ADDITIONS AND CORRECTIONS

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J. J. H. Pijpers,* E. Hendry, M. T. W. Milder, R. Fanciulli, J. Savolainen, J. L. Herek, D. Vanmaekelbergh, S. Ruhman, D. Mocatta, D. Oron, A. Aharoni, U. Banin, and M. Bonn: Carrier Multiplication and Its Reduction by Photodoping in Colloidal InAs Quantum Dots

In our earlier paper,[1] we reported carrier multiplication (CM) in colloidal InAs quantum dots (QDs) and CM in dots with a pre-population of one exciton. The occurrence of CM in relaxed InAs QDs was concluded from the results of time-resolved TeraHertz (THz). Both THz and quasi-continuous wave (quasi-CW) experiments were performed to study the CM in pre-excited dots. Recent attempts to reproduce the observations of CM using THz spectroscopy on InAs based QDs of two sizes were unsuccessful. These attempts followed transient absorption measurements in Jerusalem by one of us (S. R.) and co-workers reported elsewhere,[2] which have not yielded evidence for CM in these QDs.

Time-resolved THz measurements were repeated for InAs/CdSe/ZnSe core/shell-1/shell-2, synthesized as reported elsewhere.[3] The investigated QDs had an InAs core of 4.4 nm diameter ($E_g = 1.1$ eV) onto which one atomic layer of CdSe and four layers of ZnSe were deposited. As in our original paper, the presence or absence of CM was investigated by comparing two excitation wavelengths above and below the theoretical onset for CM (for InAs: 2.05 times E_g).[4] CM is characterized by the presence of relatively short-lived biexcitons (lifetime tens of picoseconds),[1,4,5] which are created by the absorption of one photon. However, biexcitons are also readily created by sequential multiphoton absorption. Hence, the relative yield of bi- versus single excitons has to be determined for fluences where multiexciton generation by multiphoton absorption is negligible. To disentangle effects of multiphoton excitation versus CM, we use various excitation fluences. Single excitons are long-lived (∼100 ns) as compared to the time frame of our experiment.

For the 4.4 nm particles, the scaled signals for excitation wavelengths of 400 and 800 nm, corresponding to 2.74 and 1.35 times the gap are shown in Figure 1 (offset for clarity).

The data in Figure 1 correspond to 400 and 800 nm excitation fluences, which result in approximately the same average number of absorbed photons per particle (considering the optical density of the sample at the two wavelengths and the ratio of absorption cross sections σ at 400 and 800 nm, σ_{400} nm/σ_{800} nm $= 10.0$). It is apparent from the data that there is no significant bi-exciton decay visible at low fluence. This points directly to the absence of CM. A conservative estimate, considering both the fluence dependence of the 400 nm signal, and a comparison of the signals at 400 and 800 nm at roughly the same excitation densities, provides an upper limit for CM of 10%, well below the factor of 1.6 concluded previously,[1] and also lower than the factor of 1.2 concluded in ref 6 under similar conditions.

Summarizing, we could not reproduce our earlier results and the conclusions regarding the presence of CM. One or a combination of the following effects may explain these contradictory observations:

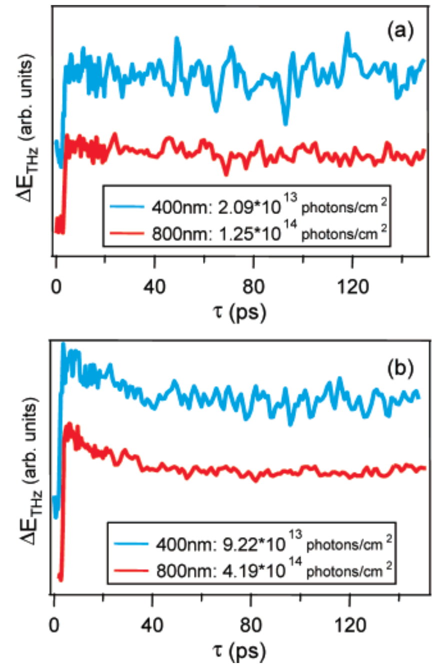

Figure 1. Decay dynamics of THz measurements on QDs with 4.4 nm core ($E_g = 1.1$ eV) at low (a) and high (b) excitation fluence.

- The QDs in previous and recent measurements were synthesized at different times. We cannot exclude the possibility that QDs from different synthesis batches have different properties, although this does not seem very likely.
- The demonstration of CM relies heavily on a highly accurate determination of the product of pump photon fluence and QD absorption cross section at various wavelengths. In our case, it was challenging to reliably determine the ratio of absorption cross sections (σ) at 400 and 800 nm excitation. In the earlier reported measurements,[1] we underestimated the value of σ_{400} nm resulting in bi-exciton generation by multiphoton absorption of pump photons at fluences where this was expected to be negligible.
- Spatial inhomogeneities in the excitation beam (resulting in local "hotspots"), may lead to multiphoton absorption, locally in the excitation beam. If this occurs, then bi-excitons may be formed in the sample, despite the fact that the average number of excitons remains low. The experimental results at 400 nm excitation presented in this correction have been measured with a so-called "diffuser" in the excitation beam.

The revised conclusion regarding CM has no effect on the other conclusions in the paper, pertaining to the twofold degeneracy of the lowest energy of the 1S electron level and its effect on THz and TA signals. Also, the reported values for bi-exciton recombination times, the bi-exciton binding energy, and tri-exciton energy shift remain valid.

References and Notes

(1) Pijpers, J. J. H.; Hendry, E.; Milder, M. T. W.; Fanciulli, R.; Savolainen, J.; Herek, J. L.; Vanmaekelbergh, D.; Ruhman, S.; Mocatta,

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