

tained is smaller than d_{14} of InSb at near-IR frequencies.⁸

In conclusion we have demonstrated that magnetoplasma effects in semiconductors can be used for phase-matched nonlinear interactions in the far infrared. We have also shown that using a pair of CO₂ lasers, it is possible to generate microwatts of coherent power at discretely tunable frequencies in the 100-cm⁻¹ range. Since the CO₂ lasers can be tuned from 9.1 to 9.8 μ and from 10.2 to 10.8 μ, ω₃ can be tuned from 10 to 150 cm⁻¹ to cover the entire far infrared. (Generation of smaller frequencies has been reported.⁹) In Faraday configuration, it should be possible to generate circularly polarized heliconlike waves in semiconductors in the submillimeter range by phase-matched nonlinear interactions. Finally there is the exciting possibility of using the magnetoplasma effects for tunable, phase-matched, parametric oscillation in the far infrared when a semiconductor such as InSb is pumped in the 10.6-μ range with the CO₂ lasers.

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LATTICE RELAXATION IN Cr AT TEMPERATURES ABOVE T_N AFTER RAPID HEATING FROM THE INTERMEDIATE TEMPERATURE PHASE

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After rapid heating of chromium from a temperature between T_{sf} and T_N to just above T_N , a marked lattice relaxation is noticed. The relaxation times and their temperature dependence indicate that the diffusion of interstitial nitrogen—possibly ordered in tetragonal domains below T_N —is responsible for the observed effect. In order to explain the macroscopic volume strain associated with nitrogen interstitials it is believed that immediately after heating the nitrogen atoms are accompanied by residual short-range spin ordering and an associated magnetostriction. A model for the formation of domains on cooling past T_N in zero field is proposed.

The present paper is the result of difficulties in obtaining reproducible thermal-expansion curves for chromium crystals when they were cooled below the Néel temperature and subsequently heated and cooled again several times. The reason for our thermal-expansion measurements on chromium—the results of which will be published later—was a desire to study in detail the expansion of single crystals near T_N for comparison with theory. In an earlier investigation¹ it was shown that the integrated effect of the anomaly (inflection) in the expansion of single crystals as measured using a quartz dilatometer

was $\Delta l/l = 2 \times 10^{-4}$. The resolution of the dilatometer was about one fourth of the total effect [the sensitivity $\Delta(\Delta l/l) = (5-7) \times 10^{-5}$ is obtained from the reported resolution “ 10^{-5} in. or better” and the specimen size, diameter 4.8 mm, height 3.6 mm²]; i.e., although the existence of an anomaly was clearly demonstrated, a detailed study remained to be performed.

Hydrostatic weighing using specimens about 0.5 cm³ in size and a weighing sensitivity of 10^{-5} g offers a sensitivity of about 2×10^{-5} in the relative volume change, giving a resolution $\Delta(\Delta l/l)$ in the relative linear expansion of better

than 1×10^{-5} , thus making the method suitable for our purposes. The usual setback of the hydrostatic weighing method—the lack of sensitivity to anisotropic effects—puts no restriction to its usefulness here since anisotropy inside magnetic domains would not show up in a macroscopic specimen where the random distribution of single- \vec{Q} domains, if these do at all exist, should assure an overall cubic symmetry, i.e., isotropy, in the expansion data.

A number of neutron-diffraction experiments by various authors, following the early detection of antiferromagnetism in polycrystalline chromium by Shull and Wilkinson³ and the demonstration of what was at first thought to be a phase-antiphase domain structure in single crystals,⁴ have established the existence of the spin density waves (SDW) proposed by Overhauser.⁵⁻¹³ However, whereas it has been shown that field cooling (critical field: about 24 kG along a cube edge¹⁴) results in a magnetic state that can be described by a single \vec{Q} vector parallel to the applied field, no definite knowledge is available about the state obtained on cooling in zero field. All neutron-diffraction measurements show a multi- \vec{Q} (triple- \vec{Q}) state if no magnetic field is applied during cooling. It is believed, yet not fully proved, that this apparent triple- \vec{Q} state is only a macroscopic feature, the microscopic structure being an assembly of domains each of which is characterized by a single \vec{Q} vector. The SDW could, in principle, be helical or plane-polarized—in each case there are six possible waves: three Q vectors, $\vec{Q}_{[100]}$, $\vec{Q}_{[010]}$, and $\vec{Q}_{[001]}$, each of which can be associated with, in the helical case, right-hand or left-hand spiraling spins or, in the linear case, the two polarization directions along those $\langle 100 \rangle$ directions perpendicular to \vec{Q} . In the single- \vec{Q} picture only one \vec{Q} and one state of polarization should be possible inside a domain. Besides this model and the mixed triple- \vec{Q} domain model a third possibility, domains simultaneously containing two plane-polarized SDW (with varying relative populations) along $\langle 100 \rangle$ directions, was indicated by diffraction experiments on a crystal subject to a strong magnetic field oriented away from the $\langle 100 \rangle$ directions⁹; this model seems most improbable in a crystal cooled in zero field.

Strong evidence for the single- \vec{Q} picture is provided by de Haas-van Alphen results¹⁵ reflecting the tetragonality of the Fermi surface proposed by Lomer.¹⁶ A corresponding tetragonality is expected in the lattice, but no such distortion

was found so far despite very careful x-ray studies.¹⁴ A missing tetragonalization of the lattice must, of course, be regarded as a disturbing fact in the chain of evidence in favor of the single- \vec{Q} model.

Some preliminary thermal-expansion measurements around T_N presented an unexpected difficulty in obtaining reproducible results on the same crystal. Whereas good reproducibility within experimental errors was obtained on rapid thermal cycling, slow runs resulted in a gradual lowering of the density-versus-temperature curve, i.e., a time-dependent lattice expansion process seemed to be superimposed.

The actual results obtained on our studies of this phenomenon fully confirmed the existence of a lattice relaxation with relaxation times of the order of hours above the Néel temperature.

The crystal studied was a commercially available iochrome single crystal,¹⁷ $7 \times 8 \times 25$ mm³, 99.996% pure, with 2 ppm nitrogen content. The hydrostatic weighing was performed using a Mettler microbalance with sensitivity 10^{-5} g.

Before each measurement the crystal was “normalized” by heat treatment for 1 h at 100°C to eliminate traces of the preceding cooling-ordering process. The crystal was then transferred to a methyl alcohol bath held at -78°C and kept in the bath for 1 h before the rapid transfer to the thermostat bath under the balance. The time needed to reach equilibrium inside the hydrostat bath, of course, restricts this simple method to being used for a volume relaxation with relaxation times greater than about five minutes.

The lattice relaxation observed can be described by

$$a_{T,t} = a_{T,0} + \Delta a_T [1 - \exp(-t/\tau)],$$

where $a_{T,t}$ is the lattice parameter at time t after heating to temperature T , $a_{T,0}$ is the lattice parameter at time zero, Δa_T is the total relaxation at temperature T , and τ is the relaxation time.

Admittedly, what is referred to as change in lattice parameter is not necessarily strictly a lattice parameter effect, since any influence from domain walls—the structure of which is so far unknown—is included here. The relaxation was measured at five different temperatures. For convenience $\ln[1 - (a_{T,t} - a_{T,0})/\Delta a_T]$ is plotted as a function of t in Fig. 1. From the straight lines thus obtained τ is easily calculated. The results are given in Table I together with the dif-

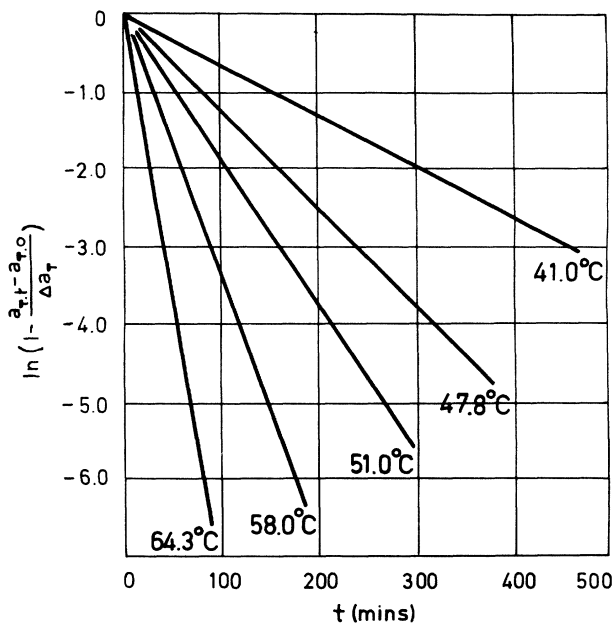


FIG. 1. Relaxation at five temperatures above the Néel point.

fusion coefficients obtained from $D = a^2/36\tau$. In Fig. 2 our results are plotted together with relaxation times due to ordered interstitial nitrogen obtained by other authors using mechanical damping and strain relaxation methods well above the Néel temperature and, in one case,¹⁸ damping due to magnetomechanical ordering of interstitial nitrogen just below T_N .^{19-25,18}

It is clearly seen from Fig. 2 that the volume relaxation observed should be attributed to the ordering of interstitial nitrogen. This could be taken as an indication (but not necessarily as a definite proof - see below) of the tetragonalization of the lattice below T_N , in accordance with the tetragonality of the Fermi surface in a linear SDW state. Now, as has been established by neutron-diffraction investigations, a crystal cooled

Table I. Results of relaxation measurements at five temperatures, including resultant calculated values of τ and the diffusion coefficient, D .

T (°C)	τ_r (sec)	D^a (10^{-21} cm ² /sec)
41.0	9200	2.5
47.8	4740	4.8
51.0	3200	7.2
58.0	1730	13.3
64.3	770	29.9

^aDiffusion coefficient of nitrogen in chromium, calculated from $D = a^2/36\tau_r$, where a is the lattice constant.

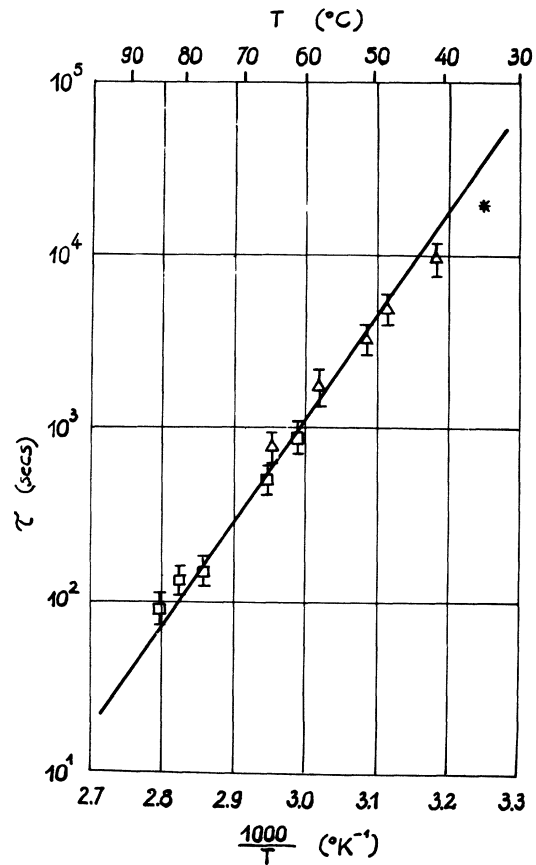


FIG. 2. Relaxation times in comparison with earlier results due to strain relaxation and magnetomechanical damping methods. Open squares and solid line from Ref. 25; asterisk from Ref. 18; and open triangles from this paper.

in zero field exhibits macroscopic cubic symmetry of the SDW, i.e., we must have a large number of single- \vec{Q} domains with democratic distribution of the different possible SDW among the domains. This makes the observed volume relaxation above the Néel temperature puzzling since a purely geometrical redistribution of interstitials from one random arrangement to another one should give rise to no macroscopic effects. We are thus led to the conclusion that on heating above T_N residual antiferromagnetic spin ordering together with an associated magnetostriction accompanies each interstitial nitrogen atom.

On the basis of our observations the following hypothetical model seems applicable:

- (1) On cooling past T_N interstitial nitrogen is responsible for the formation of domains, the direction of the \vec{Q} vector being determined by the interstitial position occupied by the nitrogen

atom. The three possible interstitial sites in the bcc lattice are $(\frac{1}{2}a, 0, 0)$, $(0, \frac{1}{2}a, 0)$, and $(0, 0, \frac{1}{2}a)$. A nitrogen atom can obviously give rise to strain fields with one of the $\langle 100 \rangle$ directions as a symmetry axis.

(2) Above T_N , after heating from a temperature below T_N the interstitial is responsible for residual magnetic ordering.

(3) Strong lattice relaxation occurs when nitrogen interstitials jump from one position to a neighboring one. When the residual ordering is localized to a very restricted region around the interstitial the strong relaxation could hardly be explained, since the localized spin ordering should accompany the interstitial even after a jump. We thus conclude that the interaction between neighboring ordered regions is strong and that the relaxation is to a great extent due to rearrangement of domain walls.

The experimental evidence obtained so far indicates a mechanism along these lines. The proposed model seems appealing, since it would answer the question of the formation and size of individual domains. The conclusion reached by Graebner and Marcus on the basis of their de Haas-van Alphen measurements¹⁵ that a crystal of very high purity cooled in zero field would consist of one single domain is in agreement with our hypothesis.

It should be remarked here that Overhauser and Arrott,⁶ in an attempt to explain the discrepancies between single crystal and polycrystal data obtained by Shull and Wilkinson,³ pointed out that when a domain could terminate on mechanical defects at a point of coincidence between the SDW nodes and lattice points, thus allowing an energetically more favorable interrelationship between the phase of the next SDW and the lattice sites, the condensation energy and the Néel temperature could be increased considerably and in addition, for sufficiently small domains, no splitting of the magnetic reflections would be obtained. Also, from the dependence of the critical temperature on volume²⁷ it is seen that a negative value of $(V_0 - V)/V_0$ (i.e., positive strain) should lead to an increasing Néel temperature. The local magnetic ordering possibly resulting from the last-mentioned effect should, of course, accompany the interstitial even after jumps to neighboring sites, as assumed above.

Finally, it should be mentioned that the possibility of a pair-breaking effect on a spin⁺, spin⁻ pair by a nonmagnetic impurity in a Lomer-Fedders-Martin^{16,28} itinerant antiferromagnet has

recently been pointed out by Zittartz.²⁸ This might combine with Overhauser-Arrott ideas to explain satisfactorily the strong influence of nitrogen interstitials on domain formation.

The model proposed here would lead to an extinction of the characteristic splitting of the magnetic superlattice neutron reflections for crystals containing approximately 10-ppm nitrogen in solution in an otherwise perfect sample. This concentration of nitrogen interstitials corresponds to a "mean free distance" for the SDW of the order of the SDW periodicity itself and would thus suppress the formation of SDW. In view of the extremely low solubility of nitrogen in chromium²⁴ a neutron diffraction study would have to be performed on a crystal quenched rapidly after heat treatment in an ammonia atmosphere. A serious consequence of a marked magnetic relaxation should be its hidden influence on almost all detailed data on the temperature dependence of various physical quantities if the time of measurement at each point is of the same order of magnitude as (or shorter than) the relaxation time at that temperature and if allowance has not been made for the crystal to relax.

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 NUCLEAR STATES OF $^{98}\text{Zr}^\dagger$

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By means of the reaction $^{96}\text{Zr}(t, p)$, we have achieved the first reported production of excited states of ^{98}Zr . Comparison of the experimental proton angular distributions with the predictions of a distorted-wave calculation has yielded spin and parity assignments for many of the states. Possible identification of neutron configurations with some of the states is discussed.

Except for half-life measurements,¹ there exists no published experimental information on the nucleus $^{98}\text{Zr}_{58}$. Since the ground state of ^{96}Zr appears to be approximately described by a closed $2d_{5/2}$ neutron subshell,² naive considerations suggest that the ^{98}Zr ground state might be approximately described by coupling two neutrons in the next $3s_{1/2}$ shell-model orbital to the ^{96}Zr ground state. However, the following two neutron orbitals, $2d_{3/2}$ and $1g_{7/2}$, lie fairly close (1 to 2 MeV) to the $3s_{1/2}$ orbital,³ and their proximity might complicate the ^{98}Zr ground state. To the extent that neutron configuration mixing appears in the ground state, excited 0^+ states will also, of course, be described by wave functions which include mixed configurations.

We have studied the ^{98}Zr nucleus by means of the reaction $^{96}\text{Zr}(t, p)^{98}\text{Zr}$. The experimental techniques have been described elsewhere.⁴ The present experiment was conducted with a 20-MeV triton beam from the Los Alamos tandem Van de Graaff facility. A preliminary run was made with a thin self-supporting target⁵ of Zr enriched to 57.4% in ^{96}Zr . Later runs were made with a target prepared in the same way, but with 82.25% enrichment in ^{96}Zr . Angular distributions were obtained with semiconductor ΔE - E telescope counters, and, in addition, spectrograph nuclear emulsion exposures were made at three angles, $\theta_{\text{lab}} = 12, 24, \text{ and } 36^\circ$. The energy resolution of the counter spectra was approximately 45 keV;

the resolution of the emulsion spectra was limited by the target thickness to approximately 25 keV.

The Q value of the ground-state transition was measured to be 3.508 ± 0.020 MeV. The calibration for this measurement was obtained from the positions of the ground-state and first-excited-state peaks of ^{18}O , obtained from the (t, p) reaction on the contaminant ^{16}O in the target, and the measured energy calibration curve of the spectrograph.

Table I shows the states observed in ^{98}Zr , with their differential cross sections at $\theta_{\text{c.m.}} \approx 37^\circ$ ($\theta_{\text{lab}} = 36.6^\circ$). The excitation energies up to 4.0 MeV are considered accurate to ± 5 keV, while the levels above 4.0 MeV have associated errors of ± 10 keV.

Distorted-wave (DW) calculations were performed for all of the neutron configurations considered to lie in the first few MeV of excitation of ^{98}Zr . The principal purpose of these calculations was to establish the L transfer for the various states observed and to obtain an estimate of the relative strength of different configurations. The DW program used for this purpose is due to Bayman and Kallio.⁶ The optical-model parameters for the triton incident channel were obtained from a computer program,⁷ which adjusted the parameters until a best fit to the measured elastic data was obtained.⁸ The proton parameters are from the work of Perey.⁷ The parameters