LORENZ-FUNCTION ENHANCEMENT DUE TO INELASTIC PROCESSES NEAR THE NÉEL POINT OF CHROMIUM

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We report the discovery of a localized enhancement of the Lorenz function in the vicinity of the Néel point of chromium. It is believed to be a manifestation of the inelastic scattering processes that occur due to the critical fluctuations in the spin-density-wave system resulting in an augmentation of the electron and spin-wave thermal conductivities.

Observations have been made of the thermal and electrical conductivities of chromium on the same polycrystalline sample in the region of the Néel point. The sample of residual resistivity ratio 178 was made from 99.999% pure material having the following impurities: Al, 1; Ca, 3; Cu, 1; Fe, 2; Mg, 1; C, 10; H_2, 0.8; and O_2, 9 ppm by weight. A senstitive longitudinal heat-flow method with differential thermocouple thermometry was employed for the thermal-conductivity work. Good resolution was achieved without undue loss of precision while maintaining a small temperature difference of 0.3 K. Full experimental details will be reported elsewhere.

The basic results between 275 and 325 K are reproduced in Fig. 1 in terms of the electrical and thermal resistivities. It is apparent at once that several qualitative features of the behavior of the electrical resistivity are found also in the thermal resistivity. In particular, below the Néel point T_N, the appearance of the spin-density-wave (SDW) periodic potentials, which cause the well-known hump-backed increment to the electrical resistivity, clearly provides resistance to the electronic thermal-conduction processes in a similar way. On the other hand, the minima of the two resistivities (ρ_{min} at 312.2 K and W_{min} at 313.5 K) not only differ significantly in temperature but occur at least a degree higher than the Néel temperature (311 K). Evidently in the region of the phase transition the details of the conduction and scattering processes which come under the influence of the fluctuations in magnetization are not the same in the two cases. Furthermore, W_{min} is deeper and sharper than in the case of the electrical resistivity (5% deep compared with 2%), falling to lower values than would be expected by simple extrapolation from higher temperatures through the region of the Néel transition. This is best appreciated by referring to Fig. 2 in which both the Lorenz function and the thermal conductivity are plotted as functions of the temperature. The thermal-conductivity enhancement destroys the monotonic temperature dependence of the Lorenz function and leads to an anomalous peak of the latter centered at about 313 K. There also occurs a weak minimum in the Lorenz function at 307.5 K that we tentatively ascribe to the effect of developments of the SDW states which inhibit phonons and so lower the <u>phonon</u> conductivity. We have

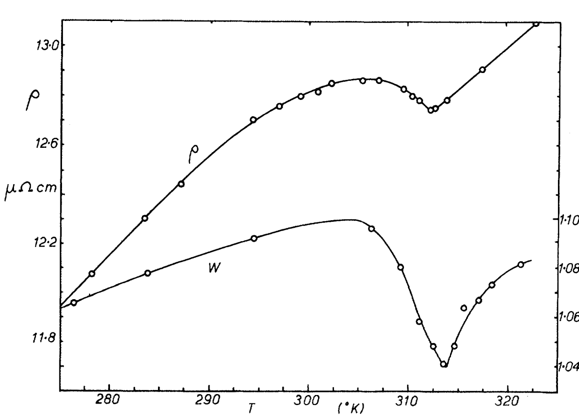

FIG. 1. The thermal and electrical resistivities of chromium between 275 and 325 K.

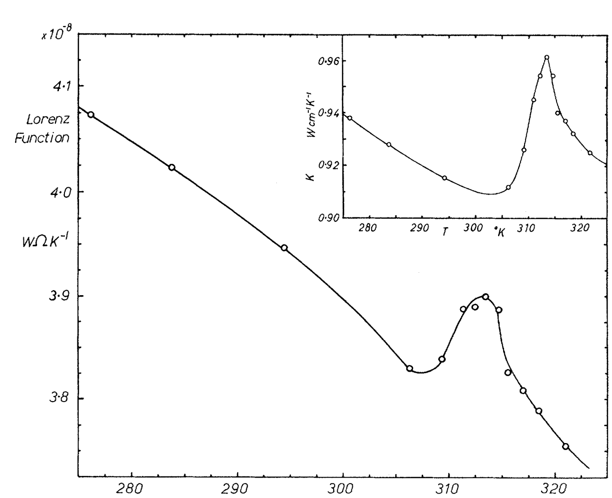

FIG. 2. The Lorenz function of chromium between 275 and 325 K. Inset: the thermal conductivity.

shown elsewhere[1] that above 220 K the thermal conductivity of the present chromium can be described by $K_{tot} = 2.80\times10^{-8}T/\rho + 79.9/T\ W\,cm^{-1}\,K^{-1}$ to within 1%. The second term demonstrates the existence of phonon conduction in this metal, and its magnitude around the 300-320 K region represents about 25% of the total conductivity. However, this analysis does not consider the consequences of the antiferromagnetic phase transition. In the proximity of this transition there is known to exist a considerable effect on the elastic moduli,[2] although it is not clear to what extent this should affect the transport properties. But if, as is likely, it alters the lattice vibration spectrum, it will affect the electron-phonon scattering probability and hence the transport properties, besides affecting the spin-phonon interaction too. In addition, of course, the critical fluctuations will diffuse the conduction electrons[3-5] and a breakdown of the Weidemann-Franz law will ensue locally for both this[3,6] and the foregoing reasons. Our explanation of the enhancement of the thermal conductivity near T_N is therefore as follows: In the vicinity of T_N critical fluctuations in the energy density of the SDW system are responsible for critical scattering of the phonons. The resulting strong phonon absorption, while lessening the phonon conductivity, enhances considerably the electron thermal conductivity and may even generate a significant spin-wave conductivity. The net result is a positive contribution to the thermal conductivity in the form of a roughly symmetrical peak.

This peak also bears a superficial resemblance to the magnetic specific-heat peak at 311.6 K reported by Beaumont, Chihara, and Morrison.[7] The Néel temperature of our chromium sample is at about 311 K according to susceptibility results[8] and to the temperature location of the electrical resistivity derivative $(d\rho/dT)_{min}$. The association of the latter [and $(dW/dT)_{min}$, too] with T_N has been explained by Suezaki and Mori[3] as being a natural consequence of the summation of the magnetic superzone and critical scattering processes acting on the conduction electrons in antiferromagnetic metals, and has been used to locate T_N successfully in chromium[5,9] and antiferromagnetic rare-earths, including terbium.[10] Salamon, Simons, and Garnier[11] have even demonstrated the coincidence of the specific heat peak and $(d\rho/dT)_{min}$ at about 311 K in their single-crystal and polycrystalline chromium specimens. Hence the supposition by several previous workers[6,12] that ρ_{min} is sited at T_N now appears invalid. By the same reasoning it should not be expected that W_{min} should locate at T_N, and the fact that ρ_{min}(312.2 K) and W_{min}(313.5 K) differ markedly in temperature,[13] whereas $(d\rho/dT)_{min}$ and $(dW/dT)_{min}$ almost coincide in temperature (close to 311 K), further supports the argument.[14] Accordingly, the difference in temperatures of ρ_{min} and W_{min} is merely a result of slightly differing effects of the critical inelastic scattering on the phonon system and on the conduction electrons. A fortiori, the Lorenz function maximizes at a temperature a little higher than T_N as well. In short, we conclude that besides the usual electrical- and thermal-resistivity increases occurring about T_N because of critical-fluctuation scattering of the conduction electrons (and which maximize where the resistivities undergo inflexions), the same fluctuations are also responsible for the enhancement of the thermal conductivity and Lorenz function at a slightly higher temperature, namely, at 313 K.

We point out that this is the first observation of an inelastic peak associated with a magnetic phase transition to be found in the thermal conductivity of any solid. However Huber[15] has speculated that a rise in the thermal conductivity near the Curie point of a ferromagnetic lattice may result because the volume of the Brillouin zone accessible to the paramagnetic spin waves increases as the temperature nears T_c. Previous to our work only narrow-deep or shallow-broad minima (and sometimes merely temperature-derivative increases) have been noted.[6,16] Such behavior occurs in insulators (e.g., EuO [16]), which have high-phonon conductivities and no electronic conductivity, and in the rare-earth or iron-group metals[16,17] which have high spin-disorder resistivities. The case of chromium is different from these other solids because of its comparatively low phonon conductivity and negligible spin-disorder resistivity. Previous work on chromium revealed only a change of slope of the thermal conductivity and Lorenz function near T_N.[18] Obviously measurements of quite high resolution and precision are needed in studies of this type.

Lastly, we point out that a continuation of these experiments using magnetic fields is being initiated. This will inform us of the role played by spin-wave conductivity near the transition temperature and by magnetic superzone scattering below it. Also, with improved accuracy, it may become possible to ascertain details of the divergent behavior occurring in dW/dT via a criticalexponent analysis. Present accuracy does not warrant such an attempt as has been tried for $d\rho/dT$.[5,9,11]

We wish to acknowledge helpful discussions with Dr. D. J. W. Geldart and N. H. Sze, and are grateful to the National Research Council of Canada for a grant in aid to this research.

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[14]We are aware that, in general, anomalies due to T_N in nonmagnetic physical properties (such as specific heat, thermal expansion, temperature coefficient of resistivity, etc.) will not be located exactly at T_N because of the effect of a tail in the spontaneous magnetization curve persisting for $T > T_N$. This is especially true of strained or alloyed specimens {K. P. Belov and Ya. Paches, Fiz. Metal. i Metalloved. 4, 48 (1957) [translation: Phys. Metals Metallog. (USSR) 1, 35 (1957)]; A. V. Voronel, S. R. Garber, A. P. Simkina, and I. A. Charkina, Zh. Eksperim. i Teor. Fiz. 49, 429 (1965) [translation: Soviet Phys.—JETP 22, 301 (1966)]}. However such effects are quite small in annealed pure-metal samples and are likely to be small in the present case as the near coincidence of the susceptibility and $d\rho/dT$ anomalies shows.

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TEMPERATURE DEPENDENCE OF THE WIDTH AND g FACTOR OF A PURELY ELECTRONIC OPTICAL TRANSITION IN MnF_2 †

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We report thermal broadening of the 18418-cm^{-1} purely electronic transition in MnF_3 split by an external magnetic field of 25 kOe. A differential broadening of the split line is observed, demonstrating the important role of Raman scattering of magnons as a mechanism for broadening this optical line at low temperatures. A temperature-dependent reduction of the effective g factor is also observed. Calculations based on a simple model give satisfactory agreement with the observations.

Previous work[1,2] in zero external field has shown Raman scattering of magnons to be an important factor in the shift and broadening of optical lines in antiferromagnetic crystals at low temperatures. We report here results for the 18418-$cm^{-1}$$[{}^6A_1 \to {}^4T_1({}^4G)]$ purely electronic transition in MnF_2 split by an external magnetic field of 25 kOe.

The temperature-dependent renormalization of magnon energies[3] and magnon g factors[4] in antiferromagnets has been the subject of several recent studies. The renormalization of the g factor in antiferromagnetic resonance has been well established for some time.[5,6] To our knowledge this Letter reports the first observation of a temperature-dependent reduction of the effective g factor