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Optical induction of magnetization and observation of fast spin dynamics in aqueous solutions of copper ions

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Abstract

Optically induced magnetization and its fast dynamics in aqueous solutions of transition-metal ions is studied by the polarization spectroscopy with the pump-probe technique. The fast spin dynamics in solutions at room temperature, which cannot be detected by the conventional magnetic-resonance methods, was observed by an all-optical method. A circularly polarized pump pulse creates the magnetization in the ground state of the copper ions in aqueous solutions of copper sulfate, and the time evolution of the magnetization in the subnanosecond region is monitored through the change of the polarization of the probe pulse. Quantum-beat free-induction decay signals are observed in transverse magnetic fields, whose Fourier transform gives the ESR spectra. The observed concentration dependence of the relaxation rate in no external magnetic field is interpreted by the spin cross relaxation due to the magnetic dipole interaction between the copper ions.

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1. Introduction

Optical induction of magnetization by short laser pulses is a very useful method to investigate fast spin dynamics in condensed matter near room temperature. In many cases, the magnetization in solids is induced thermally at low temperatures and under high magnetic fields, and the dynamic behavior of the magnetization has been observed by using the magnetic resonance methods. Spin relaxation of transition-metal ions in aqueous solutions was studied by using the NMR and ESR methods [1–4]. In these magnetic-

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resonance methods, the information on the relaxation was derived from the resonance-line profile, while the direct observation in the time domain has not been given since the time resolution of these methods is not enough to observe the decay curve of the magnetization near room temperature. The observations in the time and the frequency domains are complementary to each other to obtain the information both on the energy structure and the relaxation, and are useful for the synthetic understanding of microscopic structure and dynamics. As for the relaxation, the observation of line profile is an indirect one, and direct observation of the time evolution is very significant. The spin-lattice relaxation has been investigated in detail by the magnetic-resonance methods, but the study on the spin–spin relaxation or spin cross relaxation of magnetization in aqueous solutions has not been reported so far.

The observations of optically induced magnetization and the spin dynamics of the transition-metal ions in crystals or aqueous solutions at room temperature have been reported in the time domain [5–8]. In these experiments the magnetization was induced by circularly polarized laser pulses, and the time derivative of the optically induced magnetization was monitored by pickup coils. Although the time resolution in such experiments superior to that of the conventional magnetic-resonance methods, it is of the order of nanoseconds at the highest. If the induced magnetization is monitored by optical pulses, the time resolution can be remarkably improved because the time resolution is limited only by the temporal width of the light pulses. Then it is possible to observe the ultrafast spin dynamics in the picosecond or femtosecond region. Another advantage of the all-optical method is the sensitivity. The magnetization signals can be observed by the selective optical pumping even in low magnetic fields or near room temperature, where the signal detection by the magnetic-resonance methods is not necessarily easy because of the small thermally-induced population difference. In order to study the ultrafast spin dynamics of magnetic ions in solutions near room temperature, we applied the polarization spectroscopy with the pump-probe technique for the creation and detection of the magnetization [9]. The ultrafast spin dynamics have been studied by the similar technique in dilute magnetic semiconductors [10–14], bulk semiconductors [15–18], and metals [19–22].

In the present Letter we report on the optically induced magnetization and fast spin dynamics of copper ions in aqueous solutions at room temperature. The magnetization in the ground state of the copper ions is induced by a circularly polarized pump pulse, which can induce instantaneously a large population difference even at high temperatures and in low magnetic fields. The time evolution of the magnetization in the region of subnanoseconds is monitored through the change of the linear polarization of the probe pulse due to circular dichroism. By using this technique, a time resolution much higher than that in the conventional methods is achieved and direct observation of the fast spin dynamics in solutions is realized. In a transverse magnetic field, the precession of the magnetization around the external magnetic field is observed as quantum-beat free-induction-decay (FID) signals. The Fourier transform of the observed FID signals gives the ESR spectra. This method may be called optically induced Fourier-transform (FT) ESR spectroscopy. The line shape of the ESR spectra gives information about the molecular dynamics in the solution. In zero magnetic field, monotonous decay of the magnetization is observed. The observed decay curve depends on the concentration of the copper ions, which can be interpreted as an effect of the spin cross relaxation due to the magnetic dipole–dipole interaction between the copper ions.

2. Experiment

The experimental setup is shown in Fig. 1. The light source is a mode-locked Ti:sapphire laser/regenerative amplifier system producing 0.2 ps pulses at wavelength $\lambda \approx 790$ nm. The pump and probe pulses are provided by optical parametric amplifiers (OPA1 and OPA2). The circularly polarized pump and linearly polarized probe beams are nearly collinear and focused in the sample, aqueous solutions of copper sulfate ($CuSO_4$) in a glass cell of 1 mm thickness. The wavelengths of the pump and probe pulses are 1.3 μm and 650 nm, respectively, which are selected by considering the appropriate absorption [23]. The waist sizes of the beams at the focus are about 100 μm. The repetition rate of the laser pulses is 1 kHz, and the pulse energies of the pump and probe pulses are about 10 and 1 μJ in front of the sample cell.