

**Hansen and Echt Reply:** The authors of [1] argue that the time dependence of the electron yield can have a power-law behavior as observed, i.e., with a power less than 1, if only the energy distribution is sufficiently different from a flat one. In Fig. 1 of [1] the decay on the experimentally sampled time scale involves molecules that have energies at the *decreasing* edge of the energy distribution ( $E > 40$  eV); this produces a quasi-power-law decay with exponent less than 1. The discreteness of the photon energy can be ignored as also noted in the Comment.

The assumptions underlying Fig. 1 [1] are, however, contradicted by the experimental data. The exponent of the power law does not change when the laser fluence is increased from 36 to 94 mJ/cm<sup>2</sup> as evident from the data shown in Fig. 2(a) of Ref. [2], whereas the model proposed in [1] would predict a change in slope by more than a factor of 2. The fluence increase would cause the average number of absorbed photons to change from 10 to  $10 \times 94/36 = 26$  and shift the energy distribution so high that the decay would now involve molecules with energies at the *increasing* slope of the distribution, causing a decay which is even faster than  $1/t$  in sharp contradiction to the data. On the other hand, the yield of delayed electrons strongly increases with fluence [3], contradicting the suggestion [1] that the excitation energy may have reached an upper limit.

The model proposed in the preceding Comment fails for several reasons. The energy distribution is calculated with a Poisson distribution which assumes that the absorption cross section is constant. It is known, however, that the absorption cross section of C<sub>60</sub> is strongly dependent on the degree of excitation (see [4] for just one example). Furthermore, a constant laser fluence is assumed. In the experiment a *Q*-switched, multimode Nd:YAG laser is employed which is characterized by large temporal and spatial fluctuations of the power density within each laser pulse, resulting in much broader energy distributions.

Another important issue where we disagree with the authors of the preceding Comment concerns the frequency factors. In fact, we believe that the large discrepancies among the various experimental determinations of the C<sub>2</sub> dissociation energy  $D$  [5] mainly arise from the use of different frequency factors in the expressions being used for the rate constants. By contrast, our analytical procedure does not require assumptions about the detailed form of the rate constants. Although we do not agree with the estimate of the electron emission frequency factor in [1], a more serious error is committed when using a simple formula based on the Rice-Ramsberger-Kassel-Marcus (RRKM) theory with an incorrect transition state to estimate the C<sub>2</sub> loss frequency factor. An error in the frequency factor will translate into an error in the absolute energy scale when data such as in [6] are used to extract binding energies. The error depends on the relative value of the correct frequency factor and the one actually used. It can be estimated using the Weisskopf expression [7]. Alternatively, we can use

the frequency factor given in [6] for the so-called “orbiting transition state.” This is the one consistent with the measured kinetic energy release data [8]. Including also the rotational phase space of the C<sub>2</sub> multiplies this frequency factor by the rotational partition function which is 440 for a final temperature of 0.2 eV. Since the derived dissociation energy  $D$  scales with the value of  $\ln(\omega_a t)$ , where  $t$  is a typical measurement time and  $\omega_a$  the C<sub>2</sub> loss frequency factor, the resulting C<sub>2</sub> dissociation energy of C<sub>60</sub><sup>+</sup> can be estimated. The result is that  $D(\text{C}_{60}^+)$  is equal to  $9.20 + 7.06 \text{ eV} \times \ln(440)/25.6 = 10.9 \text{ eV}$ . Except for the rotational partition function, all data in this calculation are provided by Ref. [6]. Correcting for the 0.5 eV difference between ionic and neutral dissociation energies finally gives  $D(\text{C}_{60}) = 11.4 \text{ eV}$ .

A proper treatment of the evaporative process with the correct frequency factor, using experimental data like those reported by Märk and co-workers [6], is thus found to give a C<sub>2</sub> dissociation energy close to the one we derived from a completely different type of experiment and close to theoretical results which consistently give values at about 11 eV [9]. The agreement suggests that a critical revision of previously published experimental data pertaining to statistical dissociation of C<sub>60</sub> is warranted.

Klavs Hansen\*

Institute of Physics and Astronomy  
University of Aarhus  
DK-8000 Aarhus C, Denmark

Olof Echt†

Department of Physics  
University of New Hampshire  
Durham, New Hampshire 03824

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\*Electronic address: klh@dfi.aau.dk

†Electronic address: olof.echt@unh.edu

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