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THE VIBRATIONAL RELAXATION PROCESSES IN $BeAl_2O_4:Cr^{3+}$

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The vibrational relaxation processes in $BeAl_2O_4:Cr^{3+}$ were studied by analysing the rise time of luminescence which was found to be 100 ps when excited by 532 nm.

INTRODUCTION

Chromiun-doped $BeAl_2O_4$ is a kind of new laser materials in which laser action occurs not only on sharp R-lines but also in the vibronic side-band where the lasing is broadly tunable[1,2], so the electron-LO phonon interaction is important in this material, we try to study the dynamic processes of electron-LO phonon interaction. In $BeAl_2O_4:Cr^{3+}$ the transition-metal ion Cr^{3+} substitudes Al^{3+} into two inequivalent crystal sites 75% in mirror sites(Cs) and the rest in inversion sites(Ci). According to selection rule, the R-lines in Cs site(Rs) are electric-dipole transition and the R-lines in Ci site(Ri) are magnetic-dipole transition[2], so the Rs-lines are especially strong in the luminescence spectrum, we pay attention to study their luminescence dynamic properties.

EXPERIMENT

We excited the sample $BeAl_2O_4:Cr^{3+}$ by means of an active-mode-locked YAG laser (pulse width 100 ps, second harmonic wave length 532nm), and then used a set of interference filters and a high speed streak camera to study the luminescence dynamic process of Rs-lines. As the luminescence life times of Rs-lines are very long (ms), the streak camera should use the gate mode function to sample a special period of the luminescence process.

THEORY

When the luminescence centers are excited by laser pulse 532 nm, the electrons come into high excited states[2]. Then two processes will take place (1) hot luminescence, the luminescence of electron in high excited states; (2) vibrational relaxation, the electron in higher excited states relaxes down to lower excited states with emission of LO phonons. As the vibrational relaxation is very quick (pico-second process), the highly excited electrons relax down to lower excited states, the hot luminescence is very weak compared with the electron-LO phonon interaction. After relaxed down to the lowest excited states ^2E, luminescence Rs-lines appears. The dynamic process can be expressed as follow

$$\begin{cases} dN_1/dt = -Tn^{-1}N_1 \\ dN_2/dt = Tn^{-1}N_1 - Te^{-1}N_2 \end{cases}$$

where N_1 is the electron population in higher excited states($4T_2$), N_2 is that in the lowest excited states(^2E). Te is the life time of energy level ^2E, Tn is the interaction time of electron-LO phonon. By solving the equations we have

$$N_1 = N_0[1-\exp(-t/T)],\ Tn \ll Te$$

where $1/T = 1/Tn - 1/Te$. The luminescence of Rs-lines is given as

$$I = Te^{-1} N_0[1-\exp(-t/T)]$$

By studying the time developing process of Rs-lines, we can get some information about the time scale of the electron-LO phonon interaction.

RESULTS AND DISCUSSION

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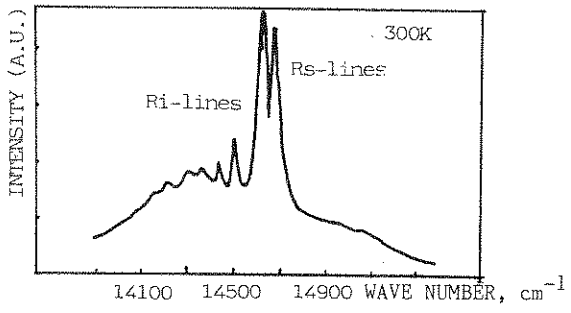

FIGURE 1
Luminescence spectrum of $BeAl_2O_4:Cr^{3+}$ excited by 488 nm.

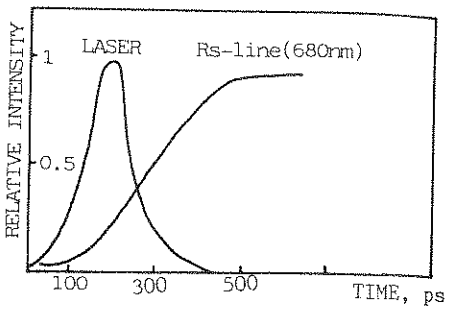

FIGURE 2
Time resolved luminescence spectrum of Rs-lines

First we used a Raman Spectrograph to measure the luminescence spectrum of $BeAl_2O_4:Cr^{3+}$ excited by Ar^+-laser 488 nm at room temperature. The result is given in Figure 1.

The vibronic sidebands are electric-dipole-LO phonon transitions of Cr^{3+} at Ci site. The LO phonon energy is 250 cm^{-1}.

Then we studied the time resolved luminescence spectrum of Rs-lines, the result is shown in Figure 2.

As the laser pulse is 100 ps wide, we take a deconvolution method to get the real rising time of Rs-lines. Suppose F(t) is the laser pulse, D(t) is the observed luminescence rising, G(t) is the real rising.

$$D(t) = \int_0^t F(x)G(t-x)dx$$

Intergrate both sides between the time limits 0 and t, put $G(t)=k[1-\exp(-t/T)]$ into the equation. Note[4]

$$k\int_0^t \exp(-y/T)\int_0^y F(x)\exp(x/T)dxdy$$
$$= kT\int_0^t F(x)dx - kT\int_0^t \exp[-(t-x)/T]F(x)dx$$
$$= TD(t)$$

Then we have

$$Z(t) = -TW(t)+k$$

where $Z(t) = \int_0^t D(x)dx / \int_0^t\int_0^y F(x)dxdy$

$W(t) = D(t) / \int_0^t\int_0^y F(x)dxdy$

From the experimental results, we calculated Z(t), W(t), and drew a curve of W(t) versus Z(t). The curve must be a straight line with slope -T. In this way, we get Th=100 ps. As we know the high excited electrons have excess energy of 4050cm^{-1}, the LO phonon energy is 250cm^{-1}, there are about 4050/250≈16 vibrational levels between high excited states and ^2E states, so the highly excited electrons relax down along the configurational curve by emitting LO phonons, the total relaxation time before luminescence from the lowest excited states(^2E) to the ground state, is about 100ps.

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