Time-Resolved Optical Observation of Spin-Wave Dynamics

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We have created a nonequilibrium population of antiferromagnetic spin waves in Cr_2O_3, and characterized its dynamics, using frequency- and time-resolved nonlinear optical spectroscopy of the exciton-magnon transition. We observe a time-dependent pump-probe line shape, which results from excitation induced renormalization of the spin-wave band structure. We present a model that reproduces the basic characteristics of the data, in which we postulate the optical nonlinearity to be dominated by interactions with long-wavelength spin waves, and the dynamics to be due to spin-wave thermalization.

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Optical magnetic excitations have been studied extensively in several magnetic oxides [1,2], and have recently attracted interest in studies of low-dimensional correlated electron systems [3,4]. In this Letter we demonstrate how time-resolved, nonlinear optical spectroscopy (TR-NLOS) of optical magnetic excitations may be used to investigate the interactions and dynamics of short-wavelength magnetic modes in strongly correlated systems. These short-wavelength excitations are the most difficult to treat theoretically, and even in three dimensions their mutual interaction is not fully understood. We present results of femtosecond pump-probe spectroscopy of the exciton-magnon (X-M) absorption feature in the antiferromagnetic oxide Cr_2O_3 [5], with both temporal and spectral resolution. This optical absorption feature allows us to excite antiferromagnetic spin waves directly, with nonequilibrium occupation distributions weighted toward large momenta, high energies, and with sufficient density to observe interaction effects. We have observed a novel nonlinear optical effect associated with the nonequilibrium occupation dependence of the spin-wave (SW) dispersion relation. In semiconductor physics, it has been demonstrated that important and nontrivial physics of correlated many-particle systems can be clarified through careful attention to dynamics, using TR-NLOS [6]. Our work indicates that TR-NLOS can be used to directly manipulate and study magnetic excitations in strongly correlated insulators, in addition to the charged excitations usually probed with optics.

In the periodic lattice of a magnetic crystal the concept of a SW appears naturally when the fermionic spin Hamiltonian is expressed in terms of boson operators [7]. Controlled approximations exist for distributions of long-wavelength SWs at low excitation densities, but away from these conditions theory must be guided by experimental observations. Neutron spectroscopy and linear optical spectroscopy have provided some of the best evidence for the existence of spin-wave renormalization (SWR) at elevated temperatures [8,9], but as previous measurements were largely limited to thermally occupied SWs, not much is known about the interactions among excitations at short wavelengths. Theory suggests that the interactions may undergo qualitative changes as the zone boundary is approached [10]. In our experiments, using laser excitation of the X-M absorption, we are able to macroscopically occupy a strongly nonthermal distribution of SWs, with occupation heavily weighted toward the zone boundary.

The X-M transition may be understood as a magnon sideband to an exciton, and is similar in character to the well-known two-magnon absorption. In a cubic environment, the three electrons per Cr^{3+} site possess a ground state with $^4A_2$ symmetry and a lowest excited ^2E state at $\sim$1.7 eV. In a lattice, these levels couple via superexchange interactions J and J', of order 50 meV. The ground state multiplet develops into antiferromagnetic SWs, and the excited state multiplets into magnetic exciton bands. The total spin projection S_z is preserved by the two spin excitations, so the spin selection rule $\Delta m = \Delta m_X + \Delta m_M = 0$ is satisfied. To conserve momentum the photon is absorbed by an exciton and a magnon of equal and opposite momentum, $\mathbf{k}_X + \mathbf{k}_M \sim 0$ [11]. The excitation is largely localized to neighboring sites, and consequently draws its spectral weight from states over the entire Brillouin zone. Neglecting interaction between the nearby exciton and magnon for clarity, the X-M absorption line shape is given approximately by the joint density of states (JDOS) [5],

$$\rho_{e\text{-}m}(\omega) = \sum_{\mathbf{k}} \delta(\omega - \omega^e_{\mathbf{k}} - \omega^m_{-\mathbf{k}}). \qquad (1)$$

The SW dispersion is stronger than that of the exciton and provides the dominant contribution to the line shape. The absorption feature may be qualitatively understood as the SW DOS, shifted up rigidly in energy by the exciton energy. This allows us to excite SWs and subsequently

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monitor their density of states (DOS), using a pulsed near-infrared laser.

We have performed pump-probe spectroscopy of the X-M transition, using 100 fs pulses emanating at 76 MHz from a Ti:sapphire laser tuned to $\hbar\omega \approx 1.765$ eV. The beam is split in a 10:1 (pump:probe) ratio and the time delay, $\Delta t = t_{\text{probe}} - t_{\text{pump}}$ is controlled with a delay line. Both pump and probe are focused through a microscope objective to a 6 μm diameter spot at the sample, which is held at 10 K with a cold finger cryostat. We estimate the average temperature at the sample to be $\leq$30 K. The sample is a 2.2 μm thick film of Cr_2O_3 grown epitaxially on sapphire, by evaporation of Cr in a reactive oxygen environment. The c axis of both Cr_2O_3 and sapphire is normal to the film surface. At the peak of the X-M absorption line, the internal transmissivity of the sample is 52%.

After the sample we measure $\Delta T \times I_{\text{inc}} = I_{\text{on}} - I_{\text{off}}$, the change in the transmitted probe intensity as the pump is chopped mechanically. In the small signal regime, the normalized change in transmission reproduces the differential absorption of the sample, by the relation $\Delta T/T \approx -\Delta\alpha L$. We measure $\Delta T/T \simeq (I_{\text{on}} - I_{\text{off}})/I_{\text{off}}$ as a function of wavelength and time delay, and substract from this a small background contribution which persists at negative time delay because of the finite repetition rate. At excitation densities of $\sim 10^{-3}$/Cr, we observe the complex spectral feature shown in Fig. 1 for three different time delays. The data are well described in terms of two components: a spectrally featureless photoinduced absorption (PIA), which shifts the overall $\Delta T/T$ toward negative values and is relatively time independent over 100 ps, and a derivativelike line shape, which is weakly evident at $\Delta t = 0$ and grows as a spectral unit as Δt increases.

The magnitude and qualitative line shape shown in Fig. 1 may be explained by recognizing that the absorption of each photon creates a SW, which in turn renormalizes the overall SW band structure through interaction. In a cubic ferromagnet with occupation at long wavelengths, this renormalization takes the form

$$\omega_{\mathbf{k}}(\{n_{\mathbf{k}'}\}) = \omega_{\mathbf{k}}^0\left[1 - \frac{1}{zJS^2N}\sum_{\mathbf{k}'}\langle n_{\mathbf{k}'}\rangle\omega_{\mathbf{k}'}\right] = \omega_{\mathbf{k}}^0[1 - \kappa\mathcal{E}_{\text{tot}}], \quad (2)$$

where $\mathcal{E}_{\text{tot}}$ is the total energy of the excited SWs [7]. Similar, more complicated expressions hold for antiferromagnets and for different crystal structures. In our experiments, the X-M line shape reflects this renormalization, as $\alpha(\omega,\{n_{\mathbf{k}}\}) \simeq \rho_{e\text{-}m}(\omega,\{n_{\mathbf{k}}\})$ now depends on the time-dependent distribution of photoexcited SWs, and the pump-probe experiment measures the time evolution of $\sum_{\mathbf{k}} \frac{d\alpha}{dn_{\mathbf{k}}} n_{\mathbf{k}}(t)$. In principle, the exciton dispersion relation should also be renormalized for the same reasons, but the exciton bandwidth is a factor of 2 narrower than that of the SWs, and its dispersion is weakest near the zone boundary, so this effect contributes little to the overall line shape change. Numerical simulations confirm these arguments. Since α is proportional to the integrated JDOS given in Eq. (1), the total derivative $\frac{d\alpha}{dn_{\mathbf{k}}}$ includes two distinct contributions, one from the level shifts $\frac{d\omega_{\mathbf{k}}}{dn_{\mathbf{k}}}$ and the other from the change in the integration volume associated with the level shifts. Qualitatively, the level shifts produce an overall redshift in the energy, while the change in the integration volume reduces the spectral weight.

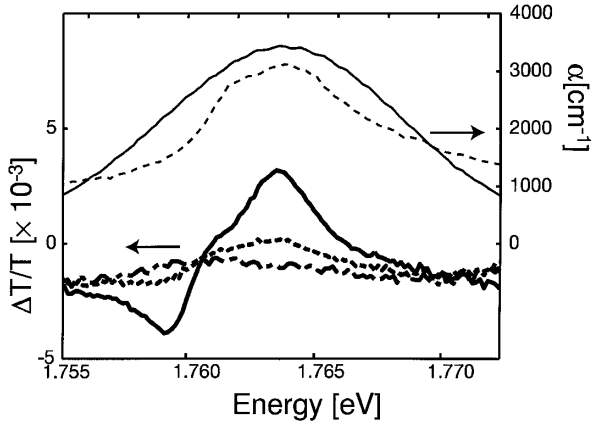

FIG. 1. Pump-probe spectrum at $\Delta t = 0.3$ (heavy dash-dotted line), 10 (heavy dotted line) and 50 ps (heavy solid line), with units given on the left, shown together with the absorption spectrum (light dashed line), with units given on the right, and the incident laser spectrum (light solid line), shown for reference in arbitrary units.

It is interesting to compare the pump-probe signal at long times to the change in the linear absorption induced by raising the temperature. The SW energy absorbed from the laser is only $\sim$2% of the total absorbed energy, so from the total incident energy density of 5.6 J/cm^3 only $\sim$100 mJ/cm^3 is absorbed by the SW system. From the known SW dispersion relation measured by neutrons [12], we may calculate the SW contribution to the specific heat, $C_p \simeq C_v = \frac{2\pi}{15}\frac{k_B^4}{D^3}T^3$, where $D \simeq 1.25 \times 10^{-28}$ J/cm. The energy density of one laser pulse thus corresponds to a temperature change of $\sim$30 K in the magnetic system. In Fig. 2, we compare the saturated pump-probe line shape to that expected from a 30 K temperature change, based on the measured temperature dependence of the linear absorption. For additional comparison, we also show the line shape calculated from Eqs. (1) and (2), using a scale factor κ derived from the temperature-dependent absorption spectra. The pump-probe spectrum has been scaled up by a factor of 3, which may reasonably be attributed to the simplifications of Eq. (2).

The agreement among these curves confirms our assignment of the line shape to SWR, and shows that the energy density in the magnetic system at a long time delay is comparable to that absorbed directly by the spin system from the pump beam. The magnetic excitons absorb the majority of the laser energy, serving as a reservoir.

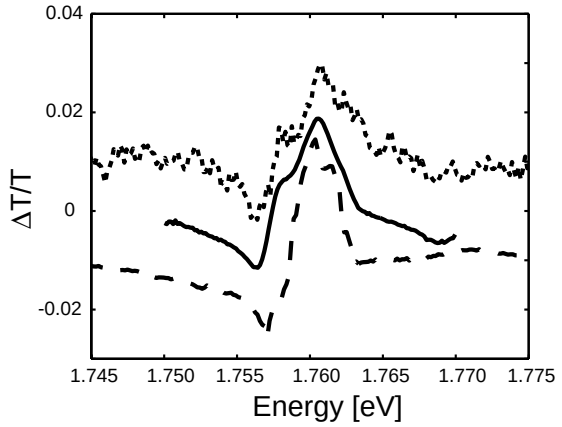

FIG. 2. Comparison of the saturated pump-probe line shape with simulated and actual changes due to a thermalized energy density. Solid line: pump-probe line shape, $\times 3$; dotted line: thermal difference spectrum; dashed line: calculation using Eqs. (1) and (2).

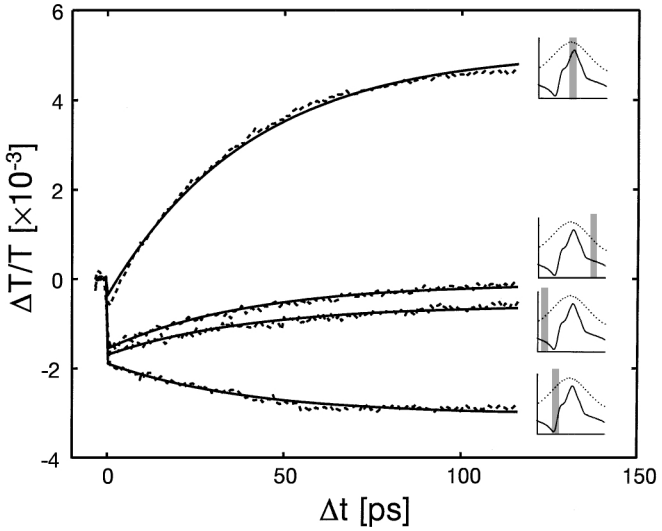

FIG. 3. Time-resolved pump-probe signal at different wavelengths over energy bandwidth $\hbar\Delta\omega = 20$ meV. Probe energy ranges are indicated in the insets accompanying each curve, and correspond to the following average energies, listed from top to bottom: 1.764, 1.771, 1.754, and 1.759 eV.

The excess energy is transferred rapidly to quenching sites, whereupon it is dissipated over 100 ns to several microseconds via nonradiative processes [13]. In the steady state, a large number of these defect states will be excited, together with the steady state phonon and SW distributions. As a test for indirect energy transfer to the magnetic system via defects and phonons, we have performed two-color pump-probe experiments using a Coherent RegA 9000 regenerative amplifier system to create high intensity pump pulses at 1.5 eV, well away from the X-M absorption feature. We focused a portion of this beam onto a 3 mm sapphire crystal to generate a white light continuum probe, and used an interference filter to select a 15 meV spectral range spanning the X-M absorption. For absorbed pulse energy densities ranging from 1–100 times those used in the degenerate pump-probe experiments, we observed no measurable change in the X-M absorption, indicating that the mechanisms for energy transfer from defect absorption into the magnetic system occur on time scales much longer than those of interest here. We conclude that the magnetic system behaves as a quasiclosed system at least during the first nanosecond, and that the dynamics which we observe is related to an intrinsic internal thermalization of the optically induced, nonequilibrium SW population.

We show the time evolution directly in Fig. 3, where we plot the response at different wavelengths, keeping the laser center wavelength fixed. As in Fig. 1, we have subtracted a small background component, present at negative time delay, which is due to the steady state heating of the sample. When the probe frequency is outside the X-M line $\Delta T/T$ exhibits prompt PIA with a decay time of at least 500 ps, not shown here. When the probe frequency is inside the X-M line $\Delta T/T$ exhibits both prompt PIA and picosecond dynamics. The initial distributions of SWs created by the laser are weighted heavily toward the zone boundary, because of the factor of $4\pi k^2$ in the JDOS integral given in Eq. (1). If all SWs contributed equally to the renormalization, as suggested by Eq. (2), one would expect the SWR and hence the X-M line shape to undergo an abrupt change at $\Delta t = 0$, and remain unchanged during the internal thermalization of the SW system. This clearly is not the case: the initial population of $k \approx \pi/a$ SWs contributes little to the X-M renormalization line shape.

The exciton and magnon are initially created on neighboring sites and should interact moderately with each other, but the difference in their relative group velocities of ~ 10 Å/ps indicates that they should be well separated after a picosecond or less, so we do not believe that the observed change is associated with the decay of the X-M composite. It is also possible that the process of optical absorption in the presence of large k excitations is not well described by the particular approximation used here, but must include additional many-particle interactions. Such effects, however, would need to suppress the contribution of SWR to the pump-probe line shape by an order of magnitude. The simplest explanation for our result is that occupation at large k produces weaker overall SWR than those at the zone center. Such strong k dependence in SW interactions has long been indicated theoretically, and the variation of the interaction at short wavelengths may be so strong that the overall interaction effects cancel [14].

Regardless of the underlying reason for the difference between zone center and zone boundary SW occupation, we can describe the dynamical response phenomenologically by dividing the occupied SW states into two different populations, those at the zone boundary (b) and those at the zone center (c), with a boundary in reciprocal space

chosen to reproduce the experiments. We assume that the decay of the initial nonequilibrium energy density $\mathcal{E}_b = \sum_{\mathbf{k}\in\mathbf{k}_b} n_{\mathbf{k}}\epsilon_{\mathbf{k}}$ into the thermalized energy density $\mathcal{E}_c = \sum_{\mathbf{k}\in\mathbf{k}_c} n_{\mathbf{k}}\epsilon_{\mathbf{k}}$ is governed by a single thermalization time, τ, and the decay over long times is set by an overall energy decay time, $\mathcal{T}$. Clearly, τ provides a measure of the interactions coupling the zone boundary spin waves to those at lower energy, both directly and via phonons. This process may be described by the following phenomenological rate equations:

$$\frac{d\mathcal{E}_b}{dt} = -\mathcal{E}_b/\tau - \mathcal{E}_b/\mathcal{T}\,, \qquad \frac{d\mathcal{E}_c}{dt} = \mathcal{E}_b/\tau - \mathcal{E}_c/\mathcal{T}\,. \tag{3}$$

If we further assume that only the energy density due to zone center SWs $\mathcal{E}_c$ is involved in the renormalization line shape, we obtain the following equation for the time-dependent pump-probe signal, valid for each energy within the X-M absorption region:

$$\Delta T/T = a_1\Theta(t) + a_2[1 - \exp(-t/\tau)]\exp(-t/\mathcal{T})\,, \tag{4}$$

where a_1 and a_2 are prefactors which depend on the spectral region of interest. The step function $\Theta(t)$ is required to account for the PIA, which we have taken to be time independent, consistent with experiments away from the X-M peak. The weak structure at early times, due to SWR, is also included in a_1, though in principle this may be accounted for by an additional term. We have divided the 10 nm spectral range given by our laser spectrum into ten ranges of equal width, separated by 1 nm, and measured the temporal pump-probe response in each range. We then found the best global fits to the data of Eq. (4), in which a_1 and a_2 are allowed to vary with probe wavelength, and $\tau = 40 \pm 10$ ps and $\mathcal{T} = 2 \pm 1$ ns are constrained to be the same for all ten wavelengths. The results for four of them are shown by the theoretical curves in Fig. 3. This simple model captures very well the observed dynamics.

In summary, we have generated a macroscopic, nonequilibrium population of SWs and observed its dynamics, using pulsed laser spectroscopy. At long times, the change in the absorption line shape is well understood by assuming that the photogenerated SWs induce a renormalization of the SW dispersion relation. At short times, our results deviate sharply from the predictions of this model, indicating that short wavelength SWs do not contribute to the renormalization line shape. The SWs form a quasiclosed system over 100 ps time scales. The evolution of this nonequilibrium population is consistent with a simple model of SW thermalization, and the thermalization time characterizes intrinsic spin-wave coupling. The effects reported here provide us with information on elementary magnetic excitations that is inaccessible through conventional techniques, which typically probe only thermally occupied magnetic excitations. Moreover, the technique may be used quite generally in magnetic insulators, and may be applied to a wide variety of optical magnetic excitations, including two-magnon excitations and the recently assigned phonon-bimagnon feature in the undoped cuprates [3]. By using multiple wavelength laser sources, this technique may be used in conjunction with excitations across the Mott gap to probe directly the interactions between charge and spin excitations in magnetic solids.

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