Comment on "Thermal Response of Metals to Ultrashort-Pulse Laser Excitation"

Recently, Corkum, Brunel, Sherman, and Srinivasan-Rao[1] (CBSS) reported on a method to determine the electron-phonon coupling constant g in Eqs. (1) and (2) of Ref. 1, in metals, based on a comparison of the experimental damage threshold for different laser pulse widths with a heat-transport model. The damage threshold was equated with the surface-melting threshold and used as an input to the model to solve for g.

Using this approach, CBSS concluded that, for copper, g has a value 10 times smaller than an estimate based on our femtosecond time-resolved experiments[2] and 20 times smaller than that calculated in Ref. 3. CBSS state that inelastic-scattering rates by impurities and surfaces in our samples far exceed the electron-phonon scattering rate. To avoid these stated sample problems, they used a standard copper laser mirror as their sample.

With regard to sample quality, while defects and surface scattering can somewhat contribute to the electron relaxation process, such a contribution can become dominant only at very low temperatures.[4] This is not the case in our samples (~200-Å evaporated copper films with an average grain size of a few hundred Å) since they were initially at room temperature. While no details were given on surface preparation of the laser mirrors used by CBSS as samples, they are typically produced by mechanical polishing or single-point diamond turning of polycrystalline material. Mechanical polishing of copper produces an amorphous surface layer with a typical grain size of ~10 Å up to a depth of 100 Å and $\lesssim 30$ Å up to a depth of 2000 Å.[5] Diamond turning also introduces further defects to the original polycrystalline surface. Therefore, the argument regarding the much lower level of defects in their samples compared to ours is unproven. Moreover, the statement regarding the mirror surface flatness is irrelevant since what matters in electron-surface scattering is smoothness in the order of the wavelength to the conduction electrons (i.e., a few to several Å). A high-quality copper laser mirror would typically have surface flatness of $\lambda/20$ at 10.6 μm and rms surface roughness of ~50 Å. Another point regarding sample quality is the problem of oxide formation on the surface of the copper mirror, used as a sample, which could affect its damage threshold.

Aside from sample quality, CBSS equate the threshold for multishot damage to that of surface melting. While it is generally assumed that the single-shot damage threshold for the relatively longer pulses (~100 ns) is compatible to the surface-melting threshold[6] for the shorter pulses, it is possible that there are many other mechanisms of surface damage with thresholds different from those of melting. These include vaporization, slip-band formation, ripple formation, pitting, and cratering.[6] Therefore, equating the surface-damage threshold to that of melting for the shorter pulses used by CBSS (2.5 and 50 ps) is unjustified. Also, it is well demonstrated that the multishot damage threshold could be different from that of a single shot.[7,8] Thus, even for the longer pulses, equating the multishot damage threshold to that of melting is also unjustified. Moreover, the four data points of Ref. 7 that are reproduced in Fig. 1(a) of CBSS were for single-shot damage of the mirror surface when irradiated with $\lambda = 10.6$ μm rather than multishot damage at $\lambda = 9.3$ μm as for data reported by CBSS in the same figure. Further complications arise in defining the criteria of surface damage. CBSS assessed damage by observing a change in reflectivity, observing visible sparks on the surface, or using ~1-μm resolution optical microscopy. It is not clear if their observed damage threshold was independent of the damage criterion. It would seem likely that the threshold for observing surface damage with 1-μm resolution is lower than the threshold for observing sparks on the mirror surface.

In view of these comments, the conclusions of CBSS regarding the value of g are questionable.

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[1] P. B. Corkum, F. Brunel, N. K. Sherman, and T. Srinivasan-Rao, Phys. Rev. Lett. **61**, 2886 (1988).

[2] H. E. Elsayed-Ali, T. B. Norris, M. A. Pessot, and G. A. Mourou, Phys. Rev. Lett. **58**, 1212 (1987).

[3] P. B. Allen, Phys. Rev. Lett. **59**, 1460 (1987).

[4] M. L. Roukes, M. R. Freeman, R. S. Germain, R. C. Richardson, and M. B. Ketchen, Phys. Rev. Lett. **55**, 422 (1985).

[5] For example, Z. G. Pinsker, *Electron Diffraction* (Butterworths, London, 1953), Chap. 12.

[6] M. Sparks and E. Loh, Jr., J. Opt. Soc. Am. **69**, 847 (1979); **69**, 859 (1979).

[7] J. F. Figueira and S. J. Thomas, IEEE J. Quantum Electron. **18**, 1381 (1982).

[8] Y. Jee, M. F. Becker, and R. M. Walster, J. Opt. Soc. Am. B **5**, 648 (1988).

