RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 33, NUMBER 6 JUNE 1986

Squeezing in nondegenerate four-wave mixing

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(Received 16 September 1985)

Ways to minimize spontaneous-emission noise due to quantum fluctuations in the medium are a major consideration in experiments designed to produce squeezed states of light. Motivated by the recent experiments of Slusher *et al.* we present a fully quantized model of nondegenerate four-wave mixing. We show that the spontaneous-emission noise, important at higher intensities in the degenerate case, may be minimized in the nondegenerate case for certain regimes of cavity and medium parameters.

I. INTRODUCTION

There is presently a considerable experimental[1–4] effort to generate a squeezed state of light.[5] Experimental efforts by Shapiro and co-workers,[3] Levenson and co-workers,[2,4] and Slusher *et al.*[1] have concentrated on four-wave mixing schemes,[6–9] first predicted to give squeezing by Yuen and Shapiro.[6] However, their model neglected quantum fluctuations due to the medium. A fully quantized treatment of a medium modeled as N two-level atoms was carried out by Reid and Walls for degenerate four-wave mixing[8] and optical bistability.[10] Calculation of the output squeezing at the pump frequency showed two effects due to the medium that must be minimized if one is to obtain large squeezing; atomic loss, more important at low intensities, and spontaneous emission, important at higher intensities, as one begins to saturate the transition. The implication for a two-level medium in the totally degenerate scheme is that high pump intensities, high cavity cooperativity parameters, and high atomic detunings were required.

In this Rapid Communication we examine a means of minimizing the spontaneous emission induced from a two-level atomic medium at higher intensities. The initial calculations of Reid and Walls were limited to the degenerate situation, but the present experiments employ a nondegenerate four-wave mixing scheme. Thus, this Communication is motivated by recent comments made by Slusher *et al.*[1] that spontaneous emission is reduced in the nondegenerate four-wave mixing scheme they employ. Our work reveals for which regime of cavity and medium parameters this is indeed true. Results indicate a more optimistic picture for squeezing in terms of parameters attainable in experimental situations.

II. QUANTUM THEORY

We begin with a general description of nondegenerate four-wave mixing in an optical cavity. The medium is modeled as N two-level atoms with resonance frequency ω_0 and is interacting with three cavity modes of frequencies ω_1, ω_2, and ω_3. The cavity mode spacing is $\epsilon=\omega_2-\omega_1=\omega_1-\omega_3$. The amplitudes of the cavity modes are denoted by α_j. All three cavity modes are assumed to have the same cavity damping rate κ. The central cavity mode α_1 is driven by an external coherent input field ε of frequency ω_L. The cavity detuning $\omega_1-\omega_L$ is assumed to be much smaller than the cavity mode spacing ϵ. The atomic longitudinal and transverse decay rates are $\gamma_\parallel$ and $\gamma_\perp$, respectively. The collision parameter $f=\gamma_\parallel/2\gamma_\perp$ is one for pure radiative damping and zero in the large collisional limit.

We proceed as in Reid and Walls[8] to derive quantum c number Langevin equations for the field mode amplitudes. We are interested in the equations describing the gain of the weak field modes α_2,α_3 in the presence of a very strong pump mode α_1. We thus treat α_1 to all orders, describing completely the saturation of the medium, while the expressions for the weak-field modes α_2,α_3 are kept to first order only.

In the limit of a high Q cavity ($\kappa \ll \gamma_\perp,\gamma_\parallel$) one is justified in adiabatically eliminating the atomic variables to obtain final equations for the field modes as follows.

$$\dot{\alpha}_1=\varepsilon-\kappa(1+i\phi)\alpha_1\frac{2C\kappa\alpha_1}{(1+i\Delta_1)\Pi}+F_1(t)\ , \tag{1a}$$

$$\dot{\alpha}_2=-\gamma(\delta)\alpha_2+\chi(\delta)\alpha_3^\dagger+F_2(t)\ , \tag{1b}$$

$$\dot{\alpha}_3=-\gamma(-\delta)\alpha_3+\chi(-\delta)\alpha_2^\dagger+F_3(t)\ , \tag{1c}$$

where we write

$$\gamma(\delta)=\kappa(1+i\phi)+\gamma_R(\delta)+i\gamma_I(\delta)\ ,$$

$$\chi(\delta)=\chi_R(\delta)+i\chi_I(\delta)\ ,$$

$$\Pi=\left(1+\frac{X}{1+\Delta_1^2}\right)\ ,$$

and the nonzero noise correlations for the sidebands are

$$\begin{aligned}&\langle F_2(t)F_3(t')\rangle=R\delta(t-t'),\quad R=R_R+iR_I\ ,\\&\langle F_2^\dagger(t)F_3^\dagger(t')\rangle=R^*\delta(t-t')\ ,\\&\langle F_2(t)F_2^\dagger(t')\rangle=\langle F_3(t)F_3^\dagger(t')\rangle=\Lambda\delta(t-t')\ .\end{aligned} \tag{2}$$

The parameters γ,χ,R, and Λ are functions of the following scaled variables: the cavity detuning $\phi=(\omega_1-\omega_L)/\kappa$, the detuning of the pump from the medium $\Delta_1=(\omega_0-\omega_L)/\gamma_\perp$, the detuning of the sidebands from the pump $\delta=-\epsilon/\gamma_\parallel$, the cavity cooperativity parameter $C=g^2N/2\gamma_\perp\kappa$, the collisional parameter f, and the scaled intracavity steady-state pump intensity $X=|\alpha_1|^2/n_0$. $n_0=\gamma_\parallel\gamma_\perp/4g^2$ is the resonant saturation intensity and $|\alpha_1|^2$ is determined by the optical bistability state equation[11] which is the steady-state deterministic solution of (1a). The full explicit solutions are presented in the Appendix.

We have assumed in the derivation of Eq. (1) that

$\Delta k = 2k_1 - k_2 - k_3$ is zero. This is always satisfied for the completely forward configuration, but not, in general, for alternative relative orientations of the field propagation vectors.

III. RESULTS

The output light is peaked about the frequencies ω_L and $\omega_L \pm \epsilon$. We have calculated the squeezing in the output light at the two sideband frequencies. Squeezing is observed in a homodyne detection scheme where the output of the two sidebands at frequency $\omega_L \pm \epsilon$ beat with a local oscillator $\varepsilon_{LO} = \varepsilon e^{i\theta}$ at frequency ω_L. The local oscillator is obtained by phase shifting the external driving field. The sidebands and the local oscillator mix on the surface of a photodetector giving a photocurrent $i(\epsilon)$. The spectrum of fluctuations in this photocurrent $\langle i^2(\epsilon)\rangle$ is measured with a spectrum analyzer. $V(X_\theta,\epsilon) = \frac{1}{2}\langle X_\theta^\dagger X_\theta + X_\theta X_\theta^\dagger\rangle - \langle X_\theta\rangle\langle X_\theta^\dagger\rangle$ is the spectral "variance" of the quadrature phase $X_\theta = a_{2\,\mathrm{out}} e^{-i\theta} + a_{3\,\mathrm{out}} e^{i\theta}$ and is directly proportional to $\langle i^2(\epsilon)\rangle$. Squeezing is characterized by $V(X_\theta,\epsilon) < 1$.

The key question is the orders of magnitude required for the experimental parameters Δ_1, δ, $2C$, f, and X to obtain squeezing. The recent studies by Reid and Walls of degenerate four-wave mixing[8] and optical bistability[10] in a two-level medium have pointed to at least three important physical effects due to the medium which may limit the squeezing. These are the loss γ_R, the spontaneous emission Λ, and the degree f of collisional phase damping. The general principle is to enhance the nonlinear gain terms χ_I, R_I responsible for squeezing over both the loss γ_R (important at lower intensities) and the dephasing quantum noise terms (important at higher intensities). In such a regime of parameter space the four-wave mixing may be described by an idealized Hamiltonian based on a classical susceptibility for the medium.[6,12]

In the degenerate case, the general requirements were to operate in the dispersive regime ($\Delta_1 >> 1$), well below saturation ($X << \Delta_1^2$) and to have pure radiative damping ($f = 1$). In this limit the gain to loss ratio of the medium $|\chi|/\gamma_R \approx X/\Delta_1$ could be enhanced by increasing the pump intensity X, except that one had to be careful not to induce additional spontaneous emission Λ due to increased population of the upper level. The requirement that $\Lambda/|\chi|$ be small is $X^2/\Delta_1^3 << 1$. In order to attain threshold ($|\chi|^2 \geq |\gamma|^2$) without needing to increase X such that spontaneous emission was induced, the cooperativity parameter $2C$ needed to be of the right order ($2C \gtrsim \Delta_1$). Yet one could not increase C to the extent that collisional damping was important, or that the atomic loss due to the medium was significant compared to the cavity loss (one needed $2C << \Delta_1^2$). The combined effect of both the atomic loss γ_R and spontaneous emission Λ was to demand high detunings ($\Delta_1 \sim 10^3$), high pump intensities ($X \sim \Delta_1$), and high C values ($C \gtrsim 10^4$).

The nondegenerate scheme ($\delta \neq 0$) offers advantages over the degenerate situation. The dephasing spontaneous-emission noise Λ is due to the pump saturating the two-level atom and is peaked about the pump frequency. Thus, we look for a reduction in this noise as we increase δ. The limit of interest is that of large detuning of the pump relative to the atomic resonance ($\Delta_1 >> 1$), pure radiative damping ($f = 1$), and a detuning δ of the sidebands relative to the pump, but such that $\delta < \frac{1}{2}\sqrt{\Delta_1^2 + 2X}$. Then one finds a regime where γ and χ are essentially unchanged from the degenerate case, yet the desqueezing term Λ is reduced by the factor $(1+\delta^2)$. The condition to avoid onset of spontaneous emission as one increases the pump intensity is now somewhat looser; $X^2/\Delta_1^3 << (1+\delta^2)$. Thus, one can afford higher pump intensities (X/Δ_1) relative to the pump detuning. The implication is that squeezing becomes possible at lower pump detunings and hence, most importantly, for lower values of the cooperativity parameter C. There is also a somewhat lesser reduction in pump powers necessary.

The dispersion term γ_I incorporates the (linear and nonlinear) change in the refractive indices due to the medium and also any would-be phase mismatch Δk that may be present in configurations where the k_j vectors have relative orientations. We analyze in the first instance the simplest situation where the Δk is chosen to allow perfect phase matching in the medium, that is, so that $\gamma_I = 0$ for both sidebands. Also, the pump mode α_1, distinguishable from the sidebands by a different k direction, may or may not be in a cavity, but any cavity detuning ϕ is zero. Thus no bistability exists in this situation.

The variance of fluctuations at the sideband frequencies $V(X_\theta,\epsilon)$ for this particular case is plotted in Figs. 1 and 2 for a scaled detuning $\Delta_1 = 100$ of the pump from the atomic resonance. Solutions hold only about a stable steady state ($\alpha_2 = \alpha_3 = 0$), and the stop in the curves in Fig. 1 indicates the onset of oscillation (threshold corresponds to $|\chi| = \kappa + \gamma_R$). The advantage obtainable in the nondegenerate case is immediately apparent. In the degenerate case, $\delta = 0$, the conditions $X/\Delta_1 >> 1$ and $X^2/\Delta_1^3 << 1$ are optimized at $X \sim 400$ (Fig. 1). The optimal C value is, in fact, that plotted ($2C \sim 3500$) in Fig. 1. Even at the relatively high detuning $\Delta_1 \sim 100$ perfect optimization has not been possible for $\delta = 0$, and the effect of atomic loss is still to limit the squeezing attainable. Thus in the degenerate situation, one would have to increase the detuning to improve squeezing ($\Delta_1 \sim 10^4$, $2C \sim 10^6$, $X \sim 10^5$).

The conditions for the nondegenerate situation $X/\Delta_1 >> 1$ and $X^2/\Delta_1^3 << 1+\delta^2$ mean that, for a given detuning Δ_1, one can increase intensities to a regime where loss is not important and still obtain good squeezing, as shown in Fig. 2, for a C value of 500. Thus, good squeezing is obtainable

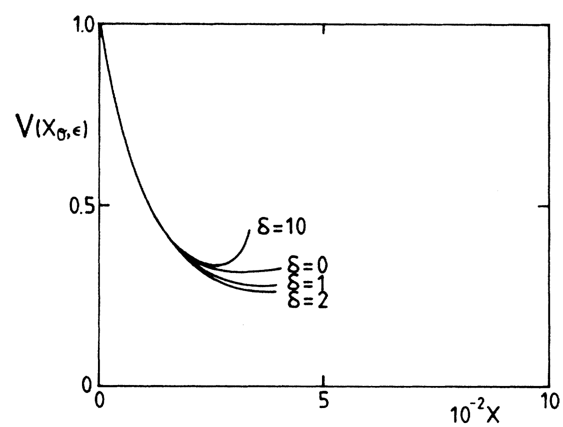

FIG. 1. High-loss situation (perfect phase matching): Nondegeneracy δ has little effect on squeezing. $V(X_\theta,\epsilon)$ vs scaled pump intensity X. $\Delta_1 = 100$, $2C = 3500$, $\phi = 0$.

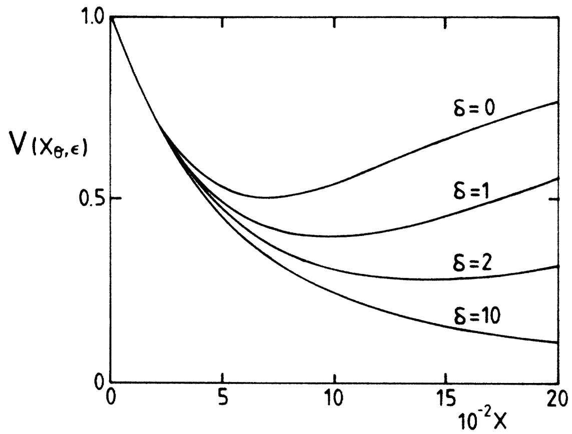

FIG. 2. Low-loss situation (perfect phase matching): Nondegeneracy δ has major effect on squeezing. $V(X_\theta,\epsilon)$ vs scaled pump intensity X. $\Delta_1=100$, $2C=500$, $\phi=0$.

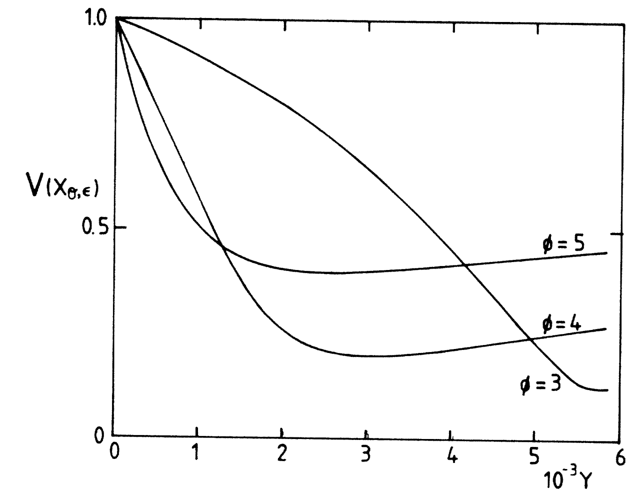

FIG. 3. Low-loss situation (forward configuration): The effect of cavity detuning ϕ. $V(X_\theta,\epsilon)$ vs scaled external pump intensity $Y=|\varepsilon/\kappa|^2/n_0$. $\Delta_1=100$, $2C=500$, $\delta=10$.

for lower detunings Δ_1 and hence, also for much lower C values. To avoid the regime of high atomic loss relative to the cavity loss, which still limits the squeezing possible and for which the nondegenerate situation is no advantage, we still stringently require $2C \ll \Delta_1^2$. This feature is illustrated in Fig. 1, where $2C \sim \Delta_1^2$ and for which little improvement is seen upon increasing δ. One still needs $2C \sim \Delta_1$ (to avoid reaching total saturation before a reasonable order of X is obtained), but the sensitivity to this latter criterion is very much reduced, and good squeezing is possible for a wider range of C values compared to the degenerate case. The advantage in terms of pump powers is not so significant, since we have to pump the medium harder to gain advantage. However, the lower detunings needed do imply a net reduction in minimum powers needed to attain the same amount of good squeezing.

Perfect phase matching is not always achievable. However, the presence of the dispersive term γ_I, particularly with the nonzero cavity detuning ϕ, does not necessarily reduce squeezing. The threshold condition is changed ($|\chi|^2=|\gamma|^2$) and also the phase θ required for optimal squeezing will, in general, be different. The latter feature may well be an advantage in experiments where a particular choice of θ is required to reduce nondesirable phase sensitive noise.[13]

Next we consider the completely forward configuration in which all modes propagate in a single ring cavity driven by an external pump, detuned ϕ cavity linewidths with respect to the internal pump α_1. Equations (1) derived describe this situation directly. This system can show bistability with respect to the external driving field intensity. The bistability criteria and the squeezing in the output resonant pump mode have been examined previously.[10,11] A rather simplified criterion for bistability is to require a cavity detuning such that $\phi < 2C/\Delta_1$, provided C is large in terms of the detuning Δ_1. For the situation we are currently presenting, where $|\delta| \ll \frac{1}{2}\sqrt{\Delta_1^2+2X}$, the δ and χ for the sidebands are essentially unchanged from the values at $\delta=0$. Hence, the sidebands and pump have identical stability/instability regimes, the stable regions corresponding to $|\chi|^2<|\gamma|^2$. The steady-state deterministic solutions for the pump is the optical bistability state equation,[11] while for the sidebands the solution is $\alpha_2=\alpha_3=0$. In the idealized situation discussed above we have $\Delta_1 \gg 1$ (dispersive limit), $\gamma_R=0$ or negligible atomic loss both relative to the cavity loss ($2C \ll \Delta_1^2$) and to the gain ($X>\Delta_1$), $X \ll \Delta_1^2$ (low saturation), $X^2 \ll (1+\delta^2)\Delta_1^2$ (low spontaneous emission), and $|\delta| \ll \frac{1}{2}\sqrt{2X+a^2}$. The use of effective Hamiltonians becomes valid, and the system is identical to that studied recently by Levenson *et al.*[13] (but where ϕ becomes $\phi-2C/\Delta_1$ incorporating the linear dispersion). For this idealized situation, perfect squeezing is attainable at the sideband frequencies at the critical points where the pump (and sidebands) become unstable.

Full solutions for squeezing in the case $\Delta_1=100$ are presented in Fig. 3. In the low C ($2C \approx \Delta_1$) situation the nondegenerate scheme is advantageous over the degenerate ($\delta=0$) scheme. In fact, for such low C values (compared to Δ_1^2) the internal intensity X (a minimum of $X \sim 4000$ for $\phi \sim 4.5$) required to reach bistability under the ideal conditions is high enough to cause saturation ($X/\Delta_1^2 \sim 0.4$) and bistability is not observed. In fact, the idealized situation studied by Levenson *et al.*[13] is not attainable for $\Delta_1=100$. Figure 3 shows the dependence of the squeezing on the cavity detuning ϕ. Unlike the degenerate situation, where one found a high sensitivity with respect to ϕ (the optimal ϕ being the highest still giving bistability), the nondegenerate case allows good squeezing for a relatively large range of ϕ.

For higher C values one obtains bistability with appropriate ϕ. In this case one obtains some advantage in the nondegenerate situation only if operating in an appropriate regime on the upper branch. However, a high absolute value of loss compared to the cavity loss ($2C$ not much less than Δ_1^2) will tend to limit the order of squeezing possible.

IV. CONCLUSION

We have considered nondegenerate four-wave mixing via an N two-level atomic medium in a cavity in which the weak-field modes are detuned from the pump by 2δ atomic natural linewidths in the pure radiative damping limit. The squeezing in the output field at the weak-field frequencies is computed. The key parameters describing the system are the detuning Δ_1 of the pump from the atoms in natural atomic linewidths, and the intracavity intensity X scaled in terms of the resonant saturation intensity n_0. We have established, in a limit where $|\delta| \ll \frac{1}{2}\sqrt{a^2+2X}$, approximate guidelines so that unfavorable noise contributed from the medium is minimal. These are $\Delta_1 \gg 1$, $X \gg \Delta_1$, $X \lesssim \Delta_1^2$, $X^2 \ll (1+\delta^2)\Delta_1^2$, $2C \ll \Delta_1^2$, and $2C \gtrsim \Delta_1$. In a completely forward configuration, where perfect phase matching in the medium is not possible, one can attain good squeezing by allowing a cavity detuning between the external and internal driving fields of ϕ cavity linewidths, of the order $\phi \lesssim 2C/\Delta_1$.

The important feature is that the frequency shift δ allows spontaneous emission contributed from the medium and peaked about the pump frequency to be minimal. The result is better squeezing possible at lower detunings Δ_1 and ϕ, much lower C values, lower pump intensities, and also a broader range of these parameters, thus considerably improving possibilities of obtaining squeezing in such experiments.

ACKNOWLEDGMENT

We wish to thank the New Zealand University Grants Committee for financial support.

APPENDIX

Defining

$$\Delta_2=\Delta_1+2\delta f,\quad \Delta_3=\Delta_1-2\delta f \;,$$

the solutions are

$$\gamma_R(\delta)=\frac{2C\kappa\{1+X[a-c+\Delta_2(d-b)]+X^2[bd-ac+\Delta_2(ad+cb)]\}}{(1+\Delta_2^2)\Pi(0)|\Pi(\delta)|^2}\;,$$

$$\gamma_1(\delta)=\frac{2C\kappa\{-\Delta_2+X[d-b-\Delta_2(a-c)]+X^2[ad+bc-\Delta_2(bd-ac)]\}}{(1+\Delta_2^2)\Pi(0)|\Pi(\delta)|^2}\;,$$

$$\chi_R(\delta)=2C\kappa X\{e\mp\delta fq+X[ae+bq+\delta f(be-aq)]\}D^{-1},\quad \chi_I(\delta)=2C\kappa X\{q+\delta fe+X[aq-be+\delta f(ae+bq)]\}D^{-1}\;,$$

$$\Lambda=2C\kappa X^2[1+\Delta_3^2+f-f\Delta_1\Delta_3+\delta(\Delta_1+\Delta_3)+Xf/2]D^{-1},\quad R_R=-2C\kappa X[f(1+\delta^2)r+XA(r,s)+X^2fB(r,s)]D^{-1}\;,$$

$$R_I=-2C\kappa X\{-f(1+\delta^2)s+X[A(-s,r)+\Delta_2+\Delta_3-\Delta_2\Delta_3+1]+X^2f[B(-s,r)\Delta_1/4-\tfrac{1}{4}]\}D^{-1}\;,$$

where

$$D=\Pi(0)|\Pi(\delta)|^2(1+\delta^2)(1+\Delta_1^2)(1+\Delta_2^2)(1+\Delta_3^2),\quad a=\frac{2+\Delta_3^2+\Delta_2^2-\delta\Delta_2(1+\Delta_3^2)+\delta\Delta_3(1+\Delta_2^2)}{2(1+\delta^2)(1+\Delta_2^2)(1+\Delta_3^2)}\;,$$

$$b=\frac{-\delta(2+\Delta_3^2+\Delta_2^2)-\Delta_2(1+\Delta_3^2)+\Delta_3(1+\Delta_2^2)}{2(1+\delta^2)(1+\Delta_2^2)(1+\Delta_3^2)},\quad c=\frac{(\Delta_3-\Delta_1)(\Delta_3+\Delta_1-\delta+\delta\Delta_3\Delta_1)}{2(1+\delta^2)(1+\Delta_3^2)(1+\Delta_1^2)}\;,$$

$$d=\frac{(\Delta_3-\Delta_1)[1-\Delta_3\Delta_1+\delta(\Delta_3+\Delta_1)]}{2(1+\delta^2)(1+\Delta_3^2)(1+\Delta_1^2)},\quad e=1+\Delta_3\Delta_1+\Delta_2\Delta_3-\Delta_2\Delta_1+\delta(\Delta_3-\Delta_1-\Delta_2-\Delta_2\Delta_3\Delta_1)\;,$$

$$q=\Delta_3-\Delta_1-\Delta_2-\Delta_2\Delta_3\Delta_1-\delta(1+\Delta_3\Delta_1+\Delta_2\Delta_3-\Delta_2\Delta_1),\quad \Pi(\delta)=\Pi_R+i\Pi_I,\quad \Pi_R=1+aX,\quad \Pi_1=bX\;,$$

$$A(r,s)=f(1+\delta^2)2ar-\frac{f(rg+ms)}{(1+\Delta_2^2)(1+\Delta_3^2)}-1+\Delta_2\Delta_3\;,$$

$$B(r,s)=\tfrac{1}{4}+(1+\delta^2)(a^2+b^2)r-\frac{[rag+rbh+ams+bns-r(1-\Delta_2\Delta_3)/4+s(\Delta_2+\Delta_3)/4]}{(1+\Delta_2^2)(1+\Delta_3^2)}\;,$$

$$r=1-\Delta_2\Delta_3-\Delta_1\Delta_2-\Delta_1\Delta_3,\quad s=\Delta_2+\Delta_3+\Delta_1-\Delta_1\Delta_2\Delta_3,\quad 2g=(1+\Delta_3^2)(1-\Delta_2\delta)+(1+\Delta_3\delta)(1+\Delta_2^2)\;,$$

$$2h=-(1+\Delta_3^2)(\Delta_2+\delta)+(1+\Delta_2^2)(\Delta_3-\delta),\quad 2m=-(1+\Delta_3^2)(\Delta_2+\delta)-(1+\Delta_2^2)(\Delta_3-\delta)\;,$$

$$2n=-(1+\Delta_3^2)(1-\Delta_2\delta)+(1+\Delta_2^2)(1+\Delta_3\delta)\;.$$

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