Four-wave mixing and its relaxation effect in liquid crystals

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We have observed degenerate and nondegenerate four-wave mixing in liquid crystals MBBA and p-methoxybutyl oxyazobenzene in their isotropic phases (at temperatures higher than 42°C and 70°C respectively). The experimental setups are shown in Fig. 1 and Fig. 2. respectively. Beam 4 is the mixed output. The mixing efficiency (intensity ratio of beam 4 to beam 3) was about 2×10^{-3} and 1×10^{-4} respectivcly. The parameters for the SHG of YAG laser are as follows: wave length, 5320 Å; pulse width, 14ns; output energy, 5-6mJ; cross-section of the beams, about 1 mm². Those for the flash-lamp-pumped dye laser are: wavelength 5800 Å; pulse width, 0.5μs; output energy, 2 mJ. The SHG of the YAG laser and the dye laser output could be synchronized or adjusted to have a relative delay. The angle between beams 1 and 3 was about 0.08 rad. The fluctuation of the sample's temperature was less than 0.01°C.

Using the same setup as shown in Fig. 2 and adjusting the relative delay between the SHG pulses (i.e. beams 1 and 3) and the dye laser pulse (i.e. beam 2), a pronounced relaxation effect in four-wave mixing have also been observed in both kinds of liquid crystal. The following results were obtained:

(1) When the SHG pulse occurred before the dye laser pulse (up to 10 μs), mixed output could be observed, and its waveform was similar to that of the dye laser (Fig. 3).

(2) When the SHG pulse was on the front edge or on the back edge of the dye laser pulse, the waveform of the mixed output was such that before the arrival of the SHG pulse there was no mixed output, although there was an action of the dye laser (Fig. 4 and Fig. 5).

(3) When the SHG pulse occured after the dye laser pulse without any overlapping, mixed output did not appear at all.

In order to explain the results described above, two mechanisms must be introduced together. One of them is the thermal effect. That is, in liquid crystals, a thermally induced index grating may be produced by the action of beams 1 and 3. Thus, while beam 2 is switched on during the relaxation period of that grating, beam 4 can be produced by Bragg reflection. This explains why the relaxation time of the four-wave mixing is so long in comparision with that of molecular reorientation. The other mechanism is the nonlinear polarization effect due to molecular reorientation. That is, coincident action on liquid crystals by the electric fields of beams 1 and 3 results in molecular reorientation and ordering to some degree. Then, during the relaxation period of that ordering, nonlinear polarization with frequency ω may be produced when there is a coincident action of beam 2. Beam 4 is just the radiation from this polarization.

In order to get rid of the thermal effect, the polarization discrimination method was adopted. That is, the linear polarizations of beams 1 and 2 were made parallel to each other but orthogonal to that of beam 3. In this case, the thermal grating described above

cannot be induced, and the mixed output is then only due to the nonlinear polarization corresponding to the susceptibility x_{1221} related to molecular reorientation. This experiment has been performed and beam 4 observed. The polarization direction of beam 4 was indeed the same as that of beam 3. The measurement of the relaxation time is still in progress.

液晶中的四波混频及其弛豫效应

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我们已经在 MBBA 和氧化偶氮苯两种液晶的各向同性相(温度分别在 42°C和 70°C以上)观察到了简并和非简并四波混频。混频效率分别约为 2×10^{-3} 和 1×10^{-4}。YAG 激光倍频的波长为 5320Å，脉宽 14ns，输出能量 5～6mJ，光束截面约为 1mm²。闪光灯泵浦的染料激光的波长为 5800Å，脉宽 0.5μs，输出能量 2mJ。两台激光器的输出可以同步或调节相对延时。两光束间的夹角约为 0.08 rad。样品温度起伏小于 0.01°C。

改变倍频脉冲和染料激光脉冲之间的相对延时，在这两种液晶中都观察到了四波混频的明显的弛豫效应。结果为：

(1)当倍频脉冲出现在染料激光脉冲之前时(直到超前 10μs)都能观察到混频输出，其波型与染料激光波形相似。

(2)当倍频脉冲落在染料激光脉冲前沿或后沿时，倍频脉冲到达之前虽然也有染料激光作用，却没有混频输出。

(3)当倍频脉冲出现在染料激光脉冲之后没有重迭时，就完全不出现混频输出。

为解释上述结果，必须同时引入两种机构。一为热效应。即在液晶中由于两光束的作用产生热致折射率光栅。若在这一光栅的弛豫时间之内打开染料激光脉冲光束，即可由布喇格反射得到混频输出光束。这就解释了为什么四波混频的弛豫时间要比分子重新取向的弛豫时间长得多。另一种机构是由于分子重新取向而产生的非线性极化效应。即两光束电场同时作用于液晶，使液晶分子重新取向，并达到一定程度的有序化。在这一有序化的弛豫时间内，若有染料激光脉冲光束同时作用，即可以产生频率为 ω 的非线性极化。混频输出光束正是由这一极化产生的辐射。

为消除热效应，采用了偏振鉴别方法。令两光束取相互平行的偏振方向，而与倍频脉冲光束的偏振方向相垂直。此时不能产生上述的热致光栅，因此混频输出仅由与极化率 x_{1221} 相对应的非线性极化所产生。用这一实验方法已经观察到混频输出光束。并且混频输出光束的偏振方向确实与倍频脉冲光束相同。目前正在测量弛豫时间。