Coalescence of Phenomenological Laser Damage, Materials Properties, and Laser Intensity: Moving Toward Quantitative Relationships

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Abstract

Many of the investigations of contamination related laser damage to date have been largely phenomenological. These papers can provide some direction in the quest for determining and maintaining adequate laser cleanliness levels. Purely optical analyses of laser contamination effects likewise provide direction. Neither the phenomenological nor the purely classical optical methods provide a solid basis for the determination of a safe or acceptable contamination level. This is borne out by the present state of the art. Investigation of the purely physical, optical, or chemical properties of the contamination is insufficient and and incomplete. It is necessary to look at the combined physical, chemical, and optical properties of the laser system, which includes the contamination. Determination of some key properties of the laser/contaminant system and their interactions has been carried out and the results are promising.

Introduction

To adequately study the issue of contamination effects of laser damage, it is requisite to coalesce knowledge from at least the following areas of study in a unified approach:

> Experimental laser damage studies, Laser damage theory, Laser physics, Classical Electromagnetic theory, Quantum Electromagnetic theory, Physical Chemistry, Photochemistry, Chemical Physics, Inorganic Chemistry, Organic Chemistry, Glass Chemistry, Surface Chemistry, Surface Physics, Contamination engineering, Materials engineering, Thermal engineering, Polymer science.

This is a difficult process as the terminology and in many cases the units vary between the various fields. In addition, it is not uncommon for the different groups to make different standard assumptions. It is also not uncommon for the base assumptions to be forgotten, and that the critical behavior of interest may negate the assumptions of the theory and thus the applicability of the theory.

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Treatments of Laser Damage Behavior

Classical Modeling

In classical optics, linear behavior is assumed, making the assumption that ray tracing and linear behavior is all that matters. This is inadequate for application to lasers as neither the unary nature of ray tracing nor the linear optical behavior is consistent with laser operation. Classical chemical thermodynamics is inadequate because in the surface interactions, the number of molecules and the distribution of the energy states violate the assumptions of chemical thermodynamics.

The typical homogeneous material continuum Maxwellian treatment of laser optical damage violates a number of assumptions of Maxwell's assumptions. First, Maxwell assumed that surface interactions were insignificant. It is well known that surface behavior due to surface fields is significant in laser damage phenomena. Second, Maxwell assumed that the material could be treated as a constant wavefunction or a constant tensorial function. In the case of a laser damage scenario, the laser optic is not damaged prior to the damage event, and it is damaged after the event, further the damage event results in a material that is more sensitive to damage than the initial part. Third, it is assumed that the bulk material is homogeneous in its interactions with the laser beam. It is well known that inhomogeneities of a variety of types result in significantly decreased laser damage thresholds. Maxwell in fact points out that in the case of crystalline defects that the homogeneity assumption may be invalid.[1] Fourth, in the case of contamination related laser damage, Maxwell did not deal with molecules and atoms, and thus is inadequate for the description of systems consisting of different types of molecules.

It is apparent that the common models of laser optical systems, and laser optical damage are by violation of their fundamental assumptions are therefore inadequate to describe laser damage. These models cannot describe laser optical damage because the principle assumptions of the models are violated and the models are thus negated. In all the previously mentioned cases it is apparent that a number of limiting assumptions are made that exclude their use in laser optical damage modeling. Thus, it is apparent that one needs to eliminate many of the restrictions to the models.

Improved Theoretical Modeling of Laser Optical Damage

The next step in modeling of laser optical damage using existing proven modeling systems is to jump to a significantly less limited model, that addresses both the energetics and the material behavior in the system. Schrödinger theorized that it is possible to describe the energetics, composition, chemical reactivity and configuration of a system in the form of a quantized wavefunction.

Schrödinger stated that if a properly described wave function describes a system, then it will describe the system. Although this is a cyclic definition, if a model adequately describes a system, it describes the system, QED. The catch with this approach is that it is

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[1] J. C. Maxwell, Philos. Trans. R Soc London, 155, 459-512, 1865.

not possible to solve a wavefunction in closed form for any system beyond the hydrogen atom. As a result, there have been a number of approximations made that allow the solution of approximate models to allow the full utility of the technique. These include the Born-Oppenheimer approximation, which states that the interaction of the nucleus with the electrons in the system does not result in significant changes in the positions of the nuclei. This approximation typically works well. The LCAO approximation stating that the total system can be approximated as a linear combination of atomic orbitals. The Hartree-Fock approximation, assumes that the net interactions of all electrons in a molecule can be approximated by treating the electrons individually, interacting with an average electron distribution. This results in a slightly higher energy than the real system where the individual electrons will interact with the remaining electrons on an individual basis. This is known to give reasonably accurate results for absolute energy values and more consistent values for differential values.

The main issue with the application of molecular and electronic modeling of laser optical systems is the computational intensiveness of the systems. While it is reasonably simple to describe the system as a series of wavefunctions in linear combination, the solution of the system of equations is not feasible at this time nor in the forseeable future. This does not mean that the technique is not viable. By selecting small subsets of atoms and molecules to model the interactions present in the system, it is possible to gain great insight into the interactions of light, surfaces, and molecules within laser optical systems.

Insight Gained from Molecular Modeling

The process of modeling the molecular interactions with laser intensity fields, has been carried out for many years, mostly on isolated molecules. This modeling is based upon the rudimentary interactions of the electronic wave functions with oscillating electromagnetic fields. In the case of a full electron model, the interactions are well modeled.

At a fundamental level the interaction of laser light with a dielectric or a molecule is defined by the interaction of the monopole and multipole moments both permanent and induced, of the matter with the field gradients[2]. These interactions fully account for the time dependent interactions of the oscillating electromagnetic fields of the molecular orbitals, the inherent fields within the molecule and the external fields present at the point, all in a time dependent manner.

The interaction of the poles with the field gradients perturbs both the matter and the internal and external field gradients. This is much more representative of the net effect of the immediate electric fields within the system than the assumption that the field is an average of the whole system. In the case of a purely linear system, the average of the interactions is a reasonable approximation for low intensity electromagnetic radiation. In the case of interaction of a system within a non-linear regime due to localized internal stresses, the average of the interactions is a very poor representation. In a largely anisotropic system, the presence of point defects or dislocations in the medium will result

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[2] A. D. Buckingham, Phil. Trans. of the Royal Society A, (293) 1402, 239-247, 1979.

in large internal fields, often exceeding 10^7 volts/ meter. This field intensity will distort the molecular orbitals. This distortion results in the induction of large anisotropies in the molecular orbitals within the structure of the material. The induced asymmetry in the immediate region of the defect will result in the non-linear optical properties being greatly amplified. The internal field will induce a radially decaying induced polarization gradient in the surrounding material. The incidence of a laser beam will increase the distortion at the distorted orbitals, even within the decaying region. The laser field will further distort the molecular polarization field, increasing the distortion across a wider area with a larger peak distortion. This results in the observed exponential like growth in the size of laser optic damage propagation seen in lasers having pulse lengths significantly greater than the vibrational periods of the atoms within the system.

Gradients that result in the loss of symmetry and isotropic behavior can be internal or external gradients. Internal gradients that results in the distortion of the electron densities include stress, thermal, compositional, morphological, crystallographic and excitational gradients. The external gradients in laser systems that result in electronic distribution distortions are electric and magnetic distortions. These effects are all time dependent and all interact.

It must be remembered that the symmetry that results in the degeneracy of molecular orbitals is due to the electron distributions. These electron distributions and densities largely control the physical interactions of the molecules. Thus, physical forces will also distort the electronic distributions. The higher order polarizabilities of materials are more greatly affected by the imposition of force on a molecule. The dipole moment and its effect upon the properties of matter is significantly less affected by the immediate fields surrounding a molecule than are the quadrupole and octupole moments, this is largely due the loss of symmetry, and its affect on two or more, rather than just one axis.

As these interactions are of the multipole type, due to the charge distribution non-degeneracy, caused by electronic asymmetry generated from the loss of symmetry, the effects grow quite rapidly. Further, there is no inherent restriction of size based upon the wavelength of the radiation interacting with the material. Thus, a point defect in a crystal caused by the impingement of a high-energy ion can result in the generation of a latent damage initiation site, if the site does not decay back to its original condition. Further, frozen in stresses as occurs in glasses and crystals can initiate the enhancement of non-linear behavior in materials.

It should be noted that the gradient induced asymmetry due to the internal field distortion interacts with the external field of the laser increasing the distortion and asymmetry. This will propagate at a speed approaching the speed of light. This distortion of the initial site induces distortion of the surrounding sites, and the distortion of the surrounding sites induces distortion in the sites surrounding them, including the original site. This results in the near exponential growth of damage seen in laser damage events. This propagates in a time dependent manner.

Conclusion:

Virtually all laser optical damage can be described based upon the rudimentary time dependent interactions of the monopole and multipole interactions of the matter in the optic with the laser photons. Much of the existing laser damage theory presently in use cannot describe the behavior of matter within the laser beam beyond a very limited regime due to the violation of one or more of the fundamental assumptions of the base theory upon which the overall theory is based.

The interactions of the inhomogeneities within systems with the high intensity laser beam results in a perturbation of the non-linear optical properties of the material that exceeds the level that could reasonably be approximated by the initial materials properties or the average properties of the material as a whole. As an example the adsorption of a molecule on a silver sol particle will result in the surface enhanced Raman effect, which results in a change in the Raman susceptibility of about fourteen orders of magnitude, as measured and as calculated. [3,4]

The perturbation of a total internal reflection Nd:YAG slab by the thermal and excitational gradients, due to the point focusing of the pump laser beam, resulted in self focusing damage of the optic at levels significantly below the threshold for small scale self focusing, based upon existing theory. The laser damage community, including the recognized experts in damage occurring in total internal reflection systems, has considered the damage mechanism indescribable. The damage sites resulting from this damage were 50 nanometers in diameter as measured by scanning probe microscopy. The damage probability was determined to be directly related to the presence of a circulating beam within the cavity and the location of the point foci of the laser diode images. This interaction was tracked by changing the pump lens position and firing ten million laser shots per data point, including reproduction of key data points. The induced gradients in the laser slab result in asymmetry of the internal fields, greatly reducing the required external field required to induce small scale self-focusing. Thus, the probability of small scale self-focusing increased significantly at the existing intensity levels within the laser, resulting in damage.

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[4] H. Nakai and H. Nakatsuji, J. Chem. Phys. 103 (6) 2286-2294, 1995.

Thus, based upon a reasonable selection of a model of the state of the matter within a laser field, the interactions of laser beam intensity and materials behavior can be treated in a micro-canonical manner. This will provide a representative computed value for the optical properties of the combined system.

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