrum of $A_1(\text{TO})$ mode. According to Raman selection rule, when the polarization direction of incident light is parallel to the c -axis of m -plane AlN single crystal, the scattering light corresponding to E_2^2 mode will disappear. Returning to the results obtained from the angle-resolved polarization Raman spectrum of m -plane AlN, it is clear that when the sample is rotated by 90° , the polarization direction of incident light will be parallel to the c -axis.

As described above on the Raman scattering intensities of $A_1(\text{TO})$, $E_1(\text{TO})$ and E_2^2 vibration modes, an “extinction phenomenon” can be observed when the rotation angle θ is equal to 90° . At this moment, a scattering light with specific wavelength can be obtained. According to the vibration frequency at 607.87 cm^{-1} of $A_1(\text{TO})$ mode at

room temperature, a corresponding wavelength of scattering light at 502.92 nm can be obtained. In order to verify the polarization characteristics of scattering light, we designed a specific measurement geometry as shown in Fig. 2a [31]. There are two procedures. Firstly, fixing the polarization direction of pump light parallel to the c -axis of AlN; secondly, changing the polarization orientation of the polarizer placed in the collection optical path to verify the depolarization characteristics of scattering light. Results are shown in Fig. 2b, that is, when the polarization direction of polarizer is along the c -axis of AlN, a strong scattering light can be observed, which is absolutely different from the situation when the polarization direction of polarizer is along the b -axis, in which $A_1(\text{TO})$ mode seems to be forbidden. This phenomenon is also consistent with the description of Raman selection rule. It is explicit that there is no depolarization phenomenon observed in the $A_1(\text{TO})$ mode when the polarization direction of pump light is along the c -axis of AlN, which directly indicates that the scattering light with

Figure 2 Verification of polarization characteristics of scattering light. (a) Schematic diagram of laser wavelength tuning. (b) Output light spectrum when the polarization direction of scattering light is parallel to the b -axis and c -axis of AlN SC, respectively. (c, d) Output light spectra and intensities under the pump light with different excitation densities.

a wavelength of 502.92 nm has apparent linear polarization characteristics.

We also measured the intensities of scattering light excited by a 488-nm laser with different excitation densities. In order to measure the optical power density, we used an optical power meter whose model is NOVA II PD300 UV. Measured results are shown in Fig. 2c. With the decrease of excitation density, the intensity of scattering light also reduces rapidly. Then, we extracted the light intensities of $A_1(\text{TO})$ mode under the pump light with different excitation densities, whose results are plotted in Fig. 2d. Obviously, there is a threshold excitation density P_{th} , that is, only when the excitation density of 488-nm pump light is larger than P_{th} , can $A_1(\text{TO})$ mode be excited.

We measured the output light spectra of $A_1(\text{TO})$ mode at different temperatures, and the results are plotted in Fig. 3. During the measurement, the polarization direction of pump light is always parallel to the c -axis of AlN SC. We normalized the output light spectra at different temperatures. Then we extracted the wavelengths and FWHMs of output light at different temperatures, and the

extraction results are shown in Fig. 3c and d. For the wavelength, it can be observed that the wavelength of scattering light will redshift with the decrease of temperature, that is, its wavelength increases from 502.90 nm at 420 K to 503.05 nm at 80 K. Besides, its change trend is also different in different temperature ranges. When the temperature of sample is higher than 250 K, its wavelength is almost linearly redshift with an accuracy of 0.0006 nm K^{-1} as the temperature decreases; when the temperature is lower than 250 K, its wavelength will increase with the decrease of temperature in a nonlinear law. For the FWHM, it can be found that the scattering light has a quite narrow FWHM, which decreases in an almost linear way from 0.17 nm at 420 K to 0.11 nm at 80 K. This also suggests that Raman scattering light has a great monochromaticity close to that of commercial lasers.

In this letter, we realized wavelength modulation in m -plane AlN SC by means of Raman scattering effect. Based on the Raman selection rule, when an “extinction phenomenon” appears in $E_1(\text{TO})$ and E_2^2 vibration modes, only $A_1(\text{TO})$ mode can still be observed. With the help of

Figure 3 Temperature effect of output light. (a) Output light spectra under different temperatures. (b) Normalized output light spectra under different temperatures. (c, d) Extracted wavelengths and FWHMs of output light under different temperatures.

a specific optical measurement geometry, we confirm that the output light corresponding to $A_1(\text{TO})$ mode has perfect liner polarization characteristics with a degree of polarization approaching 1. Besides, the extremely narrow FWHM indicates that Raman scattering light also has great monochromaticity. Based on these experimental results, it can be expected that Raman scattering effect may be a feasible method to realize laser wavelength tuning. What's more, based on the theory that phonon frequency is limited by the temperature, laser wavelength tuning with an accuracy of 0.0006 nm K^{-1} was achieved *via* temperature regulation. Theoretically speaking, changing the laser medium can realize arbitrary wavelength tuning, which is an important reference to achieving monolithic multi-channel coherent optical communication.

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Author contributions Zhu Y designed the experimental details. Zheng W directed this program. Zhu Y, Zhu S, Ding Y and Jia L led the experimental work. Huang F participated in the discussion. Zhu Y contributed to the writing of the manuscript.

Conflict of interest The authors declare that they have no conflict of interest.

Supplementary information Supplementary information and supporting data are available in the online version of the paper.

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AlN单晶中的激光调谐

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摘要 激光波长调谐对于在压力检测、强电场监测、痕量气体探测以及可见光通讯领域中提高动态波长分配效率起着重要作用. 拉曼散射是一种具有频移特性的非线性光学效应, 这表明拉曼散射可能是一种实现激光波长调谐的可行方法. 在本文中, 我们发现特定的测试结构下, 氮化铝的 $A_1(\text{TO})$ 模式对应的散射光具有线偏振特性. 此外, 通过温度调制, 我们在氮化铝中实现了高精度的散射光波长调谐. 基于实验结果, 具有高热导率和高稳定性的氮化铝可能是一种实现激光波长调谐的良好载体.