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光泵氨分子振动基态远红外激光的增益系数*

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摘要 从密度矩阵运动方程出发，详细地推导了光泵氨分子振动基态远红外激光的增益系数表达式.

关键词 光泵远红外激光、振动基态反演跃迁，NH_3 分子.

氨分子 增益系数

THE GAIN COEFFICIENT OF OPTICALLY PUMPED NH$_3$ MOLECULES FAR-INFRARED LASER IN VIBRATION GROUND STATE*

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Abstract *The density matrix equation of three-level system for NH_3 molecules far-infrared* (FIR) Raman transition in vibration ground state was solved. The gain coefficient of NH_3 molecules far-infrared laser in vibration ground state was obtained.

Key words optically pumped far-infrared (FIR) laser, vibration ground-state retrieval transition, NH_3 molecules.

引言

NH_3 分子振动激发态的远红外激光(对应正三能级系统)容易获得，相关的实验和理论研究很多[1~4]. 而获得 NH_3 分子振动基态的远红外激光(对应倒三能级系统)的效率非常低，至今只有实验报道[5]，缺乏相应的理论研究. 本文以 CO_2-10R(14)泵浦超辐射式 NH_3 分子远红外激光为例，从密度矩阵运动方程出发，详细地推导了光泵氨分子振动基态远红外激光的增益系数表达式. 这一工作对于进一步深入研究光泵 NH_3 分子振动基态的远红外激光特性具有一定的意义.

1 模型

与 CO_2-10R(14)泵浦激光相接近的 NH_3 分子能级结构如图 1 所示，其中中红外振动吸收跃迁为 G-V_2: aR(1,1)，相应的远红外跃迁为 G: sP(2,1)，其波长为 265.5μm.

上述光泵激光过程实际是偏频拉曼过程，其跃迁过程可以简化为图 2 所示模型.

2 增益系数表达式的推导

根据密度矩阵理论可知，其运动方程为

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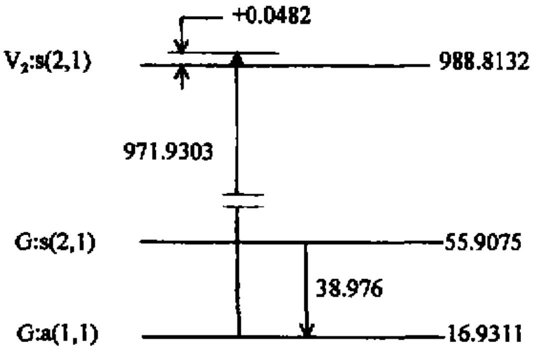

图 1　与 CO_2-10R(14)相匹配的 NH_3 分子能级图(单位:cm^{-1})
Fig. 1 The NH_3 molecules energy levels matching with CO_2-10R(14) (unit:cm^{-1})

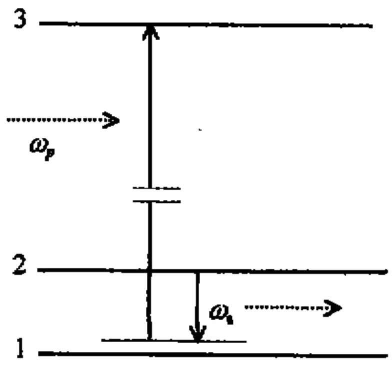

图 2　倒三能级系统的跃迁模型
Fig. 2 Raman transitions of OPFIRL in reversed three-level system

$$
\begin{aligned}
&i\hbar\frac{\partial\rho_{jk}}{\partial t}=\hbar\,\omega_{jk}\rho_{jk}+[H',\rho]_{jk}-\frac{i\hbar}{\tau_{jk}}\rho_{jk},\\
&i\hbar\frac{\partial\rho_{jj}}{\partial t}=[H',\rho]_{jj}-\frac{i\hbar}{\tau_{jj}}(\rho_{jj}-\rho^e_{jj}),
\end{aligned}
\tag{1}
$$

式中 $j,k=1,2,3;j\neq k$.

在倒三能级系统中,能级 3 与能级 2 之间为禁戒跃迁,只有驰豫过程,所以偶极短元 $\mu_{23}=0$,于是,可以令 $\mu_{13}=\mu_p,\mu_{12}=\mu_s,\mu_{11}=\mu_{22}=\mu_{33}=\mu_{23}=0,\mu_{jk}=\mu^*_{kj}$. 在电偶极近似下,$H'=-\mu E(t)$,则密度矩阵的对角元满足

$$
\begin{aligned}
i\hbar\frac{\partial\rho_{11}}{\partial t}&=\frac{i\hbar}{\tau_1}(\rho^e_{11}-\rho_{11})+[\mu_{21}\rho_{12}+\mu_{31}\rho_{13}-\\
&\qquad\mu_{12}\rho_{21}-\mu_{13}\rho_{31}]E(t),\\
i\hbar\frac{\partial\rho_{22}}{\partial t}&=\frac{i\hbar}{\tau_2}(\rho^e_{22}-\rho_{22})+[\mu_{12}\rho_{21}+\mu_{21}\rho_{12}]E(t),\\
i\hbar\frac{\partial\rho_{33}}{\partial t}&=\frac{i\hbar}{\tau_3}(\rho^e_{33}-\rho_{33})+[\mu_{13}\rho_{31}+\mu_{31}\rho_{13}]E(t).
\end{aligned}
\tag{2}
$$

而非对角元满足

$$
\begin{aligned}
&\hbar\left[i\frac{\partial}{\partial t}+\frac{i}{\tau_{12}}-\omega_{12}\right]\rho_{12}=(\mu_{12}\rho_{11}-\mu_{12}\rho_{22}-\mu_{13}\rho_{32})E(t),\\
&\hbar\left[i\frac{\partial}{\partial t}+\frac{i}{\tau_{13}}-\omega_{13}\right]\rho_{13}=(\mu_{13}\rho_{11}-\mu_{12}\rho_{23}-\mu_{13}\rho_{33})E(t),\\
&\hbar\left[i\frac{\partial}{\partial t}+\frac{i}{\tau_{23}}-\omega_{23}\right]\rho_{23}=(\mu_{13}\rho_{21}-\mu_{21}\rho_{13})E(t).
\end{aligned}
\tag{3}
$$

共轭项 $\rho_{jk}=\rho^*_{kj}$,($jk=21,31,32$).

设与分子系统相互作用的电场只有泵浦场和信号场,且 $\omega_p\approx\omega_{31},\omega_s\approx\omega_{21}$,即

$$
E(t)=\frac{1}{2}E_p\exp(i\omega_p t)+\frac{1}{2}E_s\exp(i\omega_s t)+\text{c.c.}
\tag{4}
$$

设非对角元有如下形式的尝试解:

$$
\begin{aligned}
\rho_{12}&=P_{12}\exp(i\omega_s t),\\
\rho_{13}&=P_{13}\exp(i\omega_p t),\\
\rho_{23}&=P_{23}\exp[i(\omega_p-\omega_s)t].
\end{aligned}
\tag{5}
$$

其中 P_{ij}($ij=12,13,23$)为时间的缓变函数. 在旋波近似下,方程(2)和(3)可分别简化为

$$
\begin{aligned}
&\tau_1\frac{\partial\rho_{11}}{\partial t}+\rho_{11}+i\left[\frac{\tau_1}{\tau_{12}}B^*_sP_{12}+\frac{\tau_1}{\tau_{13}}B^*_pP_{13}-\text{c.c.}\right]\\
&\qquad=\rho^e_{11},\\
&\tau_2\frac{\partial\rho_{22}}{\partial t}+\rho_{22}+i\left[\frac{\tau_2}{\tau_{12}}B^*_sP_{12}-\text{c.c.}\right]\\
&\qquad=\rho^e_{22},\\
&\tau_3\frac{\partial\rho_{33}}{\partial t}+\rho_{33}+i\left[\frac{\tau_3}{\tau_{13}}B^*_pP_{13}-\text{c.c.}\right]\\
&\qquad=\rho^e_{33};
\end{aligned}
\tag{6}
$$

$$
\begin{aligned}
&[(\omega_{21}-\omega_s)\tau_{12}+i]P_{12}\\
&\qquad=B_s(\rho_{11}-\rho_{22})-\frac{\tau_{12}}{\tau_{13}}B_pP_{32},\\
&[(\omega_{31}-\omega_p)\tau_{13}+i]P_{13}\\
&\qquad=B_p(\rho_{11}-\rho_{33})-\frac{\tau_{13}}{\tau_{12}}B_sP_{23},\\
&[(\omega_{31}-\omega_p)\tau_{23}-(\omega_{21}-\omega_s)\tau_{23}+i]P_{23}\\
&\qquad=\frac{\tau_{23}}{\tau_{13}}B_pP_{21}-\frac{\tau_{23}}{\tau_{12}}B^*_sP_{13},\\
&P_{jk}=P^*_{kj}.
\end{aligned}
\tag{7}
$$

其中 $B_s=\tau_{12}\mu_sE_s/2\hbar,B_p=\tau_{13}\mu_pE_p/2\hbar,B^*_s$ 和 B^*_p 分别为 B_s 和 B_p 的共轭量,而 $\mu_pE_p/2\hbar$和 $\mu_sE_s/2\hbar$分别称之

为浦信号和远红外信号的 Rabi 频率.

对 NH_3 气体，可以认为所有的横向驰豫时间 τ_{jk} 相等，即 $\tau_{12}=\tau_{13}=\tau_{23}=T_a$；设泵浦场与信号场最初位相相同，则 $B_s=B_s^*$，$B_p=B_p^*$. 又因为脉冲泵激光器（TEA CO_2），其泵脉冲持续时间一般为 100ns，而 $\tau_j(\tau_j>\tau_{jk})$ 的数量级一般为 10ns，因而可近似地把系统看成稳定状态，即 $\frac{\partial\rho_{11}}{\partial t}=\frac{\partial\rho_{22}}{\partial t}=\frac{\partial\rho_{33}}{\partial t}=0$，则方程(6)和(7)可变为

$$\begin{aligned}
&\rho_{11}+i\left[\frac{T_1}{T_a}B_sP_{12}+\frac{T_1}{T_a}B_pP_{13}-c.c.\right]=\rho_{11}^{*},\\
&\rho_{22}+i\left[\frac{T_2}{T_a}B_sP_{12}-c.c.\right]=\rho_{22}^{*},\\
&\rho_{33}+i\left[\frac{T_3}{T_a}B_pP_{13}-c.c.\right]=\rho_{33}^{*},\\
&[(\omega_{21}-\omega_s)T_a+i]P_{12}\\
&\quad=B_s(\rho_{11}-\rho_{22})-B_pP_{32},\\
&[(\omega_{31}-\omega_p)T_a+i]P_{13}\\
&\quad=B_p(\rho_{11}-\rho_{33})-B_sP_{23},\\
&[(\omega_{31}-\omega_p)T_a-(\omega_{21}-\omega_s)T_a+i]P_{23}\\
&\quad=B_pP_{21}-B_sP_{13}.
\end{aligned}\tag{8}$$

加上上述非对角元共轭 3 个方程，令

$$\begin{aligned}
L_{12}&=(\omega_{21}-\omega_s)T_a+i=y+i,\\
L_{13}&=(\omega_{31}-\omega_p)T_a+i=x+i,\\
L_{23}&=(\omega_{31}-\omega_p)T_a-(\omega_{21}-\omega_s)T_a+i\\
&=x-y+i.
\end{aligned}$$

其中 $y=(\omega_{21}-\omega_s)T_a$ 称为信号失谐量（或称为信号频偏），$x=(\omega_{31}-\omega_p)T_a$ 称为泵失谐量（亦称为泵频偏）. 方程(8)可写成矩阵形式. 根据矩阵的特点，方程组可以简化为

$$\begin{bmatrix}y_{11}&y_{12}&y_{13}\\y_{21}&y_{22}&y_{23}\\y_{31}&y_{32}&y_{33}\end{bmatrix}\begin{bmatrix}Im(P_{12})\\Im(P_{13})\\Im(P_{23})\end{bmatrix}=\begin{bmatrix}m_1\\m_2\\m_3\end{bmatrix}.\tag{9}$$

根据克兰姆法则，可得 P 的虚部解析式. 其中

$$\begin{aligned}
&y_{11}=y^2+B_p^2+1+\frac{2B_s^2}{T_a}(T_1-T_2),\\
&y_{12}=y_{21}=B_pB_s+\frac{2T_1}{T_a}B_sB_p,\\
&y_{13}=y_{31}=B_p(x-2y),\\
&y_{22}=x^2+B_s^2+1+\frac{2B_p^2}{T_a}(T_1-T_3),\\
&y_{23}=y_{32}=B_s(2x-y),\\
&y_{33}=B_p^2+B_s^2+(x-y)^2+1,\\
&m_1=B_s(\rho'_{22}-\rho'_{11}),\\
&m_2=B_p(\rho'_{33}-\rho'_{11}),\\
&m_3=0.
\end{aligned}$$

倒三能级系统中跃迁引起的电极化强度为

$$\begin{aligned}
P'&=N_v\langle\mu\rangle=N_vtr(\rho\mu)\\
&=N_v(\rho_{12}\mu_{21}+\rho_{13}\mu_{31}+\rho_{21}\mu_{12}+\rho_{31}\mu_{13})\\
&=P_s+P_p,
\end{aligned}$$

所以

$$\begin{aligned}
P_s&=2N_vRe(p_{11}\mu_s)\\
&=2N_v\mu_sRe[p_{12}\exp(i\omega_st)]\\
&=2N_v\mu_s[Re(p_{12})\cos(\omega_st)-Im(p_{12}\sin(\omega_st)].
\end{aligned}\tag{10}$$

$$\begin{aligned}
P_p=2N_v\mu_p[&Re(p_{13})\cos(\omega_pt)\\
&-Im(p_{13})\sin(\omega_pt)].
\end{aligned}\tag{11}$$

因为开放腔和无腔式激光器样品管内的光场可近似地看作平面波，则 $P'=\varepsilon_0\chi E$，因此远红外信号（频率为 ω_s）的诱导极化强度 $P_s(\omega_s)$ 为实数，即

$$\begin{aligned}
P_s&=Re(\varepsilon_0\chi E)\\
&=Re\left[\varepsilon_0(\chi'+i\chi'')\frac{1}{2}E_s\exp(i\omega_st)+\text{c.c.}\right]\\
&=\frac{1}{2}\varepsilon_0\chi'E_s\cos(\omega_st)-\frac{1}{2}\varepsilon_0\chi''E_s\sin(\omega_st),
\end{aligned}\tag{12}$$

$$\chi''=\frac{4N_v\mu_s}{\varepsilon_0E_s}Im(P_{12}).\tag{13}$$

即极化率的虚部与密度矩阵非对角元的虚部有关. 激光介质的增益系数为

$$\begin{aligned}
G_s&=\frac{4N_v\mu_s\omega_s}{\varepsilon_0\eta cE_s}Im(P_{12})-\alpha_s\\
&=\frac{2N_v\mu_s^2\omega_sT_a}{\varepsilon_0\eta c\hbar\,B_s}Im(P_{12})-\alpha_s.
\end{aligned}\tag{14}$$

同理可得

$$G_p=\frac{4N_v\mu_s^2\omega pT_a}{\varepsilon_0\eta c\hbar\,B_p}Im(P_{13})-\alpha_p.\tag{15}$$

于是，根据式(14)和(15)即可求出倒三能级系统的远红外信号增益系数 G_s 和泵浦信号的吸收系数 G_p. 式(14)和(15)表明，G_s 和 G_p 与有效工作分子数密度、激活介质的折光系数、激光的归一化 Rabi 频率、跃迁复电偶极矩、激光损耗率以及中红外泵浦失谐量和远红外信号失谐量有关. 利用上式理论公式，采用迭代法对光泵氨分子和甲醇分子振动基态远红外激光的频谱曲线和工作参数进行了理论计算，并进行了相关的实验测量，其结果表明理论和实验符合得很好[6]. 因此，本文导出的式(14)和(15)对于进一步深入研究振动基态的光泵远红外激光特性及工作参数优化具有重要意义.

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