VOLUME 25, NUMBER 10

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Phonon contribution to the frequency-dependent part of the relaxation time involved in the intraband optical conductivity of noble metals

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It is shown that at room temperature the phonon contribution to the frequency-dependent part of the relaxation time involved in the intraband optical conductivity of noble metals is comparable to that of the electron-electron scattering as determined from dc measurements by Parkins, Lawrence, and Christy.

The frequency dependence of the relaxation time $\tau(\omega)$ involved in the intraband optical conductivity of the noble metals has been carefully analyzed by Parkins, Lawrence, and Christy.¹ For infrared photon energies $\hbar\omega$,

$$\frac{1}{\tau(\omega)} \simeq \alpha + \beta \hbar^2 \omega^2 , \qquad (1)$$

where α and β are adjustable parameters. The empirical values of β (β_{expt}) for Cu, Ag, and Au are shown in Table I; β_{expt} seems to be temperature independent, at least in the measured range 78–425 K.

Both electron-electron scattering² and electron scattering by disordered ions^{3,4} lead to a frequency-dependent relaxation time. The ion disorder may be due to phonons, impurities, grain boundaries, and structural disorder (liquid or amorphous phases). Parkins, Lawrence, and Christy¹ as well as Beach and Christy⁵ indicated that the quadratic dependence on ω and the temperature independence of β_{expt} in Eq. (1) suggest that the electron-electron scattering is the dominant effect. However, the coefficients β_{expt} exceed by a factor of 2 to 3 the theoretical estimates $\beta_{e-e,theor}$ and by an order of magnitude the values $\beta_{e-e,theor}$ inferred in a rather model-independent way from dc measurements of the electrical and thermal resistivities (Table I).¹ Thus, the presence of some kind of structural disorder cannot be excluded as a possible explanation for the large temperature-independent values of β_{expt} observed.⁴ Parkins, Lawrence, and Christy¹ pointed out that if this is indeed the case then, in sufficiently perfect samples, β_{expt} should be consistent with the dc measurements, that is, with the values $\beta_{e-e,dc}$.

We show here that around and above room temperature the phonon contribution to β (β_{ph}) is comparable to $\beta_{e-e,dc}$, and thus β_{expt} should rather be consistent with the sum $\beta_{e-e,dc} + \beta_{ph}(T)$. We

TABLE I. Coefficient β in units of $10^{13} \sec^{-1} eV^{-2}$.

	Cu	Ag	Au
β_{expt} (average) ^a	2.1	1.4	2.4
$\beta_{e-e, \text{theor}}^{a}$	0.64	0.73	0.76
$\beta_{e-e,dc}^{b}$	0.19	0.25	~0.49
$\beta_{\rm ph}$ (78 K)	0.06	0.08	0.04
$\beta_{\rm ph}$ (295 K)	0.20	0.11	0.28
$\beta_{\rm ph}$ (425 K)	0.29	0.22	0.44

^aValues taken from Table IV of Ref. 1.

^bValues taken from Table V and Eq. (16) of Ref. 1.

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evaluated $\beta_{ph}(T)$, which had not been considered before in this context.

A quantum-theoretical treatment of the dielectric function $\epsilon(\omega)$ of an electron gas interacting with disordered ions leads to³

$$\boldsymbol{\epsilon}(\boldsymbol{\omega}) = 1 - \left[\frac{\omega_p}{\omega}\right]^2 [1 - \gamma(\boldsymbol{\omega}) + i\boldsymbol{\xi}(\boldsymbol{\omega})/\boldsymbol{\omega}]^{-1}, \qquad (2)$$

where ω_p is the plasma frequency of the freeelectron gas and $\gamma(\omega) - i\xi(\omega)/\omega$ is a complex function of ω that is expressed in terms of an integral over the scattering momentum transfer q, and whose integrand involves, besides some phase factors, the electron-ion pseudopotential V_q and the structure factor S(q) of the ions in the metal.

At infrared energies the relaxation frequency $\xi(\omega)$, which is to be identified with $\tau^{-1}(\omega)$, can be approximated by

$$\xi(\omega) = \alpha_{\rm ph} + \beta_{\rm ph} \hbar^2 \omega^2 , \qquad (3)$$

where α_{ph} coincides with α if the static electronelectron and impurity scattering are negligible. We calculated β_{ph} for Cu, Ag, and Au at room temperature with the use of pseudopotentials reported

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by Moriarty⁶ and a model structure factor S(q) in the one-phonon approximation with Einstein dispersion $\omega_q = k_B T_E / \hbar$.⁷ The Einstein temperature T_E was adjusted to reproduce the empirical static resistivity (or $\alpha_{\rm ph}$) at room temperature. The results are shown in Table I. The values of $\beta_{\rm ph}$ reported at 78 and 425 K in Table I were obtained under the assumption that $\beta_{\rm ph}$ is proportional⁸ to $\alpha_{\rm ph}$; the corresponding experimental values of $\alpha_{\rm ph}$ were taken from Table III of Ref. 1.

In conclusion, it is apparent from Table I that the temperature-dependent contribution of phonon scattering to the coefficient β in Eq. (1) is significant, and that already at room temperature it becomes comparable to that of electron-electron scattering as determined from dc measurements.

The work of G. C. was partially supported by Comision de Fomento de Actividades Académicas—Instituto Politécnio Nacional (COFAA-IPN). J. S. H. acknowledges the partial support of the Organization of American States (OAS), Secretaria de Patrimonio y Fomento Industrial (SEPAFIN, Mexico) and Consejo Nacional de Ciencia y Tecnología (CONACyT, Mexico).

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- ⁸This proportionality is exact within the Einstein approximation for the phonon dispersion and we conjecture that it should be very approximate in general for the solid. However, this relation between β_{ph} and α_{ph} cannot be extrapolated from the solid to the liquid or amorphous state; this may lead to the overestimation of β_{ph} by an order of magnitude. In the liquid state β_{e-e} is negligible compared with the contribution to $\beta(\sim 5-10\times 10^{13} \text{ sec}^{-1} \text{ eV}^{-2})$ coming from the structural disorder.