LETTERS TO THE EDITOR
The Letters to the Editor section is divided into three categories entitled Notes, Comments, and Errata. Letters to the Editor are limited to one and three-fourths journal pages as described in the Announcement in the 1 January 1999 issue.

NOTES

Cavity ring down spectroscopy on solid C$_{60}$

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Following the success of cavity ring down (CRD) spectroscopy for quantitative optical absorption studies on gas-phase species, various experimental schemes are currently being explored to make this technique applicable to the study of thin solid films as well. A straightforward way to proceed is to put a thin solid film on an optically transparent substrate inside the optical cavity such that the various (embedded) cavities are all optically stable; reflection losses from the substrate will not be noticed as overall losses from the cavity in this case and one can selectively measure the absorption or optical rotation in the solid. We used this approach to measure the polarization rotation at a fixed wavelength in this case and one can selectively measure the absorption or optical rotation in the solid. We used this approach to measure the absorption spectrum of a thin solid film via CRD spectroscopy by the measurement of the absorption spectrum of a 20–30 nm thick C$_{60}$ film deposited on a 3 mm thick ZnSe substrate in the 8.5 μm region.

A scheme of the experimental setup is shown in Fig. 1. Tunable pulsed radiation from the free-electron laser for infrared experiments (FELIX) in Nieuwegein, The Netherlands, is used for this study. FELIX produces IR radiation that is continuously tunable over the 5–250 μm range with a minimum achievable bandwidth (FWHM) of 0.3% of the central frequency. The light output consists of macropulses of about 5 μs duration containing up to 50 mJ of energy. Each macropulse consists of a train of micropulses with a duration of 0.3–5 ps, separated by 1 ns. In the present experiment FELIX runs at a 5 Hz repetition rate, and the full macropulse is used to excite the optical cavity in the 8–9 μm region. If required, a higher peak intensity in the optical cavity can be obtained when pulse-stacking, or synchronous pumping, is performed, as recently demonstrated using the Stanford FEL. The temporal shape of the FELIX pulse is well-approximated by a flat-topped profile with an exponentially decaying tail with a time-constant of 360±20 ns (ring down time of the 6 m long FELIX cavity, implying a Q-factor of less than 20 for the laser cavity). A long pass filter (7.2 μm cutoff wavelength) is used to prevent the higher harmonics that might be present in the FELIX-beam from entering the ring down cavity.

The ring down cavity is formed by two identical plano-concave mirrors with a diameter of 25.4 mm and a radius of curvature of −1.0 m placed a distance d = 36 cm apart. With a specified reflectivity R of the mirror coatings of 0.9998 at 8.5 μm (Laser Power Optics), a ring down time of the empty cavity at 8.5 μm is shown, together with a linear fit to the natural logarithm of the decaying tail.

FIG. 1. Scheme of the experimental setup. In the inset the ring down transient of the empty cavity at 8.5 μm is shown, together with a linear fit to the natural logarithm of the decaying tail.
such a way that the cavities that are formed are again optically stable, transients with a ring down time larger than 2.2 μs are found over the 8.1–8.8 μm region. This implies that the additional losses due to the presence of the ZnSe window are below 340 ppm per pass. The upper limit for the absorption losses for this particular sample of ZnSe in this wavelength region is therefore concluded to be below 1.1 × 10⁻³ cm⁻¹, which can be accurately measured using this experimental setup. With a thin film of C₆₀ deposited on the ZnSe window the CRD spectrum shown in the lower part of Fig. 2 is recorded. The baseline of the spectrum is determined by the effective cavity losses, (1 - Rₚ₀)/d, mainly due to the mirrors, the substrate and scattering losses from the thin film. Absorption of the thin film introduces an extra loss determined by κl/d, in which κ is the absorption coefficient and l the thickness of the thin film. The absorption line centered at 8.46 μm is one of the four F₁₄ IR fundamental absorptions of C₆₀. With an absorption coefficient of 0.24 μm⁻¹ and taking the mismatch between the linewidth of the C₆₀ absorption line in the thin film (2 cm⁻¹) and the width of the spectral profile of FELIX (8 cm⁻¹) into account, a sample thickness of approximately 20 nm is found, in good agreement with the 20–30 nm sample thickness found in independent FTIR measurements.

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FIG. 2. (a) Mirror reflectivity as a function of wavelength as measured via CRD in the empty cavity. (b) CRD absorption spectrum of a 20–30 nm thick C₆₀ film on a 3 mm thick ZnSe window.