Boundary condition for the optical force density

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The optical force density as a function of position and time provides fundamental information to model local and, through integration, macroscopic kinetic motion of condensed matter. Here, the boundary condition associated with the optical force density is developed and investigated using an expression stemming from the work of Einstein and Laub, and in conjunction with Maxwell's equations to describe the electromagnetic fields. Consequently, a constraint is formed that allows a unique relationship between the total force and the force density, one that is achieved by virtue of the conservation principles for physical materials and described by locally homogenized constitutive parameters. Further insight can be garnered from new experimental studies, as summarized. The mathematical steps presented form a basis for modeling various optomechanical phenomena, including optical forces in and on solid-state systems such as membranes, beams, cantilevers, and waveguides, and can be interpreted in terms of a suite of related theoretical work. This specification of the force density boundary condition is relevant for basic scientific fields including those involved with various quantum cooling issues, molecular optomechanics, photochemistry, and biophysics (including mechanotransduction). The technologies impacted encompass integrated optomechanics (silicon photonics, where new optical device concepts can be enabled), communication systems (in which optical forces could supplant electronic switching), remote control and actuation, propulsion, sensing, and navigation.

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I. INTRODUCTION TO OPTICAL FORCES

While there have been many important papers written on the theory of electromagnetic forces, we still lack complete understanding of how photons interact mechanically with materials. This has consequences for the interpretation of experimental data and for optomechanics in condensed matter, as well as for a range of applications. In the work presented here, a force density theory that has been used to explain key experiments and is based on the work of Einstein and Laub [1] is used to develop the boundary conditions that are appropriate at material interfaces and for optical frequencies. This leads to a theory for the total force and pressure that can be further evaluated through various experiments, and the concepts can be applied to other models.

Since a prediction of an optical force by Maxwell [2] and subsequent measurement [3,4], many basic developments have occurred. Notable among these, after the invention of optical tweezers in the 1980s [5], optical manipulation has become important in biology, physical chemistry, and condensed-matter physics [6]. Optical rheology and mechanotransduction in cells has enabled new experimental regimes [7], and the range of forces realizable [6,8–13] are useful for biological and macromolecular systems research [14–16]. In addition, optomechanics has led to surprising findings in classical statistical mechanics [17], including anomalous attraction [18], oscillatory colloidal interactions [19–22], and hydrodynamic fluctuations [23]. Op-

tical traps have been studied extensively for use in achieving Bose-Einstein condensates [24,25] and in regards to photon momentum exchange [26]. Nonclassical states have been investigated using optical traps as a means to enhance sensitivity [27] and for atomic clocks [28]. There have been studies of ways to regulate the phonon spectra in solidstate materials and devices during the past decade or so. Membrane cooling for cavity optomechanics facilitates improved sensing or a platform for quantum signal processing [29,30]. In Tamm plasmon resonators, efficient light-sound transduction has been shown [31], as has been found in distributed Bragg reflector GaAs/AlAs vertical cavities [32]. Such optical-phonon interactions can facilitate nonreciprocal elements, important in realizing optical isolators (a challenge for integrated photonics) [33–35] and directional amplifiers [36]. More generally, there are interesting lines of physics related to topological phases of sound and light [37].

Optical forces in nanostructured media are relevant in a variety of technologies. Nano-optomechanical actuators have been demonstrated that have the potential to impact optical signal processing [38], and a Si microdisk provided high-frequency signal processing, sensing, and metrology [39]. Other opportunities in quantum information processing [40], thermal and humidity sensing [41], optical logic gates [42], channel routing or switching, dispersion compensation, and tunable lasers [43] have also been considered. In addition, concepts for applying all-optical control to mechanical or physical devices present exciting opportunities for optomechanical systems. For example, the energy consumed in the electronic control of optical communication networks has driven interest in all-optical means of

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communication between computers [44] and in radio frequency photonics [45].

Based on the aforementioned and other related achievements, it is indeed remarkable that more than one century after the first measurements of optical force there remains such uncertainty with regard to force densities in condensed matter. By contrast, Maxwell's equations projected in Poynting's theorem is widely accepted as a means to describe electromagnetic energy. As an illustration, the physics behind photon drag with surface plasmons has recently been brought into question [46], despite the relatively long history of this phenomenon. To resolve open questions related to optical force density, a combined basic theoretical and experimental effort is needed. The key interface problem treated here relates to this goal.

Section II provides background on the force density model, and the specific boundary issues developed are introduced. The mathematical treatment in Sec. III is the key contribution of this work. Section IV considers mathematical and physical interpretations in relation to experiments, including suggestions for studies to evaluate the theory, as well as broader impacts in current application spaces. Following some conclusions in Sec. V, acknowledgments are presented.

II. OPTICAL FORCE DENSITY AND MODEL

To understand how light imparts a force throughout condensed matter, one must build a theory for the force density. In this way only, can a model be formed to determine mechanical mode excitation, as a result of the spatially dependent optical force density. The rigorous way to obtain the total force on a region of material is to integrate the force density over volume. With suitable boundary conditions, this integration can be moved across material interfaces with complete rigor. One should anticipate that there is a unique force density for a particular electromagnetic field distribution and physical problem, and hence a unique result from integration, based on the physics. The goal of the present work is to evaluate the requirement of the optical force density at material interfaces, in order to correctly connect the force density internal to a material to the external force, for example, to obtain the pressure.

The force density theory considered here stems from work done by Einstein and Laub [1], yielding results of note for the conservation of momentum in physical materials. This theory correctly predicts the important water experiment done by Ashkin and Dziedzic [47], as verified by us and previously described [48]. In that experiment, the fluid moved according to the local force density, and this was measured to produce a bulge or squeezing effect that is independent of incident laser direction [47]. This is the theory for force density that has been used to describe forces (deflections) in structured Au on SiN membranes [49]. It or related theory has been widely studied [50-54], but not in the sense of the boundary condition treated here.

At optical frequencies, where magnetism (impacting the magnetization) is negligible, it is the locally homogenized polarization (exhibited in the complex electric susceptibility or dielectric constant, possibly in tensor form) that describes the material in both an electromagnetic and force density

sense. All force formulations must contend with a spatially varying dielectric and hence interfaces. By way of example, the laser optical trap model with dielectric beads relies on a gradient force involving the spatial derivative of the electric field, E [5]. Relevant is the dipole force density of the form $(\mathbf{P} \cdot \nabla)\mathbf{E}$, where **P** is the polarization [51,52]. Treatment of this dipole term is important in such inhomogeneous material systems, as will become apparent here.

In the theoretical development that follows, the basis for the force density is derived from electromagnetic principles to make clear how kinetic force (that moves condensed matter) can be related to the field description in a way that is meaningful for optical materials. This allows boundary conditions that conserve momentum to be developed, leading to the primary contribution of the present work.

III. FORCE DENSITY BOUNDARY CONDITION

A. Electromagnetic fields and related mathematics

The starting point is assumed to be Maxwell's equations and a stationary reference frame. Consequently, a nonrelativistic form of the kinetic force density is derived using the classical form of the Abraham momentum. Using the standard assumption of sufficient boundary smoothness to allow a locally planar assumption for an interface, the usual field boundary conditions are extended to those that apply to the momentum density. As a result, a boundary condition applicable for the force density is obtained, assuming physical materials and optical frequencies. Resulting is a kinetic force density with Cauchy principal value integration to obtain local forces, including at boundaries. The implications in relation to experiments are addressed in Sec. IV.

Maxwell's equations are written with all source terms on the right-hand side, leading to

$$\nabla \times \mathbf{E} + \mu_0 \frac{\partial \mathbf{H}}{\partial t} = -\mu_0 \frac{\partial \mathbf{M}}{\partial t},\tag{1}$$

$$\nabla \times \mathbf{H} - \epsilon_0 \frac{\partial \mathbf{E}}{\partial t} = \frac{\partial \mathbf{P}}{\partial t} + \mathbf{J},$$
 (2)

$$\epsilon_0 \nabla \cdot \mathbf{E} = -\nabla \cdot \mathbf{P} + \rho, \qquad (3)$$

$$\nabla \cdot \mathbf{H} = -\nabla \cdot \mathbf{M},\tag{4}$$

with **H** being the magnetic field, **M** the magnetization, **J** the source electric current density, ρ the electric charge density, μ_0 the permeability of free space, and ϵ_0 the permittivity of free space. Taking the cross product of $\epsilon_0 \mathbf{E}$ with (1) and $\mu_0 \mathbf{H}$ with (2), and adding the resulting equations, gives

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$$\epsilon_{0}\mathbf{E} \times (\mathbf{\nabla} \times \mathbf{E}) + \mu_{0}\mathbf{H} \times (\mathbf{\nabla} \times \mathbf{H}) + \mu_{0}\epsilon_{0}\mathbf{E} \times \frac{\partial\mathbf{H}}{\partial t} - \mu_{0}\epsilon_{0}\mathbf{H} \times \frac{\partial\mathbf{E}}{\partial t} = -\mu_{0}\epsilon_{0}\mathbf{E} \times \frac{\partial\mathbf{M}}{\partial t} + \mu_{0}\mathbf{H} \times \frac{\partial\mathbf{P}}{\partial t} + \mu_{0}\mathbf{H} \times \mathbf{J}.$$
 (5)

With a little work [51], the triple cross product terms in (5) can be written in the form

$$\epsilon_{0}\mathbf{E} \times (\nabla \times \mathbf{E}) + \mu_{0}\mathbf{H} \times (\nabla \times \mathbf{H})$$
$$= \nabla \cdot \mathbf{T}_{E} + \epsilon_{0}(\nabla \cdot \mathbf{E})\mathbf{E} + \mu_{0}(\nabla \cdot \mathbf{H})\mathbf{H}, \qquad (6)$$

with

$$\mathbf{\Gamma}_E = \frac{1}{2} (\epsilon_0 \mathbf{E} \cdot \mathbf{E} + \mu_0 \mathbf{H} \cdot \mathbf{H}) \mathbf{I} - \epsilon_0 \mathbf{E} \mathbf{E} - \mu_0 \mathbf{H} \mathbf{H}$$
(7)

being the so-called Maxwell stress tensor [55], and where **I** is the identity matrix. Here, the divergence of the tensor in the (x_1, x_2, x_3) coordinate system is

$$\boldsymbol{\nabla} \cdot \mathbf{T} = \begin{bmatrix} \frac{\partial T_{11}}{\partial x_1} + \frac{\partial T_{21}}{\partial x_2} + \frac{\partial T_{31}}{\partial x_3} \\ \frac{\partial T_{12}}{\partial x_1} + \frac{\partial T_{22}}{\partial x_2} + \frac{\partial T_{32}}{\partial x_3} \\ \frac{\partial T_{13}}{\partial x_1} + \frac{\partial T_{23}}{\partial x_2} + \frac{\partial T_{33}}{\partial x_3} \end{bmatrix},$$
(8)

and the dyadic product of two vectors is defined according to $(\mathbf{ab})_{ij} = a_i b_j$ [50], giving, for example,

$$\mathbf{EE} = \begin{bmatrix} E_1^2 & E_1E_2 & E_1E_3\\ E_2E_1 & E_2^2 & E_2E_3\\ E_3E_1 & E_3E_2 & E_3^2 \end{bmatrix}.$$
 (9)

Substituting (6) into (5) gives

$$\nabla \cdot \mathbf{T}_{E} + \epsilon_{0} (\nabla \cdot \mathbf{E}) \mathbf{E} + \mu_{0} (\nabla \cdot \mathbf{H}) \mathbf{H}$$

= $-\mu_{0} \epsilon_{0} \mathbf{E} \times \frac{\partial \mathbf{H}}{\partial t} + \mu_{0} \epsilon_{0} \mathbf{H} \times \frac{\partial \mathbf{E}}{\partial t}$
 $-\mu_{0} \epsilon_{0} \mathbf{E} \times \frac{\partial \mathbf{M}}{\partial t} + \mu_{0} \mathbf{H} \times \frac{\partial \mathbf{P}}{\partial t} + \mu_{0} \mathbf{H} \times \mathbf{J}.$ (10)

We shall consider the Abraham momentum density, which is given by

$$\mathbf{g} = \frac{1}{c^2} \mathbf{E} \times \mathbf{H},\tag{11}$$

with *c* being the speed of light in vacuum. In free space, all forms of the photon momentum give $\hbar k_0$, with $\hbar = h/(2\pi)$, *h* being Planck's constant, and $k_0 = \omega/c$ the free space wave number at circular frequency ω (see Appendix). In background media, the description of a photon has been the subject of considerable attention (see Ref. [26], for example).

Taking the time derivative of (11), we have

$$\frac{\partial \mathbf{g}}{\partial t} = \mu_0 \epsilon_0 \mathbf{E} \times \frac{\partial \mathbf{H}}{\partial t} - \mu_0 \epsilon_0 \mathbf{H} \times \frac{\partial \mathbf{E}}{\partial t}.$$
 (12)

Using (10) and (12),

$$\nabla \cdot \mathbf{T}_{E} + \epsilon_{0} (\nabla \cdot \mathbf{E}) \mathbf{E} + \mu_{0} (\nabla \cdot \mathbf{H}) \mathbf{H} + \frac{\partial \mathbf{g}}{\partial t}$$
$$= -\mu_{0} \epsilon_{0} \mathbf{E} \times \frac{\partial \mathbf{M}}{\partial t} + \mu_{0} \mathbf{H} \times \frac{\partial \mathbf{P}}{\partial t} + \mu_{0} \mathbf{H} \times \mathbf{J}. \quad (13)$$

The Einstein-Laub momentum flow or stress tensor is defined as [51,53]

$$\mathbf{T} = \frac{1}{2} (\epsilon_0 \mathbf{E} \cdot \mathbf{E} + \mu_0 \mathbf{H} \cdot \mathbf{H}) \mathbf{I} - \mathbf{D} \mathbf{E} - \mathbf{B} \mathbf{H}, \qquad (14)$$

with $\mathbf{D} = \epsilon_0 \mathbf{E} + \mathbf{P}$ being the electric flux density and $\mathbf{B} = \mu_0(\mathbf{H} + \mathbf{M})$ the magnetic flux density. One arrives at (14) by considering the free space contribution, in the form of (7), and those due to polarization and magnetization, as described by a number of people (see Ref. [51]). This amounts to writing $\mathbf{T}_E + \mathbf{T}_P + \mathbf{T}_M = \mathbf{T}$, with $\mathbf{T}_P = -\mathbf{P}\mathbf{E}$ and $\mathbf{T}_M = -\mu_0\mathbf{M}\mathbf{H}$ providing the polarization and magnetization contributions, respectively (i.e., the material effects).

With use of the vector identities

$$\mathbf{P}(\mathbf{\nabla} \cdot \mathbf{P})\mathbf{E} = -\mathbf{\nabla} \cdot (\mathbf{P}\mathbf{E}) + (\mathbf{P} \cdot \mathbf{\nabla})\mathbf{E}, \qquad (15)$$

$$-(\nabla \cdot \mathbf{M})\mathbf{H} = -\nabla \cdot (\mathbf{M}\mathbf{H}) + (\mathbf{M} \cdot \nabla)\mathbf{H}, \qquad (16)$$

and incorporating (14) into (13), we have the key result

$$\nabla \cdot \mathbf{T} + \frac{\partial \mathbf{g}}{\partial t} = \mu_0 \mathbf{H} \times \frac{\partial \mathbf{P}}{\partial t} - \mu_0 \epsilon_0 \mathbf{E} \times \frac{\partial \mathbf{M}}{\partial t} + \mu_0 \mathbf{H} \times \mathbf{J}$$
$$-\rho \mathbf{E} - (\mathbf{P} \cdot \nabla) \mathbf{E} - (\mathbf{M} \cdot \nabla) \mathbf{H}, \qquad (17)$$

which we draw upon throughout the remainder of this development. Equation (17) provides a basis to consider arbitrary material responses (nonlocal in time and with material and geometric dispersion). For source-free (where the incident field, that without the material, is due to some remote source), nonmagnetic materials, (17) becomes

$$\frac{\partial \mathbf{g}}{\partial t} = -\boldsymbol{\nabla} \cdot \mathbf{T} - \left[\frac{\partial \mathbf{P}}{\partial t} \times \mu_0 \mathbf{H} + (\mathbf{P} \cdot \boldsymbol{\nabla}) \mathbf{E}\right], \quad (18)$$

where loss and hence free charge motion is incorporated into the temporal Fourier form for the polarization, which for isotropic materials becomes $\mathbf{P}(\mathbf{r}, \omega) = \epsilon_0 \chi_E(\mathbf{r}, \omega) \mathbf{E}(\mathbf{r}, \omega) =$ $\epsilon_0[\epsilon(\mathbf{r}, \omega) - 1]\mathbf{E}(\mathbf{r}, \omega)$, with complex electric susceptibility χ_E and dielectric constant ϵ .

B. Boundary conditions

Maxwell's curl equations applied to boundaries with sufficient smoothness and for physical materials (precluding a perfect electric conductor) yield the tangential field boundary conditions

$$\hat{\mathbf{n}}_{21} \times (\mathbf{H}_1 - \mathbf{H}_2) = \mathbf{J}_s = 0, \tag{19}$$

$$\hat{\mathbf{n}}_{21} \times (\mathbf{E}_1 - \mathbf{E}_2) = 0, \tag{20}$$

with $\hat{\mathbf{n}}_{21}$ being the unit normal vector directed from Region 2 into Region 1. These follow from Ampere's law (2) and Faraday's law (1), respectively, transformed into integral form with use of Stokes' theorem, and then applied to a small (rectangular) domain spanning the interface, in the usual manner. The boundary conditions in (19) and (20), i.e., continuity of tangential magnetic and electric fields, combined with the momentum density in (11), yield

$$\hat{\mathbf{n}}_{21} \cdot (\mathbf{g}_1 - \mathbf{g}_2) = 0 \tag{21}$$

as a momentum density boundary condition. Equation (21) is quite interesting, because it implies that the normal component of the Abraham momentum density is preserved across an interface, by virtue of the field boundary conditions. It also indicates that the momentum density in media with χ_E other than zero has the same form as that in vacuum (or some other material), as expected for the Abraham form. Of course, the normal components of the Poynting vector ($\mathbf{S} = \mathbf{E} \times \mathbf{H}$) are also preserved across interfaces, a statement of energy conservation based on Poynting's theorem,

$$\oint (\mathbf{E} \times \mathbf{H}) \cdot d\mathbf{s} = -\int \left(\mathbf{E} \cdot \frac{\partial \mathbf{D}}{\partial t} + \mathbf{H} \cdot \frac{\partial \mathbf{B}}{\partial t} \right) dv, \quad (22)$$

applied here to a source-free region ($\mathbf{J} = 0$) containing locally homogenized material parameters [dictating $\mathbf{D}(\mathbf{r}, t)$ and $\mathbf{B}(\mathbf{r}, t)$] and derived from Maxwell's equations with use of a vector identity and the divergence theorem.

Let us assume that the condensed matter experiencing an optical force is fixed in position for a time that is large relative to the temporal period of the optical wave. This is discussed in some detail in Sec. IV, but is reasonable for material having a spatial support on the few-nanometer scale or larger (with significant mass and where a classical field description with locally homogenized parameters can be applied). Mathematically, we can simply assume that the material is fixed in space during some time interval. As a result, the complex spatiotemporal optomechanics problem can be simplified in a manner representative of many experimental situations. Integrating (18) over suitably small spatial and temporal supports results in

$$\int_{v} \int_{t} \frac{\partial \mathbf{g}}{\partial t} dv dt = -\int_{v} \int_{t} \nabla \cdot \mathbf{T} dv dt$$
$$-\int_{v} \int_{t} \left[\frac{\partial \mathbf{P}}{\partial t} \times \mu_{0} \mathbf{H} + (\mathbf{P} \cdot \nabla) \mathbf{E} \right] dv dt.$$
(23)

More specifically, the constraint underlying (23) would be that, given a specific spatial support, a time interval is specified as being sufficiently short. Computationally, this would relate to a numerical convergence criteria. With these caveats, we proceed with evaluation of (23). Within any domain, it is straightforward to evaluate (23) using computational electromagnetics, once the locally homogenized material parameters $[\epsilon(\mathbf{r}, \omega)$ for all ω] are defined.

One can apply the divergence theorem to (23), assuming a locally smooth surface that is fixed during the relevant time interval, allowing the first term on the right to be expressed as a closed surface integral for the region defining the volume. Therefore, with consideration of an infinitesimally small region that spans an interface between Regions 1 and 2, (23) becomes

$$\int_{v} \int_{t} \frac{\partial \mathbf{g}}{\partial t} dv dt = -\oint \int_{t} T_{ij} ds_{k} dt -\int_{v} \int_{t} \left[\frac{\partial \mathbf{P}}{\partial t} \times \mu_{0} \mathbf{H} + (\mathbf{P} \cdot \nabla) \mathbf{E} \right] dv dt,$$
(24)

where the appropriate permutations of the surface integral variables are implied. Assuming finite fields [sufficient local boundary smoothness, as used to arrive at (19) and (20), and thus finite **E** and **H** and hence **g** and **P**], we have

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 $v \rightarrow 0$

$$\lim_{v \to 0} \left\{ \int_{v} \int_{t} \frac{\partial \mathbf{g}}{\partial t} dv dt \right\} = 0,$$

$$\left\{ -\oint \int_{t} T_{ij} ds_{k} dt \right\}$$
(25)

$$-\int_{v}\int_{t}\left[\frac{\partial \mathbf{P}}{\partial t} \times \mu_{0}\mathbf{H} + (\mathbf{P}\cdot\nabla)\mathbf{E}\right]dvdt \bigg\}$$
$$=\lim_{v\to 0}\left\{-\oint \int_{t}T_{ij}ds_{k}dt - \int_{v}\int_{t}(\mathbf{P}\cdot\nabla)\mathbf{E}dvdt\right\}$$
$$= 0.$$
(26)

Equations (25) and (26) are key to evaluating boundarycondition requirements for the kinetic force density, yet to be formed.

Selecting the normal components from (26) yields

$$\lim_{v \to 0} \left\{ \hat{\mathbf{n}}_{21} \cdot \oint \int_{t} T_{ij} ds_k dt \right\}$$
$$= \lim_{v \to 0} \left\{ -\hat{\mathbf{n}}_{21} \cdot \int_{v} \int_{t} (\mathbf{P} \cdot \nabla) \mathbf{E} dv dt \right\}.$$
(27)

If we assume that $\hat{\mathbf{n}}_{21} = \hat{\mathbf{y}}$ and two-dimensional (2D) Cartesian coordinates (no field variation in the third dimension), then

$$\lim_{v \to 0} \left\{ \oint \int_{t} (T_{yx} dy dz + T_{yy} dx dz) dt \right\}$$
$$= \lim_{v \to 0} \left\{ \oint \int_{t} T_{yy} dx dz dt \right\}$$
(28)
$$= \lim_{v \to 0} \left\{ \int_{v} \int_{t} -\hat{\mathbf{y}} \cdot [(\mathbf{P} \cdot \nabla)\mathbf{E}] dv dt \right\}.$$
(29)

The point here is that a Dirac δ function term that exists from the spatial derivative [formation of $(\mathbf{P} \cdot \nabla)\mathbf{E}$ at the interface] is associated with the step in T_{vv} , so that

$$\lim_{v \to 0} \left\{ \oint \int_t T_{yy} dx dz dt + \int_v \int_t \hat{\mathbf{y}} \cdot [(\mathbf{P} \cdot \nabla) \mathbf{E}] dv dt \right\} = 0.$$
(30)

Physical materials are dispersive, so (27) is correctly interpreted in the temporal Fourier domain and with spectral superposition. In phasor form, $(\mathbf{P} \cdot \nabla)\mathbf{E}$ is complex and frequency dependent, as is the Dirac δ function coefficient at an interface.

C. The kinetic force density

We now ascribe a kinetic force based on the electromagnetic field description, with consideration of the relevant physics and with the goal of predicting experimental observations. This development will utilize (17) and the associated boundary conditions that assume local smoothness.

At the interface between two materials, say free space and some solid-state material, there is in general an electric-field component normal to the interface. In the mathematical (and nonphysical) case of a perfect electric conductor, an unlimited charge can respond instantaneously, and $\hat{\mathbf{n}}_{21} \cdot \mathbf{D}_1 = \rho_s$, with ρ_s the local surface charge density. Here, Region 2 is the perfect conductor and ρ in (17) exists at the surface and is described as a Dirac δ function with respect to the normal direction. This surface charge would experience a force just as Lorentz described [56]. The angle-dependent force on a mirror, as presented by Planck [57], also follows. Likewise, for the electrostatic case with free charge ($\omega = 0$), one associates a similar force density with the surface charge density from the normal electric flux density boundary condition (and Gauss' law). We can consider both of these situations as being a local force on a charged surface, at least in the context of (17). In such situations, charge is experiencing a force, and to the degree that a local solid-state material contains such net (local) charge, one might anticipate a resulting force.

What about interfaces with neutral materials and at optical frequencies? While an electric-dipole moment is established in a material (resulting in the local, average dipole moment per unit volume, i.e., the polarization P, a complex quantity in the temporal Fourier domain), moving this material with a kinetic force requires collective motion of these dipoles (the neutral material picture) or free charge motion over length scales large relative to an underlying atomic or lattice dimension (thereby achieving a locally charged material, most rapidly from electron transport). One could speculate that the ability to translate material relates to local neutrality and to the distance and timescale for local charge transport. At optical frequencies, a simple charge transport analysis (Lorentz force with a practical laser beam power and the associated electric field that accelerates an electron) indicates that the femtosecond-timescale displacement of charge (local electron motion, within a unit cell, or free, with the timescale involved for an optical wave that moves charge back and forth) is very small. By small, this means relative to atomic scales, as can be verified for the electric field from a high-power laser and with use of the rest mass and charge for an electron [58]. Because the free charge does not respond fast enough to the optical field (a phenomenon that results in kinetic inductance), one arrives at the conclusion that free charge motion can be neglected in the boundary representation for optical forces (but not in longer-range force density phenomena, because this is exhibited in loss and hence the imaginary part of the dielectric constant [59]). The charge-neutral material at the interface will respond as an electric dipole over the atomic or unit cell (with homogenization) length scales that are small relative to the penetration depth for optical fields. One thus arrives at the conclusion that there will be no net force on these dipoles very close to the interface for physical neutral materials, and hence zero force density as the boundary is approached. Equivalently, the atoms in an infinitesimally thin material slice will be predominantly forward scattering, resulting in vanishingly small momentum transfer. All of this suggests that the force density at the interface cannot be infinite physically (in such neutral materials), and that any Dirac delta terms associated with $(\mathbf{P} \cdot \nabla)\mathbf{E}$ would not exist in reality in this situation at optical frequencies (despite the presence of a polarization surface charge density). A separate question might be whether this Dirac delta term represents an equivalent effect that is not captured classically by another means, but that position is unsatisfying. Physically, then, and at optical frequencies, we might be inclined to reject a Dirac delta term in the kinetic force density, that involved in creating motion of material and associated with the electromagnetic description.

The electromagnetic momentum density \mathbf{g} in material has a long history of debate. Without belaboring the point, this is of relevance because of the boundary treatment. We consider here a mathematical-field interpretation that, when folded into a conservation condition related to momentum, can be interpreted in a physical sense without ambiguity. Equation (21) presents a requirement on (the normal component of) the momentum density on either side of an interface based on electromagnetic boundary conditions. The momentum flow (stress) tensor (**T**) can have discontinuous components at an interface if the force density has a Dirac delta term, and this can only come from $(\mathbf{P} \cdot \nabla)\mathbf{E}$ in (18) for dielectric media. If one considers that photon momentum must flow across an interface, then the relevant components of **T** have to be continuous across the interface, i.e., the T_{ij} are conserved quantities. If one accepts an infinite Dirac delta force density at the boundary, then **T** can have discontinuous steps. To force continuity in **T** and hence momentum flow, one can add the Dirac delta contribution from $(\mathbf{P} \cdot \nabla)\mathbf{E}$ to the integral of $\nabla \cdot \mathbf{T}$. Doing so requires consideration of the external stress tensor elements while adhering to momentum conservation requirements.

This backdrop leads to an interpretation of the electromagnetic expression in (17) for the description of kinetic force density as

$$[\nabla \cdot \mathbf{T}]_{PV} + \frac{\partial \mathbf{g}}{\partial t} = \mu_0 \mathbf{H} \times \frac{\partial \mathbf{P}}{\partial t} - \mu_0 \epsilon_0 \mathbf{E} \times \frac{\partial \mathbf{M}}{\partial t} + \mu_0 \mathbf{H} \times \mathbf{J}$$
$$-\rho \mathbf{E} - [(\mathbf{P} \cdot \nabla) \mathbf{E}]_{PV} - [(\mathbf{M} \cdot \nabla) \mathbf{H}]_{PV}$$
$$= -\mathbf{f}_k, \qquad (31)$$

with \mathbf{f}_k being the kinetic force density and the implication of $[\cdot]_{PV}$, the principal value in the Cauchy sense with integration, is that the Dirac delta boundary contributions from the right have been separated and added to the divergence of the momentum flow tensor on the left, if the boundary is moved across the interface. Consequently, the kinetic force density, that associated with collective material motion, becomes

$$\mathbf{f}_{k} = \frac{\partial \mathbf{P}}{\partial t} \times \mu_{0} \mathbf{H} - \frac{\partial \mathbf{M}}{\partial t} \times \mu_{0} \epsilon_{0} \mathbf{E} + \mathbf{J} \times \mu_{0} \mathbf{H} + \rho \mathbf{E} + [(\mathbf{P} \cdot \nabla)\mathbf{E}]_{\mathrm{PV}} + [(\mathbf{M} \cdot \nabla)\mathbf{H}]_{\mathrm{PV}}.$$
(32)

Equation (32) is the key result developed here.

At optical frequencies and for physical materials, we might conclude that the force density in (32), without boundary Dirac delta terms and hence within the spatial support of material media, be applied. Electromagnetic momentum flow in the field sense is thus preserved across boundaries, as is shown in (25) and (26), in a manner that is physically satisfying. This implies that the total kinetic force becomes

$$\mathbf{F}_{k} = \int_{v} \int_{t} \left[\frac{\partial \mathbf{P}}{\partial t} \times \mu_{0} \mathbf{H} - \frac{\partial \mathbf{M}}{\partial t} \times \mu_{0} \epsilon_{0} \mathbf{E} + \mathbf{J} \times \mu_{0} \mathbf{H} + \rho \mathbf{E} + (\mathbf{P} \cdot \nabla) \mathbf{E} + (\mathbf{M} \cdot \nabla) \mathbf{H} \right] dv dt, \qquad (33)$$

where the volume integral is interpreted in the Cauchy principal value sense (so the PV subscripts are removed from the relevant terms in the kernel). Both the temporal and spatial integration requirements are addressed in Sec. IV. Briefly, the normalized time integral (preserving units) is over a period or an interval that is sufficiently small relative to the kinetics of the system, and could be over the carrier period for modulated light, so the force becomes a function of the modulation time variable (see, for example, prior work on force [52] and energy [60] in dispersive materials). The volume integral also assumes that the spatial distribution of material is fixed over the time interval considered.

It is important to note that (32) and (33) impose all relevant boundary conditions and momentum flow is conserved. That model was matched to subsequent experimental studies with membrane deflection that supported the existence of enhanced optical pressure [49]. The broader implications of this theory holding physically, and hence (32), are significant, as outlined in Sec. IV.

IV. DISCUSSION

Interpretations of the optical force boundary-condition theory developed in Sec. III are described in relation to material properties and how solutions to optomechanical problems can be formed to describe experimental situations. The role of new experiments in exploring the forces near interfaces and at the nanometer length scale is explored, and the impact on several current research trends is outlined. Throughout this section, published numerical simulation results and data from experiments are noted because they relate to the central theme of a boundary constraint for optical forces, i.e., electromagnetic force theory in condensed matter and at optical frequencies, as would be the case with use of lasers.

A. Motion of solid-state media

Consider a condensed-matter system with mass, as would apply in the laboratory setting with, say, membrane motion. Flexural modes generally accessed in SiN membranes of typical dimensions used in optomechanics experiments [49,61] have lower-order resonances below the MHz range. A thin plate model for the eigenmodes for a 100 nm thick, low-stress SiN membrane [62,63] suggests that the local linear velocity is of the order of 10^3 m/s. In one optical cycle, say 10^{-15} s, the membrane moves 10^{-12} m (10^{-2} Å). The quality factors in Au plasmonic cavities are modest, less than 100 typically [64], which results in a cavity lifetime of less than 10^{-13} s. During this time (10^{-13} s) , the membrane will move about 10^{-10} m (1 Å). Both these timescales (optical period and cavity excitation time in plasmonics [49]) suggest material motion might be neglected in the optomechanical analysis for such solid-state systems over these periods. This leads to a practical interpretation for (32) and (33) in experimental situations involving membranes, beams, cantilevers, beads, etc. Consequently, and as done previously [60], a temporal average over the carrier period presents the modulated light description and the plausible time frame for a mechanical response. Hence, the time variation measured with a sensing system for membrane deflection measures (at or below) the modulation temporal profile for the (laser) light.

B. Time-averaging in force density

The time integrations in Sec. III and specifically in (32) and (33) can be written more precisely. Modulated light and a local average over the optical (carrier) having period t_0 can be described as

$$\langle f(t) \rangle(t_n) = \frac{1}{t_0} \int_{t_n - t_0/2}^{t_n + t_0/2} f(t) dt,$$
 (34)

where f(t) is a scalar function that varies with time (and position), such as a component of the vector force density, and t_n is the time variable relevant for the mechanical system. With this definition, averages of the force density $[\langle \mathbf{f}(\mathbf{r}, t) \rangle (t_n)]$

and total force or pressure can be formed to achieve timedependent results that vary with the modulation temporal envelope. This produces a (modulated-light) time variation in the force quantities that is commensurate with mechanical motion timescales [52].

C. Implications regarding the total optical force and conservation of momentum

The fundamental way we should view optical forces in materials is through the spatially and temporally dependent force density, as exhibited in (32). This forms the basis to obtain components of force and torque through integration, and to predict motion in three-dimensional (3D) space. Conservation of momentum is thus point-wise and comes from (31).

All materials must have continuity of the normal component of the Abraham momentum density, based on the field boundary conditions and as given in (21). Likewise, the constraints involving $\partial \mathbf{g}/\partial t$ and T_{ij} , (25) and (26) respectively, must hold at interfaces, as well as along any contour within a locally homogeneous domain. Equation (25) follows directly from the constraint in (21). In a sense, more interesting is the conservation condition in (26), where the integral of a (possible) Dirac delta force density term (from $(\mathbf{P}(\mathbf{r}, t) \cdot \nabla) \mathbf{E}(\mathbf{r}, t)$, with a normal component of the electric field) must be precisely compensated by the surface integral of respective stress tensor elements $T_{ii}(\mathbf{r}, t)$. What appears not to have been widely appreciated is that the condition in (26) holds with either: (i) a Dirac delta component of the force density at all dielectric interfaces added to the bulk force density, or (ii) with the Dirac delta contribution added to the stress tensor (so that the force density is integrated to form the total force without the Dirac term). Both precisely satisfy all mathematical constraints, but only one should be physically meaningful in a given situation. The point made in Sec. III C is that only case (ii) conforms to the physics of the system (with optical frequencies and physical, neutral materials), thus driving the choice of the principal value representation and the form for the kinetic force density $[\mathbf{f}_k(\mathbf{r}, t)]$ in (32) without the Dirac delta contribution. In special cases like normal incidence on a planar interface, interpretations (i) and (ii) are identical. Therefore, based on charge transport time constants and at optical frequencies, the total kinetic force, that which can move a structure in a manner measured by spatial motion or matter, would appear to be dictated by (33).

D. External momentum flow tensor

Equations (32) and (33) are applied within the spatial support of the medium. From the left of (31), the result from $[\nabla \cdot \mathbf{T}] + \partial \mathbf{g}/\partial t$ or its integral, respectively, gives the identical result. In this case, **T** from (14) is that in the material. If one moves the integration just across a material interface with, for example, free space, **T** assumes its free space value, resulting in (7). Then, the result from the integration of the Dirac delta at the surface is added to the stress tensor integration within the material. This produces a consistent internal and external description of the kinetic force and allows correct

interpretation of the time-dependent momentum flow into the material.

E. Nonlocal-in-time fields and impact on the force density

The fields in all physical materials are a result of current and all earlier excitations in time, making the description nonlocal in time (see Ref. [65], for example). Likewise, a nonlocal temporal response should also apply for the optical force in material and in (32), where the force at each point in matter is the result of the excitation history as conveyed by the nonlocal field responses. The same nonlocal-in-time situation holds for geometry-based means to store optical energy, such as cavities in materials [66], which opens new opportunities for optical force control, such as with optimization-based aperiodic structures [67]. On this basis, the established principles of nonlocal fields in materials is reviewed, as relevant to understanding the impact in (32).

The (locally homogenized [68]) polarization is correctly written in the frequency domain at each point in space as

$$\mathbf{P}(\mathbf{r},\omega) = \epsilon_0 \chi_E(\mathbf{r},\omega) \mathbf{E}(\mathbf{r},\omega), \qquad (35)$$

with the simple case of a scalar complex electric susceptibility χ_E being assumed for the constitutive parameter. Material responses in the form of (35) are how materials are modeled either quantum mechanically or through a classical picture, and generally how they are characterized experimentally. Taking the inverse Fourier transform of (35), we have

$$\mathbf{P}(\mathbf{r},t) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} \epsilon_0 \chi_E(\mathbf{r},\omega) \mathbf{E}(\mathbf{r},\omega) e^{-i\omega t} d\omega \quad (36)$$

$$= \int_{-\infty}^{+\infty} \epsilon_0 \chi_E(\mathbf{r}, \tau) \mathbf{E}(\mathbf{r}, t - \tau) d\tau.$$
(37)

Equation (37) is of course the standard treatment of dielectric materials expressed in convolution form [65] and subject to causality associated with the velocity c, where we assume a local space result. However, here the relevance is that the force density in condensed matter is also the result of the time history of the excitation. The nonlocal-in-time concept for optical forces is thus important and essential in the correct theoretical description, and it is incorporated into the theory in (32) and (33).

The fact that materials respond nonlocally in time requires consideration of this phenomenon in all optomechanical situations, including those described by modulated light and in sinusoidal steady state (with monochromatic illumination). Because materials store energy, this impacts both the energy exchange with the outside world [60] and also the force imparted. Optical cavities in various forms provide energy storage and this impacts the force [49,66]. Materials with gain (optical activity) can have negative pressure [52,59], also by virtue of the time history of the system.

F. Momentum flow into a volume

While the force density, i.e., a pointwise result as a function of time, provides a fundamental path to kinetic information, it is instructive to reflect on on a widely disseminated presentation of momentum flow into a volume and conservation conditions in relation to the results in Sec. III. For this purpose, Jackson [65] (third edition, Sec. 6.7, pages 260-261) is primarily drawn upon. That material follows the argument in Stratton [69] (pages 156—159). By starting with the development in Sec. III, we shall arrive at Eq. (6.122) in Jackson [65], then interpret the result in terms of what is and is not implied in this volume-based conservation result. In Jackson (pages 260–261), the simple situation of charges (ρ) and current density (J) in free space is treated, avoiding interfaces and nonlocal-in-time phenomena associated with physical material (physical atomic and molecular states), where the established Lorentz force density ($\mathbf{f} = \rho \mathbf{E} + \mathbf{J} \times \mathbf{B}$) is utilized. Here, we use the more general Einstein-Laub description (Sec. III) because of an interest in condensed matter, but the same general expression [Eq. (6.122) [65]] can be found. Following this, we consider the meaning of each term in light of the spatiotemporal boundary conditions for force density to ensure momentum flow.

From (31) and suppressing the PV, we have

$$\mathbf{f}_{k}(\mathbf{r},t) + \frac{\partial \mathbf{g}(\mathbf{r},t)}{\partial t} = -\nabla \cdot \mathbf{T}(\mathbf{r},t).$$
(38)

Integrating (38) over some volume and applying the divergence theorem (under the assumption that a fixed boundary is meaningful, implying some local time picture), we have

$$\int \left(\mathbf{f}_k(\mathbf{r},t) + \frac{\partial \mathbf{g}(\mathbf{r},t)}{\partial t} \right) dv = -\oint T_{ij}(\mathbf{r},t) ds_k.$$
(39)

Using the notation of Jackson, (39) can be written as

$$\frac{d}{dt}[\mathbf{P}_{\text{mech}}(t) + \mathbf{P}_{\text{field}}(t)] = -\oint T_{ij}(\mathbf{r}, t)ds_k, \quad (40)$$

which (in mathematical form) is Eq. (6.122) in Ref. [65]. Here, $d\mathbf{P}_{mech}(t)/dt = \int \mathbf{f}_k(\mathbf{r}, t) dv$ is the sum of all mechanical momenta associated with the optical field in the volume at each instant of time and $\mathbf{P}_{\text{field}}(t)$ is the integral of the (Abraham) field momentum $\mathbf{g}(\mathbf{r}, t)$. In words, (40) says that the momentum flow into the volume (described by the righthand side) is exactly the time rate of change of the sum of the total mechanical and field momenta in this volume. There are two important points to note about (40). One is that this conservation condition relates to the total time rate of change of mechanical plus field momenta and does not in general allow the two to be distinguished, or at least the mechanical component of the force is not uniquely determined without a suitable forward model. This means that, except with specific assumptions of the situation being modeled, one cannot determine the mechanical force from photon momentum flow. The other is that, beyond this ambiguity, there is no information about the spatiotemporal distribution of the forces in the volume. This prevents a general interpretation of how matter is translated by photons (e.g., there could be a number of regions within the volume or a contiguous material that could move in some fashion). Related momentum challenges in materials has been broached in a long line of papers, and, for example, Shockley proposed the concept of hidden momentum [70,71]. Without doing justice to the enormous scope of related papers, another example is the question of whether the Abraham or Minkowski momentum may hold in materials, and preservation of the total momentum was proposed as being implied through the kinetic and canonical momenta of the light [72]. We should also note that the relevance of the boundary description has been presented [73].

In Sec. III, we considered boundary conditions that must hold at interfaces, and a special case is a condensed-matter boundary with vacuum. Equation (27) specifies the requirement for continuity of the normal component of momentum flow. Applying (40) to a small domain spanning the vacuummaterial interface produces exactly the same conclusion. Thus, in a vacuum environment, the right-hand side of (40)would have elements from the free space stress tensor in (7), but in the presence of a material boundary [defined as being sufficiently close with regard to the temporal character of $\mathbf{T}(\mathbf{r}, t)$, in a causal sense], continuity of momentum flow may require adding the contribution from $[\mathbf{P}(\mathbf{r}, t) \cdot \nabla] \mathbf{E}(\mathbf{r}, t)$, assuming a dielectric interface. In the sense that (40) provides a statement of momentum conservation by flow into a volume, the boundary conditions developed in Sec. III B must hold, including when the photons are incident from vacuum with a well-defined photon momentum ($\hbar k_0$). This result is implicit in the boundary condition of (27) and with application of the resulting spatiotemporal force density in (32). Without this step and when $\mathbf{n}_{21} \cdot [\mathbf{P}(\mathbf{r}, t) \cdot \nabla] \mathbf{E}(\mathbf{r}, t) \neq 0$, momentum flow in this context would not be enforced (with application of \mathbf{T}_E). One can thus preserve photon flow, as in the free-space example treated in Jackson. In the case of free space and with some charge and current distribution in the enclosed volume, the momentum flow integral on the right-hand side of (40)is simply given by the time domain form of the elements of the Maxwell free-space stress tensor in (7) [65], but with an optical pulse overlapping a physical boundary, Sec. III shows that additional considerations are needed to preserve the momentum flow from vacuum into and out of the material.

The electromagnetic system described by (40) may be open, so other forces can be involved [50], and we consider two such situations. The first is mathematical, where the material is fixed in position, so, referring to (39), $\mathbf{f}_k(\mathbf{r}, t) =$ $-\mathbf{f}_{s}(\mathbf{r}, t)$, with \mathbf{f}_{s} being a force due to the (coupled) system which negates $d\mathbf{P}_{mech}/dt$ associated with optical fields in (40); this will be used here. The second is where a physical system is involved, and inertia, tension or structure support diminishes the net kinetic force density inducing motion. Such coupled system forces need to be considered to solve the kinematic problem in relation to experiments. Thus, use of the right-hand side of (40) and photon momenta through an external boundary does not provide the fundamental conservation condition related to motion of media, i.e., in general, it is not possible at that level to separate field and mechanical parameters (but some simple cases of practical interest can be treated in this manner).

With pulsed light, we could have the right-hand side of (40) be zero and only $d\mathbf{P}_{\text{field}}(t)/dt$ gives rise to nonzero $d\mathbf{P}_{\text{mech}}(t)/dt$. This illustrates the need to specify temporal initial conditions to determine motion, and emphasizes the role of nonlocal material temporal responses. This prior information would incorporate earlier excitation and energy storage. Also, causality exists with the densities (through the fields) but not locally from the volume integrals—because the local field-matter interactions are not measured. We now have a sense that care is needed in applying the time domain form in (40) to infer the kinetic force. However, for ideal charges

and currents in vacuum, which are described as being local in time (Lorentz in the case of the picture in Jackson [65]), the interpretation of (40) is reasonably straightforward.

G. Sinusoidal steady state

The sinusoidal steady-state case having a single circular frequency ω is mathematically and computationally convenient and provides access to the basic constitutive parameters. Such a model could also be of practical importance because relatively coherent laser light or low-frequency modulated light may be involved. In practice, the time frame for excitation to a quasimonochromatic case may need only be long compared with a measure such as the average photon lifetime in the material, possibly encompassing structured resonances [66,74]. The mathematical requirement is that the system modeled be linear and time invariant. This means, strictly speaking, there can be no motion and hence no momentum exchange. Assuming the momentum density in (11), one can show that $\langle \partial \mathbf{g} / \partial t \rangle = 0$, regardless of the material properties (see the Appendix of Ref. [54]).

With time dependence $\exp(-i\omega t)$, used in the inverse temporal Fourier transform, and considering the source-free dielectric (nonmagnetic material case), the time-average sinusoidal steady-state kinetic force density, from (32), can be written as

$$\langle \mathbf{f}_k \rangle = \frac{\omega \mu_0}{2} \operatorname{Im} \{ \mathbf{P} \times \mathbf{H}^* \} + \frac{1}{2} \operatorname{Re} \{ (\mathbf{P} \cdot \nabla) \mathbf{E}^* \}, \quad (41)$$

where $\langle \cdot \rangle$ is the temporal average (which does not vary with time because there is no modulation), Re{ \cdot } is the real part and Im{ \cdot } is the imaginary part, and **P**, **E**, and **H** are (the polarization, electric field, and magnetic field) phasors, with complex conjugates indicated by the superscript asterisks. The average total force (or pressure with area normalization) becomes

$$\langle \mathbf{F}_k \rangle = \int_{v} \left[\frac{\omega \mu_0}{2} \operatorname{Im} \{ \mathbf{P} \times \mathbf{H}^* \} + \frac{1}{2} \operatorname{Re} \{ (\mathbf{P} \cdot \nabla) \mathbf{E}^* \} \right] dv, \quad (42)$$

with principal value interpretation of the integral.

The physical situation described by (42) is one where the entire constellation of materials (at each point in space) has been invariant for infinite time. This implies that all points in the material are fixed in space (at least in the mathematical description). In practice and in an experiment, at some time, the local laboratory time or with reference to the optical modulation or mechanical time, the system could be considered released and to respond mechanically. Prior to this time, nothing is free to move in this mathematical description as a result of the time history of momentum transfer. Upon (mathematical, physical, conceptual) release, a kinematic problem would be addressed in a different model with the initial conditions presented by geometry, material, and the excitation conditions, all resulting from particular field and force density distributions. The local time average from (41) (force density) and (42) (force) would then provide an estimate of this average over the optical period at the initiation of motion. The external excitation field after this release time point would lead to a relativistic consideration.

Returning to (40), we can understand the relevance of the sinusoidal steady-state situation in this context. The integrated

parameters over some volume in (40) leads to

$$\langle \mathbf{F}_k \rangle = \left\langle \frac{d\mathbf{P}_{\text{mech}}(t)}{dt} \right\rangle = -\oint \langle T_{ij}(\mathbf{r}, t) \rangle ds_k,$$
 (43)

because $\langle d\mathbf{P}_{\text{field}}(t)/dt \rangle = 0$. This result can now readily be interpreted based on the results of Sec. III, notable here in terms of boundary conditions for momentum flow. To conform to a steady-state field solution, the media must be immobilized, so we have an additional force (related to a system force, so $\langle \mathbf{F}_s \rangle = -\langle \mathbf{F}_k \rangle$). After having been excited for an infinite time, consider that we allow release. Equation (43) no longer holds and we must return to the form in (40) for a volumetric description. However, (40) does not allow determination of kinetic motion in and of condensed matter, so we need to apply the force density in the time domain. Conceptually, the field energy stored in material at release now contributes to the field momentum, so the kinetic force is the result of this plus the flux of incident and scattered photons expressed by the right-hand side in (40).

Equation (43) cannot be applied in isolation, otherwise the sinusoidal steady-state condition is violated, and requires the additional force $\langle \mathbf{F}_s \rangle$. Consequently, regardless of the incident photon flux with corresponding momentum ($\hbar k_0$ per photon), the structure is fixed in space (mathematically). The boundary condition for the photon flow must be imposed on the right of (43), and this is the frequency-domain form of (27). Doing so means that the light is correctly treated at the boundary, regardless of whether it is in free space. Without including contributions from $\langle [\mathbf{P}(\mathbf{r}, t) \cdot \nabla] \mathbf{E}(\mathbf{r}, t) \rangle =$ $\operatorname{Re}\left\{\left[\mathbf{P}(\mathbf{r})\cdot\nabla\right]\mathbf{E}^{*}(\mathbf{r})\right\}/2$ into the right-hand side of (43), the physical material boundary condition (such as at a vacuum interface) is violated (Sec. III). The point here is that even though motion is strictly precluded, the field-based transfer of energy into the material needs to be preserved. Rather than (43), it is the time harmonic force density in (41) that presents a more useful result in relation to experiments and through $\langle \mathbf{f}_k(\mathbf{r},\omega)\rangle$.

H. Modeling the general time-dependent optical force

Optomechanics involves the coupled electromagnetic and mechanical problems where momentum is transferred from fields to matter, and the time and space domain differential operators are not in general independent. However, one can take simplifying steps based on either the physical situation of macroscopic objects (composed of a substantial number of atoms in a condensed state) having mass and hence inertia or numerical discretization descriptions for the differential operators and hence time steps. Practical aspects of mechanical timescales broached in Sec. IV A are drawn upon here.

Consider the discretized domain typical of finite difference and finite element solutions (computational physics, in this case, computational electromagnetics), for the solution of Maxwell's equations. By way of example, solution of the two coupled curl equations (Ampere's and Faraday's laws) represented in point form has become known as the finite difference time domain (FDTD) method [75]. In FDTD, the Yee algorithm utilizes displaced Cartesian grids for formation of the curl operators and finite difference representations (leading to central differences from Taylor-series expansions). While not necessarily the most computationally efficient approach, the simplicity of this algorithm and the broad availability of inexpensive computational resources have led to its popularity, and it serves the purpose of illustrating the time-step issues in optomechanics of interest here. Materials are specified only at a set of discrete points. At each time step, a spatial grid problem is solved, and the staggered grid arrangement allows updates according to the time step. Convergence can be proven in electromagnetics with a time step (times the background wave velocity) that is sufficiently small relative to the spatial grid size (the so-called Courant condition). One can consider that the optomechanical problem is treated in this manner. Any two time steps must be small relative to all other timescales. The object can be translated only in the time step after the wave (the field) enters the material, and as a result of the momentum transfer. As a result, one can assume that the condensed-matter system is fixed in space at some time step and all the boundary conditions are enforced, both electromagnetic [(19) and (20)] and optical force density, reflected in (32). Obviously, either a fixed medium or a coupled mechanical-electromagnetic problem needs to be addressed over the relevant timescales. Extensions of the FDTD approach may in fact be a simple way to implement an algorithm for the general coupled optical and mechanical problem, although material dispersion needs to be implemented as a temporal convolution, as in (37).

I. Forces in relation to the energy density picture from Maxwell

It is interesting to read Maxwell's description of the force (on a planar surface) as being related to electromagnetic energy, leading to the "stress of radiation" [2] (pages 440–441). While this was before the mathematical details were developed, including by Lorentz [56] and Einstein and Laub [1], the position still stands true in the sense of the underlying meaning of (31), where an electromagnetic field result is equated to the negative of the kinetic force density. Later, the "Maxwell-Bartoli" pressure was presented by Nichols and Hull [4] and Lebedev [3] (because of the joint conclusion both Maxwell and Bartoli made related to special cases), with the form

$$P_{\rm MB} = \frac{\langle S_i \rangle}{c} (1 + |\Gamma|^2), \tag{44}$$

where $\langle S_i \rangle$ is the magnitude of the time-average normally incident Poynting vector (a positive quantity) and Γ is the field reflection coefficient. Neither Maxwell nor Bartoli write the precise form of (44), but rather they consider situations that allow this result to be inferred (at least in certain circumstances). The form in (44) has been widely used for understanding the pressure on opaque (nontransmissive) materials.

Equation (44) can be derived for the sinusoidal steady state (single ω) situation and under the assumption that there is a single plane wave (normally incident and reflected) from an infinite planar material interface in vacuum. This involves writing the field as a superposition of incident and reflected plane waves (in the frequency domain), with field reflection coefficient magnitude $|\Gamma|$. Thus, the frequency domain forms of (7) or (14) (free space) or the right-hand side of (43) provide for this result. Again, there can be no motion and hence no momentum exchange. With the assumption that the closed

surface integral of the components $\langle T_{ij}(\mathbf{r}, t) \rangle$ has nonzero contributions only along the incident or reflected (planar) boundary, (44) results. The maximum pressure from (44) is thus $2\langle S_i \rangle / c$, the case for a perfect mirror.

What we now understand from the treatment in Sec. III is that (44) applies in a specific set of situations and that, depending on factors involving excitation, geometry, and time-history, pressures outside of $[0, 2\langle S_i \rangle/c]$ are in principle possible (see Secs. IV C and IV E). Systems offering optical gain can have negative pressure [52] and hence do not conform to (44). Separately, asymmetric plasmonic cavities were found numerically to have a pressure greater than $2\langle S_i \rangle/c$, based on application of (32) [66] and experiments were supportive [49]. In addition, there is a proposal for a system with surface wave modes induced on the back that may yield a negative pressure [54].

Let us again return to (43), offering the relevant conditions (assuming the immobilizing force is added). With imposition of the momentum flow boundary conditions implied by (25)and (26), resulting in a modified external region description for continuity of momentum flow at surfaces, it becomes possible to exceed $2\langle S_i \rangle/c$ (depending on the geometry and incident-field conditions). Again, this can be established from the right-hand side of (43) with the enforcement of momentum flow continuity across boundaries. This frequency domain revision to momentum flow is legitimate because the material is fixed in position for infinite time and the resulting fields, force density, force and momentum must satisfy these constraints. Strictly speaking then, (44) comes from a momentum flow description and not conservation of momentum, where the latter would require specifics of the pointwise momentum exchange and hence motion. With release, this single planar interface situation (which needs to be redefined in an experimental context, where infinite geometries are not possible), having some time-average pressure ($P_{\rm MB}$, for example), would then move according to a kinetic problem solution. With motion, we no longer have the sinusoidal steady-state condition. However, what about slow motion where we approximate this to be case? In situations where only the planar interface contributes to the reflected photons and all those incident on the medium are either reflected or eventually absorbed, and a single plane wave incident and reflected, then we expect (44) to continue to apply. This is the case for planar mirrors, making the result of great practical importance. Notably, this implies a local-in-time material description (and in that manner being analogous to ρ and **J** in free space and the situation treated in Jackson [65]). We are thus left with a position that (44) is a statement of momentum flow through a surface (a mathematical boundary in free space) and for a single-plane-wave problem with a local material response-and not a general conservation statement.

J. Experimental studies

The theory in (32) and (33) conforms with early pressure measurements [3,4], the Ashkin-Dziedzic water experiment [47], the extracted pressure on nanostructured membranes from deflection measurements [49], and the picture introduced by Maxwell for the situation he considered [2]. However, additional experimental studies are important to evaluate (32)

for materials at optical frequencies. Such experiments are not easy, and there is a need to extract the relevant optical force density information from measurement of the physical observable, displacement, for example.

An optomechanics experimental effort we pursued involved a SiN membrane with patterned Au that supported a surface-plasmon mode [49]. The resonance-enhanced plasmon modes were contributors and the surface waves involved have an electric-field component perpendicular to the interface, conforming to the mathematical situation involving a Dirac δ function at the interface due to $(\mathbf{P} \cdot \nabla)\mathbf{E}$ (Sec. III). Results from those experiments (with multiple measurements and statistical and error studies) were found to be consistent with an enhanced optical pressure, one that exceeded that with the same incident-field power density normally incident on a perfect mirror. The enhancement follows from application of (32), so the consistency of the model and the experiments is notable. However, consistency is distinct from uniqueness in the force density, and additional validation experiments are in order. Physically, this enhancement involves the dimension perpendicular to the "interface" and the nonlocal-in-time excitation of the asymmetric cavity modes-and hence energy storage in the material (over a timescale short relative to that associated with mechanical motion but long compared with the optical period).

Separately, we know that the force density in (32) describes the bulging water experiment done by Ashkin and Dziedzic [47]. However, this does not specifically address a force theory at interfaces. Likewise, despite the important experiments done by Jones and Leslie [76], more studies are needed.

From an electromagnetic field basis, the force density and pressure calculations applied within a resonant plasmonic cavity and yielding enhancement [49] can satisfy all physical requirements. Interestingly, this also allows for the possibility of pulling structures with light [54] in a manner that is fundamentally different from optical tweezing of beads in a trap [5]. In this case, it is the promotion of a plasmon surface wave on the back of a membrane that could produce pulling, something that is yet to be shown experimentally. Thus, evidence of pulling would provide further support for (32).

New experiments are needed to allow evaluation of the force density with optical electric fields normal to interfaces. These could involve a laser beam illuminating a planar membrane and a suitable measurement of displacement that is then calibrated to determine relative force and hence allow inference of the force density in the appropriate region. Alternatively, experiments could focus on various 3D objects that have the appropriate modes excited. While it is reasonably straightforward to obtain subnanometer displacement precision, based upon the success achieved in sensing motion in atomic force microscopes (motion of a sensing laser on a quadrant detector was used in Ref. [49] to monitor nanometer-scale membrane displacement), the challenge here is a set of experiments that provides a uniquely interpretable force density result. Meaningful conclusions require beam characterization, adequate measurement diversity, and the adjustment of key parameters (such as polarization, position, beam angle, and material). In addition, either through the calibration or separately, parametrized models (mechanical, electromagnetic, force) may need to be utilized that also require careful consideration. A pragmatic approach would be to perform a set of experiments and apply (32), along with any necessary thermal and mechanical model, to fit to the experimental data over a substantial parameter range. The ability of a theory to explain such a suite of experiments would then lead to a position on the optical force density in condensed matter and near to interfaces.

K. Contemporary applications

Several developing application domains are presented where improved understanding of the theory for the optical force density in condensed matter will have immediate ramifications. The set summarized here is anecdotal but serves the role of illustrating the need in contemporary physics to develop a rigorous optical force density theory.

Rather than optical cooling in a vacuum chamber to reduce vibrational modes, patterned silicon nitride (SiN) membranes have been studied [77]. To reduce external coupling and achieve low mass, tethered trampoline geometries have been recently considered [61]. With a goal of achieving high Qf at room temperature, where Q is the mechanical quality factor of a resonance and f is the operating frequency, photonic crystals were fabricated in SiN membranes to achieve mechanical mode band gaps [77]. A design approach based on topology optimization has been pursued and related to trampoline-like geometries [78]. In all of this work, the structure of the material has been modified to achieve a desired mechanical property. Understanding of the theory describing optical force density could enable structured material with optical force control to better regulate vibrational modes at higher temperatures. This might also prove valuable for control with exciton condensates in bilayer transition-metal dichalcogenides [79].

Substantial use has been made of optical tweezers, traps with a high-power laser to manipulate beads, for example, and calibration procedures related to trap stiffness have been developed [80]. With a suitable model for the optical force density in a bead in solution, additional information useful for characterizing the force and torque on biological molecules [16,81] should become available.

The field of nanophotonics has been driven in large part by the small mode volumes possible with cavities formed with metal-insulator-metal (MIM) modes. Notably, the field of molecular optomechanics [82-84] offers promise to employ such plasmonic cavities filled with molecules whose assembly arrangement is enabled through laser illumination. Understanding the spatially and temporally dependent optical force density in such situations is clearly important. Plasmonic cavities in various configurations have been shown to provide enhanced Raman-scattering signals [85,86], allowing more sensitive (surface-enhanced Raman scattering—SERS) measurements for molecules in the neighborhood of these cavities. With molecular optomechanics, enhanced Raman scatter in such picocavities is proposed as a platform for coherent control, optomechanics, and quantum signal processing [82]. Coherent Raman-sideband up-conversion with a few hundred molecules in a picocavity has been presented at the terahertz-midinfrared to visible light [83], illustrating tailorable molecular or plasmonic properties within small cavities. Molecular oscillators in small cavities, a new frontier, could benefit from knowledge of the force distribution throughout the picocavity and in the constituent molecules or nanoparticles. All of these domains have a need for an established optical force density theory.

Light [87,88] and electric field [89] can induce motility (chemical reconfiguration in polymers, resulting in motion), and this has been found to be related to chemical structure and be reversible. Photoisomerizable rods can bend upon absorption of photon energy and then be returned to the original state (molecular trans \leftrightarrow cis) [87]. However, the regulation and speed of this process might be improved with optical forces in additional to optical-energy-induced chemical forces, such as through a way to initiate the motion. Optical forces in nanostructured material may thus aid photomotility, hence the need to build insight.

L. Relationship to enhanced pressure

Among the interesting optical force phenomena that results from the force density in (32) and total force in (33) is an enhanced optical pressure, that exceeding the Maxwell-Bartoli result in (44), and this depends on both polarization and structure. This result follows directly from the boundary-condition development in Sec. III. It was discovered through sinusoidal steady-state simulations with surface-plasmon mode cavities in Au [90], and then found to conform to a set of experiments with a laser illuminating patterned Au on SiN membranes and measurements of deflection (incorporating a statistical treatment and with use of optical force density and thermal models, and with parameter fitting) [49]. This effect, being nonlocal in time and associated with an asymmetric cavity, requires an excitation time that is long relative to the cavity lifetime (Sec. IV E). This is of course satisfied in sinusoidal steady state, where there can be no motion and hence no photon momentum exchange inducing such motion (Sec. III). In relation to the experiment, there is thus the requirement of a system force (Secs. IV F and IV G) that resists motion over timescales commensurate with the optical period and cavity lifetime, and these relate to the mechanical system (tension, membrane frame mount, attachment to an optical bench that also supports the laser) [49]. Section IV G considered in substantial detail the requirements for the sinusoidal steady state model and how this might relate to experimental situations. Notably, enhancement is not evident based on (44), which can easily be established from related plasmonic cavity reflectance measurements [74] for structures similar to Ref. [90], as reviewed in Sec. IV I.

A recent comment [91] on Ref. [90] describes internal force density calculations that indeed have an enhanced pressure for resonant plasmonic cavity modes, but also calculations with application of the free-space stress tensor in (7) produce no pressure enhancement (a point made previously [54]), a result that was confirmed with application of the Maxwell-Bartoli form in (44). This should be no surprise, because the free space stress tensor (7), in the frequency domain (sinusoidal steady state), and under the assumption of a single, normal plane wave incident and a single (normal) propagating plane wave reflected, leads to (44) mathematically. Earlier measurements for reflected power from similar structures (nano-imprinted Au plasmonic cavities) [74], in conjunction with (44), takes us precisely to this result (as does any consideration of a plane-wave reflection coefficient). Based on (44), it was concluded that any pressure beyond $2\langle S_i \rangle/c$ is a violation of momentum conservation. However, sinusoidal steady-state conditions preclude direct inference of (44) in regard to conversation of momentum (see the preceding discussions in this section, including Sec. IV G). The correct way to think about this is with optical momentum flow subject to the steady-state motionless requirement. Separately, interpretation of a sinusoidal steady-state force model to an experimental situation (Secs. IV G and IV J) needs to be considered in relation to the specific situation (see Secs. IVF, IV G, and IV I). Section III develops a force density boundary condition that, when interpreted in the frequency domain, leads to a meaningful way to consider the vacuum-Au interface in Ref. [90]. This leads directly to a conclusion that enhanced pressure (beyond $2\langle S_i \rangle / c$) is possible without violation of conservation principles.

The specific situation considered in Refs. [90,91] is a periodic slot array in a thick Au film supporting plasmon modes and in free space [90,91], with a normally incident plane wave (transverse magnetic 2D simulation for the fields, with magnetic field out of the plane), under sinusoidal steady-state conditions. The structure is thus infinitely wide and thick enough for there to be negligible penetration. Scatter from the structure excites metal-insulator (MI) surface waves on the top and metal-insulator-metal (MIM) modes in the slot within the unit cell. With adjustment of the geometry and fixed wavelength, the waveguide modes can resonate, resulting in substantial fields in the material and hence energy density, with a corresponding impact on the force density. With regard to further investigation of this situation, the salient points from Ref. [91] are extracted and considered in the following:

Momentum. There is a sense that maximum momentum exchange with $\hbar k_0$ per photon leads to $2\langle S_i \rangle/c$ being the maximum force density for opaque materials [91]. We learn from Sec. III that this is true only in specific situations and not that considered in Ref. [90] (see Secs. IV C, IV F, IV G, and IVI). Rather than in an integral sense, the fundamental way to enforce momentum conservation is pointwise throughout space and through the force density in the time domain, as in Sec. III. This is how (40) is developed from the Lorentz force density through integration over a volume in free space [65]. While the stationarity and boundary-condition arguments are clear for the sinusoidal steady-state situation, projecting these in the volume description of Secs. IV F and IV G is perhaps illustrative-although we need to take care in inferring conservation conditions related to kinetic force. Importantly, (32) appears to satisfy requirements for the correct force density, as developed here and based on necessary boundary conditions.

What is known. It is assumed in Ref. [91] that the measure of force from the electromagnetic description in a free space region around any object is established and irrefutable—but there may be debate about the force in the material itself. In general, this cannot be correct, because the internal and external descriptions are coupled (Sec. III). The exterior mathematical picture comes from various steps stemming from the (presumably unique) force density in material and so cannot be independent of the result inside the material. In so far as the theory related to the mechanical interaction of photons with material remains open, so too does the exterior description, because of this exact relationship from superposition. More precisely, what has been established would be the photon momentum flow in free space. Distinct is a relationship between this and kinetic motion of material.

Physical picture. In describing the maximum momentum exchange between a photon and an object as the interpretation for the sinusoidal state state simulations presented, the assumptions in the model (see Sec. IV G) and the relationship to an experimental situation is misstated in Ref. [91]. With a monochromatic field solution and corresponding force description used, it is implicit that the object is fixed in space and unchanged for infinite time. By using the term "floating" in Ref. [91], a physical situation that violates the requirements of the mathematical model is implied, which is misleading. A distinction is made between forces on part of the object (say the interior) and the total force [exterior, forced to adhere to some restriction, as from (44)]. To be precise, there is an electromagnetic field formulation that leads to a vector force density and hence components of a total force through integration (Sec. III). The object moves most fundamentally due to the spatiotemporal mechanical force density, and this provides the basic link between photons and induced mechanical action (flexural and longitudinal modes in a membrane, for instance).

Mathematics. The sentiment in Ref. [91] that using the external, free-space stress tensor [the frequency domain form of (7)] to circumvent a nonphysical Dirac δ function at surfaces [91] is fundamentally wrong. The line of mathematics starts with an electromagnetic field description, ascribes a meaningful force density, and then, through integration, one arrives at a force or, with a suitable structure and area normalization, the pressure (Sec. III). With condensed matter described by a dielectric constant, the free space stress tensor implies the presence of a Dirac delta term when the normal component of the electric field is discontinuous and there is a spatial derivative with respect to that direction—see Sec. III for the details. There is concern in Ref. [91] about numerically evaluating a Dirac term (under FDTD) [91], driving implementation of a Lorentz force density evaluation that involves $\nabla \cdot \mathbf{P}$ (that, incidentally, does not describe correctly the Ashkin-Dziedzic water experiment [47]). However, there remains a Dirac delta term at dielectric interfaces (from the derivative of a Heaviside step function describing the spatial extent of a well-defined **P**), i.e., this does not circumvent the influence of spatial derivatives of fields at interfaces, should they be relevant. One must conclude that the total force results in Ref. [91] are effectively determined using one approach, that involving a free-space stress tensor in the frequency domain [which leads to (44)], rather than there being distinct approaches, and these identical results are presented in separate figures. The point to consider here is that the mathematics (in the frequency domain here) has a firm basis that does not benefit from loose concepts of what the incident momentum per photon implies in this situation with stationary material and infinite excitation time, and that physical and experimental interpretation then has a viable foundation.

To summarize in regard to Ref. [91], Sec. III makes it clear why the enhanced pressure result in Ref. [90] satisfies momentum and boundary-condition requirements when treated in the sinusoidal steady state where the periodic plasmonic cavity is fixed in space. As described earlier in this section, the experimental situation would relate to some release point in time, and motion dictated by both the incident and internal fields. The situation in Ref. [90] is incorrectly portrayed in Ref. [91].

Consider now some broader conclusions in relation to the possibility of enhanced optical pressure (beyond $2\langle S_i \rangle/c$). The theoretical development of the force density boundary condition (the key contribution here) in Sec. III, with subsequent analysis (Sec. IV), proves that pressure enhancement is possible. It would seem prudent to emphasize force density in a path forward and not rely on inferences from a free space stress tensor (momentum flow) description in order to understand how material moves in response to electromagnetic fields. This is important when describing experimental situations involving laser illumination. Of course, formally, the purpose of a mathematical model is to describe experimental observations. Conveniently, the standard description of momentum flow into a volume can be adapted [65].

V. CONCLUSIONS

This work probes the fundamental way in which light interacts with condensed matter and transfers momentum from the photon to the material, as exhibited in the electromagnetic force density applicable in physical materials and at optical frequencies. A boundary condition relevant for the force density in materials and inhomogeneous condensed matter that provides a basis for the unique interpretation of total force is derived and motivated to be interpreted in the Cauchy principal value sense, using an example force theory that presents a representative field interface situation [(32) and (33)]. The physical argument relates to neutral materials and to the transport of free charge during the period of the electromagnetic wave to provide an interface screening charge. How far the frequency range for the application of (32) and (33) extends into the terahertz and microwave domain is both temperature and material dependent, but certainly for statics and even cryogenic temperatures, the interface surface charge densities should be considered in force calculations. New experiments are needed that allow extraction of force (density) information near to the surface in solid-state materials, and these will need to be carefully designed. With this new experimental information, it should be possible to not only better understand optical forces at nanometer length scales, but also arrive at a force theory that can be used with generality to consider systems with material and spatial dispersion. Such a model could then be broadly applied in the physical sciences and for the development of related technologies.

The momentum of a photon in vacuum is understood to be $\hbar k_0$, as can be readily found from energy principles and based on classical electromagnetics. Application of (32) enforces momentum flow across boundaries (mathematical and physical), and the boundary conditions in (25) and (26) do this for the general interface situation while allowing for the accepted vacuum photon momentum. As the optical pulse enters the material from vacuum, the theory developed here describes the boundary condition that allows momentum flow, Poynting vector, and field boundary conditions to be simultaneously satisfied. It is interesting that measurement of the photon momentum requires interaction with a sensor of some kind, and likely this involves a flux of photons entering a material that can be described by locally homogenized electromagnetic constitutive parameters (such as the spatially dependent dielectric constant). This means that the vacuum-matter interface is an essential part of sensing. Also, all macroscopic electromagnetic force densities utilize homogenized material systems. Thus, regardless of the description used, the principles developed here should apply, where the force density boundary condition is shown to be an essential part of the correct description of interfaces. This work therefore impacts the mechanical attributes of various photonic applications.

A point of note in the treatment of optical forces in condensed matter is the implication of the model in relation to motion. Frequency domain models describing sinusoidal steady state preclude motion, i.e., the system must be spatially fixed over the infinite time period. Motion is then at some initiation time and with the appropriate initial conditions that exist. In the laboratory, the complex and coupled electromagnetic and mechanical problem is greatly simplified through the assumption that measurable motion of macroscopic objects (like membranes) cannot occur during the optical period, nor during even low quality factor cavity excitation times (that exist in nanophotonic systems like plasmonic cavities). There are thus coupled force systems to consider in regard to laboratory settings, where, for example, one may have a mounted membrane being deflected by a laser beam with local material tension and a support frame on a stage on an optical bench on which a laser is mounted, i.e., the electromagnetic system is not closed. These situations have been treated here. Finally, one aspect of optomechanics that may not be widely appreciated is that the material and the structure both provide nonlocal-in-time contributions to the optical force density, meaning that the temporal history dictates the mechanical response at any given time.

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APPENDIX

The development of the photon momentum, a quantum phenomenon [92], is reviewed for a free-space plane wave. This follows a classical field description [93].

From (22), Poynting's theorem, and in vacuum, gives

$$\frac{\partial u_0}{\partial t} = \epsilon_0 \mathbf{E} \cdot \frac{\partial \mathbf{E}}{\partial t} + \mu_0 \mathbf{H} \cdot \frac{\partial \mathbf{H}}{\partial t}.$$
 (A1)

Ignoring the spatial dependence and for a monochromatic plane wave, we set $\mathbf{E} = \hat{\mathbf{e}} E_0 \cos(\omega t)$ and $\mathbf{H} = \hat{\mathbf{h}}(E_0/\eta_0) \cos(\omega t)$, where the free space wave impedance is $\eta_0 = \sqrt{\mu_0/\epsilon_0}$, and the energy density can be written from (A1) as

$$u_0(t) = \frac{1}{2}\epsilon_0 E^2 + \frac{1}{2}\mu_0 H^2 = \epsilon_0 E^2.$$
 (A2)

The free space momentum density is

$$\mathbf{g}_0 = \frac{1}{c^2} \mathbf{E} \times \mathbf{H} = \hat{\mathbf{e}} \times \hat{\mathbf{h}} \frac{\epsilon_0 E^2}{c}.$$
 (A3)

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From the instantaneous momentum density in (A3), we write the peak value as

$$g_{0p} = \frac{\epsilon_0 E_0^2}{c} = \frac{N\hbar\omega}{c} = N\hbar k_0, \qquad (A4)$$

with N photons per unit volume. This gives the vacuum photon momentum as

$$p_0 = \hbar k_0, \tag{A5}$$

in accordance with quantum theory [92].

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