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Laser-induced grating spectroscopy of high-T_c superconductors

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We propose a new spectroscopical method for the investigation of ultrafast phenomena in metallic and high-Tc superconductors by using tunable laser beams, without requiring femtosecond laser pulses. The method is based on the self-diffraction of a laser beam from a laser-induced moving grating. The theory of the method has been developed in detail for the case of electron-phonon (EP) interaction. Our theoretical predictions are in good agreement with a recent experiment. Using experimental data we estimated the value of the EP coupling constant for the $YBa_2Cu_3O_7$ film.

1. INTRODUCTION

Recently we have proposed [1] a new method for optical investigation of metals and superconductors with ultrafast time resolution by operating in the frequency domain using tunable lasers. The method is based on laser-induced moving gratings and does not require femtosecond laser pulses (complex systems and expensive).

The optical scheme describing the principles of the method is shown in figure 1. Two laser beams with different frequencies $\omega_1 \neq \omega_2$ (ω_1=const, ω_2=var) and wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ produce a nonstationary intensity distribution in the medium under investigation. This intensity exhibits a wavelike modulation with a grating vector $\mathbf{q} = \mathbf{k}_1 - \mathbf{k}_2$ and a frequency $\Omega = \omega_1 - \omega_2$. The wavelike modulated light intensity changes the optical properties of the material in the interference region resulting in a moving grating structure.

If this wavelike modulation is slow in comparison to the relaxation time, τ of the optical properties of the material ($\tau << \Omega^{-1}$), they follow the intensity change and the grating amplitude does not decrease. If $\Omega \geq \tau^{-1}$, the optical properties of the material do not follow the intensity modulation, and the grating structure becomes less contrast, and therefore the grating amplitude decreases. The grating effectiveness measured by the self-diffraction of waves ω_1 falls down for this case. In the moving grating method the relaxation velocity of the material optical properties is compared with the motion velocity of the grating (which is proportional to the frequency detuning).

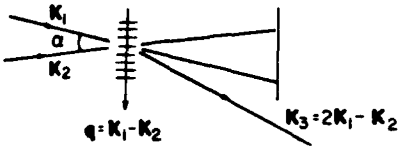

Figure 1. The scheme describing the principles of the method.

The theory of the method has been developed in detail [1,2] for the case of the investigation of electron-phonon (EP) interaction. It was based on the assumption that the relaxation within both the electron and the phonon subsystems was much faster than the rate of an energy exchange between them.

Simultaneously with our theoretical work [1] an experimental work [3] has been published, devoted to the non-linear spectroscopy of YBCO and Ni thin films by biharmonic pumping technique. Thanks to this experimental study, a unique possibility appeared to compare the theoretical predictions [1] with experimental data.

Below we shall describe the theoretical basis of the method for the investigation of EP interaction in metals and superconductors, and compare our theoretical predictions with the experimental data of Ref. [3]. Then we shall briefly discuss the possibility of using the method for the study of other relaxation mechanisms.

2. THEORY OF THE METHOD FOR STUDY OF EP INTERACTION

EP interaction plays a main role in metallic [4] and possibly an important role in high-Tc superconductors [5]. Therefore, measurements of EP interaction in superconductors are very important. In particular, such measurements can provide information about the contribution of EP interaction to the high-Tc superconductivity.

The topic of EP interaction is closely related to the thermal relaxation of hot electrons (or other carriers). Allen [6] related the relaxation rate τ_e^{-1} of the electron temperature T_e (by energy exchange between electrons and the lattice) to the factor $\lambda <\omega^2>$, which is of great interest in the theory of the superconducting transition temperature T_c:

$$\tau_e^{-1} = 3\hbar\lambda <\omega^2>/(\pi k_B T_e) \qquad (1)$$

Here $<\omega^2>$ is the second moment of the phonon spectrum, λ is the EP coupling constant. Thus, measuring the thermal relaxation of hot carriers provides information about EP interaction.

The equations for the temperature distribution in a thin film can be written in the form [1-2,7-8]

$$\partial T_e/\partial t = - \tau_e^{-1} (T_e-T_i) + f(\mathbf{r},t)/C_e \qquad (2)$$

$$\partial T_e/\partial t = \tau_i^{-1}(T_e-T_i) - \tau_{esc}^{-1} (T_i-T_{in}) \qquad (3)$$

where $\tau_i = \tau_e C_i/ C_e$ is the relaxation time of the lattice temperature T_i, $C_e= \gamma T_e$ and C_i are the electronic and the lattice specific heats, correspondingly, τ_{esc} is the time it takes for phonons to escape into the substrate, T_{in} is the initial temperature, $f(\mathbf{r},t)$ is the heat production per unit volume per unit time by the light waves $\omega_1\mathbf{k}_1$ and $\omega_2\mathbf{k}_2$,

$$f(\mathbf{r},t) = F_o+[F_1 \exp(-i(\Omega t-\mathbf{qr}))+c.c.],$$

$$F_o = \chi(J_1+J_2), \quad |F_1| = \chi(J_1J_2)^{1/2}$$

$$\chi = \hbar\omega 2kb(1-R)\exp(-2bk_o\mathbf{r}),$$

R is the reflectivity, J_i is the power density of the incident waves on the surface (in terms of [phot/cm^{2}s]), $k_o \equiv k_{01}= \omega_1/c \approx \omega_2/c$, $\omega \equiv \omega_1 \approx \omega_2$, 2kb is the absorption coefficient.

It can be shown [1,2] that the square of the modulus of the factor

$$I(\Omega)=[1+i(1+r_i/r_e)/(\Omega\tau_i)](\tau^{-1}-i\Omega)^{-1} \qquad (4)$$

describes the dependence of the power of the signal (with the wave vector $\mathbf{k}_3=2\mathbf{k}_1-\mathbf{k}_2$ and the frequency $\omega_3= 2\omega_1- \omega_2$) on the frequency detuning Ω for $|\Omega| >> \tau_{esc}^{-1}$. Here $r_{e,i} = \partial\varepsilon/\partial T_{e,i}$, ε is the permittivity,

$$\tau^{-1}=\tau_e^{-1}(T_{eo})+\tau_i^{-1}=\frac{3\hbar}{\pi k_B}(T_{eo}^{-1}+\bar{\gamma}C_i^{-1})\lambda<\omega^2>, \quad (5)$$

$\tau_e(T_{eo}) = \bar{\gamma}T_{eo}\tau_i/ C_i$ is the new value of the electronic temperature relaxation time and is a function of the heat production[1]

$$T_{eo}=T_{in}+F_o(\tau_i + \tau_{esc})/ C_i \qquad (6)$$

In the case of weak heating of the electronic system the method permits the determination of the EP interaction parameter $\lambda<\omega^2>$ without the knowledge of both the electron temperature T_e and (for $\tau_e \ll \tau_i$) the electron and lattice specific heats, $C_e = \bar{\gamma}T_e$ and C_i, respectively. In the regime of strong heating of the electron system the proposed method permits direct observation and measurement of the dependence of the thermal electron relaxation on the heat production by the external radiation.

It is obvious that in order to obtain sufficiently large signal power J_3 and to prevent film overheating it is necessary to use a long pump pulse (with pulse duration $t_p > \tau$). Therefore, in an experiment it is convenient to record the energy of the signal $k_3\omega_3$ $E_3 \sim \int_{-\infty}^{\infty} dt\, J_3(t)$. We have shown in [1] that for sufficiently long pulses the dependence of $E_3(\Omega)$ is similar to that of $J_3(\Omega)$.

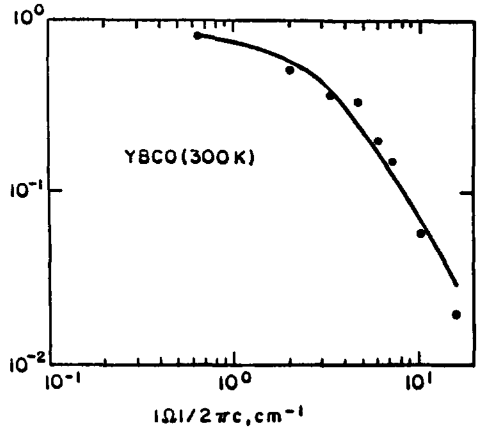

Figure 2. The dependence of the signal energy E_3 on the frequency detuning Ω using the theoretical dependence $|1- i\Omega\tau|^{-2}$, dots -experiment data [3].

3. COMPARISON WITH EXPERIMENT

We shall compare our theoretical predictions with the experimental data [3], where the scheme shown in figure 1 was used. The experimental data for YBCO in the range ~ 10 cm^{-1} and the corresponding theoretical dependence of $|I(\Omega)|^2$ (Eq.(4)) are shown in Fig.2. The agreement between the theory and the experiment is good. The theoretical curve shown in Fig.2 has been calculated only with one fitting parameter τ^{-1} that corresponds to the condition $|(1 + r_i/r_e)/(\Omega\tau_i)| \ll 1$. It can be shown that a second fitting parameter only weakly influences the curve fitting and the magnitude of τ^{-1}.

We have estimated the electron temperature in the experiment [3] as $T_{eo} \sim 10^3$K. The magnitude of the fitting parameter $\tau^{-1}/(2\pi c)$ is equal to 3 cm^{-1} for the YBCO film at T_{in}=300 K. Using this value and formula (5), we can estimate the EP coupling constant λ. We obtain λ~0.1, where we used a Debye temperature θ_D =

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[1] If one uses a pump pulse with a pulse duration $t_p < \tau_{esc}$, it is necessary to substitute in Eq. (5) $t \le t_p$ for τ_{esc} for the estimation of T_{eo} during the pulse action.

350K [9] and $\tilde{\gamma}/C_i \approx 6.10^{-4}$ K^{-1}[10] for YBCO, and the formula $< \omega^2 > = k_B^2\theta_D^2/(2\hbar)$.

4. DISCUSSION

The estimated value $\lambda \sim 0.1$ is smaller than the literature data $\lambda = 0.9$ for YBCO [8]. For a better estimation of λ from the optical measurements the exact magnitudes of both the reflectivity R (which determines the absorbed energy) and the pulse shape should be known.

Another possible factor which affects the accuracy of the determination of λ, is the deviation from the two-temperature model. Recently, the nonthermal distribution of electrons in noble metals was observed in pump-probe subpicosecond laser experiments [11,12]. Therefore, it is very interesting to generalize the theory of the laser-induced grating spectroscopy and to take into account the carrier-carrier relaxation effects. It will extend the possibilities of the method, and permit to determine not only the EP coupling constant but also the carrier-carrier relaxation parameters. The corresponding investigation is in progress.

In conclusion we proposed a new method for the investigation of ultrafast phenomena in metals and superconductors. The method is based on laser-induced moving gratings and does not demand ultrashort laser pulses. The theory of the method has been developed for the case of studying EP interaction. Our theoretical predictions conform with the experiment. We have estimated the value of the EP coupling constant for YBCO film using the experimental data of Ref. [3]. Thus, the method is real and it works.

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REFERENCES

1. B.Fainberg, Opt.Communs.,89 (1992) 403.
2. B.Fainberg, D.Huppert, Nonlinear Optics, (1993), in press.
3. V.N.Bagratashvili, V.A. Lobastov, A.N. Zherikhin, V.M.Petnikova and V.V. Shuvalov, Phys. Lett.A, 164 (1992) 99.
4. J.Bardeen, L.N.Cooper, J.R.Schrieffer, Phys. Rev. 108 (1957) 1175.
5. T.Timusk, C.D.Porter, D.B.Tanner, Phys. Rev. Lett. 66 (1991) 663; E. Altendorf, J.Chranowski, J.C. Irwin, A. O'Reilly and W.N.Hardy, Physica C, 175 (1991) 47.
6. P.B.Allen, Phys. Rev. Lett., 59 (1987) 1460.
7. S.I.Anisimov, B.L.Kapeliovich and T.L. Perelman, Sov. Phys. JETP, 39 (1977) 375.
8. S.V.Chekalin, V.M.Farztdinov, V.V. Golovlyov, V.S.Letokhov, Yu.E. Lozovik, Yu.A.Matveets and A.G.Stepanov, Phys. Rev. Lett., 67 (1991) 3860.
9. D.E. Gray (ed.), American Institute of Physics Handbook, McGraw Hill, New York, 1972, 3rd edition.
10. A.Junod, Physica C,153-155 (1988) 1078.
11. R.H.M.Groeneveld, R.Sprik, and Ad Lagendijk, Phys. Rev. B, 45 (1992) 5079.
12. W.S.Fann, R.Storz, H.W.K.Tom, and J. Bokor, Phys. Rev. Lett., 68 (1992) 2834.