Observation of nonlinear optical phenomenon in vacuum by four waves mixing

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Abstract
In the present paper, we studied possibility of observation and detection of nonlinear optical effects in free space. We studied four wave mixing process in which signal is generated opposite to the direction of probe beam. We observed that large number of photons are generated in signal beam, which should be detectable.

Keywords Nonlinear optics · Maxwell’s equations · Four wave mixing · Nonlinear Lagrangian for electromagnetic field

1 Introduction
Nonlinear interaction between electromagnetic fields in vacuum are well known in Quantum Electrodynamics (Halpern 1933; Heisenberg and Euler 1936; Schwinger 1951; Karplus and Neuman 1950, 1951; Kenna and Platzmann 1963; Heinzl 2006; Klein and Nigam 1964; King et al. 2010a). In quantum electrodynamics the interaction between two photons in vacuum and production of virtual electron positron has been already described (Halpern 1933; Heisenberg and Euler 1936; Schwinger 1951; Karplus and Neuman 1950, 1951; Kenna and Platzmann 1963) and the possibility of measuring various signals due to elastic scattering of photons of one another has been investigated theoretically (Ferrando 2007; King et al. 2010b; Tommasini and Michinel 2010; Monden and Kodama 2011; Kryuchkyan and Hatsagortsyan 2011; King and Keitel 2012). The self-interaction of photon can also be described in terms of Feynman diagram and for two photon interaction the Nonlinear Lagrangian for electromagnetic field (Karplus and Neuman 1951), which is correct to terms of order $e^4$.

$$L = \frac{1}{8\pi} [E^2 - B^2 + K((E^2 - B^2)^2 + 7(E.B)^2)], \quad K = \frac{e^4}{45\pi^4m^4}$$ (1)
in the natural system of units, $c = \hbar = 1$. Here, $E$ is electric field intensity vector, $B$ is magnetic flux density vector, $e$ is electronic charge, $m$ is electronic mass.

This Lagrangian is identical with the results of Euler and Schwinger Heisenberg and Euler (1936) and Schwinger (1951). Kenna and Platzmann (1963) are first who studied this Lagrangian to study the nonlinear interaction of light in vacuum and they obtained the modified Maxwell’s equations. Earlier some authors have been used these modified Maxwell’s equation to study Birefringence of vacuum and other effects (Heinzl 2006; Klein and Nigam 1964; King et al. 2010a; Baier and Breitenlochner 1967; Byalynicka-Birula and Byalynicki-Birula 1970; Brezin and Itzykson 1971; Prakash and Shukla 2007; Bakalov 1980). Recently Stefan Ataman introduced experimental setup to detect the QED predicted vacuum birefringence (Ataman 2018).

Earlier we used this Lagrangian (Karplus and Neuman 1950) to solve the problem of birefringence of vacuum in presence of a counterpropagating electromagnetic wave (Prakash and Shukla 2007). In earlier work we studied the birefringence of vacuum in presence of a counter propagating electromagnetic wave and found that quite large number of photons are generated in the orthogonal mode. The detection of these photons is however difficult because of non-availability of pure polarized light. The newly generated photons are far too less in number than those present initially in this mode. We note that, this problem arises due to the identity of the generated photons being the same as that of some photons present initially. We remove this in the present paper by giving a new identity to the newly generate photons. In this paper we tried to give an independent identity to the newly generated photons via momentum conservation process such as four wave mixing.

Degenerate four wave mixing was proposed by Hellworth (1977). All the four optical fields involved in this mixing process are of the same frequency $\omega$ hence the name degenerate. In this process two pump waves injected light of the same frequency $\omega$ and wave vectors $k_1(\omega)$ and $k_2(\omega)$, from opposite direction through a nonlinear medium. Then when a third wave of frequency $\omega$ (probe wave) and wave vector $k_3(\omega)$ is injected, phase conjugate light of the same frequency emerged in opposite direction. This emergent light has the wave vector as given below:

$$k_4(\omega) = k_1(\omega) + k_2(\omega) - k_3(\omega) = -k_3(\omega)$$

This is the phase matching condition. When the probe frequency is different from that of the pump wave, we have nondegenerate four wave mixing. This is a standard method for generating phase conjugate light by using gases, liquids, dielectrics, semiconductors, organic materials, and non-crystalloids etc. Degenerate four wave mixing was proposed almost the same time by Yariv and Pepper (1997). The experiment that first suggested the practical value of degenerate four wave mixing was performed in 1977 (Bloom and Bjorklund 1977). Earlier Four Wave mixing have been used to study some important effects in vacuum (Beutler et al. 2010; Silva et al. 2011; Babushkin et al. 2009; Renger et al. 2009; Dorman et al. 1998; Moulin and Bernard 1999; Lundin 2006; Bernard et al. 2000). Four wave mixing was first used in quantum optics to produce quadrature squeezed light (Yuen and Shapiro 1979). Later, several authors studied squeezing in Four wave mixing (Kumar and Shapiro 1984; Ried and Walls 1984, 1985; Yurke 1985; Shukla and Prakash 2013; Prakash and Shukla 2012, 2015; Giri and Gupta 2004). Four wave mixing has some more applications such as, detection of violation of Cauchy-Schwartz and Bell’s inequality (Nadeem et al. 1988), Quantum effects of the atom cavity interaction (Varda et al. 1987). Other schemes also have been proposed to generate quadrature squeezed light by some authors (Mishra 2010;
Prakash and Mishra 2010, 2016; Mishra et al. 2020, 2021; Mishra and Singh 2020). In this paper we study four waves mixing to observe nonlinear optics in vacuum.

Lagrangian (1) leads to the usual Maxwell equations,

\[ \nabla \mathbf{D} = 0, \quad \nabla \mathbf{B} = 0, \quad \nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t}, \quad \nabla \times \mathbf{H} = \frac{\partial \mathbf{D}}{\partial t}. \]  

(2)

but with the constitutive relations,

\[ \mathbf{D} = \mathbf{E} + K[2(\mathbf{E}^2 - \mathbf{B}^2)\mathbf{E} + 7(\mathbf{E}\cdot\mathbf{B})\mathbf{B}], \]  

(3)

\[ \mathbf{H} = \mathbf{B} + K[2(\mathbf{E}^2 - \mathbf{B}^2)\mathbf{B} - 7(\mathbf{E}\cdot\mathbf{B})\mathbf{E}]. \]  

(4)

for free space. Where, \( \mathbf{D} \) is electric flux density vector and \( \mathbf{H} \) is magnetic field intensity vector and other terms are same as defined in Eq. (1).

2 General consideration for mixing of four waves moving along or against any direction

If we consider that four participating beams travel in the same direction given by, say, the unit vector \( \hat{e}_3 \), the electric field can be written as \( \mathbf{E} = \hat{e}_3 E(t - \hat{e}_3 \cdot \mathbf{x}) \) and the magnetic field is \( \mathbf{B} = [\hat{e}_3 \times \mathbf{E}] \). For such a case, \( \mathbf{E}^2 - \mathbf{B}^2 = \mathbf{E}\cdot\mathbf{B} = 0 \) and no nonlinear effects can be seen.

In view of energy and momentum consideration, the other possible situation is where a pair of beams move forward and the other backward. If we consider a probe beam of frequency \( \omega_p \) moving along \( \hat{e}_3 \) and the two pump beams of frequency \( \omega_0 \), one moving along \( \hat{e}_3 \) and the other along \( -\hat{e}_3 \), the signal beam of frequency \( \omega_s \) moving along \( -\hat{e}_3 \) will be generated. We may now consider cases where either (i) one photon from each of forward and backward moving pump modes is absorbed or (ii) one photon of probe beam and one of backward moving pump beam are absorbed. In the former case generated signal frequency is \( \omega_s = 2\omega_0 - \omega_p \) but with a small momentum mismatch given by \( k_s - k_p = 2(\omega_p - \omega_0) \). In the latter case, again two situations may arise, viz, (A) where emitted pump photons move forward and backward moving signal photon of frequency \( \omega_s = \omega_p \) is generated with a small momentum mismatch equal to \( 2(\omega_p - \omega_0) \), or (B) where the emitted pump photon moves backward and a forward moving signal photon of frequency \( \omega_s = \omega_p \) and there is no momentum mismatch. The last case describes a phenomenon in which polarization of the probe beam changes due to presence of counter-propagating pump beam and has been studied earlier (Kenna and Platzmann 1963). Because of the momentum mismatch, we have to consider a small interaction volume, which we obtain by considering pulsed pump beams.

For consideration of generation of a signal with a new identity (frequency and direction of propagation), therefore we have to consider following two cases.

**Case I** Absorption from pump beams \( (\omega_0, \hat{e}_3) \) and \( (\omega_0, -\hat{e}_3) \) and emission into probe beam \( (\omega_p, \hat{e}_3) \) and a signal beam \( (\omega_s = 2\omega_0 - \omega_p, -\hat{e}_3) \).  

**Case II** Absorption from probe beam \( (\omega_p, \hat{e}_3) \) and pump beam \( (\omega_0, -\hat{e}_3) \) and emission into pump beam \( (\omega_0, \hat{e}_3) \) and a signal beam \( (\omega_s = \omega_p, -\hat{e}_3) \).
3 Semi classical theory

Generation of signal by nonlinear interaction of the two pump and a probe beam can be considered through the nonlinear terms in Eqs. (3) and (4). For case I, therefore, let us consider the electric field given as

\[ E = E_+ e^{i\psi_+} + E_- e^{-i\psi_-} + E_p^* e^{i\psi_p} + \text{c.c.} \]  

(5)

where c.c. stands for complex conjugate and

\[ \psi_\pm = \omega_0(t \mp x_3), \ \psi_p = \omega_p(t - x_3). \]  

(6)

Here subscripts \( \pm \) refer to the directions \( \pm e_3 \) of the two pump beams and \( p \) to the probe beam. Obviously, we have to look for the terms involving a product of \( E_+, E_-, E_p^* \) in the nonlinear terms of Eqs. (3) and (4) leading to generation of frequency \( \omega_s = 2\omega_0 - \omega_p \), in this case.

Let the generated signal beam be denoted by

\[ E_s = E_s e^{-i\psi_s} + \text{c.c.}, \quad E_s = E_s(x_3), \ \psi_s = \omega_s(t - x_3), \ \omega_s = 2\omega_0 - \omega_p \]  

(7)

We consider \( x_3 \)-dependence of \( E_s \) but not of \( E_\pm \) and \( E_p^* \) because the latter three are supposed to be intense and there is no appreciable depletion of these. This leads to

\[ B_s = -[e_3 \times E_s] e^{-i\psi_s} - \frac{i}{\omega} [e_3 \times dE_s/dx_3] e^{-i\psi_s} \]  

(8)

Equations (5–6) lead to

\[ B = [e_3 \times E_+] e^{-i\psi_+} - [e_3 \times E_-] e^{-i\psi_-} + [e_3 \times E_p^*] e^{i\psi_p} + \text{c.c.} \]  

(9)

and therefore to

\[ (E^2 - B^2)E = 4 \left( (E_+ E_-) + (E_p^* E_-) E_+ \right) e^{-i(\psi_+ + \psi_- + \psi_p)} + \text{o.t.} \]  

(10)

\[ (E^2 - B^2)B = 4 \left[ (E_+ E_-) (e_3 \times E_p^*) + (E_p^* E_-) (e_3 \times E_+) \right] e^{-i(\psi_+ + \psi_- + \psi_p)} + \text{o.t.} \]  

(11)

\[ (E \cdot B)B = 2 \left[ (e_3 \cdot [E_+ \times E_-]) (e_3 \times E_p^*) + (e_3 \cdot [E_p^* \times E_-]) (e_3 \times E_+) \right] e^{-i(\psi_+ + \psi_- + \psi_p)} + \text{o.t.} \]  

(12)

\[ (E \cdot B)E = 2 \left[ (e_3 \cdot [E_+ \times E_-]) E_p^* + (e_3 \cdot [E_p^* \times E_-]) E_+ \right] e^{-i(\psi_+ + \psi_- + \psi_p)} + \text{o.t.} \]  

(13)

where o.t. stands for other terms.

If we substitute the sums of electric fields given by Eqs. (5) and (7) for \( E \) and the sum of magnetic field given by (8) and (9) for \( B \) in the nonlinear constitutive relations (3) and (4), expressions (10–11) lead to,

\[ D = D_+ e^{-i\psi_+} + D_- e^{-i\psi_-} + D_p e^{-i\psi_p} + D_s e^{-i\psi_s} + \text{c.c.} \]  

(14)
\[ H = H_+ e^{-i\psi_+} + H_- e^{-i\psi_-} + H_p e^{-i\psi_p} + H_s e^{-i\psi_s} + \text{c.c.} \]  
\[ \text{with} \]
\[ D_s = E_s + 2K \left[ 4(E_+ - E_-)E_p^* + 4(E_p^* - E_-)E_+ + 7 \left( e_3 \left[ E_p^* \times E_- \right] \right) (e_3 \times E_+) \right] + 7 \left( e_3 \left[ E_p^* \times E_- \right] \right) (e_3 \times E_+) e^{i\Delta k s}, \]
\[ \mathcal{H}_s = [e_3 \times E_s] - i/\omega [e_3 \times dE_s/dx_3] + 2K [4(E_+ - E_-)e_3 \times E_p^* + 4(E_p^* - E_-)e_3 \times E_+ - 7\left( e_3 \left[ E_+ \times E_- \right] E_p^* \right) - 7\left( e_3 \left[ E_p^* \times E_- \right] E_+ \right)] \]

where \( \Delta k = 2(\omega_0 - \omega_p) \) give the momentum mismatch.

Substitution in \( \dot{D} \) and \( \nabla \times H \) leads to terms in signal frequency,
\[ -i\omega_s E_s - 2i\omega_p K \left[ 4(E_+ - E_-)E_p^* + 4(E_p^* - E_-)E_+ + 7 \left( e_3 \left[ E_+ \times E_- \right] \right) (e_3 \times E_p^*) \right] \]
\[ +7 \left( e_3 \left[ E_p^* \times E_- \right] E_+ \right) (e_3 \times E_+) \]
and
\[ -i\omega_s E_+ + 2dE_+/dx_3 + 2i\omega_p K \left[ 4(E_+ - E_-)E_p^* + 4(E_p^* - E_-)E_+ + 7 \left( e_3 \left[ E_+ \times E_- \right] \right) (e_3 \times E_p^*) \right] \]
\[ +7 \left( e_3 \left[ E_p^* \times E_- \right] E_+ \right) (e_3 \times E_+) \]
respectively.

Maxwell’s Eq. (2) then leads to
\[ \frac{dE_3}{dx_3} = -i(\omega_s + \omega_p) K \left[ 4(E_+ - E_-)E_p^* + 4(E_p^* - E_-)E_+ + 7 \left( e_3 \left[ E_+ \times E_- \right] \right) (e_3 \times E_p^*) \right] \]
\[ +7 \left( e_3 \left[ E_p^* \times E_- \right] E_+ \right) (e_3 \times E_+) e^{i\Delta k s} \]

If the overlap region is of length \( L \) and initially \( E_s = 0 \), at the exit,
\[ E_s(L) = -i\omega_0 KLF, \]
where
\[ F_I = 2 \left[ 4(E_+ - E_-)E_p^* + 4(E_p^* - E_-)E_+ + 7 \left( e_3 \left[ E_+ \times E_- \right] \right) (e_3 \times E_p^*) \right] \]
\[ +7 \left( e_3 \left[ E_p^* \times E_- \right] E_+ \right) (e_3 \times E_+) \]
and
\[ f_{\text{mismatch}} = \frac{e^{i\Delta k L} - 1}{ikL}, \]

For case II of generation of frequency \( \omega_s = \omega_p \) we look for the terms involving product of \( E_-, E_p, E_+ \). We start with writing,
\[ E = E^+_e e^{i\varphi} + E_-e^{-i\varphi} + E_p e^{-i\varphi} + \text{c.c.} \quad (24) \]

\[ B = [e_3 \times E^+_e]e^{i\varphi} - [e_3 \times E_-]e^{-i\varphi} + [e_3 \times E_p] e^{-i\varphi} + \text{c.c.} \quad (25) \]

and proceeding exactly in the same way. We are then lead to

\[ \frac{dE_3}{dx_3} = -2i \omega_0 KL [4(E^+_E E_-)E_P + 4(E_P E_-)E^+_E + [7(e_3, [E^+_E E_-] (e_3 \times E_P))] + 7(e_3, [E_P E_-] (e_3 \times E^+_E))] e^{i\Delta k x_3} \quad (26) \]

and therefore to \( E_3(L) = -i\omega_0 KLF_I \).

Where

\[ F_{II} = -2i \omega_0 KL [4(E^+_E E_-)E_P + 4(E_P E_-)E^+_E + [7(e_3, [E^+_E E_-] (e_3 \times E_P))] + 7(e_3, [E_P E_-] (e_3 \times E^+_E))] e^{i\Delta k x_3} \quad (27) \]

For the special cases of plane polarized light, both cases I and II lead to

\[ E_3(L) = -iP\omega_0 KL e^-e^p e f_{\text{mismatch}} \quad (28) \]

On putting \( E_\pm = E_\pm e_\pm, E_P = E_P e_P \), we get \( F_I = F_{II} = Pe \), where \((P,e) = (16, e_\pm), (-6, e_y), (28, e_\pm) \) and \((-6, e_y) \) for \((e_\pm = e_P = e_y), (e_\pm = e_y, e_y = e_y), (e_\pm = e_y, e_y = e_y, e_P = e_y) \) and \((e_\pm = e_e, e_P = e_y) \) respectively. If we consider pulses of radiation, for any pulse of energy \( E \), intensity \( I \) area \( A \), pulse length \( L \) and duration \( \tau = L \), we have

\[ I = \frac{1}{2\pi} |E|^2, |E| = I \tau A = ILA, \quad (29) \]

which leads to

\[ \frac{I_s}{I_p} = \frac{16\pi^4 P^2 K^2 e_+ e_-}{A^2 \lambda_0^2} |f_{\text{mismatch}}|^2 \quad (30) \]

### 4 Quantum theory

The Lagrangian density given in Eq. (1) leads to the Hamiltonian density in radiation gauge,

\[ \hat{H} = \hat{H}_0 + \hat{H}_{NL}, \hat{H}_0 = \frac{1}{8\pi} (\hat{E}^2 + \hat{B}^2), \quad (31) \]

\[ \hat{H} = \frac{1}{8\pi} \left[ (\hat{E}^2 + \hat{B}^2) + K \left( (\hat{E}^2 - \hat{B}^2)^2 + 2(\hat{E}^2 + \hat{B}^2)(\hat{E}^2 - \hat{B}^2) + 7(\hat{E}^2 - \hat{B}^2)^2 \right) \right] \quad (32) \]

where caps on symbols are used to emphasize that they are operators not c-numbers.

For case I we can write in the radiation gauge.
\[ \hat{A} = \sqrt{\frac{2\pi}{\omega_0 V}} \left[ \hat{e}_+ \hat{a}_+ e^{-i\omega t} + \hat{e}_- \hat{a}_- e^{-i\omega t} \right] + \sqrt{\frac{2\pi}{\omega_p V}} \hat{e}_p^\dagger \hat{a}_p^\dagger e^{i\omega_p t} + \sqrt{\frac{2\pi}{\omega_s V}} \hat{e}_s^\dagger \hat{a}_s^\dagger e^{i\omega_s t} + \text{h.c.} \]  

where \( V \) is the interaction volume and h.c. stands for Hermitian conjugate and \( \hat{a}_\pm \) and \( \hat{a}_{p,s} \) are annihilation operators for the forward and backward moving pump beam, probe beam and signal beam respectively, having phases \( \omega_+ \), \( \omega_- \), \( \omega_p \) and \( \omega_s \) defined in Eqs. (6) and (7).

We can calculate \( \hat{E} = -\hat{A}, \hat{B} = \nabla \times \hat{A} \) and then get

\[ \hat{E}^2 - \hat{B}^2 = \frac{8\pi \omega_0}{V} (\hat{e}_+ \hat{e}_-) \hat{a}_+ \hat{a}_- e^{-i(\omega_+ + \omega_-)} - \frac{8\pi \sqrt{\omega_p \omega_s}}{V} (\hat{e}_p^\dagger \hat{e}_s^\dagger \hat{a}_p^\dagger \hat{a}_s^\dagger e^{i(\omega_p + \omega_s)}) + \text{h.c.} \]  

and

\[ \hat{E}^2 + \hat{B}^2 = \frac{8\pi \omega_0}{V} (\hat{e}_+^\dagger \hat{e}_-^\dagger) \hat{a}_+ \hat{a}_- e^{-i(\omega_+ - \omega_-)} + \frac{8\pi \sqrt{\omega_p \omega_s}}{V} (\hat{e}_p \hat{e}_s) \hat{a}_p \hat{a}_s e^{i(\omega_p - \omega_s)} + \text{h.c.} \]  

In case I and II we look for terms involving \( \hat{a}_+ \hat{a}_- \hat{a}_p^\dagger \hat{a}_s^\dagger \) and \( \hat{a}_+^\dagger \hat{a}_- \hat{a}_p \hat{a}_s^\dagger \) respectively.

For interaction volume \( V = AL \), we get the Hamiltonian

\[ \mathcal{H}_{\text{NL}} = \frac{2\pi \omega_0 \sqrt{\omega_p \omega_s}}{V} K \left[ 4(\hat{e}_+ - \hat{e}_-) (\hat{e}_p - \hat{e}_s^\dagger) + 4(\hat{e}_- - \hat{e}_p^\dagger) (\hat{e}_+ - \hat{e}_s^\dagger) - 7(\hat{e}_3 - [\hat{e}_+ \times \hat{e}_-]) \right] \hat{a}_+ \hat{a}_- \hat{a}_p^\dagger \hat{a}_s^\dagger f_{\text{mismatch}} + \text{h.c.} \]  

Equation of motion of \( \hat{a}_i \) is

\[ i\dot{a}_i = [a_i, \mathcal{H}_{\text{NL}}] = \frac{2\pi \omega_0 \sqrt{\omega_p \omega_s}}{V} K \left[ 4(\hat{e}_+ - \hat{e}_-) (\hat{e}_p - \hat{e}_s^\dagger) + 4(\hat{e}_- - \hat{e}_p^\dagger) (\hat{e}_+ - \hat{e}_s^\dagger) - 7(\hat{e}_3 - [\hat{e}_+ \times \hat{e}_-]) \right] \tau \hat{a}_+ \hat{a}_- \hat{a}_p^\dagger \hat{a}_s^\dagger f_{\text{mismatch}} \]  

The total number of photons \( N \) are given by \( N = \frac{\omega}{\omega_0} = \frac{TA}{\omega_0} \). This identification leads to the signal photons \( N_s \) given in terms of photons \( N_\pm, N_p \) (each \( \gg 1 \)) of the pump and probe beams by

\[ \frac{N_s}{N_p} = \frac{16 \pi^4}{V^2 \lambda_0^2} P^2 K^2 T^2 \omega_p \omega_s N_+ N_- f_{\text{mismatch}} \]  

which is identical with classical results of \( \frac{P}{\langle \gamma \rangle} \).
5 Result and discussion

If four beams move on the same line, the principal task is their separation required for incidence and for detection. Two co-propagating beams having orthogonal polarization can be separated by use of a polarizing beam splitter. If the two copropagating beams of different frequency have the same polarization, separation may be affected by a simple prism using dispersion. If the two beams are counterpropagating and have orthogonal polarizations, a polarizing beam splitter can be used for separation. For two counterpropagating waves having the same polarization, one can use a Faraday-rotation-cell giving rotation by $\frac{\pi}{4}$ and a polarizing beam splitter for which the two directions of polarization have the direction of polarization of the given two beams as the bisector.

To make $\frac{I_s}{I_p}$ large, one may consider the two pump beams having intense light pulses of ultra-short duration. For duration $\sim 10$ fs $f_{\text{mismatch}}$ can be $\sim 1$. If we divide a 10 kJ, 10 fs pulse into two and get the counter propagating pump beams focused on area $(10 \mu^2)$, we have $\frac{I_s}{I_p} \sim 7.74 \times 10^{-16}$, for the maximum value of $P$. For this case if the probe beam has energy 1 J in the interaction volume, a pulse of signal of energy $0.774 \times 10^{-15}$ J will be produced which shall have $2 \times 10^3$ photons. This should be detectable easily.

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Declaration

Conflict of interest The author have no relevant financial interests to disclose. The authors have no conflict of interest to declare that are relevant to content of this article. All authors certify that they have no affiliations with or involvement in any organization or entity with any financial interest or non-financial interest in the subject matter or materials discussed in this manuscript. The authors have no financial or proprietary interests in any material discussed in this article.

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