Interaction of picosecond laser pulses with the electron, spin, and phonon subsystems of nickel

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(Submitted 13 July 1983)
Zh. Eksp. Teor. Fiz. **86**, 1376–1379 (April 1984)

It was observed that a section of magnetized nickel film 50–100 nm thick does not become demagnetized when heated by a light pulse of duration 5–20 psec to double the Curie temperature. A similar experiment at a pulse duration 40 nsec resulted in demagnetization of the heated section of the film. It is deduced from the results that the values of the spin-lattice and spin-electron relaxation times in the vicinity of the Curie temperature lie in the interval 10^{-9}–4×10^{-8} sec. The data obtained are discussed in the framework of the theory of relaxation phenomena in the electron, phonon, and spin systems of the ferromagnet.

Noninertial emission from metals, induced by picosecond laser pulses, was observed experimentally in Refs. 1 and 2. It was shown there that this phenomenon is determined by the deviation of the temperature T_e of the electron subsystem of the metal from the lattice temperature T_i. The possibility of deviation of T_e from T_i in metals was indicated in Ref. 3. A solution of the problem of interaction between ultrashort laser pulses and the electron and phonon subsystems of a metal was obtained in Refs. 1 and 4, where it was shown that the deviation of T_e from T_i can be substantial and reach, say for silver, $(3–4)\times10^3$ K. The conditions under which T_e and the emission follow without inertia the temporal changes of the laser-pulse power density were derived in Ref. 2.

Rapid processes that occur in the electron and phonon subsystems of a metal were investigated in Refs. 1–4. In the present paper, using as the example the interaction of picosecond laser pulses with ferromagnetic nickel films, we investigate relaxation processes in the electron, phonon, and spin subsystems of a metal. It is known[5] that nickel is a ferromagnet with collectivized electrons, and that the relaxation processes in it are the most complicated ones.

EXPERIMENTAL RESULTS

The change of the magnetization of thin nickel films irradiated by laser pulses of duration 10^{-11} and 10^{-8} sec were investigated by a setup whose optical diagram is shown in Fig. 1. The films were heated with an aluminum-yttrium-garnet laser (1) emitting at 10.6 μm and generating either single picosecond pulses of duration 5–20 psec and energy up to 10 mJ (first run of experiments) or single nanosecond pulses of 40 nsec duration and energy up to 1 J (second run). The energy density at the focus of the lens 8 was smoothly varied with the aid of attenuator 7. The laser-pulse parameters were monitored in the nanosecond-pulse regime with a type FK (coaxial) photocell 2 and a calorimeter 3, and the picosecond pulse regime with an "Agat" image-converter streak camera 4 and a photocell operating as a calorimeter. The relative error in the measurement of the radiation power did not exceed 10%. The energy distribution in the laser-pulse spot was close to Gaussian.

The change of the magnetization after applying the laser pulse to the film was measured by the magnetooptic Kerr effect using a differential photoelectric signal-recording system. The probing radiation source was the single-mode helium-neon laser 10. The probed region was smaller than the irradiated one to ensure uniformity of the illumination. The polarizing elements were Glan prisms 14. Signals from the reference and measuring photomultipliers 12 and 15 were fed to the of the differential amplifier of an S1-74 oscilloscope. An electromagnet (not shown in the figure) was used to turn the magnetic field on and off and to set its strength and polarity. The experiments were performed on nickel films 50–10 nm thick sputtered on glass substrates.

All the numerical data and estimates that follow are for films 50 nm thick. The films were first magnetized to saturation, after which the field was turned off and the rotation of the polarization plane of the probing radiation was measured in the remanent state of the film magnetization. The measurements were repeated after applying the laser pulse. To monitor the irreversible changes in the magnetic surface, when no damage to the film could be observed in an optical microscope, the film was magnetized to saturation in the opposite direction and the rotation of the polarization plane was measured again.

The streak-camera measurements of the kinetics of the emission produced by the action of picosecond pulses on the nickel film were performed in analogy with Refs. 1 and 2.

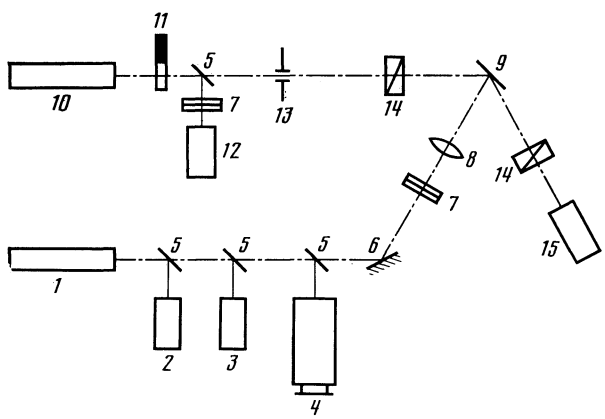

FIG. 1. Optical system of setup: 1—pulsed laser, 2—FK coaxial photocell, 3—calorimeter, 4—"Agat" image converter streak camera, 5—beam splitting plates, 6—mirror, 7—neutral light filters, 8—lens, 9—film, 10—He-Ne laser, 11—modulator, 12—reference photomultiplier, 13—diaphragm, 14—polarizer, 15—measuring photomultiplier.

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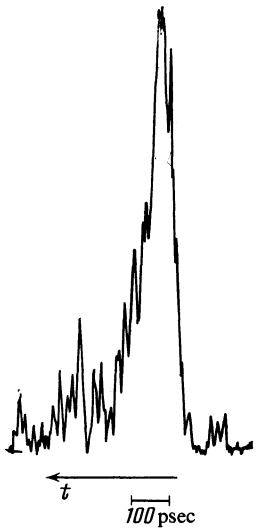

FIG. 2. Typical streak photograph of emission from nickel.

The experimental results reduce to the following.

1. Streak camera measurements have shown that a visible emission pulse is observed and, unlike in silver,[2] does not have the same shape as the initiating picosecond pulse (Fig. 2).

2. Heating a section of the film by a laser pulse of duration $\tau_p = 40$ psec and energy density $\approx 10^{-1}$ J/cm^2 softens this section. When the laser-pulse is increased by 2.5–3 times the magnetization of the heated section of the film is not restored when the field is turned on again, i.e., this section becomes irreversibly nonferromagnetic (apparently as a result of formation of the antiferromagnetic oxide NiO at temperatures close to the melting point). Further increase of the laser-pulse power causes evaporation of the Ni (Ni evaporates at $T_{ev} = 2550$ K when heated by a laser pulse[6]).

3. When a section of the film is heated by a pulse of duration $\tau_p = 5$–20 psec, no change of magnetization is observed when the power density is increased up to a value corresponding to the irreversible magnetization loss at $\tau_p = 40$ nsec. In the absence of irreversible changes in the film structure it can be assume that the lattice temperature T_i is proportional to the radiation power density. Consequently, on heating to the irreversible magnetization loss, Ti is approximately 2.5–3 times higher (see Item 2) than the Curie temperature of nickel ($T_C = 631$ K) and is close to the melting temperature ($T_m = 1726$ K).

Thus, in contrast to heating by pulses with $\tau_p = 40$ nsec, the film is not demagnetized when heated by a pulse of duration 5–20 psec all the way to more than double the Curie temperature.

DISCUSSION OF RESULTS

Since the specific heats c_e, c_i, and c_s of the electron, phonon, and spin subsystems in Ni are different at room temperature and the time of temperature equalization within each of these systems is much shorter than the characteristic times of heat-exchange between the systems,[2–4,6–10] the electron temperature T_e can deviate from the lattice and spin-system temperatures T_i and T_s. The laser energy is absorbed by the electron system and is transferred to the lattice, to the spin system, and to the film substrate. The heat exchange processes are characterized by the time $\tau_{ei} \sim 10^{-10}$ sec of equalization of the temperatures of the electron and phonon systems (Ref. 2) and by times τ_{es} and τ_{is} of the spin-electron and spin-lattice relaxation.

Since the electron subsystem of Ni has, by virtue of the presence of s- and d-like states, a larger value of c_e than in transition metals, the deviation of T_e from T_i for Ni under the action of picosecond pulses is less effective than for Ag and Cu, and the condition that the emission have no inertia (see Ref. 2), viz., $c_e/\alpha_{ei} \ll \tau_p$ ($\alpha_{ei} \approx c_i/\tau_{ei}$), is not satisfied for Ni at $\tau_p = 5$–20 nsec; this explains the difference between the streak photographs of the emission of Ni and Ag.

According to the experimental data, when a lattice is heated by picosecond pulses to 2.5–3 times the Curie temperature T_C, i.e., at $T_i > T_C$ and $T_s < T_C$, the film does not become demagnetized ($\tau_p = 5$–20 psec). During the initial time interval $t < \tau_{ei}$ we have $T_e > T_i > T_c > T_s$, after which the electron and lattice temperatures become equalized and are lowered by energy transfer to the spin degrees of freedom and to the substrate. The solution of the heat-conduction problem for the two-layer film + substrate system by the method described in Ref. 11 offers evidence that when the lattice is heated to $T_i = 1700$ K by pulses of $\tau_p = 5$–20 psec the time of cooling to T_C is $t_0 \approx 10^{-9}$ sec. Consequently, since $T_s < T_C$, it follows that $\tau_{es} > t_0$ and $\tau_{is} > t_0$. For heating by a "long" pulse, thermodynamic calculation shows that the film temperature exceeds T_C within a time comparable with the pulse duration $\tau_p = 4 \times 10^{-8}$ sec, i.e., τ_{es}, $\tau_{is} < 4 \cdot 10^{-8}$ sec. We have thus in the vicinity of TC

$$10^{-9} \text{ c} < \tau_{es},\ \tau_{is} < 4 \cdot 10^{-8} \text{ sec}. \tag{1}$$

Let us compare our data with the results of the theory of relaxation phenomena in ferromagnets.[8–10] According to Refs. 8 and 9,

$$\tau_{is} = \frac{c_i c_s}{(c_i + c_s) D_{is}}, \quad D_{is} = \frac{\pi V \hbar k_B}{60 \rho a^8}, \tag{2}$$

(ρ is the density, a is the lattice parameter, k_B is Boltzmann's constant, V is the volume) and, according to Ref. 10,

$$\tau_{es} = \frac{c_e c_s}{(c_e + c_s) D_{es}}, \quad D_{es} = 10^{-12} \frac{V}{a^3} T_e [\text{erg} \cdot \text{deg}^{-1} \cdot \text{sec}^{-1}]. \tag{3}$$

Assuming for Ni in the vicinity of T_C, on the basis of the experimental data, that $c_s = 0.3 c_i$ and $c_e = c_s$, we obtain from (2) $\tau_{is} = 2.2 \cdot 10^{-8}$ sec, and at $T_e = 10^3$ K we get from (3) $\tau_{es} = 2.3 \cdot 10^{-9}$ sec.

Thus, although the theory of relaxation phenomena in ferromagnets[7–10] was developed for small deviations from the uniformly magnetized state and is strictly speaking inapplicable at temperatures near the Curie point, the experimentally obtained interval (1) of values for τ_{es} and τ_{is} does not contradict the established notions concerning the energy relaxation between the electron and phonon systems, on the one hand, and the spin system on the other.

The authors thank Professor B. M. Stepanov for interest and for a discussion that prompted this work.

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Translated by J. G. Adashko