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A Comprehensive Study on the Optical Properties of Thin ReS_2:Au Layered Single Crystals

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1. Introduction

The transition-metal semiconductor ReS_2 is a diamagnetic indirect semiconductor and belongs to the family of transition-metal dichalcogenides (TMDCs). ReS_2 has been crystallized in a triclinic distorted structure with strong covalent bonds within a layer consisting of S-Re-S sheets and weak van der Waals interactions between layers [1]. The crystal of ReS_2:Au is shown with strip lines which are formed by metal chains along the in-plane b [010] axis. Owing to the unique optical, electrical and mechanical properties, the layered semiconductors have attracted considerable interest in basic studies and applications. In this study, the optical properties of Au-doped rhenium disulphide (ReS_2:Au), which was grown by the chemical vapor transport (CVT) method using I_2 as a transporting agent, has been investigated by temperature- and polarization-dependent thermoreflectance (TR), transmittance (T), photoluminescence (PL), and photoconductivity measurements to determine their energy gaps, crystal quality and doping effects [2]. The polarization-dependent TR and T measurements can be used to characterize the unique polarization property of samples, and identify the origin of the excitonic transitions. The polarization and temperature dependence of excitonic transitions have been performed from $\theta = 0$ to 90° and 42 to 300 K respectively, and their polarization properties were studied by photoconductivity as well [3]. We also presented the polarization dependence and doping effect by luminescent PL. The least-squares fits are used to extract the parameters that describe the temperature variations of the band-edge excitonic transitions.

2. Results and discussion

The temperature-dependent TR and T spectra of ReS_2:Au crystal with horizontal and perpendicular polarizations at selective temperatures between 42 and 300 K are illustrated in Fig. 1(a) and (b). The dashed and solid lines are respectively the experimental spectra of E||b and $E\perp b$ polarizations while the open circles are least-square fits of Lorentzian line-shape function given by [4],

$$\frac{\Delta R}{R} = \mathrm{Re}\left[\sum_{i=1}^{n} A_i^{ex} e^{j\phi_i^{ex}} \left(E - E_i^{ex} + j\Gamma_i^{ex}\right)^{-2}\right] \qquad (1)$$

where $A^{ex}{}_i$ and $\varphi^{ex}{}_i$ are the amplitude and phase of the line shape, and $E^{ex}{}_i$ and $\Gamma^{ex}{}_i$ are the energy and broadening parameter of the interband excitonic transitions. The arrows indicate six excitonic features of ReS_2:Au.

The temperature dependence TR measurement shows a blue-shift and the absorption edge also shifts toward high energy as the temperature of the sample is lowered. From the absorption spectra, there are two obvious signals at different polarization angles which were originated from totally different transitions. By comparing with the TR measurement, the observed absorption peaks are determined to be due to excitonic transitions $E^{ex}{}_1$ and $E^{ex}{}_2$.

Fig. 2(a) shows the temperature-dependent PL measurement, where the open circles are least-square fits of Gaussian function. It can be found that when the temperature is increased the intensity is decreased and the FWHM becomes wider. The temperature-dependent PC spectra are shown in Fig. 2(b). Comparing with the absorption, TR and PL, the observed PC features are determined from different origins. The result reveals the transition energies indicated with arrows.

The temperature-dependent excitonic transition energies of $E^{ex}{}_1$ and $E^{ex}{}_2$ have been fitted by Varshni semiempirical relationship and the equation proposed by O'Donnell and Chen and the results are shown in Fig. 3(a). The temperature dependence of the broadening parameters of $E^{ex}{}_1$ and $E^{ex}{}_2$ transitions, which was given by eq. (2), are extracted from the lineshape fit for ReS_2:Au and displayed in Fig. 3(b).

$$\Gamma_i(T) = \Gamma_{io} + \frac{\Gamma_{iLO}}{\left[\exp(\Theta_{iLO}/T) - 1\right]} \qquad (2)$$

where Γ_{io} is the zero temperature broadening parameter and Γ_{iLO} is caused by the exciton-LO phonon interaction, and Θ_{iLO} is the LO phonon temperature.

The angular-dependent TR, PL and PC spectra of ReS_2:Au in the Fig. 4 clearly show that the $E^{ex}{}_1$ transition feature is obviously observed at low polarization angles, while the $E^{ex}{}_2$ transition feature is presented at high polarization angles. The spectrum amplitudes of $E^{ex}{}_1$ transition gradually decrease with increasing the polarization angular. Conversely, the amplitudes of $E^{ex}{}_2$ transition grow up gradually and become the main feature at θ= 90°. For PL spectra, a small extra feature is observed at the photon energy below the E^{ex}_1 and E^{ex}_2 transitions, respectively. We believe that these two extra PL feature should be relative to the doping of gold, because in undoped sample, they did not been observed. The exciton series (E^{ex}_3 and E^{ex}_4) are observed at the high energy side, they show no obvious polarization correlation.

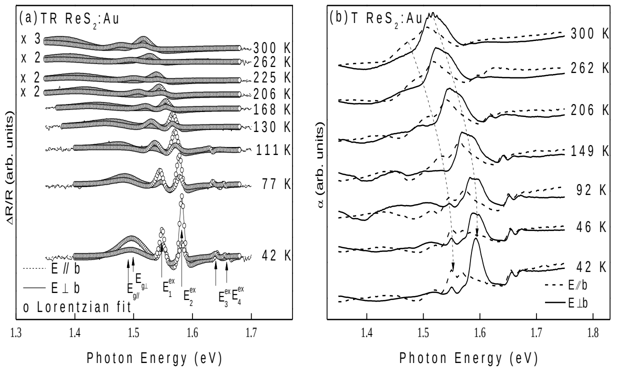

Fig. 1 The temperature-dependent polarized (a) TR and (b) T spectra of Au-doped ReS_2 in the temperature ranging between 42 and 300 K.

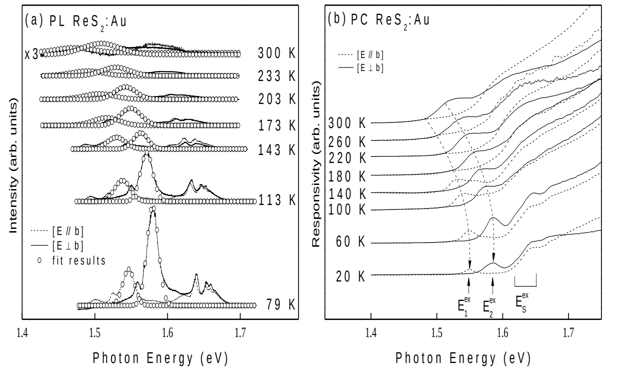

Fig. 2 The temperature-dependent polarized (a) PL and (b) PC spectra of Au-doped ReS_2 in the temperature range from 79 to 300 K and 20 to 300 K.

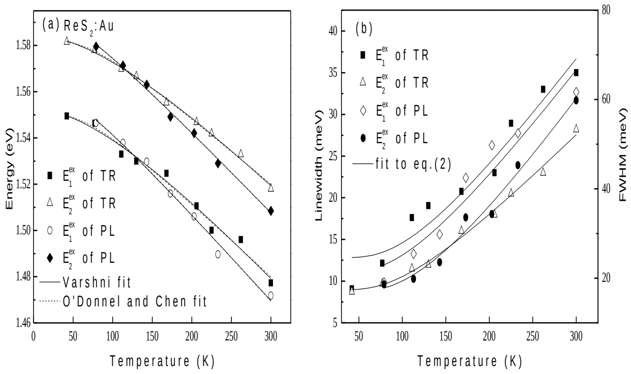

Fig. 3 (a) Temperature-dependent excitonic transition energies of Au-doped ReS_2. Solid curves indicate least-square fit to Varshni and dashed curves fit to the equation proposed by O'Donnell and Chen. (b) Temperature-dependent of linewidths of the interband transitions E^{ex}_1 and E^{ex}_2. The solid curves indicate least-square fit to eq. (2).

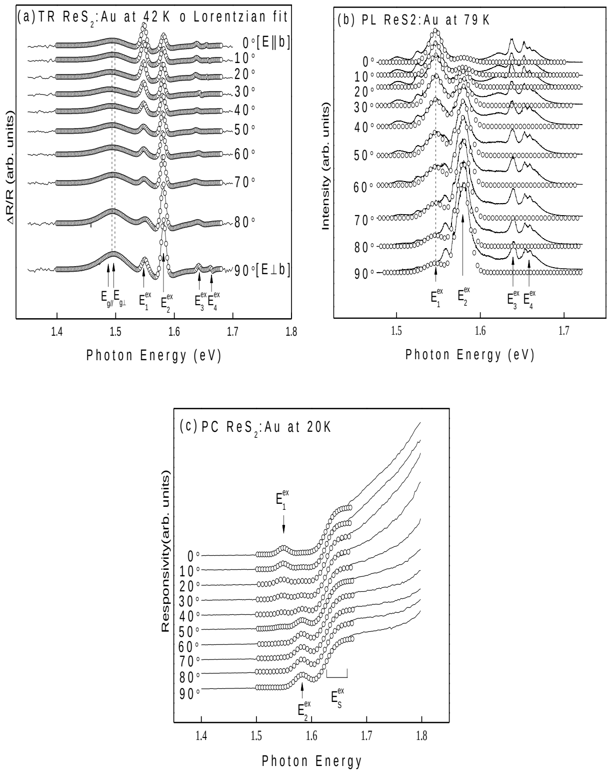

Fig. 4 Polarization-dependent (a) TR (b) PL and (c) PC spectra of Au-doped ReS_2 with polarization angles ranging from θ= 0 (E||b) to 90°(E ⊥ b).

3. Conclusions

We have measured the temperature- and polarization-dependent optical characterizations of Au-doped ReS_2 by using TR, T and PL measurements. The results provide conclusive evidence that E^{ex}_1 and E^{ex}_2 arise from different origins. A previous study indicated that E^{ex}_1 and E^{ex}_2 are closely related to the transitions of the nonbonding Re 5d t_{2g} (d_{xy},d_{x2-y2}) to anti-bonding 5d t_{2g}*band. The near-edge temperature-dependent in-plane anisotropic optical properties were studied by the polarization-dependent PC measurements in the temperature range from 20 to 300 K.

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