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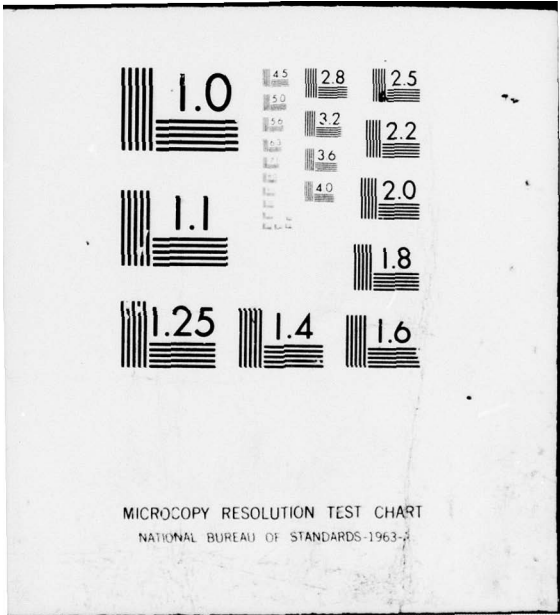
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THEORY OF THE INTERACTION OF
INFRARED RADIATION WITH SOLIDS

M. Sparks, Principal Investigator, 213/451-9916
Xonics, Incorporated
Santa Monica, California 90401

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30 November 1978

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20. Abstract

in which photons are absorbed simultaneously with processes which allow momentum conservation will be considered. Including intraband processes (valence band to final conduction band plus intraconduction band, for example) as well as the usual interband process in two-photon absorption-coefficient calculations gave good agreement (factor of 0.5 to 4) with Harvard experimental results without adjusting parameter values. Calculations showed that including a linear temperature dependence of the optical absorptance of metallic or dielectric reflectors gave much higher temperatures and damage susceptibilities in general than for the case of temperature-independent absorptance and gave thermal runaway in several interesting cases. Such simple considerations as continuity of the electric field and its slope were exploited to extract information and understanding of multilayer-dielectric reflectors, including prediction and explanation of asymmetric spectral reflectance resulting from absorption in the layers.

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a. ABSTRACT

A transport equation for electron-avalanche breakdown, including large- and small-quantum effects, was developed, checked by two derivations, compared with previous results, solved by three methods (numerical solution of partial-differential equation, casting into eigenvalue differential equation form, approximate intuitive solution), applied to simple models and to cases of practical interest, and compared with gas-breakdown results. Large-quantum processes, including interband transitions and various processes in which photons are absorbed simultaneously with processes which allow momentum conservation will be considered. Including intraband processes (valence band to final conduction band plus intraconduction band, for example) as well as the usual interband processes in two-photon absorption-coefficient calculations gave good agreement (factor of 0.5 to 4) with Harvard experimental results without adjusting parameter values. Calculations showed that including a linear temperature dependence of the optical absorptance of metallic or dielectric reflectors gave much higher temperatures and damage susceptibilities in general than for the case of temperature-independent absorptance and gave thermal runaway in several interesting cases. Such simple considerations as continuity of the electric field and its slope were exploited to extract information and understanding of multilayer-dielectric reflectors, including prediction and explanation of asymmetric spectral reflectance resulting from absorption in the layers.

b

b. STATEMENT OF WORK

The Contractor shall furnish scientific effort together with all necessary related services, facilities, supplies, and materials, to conduct the following research:

- A) Investigate the theory of dielectric breakdown by intense electric fields.
- B) Formulate a transport theory of electron-avalanche breakdown that includes the pertinent physical processes occurring in the dielectric breakdown of insulators, semiconductors and infrared transparent optical materials.
- C) Solve the transport equation to obtain the magnitude, frequency dependence, pulse-duration dependence, and variation from material to material of the breakdown irradiance, as well as the damage morphology, breakdown statistics, and the relative time in the pulse at which breakdown occurs.
- D) Relate and compare the above transport theory to experimental results and with other breakdown processes in optical materials subjected to intense infrared radiation.

c. STATUS OF THE RESEARCH EFFORT

I. SHORT SUMMARY OF ELECTRON-AVALANCHE BREAKDOWN RESULTS

A transport equation was developed intuitively and verified by two derivations. Both large-quantum processes, which were treated as differences, and small-quantum processes, which were approximated by differentials, were included. Previous equations of Holstein, Soviet researchers, and Holway and Fradin were derived from the transport equation. Writing the transport equation in the explicit form of a diffusion equation and interpreting the appropriate coefficients as generalized velocities and diffusion coefficients (in energy space rather than, say, temperature space) afforded useful intuitive understanding of results and gave two time constants that were quite useful in estimating how fast the quasi-steady-state was approached.

The small-quantum processes were considered first. Simple model problems were solved in order to establish that the methods used gave the correct exact results (where they could be obtained) and gave results that were verified by comparison of several solutions. Three types of solutions were obtained. (1) Numerical solutions of the complete partial-differential equation were obtained. (2) Consideration of the time constants mentioned above suggested that a quasi-steady-state was approached in a time short with respect to laser-pulse durations of current interest. Thus, time was eliminated from the partial differential equation. The resulting differential equation was in the form of a non-Hermitian eigenvalue problem, with the electron multiplication rate as the eigenvalue. A sufficiently simple model was found to admit an exact solution, which was quite useful in evaluating approximation methods. Comparison of approximate and exact results allowed detailed studies of

approximate methods of solution. Numerical solution of the eigenvalue equation is the most powerful tool for obtaining the electron multiplication rate. (3) An intuitive approximate quasi-steady-state solution was developed and used in conjunction with the other methods.

The full problem with all previously considered small-quantum processes included is now being solved. The results of this first phase of the investigation can be completed and the results published soon after an anticipated continuation of the program is underway. Establishing contact with previous results at this intermediate stage is important.

With this background established, the more exciting ideas will be pursued in the continuation of the program. Large-quantum processes involving interband transitions or absorption of photons in processes including events which allow momentum conservation will be included in the transport-equation solutions. Preliminary results suggest that several different physical processes may be involved in electron-avalanche breakdown in which wide ranges of experimental variables are involved, as the estimates of Sparks suggested several years ago.

A study of the closely related problem of electron-avalanche breakdown in gasses, which was carried out early in the program, was useful in formulating the present study of breakdown in solids. The gas-breakdown problem itself is of great current interest in such very-high-power lasers as cylindrical lasers and in such solid-breakdown problems as the shielding and coupling of laser radiation into solids by plasmas formed at their surfaces. A recent re-investigation of the gas-breakdown problem revealed that it may be profitable to apply the techniques and processes considered in the present study of solids to the gas-breakdown problem.

II. SHORT SUMMARY OF MULTIPHOTON-ABSORPTION CALCULATIONS

Multiphoton-absorption coefficients in solids have not been calculated correctly in the past. Most perturbation-theory calculations included interband processes (intermediate state in a conduction band above the lowest conduction band, for example), but overlooked interband processes (intermediate state in the lowest conduction band, differing from the final state by a small wave vector shift, for example). Though not stated by Keldysh and not widely appreciated, the multiphoton limit of Keldysh's well known result corresponds to intraband processes.

Values of two-photon absorption coefficients of alkali halides were calculated with both interband and intraband processes included, with intermediate states in the valence and conduction bands included, and with interference considered. No parameters were adjusted. The common procedure of avoiding numerical evaluation of sums in k space by formally neglecting the angle-dependence of the terms in the sums was used to obtain a simple closed-form expression for the two-photon absorption coefficient. The average values of the interband matrix elements (typical oscillator strengths $f \approx 1/4$) were obtained by comparing the theoretical one-photon absorption coefficients with experimental values. The velocity expression was used for the intraband matrix elements. Simple consideration of interference effects at high symmetry points in the Brillouin zone suggested that these effects were smaller than a one-dimensional analysis would predict. The resulting agreement with recent experimental values of Lui and co-workers at Harvard was very good — factors of 0.5 to 4. Greater accuracy is not expected to be attained from first-principles theories in the near future since current band-theories do not give accurate values of the matrix elements.

III. SHORT SUMMARY OF SIMPLIFIED DESCRIPTION OF DIELECTRIC REFLECTORS

The spectral-absorptance curves of quarter-wave reflectors are quite asymmetric in general as a result of absorption in the layers. The maximum electric field E in an infinite stack is in the center of the high-index (low-index) layer at the low-frequency (high-frequency) end of the high-reflection band. Thus, the absorptance minimum of a stack having much greater absorption in the high-index material occurs at $\omega > \omega_c$ (band center). Operation at $\omega > \omega_c$ could increase the damage resistance if the high-index material is easily damaged. At ω_c , E decays rapidly. As ω departs from ω_c in an infinite reflector, E penetrates deeper into the coating, with no decay at the band edges. The peak-to-peak distance in an infinite dielectric stack is constant for all frequencies in the band. By using the continuity of E and dE/dz , E can be obtained from $E = 0$ at the metallic substrate or, in an infinite stack, from $E = 0$ at the reflector surface for $\omega = \omega_c$ or $E = 0$ in the center of the high- or low-index layer at the band edges. Simple closed-form approximations for the spectral absorptance and the phase of E are accurate and sufficient for present applications.

IV. SHORT SUMMARY OF HIGH-POWER ULTRAVIOLET MATERIALS AND COMPONENTS

Windows have low theoretical failure thresholds — typically $It_p = 0.2 \text{ J/cm}^2/\text{pulse}$ for thermally induced optical distortion in currently envisioned systems ($t_p = 1 \mu\text{s}$, 10^3 pulses/s, 60 s operation, 250-350 nm, window thickness = 1 cm, $\beta_{\text{window}} = 10^{-4} \text{ cm}^{-1}$, $A_{\text{refl}} = 10^{-3}$, and $\lambda/40$ phase distortion). Dielectric reflectors have greater theoretical thresholds, $It_p \approx 10\text{-}100 \text{ J/cm}^2/\text{pulse}$ for thermally induced optical distortion, which may be approached in well designed reflectors. State-of-the-art experimental values are: 47 J/cm^2 for reflector damage, $A_{\text{refl}} = 2 \times 10^{-3}$, $A_{\text{metal uv}} \approx 0.08$, $\beta_{2 \text{ photon}} = 0.045\text{-}7.3 \text{ cm/GW}$, $\beta_{\text{window}} < 10^{-4} \text{ cm}^{-1}$, and heat-transfer coefficient $h = 10 \text{ W/cm}^2 \text{ K}$. If the absorptance is a strongly increasing function of temperature (few percent for 10^3 K rise), reflector failure thresholds will be lowered considerably, and there will be a thermal runaway even in cooled reflectors and in spherical heat diffusion. Two-photon absorption is tolerable in coatings, but should be avoided in windows (by making the absorption edge $> 2\hbar\omega$) if fast high-power adaptive optics are developed and used. Recent two-photon calculations with interband and intraband processes included agree with recent experimental values for alkali halides to within factors of 0.5 to 4.0. Isolated-spot damage may result from absorbing regions (inclusions, surface contamination, microscopic absorbers, etc.) that are either poor thermal conductors or are thermally isolated, either physically or by a short pulse duration. Predicted asymmetric spectral absorptance curves are explained simply by considering the electric-field distributions.

V. SHORT SUMMARY OF TEMPERATURE DEPENDENCE OF ABSORPTANCE
IN LASER DAMAGE OF METALLIC MIRRORS: I - MELTING

Naval Weapons Center damage thresholds for ultraclean metals (45 J/cm^2 , 100 ns, $10.6 \mu\text{m}$, Cu) are a milestone in laser-damage studies, being reproducible, with no isolated-spot damage, and showing the first excellent agreement (~ 15 percent) with a first-principles theory (developed here) for any metal or transparent material. An exact premelting temperature result for the case of optical absorptance $A = A_i + A_1 T_s$ indicates that the surface-temperature rise T_s reduces the theoretical threshold $(It_p)_m$ for raising T_s to the melting temperature T_m by a factor of 3.6 to 1.6, typically. Raising T_s to T_m is a good damage criterion for 100 ns pulses, but not for single subnanosecond pulses. The previous scaling $It_p \sim t_p^{1/2}/A$ is invalid in general, but $It_p \sim t_p^{1/2}$ is valid for $(It_p)_m$ even with $A = A_i + A_1 T_s$. Approximating a Gaussian $I(t)$ pulse by a square pulse makes $(It_p)_m$ 17 percent too small in the constant-A approximation.

VI. SHORT SUMMARY OF TEMPERATURE DEPENDENCE OF ABSORPTANCE IN LASER DAMAGE OF METALLIC MIRRORS: II - VAPORIZATION AND HEATING THE VAPOR

For conventional metallic samples, agreement between experimental damage thresholds and three theoretical thresholds — for melting m , vaporizing v , and heating to $2eV$ — is fair (factor of ~ 2 to ~ 10). The previous scaling $It_p \sim t_p^{1/2}/A$ is invalid in general, but $It \sim t_p^{1/2}$ is valid for $(It_p)_m$, $(It_p)_v$, and $(It_p)_{2eV}$ individually. Silver has the greatest theoretical threshold and figure of merit at $1.06 \mu m$, but silver, copper, and gold have approximately equal figures of merit at $10.6 \mu m$. Some clustering mechanism probably is involved in the isolated-spot damage observed in conventional samples.

d. LIST OF PUBLICATIONS AND REPORTS

- M. Sparks, P. N. Sen, D. L. Mills, T. Holstein, A. A. Maradudin, and E. Loh, Jr., "Electron Avalanche Breakdown Theory: Small-Quantum Limit." In progress, plan to submit to Physical Review.
- M. Sparks, "Multiphoton-Absorption-Coefficient Calculations." In progress, plan to submit to the Journal of the Optical Society of America.
- M. Sparks, "Nonlinear Absorption in Optical Materials," Chapter V in Physics of Transmissive Optical Materials, edited by Marvin Hass. Manuscript submitted to editor.
- M. Sparks and M. Flannery, "Simplified Description of Dielectric Reflectors." Presented at the Boulder Laser Damage Symposium. Manuscript mailed to editor.
- M. Sparks and M. Flannery, "Simplified Description of Dielectric Reflectors." Manuscript submitted to the Journal of the Optical Society of America.
- M. Sparks, "High-Power Ultraviolet Materials and Components." Paper presented at the Boulder Laser Damage Symposium. Manuscript mailed to editor.
- M. Sparks and E. Loh, Jr., "Temperature Dependence of Absorptance in Laser Damage of Metallic Mirrors: I - Melting; II - Vaporization and Heating the Vapor." Manuscript submitted to the Journal of the Optical Society of America.
- M. Sparks, "Reflector Damage by CW and Repetitively Pulsed Lasers." In progress, plan to submit to the Journal of the Optical Society of America.
- M. Sparks, E. Loh, Jr., and G. T. Johnston, "Failure of Reflectors with Temperature Dependent Absorptance." Manuscript sent to the editors of the proceedings for the High-Power Laser Optical Components Conference.
- P. N. Sen, Soviet Literature on Electron Avalanche Breakdown, working note SM-WN-791, prepared for internal use to bridge the gap in writing styles of Soviets and U. S. investigators.
- M. Sparks, Informal Semiannual Technical Report on AFOSR Contract No. F49620-78-C-0042 entitled "Theory of the Interaction of Infrared Radiation with Matter," September 1978.

e. LIST OF PROFESSIONAL PERSONNEL

Dr. Marshall Sparks, principal investigator

Dr. P. N. Sen, research scientist

Dr. D. Mills, University of California at Irvine, consultant

Dr. A. Maradudin, University of California at Irvine, consultant

Dr. T. Holstein, University of California at Los Angeles, consultant

Mr. E. Loh, Jr., student at the California Institute of Technology,
research associate

Mrs. Fumiko Tajima, student at the University of California at
Los Angeles, research associate

f. INTERACTIONS

On 26 May 1978 in a telephone conversation with Dr. Santa Mayo of the National Bureau of Standards, M. Sparks answered questions and gave suggestions about problems concerning 2-3 μm turnable lasers, OH molecules in SiO_2 , and related topics. On 3 April 1978 in a telephone conversation with Dr. Edward Borsare of W. J. Schafer Associates, M. Sparks provided information on diamond windows for the free-electron laser. The information was for use in his Department of Defense program. On 24 April 1978 in a telephone conversation with Dr. Marvin Klein of the Hughes Research Laboratory, M. Sparks provided information, suggested materials, and suggested contacts for obtaining materials for fiber optics to operate at wavelengths longer than 10 μm .

M. Sparks presented an invited paper and a contributed paper, listed in Sec. d, at the Boulder Laser Damage Symposium. He attended the Denver High-Power Laser Optical Components Conference and prepared a paper to be included in the proceedings. M. Sparks presented lectures at the lecture series of the Optical Coating Laboratory. He visited the Air Force Weapons Laboratory on 6 July 1978. He gave a talk on Laser Damage of Materials by Electron Avalanche Breakdown at the University of Southern California on 27 October 1978 a talk on Optical Problems of High Power Laser Systems for the Special Seminar Series at the Aerojet Electro Systems Company on 28 September 1978 and an invited presentation "Recent Theoretical Developments in High-Power Optics" at BK Dynamics, Inc. on 8 and 9 November 1978. He visited the Lockheed Palo Alto Research Laboratory on 2 and 3 November 1978.

The Informal Semiannual Technical Report dated 30 September 1978 was sent to P. F. Braunlich, M. Braunstein, S. M. Copley, A. A. Maradudin, and P. A. Miles on 4 October 1978.

Numerous informal technical interactions include discussions with: Dr. Joseph Apfel, Optical Coating Laboratory, Dr. Michael Bass, University of Southern California, Dr. H. E. Bennett, Naval Weapons Center, Dr. E. Bernal G., Honeywell, Inc., Dr. J. R. Bettis, Air Force, currently at the Naval Academy, Dr. Peter Braunlich, Bendix Research Laboratories, Mr. Morris Braunstein, Hughes Research Laboratories, Dr. Paul Kelly, Physics Division, NRC, Dr. Conrad Phillippi, Air Force Materials Laboratory, Dr. P. A. Miles, Raytheon Company, and Major James Stapp, Air Force Weapons Laboratory.

g. INVENTIONS AND PATENT DISCLOSURES

There were no inventions or patent disclosures during the period of the contract.