

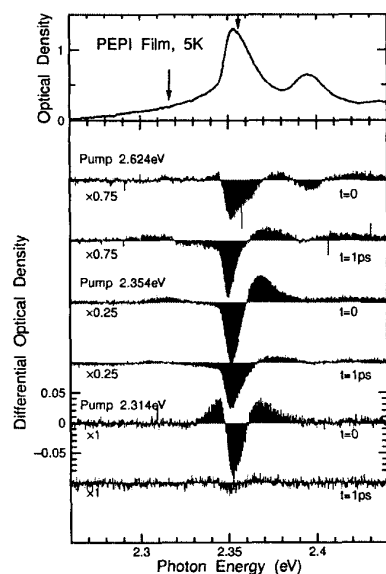
QTh125

Femtosecond pump-probe spectroscopy on two-dimensionally confined excitons in inorganic-organic layered perovskite compound

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In an inorganic-organic layered compound of $(\text{C}_6\text{H}_5\text{C}_2\text{H}_4\text{NH}_3)_2\text{PbI}_4$, a 2D exciton has a binding energy of 220 meV and an oscillator strength of 0.5 per formula unit by virtue of the spatial and the dielectric confinement by organic barrier layers.¹ In order to investigate the optical nonlinearity and its mechanism affected by these characteristics, we have performed the pump-probe spectroscopy on its spin coated films at 5K. The time resolution was about 200 fs and the pump power density was $1 \text{ MJ}/\text{cm}^2$.

Figure 1 shows differential absorption spectra at 0 and 1 ps after pumping at the lowest exciton resonance (middle), above the band gap (upper), and below the exciton resonance (lower). The linear absorption spectrum is shown for reference. By pumping at the exciton resonance, bleaching and blue shift occur for the exciton band. The blue shift consists of a slow component of picoseconds and a fast component of less than 200 fs, respectively. The former is due to the exchange interaction between excitons,^{2,3} and the latter may be attributed to the optical Stark effect. Surprisingly, the "exciton" density at $t=0$ is estimated to be $1 \times 10^{14}/\text{cm}^2$ per layer. In this excitation density, in other systems such as semiconductor quantum wells⁴ and CuCl thin films,⁵ the electron-hole plasma is the stable phase and excitons (bound state) do not survive. The fact that excitons sur-



QTh125 Fig. 1. Differential absorption spectra at 0 and 1 ps after pumping at the lowest exciton resonance (middle), above the band gap (upper), and below the exciton resonance (lower). The linear absorption spectrum is shown for reference.

vive in our layered compound indicates that excitons are stabilized by the spatial and the dielectric confinement.

By pumping free electron-hole pairs with more than the band gap energy, the exciton band is bleached, but shift and broadening are negligibly small at $t=0$. This result can be explained in two ways. The one is that both exchange interaction and long-range Coulomb interaction are not effective between excitons and free electron-hole pairs. The other is that repulsive exchange interaction is balanced by attractive long-range Coulomb interaction between these two species; due to the dielectric confinement, long-range interaction may not be screened. Then within 1 ps, blue-shift and bleaching of excitons, as well as biexciton absorption occur, as with pumping at the exciton resonance. Therefore, we conclude that free electron-hole pairs relax to the bound state within 1 ps.

By pumping below the exciton resonance, considerable broadening of the exciton band occurs during the pump pulse of 200 fs. Since the pump energy corresponds to the spectral tail of the exciton band due to the polaritonic effect, we now tentatively attribute the broadening to the scattering between photon-like and exciton-like polaritons. Interestingly, no residual effect (usually caused by really excited species) remains after the pump pulse passes through the sample.

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QTh126

Probing mixed phase space dynamics using atom optics

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Atom optics has proven to be a fruitful testbed for the experimental study of quantum dynamics in classically chaotic systems. Previous work in our group¹ has focused on a regime where the classical phase space is strongly chaotic, so that any stable phase-space structures are small, and the atomic phase-space distribution is broad, so that the dynamics are averaged over the features in phase space. However, the quantum behavior in a mixed classical phase space, where both stable and chaotic regions are present, remains an interesting and challenging subject, with much left to understand.

To probe local dynamics in mixed phase space, the atomic sample must be prepared such that it is localized in phase space. To accomplish this, cesium atoms are first prepared in a standard magneto-optic trap. They are then further cooled in a 3-D far-detuned optical lattice similar to the setup described in.² After the atoms are adiabatically released from this lattice, they have a Gaussian momentum distribution with $\sigma_p/(2\hbar k_L) = 1.4$

(where $\hbar k_L$ is the photon recoil momentum). The atoms are then adiabatically loaded into a 1-D, linearly polarized, far-detuned optical lattice, where they become localized in the nodes of the standing wave. The spatial phase of the lattice is controlled by an electro-optic modulator. We center the atomic distribution at different momenta by quickly shifting the lattice position and allowing the atoms to evolve until they return to the lattice nodes, accumulating momentum along the way. The atoms can then be centered at any desired spatial phase by either quickly shifting the lattice phase again or by allowing the atoms to drift in free space for an appropriate duration. A typical distribution (localized in both position and momentum) prepared with this technique in the deep-well regime (so that the lowest few bands are approximately harmonic oscillator states) results in a Gaussian distribution with $\sigma_p/(2\hbar k_L) = 3$; the corresponding minimum uncertainty state is about 50% populated, and is characterized by $\sigma_p/(2\hbar k_L) = 1.7$ and $\sigma_x = 0.05$ lattice periods. The atoms prepared in this manner are then ready for an interaction with a time-dependent potential.

A model system for the study of mixed phase space is realized when the atoms are subjected to an amplitude-modulated standing wave, where the intensity varies as $\sin^2(\pi t/T)$ (T is the pulse duration). There are three primary resonances in this system;³ one is at zero momentum, and the other two are located symmetrically about $p=0$. As the intensity of the standing wave increases, the resonances overlap, and the resonances appear as islands of stability surrounded by bands of chaos. We have used our method of state preparation to center the atomic distribution at various positions and at the same momentum as one of the "side" resonances. We observe trapping of the atoms in the resonance when the atomic distribution is centered on the island, but not when the atoms are centered away from the resonance, since they instead diffuse through the chaotic region.

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QTh127

An optical micro-linear accelerator for molecules and atoms

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Acceleration of molecules within a time varying electric field produced by an optical traveling wave has been proposed as a means to accelerate