Isotope effect on the Casimir force

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Isotopic dependence of the Casimir force is key to probing new physics and pushing novel technologies at the micro and nanoscale, but is largely unexplored. In 2002, an isotope effect of $10^{-4}$ was estimated for metals—orders of magnitude beyond the experimental resolution. Here, by employing the Lifshitz theory, we reveal a significant isotope effect of over $10^{-1}$ for polar dielectrics. This effect arises from the isotope-mass-induced line shift of the zone-center optical phonons and is insensitive to the linewidth. We perform numerical analyses on both the imaginary and real-frequency axes, and derive analytical formulas for predicting the isotope effect. The three-orders-of-magnitude difference between polar dielectrics and metals arises from the distinct isotopic dependence of the phonon and plasma frequencies. Our work opens up a new avenue for engineering forces at small scales and may also facilitate the quest for the fifth force of nature.

isotope effect, Casimir force, polar dielectric, phonon line shift, plasma frequency

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1 Introduction

Hendrik Casimir [1] predicted in 1948 that there exists an attraction between two perfectly conducting charge-neutral plates separated by a vacuum gap $d$. This so-called Casimir force per unit area is given by

$$F_c = \frac{\pi^2 \hbar c}{240 d^4} = -1.3 \times 10^{-27} d^{-4} \text{ N m}^2,$$

where $\hbar$ is the reduced Planck constant and $c$ is the speed of light. Similar to the van der Waals (vdW) force, the Casimir force also arises from quantum fluctuations of the electromagnetic (EM) field [2-9]. According to Casimir, the two plates form a cavity with high-reflectivity boundaries that limit the number of modes therein, leading to a net zero-point pressure that pushes inward [1]. The Casimir force finds broad applications in colloid and surface sciences, microsystems, and nanotechnologies due to the $d^{-4}$ dependence [10-13], and is of great interest to several branches of physics including elementary particles, condensed matter, and cosmology [14-16].

Over the decades, much progress has been made in the measurement [17-26], computation [27-33], and understanding [34-38] of the Casimir force. In particular, a general theory for the Casimir interaction between objects of arbitrary materials, geometries, temperatures, and separating media was proposed by Lifshitz [27] in 1956 based on the framework of fluctuational electrodynamics. With the complex permittivities of the interacting bodies as the key input, this theory has since been widely adopted and laid the foundation for engineering both the strength and sign of the...
Casimir force. To this end, many schemes have been devised, such as using micro/nanostructures [39-42], metamaterials [43-45], phase transitions [46,47], topological insulators [47,48], epsilon-near-zero materials [49], chiral objects [50], rotating particles [51], tailored separating media [52-54], and external fields [54-56].

Among all the mechanisms for tuning the Casimir force, its isotopic dependence is rarely explored but of great interest, for example, in seeking the fifth force of nature [57-59]. Previous effort only estimated a negligible isotope effect of $10^{-4}$ for metals, which was two orders of magnitude below what was experimentally resolvable [57]. In contrast, large isotope effects on the vDW force have been extensively reported in diverse phenomena including vapor pressure, adhesion, and friction [60-63]. More importantly, an isotope effect of over $10^{-3}$ on heat radiation from thermally fluctuating EM fields was recently revealed [64], which was attributed to the permittivity variation of polar dielectrics through isotope-mass-induced phonon line shift and broadening [65,66]. Together, these results motivate us to revisit the question as to how the Casimir force might change with the isotopic compositions of the interacting bodies.

In this work, we focus on the Casimir interactions between polar dielectrics (Figure 1(a)) and highlight an isotope effect of over $10^{-1}$ for bulk plates, which is 3-orders-of-magnitude larger than that for metals. Reducing plate thickness can enhance the isotope effect by several folds, especially for relatively large gaps. From the imaginary-frequency perspective, we attribute this significant isotopic dependence of the Casimir force to the atomic-mass-induced line shift of the zone-center phonons, and derive analytical formulas for predicting the isotope effect. Further, we provide additional insight on the real-frequency axis in terms of the surface phonon polaritons (SPhPs) modes. In contrast to thermal radiation [64], the phonon linewidth has a negligible influence. We conclude by showing that the difference between polar dielectrics and metals originates from the distinct isotopic-mass dependence of the phonon and plasma frequencies.

2 Methods and materials

According to the Lifshitz theory [27], the Casimir force between two parallel plates at 0 K in vacuum can be expressed as:

$$F(d) = \frac{\hbar}{2\pi}\int_0^\infty d\omega \int_0^\infty k \, dk \, |\varphi(\omega, k)|^2 \sum_{s,p} Z_{s,p}(\omega, k).$$  \hspace{1cm} (2)

In practice, eq. (2) holds well at finite temperatures as long as the separation $d$ is much smaller than the thermal wavelength

$$\lambda_{th} = \frac{\hbar}{k_0T}$$

which is about 7.6 μm at 300 K and 570 μm at liquid helium temperature [57]. Here, $F_\omega$ is the force spectrum, $k$ is the wavevector component parallel to the plates, $\gamma_0 = \sqrt{k_0^2 - k^2}$ is the perpendicular wavevector in vacuum with $k_0 = \omega / c$, and $Z$ is the exchange function given by

$$Z_{s,p}(\omega, k) = \left\{ \begin{array}{ll}
\text{Re} \left\{ \frac{r_s^A r_p^B e^{2\gamma_0d}}{1 - r_s^A r_p^B e^{2\gamma_0d}} \right\}, & k < k_0^p \\
\text{Im} \left\{ \frac{r_s^A r_p^B e^{2\gamma_0d}}{1 - r_s^A r_p^B e^{2\gamma_0d}} \right\}, & k > k_0^p
\end{array} \right.$$  \hspace{1cm} (3)

where A and B denote the two plates, and $i$ refers to the s- and p-polarized modes. Both propagating and evanescent waves are included. For bulk plates, $r_s^A(\omega, k)$ and $r_p^A(\omega, k)$ are the Fresnel reflection coefficients at the plate-vacuum interfaces which are functions of the permittivity $\varepsilon(\omega)$ of the
plates. Formulas for plates of finite thickness are provided in the Supporting Information (SI).

In light of the highly oscillating nature of the integrand in the real-frequency domain [27], \( F(d) \) in eq. (2) is usually expressed as a contour integration on the complex frequency plane along the imaginary-frequency axis \( \omega = i\xi \):

\[
F(d) = \frac{\hbar}{2\pi^2} \int_0^\infty \int_0^\infty \frac{d\xi}{\sqrt{\xi}} dk \langle \gamma(i\xi,k) \rangle \sum_{i,s,p} \gamma_{i,s,p}(i\xi,k), \tag{4}
\]

where the permittivity \( \varepsilon(i\xi) \) can be obtained via the Kramers-Kronig relation [33] as:

\[
\varepsilon(i\xi) = 1 + \frac{2}{\pi} \int_0^\infty \frac{\omega\text{Im}[\varepsilon(\omega)]}{\omega^2 + \xi^2} d\omega. \tag{5}
\]

For the theoretical study of the Casimir force between polar dielectrics such as silicon carbide [67,68], magnesium oxide [69], and aluminum oxide [70], the permittivity \( \varepsilon(\omega) \) is usually given by the Lorentz oscillator model as:

\[
\varepsilon(\omega) = \varepsilon_\infty \left[ 1 - \sum_{j} \left( \frac{\omega_{LO,j}^2}{\omega^2 - \omega_{TO,j}^2 + i\Gamma_j \omega} \right) \right], \tag{6}
\]

here, \( j \) is the oscillator index; \( \varepsilon_\infty \) is the high-frequency permittivity; \( \omega_{LO,j} \) and \( \omega_{TO,j} \) are respectively the frequencies of the transverse and longitudinal zone-center optical phonons, which shift with the reduced mass \( \mu \) of an isotope-engineered polar dielectric approximately as \( \omega \propto \mu^{-0.5} \), and \( \Gamma \) is the damping factor which increases with the isotopic mass fluctuations in ideal crystals but may vary considerably due to phonon-scattering by imperfections as the crystal quality varies [64]. In addition to phonons, charge carriers can also contribute to the permittivity of polar dielectrics upon doping or at sufficiently high energies [71-75], which are usually less sensitive to isotopes. Therefore, for simplicity and without loss of generality, we focus on materials with a single-oscillator Lorentz model to highlight the essential characteristics of the phonon-mediated isotope effect on the Casimir force.

3 Results and discussion

3.1 Calculation of the Casimir force for cBN

To begin with, we study isotope-engineered cubic boron nitride (cBN) which is a prototypical polar dielectric and has already been used to study the isotope effects on heat conduction [76] and thermal radiation [64]. Usually, natural nitrogen is treated as isotopically pure (99.6% \(^{14}\text{N} \)) while the ratio between the two boron isotopes \(^{10}\text{B} \) and \(^{11}\text{B} \) is varied. Here, we consider cBN with four representative isotope ratios: \(^{11}\text{BN} \) with pure \(^{11}\text{B} \), \(^{11}\text{BN} \) with 20% \(^{10}\text{B} \), \(^{11}\text{BN} \) with 50% \(^{10}\text{B} \), and \(^{11}\text{BN} \) with pure \(^{10}\text{B} \). Their phonon properties can be found in our previous work [64]. Without loss of generality, we first focus on the isotope-induced phonon line shift and assume a fixed damping factor of 0.5 cm\(^{-1} \) which is representative of perfect cBN crystals.

In Figure 1(b), we plot the calculated Casimir force between two c\(^{11}\text{BN} \) plates of representative thicknesses (bulk, 1 \( \mu \)m, 100 nm, and 10 nm) as a function of gap size. The same thickness is always assumed for both plates. We are concerned chiefly with the force magnitude so the negative sign is omitted unless otherwise noted. For bulk plates, \( F \) increases from about 10 nN m\(^{-2} \) at a 10 \( \mu \)m gap to 80 N m\(^{-2} \) at a 10 nm gap. The scaling is roughly \( d^{-4} \) at large gaps, since retardation is expected at distances comparable to the characteristic wavelengths of the plates [77]. At nanometer gaps, however, \( F \) scales as \( d^{-1} \). As the plates become thinner, the force decreases at large gaps but remains close to the bulk values for \( d \ll t \). This is because the Casimir force between polar dielectrics at small gaps is expected to be dominated by the cavity SPhP modes [71,78], whose penetration depths decrease with and remain comparable to the gap size [79,80].

3.2 Isotope effect on the Casimir force

We further perform calculations for other isotopic configurations and notice that the Casimir force is always the weakest (denoted as \( F_{\text{min}} \)) for the pair of c\(^{11}\text{BN} \) which features the largest reduced mass and lowest phonon frequencies of any isotope-engineered cBN (see Figure S1 in SI). With bulk c\(^{11}\text{BN} \)-c\(^{11}\text{BN} \) as the reference, we plot in Figure 1(c) variations of the Casimir force \( \Delta F = F - F_{\text{min}} \) for the symmetric pairs of c\(^{14}\text{BN} \), c\(^{15}\text{BN} \), c\(^{11}\text{BN} \), and c\(^{10}\text{BN} \), and observe a monotonically increasing force as the fraction of \(^{10}\text{B} \) increases, regardless of the gap size. We also show the asymmetric case of c\(^{11}\text{BN} \)-c\(^{10}\text{BN} \) and find that the result overlaps well with that for c\(^{10}\text{BN} \)-c\(^{10}\text{BN} \).

Based on the results above, we define the magnitude of the isotope effect for two plates of arbitrary isotopic compositions with respect to c\(^{11}\text{BN} \)-c\(^{11}\text{BN} \) as \( \alpha = \Delta F / F_{\text{min}} \times 100\% \). We focus on the isotope effect of c\(^{10}\text{BN} \)-c\(^{10}\text{BN} \) since it always yields the largest force. As shown in Figure 1(d), \( \alpha \) increases with decreasing gap size and saturates at a maximum value of about 2.8% for sufficiently small gaps regardless of the plate thickness, which is two-orders-of-magnitude larger than what was previously estimated for metals [57]. At larger separations, however, the isotope effect can be enhanced by several folds with thinner plates (Note 2 in SI). The largest enhancement appears at gap sizes around 4 \( \mu \)m.

3.3 Imaginary-frequency analysis of the isotope effect

To understand the isotope effect on the Casimir force, we
perform spectral analysis on the imaginary-frequency axis in terms of the force spectra $F_\xi$, the cavity reflectivity $R^{AB} = r^Ar^B$, and the permittivity $\varepsilon(i\xi)$. A small gap size of 10 nm is assumed for its larger isotope effect. As shown in Figure 2(a), (b) and (c) respectively, all the $F_\xi$, $R^{AB}$, and $\varepsilon(i\xi)$ curves are very similar, with a flat region at low frequencies and a sharp drop at high frequencies where the cBN plates are effectively transparent [1,81]. In addition, the isotope-induced variations $\Delta F_\xi$, $\Delta R^{AB}$, and $\Delta \varepsilon(i\xi)$ with respect to $^{11}$BN-$^{11}$BN are also shown in Figure 2 on the right axes. Notably, predominant peaks appear at around $2 \times 10^{14}$ rad/s, very close to $\omega_{\text{TO}}$ of $^{11}$BN, indicating a crucial role of the zone-center phonons. These observations allow for a qualitative understanding of the gap-dependence of the isotope effect. First, we note that the Casimir force at gap size $d$ arises mainly due to waves of frequencies below $\xi_d = 2\pi c / a$. For large gaps with $\xi_d \ll \omega_{\text{TO}}$, the isotope barely affect the force. As $d$ reduces, $\xi_d$ increases towards and passes $\omega_{\text{TO}}$, leading to a monotonically increasing enhancement is expected.

3.4 Analytical prediction of the isotope effect

Mathematically, we can further establish analytical formulas for predicting the isotope effect at the small-damping limit. Details of the derivation can be found in Note 3 (SI). Briefly, with $\Gamma \to 0$, $\text{Im}[\varepsilon(\omega)] / \omega$ approaches the Dirac $\delta$-function centered at $\sqrt{1+\beta} \omega_{\text{TO}}$, where $\beta$ denotes the isotope-induced relative phonon line shift [64] given by $\beta = \Delta \omega_{\text{TO}} / \omega_{\text{TO}} \approx \Delta m_{\text{iso}} / \mu_{\text{iso}}$. Compared with $^{11}$B, the line shift $\beta$ is 2.8% and 1.4% for $^{10}$B and $^{8}$B, respectively. With the Dirac $\delta$-approximation, the isotope effect on $\varepsilon(i\xi)$ can be written as:

$$\varepsilon_p(i\xi) = \varepsilon\left(1 + \frac{\xi}{1+\beta}\right).$$

(7)

That is to say, a line shift $\beta$ in the real-frequency domain leads to a stretch of the imaginary-frequency permittivity by a factor of $1+\beta$.

In order to derive the formula for predicting the isotopic dependence of the Casimir force, we begin with the isotope effect on the reflection coefficients. As an example, we consider the $s$-polarized waves with

$$r(i\xi,k) = |r_p(i\xi,k)|^2 = |r_p(i\xi,k)|^2,$$

(8)

where $\gamma$ is the perpendicular wavevector with $|\gamma| = \sqrt{\varepsilon(i\xi) \xi^2 / c^2 + k^2}$. Based on eq. (7), the isotope-modified $|\gamma|$ can be expressed as:

$$|\gamma_p(i\xi,k)|^2 = (1+\beta) |\gamma(i\xi,k)|^2.$$ 

(9)

Therefore, we obtain the isotope effect on $r(i\xi,k)$ for the $s$-polarization as:

$$r_p(i\xi,k) = r(i\xi,k) \sqrt{\frac{i\xi}{1+\beta}} \frac{k}{1+\beta}.$$ 

(10)

Likewise, eq. (10) also holds for $p$-polarized waves.

Next, we proceed to the exchange function. For the symmetric case with the two bulk plates having the same isotopic composition, the isotope-modified exchange function can be expressed in a similar form as:

$$Z_p(i\xi,k,d) = Z_p(i\xi,k,d) = \left|\frac{i\xi}{1+\beta} \frac{k}{1+\beta} \cdot (1+\beta)d\right|^2.$$ 

(11)

By further defining $\xi_p = \xi/(1+\beta)$, $k_p = k/(1+\beta)$, and $d_p = (1+\beta)d$, we can then obtain an analytical formula for the isotope-engineered Casimir force between two identical plates as:

**Figure 2** (Color online) Understanding the isotope effect on the imaginary-frequency axis. (a) Force spectra at a 10 nm-gap for pairs of $c^{11}$BN, $c^{10}$BN, $c^{8}$BN, and $c^{11}$BN-$c^{11}$BN. (b) Corresponding reflectivity spectra at normal incidence. (c) Permittivities at imaginary frequency. Right axes show variations with respect to (pairs of) $c^{11}$BN with the peak position marked.
\[ F_{\beta}(d) = \frac{\hbar (1 + \beta)^4}{2\pi^2} \int_0^\infty d\xi \int_0^\infty k_d dk_d \times \left| \gamma(\xi^2, k_d) \right| \sum_{i=\alpha}^p Z_i(\xi^2, k_d, d_d)
\]
\[ = (1 + \beta)^4 F[(1 + \beta)d], \quad (12) \]

which agrees well with our exact calculations as shown in Figures 1(d) and S3 (SI).

Recall that \( F(d) \propto d^{-4} \) at small gaps, we then have \( F_d(d) \approx (1 + \beta)F(d) \) so the isotope effect is simply \( \alpha \approx \beta \), as already demonstrated for \( ^{10}\text{BN}-^{11}\text{BN} \). As the gap size increases, the scaling law gradually approaches \( F(d) \propto d^{-4} \) so \( \alpha \) decreases and eventually goes to 0 with \( F_d(d) = F(d) \). Eq. (8) also holds in the general case of pairing plates A and B, although the stretch factor is then \( \gamma(1 + \beta^A)(1 + \beta^B) \), which explains the similarity between \( ^{11}\text{BN}-^{10}\text{BN} \) and \( ^{18}\text{BN}-^{10}\text{BN} \).

### 3.5 Real-frequency analysis of the isotope effect

Despite the success on the imaginary-frequency axis, we have not been able to relate the isotope effect to the SPhPs of cBN, which are expected to dominate the Casimir force at small gaps [71,78]. To this end, we perform modal analysis in the real-frequency domain which is challenging in general but proves feasible in our case. The force spectra \( F_\omega \) in eq. (2) for various bulk pairs at \( d = 10 \) nm are plotted in Figure 3(a), which feature peaks and dips around the frequencies where \( \text{Re}[\varepsilon(\omega)] = -1 \), thus confirming the dominant role of the SPhPs in the near field. Here, the dips represent attractive forces (negative) due to the symmetric cavity SPhP modes as the SPhPs on the two vacuum interfaces couple, while the peaks indicate repulsion arising from the antisymmetric cavity modes [71]. By integrating \( F_\omega \) across the Reststrahlen band (Figure 3(b)), we obtain net attractive forces in good agreement with those calculated using eq. (4) for \( d < 100 \) nm (Figure S4 in SI). In addition, we notice that the width of the highly reflective Reststrahlen band scales as \( (1 + \beta)(\omega_{10} - \omega_{T0}) \), which may offer a more intuitive picture for the magnitude of the isotope effect at small gaps.

With the fundamental role of the phonon line shift revealed, we proceed to a brief discussion of the line broadening. For perfect cBN crystals, the first-principles calculated damping factor \( \Gamma \) increases with atomic mass fluctuations from \(-0.5\text{ cm}^{-1} \) for \(^{11}\text{BN} \) and \(^{10}\text{BN} \) to \(-1\text{ cm}^{-1} \) for \(^{18}\text{BN} \). However, \( \Gamma \) may be much larger in real samples due to various imperfections [64]. In Figure 4(a), we plot the Casimir force for pairs of bulk \(^{11}\text{BN} \), \(^{18}\text{BN} \), and \(^{10}\text{BN} \) at a 10 nm-gap versus \( \Gamma \) from 0.5 to 100 cm\(^{-1}\). All three force curves nearly change at \( \Gamma < 10 \text{ cm}^{-1} \) (\(-0.01\omega_{T0}\) and drop slightly for larger \( \Gamma \) (\(-4\% \) at 100 cm\(^{-1}\)). The corresponding isotope effect for \(^{10}\text{BN}-^{10}\text{BN} \) is also shown on the right axis, which remains almost constant. To explore the underlying mechanism, we plot in Figure 4(b) the exchange functions for \(^{11}\text{BN}-^{11}\text{BN} \) and \(^{10}\text{BN}-^{10}\text{BN} \) at three representative \( \Gamma \) (0.5, 1, and 2 cm\(^{-1}\)) with \( k = 10^6 \text{ m}^{-1} \) (\(d^{-2}\)). Similar to \( F_\omega \) in Figure 3(a), \( Z_f(\omega, k) \) features attractive dips and repulsive peaks, the widths of which broaden with \( \Gamma \) while the heights reduce as \( \Gamma^{-1} \). Therefore, the mode-resolved force remains the same upon frequency integration, which explains the negligible impact of \( \Gamma \) on the Casimir force and its isotope effect. On the imaginary frequency axis, this insensitivity can also be anticipated by noting that eq. (7) holds as long as \( \Gamma \) is sufficiently small.

### 3.6 Polar dielectrics versus metals

Apart from cBN, we also consider isotope-engineered lithium hydride (LiX with Li = \( ^6\text{Li} \), \( ^7\text{Li} \) and X = H, D, T) which offers some of the largest phonon line shifts among polar dielectrics [64,82]. The isotope effects calculated via the Lifshitz theory for 5 symmetric LiX pairs and 15 asymmetric pairs at a 10-nm gap are shown in Figure 5(a), with \(^7\text{LiT}^7\text{LiT}\)
as the reference and the average $\Delta \mu / \mu$ of the two plates on the $x$-axis. Indeed, the isotope effect for LiX is significant and reaches up to 55.6%. Further, we assume a hypothetical polar dielectric based on the permittivity model of LiX, and plot the exactly calculated isotope effect together with the prediction of eq. (8) for small gaps ($\alpha \approx \beta \approx \Delta \mu / \mu$). All the data agree well with each other except for some minor deviations in a few asymmetric cases. In addition, we perform similar calculations for several real metals (Li, nickel, and copper) and a hypothetical metal based on the Drude model of Li [64]. Negligibly small isotope effects on the order of $10^{-4}$ are obtained, in agreement with the estimation by Krause and Fischbach [57]. This is because for metals the plasma frequency shifts with the isotopic mass variation but with a reduction factor of about $10^{-3}$, that is, $\beta_{\mu} \approx \Delta m / m \times 10^{-3}$ [64].

With the basic characteristics of the isotope effect on the Casimir force systematically understood for polar dielectrics and metals based on the classic Lorentz and Drude model, respectively, we now briefly revisit polar dielectrics considering the electronic contribution to the permittivity. As mentioned earlier, for doped samples, a Drude term is often added to account for the free carriers [74,75]. Moreover, at energies above the bandgap, electronic excitations can further contribute to the force [71]. However, a rigorous treatment of these electronic factors remains inaccessible due to the unavailability of their exact isotopic dependence. Qualitatively, with a relatively small sensitivity to the isotopic composition, they tend to make polar dielectrics behave more similar to metals by increasing the Casimir force and reducing the isotope effect, especially at small gaps where high-frequency electronic contributions may be large. This is expected in light of the physical mechanisms already revealed.

4 Conclusions

In summary, we have systematically explored the isotope effect on the Casimir force between two plates in vacuum using the Lifshitz theory. In particular, we reveal a significant isotope effect of over $10^{-3}$ for polar dielectrics, which is three-orders-of-magnitude larger than the isotope effect for metals. This is understood via the reflectivity and permittivity variations along both the real and imaginary frequency axes, which originate from isotope-mass-induced zone-center phonon line shifts. Both the Casimir force and its isotope effect are insensitive to the phonon linewidth, in striking contrast to the case of thermal radiation [64]. Reducing the plate thickness enhances the isotope effect at large gaps but only by a few folds. The essential difference between polar dielectrics and metals lies in the distinct isotopic dependence of the phonon and plasma frequencies. We derive formulas that allow the isotope effect on the Casimir force to be accurately yet conveniently predicted from the relative frequency shift. To experimentally observe the large isotope effect that we theoretically predict, special attention should be paid to the sample quality, temperature, and gap size. Our work opens up new possibilities in engineering forces at small length scales and may also help probe new physics, in particular, the fifth force of nature.

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Conflict of interest The authors declare that they have no conflict of interest.

Supporting Information

The supporting information is available online at http://phys.scichina.com and https://link.springer.com. The supporting materials are published as submitted, without typesetting or editing. The responsibility for scientific accuracy and content remains entirely with the author.

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