Title: Observation of Bloch-Siegert shift in an atomically thin crystal

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Introduction. Coherent light-matter interaction can be used to manipulate the energy levels of atoms, molecules and solids. When light with frequency ω is detuned away from a resonance ω_0, repulsion between the photon-dressed (Floquet) states can lead to a shift of energy resonance. The dominant effect is the optical Stark shift ($\propto 1/(\omega_0 - \omega)$), but there is an additional contribution from the so-called Bloch-Siegert shift ($\propto 1/(\omega_0 + \omega)$). Although it is common in atoms and molecules, the observation of Bloch-Siegert shift in solids has so far been limited only to artificial atoms since the shifts were small (<1 μeV) and inseparable from the optical Stark shift. Here we observe an exceptionally large Bloch-Siegert shift (~10 meV) in monolayer WS_2 under infrared optical driving by virtue of the strong light-matter interaction in this system. Moreover, we can disentangle the Bloch-Siegert shift entirely from the optical Stark shift, because the two effects are found to obey opposite selection rules at different valleys. By controlling the light helicity, we can confine the Bloch-Siegert shift to occur only at one valley, and the optical Stark shift at the other valley. Such a valley-exclusive Bloch-Siegert shift allows for enhanced control over the valleytronic properties in two-dimensional materials, and offers a new avenue to explore quantum optics in solids.

The fundamental interaction between light and matter can be understood within the framework of a two-level system. When driven by off-resonant light $\hbar\omega < \hbar\omega_0$ $(= E_0)$, there are two pairs of photon-dressed (Floquet) states which contribute to the state repulsion with the original states – one pair between the original states (Fig. 1a) and the other pair outside the original states (Fig. 1b). The former case leads to a shift of transition energy called the optical Stark (OS) shift, which increases linearly with the light intensity ($\mathcal{E}_0^2$) and inversely with the detuning energy, $\Delta E_{\mathrm{OS}} \propto \mathcal{E}_0^2/(E_0 - \hbar\omega)$. The latter case also leads to a shift, called the Bloch-Siegert (BS) shift, but it has a different energy dependence, $\Delta E_{\mathrm{BS}} \propto \mathcal{E}_0^2/(E_0 + \hbar\omega)$. Although the Bloch-Siegert shift is negligible at small detuning, it can become comparable, and serves as an important correction, to the optical Stark shift at large detuning.

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We report on the observation of an unprecedentedly large Bloch-Siegert shift ($\Delta E_{\mathrm{BS}} \sim 10$ meV), which can be entirely separated from the optical Stark shift. Such a large and exclusive Bloch-Siegert shift is realized in a monolayer of transition-metal dichalcogenide (TMD) tungsten disulfide (WS_2). This is possible because this material system possesses two distinctive features. First, it exhibits strong light-exciton interaction at the two time-reversed valleys (K, K') in the Brillouin zone. Secondly, the two valleys possess finite and opposite Berry curvatures due to the lack of inversion symmetry, giving rise to distinct optical selection rules and related valleytronic properties. That is, the optical transition at the K (K') valley is coupled exclusively to left-handed σ^- (right-handed σ^+) circularly polarized light. This leads to a unique material platform that allows us to separate the Bloch-Siegert shift from the optical Stark shift by using circularly polarized light.

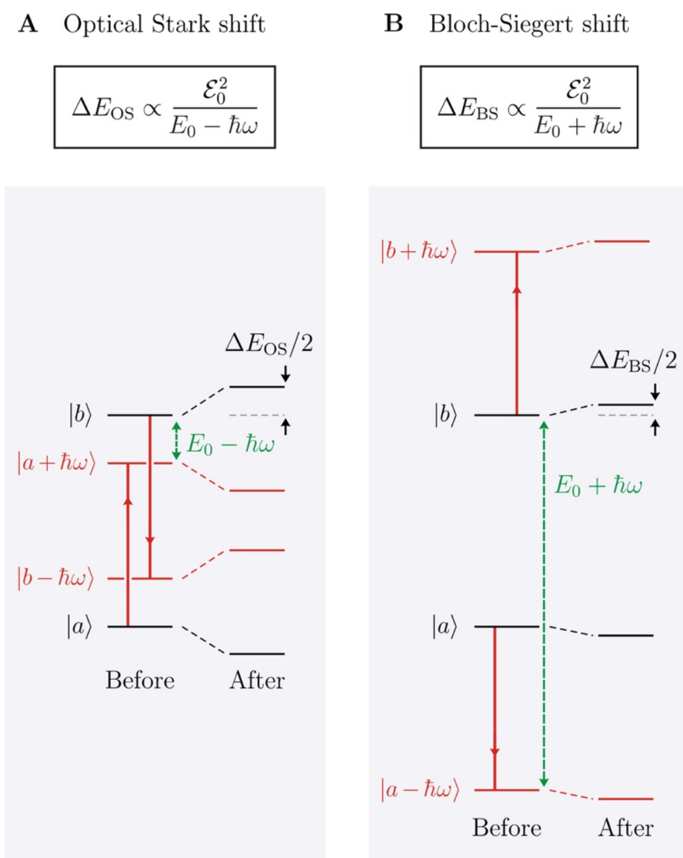

Figure 1. Comparison of the optical Stark shift and the Bloch-Siegert shift in a two-level system. (a) Energy diagram for optical Stark (OS) shift. $|a\rangle$ and $|b\rangle$ denote the two original states with resonance energy E_0 before they are optically driven. $|a+\hbar\omega\rangle$ and $|b-\hbar\omega\rangle$ are photon-dressed (Floquet) states driven by the co-rotating optical field. Hybridization between these Floquet and original states causes the resonance energy to blueshift by ΔE_{OS}, which is proportional to the light intensity ($\mathcal{E}_0^2$) and inversely proportional to $(E_0 - \hbar\omega)$. **(b)** Energy diagram for Bloch-Siegert (BS) shift. $|a-\hbar\omega\rangle$ and $|b+\hbar\omega\rangle$ are two different Floquet states driven by the counter-rotating optical field. Hybridization between these Floquet and original states causes the Bloch-Siegert shift, with magnitude ΔE_{BS} inversely proportional to $(E_0 + \hbar\omega)$.