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四能级系统极化拍频的不对称特性*

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在泵光为窄带或宽带的情形下, 对四能级系统中极化拍频的不对称特性进行了研究, 发现这种不对称特性是由于光学元件色散所导致的零延时的漂移引起的.

PACC: 4265; 3290

1 引　言

利用时延双频四波混频(FWM)技术研究物质的超快过程是现在非常热门的一个课题, 1980 年, Eichler 等人提出了 FWM 信号强度与自相干函数的平方成正比的自相干理论[1], 1985 年, Beach 又在时延四波混频实验中发现了混频信号不对称的特性[2]. 后来, 付盘铭等人又用自相干函数理论研究了两个非相干光源之间的拍频[3], 并分别在泵光为高斯线型和洛仑兹线型两种情况下, 通过信号强度与含两个频率分量光场的相关函数解释了时延激光感生双光栅(TDLIDG)实验中的不对称现象. 本文分别在窄带和宽带非相干光时研究了四能级系统中由于不对称光路中元件色散造成两个自相干过程零延时之间的偏差, 并解释由此偏差造成的调制信号不对称的现象.

2 结　论

四能级系统(如图 1)中, $|0\rangle$ 为基态, $|1\rangle$ 为中间态, $|2\rangle$, $|3\rangle$ 为激发态. 泵光的几何配制如图 2 所示, 光束 1 的频率为 ω_1, 光束 2, 3 分别包含两个频率 ω_2, ω_3, ω_1, ω_2 和 ω_3 分别对应于从 $|0\rangle$ 到 $|1\rangle$, $|1\rangle$ 到 $2\rangle$ 和 $|1\rangle$ 到 $|3\rangle$ 的跃迁频率 Ω_1, Ω_2 和 Ω_3. 光束 1, 2 中可发生 $\omega_1+\omega_2$ 的第一个双光子过程. 光束 3(ω_2)探测, 产生频率为 ω_1 的信号; 同时光束 1, 2 中也可发生 $\omega_1+\omega_3$ 的第二个双光子过程. 经光束 3(ω_3)探测, 产生频率为 ω_1 的信号. 光束 4 中两个双光子非简并四波混频信号在探测器上形成拍频信号.

由于光束 1, 4 的方向几乎分别与光束 2, 3 相反, 且我们只考虑与延时 τ 有关的信号, 故可建立复电场如下:

$$E_{P_1} = \varepsilon_1 \exp[i(\boldsymbol{k}_1 \cdot \boldsymbol{r} - \omega_1 t)],$$

$$E_{P_2} = \varepsilon_2 u_2(t) \exp[i(\boldsymbol{k}_2 \cdot \boldsymbol{r} - \omega_2 t)] + \varepsilon_3 u_3(t) \exp[i(\boldsymbol{k}_3 \cdot \boldsymbol{r} - \omega_3 t)],$$

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$$E_{P_3} = \varepsilon_2' u_2(t-\tau)\exp[\mathrm{i}(\boldsymbol{k}_2'\cdot\boldsymbol{r}-\omega_2 t+\omega_2\tau)] + \varepsilon_3' u_3(t-\tau+\delta\tau)\exp[\mathrm{i}(\boldsymbol{k}_3'\cdot\boldsymbol{r}-\omega_3 t+\omega_3\tau-\omega_3\delta\tau)].$$

式中 ε_i, $\boldsymbol{k}_i$(ε_i', $\boldsymbol{k}_i'$)分别为 ω_i 分量的光场振幅和光波矢量. $u_i(t)$是描述光场相位和振幅涨落的无量纲统计因子, τ 为光束 2,3 之间的时延, $\delta\tau$ 为两自相干过程间零延时的偏差.

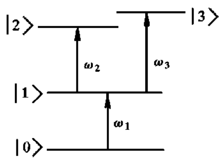

图 1　四能级位形图

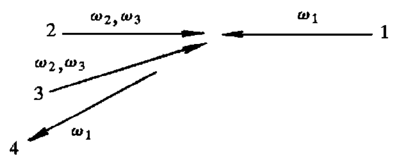

图 2　几何配制图

根据光与四能级系统相互作用的物理机制,可得如下微扰链:

(Ⅰ)　$\rho_{00}^{(0)} \xrightarrow{\omega_1} \rho_{10}^{(1)} \xrightarrow{\omega_2} \rho_{20}^{(2)} \xrightarrow{-\omega_2} \rho_{10}^{(3)}$,

(Ⅱ)　$\rho_{00}^{(0)} \xrightarrow{\omega_1} \rho_{10}^{(1)} \xrightarrow{\omega_3} \rho_{30}^{(2)} \xrightarrow{-\omega_3} \rho_{10}^{(3)}$.

在多普勒增宽系统中,利用密度矩阵动力学方程,可求解以上微扰链得

$$\begin{aligned}\rho_{10}^{(\mathrm{I})} = & \frac{-\mathrm{i}\mu_1\mu_2^2}{\hbar^3}\varepsilon_1\varepsilon_2(\varepsilon_2')^*\exp\{\mathrm{i}[(\boldsymbol{k}_1+\boldsymbol{k}_2-\boldsymbol{k}_2')\cdot\boldsymbol{r}-\omega_1 t-\omega_2\tau]\} \\ & \times\int_0^\infty \mathrm{d}t_3\int_0^\infty \mathrm{d}t_2\int_0^\infty \mathrm{d}t_1\exp\{-\mathrm{i}\boldsymbol{\theta}\cdot[\boldsymbol{k}_1(t_1+t_2+t_3)+\boldsymbol{k}_2(t_2+t_3)-\boldsymbol{k}_2' t_3]\} \\ & \times\exp[-(\Gamma_{10}+\mathrm{i}\Delta_1)t_3]\exp[-(\Gamma_{20}+\mathrm{i}\Delta_1+\mathrm{i}\Delta_2)t_2] \\ & \times\exp[-(\Gamma_{10}+\mathrm{i}\Delta_1)t_1]u_2(t_1-t_2-t_3) \\ & \times u_2^*(t-t_3-\tau),\end{aligned}\tag{1}$$

$$\begin{aligned}\rho_{10}^{(\mathrm{II})} = & \frac{-\mathrm{i}\mu_1\mu_3^2}{\hbar^3}\varepsilon_1\varepsilon_3(\varepsilon_3')^*\exp\{\mathrm{i}[(\boldsymbol{k}_1+\boldsymbol{k}_3-\boldsymbol{k}_3')\cdot\boldsymbol{r}-\omega_1 t-\omega_3\tau+\omega_3\delta\tau]\} \\ & \times\int_0^\infty \mathrm{d}t_3\int_0^\infty \mathrm{d}t_2\int_0^\infty \mathrm{d}t_1\exp\{-\mathrm{i}\boldsymbol{\theta}\cdot[\boldsymbol{k}_1(t_1+t_2+t_3)+\boldsymbol{k}_3(t_2+t_3)-\boldsymbol{k}_3' t_3]\} \\ & \times\exp[-(\Gamma_{10}+\mathrm{i}\Delta_1)t_3]\exp[-(\Gamma_{30}+\mathrm{i}\Delta_1+\mathrm{i}\Delta_3)t_2] \\ & \times\exp[-(\Gamma_{10}+\mathrm{i}\Delta_1)t_1]u_3(t-t_2-t_3) \\ & \times u_3^*(t-t_3-\tau+\delta\tau).\end{aligned}\tag{2}$$

式中 $\boldsymbol{v}$ 为原子速度, μ_1, μ_2, μ_3 分别为$|0\rangle$到$|1\rangle$, $|1\rangle$到$|2\rangle$, $|1\rangle$到$|3\rangle$的跃迁偶极矩阵元. Γ_{10}, Γ_{20}, Γ_{30}为$|0\rangle$到$|1\rangle$, $|0\rangle$到$|2\rangle$, $|0\rangle$到$|3\rangle$的横向弛豫率. $\Delta_1=\Omega_1-\omega_1$, $\Delta_2=\Omega_2-\omega_2$, $\Delta_3=\Omega_3-\omega_3$.

对于相位共轭 FWM 信号的非线性极化强度有

$$P^{(3)} = N\mu_1\int_{-\infty}^{+\infty}\mathrm{d}\boldsymbol{v}w(\boldsymbol{v})\rho_{10}^{(3)}(\boldsymbol{v}).$$

N 为原子个数，对于多普勒增宽系统，$w(\boldsymbol{v})=\frac{1}{\sqrt{\pi}u}\exp[-(\boldsymbol{v}/u)^2]$. FWM 信号正比于 $P^{(3)}$绝对值平方的平均值. FWM 信号强度与六阶相干函数有关，而我们采用相位共轭几何配制且只关心与 τ 有关的信号，假定抽运光 2, 3 是多模热源的混沌统计模式，这样 $I(\tau)\propto|\langle P^{(3)}\rangle|^2$ 仅包含二阶相干函数，若抽运光为洛仑兹线型，则

$$\langle u_i(t_1)u_i^*(t_2)\rangle=\exp(-\alpha_i|t_1-t_2|)\qquad(i=1,2)$$

式中 $\alpha_i=\frac{1}{2}\delta\omega_i$, $\delta\omega_i$ 为 ω_i 的激光线宽.

我们可得
$$\langle P^{(3)}\rangle=P^{(\mathrm{I})}+P^{(\mathrm{II})},$$
式中

$$\begin{aligned}P^{(\mathrm{I})}=&S_1(\boldsymbol{r})\exp[-\mathrm{i}(\omega_1t+\omega_2\tau)]\int_{-\infty}^{+\infty}\mathrm{d}\boldsymbol{v}w(\boldsymbol{v})\int_0^{\infty}\mathrm{d}t_3\int_0^{\infty}\mathrm{d}t_2\int_0^{\infty}\mathrm{d}t_1\exp[-\mathrm{i}\theta_I(\boldsymbol{v})]\\&\times\exp[-(\Gamma_{10}+\mathrm{i}\Delta_1)t_3]\exp[-(\Gamma_{20}+\mathrm{i}\Delta_2+\mathrm{i}\Delta_1)t_2]\\&\times\exp[-(\Gamma_{10}+\mathrm{i}\Delta_1)t_1]\exp(-\alpha_2|t_2-\tau|),\end{aligned}\tag{3}$$

$$\begin{aligned}P^{(\mathrm{II})}=&S_2(\boldsymbol{r})\exp[-\mathrm{i}(\omega_1t+\omega_3\tau-\omega_3\delta\tau)]\int_{-\infty}^{+\infty}\mathrm{d}\boldsymbol{v}w(\boldsymbol{v})\\&\times\int_0^{\infty}\mathrm{d}t_3\int_0^{\infty}\mathrm{d}t_2\int_0^{\infty}\mathrm{d}t_1\exp[-\mathrm{i}\theta_{\mathrm{II}}(\boldsymbol{v})]\\&\times\exp[-(\Gamma_{10}+\mathrm{i}\Delta_1)t_3]\exp[-(\Gamma_{30}+\mathrm{i}\Delta_3+\mathrm{i}\Delta_1)t_2]\\&\times\exp[-(\Gamma_{10}+\mathrm{i}\Delta_1)t_1]\exp(-\alpha_3|t_2-\tau+\delta\tau|),\end{aligned}\tag{4}$$

式中

$$S_1(\boldsymbol{r})=-\frac{\mathrm{i}N\mu_1^2\mu_2^2}{\hbar^3}\varepsilon_1\varepsilon_2(\varepsilon_2')^*\exp[\mathrm{i}(\boldsymbol{k}_1+\boldsymbol{k}_2-\boldsymbol{k}_2')\cdot\boldsymbol{r}],$$

$$S_2(\boldsymbol{r})=-\frac{\mathrm{i}N\mu_1^2\mu_3^2}{\hbar^3}\varepsilon_1\varepsilon_3(\varepsilon_3')^*\exp[\mathrm{i}(\boldsymbol{k}_1+\boldsymbol{k}_3-\boldsymbol{k}_3')\cdot\boldsymbol{r}],$$

$$\theta_{\mathrm{I}}(\boldsymbol{v})=\boldsymbol{v}\cdot[\boldsymbol{k}_1(t_1+t_2+t_3)+\boldsymbol{k}_2(t_2+t_3)-\boldsymbol{k}_2't_3],$$

$$\theta_{\mathrm{II}}(\boldsymbol{v})=\boldsymbol{v}\cdot[\boldsymbol{k}_1(t_1+t_2+t_3)+\boldsymbol{k}_3(t_2+t_3)-\boldsymbol{k}_3't_3]$$

分别对 t_1, t_2, t_3 积分，可得极化强度.

(1) 激光为窄带时情形，即($\alpha_2\ll\Gamma_{20}$, $\alpha_3\ll\Gamma_{30}$)，有信号强度为

$$\begin{aligned}I(\tau)\propto&|B_1|^2\exp(-2\alpha_2|\tau|)+|\eta B_2|^2\exp(-2\alpha_3|\tau-\delta\tau|)\\&+\exp(-\alpha_2|\tau|)\exp(-\alpha_3|\tau-\delta\tau|)\\&\times\{\eta B_1^*B_2\exp[-\mathrm{i}(\omega_3-\omega_2)\tau+\mathrm{i}\omega_3\delta\tau]+\eta^*B_1B_2^*\exp[\mathrm{i}(\omega_3-\omega_2)\tau-\mathrm{i}\omega_3\delta\tau]\}.\end{aligned}\tag{5}$$

其中，在忽略多普勒效应时，

$$B_1=\frac{1}{(\Gamma_{10}+\mathrm{i}\Delta_1)^2}\cdot\frac{1}{\Gamma_{20}+\mathrm{i}(\Delta_2+\Delta_1)},\qquad B_2=\frac{1}{(\Gamma_{10}+\mathrm{i}\Delta_1)^2}\cdot\frac{1}{\Gamma_{30}+\mathrm{i}(\Delta_3+\Delta_1)},$$

$$\eta=\frac{S_2(\boldsymbol{r})}{S_1(\boldsymbol{r})}\approx\frac{\mu_3^2}{\mu_2^2}\left[\frac{\varepsilon_3(\varepsilon_3')^*}{\varepsilon_2(\varepsilon_2')^*}\right].$$

(5)式表明在窄带情形下,信号强度反映了外部激光的特性而与能级系统无关.由于 $\delta\tau$ 的存在引起信号的峰值不出现在 $\tau=0$ 时.

(2) 分析信号强度的另一种方法.在多普勒极限情形下,(即 $k_1 u\to\infty$)有

$$\int_{-\infty}^{+\infty}\mathrm{d}\boldsymbol{v}w(\boldsymbol{v})\exp[-\mathrm{i}\theta_{\mathrm{I}}(\boldsymbol{v})]\approx\frac{2\sqrt{\pi}}{k_1 u}\delta(t_1+t_2+t_3-\xi_1 t_2),\tag{6}$$

$$\int_{-\infty}^{+\infty}\mathrm{d}\boldsymbol{v}w(\boldsymbol{v})\exp[-\mathrm{i}\theta_{\mathrm{II}}(\boldsymbol{v})]\approx\frac{2\sqrt{\pi}}{k_1 u}\delta(t_1+t_2+t_3-\xi_2 t_2),\tag{7}$$

式中 $\xi_1=k_2/k_1$, $\xi_2=k_3/k_1$.

我们将(6)(7)两式代入(3)(4)式中,且假定 $\xi_1>1$, $\xi_2>1$, $\delta\tau>0$,可得极化强度平均值.

(i) $\tau>\delta\tau$ 时,有

$$\begin{aligned}\langle P^{(3)}\rangle &= P^{(\mathrm{I})}+P^{(\mathrm{II})}\\ &=\left(\frac{2\sqrt{\pi}}{k_1 u}\right)S_1(\boldsymbol{r})\exp[-\mathrm{i}(\omega_1 t+\omega_2\tau)](\xi_1-1)\Big\{\frac{\exp(-\alpha_2|\tau|)}{(\Gamma_{20}^a-\Gamma_{10}-\alpha_2+\mathrm{i}\Delta_2^a)^2}\\ &+\exp[-(\Gamma_{20}^a-\Gamma_{10}+\mathrm{i}\Delta_2^a)|\tau|]\left[\frac{-\tau(\Gamma_{20}^a-\Gamma_{10}-\alpha_2+\mathrm{i}\Delta_2^a)-1}{(\Gamma_{20}^a-\Gamma_{10}-\alpha_2+\mathrm{i}\Delta_2^a)^2}\right.\\ &\left.+\frac{\tau(\Gamma_{20}^a-\Gamma_{10}+\alpha_2+\mathrm{i}\Delta_2^a)+1}{(\Gamma_{20}^a-\Gamma_{10}+\alpha_2+\mathrm{i}\Delta_2^a)^2}\right]\Big\}+\frac{2\sqrt{\pi}}{k_1 u}S_2(\boldsymbol{r})\exp[-\mathrm{i}(\omega_1 t+\omega_3\tau-\omega_3\delta\tau)](\xi_2-1)\\ &\times\Big\{\frac{\exp(-\alpha_3|\tau-\delta\tau|)}{(\Gamma_{30}^a-\Gamma_{10}-\alpha_3+\mathrm{i}\Delta_3^a)^2}+\exp[-(\Gamma_{30}^a-\Gamma_{10}+\mathrm{i}\Delta_3^a)|\tau-\delta\tau|]\\ &\times\left[\frac{-(\tau-\delta\tau)(\Gamma_{30}^a-\Gamma_{10}-\alpha_3+\mathrm{i}\Delta_3^a)-1}{(\Gamma_{30}^a-\Gamma_{10}-\alpha_3+\mathrm{i}\Delta_3^a)^2}+\frac{(\tau-\delta\tau)(\Gamma_{30}^a-\Gamma_{10}+\alpha_3+\mathrm{i}\Delta_3^a)+1}{(\Gamma_{30}^a-\Gamma_{10}+\alpha_3+\mathrm{i}\Delta_3^a)^2}\right]\Big\}.\end{aligned}\tag{8}$$

上式中 $\Gamma_{20}^a=\Gamma_{20}+\xi_1\Gamma_{10}$, $\Delta_2^a=\Delta_2+\xi_1\Delta_1$, $\Gamma_{30}^a=\Gamma_{30}+\xi_2\Gamma_{10}$, $\Delta_3^a=\Delta_3+\xi_2\Delta_1$.

(ii) $0<\tau<\delta\tau$ 时,有

$$\begin{aligned}\langle P^{(3)}\rangle &= P^{(\mathrm{I})}+P^{(\mathrm{II})}\\ &=\left(\frac{2\sqrt{\pi}}{k_1 u}\right)S_1(\boldsymbol{r})\exp[-\mathrm{i}(\omega_1 t+\omega_2\tau)](\xi_1-1)\Big\{\frac{\exp(-\alpha_2|\tau|)}{(\Gamma_{20}^a-\Gamma_{10}-\alpha_2+\mathrm{i}\Delta_2^a)^2}\\ &+\exp[-(\Gamma_{20}^a-\Gamma_{10}+\mathrm{i}\Delta_2^a)|\tau|]\left[\frac{-\tau(\Gamma_{20}^a-\Gamma_{10}-\alpha_2+\mathrm{i}\Delta_2^a)-1}{(\Gamma_{20}^a-\Gamma_{10}-\alpha_2+\mathrm{i}\Delta_2^a)^2}\right.\\ &\left.+\frac{\tau(\Gamma_{20}^a-\Gamma_{10}+\alpha_2+\mathrm{i}\Delta_2^a)+1}{(\Gamma_{20}^a-\Gamma_{10}+\alpha_2+\mathrm{i}\Delta_2^a)^2}\right]\Big\}+\frac{2\sqrt{\pi}}{k_1 u}S_2(\boldsymbol{r})\exp[-\mathrm{i}(\omega_1 t+\omega_3\tau-\omega_3\delta\tau)](\xi_2-1)\\ &\times\exp(-\alpha_3|\tau-\delta\tau|)\frac{1}{(\Gamma_{30}^a-\Gamma_{10}-\alpha_3+\mathrm{i}\Delta_3^a)^2}.\end{aligned}\tag{9}$$

(iii) $\tau<0$ 时,有

$$\langle P^{(3)}\rangle = P^{(\mathrm{I})}+P^{(\mathrm{II})}$$

$$= \left(\frac{2\sqrt{\pi}}{k_1 u}\right) S_1(\boldsymbol{r})\exp[-\mathrm{i}(\omega_1 t + \omega_2 \tau)](\xi_1 - 1)\exp(-\alpha_2|\tau|)\frac{1}{(\Gamma_{20}^a - \Gamma_{10} - \alpha_2 + \mathrm{i}\Delta_2^a)^2}$$

$$+ \frac{2\sqrt{\pi}}{k_1 u} S_2(\boldsymbol{r})\exp[-\mathrm{i}(\omega_1 t + \omega_3 \tau - \omega_3 \delta\tau)](\xi_2 - 1)\exp(-\alpha_3|\tau - \delta\tau|)\frac{1}{(\Gamma_{30}^a - \Gamma_{10} - \alpha_3 + \mathrm{i}\Delta_3^a)^2}. \tag{10}$$

下面分别从泵光为窄带和宽带考虑信号强度.

泵光为窄带($\alpha_2 \ll \Gamma_{20}$, $\alpha_3 \ll \Gamma_{30}$),此时不论 τ 取何值,都有

$$I(\tau) \propto \frac{(\xi_1 - 1)^2\exp(-2\alpha_2|\tau|)}{[(\Gamma_{20}^a - \Gamma_{10})^2 + (\Delta_2^a)^2]^2} + \frac{|\eta|^2(\xi_2 - 1)^2\exp(-2\alpha_3|\tau - \delta\tau|)}{[(\Gamma_{30}^a - \Gamma_{10})^2 + (\Delta_3^a)^2]^2} + \exp(-\alpha_2|\tau|)$$
$$\times \exp(-\alpha_3|\tau - \delta\tau|)\{q\exp[-\mathrm{i}(\omega_3 - \omega_2)\tau + \mathrm{i}\omega_3\delta\tau] + q^*\exp[\mathrm{i}(\omega_3 - \omega_2)\tau - \mathrm{i}\omega_3\delta\tau]\}. \tag{11}$$

式中 $q = \dfrac{\eta(\xi_1 - 1)(\xi_2 - 1)}{[(\Gamma_{20}^a - \Gamma_{10}) - \mathrm{i}\Delta_2^a]^2[(\Gamma_{30}^a - \Gamma_{10}) + \mathrm{i}\Delta_3^a]^2}$,

此时结果与(5)式结果类似,反映了激光的特性,而与能级结构无关.信号关于 $\tau = 0$ 不对称.

泵光为宽带($\alpha_2 \gg \Gamma_{20}$, $\alpha_3 \gg \Gamma_{30}$),此时若 $\tau > 0$,则假定 $\tau \gg \alpha_2^{-1}$, $\tau \gg \alpha_3^{-1}$, 当 $\tau > \delta\tau$ 时

$$I(\tau) \propto \left[\frac{\alpha_2(\xi_1 - 1)\tau}{\alpha_2^2 + (\Delta_2^a)^2}\right]^2 \exp[-2(\Gamma_{20}^a - \Gamma_{10}]|\tau|] + |\eta|^2\left[\frac{\alpha_3(\xi_2 - 1)(\tau - \delta\tau)}{\alpha_3^2 + (\Delta_3^a)^2}\right]^2$$
$$\times \exp[-2(\Gamma_{30}^a - \Gamma_{10})|\tau - \delta\tau|] + \left[\frac{\alpha_2(\xi_1 - 1)\tau}{\alpha_2^2 + (\Delta_2^a)^2}\right]\left[\frac{\alpha_3(\xi_2 - 1)\tau}{\alpha_3^2 + (\Delta_3^a)^2}\right]$$
$$\times \tau(\tau - \delta\tau)\exp[-(\Gamma_{20}^a - \Gamma_{10})|\tau|]\exp[-(\Gamma_{30}^a - \Gamma_{10})|\tau - \delta\tau|]$$
$$\times \{\eta\exp[-\mathrm{i}(\Omega_3 - \Omega_2)\tau - \mathrm{i}(\xi_2 - \xi_1)\Delta_1\tau + \mathrm{i}(\Omega_3 + \xi_2\Delta_1)\delta\tau]$$
$$+ \eta^*\exp[\mathrm{i}(\Omega_3 - \Omega_2)\tau + \mathrm{i}(\xi_2 - \xi_1)\Delta_1\tau - \mathrm{i}(\Omega_3 + \xi_2\Delta_1)\delta\tau]\}, \tag{12}$$

此时表明宽带 $\tau > \delta\tau$ 时,反映了能级的结构,信号峰值右移.当 $0 < \tau < \delta\tau$ 时,

$$I(\tau) \propto \left[\frac{\alpha_2(\xi_1 - 1)\tau}{\alpha_2^2 + (\Delta_2^a)^2}\right]^2 \exp[-2(\Gamma_{20}^a - \Gamma_{10})|\tau|]. \tag{13}$$

拍频信号变为第一个双光子非简并四波混频过程的自相干强度.当 $\tau < 0$ 时,

$$I(\tau) \propto \frac{(\xi_1 - 1)^2\exp(-2\alpha_2|\tau|)}{[\alpha_2^2 + (\Delta_2^a)^2]^2} + \frac{|\eta|^2(\xi_2 - 1)^2\exp(-2\alpha_3|\tau - \delta\tau|)}{[\alpha_3^2 + (\Delta_3^a)^2]^2}$$
$$+ \exp(-\alpha_2|\tau|)\exp(-\alpha_3|\tau - \delta\tau|)\{q'\exp[-\mathrm{i}(\omega_3 - \omega_2)\tau + \mathrm{i}\omega_3\delta\tau]$$
$$+ (q')^*\exp[\mathrm{i}(\omega_3 - \omega_2)\tau - \mathrm{i}\omega_3\delta\tau]\}. \tag{14}$$

式中 $$q' = \frac{\eta(\xi_1 - 1)(\xi_2 - 1)}{(\alpha_2 - \mathrm{i}\Delta_2^a)^2(\alpha_3 + \mathrm{i}\Delta_3^a)^2}.$$

由于 $\tau < 0$ 时,泵光 2,3 中相位相关子脉冲时间上发生了重叠,泵光 2,3 变为互相干,故类似于窄带情形(5)式.

(3) 从光子回波角度讨论宽带情形. 由相位匹配条件可知,链(Ⅰ)(Ⅱ)中均可发生光子回波.在多普勒极限增宽下($k_1 u \to \infty$),且 $\xi_1 > 1$, $\xi_2 > 1$.

$$\int_{-\infty}^{+\infty} \mathrm{d}\boldsymbol{v} w(\boldsymbol{v}) \exp[-\mathrm{i}\theta_{\mathrm{I}}(\boldsymbol{v})] \approx \frac{2\sqrt{\pi}}{k_1 u} \delta(t_1 + t_2 + t_3 - \xi_1 t_2),$$

$$\int_{-\infty}^{+\infty} \mathrm{d}\boldsymbol{v} w(\boldsymbol{v}) \exp[-\mathrm{i}\theta_{\mathrm{II}}(\boldsymbol{v})] \approx \frac{2\sqrt{\pi}}{k_1 u} \delta(t_1 + t_2 + t_3 - \xi_2 t_2).$$

由于泵光为宽带非相干光,

$$\exp(-\alpha_2 | t_2 - \tau |) \approx \frac{2}{\alpha_2} \delta(t_2 - \tau), \tag{15}$$

$$\exp(-\alpha_3 | t_2 - \tau + \delta\tau |) \approx \frac{2}{\alpha_3} \delta(t_2 - \tau + \delta\tau). \tag{16}$$

将(6),(7),(15),(16)式代入(3),(4)可得极化强度.在 $\tau > \delta\tau$ 时有

$$\begin{aligned} I(\tau) \propto & [(\xi_1 - 1)\tau]^2 \exp[-2(\Gamma_{20}^{a} - \Gamma_{10})|\tau|] + |\eta|^2 [(\xi_2 - 1)(\tau - \delta\tau)]^2 \\ & \times \exp[-2(\Gamma_{30}^{a} - \Gamma_{10})|\tau - \delta\tau|] + [(\xi_1 - 1)(\xi_2 - 1)]\tau(\tau - \delta\tau)\exp[-(\Gamma_{20}^{a} - \Gamma_{10})|\tau|] \\ & \times \exp[-(\Gamma_{30}^{a} - \Gamma_{10})|\tau - \delta\tau|]\{\eta \exp[-\mathrm{i}(\Omega_3 - \Omega_2)\tau - \mathrm{i}(\xi_2 - \xi_1)\Delta_1\tau + \mathrm{i}(\Omega_3 + \xi_2\Delta_1)\delta\tau] \\ & + \eta^* \exp[\mathrm{i}(\Omega_3 - \Omega_2)\tau + \mathrm{i}(\xi_2 - \xi_1)\Delta_1\tau - \mathrm{i}\Omega_3 + \xi_2\Delta_1)\delta\tau]\}. \end{aligned} \tag{17}$$

此时与(12)式结果完全类似.

在 $0 < \tau < \delta\tau$ 时, $\frac{2}{\alpha_3}\delta(t_2 - \tau + \delta\tau) = 0$, 故(Ⅱ)链不发生光子回波,光束 1,2 间仅含 ω_3 分量的脉冲间变为相关情形.此时信号中只含(Ⅰ)链的自相干强度.

$$I(\tau) \propto [(\xi_1 - 1)\tau]^2 \exp[-2(\Gamma_{20}^{a} - \Gamma_{10})\tau],$$

当 $\tau < 0$ 时, $\frac{2}{\alpha_2}\delta(t_2 - \tau) = 0$, 故(Ⅰ)(Ⅱ)均不发生光子回波.此时由于相关子脉冲时间上发生重叠,光束 1,2 间变为相互相干,信号的时间特性与窄带的情形类似.

3 实验与结论

在以 3S 为基态,3P 为中间态,6S 和 5D 为激发态的钠蒸气四能级系统中完成了双频时延四波混频实验,让脉宽为 6 ns,线宽为 0.1 nm 的三台染料激光器波长 589 nm,515.4 nm,498.3 nm 分别对应于 $3S_{1/2}$ 到 $3P_{\frac{3}{2}}$,$3P_{\frac{3}{2}}$ 到 $6S_{\frac{1}{2}}$,$3P_{\frac{3}{2}}$ 到 $5D_{\frac{3}{2},\frac{5}{2}}$ 的偶极跃迁.得拍频信号周期 $T = 2\pi|\omega_3 - \omega_2|^{-1} = 50$ fs(见图 5).调制频率 $\omega_d = \omega_3 - \omega_2 = 126\ \mathrm{ps}^{-1}$(见图 6).

图 3、图 4 分别对应于泵光 2,3 仅含 ω_2 分量的第一个双光子自相干过程,泵光 2,3 仅含 ω_3 分量的第二个双光子自相干过程.从图中可以看出,第一个自相干过程的信号强度关于 $\tau = 0$ 对称,而第二个自相干过程的信号强度关于 $\tau = \delta\tau$ 对称.由于两光源自相关零延时的偏差将导致拍频信号强度峰值右移(如图 5 所示).

在四能级系统极化拍频实验中,由于不对称光路的元件色散导致两个自相干过程零延时存在偏差,致使信号强度峰值右移,且随着频差 $\omega_3 - \omega_2$ 的增加,不对称现象越来越明显.但其调制频率的测量并不受影响.也就是采用相位共轭几何配制,容许有一定机械振动,光学元件的畸变及其他微扰,而不影响测量精度.

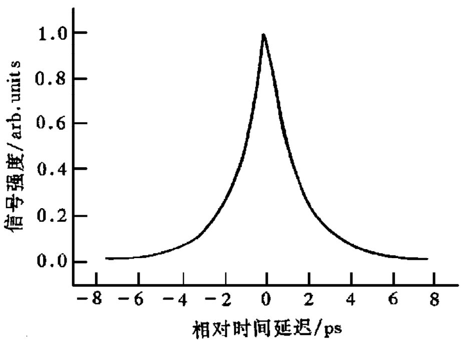

图 3 泵光仅含 ω_2 时 FWM 信号随延时的变化

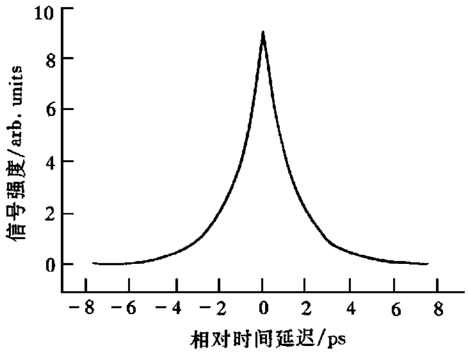

图 4 泵光仅含 ω_3 时 FWM 信号随延时的变化

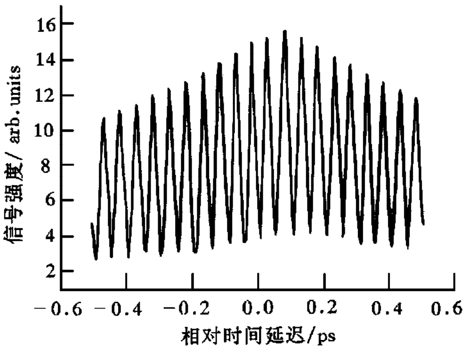

图 5 拍频信号随延时的变化

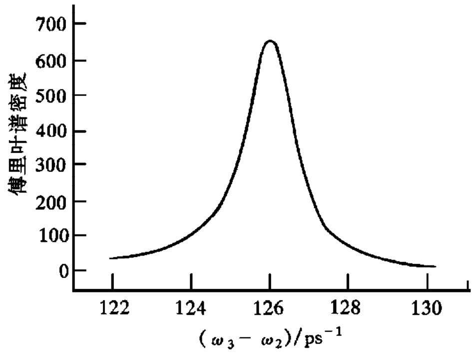

图 6 延时 τ 在 15 ps 范围中的实验数据傅里叶图谱

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ASYMMETRIC BEHAVIOR OF THE POLARIZATION BEATS SIGNAL IN A FOUR-LEVEL SYSTEM*

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ABSTRACT

We have studied the asymmetric behavior of the polarization beats in a four-level system when the pump beams have either narrowband or broadband linewidth, we attribute this asymmetry to the shift of the zero time delay which is due to the dispersion of the optical components.

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