Surface second-harmonic generation based on periodically poled $LiNbO_3$ nonlinear optical crystal

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We have generated a second-harmonic generation (SHG) of a Q-switched microchip Nd:YAG laser on the surface of a periodically poled $LiNbO_3$ (PPLN) nonlinear crystal near the grazing incidence angle. Three individual SHG waves as transmitted homogeneous, inhomogeneous and reflected radiations have been generated and their intensities are measured and characterized within a desirable range of about 10 different incidence angles of the Nd:YAG laser as pump source on the PPLN surface. The basic of surface nonlinear radiation is also investigated and similar results are calculated and extracted from the theory. Comparison between calculated and measured data shows that they are in good agreement with each other.

Keywords: surface SHG, Nd:YAG laser, PPLN nonlinear crystal

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1. Introduction

Nonlinear optical technique has been introduced as a powerful and adequate tool for the surface study of materials through the interaction of laser beam with the adsorbed particles on the surface.[1] Particularly, second-harmonic generation (SHG) and sum-frequency generation (SFG) on the surface of materials having noncentrosymetric property were employed as an adequate diagnostic instrument by a number of research groups.[2]

Generating sum-frequency radiation on the surface is so complicated compared with the SHG wave due to the necessity of two input waves and also to the difficulty in focusing those two beams on the same small spot.

Historically, in early 1960 surface SHG was first generated in calcite.[3] Thereafter, surfaces of many different materials such as metals, semiconductors, oxides and even liquids were used to generate surface SHG radiation.[4] At the same time the basic of surface nonlinear generation was studied by Bloembergen[5] and a comprehensive theory of surface nonlinear optics was established in Ref. [6]. Soon after the microscopic prospects of surface interaction with the optical beam were considered and published in several papers.[7−9] However, surface SHG has been used to detect the formation of individual monolayers[10] and it was also used as a surface probe of molecular adsorption and orientation.[11,12] Many physical and chemical interactions on the surface of metals could be traced by this method too.[13] The study of the vibrational properties of adsorbed molecules in either frequency or time domain also becomes possible by surface SHG laser beam.[14] Furthermore the molecular orientation as well as material site symmetry can also be determined by SHG surface beam.[6] Recently, the majority of materials from semiconductors to metals such as silicon and many nonlinear crystals have been introduced for the study of different interface mechanisms such as solid–solid, liquid–liquid and liquid–gas interfaces.[15,16] In this context Lithium Niobate ($LiNbO_3$) is one of the most important nonlinear optical crystals capable of generating optical radiation in a spectral region from visible to mid-infrared (MIR).[17] The conversion efficiency of this nonlinear radiation can be enhanced when the crystal is used in its special structure known as periodically poled $LiNbO_3$ (PPLN). Even though the PPLN crystal has been used in many experimental apparatus for nonlinear optical generations, it is not considered for nonlinear surface generation. While the generation of the SHG radiation on the surface of a PPLN requires more complicated apparatus, it provides more intense SHG radiation than the same radiation generated by bulk $LiNbO_3$.[18,19]

In a periodical structure, the individual do-

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main of the nonlinear polarization emits Cerenkov-like wave[20] in a direction which is determined by either the material dispersion or the spatial period of PPLN structure.[21]

In the present paper, the SH radiation of Nd:YAG laser as a pump source from the surface of a PPLN crystal is investigated and the intensities of the reflected and transmitted SHG beams are measured and characterized for different input pump powers and incident angles on the crystal surface. The theoretical model is also studied and computer generated graphs are given for comparison.

2. Theoretical background

A general geometrical diagram of the nonlinear surface wave generation at the oblique incidence is indicated in Fig. 1.

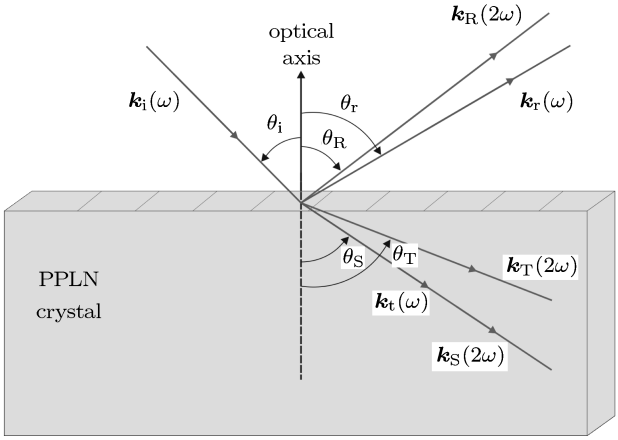


Fig. 1. Geometrical diagram of fundamental and second harmonic wave vectors generated on the surface of a PPLN nonlinear optical crystal.

As investigated by the Bloembergen–Pershan (BP) theory,[22] three different radiations with the second harmonic (SH) frequency are generated on the surface through nonlinear interaction with the material which is subjected to the incident laser beam. Those surface generated waves include an SH beam reflected at angle $\theta_{\rm R}$ and two SH waves transmitted at angles $\theta_{\rm T}$ and $\theta_{\rm S}$, respectively. We expect that when the angle of incidence $\theta_{\rm i}$ is gradually increased, the transmitted SH beam with a wave vector of $k_{\rm S}(2\omega)$, known as inhomogeneous harmonic wave, is suppressed and the homogeneous wave $k_{\rm T}(2\omega)$ grows simultaneously. This behaviour is observed in our experimental investigation too. Those two transmitted SH waves are also spatially resolved and separated within the minimum angle as low as a few degrees. When the incident laser beam arrives at the surface of material nearly at two particular critical angles specified by $\theta_{\rm cr}^{\rm T}(2\omega)$ and $\theta_{\rm cr}^{\rm S}(2\omega)$, the intensities of homogeneous and inhomogeneous transmitted SH waves decrease to zero, respectively.

The basic theory behind the surface SHG has been investigated in Ref. [23]. The electric fields associated with those generated SH waves are

$$\begin{aligned} E_{\rm R}(2\omega) &= 4\pi P^{\rm NLS} F_{\rm R}^{\rm NL}, \\ E_{\rm S}(2\omega) &= 4\pi P^{\rm NLS} F_{\rm S}^{\rm NL}, \\ E_{\rm T}(2\omega) &= 4\pi P^{\rm NLS} F_{\rm T}^{\rm NL}, \end{aligned} \tag{1}$$

where according to BP theory, $F_{\rm R}^{\rm NL}$, $F_{\rm T}^{\rm NL}$ and $F_{\rm S}^{\rm NL}$ are known as Fresnel factors which are given by the following relations:[24]

$$\begin{aligned} F_{\rm R}^{\rm NL} &= \frac{\sin\theta_{\rm S}\sin^2\theta_{\rm T}\sin(\alpha+\theta_{\rm S}+\theta_{\rm T})}{\varepsilon_{\rm R}(2\omega)\sin(\theta_{\rm T}+\theta_{\rm R})\cos(\theta_{\rm T}-\theta_{\rm R})\sin(\theta_{\rm S}+\theta_{\rm T})}, \\ F_{\rm S}^{\rm NL} &= \frac{\sin\alpha}{\varepsilon_{\rm S}-\varepsilon_{\rm T}}, \\ F_{\rm T}^{\rm NL} &= \frac{-\varepsilon_{\rm S}^{1/2}\sin\alpha}{\varepsilon_{\rm T}^{1/2}(\varepsilon_{\rm S}-\varepsilon_{\rm T})} + \frac{\varepsilon_{\rm R}^{1/2}}{\varepsilon_{\rm T}^{1/2}} F_{\rm R}^{\rm NL}, \end{aligned} \tag{2}$$

with

$$\begin{aligned} \varepsilon_{\rm R}^{1/2}(2\omega) &= n_{\rm liq}(2\omega), \\ \varepsilon_{\rm S}^{1/2}(2\omega) &= n_{\rm PPLN}(\omega), \\ \varepsilon_{\rm T}^{1/2}(2\omega) &= n_{\rm PPLN}(2\omega). \end{aligned} \tag{3}$$

The SHG reflected and transmitted angles can be connected together with the well known nonlinear Snell's law as

$$\begin{aligned} n_{\rm air}(\omega)\sin\theta_{\rm i} &= n_{\rm liq}(2\omega)\sin\theta_{\rm R} = n(\omega)\sin\theta_{\rm S} \\ &= n(2\omega)\sin\theta_{\rm T}, \end{aligned} \tag{4}$$

where the angle between the nonlinear polarization $P^{\rm NLS}(2\omega)$ in the plane of incidence and the direction of the source vector is specified by α.

When the incidence angle varies in an appropriate range, the intensity of the SHG waves can also be estimated through the following expressions:

$$\begin{aligned} I_{\rm R}(2\omega) &\propto \left|F_{\rm R}^{\rm NL}\right|^2 \times \cos\theta_{\rm R}(\cos\theta_{\rm i})^{-1}, \\ I_{\rm S}(2\omega) &\propto \left|F_{\rm S}^{\rm NL}\right|^2 \times \cos\theta_{\rm S}(\cos\theta_{\rm i})^{-1}, \\ I_{\rm T}(2\omega) &\propto \left|F_{\rm T}^{\rm NL}\right|^2 \times \cos\theta_{\rm T}(\cos\theta_{\rm i})^{-1}, \end{aligned} \tag{5}$$

where $I_{\rm R}$ is the reflected SHG intensity and $I_{\rm S}$ is the inhomogeneous transmitted SHG intensity.

The schematic diagram of $I_{\rm R}(2\omega)$ and $I_{\rm S}(2\omega)$ intensities given in the above equation is depicted in Fig. 2.

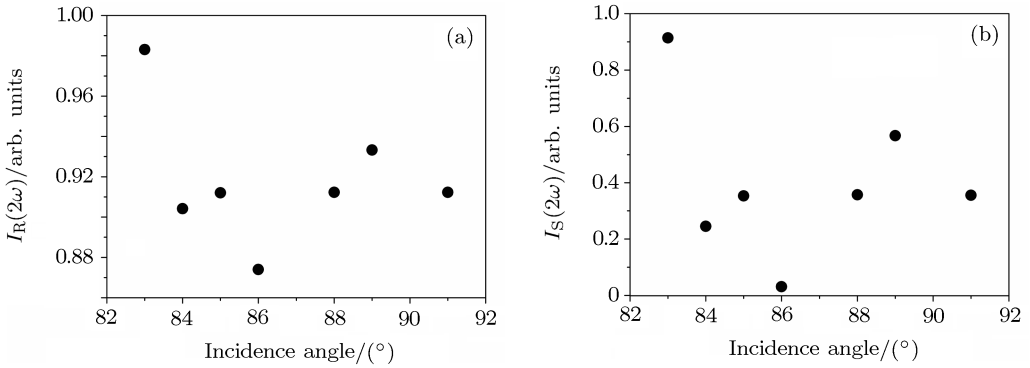


Fig. 2. Calculated intensity of (a) reflected SHG wave and (b) inhomogeneous transmitted SHG wave versus incidence angle of input Nd:YAG laser pump. Calculations are performed at room temperature.

As can be seen, reflected and inhomogeneous transmitted waves can be generated simultaneously with slightly different intensities. The critical angle for inhomogeneous wave occurs at near $\theta_i = 86°$ as shown in Fig. 2(b).

3. Experimental setup

The surface SHG experimental setup is shown in Fig. 3 where a microchip Q-switched Nd:YAG laser was used as a pump source.

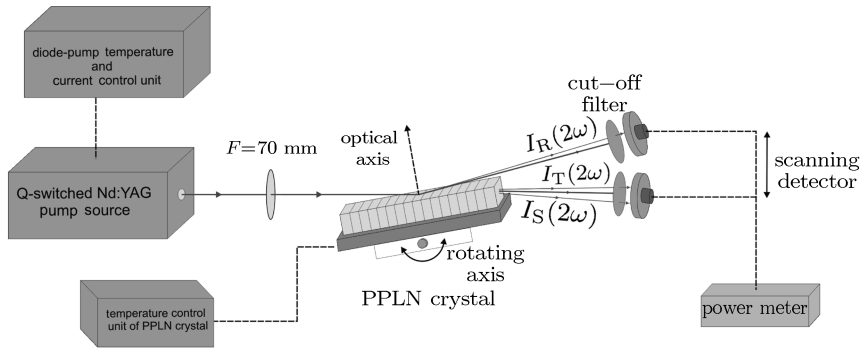


Fig. 3. Experimental arrangement of surface SHG radiation on the surface of a PPLN nonlinear crystal.

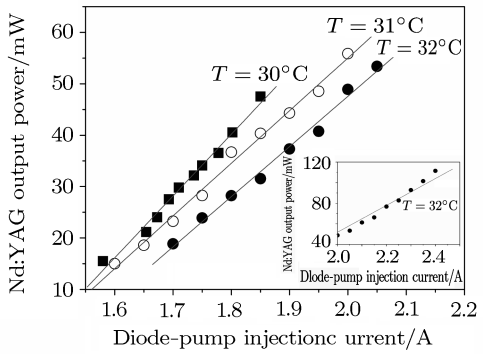


Fig. 4. Variations of Nd:YAG output power with diode-pump injection current for three different temperatures. The inset shows the Nd:YAG output power beyond $i = 2$ A at $T = 32$ °C.

A graded index lens was used to collect the output radiation of a high power diode laser ($P_{diode} = 2$ W) into a 4 mm Nd:YAG crystal embraced by a Cr^{+4}:Nd:YAG element as Q-switched element. This arrangement provided about $P_{max} = 170$ mW average output power. Figure 4 illustrates the variation of the Nd:YAG output power when the temperature and the injection current of its pump diode-laser were changed in a desirable range.

The beam size of the Nd:YAG laser was measured to be $\omega = 1$ mm by using a knife-edge technique and by fitting a Gaussian profile to the measured values. The output beam of the pump source was then imaged on the surface of a PPLN nonlinear optical crystal by a plano-convex lens with a focal length of $f = 70$ mm. Therefore the Nd:YAG laser beam size is reduced to smaller size $\omega = 0.31$ mm which was measured by the same procedure. The crystal (OPO3-40, Thorlabs Co.) was placed on a scaled rotating plate that

provided a very smooth tilt for the crystal in the vertical plane around rotating axis as depicted in the figure. The variation of the incidence angle, $\theta_{\rm i}$, of the Nd:YAG laser on the PPLN surface was then provided by this rotation angle. Behind the crystal a cut-off filter was used to block the fundamental pump beam at $\lambda = 1064$ nm. When the crystal was rotated with respect to the incident pump beam, the intensities of two transmitted and reflected SHG radiations were directly recorded by a PM100 power metre provided by Thorlabs Co. as a detector with an average responsivity of 0.35 A/W and a response time of less than 1 μs. The detector was simultaneously moved vertically to record the radiated SHG intensities behind the crystal. The beam quality of the SHG generated wave is also measured to be $M^2 \sim 1.15$ which is not too far from the value obtained from a Gaussian profile. The M^2 measurement was performed for reflected SHG, $I_{\rm R}(2\omega)$, by measuring both the number of separate planes around the beam waist of the laser and the cone half-angle of the diverging SHG-laser beam.[25]

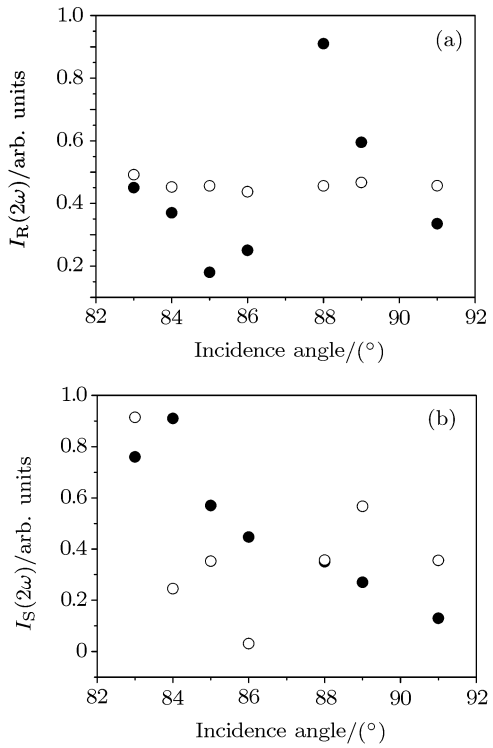


Fig. 5. Comparisons between experimental (•) and calculated (○) data for intensities of (a) $I_{\rm R}(2\omega)$ and (b) $I_{\rm S}(2\omega)$ waves for different incidence angles of input Nd:YAG laser pump.

Figures 5(a) and 5(b) show the recorded intensities of inhomogeneous transmitted and reflected SHG waves, $I_{\rm S}(2\omega)$ and $I_{\rm R}(2\omega)$, near their relevant critical angles, respectively. As specified in the figures, both $I_{\rm S}(2\omega)$- and $I_{\rm R}(2\omega)$-waves are generated on the surface of the PPLN nearly at the same angles, which is expected by the BP theory shown in Fig. 2. However, the intensities of the measured $I_{\rm S}(2\omega)$ and $I_{\rm R}(2\omega)$ radiations are slightly sensitive to the rotating angle of the crystal.

The intensity of the homogeneous transmitted SHG wave, $I_{\rm T}(2\omega)$, was also measured and the results are indicated in Fig. 6. The same behaviours as those of $I_{\rm R}(2\omega)$ and $I_{\rm S}(2\omega)$ are observed in this measurement.

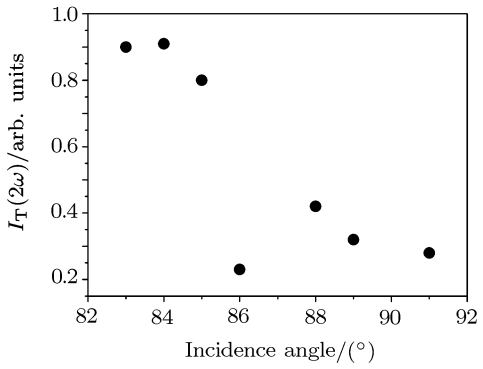


Fig. 6. Measured intensities of the homogeneous transmitted SHG wave for different incidence angles of input Nd:YAG laser pump.

4. Characterization of surface SH-generated

As indicated in Fig. 4, the output power of the Nd:YAG laser is dependent on the temperature and on the current of its high power diode pump. Consequently, the intensity of the surface SHG radiation can be modulated by the output power of the Nd:YAG pump source. Figures 7(a) and 7(b) show the powers of the transmitted and the reflected SHG waves are linearly increased respectively when the Nd:YAG output power is modulated by varying its diode-pump injection current.

Those SHG powers can also be tuned by changing the temperature of the diode pump within a specific range. Figure 8 shows that the $I_{\rm R}(2\omega)$ is linearly proportional to the pump-diode temperature.

However, it should be noted that the dependences of $I_{\rm T}(2\omega)$, $I_{\rm S}(2\omega)$ and $I_{\rm R}(2\omega)$ intensities on PPLN

temperature have not been fully investigated.

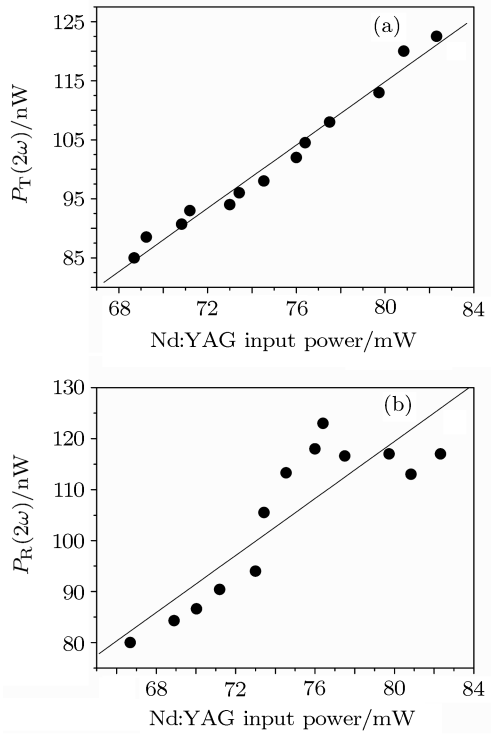


Fig. 7. Power variations of (a) homogeneous transmitted SHG power $P_T(2\omega)$ and (b) reflected SHG power $P_R(2\omega)$ with Nd:YAG input power. The temperature of the Nd:YAG diode pump is fixed at $T = 32.9°$C and the SHG powers are measured at an incidence angle of $\theta_i = 90°$.

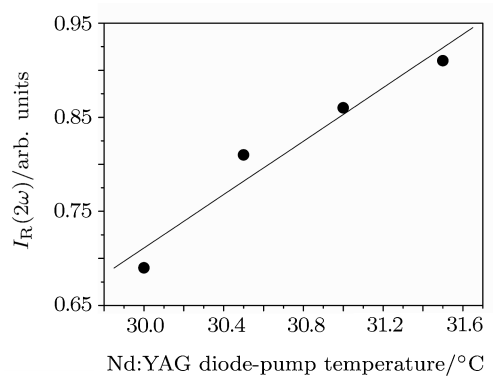


Fig. 8. Intensity variation of reflected SHG radiation $I_R(2\omega)$ with Nd:YAG diode-pump temperature, with the diode-pump injection current kept at $i = 2.13$ A.

Even though the nonlinear frequency conversion processes suffer the low efficiency, however, by carefully choosing a nonlinear crystal and precisely adjusting the experimental arrangement, SHG efficiency can be enhanced to a certain level. Figure 9 shows the measured conversion efficiency, η, of those surface SHG radiations when the PPLN crystal is turned with respect to the incidence of Nd:YAG pump radiation as clarified in the experimental set up.

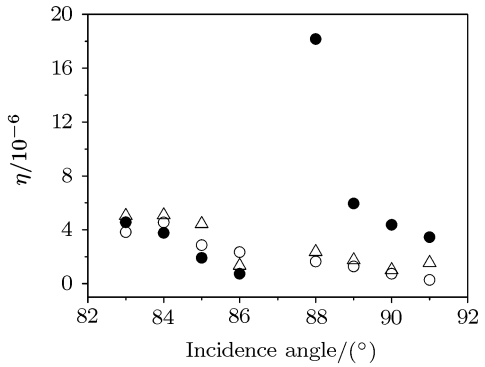


Fig. 9. Measured conversion efficiency of the generated surface SHG beams (•) η_s ($= P_S/P_P$), (△) η_T ($= P_T/P_P$) and (○) η_R ($= P_R/P_P$) when the PPLN crystal is rotated with respect to the incident Nd:YAG laser beam.

Finally it is found that the total transmitted SHG radiation, $I_{total}(2\omega)$, shown in Fig. 10, also varies with PPLN rotating angle.

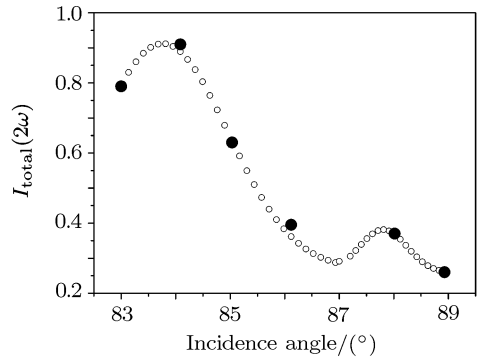


Fig. 10. Total transmitted SHG intensity (•) as a function of incidence angle of Nd:YAG pump beam, with an experimental phase-matching curve (○) fitted to the measured values.

As shown in the figure, the maximum intensity of $I_{total}(2\omega)$ occurs at $\theta_i^m = 84.36°$ which is nearly equal to the perfect phase-matching angle of the bulk $LiNbO_3$.

5. Conclusion

In this work, a surface SH radiation is generated based on a PPLN nonlinear crystal. Three individual SHG waves referred to as inhomogeneous transmitted, inhomogeneous transmitted and reflected SHG waves are generated and their corresponding intensities $I_T(2\omega)$, $I_S(2\omega)$ and $I_R(2\omega)$ are also measured

within a desirable range of rotating angle of the crystal. The results are confirmed by BP theory and the measurements and the calculations are found to be in good agreement with each other. However, the intensities of those generated SHG waves are slightly different from the calculated ones and they are not sensitive to the PPLN temperature.

From those generated SH waves it follows that the intensity of the transmitted inhomogeneous wave, $I_S(2\omega)$, is linearly proportional to the rotating angle of the PPLN crystal. The measured conversion efficiency of generated SHG waves is in a level of nWatts which is quite within the acceptable range that is reported in many scientific documents.

It is worth noting that the intensity of the PPLN surface-generated SH waves is strongly dependent on the input parameters of Nd:YAG pump source such as its injected diode-pump current and temperature.

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