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158 - Chiroptical signal enhancement and intrinsic limitations

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Abstract Body: Despite its unique capability of distinguishing molecular handedness, chiroptical spectroscopy suffers from the weak signal problem, which has restricted more extensive applications. Quasi-null-polarization detection (QNPD) method has been shown to be useful for enhancing the chiroptical signal. Here, the underlying enhancement mechanism in the QNPD method combined with heterodyne detection scheme is elucidated. It is experimentally demonstrated that the optical rotatory dispersion (ORD) signal can be amplified by a factor of ~400, which is the maximum enhancement effect achievable with our femtosecond laser setup. The upper limit of the QNPD enhancement effect of chiroptical measurements could, in practice, be limited by imperfection of polarizer and finite detection sensitivity. However, we show that there exists an intrinsic limit in the enhancement with the QNPD method due to the weak but finite contribution from homodyne chiroptical signal. This is experimentally verified by measuring optical rotation of linearly polarized lights with QNPD scheme. We further provide discussions on the connection between this intrinsic limitation in the QNPD scheme for enhanced detection of weak chiroptical signal and those in optical enantioselectivity and Raman optical activity with structured chiral field. We anticipate that the present work would be useful in further developing time-resolved nonlinear chiroptical spectroscopy.