Ultrafast optical excitation of magnons in 2D antiferromagnetic semiconductors via spin torque mediated by unbound electron-hole pairs and excitons: Signatures in magnonic charge pumping

Jalil Varela-Manjarres, Yafei Ren, and Branislav K. Nikolic

University of Delware, University of Delaware, 19 Yale Drive Apt C, NEWARK, USA

Recent experiments observing how femtosecond laser pulse (fsLP) excites magnons in two-dimensional (2D) antiferromagnetic (AF) semiconductors—such as CrSBr, $NiPS_3$, and $MnPS_3$, or their van der Waals heterostructures—suggest an important role played by excitons. However, microscopic details of such an effect remain obscure, as resonant coupling of magnons, living in the sub-meV energy range, to excitons, living in the ~ 1 eV range, can hardly be operative. Here, we develop a quantum transport theory of this effect, in which the time-dependent nonequilibrium Green's function (TDNEGF) for electrons driven by fsLP is coupled self-consistently to the Landau-Lifshitz-Gilbert (LLG) equation describing classical dynamics of localized magnetic moments (LMMs) residing on magnetic atoms of 2D AF semiconductors. This theory explains how fsLP, of central frequency *above* the semiconductor gap, generates a photocurrent that becomes spin-polarized due to the background of LMMs, which, in turn, exerts spin-transfer torque (STT) onto LMMs as a genuinely nonequilibrium spintronic mechanism. By analyzing the collective motion of LMMs via the windowed Fast Fourier transform (FFT), we extract frequencies of excited magnons, as well as their lifetime governed by *nonlocal* damping with the LLG equation due to (explicitly included via TDNEGF) electronic bath. The TDNEGF part of the loop can also include excitons, which is achieved in the present study by utilizing the off-diagonal elements of the nonequilibrium density matrix as the mean-field treatment of Coulomb-binding of electron-hole pairs into excitons. Finally, our theory makes it possible to predict that excited magnons will *pump* time-dependent charge currents into the attached electrodes, or locally within the 2D AF semiconductor, thereby emitting electromagnetic radiation. The windowed FFT of these two signals contains imprints of excited magnons, as well as the possible presence of excitons, which could be exploited as a novel probe in future experiments.

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